Experimental investigation on membrane fouling mitigation by solid-liquid fluidization

Wang, Jingwei

2018

Wang, J. (2018). Experimental investigation on membrane fouling mitigation by solid-liquid fluidization. Doctoral thesis, Nanyang Technological University, Singapore.

<http://hdl.handle.net/10356/75822>

<https://doi.org/10.32657/10356/75822>

NANYANG
TECHNOLOGICAL
UNIVERSITY

**EXPERIMENTAL INVESTIGATION ON
MEMBRANE FOULING MITIGATION BY
SOLID-LIQUID FLUIDIZATION**

WANG JINGWEI

**INTERDISCIPLINARY GRADUATE SCHOOL
NANYANG ENVIRONMENT AND WATER RESEARCH
INSTITUTE**

2018

EXPERIMENTAL INVESTIGATION ON MEMBRANE FOULING MITIGATION BY SOLID-LIQUID FLUIDIZATION

WANG JINGWEI

WANG JINGWEI

Interdisciplinary Graduate School

Nanyang Environment & Water Research Institute

A thesis submitted to the Nanyang Technological University
in partial fulfilment of the requirements for the degree of
Doctor of Philosophy

2018

ACKNOWLEDGMENTS

First of all, I would like to express my sincere appreciation to my main supervisor Assistant Professor Chew Jia Wei. Thank her for accepting me as a PhD student, and giving me the opportunity to conduct this research work. Without her invaluable guidance, support, supervision and encouragement, my research works cannot go through smoothly. Inspired by her spirits, I can then plunge into my work with immense zeal. And I also want to express my sincere gratitude to my co-supervisors Professor Anthony Gordon Fane and Professor Liu Yu for giving the crucial advices and sharing their research experiences, all of which steer my research work in a promising direction. I cordially thank my mentor Professor Wang Rong for her kind supports and encouragement.

Secondly, I would like to acknowledge the Interdisciplinary Graduate School (IGS) and Nanyang Environment and Water Research Institute (NEWRI) for funding my PhD scholarship. Thank Singapore Membrane Technology Centre (SMTC) and School of Chemical and Biomedical Engineering (SCBE) for the research funding support. Moreover, this study is also supported by research grant (1102-IRIS-02-01) provided by the Singapore National Research Foundation under its Environmental & Water Technologies Strategic Research Programme and administered by the Environment & Water Industry Programme Office (EWI) of the PUB.

Thirdly, I greatly express my thanks to project members from Singapore Membrane Technology Centre (SMTC) including Dr. Wu Bing, Mr. Lim Weikang, Ms. Wang Yuanyuan, Mr. Guo Qiming, and Dr. Li Yifei. Thank the

contributions from FYP student Kerin Guo Chun'en, URECA students Sim Hui Qi, Chen Wen, and Agarwal Vipul, IRP student Ma Yunqiao, and undergraduate students Toh Jia Yuan and Tanmay Netke.

Fourthly, I also appreciate the technicians and administrative staff from NEWRI and SMTC laboratory, especially Dr. Han Yuan, Ms. Hera Catharina Adam, Ms. Zan Ong Chin Chin, Mr. Jacky Khoo Song Sheng, Mr. Joseph Kwan Zheng Xin, and Ms. Lim Miow Wah. Without their kind assistance, I would not go smoothly and work efficiently.

Last but not least, this research work is dedicated to my beloved family and my girlfriend Dr. Cai Weiwei who always encourage me with their everlasting love when I face difficulties and challenges.

Wang Jingwei

January 2018

TABLE OF CONTENTS

ACKNOWLEDGMENTS	I
TABLE OF CONTENTS	III
SUMMARY	VIII
LIST OF PUBLICATIONS	XI
LIST OF TABLES	XIII
LIST OF FIGURES	XIV
NOMENCLATURE.....	XXVIII
Chapter 1 Introduction	1
1.1 Background.....	1
1.2 Objectives and scope	3
1.3 Organization of the thesis	4
Chapter 2 Literature review	6
2.1 Membrane fouling in MBRs	6
2.2 Unsteady-state shear to mitigate membrane fouling.....	10
2.2.1 Fouling mitigation by gas sparging	11
2.2.2 Fouling mitigation by vibrating systems	16
2.2.3 Fouling mitigation by pulsatile flow.....	19
2.2.4 Fouling mitigation by particle scouring.....	21
2.2.4.1 Gas-liquid-solid fluidization	22
2.2.4.2 Solid-liquid fluidization	25
2.3 Anaerobic fluidized bed membrane bioreactor.....	26
2.3.1 Reactor design and membrane configurations.....	28
2.3.2 Effect of fluidized media	31
2.3.3 Effect of temperature	33
2.3.4 Membrane integrity.....	34
2.4 Summary.....	37
Chapter 3 Characterizing the scouring efficiency of Granular Activated Carbon particles in membrane fouling mitigation via wavelet decomposition of accelerometer signals.....	39

3.1 Introduction.....	39
3.2 Materials and methods	40
3.2.1 Experimental setup	40
3.2.2 Materials	42
3.2.3 Accelerometer and Filtration data acquisition	43
3.2.4 Analysis of accelerometer signals.....	43
3.3 Results and discussion	49
3.3.1 Determination of minimum fluidization velocity (U_{mf}).....	49
3.3.2 Accelerometer results	50
3.3.3 Filtration results	57
3.3.4 Energy consumption	59
3.4 Conclusions.....	61
Chapter 4 Correlating the hydrodynamics of fluidized Granular Activated Carbon (GAC) with membrane-fouling mitigation	62
4.1 Introduction.....	62
4.2 Materials and methods	63
4.2.1 Experimental setup	63
4.2.2 Materials	66
4.2.3 Image analysis.....	67
4.3 Results and discussion	69
4.3.1 Determination of minimum fluidization velocity (U_{mf}).....	69
4.3.2 Fouling results.....	70
4.3.3 Influence of superficial liquid velocity (U_l) on GAC particle velocity (V_x and V_y)	71
4.3.4 GAC particle concentration	78
4.3.5 GAC particle momentum.....	80
4.3.6 Correlating GAC particle behavior with membrane-fouling mitigation	82
4.4 Conclusions.....	85
Chapter 5 Effect of fluidized granular activated carbon (GAC) on critical flux in the microfiltration of particulate foulants	87

5.1 Introduction.....	87
5.2 Materials and methods	89
5.2.1 Experimental setup	89
5.2.2 Materials	92
5.2.3 Determination of critical flux	92
5.2.4 Calculation of power requirement	94
5.3 Results and discussion	96
5.3.1 Minimum fluidization velocity (U_{mf}).....	96
5.3.2 Critical flux.....	97
5.3.3 Influence of various parameters (namely, superficial liquid velocity (U_l), height (h/H) and particle diameter (d_p)) on critical flux (J_c)... 100	
5.4 Conclusions.....	109
Chapter 6 Effect of bubble characteristics on critical flux in the microfiltration of particulate foulants	111
6.1 Introduction.....	111
6.2. Materials and methods	113
6.2.1 Experimental setup	113
6.2.2 Particulate foulant	115
6.2.3 Determination of local critical flux ($J_{c,local}$).....	117
6.2.4 Characterization of the bubbles	119
6.3. Results and Discussion	121
6.3.1. Local critical flux ($J_{c,local}$)	121
6.3.2. Effect of operation conditions on local critical flux ($J_{c,local}$).....	124
6.3.3. Influence of operating conditions on bubble behavior	125
6.3.4. Correlating bubble characteristics with membrane fouling.....	130
6.3.5. Bubble flow patterns.....	133
6.3.6. Energy consumption	138
6.4. Conclusions.....	142
Chapter 7 Relationship between scouring efficiency and overall concentration of fluidized granular activated carbon (GAC) in microfiltration	145

7.1 Introduction.....	145
7.2 Materials and methods	148
7.2.1 Experimental setup	148
7.2.2 Materials	152
7.3 Results and Discussion	153
7.3.1 Minimum fluidization velocity (U_{mf}).....	153
7.3.2 Determination of critical flux (J_c) via DOTM	155
7.3.3. Local critical flux ($J_{c,local}$)	157
7.3.4. Power requirement (P_r).....	161
7.3.5. Foulant type	168
7.3.6. Comparative analysis of the overall GAC concentration and GAC particle diameter with respect to membrane fouling mitigation	173
7.4 Conclusions.....	175
Chapter 8 Characteristics of non-spherical fluidized particles in a fluidized bed – membrane bioreactor: Effect on critical flux.....	177
8.1. Introduction.....	177
8.2. Materials and methods	180
8.2.1 Experimental setup	180
8.2.2 Materials	182
8.2.3 Determination of local critical flux ($J_{c,local}$)	184
8.2.4 Characterization of fluidization hydrodynamics.....	185
8.3. Results and Discussion	191
8.3.1. Local critical flux ($J_{c,local}$)	191
8.3.2. Effect of particle sphericity (ϕ) of fluidized rice media on local critical flux ($J_{c,local}$)	193
8.3.3. Effect of particle sphericity (ϕ) of fluidized rice media on hydrodynamics	195
8.3.4. Correlating membrane fouling mitigation with hydrodynamics.....	201
8.3.5. Overall critical flux ($J_{c,overall}$) with respect to hydrodynamics parameters and energy requirement	204
8.4. Conclusions.....	208

Chapter 9 Monitoring local membrane fouling mitigation by fluidized GAC in lab-scale and pilot-scale AnFMBRs	211
9.1 Introduction.....	211
9.2 Materials and methods	213
9.2.1 Lab-scale setup	213
9.2.2 Pilot-scale setup	214
9.2.3 Materials	218
9.2.4 Accelerometer signals.....	218
9.3. Results and Discussion	223
9.3.1. Wavelet decomposition of accelerometer signals.....	223
9.3.2. Using the accelerometer signals from a lab-scale reactor to study the effect of superficial liquid velocity (U_l) and overall GAC concentration	228
9.3.3. Using the accelerometer signals from a pilot-scale reactor to study the effect of inlet liquid flow rate (Q) and overall GAC mass	230
9.3.4. Monitoring the pilot-scale AnFMBRs	233
9.4. Conclusions.....	236
Chapter 10 Conclusions and Recommendations.....	239
10.1. Conclusions.....	239
10.2 Recommendations.....	242
REFERENCES.....	245

SUMMARY

Particle fluidization is a promising unsteady-state shear means of mitigating membrane fouling. It has the potential to lower energy requirement and prolong membrane lifespan in membrane-based wastewater treatment processes. In particular, the particles facilitate back-transport in the polarization layer and also play the role of mechanical scouring agents, the latter of which is dominant for millimeter-sized particles.

In this study, an accelerometer was first successfully used to unveil the dynamics of GAC particles with three particle diameters (namely, 1.20 mm, 1.85 mm, and 2.18 mm). The energy contained in the accelerometer signal representing the solid phase was consistent with the extent of fouling mitigation in the filtration tests. The smallest GAC particle with a particle diameter of 1.20 mm conferred the least scouring efficiency, while both the larger particles with diameters of 1.85 mm and 2.18 mm provided similar scouring efficiency. The impact of particle diameter is due to the balance between particle inertia, and the difference between superficial liquid velocity and minimum fluidization velocity. Furthermore, in a practical pilot-scale AnFMBR with wastewater, the accelerometer was shown to be able to diagnose the de-fluidization of the GAC particles due to severe clogging issues, which thereby eliminated the fouling mitigation capacity. Trends in the trans-membrane pressure (TMP) and flux data did not indicate de-fluidization of the GAC particles.

In order to further improve this technique in terms of scouring efficiency and energy requirement, the hydrodynamics of the fluidized GAC particles, and

correspondingly the relationship between the particle behavioural characteristics and the extent of membrane-fouling mitigation were investigated. Directly correlating particle behavior with membrane fouling trends showed that higher particle velocities, particle concentration and particle momentum generally all showed a negative correlation with $dTMP/dt$. Of the three characteristics (namely, particle velocity, concentration and momentum), the negative correlation between $dTMP/dt$ and particle momentum was most pronounced, which suggests that momentum transfer between the GAC particles and the membrane represents a key mechanism effecting the scouring to diminish membrane fouling.

More in-depth studies of the improvement of micrometer-sized particulate (namely, polystyrene foulant) critical flux in the presence of fluidized GAC particles was studied, whereby a Direct Observation Through the Membrane (DOTM) technique was used to measure the membrane critical flux locally and overall. It was found that (i) the fluidized GAC particles increased critical flux by an order-of-magnitude relative to that with tangential liquid shear alone; (ii) the overall critical flux increased with the increase of GAC particle diameter or power input; and (iii) a higher power input was required for sufficient bed expansion to enable more consistent critical flux values over the whole membrane height.

Accordingly, the impact of different overall GAC concentrations, which give different expanded bed heights, on the extent of fouling mitigation was investigated in this study. The results indicate that the highest overall concentration was advantageous in terms of enabling (i) higher membrane fouling mitigation at a lower power required per unit permeate (P_p) for the

polystyrene foulant but not bentonite foulant; and (ii) more similar local critical flux values over the three heights along the membrane investigated, which implies more uniform fouling mitigation across the membrane. The relative benefits of adjusting overall GAC concentration versus the particle diameter of the GAC media were also critically assessed.

Finally, the effect of the sphericity (ϕ) of the fluidized media on the extent of fouling mitigation was studied. Three types of millimeter-sized rice media were used as the fluidized media. Relative to GAC media, the rice media had similar particle densities and similar particle diameters, but offered much lower sphericities (lowest sphericity in this study was 0.54) due to the greater aspect ratios. The results indicate that the correlation between sphericity of the fluidized media and hydrodynamics (and the attendant fouling mitigation) was not straightforward, likely tied to the influence of drag on the countering effects of media-membrane contact and particle rotation.

In conclusion, this thesis gave a deep understanding of the fluidized GAC particle behavioral characteristics and the correlation with the extent of membrane-fouling mitigation in solid-liquid fluidization membrane filtration systems. The parameters investigated included GAC particle diameter (d_p), overall GAC concentration, fluidized particle sphericity (ϕ), superficial liquid velocity (U_l) thereby the power requirement (P_r), and height along the vertically aligned membrane. The findings are expected to be valuable in the optimization of membrane fouling mitigation towards more energy-efficient in a solid-liquid fluidization membrane filtration system.

LIST OF PUBLICATIONS

1. **Jingwei Wang**, Anthony G. Fane, Jia Wei Chew. Effect of bubble characteristics on critical flux in the microfiltration of particulate foulants. *Journal of Membrane Science*, Volume 535, 1 August 2017, Pages 279-293.
2. **Jingwei Wang**, Bing Wu, Yu Liu, Anthony G Fane, Jia Wei Chew. Effect of fluidized granular activated carbon (GAC) on critical flux in the microfiltration of particulate foulants. *Journal of Membrane Science*, Volume 523, 1 February 2017, Pages 409 – 417.
3. **Jingwei Wang**, Farhad Zamani, Andy Cahyadi, Jia Yuan Toh, Shiliang Yang, Bing Wu, Yu Liu, Anthony G Fane, Jia Wei Chew. Correlating the hydrodynamics of fluidized granular activated carbon (GAC) with membrane-fouling mitigation. *Journal of Membrane Science*, Volume 510, 15 July 2016, Pages 38 – 49.
4. **Jingwei Wang**, Bing Wu, Shiliang Yang, Yu Liu, Anthony G Fane, Jia Wei Chew. Characterizing the scouring efficiency of Granular Activated Carbon (GAC) particles in membrane fouling mitigation via wavelet decomposition of accelerometer signals. *Journal of Membrane Science*, Volume 498, 15 January 2016, Pages 105 – 115.
5. **Jingwei Wang**, Anthony G. Fane, Jia Wei Chew. Relationship between scouring efficiency and overall concentration of fluidized granular activated carbon (GAC) in microfiltration. *Chemical Engineering Research and Design*, Volume 132, April 2018, Pages 28-39.
6. **Jingwei Wang**, Anthony G. Fane, Jia Wei Chew. Characteristics of non-spherical fluidized media in a fluidized bed – membrane bioreactor:

- Effect of particle sphericity on critical flux. *Separation and Purification Technology*, Volume 202, 31 August 2018, Pages 185-199.
7. **Jingwei Wang**, Bing Wu, Yu Liu, Anthony G. Fane, Jia Wei Chew. Monitoring local membrane fouling mitigation by fluidized GAC in lab-scale and pilot-scale AnFMBRs. *Separation and Purification Technology*, Volume 199, 30 June 2018, Pages 331-345.
8. **Jingwei Wang**, Anthony G. Fane, Jia Wei Chew. Understanding the anaerobic fluidized membrane bioreactor for wastewater treatment. *European Water*, Issue 58, 2017, Pages 371-374. (Conference paper)
9. Shiliang Yang, Andy Cahyadi, Yuhao Sun, **Jingwei Wang**, Jia Wei Chew. CFD-DEM investigation into the scaling up of spout-fluid beds via two interconnected chambers. *AIChE Journal*. Volume 62, Issue 6, June 2016, Pages 1898 – 1916.
10. Shiliang Yang, Andy Cahyadi, **Jingwei Wang**, Jia Wei Chew. DEM study of granular flow characteristics in the active and passive regions of a three-dimensional rotating drum. *AIChE Journal*. Volume 62, Issue 11 November 2016, Pages 3874 – 3888.
11. Shiliang Yanga, Yuhao Sun, **Jingwei Wang**, Andy Cahyadi, Jia Wei Chew. Influence of operating parameters and flow regime on solid dispersion behavior in a gas-solid spout-fluid bed. *Chemical Engineering Science*. Volume 142, 13 March 2016, Pages 112–125.

LIST OF TABLES

Table 2-1. The removal efficiency and energy requirement of AnFMBRs with a single staged and two staged systems.	30
Table 4-1. Particle coordinates of the in-focus particles in Fig. 4-3 and the corresponding particle velocities. (x_1, y_1) are particle coordinates obtained from Fig. 4-3a, and (x_2, y_2) are particle coordinates obtained from Fig. 4-3b. Distance moved $(\Delta x, \Delta y)$ was derived by $(x_2 - x_1, y_2 - y_1)$, while particle velocities v_x and v_y were respectively calculated by $\Delta x / 0.01$ and $\Delta y / 0.01$	69
Table 6-1. Bubble parameters obtained from Fig. 6-4.....	121
Table 8-1. Pros and cons of fluidizing smaller GAC relative to larger ones for mitigating membrane fouling.....	178
Table 8-2. Rice media parameters	184
Table 8-3. Particle behavioral characteristics obtained from Figs. 8-5 and 8-6.	190
Table 9-1. Operating conditions for the pilot-scale two-stage wastewater treatment.	217

LIST OF FIGURES

Figure 2-1. Formation and removal of removable and irremovable fouling in MBRs. Adapted from [32].	8
Figure 2-2. The parameters and relationships that influence membrane fouling performance in an anaerobic membrane bioreactor. Adapted from [65].	9
Figure 2-3. Bubble flow pattern in tubular channels. Adapted from [6].	12
Figure 2-4. Wall shear stress distribution around a gas slug rising in stationary liquid in a vertical tubular channel, results were measured using the electrochemical shear method. Adapted from [106].	13
Figure 2-5. Flux enhancement versus gas/liquid ratio with different feed types. Adapted from [6].	14
Figure 2-6. Three-dimensional (3D) plot correlating gas (U_G) and liquid superficial velocities (U_L) with mean flux enhancement in a vertically (a) flat-sheet membrane module and (b) hollow fiber membrane module. Adapted from [6].	15
Figure 2-7. Schematic of membrane vibration in a VSEP system. Adapted from [114].	16
Figure 2-8. Schematic of submerged hollow fiber membrane modules by vibrations (a) transversely and (b) longitudinally. (a) adapted from [121] and (b) adapted from [116].	18
Figure 2-9. Velocity profiles with and without flow pulsation. Adapted from [127].	20
Figure 2-10. Energy consumption for different membrane filtration systems. Adapted from [51].	22
Figure 2-11. Photos of membrane modules after the operations of: (A) without carriers and (B) 1% additional scouring media. Adapted from [138].	24
Figure 2-12. Schematic of two staged anaerobic fluidized bed membrane bioreactor (AnFMBR). Adapted from [153].	28
Figure 2-13. Structure of the distributor used in rectangular AnFMBRs.	29

Figure 2-14. SEM and AFM images of membrane surfaces in a bench scale AnFMBR setup. (a) and (c) with GAC fluidization; (b) and (d) without GAC fluidization. Adapted from [157].	35
Figure 2-15. Membrane surface damage with exposed braid near the bottom of the pilot AFMBR after 2 years operation. Adapted from [169].	36
Figure 2-16. SEM images of membrane surface using small GAC (50% v/v) as scouring media. The feed is 300 mg/L sodium alginate solution. Adapted from [188].	37
Figure 3-1. (a) Schematic of experimental setup, and (b) Distributor in the liquid-solid fluidization reactor to ensure uniform fluidization.	41
Figure 3-2. Typical experimental accelerometer signal collected at 40,000 Hz at a superficial liquid velocity (U_l) of 0.0085 m/s for (a) liquid-solid fluidization of GAC particles with particle diameter (d_p) of 2.18 mm, (b) liquid-solid fluidization of GAC particles with particle diameter (d_p) of 1.85 mm, (c) liquid-solid fluidization of GAC particles with particle diameter (d_p) of 1.20 mm, and (d) liquid only (i.e., without GAC).	45
Figure 3-3. Average values of accelerometer signal for different GAC sizes	46
Figure 3-4. Wavelet decomposition analysis of collected signal into various scales [211].	46
Figure 3-5. Raw signal of accelerometer collected at 40,000 Hz and corresponding approximations (A_j) and details (D_j) at various scales (j) of wavelet decomposition for GAC with a particle diameter (d_p) of 1.20 mm and at a superficial liquid velocity (U_l) of 0.0085 m/s.	48
Figure 3-6. Fluidization curves for GAC particles with diameters (d_p) of (a) 2.18 mm, (b) 1.85 mm, and (c) 1.2 mm.	50
Figure 3-7. Wavelet energy distribution of accelerometer sub-signals with different sampling frequency (namely, 10 kHz, 20 kHz, 30 kHz, and 40 kHz), data obtained the fluidization of GAC particles with diameter of 2.18 mm at $U_l = 0.0053$ m/s.	51

Figure 3-8. Energy associated with each detail signal obtained via wavelet decomposition at various superficial liquid velocities for (a) GAC particles with diameter of 2.18 mm, (b) GAC particles with diameter of 1.85 mm, (c) GAC particles with diameter of 1.20 mm, and (d) liquid flow alone (i.e., without GAC particles).....	53
Figure 3-9. Energy associated with each detail signal obtained via wavelet decomposition at superficial liquid velocities of (a) 0.0042 m/s (i.e., lower than the U_{mf} of the smallest GAC particle), (b) 0.0053 m/s (i.e., U_{mf} of the GAC particle with diameter of 1.20 mm), (c) 0.0074 m/s (i.e., U_{mf} of the GAC particle with diameter of 1.85 mm), and (d) 0.0085 m/s (i.e., U_{mf} of the GAC particle with diameter of 2.18 mm).	55
Figure 3-10. Energy associated with detail signals D_1 to D_3 obtained via wavelet decomposition ($E_{D1} + E_{D2} + E_{D3}$) as a function of liquid velocity (U_l).	57
Figure 3-11. (a) TMP versus time in the absence and presence of fluidized GAC at two different U_l values; (b) Relationship between $dTMP/dt$ and $E_{D1} + E_{D2} + E_{D3}$	59
Figure 3-12. Energy associated with detail signals D_1 to D_3 obtained via wavelet decomposition ($E_{D1} + E_{D2} + E_{D3}$) as a function of particle diameter (d_p).	60
Figure 4-1. Schematic of fluidized bed – membrane filtration experimental setup	64
Figure 4-2. Schematic and dimensions of (a) fluidized bed – membrane filtration module, and (b) distributor. Dimensions are in units of mm.	65
Figure 4-3. Images extracted from a video clip, whereby the particle diameter (d_p) was 1.85 mm and the superficial liquid velocity (U_l) was 0.06 m/s. (a) and (b) are 0.01 s apart; only particles in focus as identified on the images were analyzed.	68
Figure 4-4. Fouling rate values of GAC particle velocity with diameters (a) 1.85 mm, (b) 1.55 mm and (c) 1.015 mm at different heights and superficial velocities.	71

Figure 4-5. Particle velocity distribution in the horizontal (x) direction (V_x). Each of the three columns represents a particle diameter (d_p), while each of the five rows represents a dimensionless height of the fluidized bed - membrane module (h/H , where h is the height assessed while H is the total module height of 194 mm (Fig. 4-2)).....	73
Figure 4-6. Particle velocity distribution in the vertical (y) direction (V_y). Each of the three columns represents a particle diameter (d_p), while each of the five rows represents a dimensionless height of the fluidized bed - membrane module (h/H , where h is the height assessed while H is the total module height of 194 mm (Fig. 4-2)).	74
Figure 4-7. Root-mean-square particle velocities in the x -direction ($V_{rms,x}$; first column) and y -direction ($V_{rms,y}$; second column) at different U_l . Each of the three rows represents a d_p investigated.	76
Figure 4-8. V_p versus U_l trends at five heights (h/H) for three different d_p : (a) 1.85mm, (b) 1.55 mm and (c) 1.015 mm.	78
Figure 4-9. GAC particle concentration in terms of particle number per m^2 versus U_l trends at five heights (h/H) for three different d_p : (a) 1.85mm, (b) 1.55 mm and (c) 1.015 mm.	80
Figure 4-10. Particle momentum (p) per m^2 versus U_l trends at five heights (h/H) for three different d_p : (a) 1.85mm, (b) 1.55 mm and (c) 1.015 mm.	82
Figure 4-11. $dTMP/dt$ versus (a) V_p , (b) particle concentration, (c) particle momentum (p), and (d) energy requirement per unit permeate (P_p).	84
Figure 5-1. Schematic of the vertical-DOTM setup.	91
Figure 5-2. (a) Schematic of the fluidized bed - membrane filtration cell. All dimensions are in units of mm; (b) Photograph of the stainless steel spacer.	91
Figure 5-3. DOTM images of polystyrene particles deposited on the membrane surfaces: (a) in the presence of fluidized GAC particles, and (b) in the absence of GAC particles.....	94

Figure 5-4. Bed height (Δz) versus superficial liquid velocity (U_l) for GAC particles with diameters (d_p) of 1.85 mm, 1.55 mm and 1.015 mm	97
Figure 5-5. Trends of permeate flux and trans-membrane pressure (TMP) with respect to time for the fluidized GAC particles with $d_p = 1.85$ mm at $U_l = 0.032$ m/s (3.5 times U_{mf}). The critical flux was determined to be 45.8 L/(m ² h) by both the TMP and DOTM methods.	98
Figure 5-6. DOTM images at $U_l = 0.032$ m/s (3.5 times U_{mf}) and $h/H = 0.46$ for the fluidized GAC particles with $d_p = 1.85$ mm at the 10 th minute of the flux of (a) 15.7 L/(m ² h) (i.e., below critical flux), (b) 45.8 L/(m ² h) (i.e., at critical flux), and (c) 60.4 L/(m ² h) (i.e., above critical flux).	99
Figure 5-7. Rate of change of the number of deposited polystyrene particles (dN/dt) versus flux at $U_l = 0.032$ m/s and $h/H = 0.46$. Critical fluxes were determined as 45.8 L/(m ² h) and 5 L/(m ² h) for the cases of fluidized GAC with $d_p = 1.85$ mm and when GAC was absent, respectively.	100
Figure 5-8. Critical flux (J_c) with respect to superficial liquid velocity (U_l) at $h/H = 0.29$ for the three particle diameters (d_p) investigated.	102
Figure 5-9. High-speed video images of fluidized GAC particles with $d_p = 1.85$ mm at the superficial liquid velocities (U_l) of (a) 0.027 m/s (i.e., $U_l/U_{mf} = 3$), and (b) 0.041 m/s (i.e., $U_l/U_{mf} = 4.5$). Images were taken at $h/H = 0.29$	103
Figure 5-10. Critical flux (J_c) at various heights (h/H) and superficial liquid velocities (U_l) for GAC particles with diameters (d_p) of (a) 1.85 mm, (b) 1.55 mm, and (c) 1.015 mm.	104
Figure 5-11. Overall critical flux ($J_{c,overall}$) versus power requirement with GAC particle diameters of 1.85 mm, 1.55mm, and 1.015 mm.	106
Figure 5-12. The $J_c/J_{c,overall}$ ratio at various heights (h/H) versus power requirement (P_r) for GAC particles with diameters (d_p) of (a) 1.85 mm, (b) 1.55mm, and (c) 1.015 mm.	107

Figure 5-13. Three-dimensional (3D) plot correlating power requirement (P_r) and GAC particle diameter (d_p) with the overall critical flux ($J_{c,overall}$).	109
Figure 6-1. Schematic of the vertical DOTM setup.....	116
Figure 6-2. (a) Schematic of the membrane filtration cell, with the circled region depicted in the photo in (b); (b) Photograph of the inlet of the feed chamber to distribute the bubbles; (c) components of membrane filtration cell.....	117
Figure 6-3. Sequential images 0.2 s apart of the membrane area assessed at $h/H = 0.29$. The black lines represent the contact areas between the bubble and the membrane. The permeate flux was 109 L/(m ² h) (which was below the local critical flux ($J_{c,local}$)), while the liquid (Q_l) and gas (Q_g) flow rates were respectively 0.6 L/min and 0.3 L/min.	119
Figure 6-4. Sequential images taken 0.0033 s apart of the bubbles at $h/H = 0.71$. The liquid flow rate (Q_l) was 1.0 L/min, gas flow rate (Q_g) was 0.5 L/min, and permeate flux was 0 L/(m ² h).	121
Figure 6-5. Images captured at the end of the 10 th minute at the targeted permeate fluxes, which correspond to (a) below the local critical flux ($J_{c,local}$), (b) the local critical flux ($J_{c,local}$), and (c) above the local critical flux ($J_{c,local}$). The liquid flow rate (Q_l) was 0.6 L/min and the gas flow rate (Q_g) was 0.3 L/min.....	123
Figure 6-6. (a) Plot of the rate of change of the number of deposited polystyrene particles (dN/dt) with respect to permeate flux to determine the local critical flux ($J_{c,local}$), which was 139.1 L/(m ² h) in this case; (b) Trends of trans-membrane pressure (TMP) and permeate flux with respect to time. The liquid flow rate (Q_l) was 0.6 L/min, the gas flow rate (Q_g) was 0.3 L/min, and the height (h/H) was 0.29.	123
Figure 6-7. Two sequential images 5 s apart showing the deposition of bubbles on the membrane. The permeate flux was 197.8 L/(m ² h) (i.e., above $J_{c,local}$), $Q_l = 0.6$ L/min, $Q_g = 1.0$ L/min and $h/H = 0.29$	124
Figure 6-8. Local critical flux ($J_{c,local}$) versus dimensionless heights (h/H) at (a) a constant liquid flow rate (Q_l) of 0.6 L/min and different gas flow rates	

(Q_g); and (b) a constant gas flow rate (Q_g) of 0.5 L/min and different liquid flow rates (Q_l)	125
Figure 6-9. Probability distributions of bubble velocity (V_b) at three dimensionless heights (namely, $h/H = 0.29, 0.50$ and 0.71).	126
Figure 6-10. Effect of gas flow rate (Q_g) with respect to height (h/H) at a constant Q_l of 0.6 L/min on: (a) mean bubble velocity ($V_{b,rms}$), (b) bubble area per unit area assessed (A_b), (c) number of bubbles per unit area assessed (N_b) and (d) mean area per bubble (A_b/N_b). Effect of liquid flow rate (Q_l) with respect to height (h/H) at a constant Q_g of 0.5 L/min on: (e) mean bubble velocity ($V_{b,rms}$), (f) bubble area per unit area assessed (A_b), (g) number of bubbles per unit area assessed (N_b), and (h) mean area per bubble (A_b/N_b).	129
Figure 6-11. Bubble momentum (P_b) versus gas flow rate (Q_g) at various dimensionless heights (h/H) at a constant liquid flow rate (Q_l) of 0.6 L/min.	131
Figure 6-12. Correlating the local critical flux ($J_{c,local}$) with (a) mean bubble velocity ($V_{b,rms}$), (b) bubble area per unit membrane area (A_b), (c) number of bubbles per unit membrane area (N_b), (d) mean area per bubble (A_b/N_b), and (e) bubble momentum (P_b).	133
Figure 6-13. Bubble flow patterns in the microfiltration cell at $Q_l = 0.6$ L/min. The top and bottom rows present the upper ($h/H = 0.53 - 1$) and lower sections ($h/H = 0 - 0.45$), respectively, of the membrane cell.	135
Figure 6-14. Example of the trajectory of three bubbles in the cell at $Q_g = 0.6$ L/min and $Q_l = 0.3$ L/min. h/H is the dimensionless height, whereby h is the height assessed and H is the total cell height of 146 mm; and w/W is the dimensionless width whereby w is the width assessed and W is the total cell width of 54 mm.	136
Figure 6-15. Sequential images 0.0033 s apart of the bubble coalescence phenomenon.	136
Figure 6-16. Overall critical flux ($J_{c,overall}$) versus gas to total flow rates ratio (θ ; Eq. (6-6)).	137

Figure 6-17. Ratio of local to overall critical flux critical flux ($J_{c,local}/J_{c,overall}$) versus power requirement (P_r) at the three dimensionless heights (h/H) investigated.....	140
Figure 6-18. Overall critical flux ($J_{c,overall}$) versus power requirement (P_r) for microfiltration systems employing bubbling and fluidized GAC particles as means of membrane fouling mitigation.	140
Figure 7-1. Schematic of fluidized bed membrane filtration cell - DOTM setup.	149
Figure 7-2. (a) Components of the solid-liquid fluidization membrane filtration cell; and (b) Schematic of the four different plastic sheets used for the experiments: the first three plastic sheets had Anopore membranes (diameter of 47 mm) attached for the DOTM tests (h/H is the ratio of the height assessed to the total membrane cell height of 146 mm), while the fourth plastic sheet had a PVDF membrane attached for the dTMP/dt test.....	151
Figure 7-3. Particle size distributions (PSDs) of polystyrene and bentonite particles	153
Figure 7-4. Bed height (z) versus superficial liquid velocity (U_l) for overall GAC ($d_p = 1.55$ mm) concentrations of 50 g/L, 100 g/L and 150 g/L, which corresponded respectively to initial packed bed heights of 0.019 m, 0.035 m and 0.057 m. The vertical dotted line represents the minimum fluidization velocity (U_{mf}).	155
Figure 7-5. (a) DOTM images at the end of the 1 st and 10 th minute interval for four permeate fluxes at $h/H = 0.29$, $U_l = 0.021$ m/s and overall GAC ($d_p = 1.55$ mm) concentration of 150 g/L; (b) the rate of the change of the number of deposited polystyrene particles (dN/dt) versus flux at $h/H = 0.29$, $U_l = 0.021$ m/s and overall GAC ($d_p = 1.55$ mm) concentration of 150 g/L, which indicates the local critical flux ($J_{c,local}$) was 81 L/(m ² h).	157
Figure 7-6. Local critical flux ($J_{c,local}$) of three overall GAC ($d_p = 1.55$ mm) concentrations with respect to superficial liquid velocities (U_l) at (a) $h/H = 0.29$, (b) $h/H = 0.46$, and (c) $h/H = 0.62$. The foulant was	

polystyrene particles. The hollow symbols represent the cases whereby the fluidized GAC was not in contact with the membrane, hence the critical flux was similar to that in the system without GAC.	
.....	160
Figure 7-7. Dimensionless height (h/H) versus local critical flux ($J_{c,local}$) for fluidized GAC ($d_p = 1.55$ mm) media with overall concentrations of (a) 50 g/L, (b) 100 g/L and (c) 150 g/L at different superficial liquid velocities (U_l).	161
Figure 7-8. Overall critical flux ($J_{c,overall}$) versus (a) superficial liquid velocity (U_l), (b) power requirement (P_r), and (c) power requirement per unit permeate (P_p) for the three overall concentrations of GAC ($d_p = 1.55$ mm) investigated using polystyrene particles as foulants.....	164
Figure 7-9. Ratios of $J_{c,local} / J_{c,overall}$ versus power requirement (P_r) at various heights (h/H) along the membrane for overall GAC ($d_p = 1.55$ mm) concentrations of (a) 50 g/L, (b) 100 g/L, and (c) 150 g/L.....	166
Figure 7-10. Superficial liquid velocity (U_l), pressure drop (ΔP_{bed}), and power requirement (P_r) versus overall GAC concentrations for a constant expanded bed height of 0.104 m.	167
Figure 7-11. dTMP/dt versus (a) superficial liquid velocity (U_l), (b) power requirement (P_r), and (c) power requirement per unit permeate (P_p) for the three overall concentrations of GAC ($d_p = 1.55$ mm) investigated using 2 g/L of bentonite feed at a constant permeate flux 30 L/(m ² h).	170
Figure 7-12. dTMP/dt versus the ratio of feed suspension to GAC concentration for GAC media with diameter of 1.55 mm at constant fluidization bed height of 0.104 m.	172
Figure 7-13. GAC deposition amount per unit membrane versus the ratio of U_l to U_{mf} at an overall GAC concentration of 100 g/L and TMP of 30 kPa.	173
Figure 8-1. Schematic of experimental setup.	182
Figure 8-2. (a) Photograph of the three types of rice media used in the experiments; and (b) characteristic dimensions of an ellipsoid. ...	184

Figure 8-3. DOTM images of the deposited polystyrene particles (pink circles) after 10 minutes at 86.9 L/(m ² h), which was above the critical flux of 48.4 L/(m ² h). Particle sphericity (ϕ) and equivalent spherical diameter (d_s) of the fluidized rice were 0.54 and 3.25 mm, respectively (Type 1). The images are 1 s apart.	185
Figure 8-4. Fluidized bed height (L) versus superficial liquid velocity (U_l) for the three types of rice media for the determination of minimum fluidization velocities (U_{mf}).....	187
Figure 8-5. Images taken 0.01 s apart of the fluidized rice media ($\phi = 0.54$; $d_s = 3.25$ mm; Type 1) at h/H of 0.62 and superficial liquid velocity (U_l) of 0.061 m/s ($U_l/U_{mf} = 2.8$).	189
Figure 8-6. Processed images from Fig. 5, with the green color denoting the areas of the fluidized rice media that were in contact with the membrane.	189
Figure 8-7. (a) Rate of change of the number of deposited polystyrene particles (dN/dt) with respect to permeate flux to determine the local critical flux ($J_{c,local}$), which was 48.4 L/(m ² h) in this case; and (b) Evolution of trans-membrane pressure (TMP) during the flux-stepping experiment. The superficial liquid velocity was 0.053 m/s (i.e., $U_l/U_{mf} = 2.4$), and ϕ and d_s of the fluidized rice were respectively 0.54 and 3.25 mm (Type 1), and h/H was 0.46.....	192
Figure 8-8. DOTM images at the end of the 10 th minute at (a) below the local critical flux ($J_{c,local}$), (b) the local critical flux ($J_{c,local}$), and (c) above the local critical flux ($J_{c,local}$). The superficial liquid velocity was 0.053 m/s (i.e., $U_l/U_{mf} = 2.4$), and ϕ and d_s of the fluidized rice were respectively 0.54 and 3.25 mm (Type 1), and h/H was 0.46.	193
Figure 8-9. Plots of local critical flux ($J_{c,local}$) and critical flux ratio ($J_{c,local}/J_{c,overall}$) versus superficial liquid velocity (U_l) for fluidized rice media with sphericity (ϕ) values of 0.54, 0.60 and 0.77 at various heights (h/H).	195
Figure 8-10. Probability distributions of particle velocity (V_p).	196
Figure 8-11. Probability distributions of particle rotation velocity (V_θ).....	197

Figure 8-12. Plots of average particle velocity ($V_{p,rms}$) and average particle rotation velocity ($V_{\theta,rms}$) versus superficial liquid velocity (U_l) for fluidized rice media with sphericity (ϕ) values of 0.54, 0.60 and 0.77 at various heights (h/H).....	199
Figure 8-13. Plots of contact area per unit membrane area (A_p) and particle momentum per unit membrane area (P_p) versus superficial liquid velocity (U_l) for fluidized rice media with sphericity (ϕ) values of 0.54, 0.60 and 0.77 at various heights (h/H).....	201
Figure 8-14. Correlating the local critical flux ($J_{c,local}$) with mean particle velocity ($V_{p,rms}$), mean particle rotation velocity ($V_{\theta,rms}$), contact area per unit membrane area (A_p), and particle momentum per unit membrane area (P_p) for the fluidized rice media of different sphericities (ϕ).	204
Figure 8-15. Correlating overall critical flux ($J_{c,overall}$) with (a) overall particle velocity ($V_{p,overall}$), (b) overall particle rotation velocity ($V_{\theta,overall}$), (c) overall particle contact area per unit membrane area ($A_{p,overall}$), and (d) overall particle momentum per unit membrane area ($P_{p,overall}$).	206
Figure 8-16. Correlating overall critical flux ($J_{c,overall}$) with power requirement (P_r) for the fluidized rice media and GAC media (higher sphericity (ϕ) of 0.83; lower overall concentration of 100 g/L in Chapter 5).	208
Figure 9-1. Schematic of the lab-scale AnFMBR reactor. The black dots on the reactor represent positions where accelerometer signals were taken; signals at different sides of the reactor (namely, West, South and East) were taken to determine any mal-distribution of the fluidized GAC particles.....	214
Figure 9-2. Schematic of the pilot-scale two-stage AnFMBR [13], comprising of the first-stage AnFBR and two parallel second-stage AnFMBRs, with one using flat-sheet membranes (FS-AnFMBR) and the other hollow-fiber membranes (HF-AnFMBR).	216
Figure 9-3. Photograph of the pilot-scale two-stage wastewater treatment [13] facility housed in the Ulu Pandan Water Reclamation Plant	

(Singapore), comprising of the first-stage AnFBR and two parallel second-stage AnFMBRs, with one using flat-sheet membranes and the other hollow-fiber membranes.	216
Figure 9-4. (a) Dimensions of the pilot-scale AnFMBR with flat-sheet membranes (FS-AnFMBR); and (b) photograph of the flat-sheet membrane cassette inside the FS-AnFMBR (view from the south side).....	217
Figure 9-5. Accelerometer signals obtained in lab-scale reactor as a function of time at h/H of 0.57, superficial liquid velocity (U_l) of 0.033 m/s, overall GAC concentration of 150 g/L, and permeate flux of 0 L/(m ² h). Each row represents each side of the reactor that the signals were collected at, while the left and right columns represent respectively the absence and presence of fluidized GAC.	220
Figure 9-6. Accelerometer signals obtained in pilot-scale FS-AnFMBR (with a new membrane cassette and DI water) as a function of time at h/H of 0.57, inlet flow rate of 67 m ³ /h, 50 kg of GAC particles, and permeate flux of 0 L/(m ² h). Each row represents each side of the reactor that the signals were collected at, while the left and right columns represent respectively the absence and presence of fluidized GAC.	221
Figure 9-7. Wavelet decomposition analysis of accelerometer signal into various scales [272].	222
Figure 9-8. Energy of the detail signal at each scale (ED_j ; Eq. (9-1)) obtained via wavelet decomposition with respect to scale (j) for the lab-scale accelerometer signals in Fig. 9-5. Specifically, the accelerometer signals were obtained in lab-scale reactor as a function of time at h/H of 0.57, superficial liquid velocity (U_l) of 0.033 m/s, overall GAC concentration of 150 g/L, and permeate flux of 0 L/(m ² h). Each row represents each side of the reactor that the signals were collected at, while the left and right columns represent respectively the absence and presence of fluidized GAC.	224

- Figure 9-9. Energy of the detail signal at each scale (ED_j ; Eq. (9-1)) obtained via wavelet decomposition with respect to scale (j) for the pilot-scale accelerometer signals in Fig. 9-6. Specifically, the accelerometer signals were obtained in pilot-scale FS-AnFMBR (with a new membrane cassette and DI water) as a function of time at h/H of 0.57, inlet flow rate of $67 \text{ m}^3/\text{h}$, 50 kg of GAC particles, and permeate flux of $0 \text{ L}/(\text{m}^2 \text{ h})$. Each row represents each side of the reactor that the signals were collected at, while the left and right columns represent respectively the absence and presence of fluidized GAC.227
- Figure 9-10. Dimensionless height (h/H) versus the sum of the ED_1 , ED_2 and ED_3 values at different sides of the lab-scale reactor at a constant overall GAC concentration of 200 g/L and (a) $U_l = 0.024 \text{ m/s}$, (b) $U_l = 0.028 \text{ m/s}$, and (c) $U_l = 0.033 \text{ m/s}$230
- Figure 9-11. Dimensionless height (h/H) versus the sum of the ED_1 , ED_2 and ED_3 values at different sides of the lab-scale reactor at a constant U_l value of 0.033 m/s and overall GAC concentrations of (a) 100 g/L, (b) 150 g/L, and (c) 200 g/L in the lab-scale reactor.230
- Figure 9-12. Dimensionless height (h/H) versus the sum of the ED_1 , ED_2 and ED_3 values at different sides of the pilot-scale FS-AnFMBR with a new membrane cassette (Fig. 9-4b) and DI water. Each column represents a different inlet liquid flow rate (Q), while each row represents a different overall GAC mass.232
- Figure 9-13. Evolution of TMP and flux for the (a) FS-AnFMBR reactor with $Q = 42 \text{ m}^3/\text{h}$; and (b) HF-AnFMBR reactor with $Q = 30 \text{ m}^3/\text{h}$. For each reactor, the amount of GAC present was 150 kg and the targeted permeate flux was $5.5 \text{ L}/(\text{m}^2 \text{ h})$234
- Figure 9-14. Dimensionless height (h/H) versus the sum of the ED_1 , ED_2 and ED_3 values obtained at the west side of the pilot-scale AnFMBRs at a targeted permeate flux of $5.5 \text{ L}/(\text{m}^2 \text{ h})$ for each reactor: (a) One of the trends represent the data from the FS-AnFMBR with new membranes, DI water, $Q = 46 \text{ m}^3/\text{h}$ and 75 kg of GAC, while the other two represent the data obtained towards the end of the

campaign (i.e., at 291 days for the FS-AnFMBR and 350 days for the HF-AnFMBR; Fig. 9-13); (b) One trend represents the data from the FS-AnFMBR when defluidization occurred due to diminished inlet flow, while the other when fluidization was re-established when the targeted inlet flow rate was enabled; in both cases, the HF-AnFMBR has been operated for more about 350 days at $Q = 30 \text{ m}^3/\text{h}$ and 150 kg of GAC.....	235
Figure 9-15. Evolution of $E_{D1}+E_{D2}+E_{D3}$ values for the (a) FS-AnFMBR with new membranes, 75 kg of GAC, $Q = 46 \text{ m}^3/\text{h}$, and a DI water feed, and (b) HF-AnFMBR with 150 kg GAC, $Q = 30 \text{ m}^3/\text{h}$, and wastewater at about 350 days of operation. Both acceleration signals were collected at the west side of the reactors.	236

NOMENCLATURE

a	Acceleration signal measured by accelerometer (m/s^2).
A	Cross-sectional area of the reactor (m^2).
A_b	Area of bubbles per unit membrane area (mm^2/mm^2).
A_b/N_b	Mean bubble area per bubble (mm^2).
A_j	Lower-frequency approximations at scale j wavelet decomposition.
$A_{p,overall}$	Overall particle contact area per unit membrane area (mm^2/mm^2).
$C_{bentonite}/C_{GAC}$	The ratio of bentonite concentration to overall GAC concentration (-).
d_c	Depth of the cell (m).
d_s	Volume-based equivalent spherical diameters (mm).
dC/dt	The percent foulant coverage between the two images (%).
d_{cmin}	Diameter of the minimum circumscribed circle around the particle (mm).
D_j	Higher-frequency details at scale j of wavelet decomposition.
dN/dt	Rate of change of the number of deposited polystyrene particles (-).
d_p	Particle diameter (mm).
$d\text{TMP}/dt$	The rate of change of trans-membrane pressure (kPa/h).
E_{Dj}	Energy associated with each detail signal obtained via wavelet decomposition.
$E_{D1} + E_{D2} + E_{D3}$	Energy associated with detail signals D_1 to D_3 obtained via wavelet decomposition.

g	Gravitational acceleration (9.8 m/s ²).
h	Height assessed (m).
H	Total cell height (m).
h/H	Axial position of the vertically aligned membrane (-).
m_b	Bubble mass (kg).
N_b	Number of bubbles per unit membrane area (number/mm ²).
J_c	Membrane critical flux (L/(m ² h).
$J_{c,local}$	Local critical flux (L/(m ² h).
$J_{c,overall}$	Overall critical flux (L/(m ² h).
p	Particle momentum (kg/m/s).
$P(V)$	Probability of a particle having a particle velocity V (-).
$P(V_b)$	Probability of a bubble having a bubble velocity of V_b (-).
P_b	Bubble momentum (kg m ⁻¹ s ⁻¹).
ΔP_{bed}	Pressure drop across the solid-liquid fluidized bed (Pa).
p_i	Inlet pressure (kPa).
P_p	Power requirement per unit permeate (kWh/m ³).
PSD	Particle size distribution (-).
$P_{p,overall}$	Overall particle momentum per unit membrane area (kg/m/s).
P_r	Power requirement (kW).
Q_g	Gas flow rate (L/min).
Q_l	Liquid flow rate (L/min).
Q_p	Permeate flow rate (m ³ /s).
u_g	Superficial gas velocity (m/s).
U_l	Superficial liquid velocity (m/s).
U_{mf}	Minimum fluidization velocity (m/s).

$\dot{V}$	The volumetric flow rate (m ³ /s).
V_{arith}	Arithmetic means of particle velocity distribution (mm/s).
V_b	Bubble velocity (mm/s).
$V_{b,rms}$	Mean bubble velocity (mm/s).
V_{rms}	Root-mean-square means of particle velocity distribution (mm/s).
V_x	Particle velocities in the horizontal (x) direction (mm/s).
V_y	Particle velocities in the vertical (y) direction (mm/s).
V_p	Particle velocity (mm/s).
$V_{p,overall}$	Overall particle velocity (mm/s).
V_θ	Particle rotation speed (°/s).
$V_{\theta,rms}$	Mean rotation speed (°/s).
$V_{\theta,overall}$	Overall particle rotation velocity (°/s).
z, L	Fluidized bed height (m).

Greek symbols

α	Ratio of specific heat at constant pressure to that at constant volume (-).
σ	Standard deviation (-).
ρ_{air}	Air density (kg/m ³).
ρ_s	The sludge density in MBR (kg/m ³).
γ	Shear intensity (s ⁻¹).
μ	Liquid viscosity (0.001 Pa·s).
ε_{mf}	Bed voidage at minimum fluidization condition (-).
φ	Particle sphericity (-).
θ	A ratio representing the dominance of the gas flow rate (Q_g) with respect to the total flow rates (i.e., sum of Q_g and Q_l).

Chapter 1 Introduction

1.1 Background

Membrane fouling, which leads to a decrease in filtration efficiency and increase in energy costs to sustain productivity, is inevitable in membrane-based filtration processes. To mitigate membrane fouling, unsteady-state shear means have been identified as cost-effective [1-4], the underlying principle of which is in diminishing the hydraulic resistance imposed by the boundary layer and foulants on the membrane. Current attempts at mitigating fouling involve inducing tangential shear forces on the membrane surface, such as aeration [3, 5, 6], vibration or rotation [2, 4, 7, 8], pulsatile flows [4, 9-11], and particle fluidization [12, 13]. The focus of the current work is on particle fluidization, whose beneficial impact on membrane filtration was reported as early as in 1970 [14] and recently gained significant traction particularly for the larger millimeter-sized particles [13, 15-24]. Specifically for granular activated carbon (GAC) particles, besides the adsorption capacity, the fluidized GAC particles are known to mechanically scour the membrane fouling layer and improve the back transport of solutes by the movement of particles in the boundary layer [12]. While powder activated carbon (PAC) particles, which are an order-of-magnitude smaller than GAC, offer more surface area for more effective adsorption, GAC has been reported to be more effective in membrane fouling mitigation in the longer term [13].

The use of particle fluidization as a means of fouling mitigation has flourished specifically in anaerobic fluidized membrane bioreactors (AnFMBRs) in the past couple of years [13, 15-24]. In particular, the main limitation for the application of anaerobic wastewater treatment is its poor effluent quality, which falls short in meeting the rigorous standards, but can be circumvented by coupling with membrane bioreactors (MBRs) [25]. Unfortunately, the key obstacle in the implementation of MBRs is membrane fouling, which reduces the membrane permeability and increases the operation cost [26-29]. Kim et al. [13] proposed a two-stage anaerobic fluidized bed bioreactor for wastewater treatment, with GAC particles as the fluidized media, which gives high effluent water quality at a lower energy cost than gas sparging. Specifically, the first stage is an anaerobic fluidized-bed bioreactor (AnFBR), while the second stage is an anaerobic fluidized bed membrane bioreactor (AnFMBR). The fluidized GAC particles mitigate membrane fouling by playing both the roles of bio-carriers (i.e., adsorption and degradation of organic foulants) and scouring agents (i.e., mechanically abrade the membrane surface). Since then, investigations revolving around the AnFMBR have aimed to optimize various process parameters, such as temperature [18, 20], type of pre-treatment [15, 16, 24], reactor configuration [21], and fluidized media [30]. To date, the hydrodynamics of the solid-liquid fluidization of the GAC particles is not understood, although the physical behavior of the liquid and solid phases is key to the fouling mitigation phenomenon. Hence, the current study aims at bridging the gap in the knowledge base of the dynamics of the solid particles in effecting the scouring mechanism on membranes to mitigate fouling.

1.2 Objectives and scope

The main objectives of this thesis are:

- To correlate the dynamics of fluidized GAC behavior with fouling mitigation efficacy in solid-liquid fluidization membrane filtration systems, and optimize the extent of fouling mitigation and energy requirement.
- To characterize the hydrodynamics of fluidized media locally (namely, particle velocity, particle rotation velocity, particle contact area and particle momentum), and correlate the hydrodynamics with the effectiveness of membrane fouling mitigation.
- To determine the critical flux values of a fluidized bed membrane filtration system using a novel adaptation of DOTM for a vertical membrane module, and assess the impact of various operational parameters include GAC particle diameter (d_p), overall GAC concentration, superficial liquid velocity (U_l) and thereby power requirement (P_r) on the critical flux (J_c) at various heights of the membrane.
- To further look into the tradeoff based on energy requirement (P_r) and scouring efficiency considerations in optimizing the GAC particle diameter, overall GAC concentration and fluidized particle sphericity.

1.3 Organization of the thesis

This thesis comprises ten chapters:

- (1) Chapter 1 gave an overview of current research background and specific objectives of this study.
- (2) Chapter 2 provided a comprehensive literature review, including the membrane fouling in MBRs, the unsteady-state shear means to mitigate membrane fouling, and the latest findings of anaerobic fluidized bed membrane bioreactors (AnFMBR).
- (3) Chapter 3 investigated the scouring efficacy of three GAC particles with different particle diameters (namely, 1.20 mm, 1.85 mm, and 2.18 mm). An accelerometer was successfully used to reveal the dynamics of GAC particles with different particle diameters. The impact of particle diameter on the extent of fouling mitigation was studied.
- (4) Chapter 4 dedicated to understanding the hydrodynamics of the fluidized GAC particles, and correspondingly the relationship between the particle behavioural characteristics and the extent of membrane-fouling mitigation.
- (5) Chapter 5 aimed at understanding the improvement of fine particle critical flux (J_c) in the presence of fluidized GAC via the Direct Observation Through the Membrane (DOTM) technique.

- (6) Chapter 6 targeted at understanding of the relationship between the bubble characteristics and critical flux. A comparison between the types of two-phase flow systems employed for fouling mitigation (namely, bubbling and fluidized GAC particles) was carried out.
- (7) Chapter 7 studied the impact of overall GAC concentrations on the extent of fouling mitigation. Monodisperse millimeter-sized GAC (i.e., $d_p = 1.55$ mm) was used as the fluidized media and the particulate foulants were micrometer-sized polystyrene and bentonite particles.
- (8) Chapter 8 expected to be valuable in the optimization of the properties of the fluidized media towards more energy-efficient membrane fouling mitigation. The effect of the fluidized particle sphericity (ϕ) on the extent of fouling mitigation was investigated.
- (9) Chapter 9 aimed at evaluating the feasibility of using a non-invasive accelerometer for a practical pilot-scale AnFMBR, in order to provide a means for real-time monitoring of such processes.
- (10) Chapter 10 summarized the key conclusions and major findings of this thesis study. Recommendations for future works targeted at the optimal fluidization scenario were proposed.

Chapter 2 Literature review

2.1 Membrane fouling in MBRs

Due to the significant advantages over the conventional activated sludge processes, membrane bioreactors (MBRs) have become increasingly popular for wastewater treatment and reuse. However, as with all membrane-based filtration processes, membrane fouling remains a major hurdle that leads to reduced permeability and increased operation cost [31, 32]. Membrane fouling is undesirable because it worsens permeate flux, disrupts membrane selectivity or permeate quality, and increases membrane maintenance and replacement costs. Hence, there has been a large amount of investigations on membrane fouling over the years. Most of studies have been focused on the knowledge of foulant characteristics, membrane fouling mechanism, and fouling control strategies [33-42], while other investigations were focus on indirect (i.e., pretreatment of feed flow [43, 44], optimization of process parameters [40], novel membranes materials or membrane surface modification with anti-fouling characteristics [45-47]), or direct fouling control processes (i.e., chemical- [48-50], physical- [51, 52], biological-/biochemical-, or electrical-based [53] cleaning processes).

Membrane fouling is a complicated issue, that has not been clearly understood yet. The form of membrane fouling in MBRs can be caused by concentration polarization, external fouling and internal fouling [32, 54, 55]. Concentration polarization (CP) is defined as the phenomenon of the concentration of the solute or particle in a thin liquid layer near the membrane

surface is higher than that in the bulk [56-58]. It not only increases the risk of membrane fouling but also worsens the permeability of the membrane, as the resistance of liquid flow is enhanced. The alleviation of CP can be achieved by the increase of liquid velocity in a cross-flow membrane filtration system. External fouling, which is easier to be removed than internal fouling, is caused by the deposition of particulates, colloids, and macromolecules on membrane surfaces. This type of fouling can be also classified into two kinds of fouling layers, namely, cake layer and gel layer [59, 60]. The cause of internal fouling is by the adsorption of fouling onto membrane or the pore blocking (i.e., deposition of foulants in the internal membrane structure).

Owing to the attachment strength, membrane fouling can be simply classified into the removable and irremovable fouling [32]. As shown in **Fig. 2-1**, the removable fouling, which is formed by loosely attached foulants is easy to be removed by physical cleaning such as backwashing. However, due to the strong attachment with foulants or pore blocking, it is very hard for irremovable fouling to be eliminated by physical cleaning, and the chemical cleaning (e.g., sodium hypochlorite) is required. Commonly, we can identify the removable fouling as the formation of cake layer, and the irremovable fouling as the formation of pore blocking. The influence of membrane fouling can be attributed to many factors, such as the components in the reactor including extracellular polymeric substances (EPS) or soluble organic substances [61], sludge characteristics [62], biomass concentration [62, 63], sludge rheology [63], and inorganic precipitates [54, 64]. These in turn can be influenced by many parameters in a membrane bioreactor. As shown in **Fig. 2-2**, in an anaerobic

membrane bioreactor (AnMBR), membrane fouling is governed by the relationships of variable factors, include the hydrodynamic conditions, reactor operating conditions, membrane configuration and types, biological process, permeate performance targets and the chemical treatment process.

Figure 2-1. Formation and removal of removable and irreversible fouling in MBRs. Adapted from [32].

Figure 2-2. The parameters and relationships that influence membrane fouling performance in an anaerobic membrane bioreactor. Adapted from [65].

Overall, membrane fouling hassles the permeability of the membrane, hence increases the area of membranes required per volume of reactor or the driving forces to rescue permeability targets, both inevitably increase the capital cost. The operation below the critical flux is preferred for sustainable MBR operation [66]. However, fouling is an inevitable consequence of filtration process, which cannot be mitigated only through optimum membrane operating conditions (e.g., permeate flux lower than critical flux) without the incorporation of fouling control process (e.g., gas sparging, particle scouring, adsorbent, electric field, etc). Despite the advance understanding on foulant characteristics and specific-foulant fouling mechanism, optimization of fouling control processes is hampered by the lack of understanding on: (i) the complex foulant interactions with its environment (i.e., membrane, other foulants, microbes,

fouling mitigation agents), and (ii) foulant's spatial and temporal developments which affect influent rheology during long-term filtration [67]. Recently-developed fouling control processes (e.g., *in-situ* electrocoagulation or electrophoresis, quorum quenching, EPS and SMP degradation by enzyme or microbes, anti-fouling nanomaterial-based membrane, etc) emphasize fouling prevention, rather than fouling reduction mechanism of conventional fouling control processes (e.g., membrane relaxation, backwashing, aeration). Novel anti-fouling membrane, and biological-/biochemical- and electrical-based membrane cleaning processes require further implementation in pilot- and industrial-scale, and long-term filtration tests. On the other hand, recent research on the physical-based membrane cleaning processes (e.g., pneumatic- and hydraulic-based, particle fluidization) are focus on the optimization of fouling control with respect to energy usage through hydrodynamic studies, both experimentally [68, 69] and using CFD (Computational Fluid Dynamics) simulation [70-73].

2.2 Unsteady-state shear to mitigate membrane fouling

As promising means of energy-efficient fouling mitigation, unsteady-state shear methods have been identified as techniques to employ, without which the recovery of permeability will inevitably worsen the fouling problem [1]. Unsteady-state shear methods include bubbling [5, 6, 74], vibration [7, 75, 76], pulsatile flow [9, 11], and more recently particle fluidization [13].

2.2.1 Fouling mitigation by gas sparging

The beneficial impact of bubbles on membrane filtration processes has been reported as early as in 1989 by Imasaka et al. [77], who injected the produced methane of an anaerobic digester into the tubular ceramic membrane modules, and found that membrane fouling was reduced at a reasonable energy cost. Since then, significant research on the use of bubbly flow particularly on aerobic membrane bioreactors has flourished [6, 78-85]. The introduction of bubbles has been reported to improve the permeate flux by 43% [86] and the critical flux [87] by up to 1.7 times [81]. Unsteady-state shear whereby bubbling (i.e., gas sparging) is found easily to mitigate the cake layer fouling, but to be difficult in mitigating the irreversible fouling (i.e., pore blocking) [1, 88, 89].

Previous studies have indicated that the presence of bubbles conferred shear stress whereby the creation of wakes and secondary flows, thus to disrupt the polarization layer and mitigate membrane fouling [90-94]. Besides, some of the bubbles can penetrate to the concentration polarization layer, thereby disturbing it and enhancing the mass transfer coefficient [89]. Moreover, it was also found that the presence of bubbles enhanced the back-wash efficiency [95, 96]. The shear intensity (γ) conferred by bubbles in MBRs can be expressed as [88, 97]:

$$\gamma = \left(\frac{\rho_s g Q_g}{\mu} \right)^{1/2} \quad (2-1)$$

Where ρ_s is the sludge density in MBR, g is the constant of gravity, Q_g is the bubble flowrate and μ is the apparent viscosity.

Bubble flow patterns (shown in **Fig. 2-3**), which are inevitably tied to the effectiveness of fouling mitigation have been categorized into the following according to gas flow rates [5, 6, 85, 98]: (i) bubble flow, whereby small discrete bubbles are uniformly distributed in the liquid phase; (ii) plug flow, whereby gas phase occurs in intermittent plugs or pistons with defined phase boundaries, sometimes designated as Taylor bubbles; (iii) slug flow, whereby each bubble size spans more than 75% of the width of the filtration module; (iv) churn flow, whereby the slugs are narrower and more distorted, and the flow is more chaotic, frothy and disordered; (v) annular flow, whereby the core and annulus are occupied by the gas and liquid phases, respectively. To optimize the fouling mitigation by bubbles, the influence of gas flow rates [78, 97, 99-101], bubble size and frequency [82, 102-105], and bubble shapes and orientations [93] have been investigated. Shear distribution in a bubble flow channel is shown in **Fig. 2-4**. The thin liquid film between the bubble and the membrane has the most shear strength, then the associated wake and vortices regions.

Figure 2-3. Bubble flow pattern in tubular channels. Adapted from [6].

Figure 2-4. Wall shear stress distribution around a gas slug rising in stationary liquid in a vertical tubular channel, results were measured using the electrochemical shear method. Adapted from [106].

The extent of fouling mitigation whereby bubbling can be influenced by many factors, including the gas and liquid velocities, bubble size, bubble shape, membrane configuration, and the motion of bubbles. Typically, larger bubbles and higher gas flowrates were more beneficial for fouling mitigation [1, 81, 89, 107, 108]. **Fig. 2-5** shows a clear increasing trend as gas/liquid ratio (θ) increased, particularly for the microfiltration. As seen from **Eq. 2-1**, higher gas flowrates are associated with higher shear stress, which would benefit the fouling mitigation extent. It was suggested by Wibisono et al. [6] that higher pore size of MF membranes was more beneficial from the bubble shear, rather than UF membranes, and cake formed fouling layer was more readily to be removed than solutes. Comparing the fouling mitigation extents with different membrane configurations, hollow fiber membrane modules can achieve higher flux

enhancement than flat-sheet membrane modules, as seen the results summarized in **Fig. 2-6**. This is due to the smaller hydraulic diameter for hollow fiber membranes, thereby higher wall shear stress achieved. Although the gas velocity (U_G) has positive correlation with fouling mitigation extents for both flat-sheet and hollow fiber membranes, the liquid velocity (U_L) is less effective for flat-sheet membrane modules (**Fig. 2-6a**). This highlights the importance of bubbling shear rather than liquid in gas-liquid flow systems.

Figure 2-5. Flux enhancement versus gas/liquid ratio with different feed types. Adapted from [6].

Figure 2-6. Three-dimensional (3D) plot correlating gas (U_G) and liquid superficial velocities (U_L) with mean flux enhancement in a vertically (a) flat-sheet membrane module and (b) hollow fiber membrane module. Adapted from [6].

2.2.2 Fouling mitigation by vibrating systems

The concept of vibrating systems was first proposed by Culkin and Armando [109] in 1992, and named as Vibratory Shear-Enhanced Process (VSEP). In this VSEP system, a stack of circular flat-sheet membranes separated by gaskets is fixed on a shaft that is oscillated at frequency of 60 Hz. **Fig. 2-7** depicts the schematic of a flat-sheet membrane vibrating in a VSEP system. This type of membrane separator allows a change of flow regime from laminar to turbulence, and provides an unsteady-state shear of $150,000 \text{ s}^{-1}$ on membrane surface. Besides, secondary flows and vortices are generated to enhance the back diffusions of solutes and other foulants, and the mixing of concentration polarization (CP), thereby mitigating membrane fouling. Since, the VSEP systems have been successfully employed in MF [110], UF [110], NF [111] and RO [112, 113] setups.

Figure 2-7. Schematic of membrane vibration in a VSEP system. Adapted from [114].

The benefits of vibrating membrane modules have been claimed to enhance the membrane critical flux [115, 116] and generate turbulence [117] on membrane surfaces. The fouling mitigation was found to be more effective with the onset of turbulence generated when the oscillation amplitude and frequency achieved a critical value [118]. The shear exerts on the membrane holds a key mechanism in vibrating systems. For a sinusoidal surface vibration having a frequency of f and angular frequency of ω , the shear rate can be calculated from [119, 120]:

$$\gamma(r, t) = 2r\omega(\pi f)^{1.5}\nu^{-0.5}[\cos(2\pi ft) - \sin(2\pi ft)] \quad (2-2)$$

where ν is the fluid kinematic viscosity, and r is the distance from the axis in **Fig. 2-7**.

Recently, the membrane vibration systems are advanced whereby vibrating the submerged hollow fiber membranes. The schematic of experimental setup is shown in **Fig. 2-8**. Different from the flat-sheet membrane modules, which can only vibrate the membrane axially, hollow fiber membranes can vibrate transversely or longitudinally. Li et al.[121] compared the transverse and longitudinal vibrations using same membrane module, whereby changing the orientation of membrane module. Results showed that transverse rather than longitudinal vibration had more benefits on fouling mitigation, even at low vibration frequency of 1 Hz. This contrasts with the results by Low et al.[122] that the longitudinal vibration was more beneficial for fouling mitigation due to the unequalled shear generated by transverse vibration, but is in consistency with

the results by Kola et al. [123, 124] that the transverse vibrations were more effective to enhance the flux due to the generation of secondary flows.

Figure 2-8. Schematic of submerged hollow fiber membrane modules by vibrations (a) transversely and (b) longitudinally. (a) adapted from [121] and (b) adapted from [116].

In conclusion, the mitigation of membrane fouling via vibration means can be attributed by the aspects of the back transport of solutes and foulants by pulsation, unsteady state shear by the change from laminar to turbulent flow regime, and the associated secondary flows and vortices. The optimization of the vibration systems can be benefited from the combined effects of shear rates and the associated secondary flows. Higher vibration frequency and amplitude generally give more fouling mitigation effectiveness. Vibrating the hollow fiber membranes transversely is generally more effective than longitudinally. In consistency with the fouling mitigation means via gas sparging, the vibrating systems have more effective in the mitigation of cake layer fouling rather than the irremovable fouling (i.e., pore blocking).

2.2.3 Fouling mitigation by pulsatile flow

Pulsation flow is another promising means to provide the unsteady state shear near the membrane, whereby altering the fluid dynamics. As seen in **Fig. 2-9**, the velocity profile is shifted from parabolic to bimodal shape that in turn causing the maximum velocity moved from the centreline to near the walls. A pulsatile pressure waveform is generated in this pulsation flow [1, 125] that can cause a negative TMP resulting in the back-flushing. Thereby the key mechanism for pulsation flow to mitigate membrane fouling can be attributed from two aspects: (i) the unsteady state shear on membrane surface, hence to decrease the concentration polarization layer, and (ii) the back flushing, which benefits from the pulsatile pressure wave to enhance the back transport of the foulants. It was even reported by Rodgers and Sparks [126] that the oscillated back-flushing is more effective than the shear in mitigating the membrane fouling.

Figure 2-9. Velocity profiles with and without flow pulsation. Adapted from [127].

Investigations based on the flow pulsation have been suggested to increase the permeability of the membrane in MF [125, 128], UF [11, 126], and RO [129] systems. Moreover, the use of pulsatile flow was claimed to prevent the development of cake fouling layer, reduce the filtration resistance, and increase the salt rejection [1, 125, 129]. When stable fouling layer formed at steady pressure period, the minimum pressure can destabilize the cake and reverse the permeate acted as back wash. This drives the deposits and foulants near the membrane surface back to the bulk flow, and alleviate the membrane fouling. The optimization the pulsation flow means includes the parameters of the range of pressure (i.e., the minimum and maximum pressure values), and the duration and frequency of the pulsatile flow.

However, it is difficult to find the available model predicting the shear rate under the pulsatile flow. The main problem for the pulsation flow means is

that the pulsatile pressure is ineffective in changing the streamlines nearer the membrane surface. The concentration gradient still occurs near the membrane. Besides, the energy consumption is another issue need to be concerned in this fouling control method. The induce of flow pulsations inevitably enhanced the energy consumption and also the maintenance cost as the entire flow rather the near membrane need to be accelerated.

2.2.4 Fouling mitigation by particle scouring

Albeit the gas sparging has been adopted as the most popular means to mitigate membrane fouling in MBRs, the inducing of bubbles/biogas into the reactor inevitably increased the energy consumption. The use of scouring media to mechanically exert the shear on membrane surface has been reported as a promising means to control the membrane fouling at relatively lower energy consumption [13, 51, 130, 131]. This scouring agent can be combined with both of the aerobic and anaerobic MBRs, whereby fluidizing the media via bubbles or liquid recirculation. The combination of scouring media with aerobic MBR was reported to reduce the gas required by higher than 50% at the same fouling mitigation effectiveness [132-134], which significantly saved the energy cost. It should be worth noting that the aeration in MBRs requires the energy around 40% of the total energy cost for the operation the systems [32, 51, 135].

More recently, a new approach of anaerobic fluidized bed membrane bioreactor (AnFMBR) was proposed by Kim et al. [13] whereby fluidizing the granular activated carbon (GAC) in an anaerobic MBR. In which, membrane fouling was controlled to quite a low level with energy consumption much less than gas sparging. **Fig. 2-10** depicts the energy consumption of fouling mitigation

by different unsteady state shear means using typical operation conditions. As noted above, the membrane filtration systems combined with scouring media need less energy requirement compared with others, while the combination of particle scouring with anaerobic MBR consumed the least.

Figure 2-10. Energy consumption for different membrane filtration systems. Adapted from [51].

2.2.4.1 Gas-liquid-solid fluidization

The use of gas-liquid-solid fluidization means to mitigate membrane fouling is typically implemented in aerobic MBRs, whereby scouring media are added in the conventional MBR and fluidized by the aeration. Unless biogas can be induced, this fouling mitigation method could be employed in anaerobic MBR. It has been reported that presence of additional fluidized particles could control the membrane fouling at the relatively low level with energy consumption much lower than that of bubbling or the bulk recirculation alone [136-138]. For

instance, Siembida et al. [139] employed the scouring particles in MBR and found that the critical flux was enhanced than without the particles, this allowed the permeate flux to increase by 20% than aeration only, and fouling was controlled to an expected level without any cleanings of the membrane in a long term operation (i.e., more than one year). Fu et al. [140] found that the use of particle scouring in aerobic MBR can extend the operation time 8 times than aeration only. Benefit from the mechanical scouring conferred by fluidized particles, membrane fouling mitigation is enhanced, thereby the corresponded gas requirement to control the fouling is lower. This significantly reduced the energy consumption, as the portion of aeration is the main cost of an aerobic MBR operation [32, 51, 135]. For instance, Kurita et al. [133, 141] reported that the aeration rate could be reduced more than 50% combined with fluidized media.

Typically, these fluidized particles in aerobic MBRs can be the plastic media with the density near to water, like the polyether sulphone (PET), polyethylene (PE), and polypropylene (PP), etc. or larger densities, like the GAC, silica and glass beads. The selection of scouring particles can be the non-porous or porous materials and act as the biocarriers and scouring agent. The presence of scouring particles controls membrane fouling whereby (i) provide the shear rate on membrane surface directly, hence to remove the cake fouling; and (ii) generate the turbulence and enhance the back-transport of solutes and foulants.

Huang et al. [138] systematically optimized the dosage of carriers in MBR and found that the additional scouring particles have two effects. One is the expectedly positive effect on membrane fouling removal whereby particle scouring. As seen from **Fig. 2-11**, cake layer fouling is mitigated obviously with

the presence of scouring particles. Another is the unexpectedly negative effect on the breakage of sludge flocs, which potentially enhanced the membrane fouling. In this case, the increase of carrier's dosage gave the double effects of them. Thereby, there existed an effective carrier dose range for mitigating membrane fouling, and this was tied to the sludge concentrations.

Figure 2-11. Photos of membrane modules after the operations of: (A) without carriers and (B) 1% additional scouring media. Adapted from [138]

It can be concluded that this gas-liquid-solid fluidization means is more effective than the gas sparging or liquid flow alone in MBRs, as the additional scouring media can cross the laminar flow layer and exert the shear directly on membrane surfaces. Membrane fouling is controlled whereby the mechanical scouring conferred by fluidized particles, and the turbulence generated to enhance the back-transport of foulants. However, the shear is difficult to be predicted in this process, and the optimization is tied to the amount of gas injection and dosage of fluidized media. Besides, characteristics of the sludge flocs may be changed by the scouring effect, which is another factor need to be considered for the fouling mitigation effectiveness.

2.2.4.2 Solid-liquid fluidization

As mentioned previously (**Fig. 2-10**), the use of fluidized particles with the recirculation of bulk liquid can achieve the least energy consumption in membrane fouling mitigation. Thereby, solid-liquid fluidization means of fouling mitigation is acknowledged towards the most energy positive method over the past decades [13, 23, 51, 142]. The most typically used is the combination of fluidized bed with anaerobic MBR, whereby additional particles (i.e., GAC particles) are fluidized by the recirculation. The mechanical scouring conferred by fluidized media is the key for membrane fouling mitigation. This allows the anaerobic reactor could fast process the wastewaters meeting the rigorous effluent requirements. Moreover, the produced biogas (i.e., methane) can be further employed for energy consumption, hence to reduce the energy cost in a very low level comparing with other fouling control methods.

Minimum fluidization velocity (U_{mf}) is an important parameter in solid-liquid fluidization, and defined at which the upward fluid drag balances the bed weight [143-146]. The recirculation liquid velocity must be higher than U_{mf} , thus to make sure the scouring particles are fluidized in the reactor. It can be experimentally measured via the pressure drop method [147] or calculated based on the empirical Ergun equation [148]:

$$\frac{\Delta P_{bed}}{\Delta z} = \frac{150U_{mf}\mu(1-\varepsilon_{mf})^2}{\varphi^2 d_p^2 \varepsilon_{mf}^3} + \frac{1.75U_{mf}^2 \rho(1-\varepsilon_{mf})}{\varphi d_p \varepsilon_{mf}^3} \quad (2-3)$$

Where ΔP_{bed} is the pressure drop across the bed, Δz is the height of the bed, μ is liquid viscosity (0.001 Pa·s), ε_{mf} is the bed voidage at U_{mf} , φ is the particle

sphericity, and d_p is the equivalent of the particle diameter. The energy consumption (P_r) in a solid-liquid fluidized bed can be calculated from [149]:

$$P_r = \Delta P_{bed} U_l A \quad (2-4)$$

where A is the cross-sectional area of the reactor.

There's no standard model to calculate the shear rate in a solid-liquid fluidization bed. Zamani et al. [1] predicted a model based on the empirical equation of the shear in two phase flow system, and expressed as:

$$\gamma = \left(\frac{U_l(1-\varepsilon)(\rho_p - \rho)g}{\mu} \right)^{1/2} \quad (2-5)$$

where U_l is the superficial liquid velocity, ε is bed voidage, and $(\rho_p - \rho)$ is the density difference between fluidized media and the liquid. Therefore, the shear in a solid-liquid fluidization is positively correlated with superficial liquid velocity and particle concentration, both tied to the power requirement. Optimization of fouling mitigation means via solid-liquid fluidization is the tradeoffs between energy consumption and fouling mitigation effectiveness.

2.3 Anaerobic fluidized bed membrane bioreactor

As the target of this thesis is to understand the fouling mitigation by solid-liquid fluidization, which is based on the process of AnFMBR, an overview brief of AnFMBR and the relative investigations since proposed will be given here.

Anaerobic wastewater treatment has several benefits over the conventional aerobic treatment process. For instance, the anaerobic process has the potential to eliminate the air injection, which typically cost half of the total

energy consumption [32, 51], reduce the solids (i.e., sludge wasting), and produce the usable fuel (i.e., methane and hydrogen) from the organic carbon [150, 151]. However, the shortcomings of anaerobic process are apparent, including the large footprint required, odor issues, temperature requirement, and poor effluent quality, while these could be solved by the combination of MBR. Membrane fouling is then becoming another critical challenge of this technology. Anaerobic fluidized membrane bioreactor (AnFMBR) is acknowledged having the most potential to mitigate membrane fouling at very low energy cost [13, 152].

The process of AnFMBR was first proposed by Kim et al. [153], which is composed of a first stage of anaerobic fluidized bed reactor (AnFBR), and a second stage of anaerobic fluidized bed membrane bioreactor (AnFMBR). The schematic of the two-staged AnFMBR is shown in **Fig. 2-12**. Granular activated carbon (GAC) was used as fluidized media. The function of fluidized GAC was demonstrated to adsorb the organic matters, act as biocarriers, and provide the mechanical scouring on membrane surface. However, the adsorption property can be exhausted soon at the initial days, and the scouring of fluidized GAC was the key to control membrane fouling. Total energy consumption was reported 0.058 kWh/m³ for this process, which is much less than gas sparging. Since, studies targeted at improving the operation of such anaerobic fluidized bed membrane bioreactors (AnFMBR) have focused on the design [15, 24, 154, 155], operating conditions [156, 157], fluidized media [14, 131, 158, 159], operating temperature [15, 156, 160], treatment of different types of wastewater [15, 19, 154, 161, 162], removal of trace organics [163, 164], GAC size and packing

amount [131, 157, 165-167], membrane type [168] and the integrity of the membrane [157, 165, 169].

Figure 2-12. Schematic of two staged anaerobic fluidized bed membrane bioreactor (AnFMBR). Adapted from [153].

2.3.1 Reactor design and membrane configurations

The design of the reactor and membrane configurations has significantly impact on the hydrodynamics of fluidized media, which tied to the fouling mitigation effectiveness. The shape of AnFMBR reactors can be the rectangular or cylinder one. The distributor employed in AnFMBRs is usually designed with a number of pairs of nozzles are directed downwards in an inverted-V configuration and spread evenly along a central pipe traversing the reactor bottom, as shown in **Fig. 2-13**.

Several efforts have been put to test the feasibility of a single staged AnFMBR instead of two-staged, hence to further reduce the system cost. **Table 2-1** showed the performances achieved and energy consumptions using a single staged or two-staged AnFMBR. A single stage AnFMBR has the ability to achieve the high removal efficiency as two-staged AnFMBR, while the energy consumption is lower. Wu et al. [154] systematically compared the performance of a single-stage with two-stage anaerobic fluidized bed bioreactors treating municipal wastewater and found that more than 97% of total COD and almost 100% of suspended solids could be removed in these two systems. Results indicated that the first-stage AnFBR was unnecessary to reduce the costs treating municipal wastewater.

Figure 2-13. Structure of the distributor used in rectangular AnFMBRs.

Table 2-1. The removal efficiency and energy requirement of single staged and two staged AnFMBR systems treating wastewater.

Filtration system	Feed	Removal efficiency	Energy consumption	Reference
Two-staged AnFMBR	Municipal wastewater	COD 95%; pharmaceuticals>90%	Not measured	[163]
Two-staged AnFMBR	Domestic wastewater	COD 84-91%; BOD ₅ 92-94%;	0.048 -0.053 kW h/m ³	[16]
Two-staged AnFMBR	Domestic wastewater	COD 90%-94%; BOD ₅ 90%-98%	0.227 kW h/m ³	[170]
Single stage AnFMBR	Domestic wastewater	COD 89%; BOD ₅ 94%	More than 0.049 kW h/m ³	[18]
Single stage AnFMBR	Municipal wastewater	COD 84%; BOD ₅ 92%	0.047 kW h/m ³	[19]
Single stage AnFMBR	Synthetic wastewater	sCOD 61%-93%; TSS 100%	0.0387 kW h/m ³	[168]

Membrane modules of AnFMBRs are typically the submerged flat sheet and hollow fiber membranes [13, 19-21, 24, 130-132, 152, 154, 156, 157, 160-165, 171-176]. Recently, Düppenbecker et al. [142, 177, 178] tested the performance of AnFMBR using tubular membranes, whereby glass beads with particle diameter of 1.5 mm were fluidized inside a tubular ceramic membrane. Due to the directly mechanical shear by fluidized media, there may cause membrane integrity issues for plastic membranes in a typical AnFMBR reactor [165, 169]. Thereby, a more strength of flat-tubular ceramic membrane was first

tested by Aslam et al. [168] in a single stage AnFMBR that achieved over one year operation. Albeit the flat-sheet and hollow fiber membrane modules can expectedly achieve the steady operation of AnFMBRs with high effluent quality at low energy consumption, it is possible for hollow fiber membranes held some looseness, hence to generate a forth and back movement from the contact with fluidized particles. This type of design may further improve the fouling mitigation effectiveness in comparison with flat-sheet membranes. However, the feasibility of hollow fiber looseness in a AnFMBR has not been studied yet. It was indicated by Wu et al. [157] a spacing distance between the fibres was better relative to that without spacing. Design of hollow fiber modules (i.e., packing density, spacing, looseness, and length) has non-negligible effect on fouling mitigation in AnFMBRs.

2.3.2 Effect of fluidized media

As the key of fouling mitigation by AnFMBR is the mechanical scouring conferred by fluidized media, the effects of fluidized media seems significantly important. Parameters of the media include the particle size, density, concentration, shape and materials, all of which are tied to the hydrodynamic characteristics. Larger size, and higher concentration generally give higher fouling mitigation effectiveness, however, at the cost of energy consumption. Tradeoffs involved between fouling mitigation and energy requirement are necessary to be considered in the optimization of fluidized media effect.

In order to understand the relationship between membrane fouling and the packing amount of GAC, Kim et al. [13] applied three different GAC amount of 225, 300 and 450 g in a lab AnFMBR reactor to observe the fouling rate, and

found that the higher GAC dosage resulted in better fouling control. Wu et al. [157] found that higher GAC amount and larger size are more effective in membrane fouling mitigation in a hollow fiber AnFMBR system. Aslam et al. [131] reported that an increase of GAC dosage up to 50% (v/v) could give a greater fouling reduction, but the energy requirement was higher. However, a further increase to 70% (v/v) did not provide any beneficial effect on fouling mitigation, due to the increasing collisions between fluidized media which hinder the movement. Huang et al. [138] demonstrated that the performance of fluidized media should be dependent upon bulk characteristics in the aerobic fluidized bed membrane bioreactor. An increase of fluidized media concentration can positively provide the mechanical scouring on membrane surface, while adversely break the sludge flocs into micro and even nano-scaled colloidal materials causing severe fouling [165]. For AnFMBR, a systematic understanding of fluidized media concentration is still lacked, particularly with the multi effects of the feed concentration, particle size and energy consumption.

Regarding fluidized media materials, the alternatives like the silica, polyethylene terephthalate (PET) and glass beads were employed and all of them achieved the expected fouling reduction [131, 132, 158, 177]. Interestingly, although GAC is presumably superior due to its adsorption of organics and ability to function as a biocarrier in addition to the mechanical scouring capacity [51], the use of fluidized glass beads, which were larger and denser than GAC, has been reported to reduce membrane fouling by an impressive 95% in a tubular ceramic membrane [158], and non-porous fluidized media such PET beads, which were larger and less dense than GAC, was reported to mitigate membrane

fouling better than fluidized GAC [131]. The use of other biocarriers are expected in AnFMBR setups.

In relationship with media shape (i.e., sphericity), it has not been identified yet in AnFMBRs. In fact, there are various shapes of fluidized media especially in aerobic membrane bioreactors with lesser density than GAC [138, 179-187] should be considered for AnFMBRs. For instance, the cylindrical shape of fluidized particles was reported able to promote the contact between scouring particles and membrane surface [138].

2.3.3 Effect of temperature

Investigations about the effect of temperature were limited in AnFMBRs, although it has been reported in MBR that temperature higher than 35 °C and lower than 25 °C could affect the fouling intensity, since the scouring efficiency [160]. Shin et al. [20] operated a pilot-scale anaerobic fluidized membrane bioreactor for 485 d without chemical cleaning at seasonal temperatures of 9-30 °C treating with domestic wastewater. The average COD and BOD values in effluent were reported never higher than 23 mg/L and 9 mg/L, respectively. Yoo et al. [156] tested the temperature effect on organic removal in a lab scale AnFMBR by decreasing the temperate from 25 to 10 °C. Results indicated that no noticeable negative effects of low temperature on organic removal were found, although the fouling rate (i.e., TMP) increased. Thereby, the process of AnFMBR has a potential to treat the wastewaters seasonally.

2.3.4 Membrane integrity

Although the mechanical scouring conferred by fluidized GAC benefitted fouling mitigation, it negatively impacted membrane integrity [165, 169], especially for plastic membranes. In lab-scale AnFMBR studies, GAC-induced membrane integrity loss was negligible [157, 165]. However, Shin et al. [169] observed some damage on the used PVDF hollow fiber membrane after 2 years operation in a pilot-scale AnFMBR, which operated continuously with a permeate flux of 6.1–7.5 L/(m² h) and superficial liquid velocity of 75 m/h (i.e., 0.02 m/s) without chemical cleaning. **Fig. 2-14** shows the examples of membrane surface tests with and without GAC fluidization measured by Wu et al. [157] in a lab-scale AnFMBR. **Fig. 2-15** comparatively gives the examples of membrane surface damage from pilot-scale AnFMBR by fluidized GAC after 2 years operation. Another study by Shin et al. [172] developed a procedure to evaluate the long-term integrity of membranes for selecting membranes and reactor operating conditions to better ensure long-term operating performance of the systems when scouring media is used for fouling control.

Figure 2-14. SEM and AFM images of membrane surfaces in a bench scale AnFMBR setup. (a) and (c) with GAC fluidization; (b) and (d) without GAC fluidization. Adapted from [157].

Figure 2-15. Membrane surface damage with exposed braid near the bottom of the pilot AnFMBR after 2 years operation. Adapted from [169].

Larger GAC was found to severely affected membrane structural integrity, and membrane damage was much less with the smaller GAC particles than with the larger ones [172]. GAC size used in AnFMBR studies usually ranged from 1 mm to 3 mm, below which GAC particles may act as foulant causing negative effects [130, 157] (clearly seen in **Fig. 2-16**), and above which membrane damages caused [51, 161]. To hurdle this, Aslam et al. [168] first tested the performance of ceramic membrane (i.e., made of aluminum dioxide) in a lab scale AnFMBR operating for 395 days. The permeate flux was achieved up to 17 L/(m² h) with good effluent quality, which may tackle the membrane integrity issue by fluidized GAC particles.

Figure 2-16. SEM images of membrane surface using small GAC (50% v/v) as scouring media. The feed is 300 mg/L sodium alginate solution. Adapted from [188].

Although there are some less denser particles, like PET beads, could can be used in AnFMBR, with lesser membrane integrity affected, the hydrodynamics of these particles (namely, particle shear velocity, particle concentration, and particle momentum) are not clear yet, which directly tied to the fouling mitigation effectiveness. More denser particles, like glass beads were found to severely damage even the ceramic membrane by abrasion [158, 177] in AnFMBRs.

2.4 Summary

Membrane bioreactors (MBRs) are increasingly applied for wastewater treatment; unfortunately, a key hurdle to the efficient operation of MBRs lies in membrane fouling, which leads to severe flux declines and correspondingly higher energy requirements to restore the flux. Unsurprisingly, studies on optimizing and understanding the associated effects on membrane-fouling mitigation are aplenty.

This review focused on the unsteady-state shear means to mitigate membrane fouling, that induces the shear on the membrane surface to exert the scouring and enhance back-transport. Among which, the process of solid-liquid fluidization of GAC particles (i.e., particle diameter on the order of 1 mm) in anaerobic MBR termed as AnFMBR has been presented as being able to confer membrane-fouling mitigation with energy requirement at least an order-of-magnitude lower than that of bubbling. Since then, efforts on process optimization and indirectly deducing the particle-membrane interaction have been reported, but none has focused on revealing the particle hydrodynamics, which is key to deducing the scouring mechanism, and the target of this thesis work. Accordingly, understanding the relationship between particle behaviour and membrane fouling, in order to further improve the fouling mitigation efficacy and energy efficiency of GAC fluidization in AnMBRs is warranted.

Chapter 3 Characterizing the scouring efficiency of Granular Activated Carbon particles in membrane fouling mitigation via wavelet decomposition of accelerometer signals¹

3.1 Introduction

In fluidization, non-invasive characterization techniques are superior due to the lack of disruption to the hydrodynamics behavior characterized, although difficulties with data interpretation and lower levels of discrimination are recognized [189-192]. The non-invasive technique utilized here - an accelerometer - falls under the category of monitoring via ‘passive acoustics’, which is the measurement of the acoustic emission (AE) created by the process itself [192], in this case by the liquid flow, and particles colliding with each other and with vessel walls. Due to the complex nature of the signals, AE is largely used as fingerprints in describing a specific set of conditions without necessarily knowing or understanding their cause [192]. Efforts harnessing such acoustic methods have been reported to determine the various facets of fluidization, for example, the dynamic behavior of the particles [193, 194], the onset of unstable fluidization [195], general bed dynamics and regime transition [194, 196-200], average size of particles [201], and determining the minimum fluidization

¹ The content in this chapter has been published as:

Jingwei Wang, Bing Wu, Shiliang Yang, Yu Liu, Anthony G Fane, Jia Wei Chew. Characterizing the scouring efficiency of Granular Activated Carbon (GAC) particles in membrane fouling mitigation via wavelet decomposition of accelerometer signals. *Journal of Membrane Science*, volume 498, 15 January 2016, Pages 105–115.

velocity (U_{mf}) [202, 203]. The sensitivity of AE signals to particle movement, coupled with the data analysis method of wavelet decomposition to distinguish the various frequency ranges, makes the accelerometer an ideal technique for the goal in the current work of unveiling the scouring capability of the fluidized GAC particles in mitigating the fouling of membranes.

The objective of this chapter is threefold: (1) use an accelerometer to determine the impact of GAC particle diameter on fluidized bed dynamics, (2) correlate the dynamics of the GAC particles with membrane fouling tendency in filtration tests, and (3) assess the tradeoff based on energy requirement and scouring efficiency considerations in optimizing the GAC particle diameter.

3.2 Materials and methods

3.2.1 Experimental setup

The schematic of the liquid-solid fluidization-membrane filtration setup used in this work is shown in **Fig. 3-1a**. The reactor had an inner diameter of 100 mm and height of 470 mm, and an expanded section with an inner diameter of 135 mm at the top to reduce entrainment. The distributor, designed to ensure uniform liquid flow, was such that five pairs of nozzles were directed downwards in an inverted-V configuration and spread evenly along a central pipe traversing the reactor bottom, as shown in **Fig. 3-1b**. A Cole-Parmer magnetic drive centrifugal pump was used to fluidize the GAC particles in deionized (DI) water. The superficial liquid velocity (U_l) was measured by a variable-area flowmeter (Cole-Parmer, 1.5-20 LPM), while the pressure drop across the fluidized bed was measured by a differential pressure transmitter (Cole-Parmer, 0-1 psi). A

piezoelectric accelerometer with a sensitivity of 100 mV/g procured from National Instruments was used to measure acceleration signals non-invasively to avoid any disruption to the bed dynamics. Specifically, the accelerometer was secured to the wall of the setup at a height of 84 mm above the distributor. Both the pressure drop and acceleration signals were recorded on a computer via the Labview 2013 software.

The flat-sheet membrane module had a width of 80 mm, height of 120 mm, and thickness of 15 mm. The module held two flat-sheet membranes, such that the effective membrane area was 0.0081 m². The module was secured in the fluidized bed at a height of 10 mm above the distributor, such that the entire module was fully submerged in the expanded bed of fluidized particles.

Figure 3-1. (a) Schematic of experimental setup, and (b) Distributor in the liquid-solid fluidization reactor to ensure uniform fluidization.

3.2.2 Materials

The GAC particles (FILTRASORB 300, Calgon Carbon corporation, Pittsburgh, PA USA) had a wide particle size distribution (PSD), with sizes ranging from 0.4 mm to 2.36 mm. To investigate the impact of particles sizes without the influence of polydispersity, which is well-acknowledged to affect fluidization behavior [204], the raw GAC was sieved into various size fractions using Cole-Parmer U.S. Standard Brass Test Sieves. The most dominant particle diameters obtained via Ro-Tap sieve-shaker were in the range of 1-2.35 mm, from which three particle diameters (d_p) (namely, 1.20 ± 0.20 mm, 1.85 ± 0.15 mm and 2.18 ± 0.18 mm) were investigated. After the size classification, the GAC particles were washed with DI water to remove the remnant ash, then dried at 105 °C for 24 h. The same concentration of GAC of 100 g/L was used for every test. Because of the short duration of fluidization (< 30 min), the DI water remained clear visually, hence attrition effects that may lead to a decrease in the particle diameter investigated were assumed to be negligible.

For each filtration test, two pieces of polyvinylidene fluoride (PVDF) flat-sheet membranes purchased from Merck Millipore Ltd with a nominal pore size 0.22 μm were used. The model foulant solution used was 2 g/L Bentonite. Preparing the foulant solution involved adding 8 g of inorganic Bentonite (Sigma-Aldrich) to 4 L of DI water, after which the suspension was first ultrasonicated for 20 min then agitated with a magnetic stirrer at 500 rpm for 20 min.

3.2.3 Accelerometer and Filtration data acquisition

As a precursor to any accelerometer and filtration measurement, the minimum fluidization velocity (U_{mf}) was first determined for each particle diameter (d_p) investigated via the fluidization curve, which is a plot of pressure drop across the bed versus superficial liquid velocity (U_l).

At the start of each experiment, U_l was set at the desired value for about 20 min to achieve steady-state before any data were collected. Accelerometer signals were recorded for 30 s at a sampling frequency of 40,000 Hz. For the filtration tests, the permeate flux was maintained at 30 L/(m² h), during which both the suction pressure and permeate mass were recorded every 15 s.

3.2.4 Analysis of accelerometer signals

The accelerometer signals as a function of time are presented in **Fig. 3-2** for the liquid-solid fluidization of GAC particles of various sizes (**Fig. 3-2a - c**) and for liquid flow alone (**Fig. 3-2d**). Although the mean values appear similar, the frequency of the signal is distinctly different on each sub-plot. To verify if the straightforward method of averaging is able to distinguish the varying impact of the four systems (**Fig. 3-2a - d**), **Fig. 3-3** plots the mean of the accelerometer signal versus superficial liquid velocity (U_l), with each line representing each of the three GAC particle diameters (d_p) investigated and liquid flow alone (i.e., without GAC). Two observations are noted: (i) all four lines are flat, implying that the impact of U_l is negligible; and (ii) among the four systems, the two systems with $d_p = 1.20$ mm and $d_p = 1.85$ mm appear to give similar values, while the system without GAC and the one with $d_p = 2.18$ mm give similar values,

which would suggest a non-monotonic impact of acceleration with respect to d_p . The first observation in particular flags the use of the mean values for the analysis of the scouring effect of the GAC particles, because variations due to changes in U_l , especially in the system without GAC, is well-acknowledged. The underlying reason lies in the insignificant differences in the magnitude of the mean values, which suggests that another analysis method has to be implemented.

Notably, because the frequency of the signals are clearly different (**Fig. 3-2**), a signal analysis method based on extracting information from the signal frequency is needed. Wavelet decomposition presents a good method of extracting different frequency ranges to interpret the various constituent dynamics in fluidized bed systems [205-210]. More specifically, wavelet decomposition provides a means of representing different frequencies of the raw accelerometer signal by repeatedly breaking down the signal into higher-frequency details (D) and lower-frequency approximations (A), as depicted in **Fig. 3-4**. At the first scale of decomposition (Scale 1), the signal of N Hz is decomposed into the first scale of approximation (A_1) and the first scale of detail (D_1), whereby A_1 and D_1 contain half of the lower (i.e., $0 - \frac{N}{2}$ Hz) and higher (i.e., $\frac{N}{2} - N$ Hz) frequency ranges, respectively. Subsequently, as the scale increases from j to $j+1$, the approximation signal A_j is decomposed into approximation A_{j+1} and detail D_{j+1} signals, which contain half of the lower and higher frequency ranges corresponding to A_j , respectively. All accelerometer signals were analyzed via wavelet decomposition using Matlab 2013a. **Fig. 3-5** displays the approximation and detail signals at various scales, illustrating (i) the decrease in signal frequency with the increase of scale, and that (ii) negligible frequency

information is present from scale 10 onwards, hence the maximum scale of decomposition in this work is capped at 13.

Figure 3-2. Typical experimental accelerometer signal collected at 40,000 Hz at a superficial liquid velocity (U_l) of 0.0085 m/s for (a) liquid-solid fluidization of GAC particles with particle diameter (d_p) of 2.18 mm, (b) liquid-solid fluidization of GAC particles with particle diameter (d_p) of 1.85 mm, (c) liquid-solid fluidization of GAC particles with particle diameter (d_p) of 1.20 mm, and (d) liquid only (i.e., without GAC).

Figure 3-3. Average values of accelerometer signal for different GAC sizes

Figure 3-4. Wavelet decomposition analysis of collected signal into various scales [211].

The method of wavelet decomposition is hence such that higher frequencies are represented at lower scales, while lower frequencies are represented at higher scales. Thereby, the dominance of each frequency (i.e., scale) in the signal collected can be quantified by the energy of the detail signal at each scale (D_j), defined as follows [206, 209, 211]:

$$E_{Dj} = \sum_{t=1}^N |D_j(t)|^2 \quad (3-1)$$

where E is the energy of the signal, D is a detail signal at a specified scale, j is the scale of decomposition, N is the total number of data points and t is the time index of the collected data. Based on the knowledge of the physical meaning of each scale, the energies contained in each scale can be expressed graphically to illustrate the dominance of the frequency ranges, thereby allowing differentiation between the contributions of the liquid and solid phases.

Figure 3-5. Raw signal of accelerometer collected at 40,000 Hz and corresponding approximations (A_j) and details (D_j) at various scales (j) of wavelet decomposition for GAC with a particle diameter (d_p) of 1.20 mm and at a superficial liquid velocity (U_l) of 0.0085 m/s.

3.3 Results and discussion

3.3.1 Determination of minimum fluidization velocity (U_{mf})

The minimum fluidization velocity (U_{mf}) is experimentally determined using fluidization curves, which are plots of dynamic pressure drop across the bed versus superficial liquid velocity [212-214], as shown in **Fig. 3-6** for each particle diameter (d_p) investigated. Specifically, U_{mf} is identified as the superficial liquid velocity (U_l) at which the pressure drop across the bed plateaus. The U_{mf} values are necessary to ensure complete fluidization of all the particles in the bed to confer the scouring effect in the filtration experiments. As illustrated in **Fig. 3-6**, the U_{mf} values for GAC particles with particle diameters of 2.18 mm, 1.85 mm and 1.2 mm were determined as 0.0085 m/s, 0.0074 m/s and 0.0053 m/s, respectively. On one hand, the larger U_{mf} values associated with the larger GAC particles imply higher energy consumption. On the other hand, the greater shear induced by the larger GAC particles leads to more effective mitigation of fouling [30]. Therefore, an optimal GAC particle diameter (d_p) should exist that balances increasing energy cost and increasing scouring efficiency with increasing d_p .

Figure 3-6. Fluidization curves for GAC particles with diameters (d_p) of (a) 2.18 mm, (b) 1.85 mm, and (c) 1.2 mm.

3.3.2 Accelerometer results

Various frequencies of data acquisition (namely, 10 kHz, 20 kHz, 30 kHz, and 40 kHz) were investigated to ensure adequate resolution between the solid and liquid phases, since both phases contribute to the accelerometer signal. Specifically, the solid phase corresponds to higher frequency and hence lower scale, while the liquid phase corresponds to lower frequency and hence higher scale. In this case, the data acquisition frequency is limited by the maximum sampling rate of the National Instrument data acquisition module NI 9234 for the accelerometer, which is specified at 51.2 kHz.

Fig. 3-7 shows the energy associated with each detail signal obtained via wavelet decomposition (**Eq. 3-1**) for various data acquisition frequencies. At the lower frequencies (i.e., 10 kHz and 20 kHz), only one peak is observed, implying that higher frequencies are needed in resolving the impact of the solid and liquid phases. Notably, the magnitude of the energy gives an indication of the shear imposed on the accelerometer. When the frequency is increased to 30 kHz, two peaks become apparent, one at $D_1 - D_3$ corresponding to the solid phase while the other at $D_5 - D_7$ corresponding to the liquid phase. A further increase of the frequency to 40 kHz shows that the trend is similar to that at 30 kHz, but expectedly at higher E_{Dj} values. Because a further increase of frequency to 50 kHz limits the duration of data acquisition due to the large file size and in view of the similarity of trends beyond 30 kHz, the data acquisition frequency of 40 kHz was used in all analyses.

Figure 3-7. Wavelet energy distribution of accelerometer sub-signals with different sampling frequency (namely, 10 kHz, 20 kHz, 30 kHz, and 40 kHz), data obtained the fluidization of GAC particles with diameter of 2.18 mm at $U_l = 0.0053$ m/s.

Fig. 3-8a - c show the energy associated with each detail signal obtained via wavelet decomposition at various superficial liquid velocities (U_l) for the liquid-solid fluidization of GAC particles with diameters (d_p) of 2.18 mm, 1.85 mm and 1.2 mm, respectively. **Fig.3-8d** shows the corresponding energy plots for liquid flow alone (i.e., in the absence of particles). For liquid flow alone (**Fig. 3-8d**), or in cases where GAC particles are present but the U_l values are lower than the characteristic U_{mf} of the particles (**Fig. 3-8a - c**), only one peak at $D_5 - D_7$ exists. On the other hand, when the superficial liquid velocity (U_l) is above the characteristic U_{mf} of the particles (**Fig. 3-8a - c**), two peaks at $D_1 - D_3$ and at $D_5 - D_7$ corresponding to the two phases are observed. For example, for the case of GAC particles with $d_p = 2.18$ mm shown in **Fig. 3-8a**, two peaks appear when U_l is at or above 0.0085 m/s, which is the U_{mf} of the GAC particles with $d_p = 2.18$ mm. Collectively, the plots in **Fig. 3-8** indicate conclusively that the peak at $D_1 - D_3$ is associated with the solid phase while the other at $D_5 - D_7$ is associated with the liquid phase.

Figure 3-8. Energy associated with each detail signal obtained via wavelet decomposition at various superficial liquid velocities for (a) GAC particles with diameter of 2.18 mm, (b) GAC particles with diameter of 1.85 mm, (c) GAC particles with diameter of 1.20 mm, and (d) liquid flow alone (i.e., without GAC particles).

To further explore the impact of GAC particle diameter (d_p), each subplot in **Fig. 3-9** shows the energy associated with each detail signal obtained via wavelet decomposition at the same U_l for the liquid-solid fluidization of the three GAC particle sizes and for liquid flow alone. The U_l chosen for each of the four subplots is such that one is lower than the U_{mf} value of the smallest GAC particle (**Fig. 3-9a**), while the other three are the U_{mf} values corresponding to each of the d_p investigated (**Fig. 3-9b - d**). **Fig. 3-9a** shows only the peaks at $D_5 - D_7$ associated with the liquid phase for all four systems investigated, because none of the GAC particles was fluidized at the U_l of 0.0042 m/s. In **Fig. 3-9b**, where

the U_l implemented was that of the U_{mf} of the smallest GAC particles (i.e., $d_p = 1.20$ mm) investigated, the only peaks at $D_1 - D_3$ associated with the solid phase belong to the smallest GAC particles. As for **Fig. 3-9c**, where the U_l implemented was that of the U_{mf} of the GAC particles with diameter of 1.85 mm, peaks at $D_1 - D_3$ is clearly observed for both GAC particles with $d_p = 1.20$ mm and $d_p = 1.85$ mm. Furthermore, it is also apparent that the peaks at $D_1 - D_3$ are higher for $d_p = 1.85$ mm because, although the attendant lower $U_l - U_{mf}$ should confer lower mobility of the particles, the greater inertia of the larger particles dominates the acoustic effect. Finally for **Fig. 3-9d**, peaks at $D_1 - D_3$ are seen for all three GAC particle sizes but not for the liquid only system. In this case, the GAC particles with $d_p = 1.8$ mm persist to exhibit the highest peaks at $D_1 - D_3$, presumably due to an optimal balance between $U_l - U_{mf}$ and inertia: whereas the smallest GAC particles (i.e., $d_p = 1.20$ mm) have the greatest $U_l - U_{mf}$ but least inertia, the largest GAC particles (i.e., $d_p = 2.18$ mm) have the least $U_l - U_{mf}$ but greatest inertia.

Figure 3-9. Energy associated with each detail signal obtained via wavelet decomposition at superficial liquid velocities of (a) 0.0042 m/s (i.e., lower than the U_{mf} of the smallest GAC particle), (b) 0.0053 m/s (i.e., U_{mf} of the GAC particle with diameter of 1.20 mm), (c) 0.0074 m/s (i.e., U_{mf} of the GAC particle with diameter of 1.85 mm), and (d) 0.0085 m/s (i.e., U_{mf} of the GAC particle with diameter of 2.18 mm).

Since **Fig. 3-8** and **Fig. 3-9** show clearly that the contribution of the solid phase of interest here is represented by E_{D1} , E_{D2} and E_{D3} , the sum of these three values was obtained to ascertain the scouring extents of the various GAC particle sizes at various superficial liquid velocities (U_l), after which the correlation with fouling rate is examined. Accordingly, **Fig. 3-10** presents the sum of E_{D1} , E_{D2} and E_{D3} versus U_l for liquid alone (i.e., without GAC) and liquid-solid fluidization of the three GAC particle diameters (d_p). Notably, the accelerometer signals provide well for determining U_{mf} values [202, 203], as observed in the non-zero $E_{D1} + E_{D2} + E_{D3}$ values only when $U_l \geq U_{mf}$ for each d_p . Two further

pieces of evidence indicate that the contribution of the solid phase is represented by E_{D1} , E_{D2} and E_{D3} : (i) for the system without GAC, the sum of E_{D1} , E_{D2} and E_{D3} expectedly shows negligible values, and (ii) for the systems with GAC, the sum of E_{D1} , E_{D2} and E_{D3} for each d_p only starts to show non-negligible values beyond the U_{mf} value characteristic of the d_p . Two more noteworthy observations should be highlighted for when all three particle sizes were completely fluidized beyond $U_l = 0.0085$ m/s (i.e., U_{mf} corresponding to $d_p = 2.18$ mm). First, the $E_{D1} + E_{D2} + E_{D3}$ values are the lowest for $d_p = 1.20$ mm, because although the associated $U_l - U_{mf}$ is the greatest, the inertia is the least, the latter of which presumably dominates in this case. Second, the $E_{D1} + E_{D2} + E_{D3}$ values are similar for both $d_p = 1.85$ mm and $d_p = 2.18$ mm, which indicates that the greatest inertia conferred by the largest particles is countered by the lower $U_l - U_{mf}$ value; therefore, $d_p = 1.85$ mm seems to represent the optimal size in terms of the non-negligible impact of the particles at lower U_l (e.g., $U_l = 0.0074$ m/s and $U_l = 0.0085$ m/s, whereby $E_{D1} + E_{D2} + E_{D3}$ is higher for $d_p = 1.85$ mm than $d_p = 2.18$ mm) and hence lower energy costs.

Figure 3-10. Energy associated with detail signals D_1 to D_3 obtained via wavelet decomposition ($E_{D1} + E_{D2} + E_{D3}$) as a function of liquid velocity (U_l).

3.3.3 Filtration results

The scouring effect of various GAC particle sizes was investigated in filtration experiments. **Fig. 3-11a** shows the evolution of Trans-membrane Pressure (TMP) with time in the absence and presence of GAC at two U_l values, which corresponds to the U_{mf} values of the smallest and largest d_p investigated. Four observations are worth noting. Firstly, comparing the two systems without GAC (i.e., liquid flow alone), the TMP rise with time was expectedly greater for the lower U_l of 0.0053 m/s due to the associated lesser shear on the membrane. Secondly, the TMP rise for the two systems without GAC was significantly greater than that in the presence of fluidized GAC, which attests to the mechanical scouring capability of GAC and agrees well with the higher $E_{D1} + E_{D2} + E_{D3}$ values reflective of the solid phase (**Fig. 3-10**). Thirdly, the TMP rise with time for GAC particles with $d_p = 1.20$ mm at the characteristic U_{mf} of 0.0053

m/s was greater than that for GAC particles with $d_p = 2.18$ mm at the characteristic U_{mf} of 0.0085 m/s, which agrees with the higher $E_{D1} + E_{D2} + E_{D3}$ value of the latter in **Fig. 3-10**. This indicates that the larger GAC particles performed better at fouling mitigation albeit at a higher energy cost. Fourthly, comparing the TMP rise with time at the same U_l of 0.0085 m/s for the two d_p investigated of 1.20 mm and 2.18 mm reveals approximately similar trends of TMP rise, which agrees with the $E_{D1} + E_{D2} + E_{D3}$ values in **Fig. 3-10**. **Fig. 3-11b** further shows the relationship between dTMP/dt and $E_{D1} + E_{D2} + E_{D3}$ for the five datasets in **Fig. 3-11a**. Specifically, dTMP/dt was obtained by dividing the TMP rise in the 60 minutes investigated by 60 minutes. **Fig. 3-11b** indicates that a slight increase in $E_{D1} + E_{D2} + E_{D3}$ conferred by the fluidizing the smallest GAC ($d_p = 1.20$ mm) at the corresponding U_{mf} results in a significant improvement in dTMP/dt, while further increases in $E_{D1} + E_{D2} + E_{D3}$ either by fluidizing the largest GAC ($d_p = 2.18$ mm) at the corresponding U_{mf} or the smallest GAC ($d_p = 1.20$ mm) at a higher velocity gave slight further improvement in dTMP/dt. It should be noted that this may be unique to Bentonite, which is relatively easier to remove than other foulants.

Figure 3-11. (a) TMP versus time in the absence and presence of fluidized GAC at two different U_l values; (b) Relationship between $d\text{TMP}/dt$ and $E_{D1} + E_{D2} + E_{D3}$.

3.3.4 Energy consumption

For a fluidized bed, the power requirements can be evaluated by the following equation [215]:

$$P = \Delta P_{bed} \dot{V} \quad (3-2)$$

where P_r is the power requirement (W), ΔP_{bed} is the pressure drop across the liquid-solid fluidized bed (Pa), and $\dot{V}$ is the volumetric flow rate (m^3/s). For a given permeate flow rate Q_p (m^3/s), the power requirement per unit permeate, P_p , in terms of kWh/m^3 can be expressed as:

$$P_p = \frac{P_r}{3600Q_p} = \frac{\Delta P_{bed} \dot{V}}{3600Q_p} \quad (3-3)$$

Accordingly, **Fig. 3-12** is a plot of $E_{D1} + E_{D2} + E_{D3}$ versus d_p . Each line represents the same P_p incurred by the three particle diameters per **Eq. 3-3**; because ΔP_{bed} values were constant for the fully fluidized conditions investigated and Q_p were held constant for all the filtration tests, each line simply represents a different $\dot{V}$ condition. Clearly, for the P_p value, all three particle

diameters (d_p) investigated conferred similar scouring effects by the particles (i.e., similar $E_{D1} + E_{D2} + E_{D3}$ values) at low $E_{D1} + E_{D2} + E_{D3}$ values, whereas the larger particles (i.e., $d_p = 1.85$ mm and $d_p = 2.18$ mm) provided greater scouring capacity than the smallest particle of $d_p = 1.20$ mm at higher $E_{D1} + E_{D2} + E_{D3}$ values. In other words, the impact of d_p is insignificant with regards to energy cost incurred in mitigating fouling at lower scouring efficiency, but the GAC particles with larger particle diameters (d_p) demand less energy at higher scouring efficiency. As noted earlier, the Bentonite foulant investigated here is relatively easier to remove, hence the scouring (**Fig. 3-11b**) and P_p required is presumably lower in this case, in which case **Fig. 3-12** shows that d_p exerts insignificant impact on scouring efficiency; however, for more adhesive foulants, higher P_p may be required, in which case the larger particles may be more effective. More tests on different foulants are needed, which is part of an on-going investigation.

Figure 3-12. Energy associated with detail signals D_1 to D_3 obtained via wavelet decomposition ($E_{D1} + E_{D2} + E_{D3}$) as a function of particle diameter (d_p).

3.4 Conclusions

The scouring efficiency of GAC particles with three particle diameters (namely, 1.20 mm, 1.85 mm, and 2.18 mm) has been investigated in this study. Three highlights should be noted. First, accelerometer signals were analyzed via wavelet decomposition to successfully resolve the contributions of the liquid and solid phases in the liquid-solid fluidization system, in view of the physical understanding that the liquid phase constitutes the lower-frequency while the solid phase constitutes the higher-frequency ranges of the signal. Results show that the GAC with smallest particle diameter of 1.20 mm conferred the least scouring efficiency, while both the larger GAC particles with diameters of 1.85 mm and 2.18 mm provided similar scouring efficiency, which is due to the balance between particle inertia, and the difference between superficial liquid velocity and minimum fluidization velocity ($U_l - U_{mf}$). Second, the energy contained in the frequency range of the accelerometer signal corresponding to the solid phase was found to correlate well with the fouling mitigation extents in the filtration results. Specifically, the higher the representative energy of the solid phase (i.e., at lower wavelet decomposition scales) is, the slower the rise in TMP with time (i.e., more effective fouling mitigation). Third, the energy required to impact membrane fouling mitigation by the GAC particles with various diameters was calculated. At lower scouring efficiency, the energy requirement of all three GAC particles were similar. At higher scouring efficiency, the energy requirement of the smallest d_p of 1.20 mm was the greatest, while that of the larger ones were similar.

Chapter 4 Correlating the hydrodynamics of fluidized Granular Activated Carbon (GAC) with membrane-fouling mitigation²

4.1 Introduction

In Chapter 3, we used an accelerometer to characterize wall-suspension interactions during GAC fluidization. It was found that a trade-off exists between particle scouring efficiency and energy consumption. Although a larger particle confers more effective scouring due to inertial forces, the energy required to fluidize the particle is also commensurately higher. If the energy used to fluidize the larger particle is used for smaller particles, the greater mobility gained by the smaller particles may confer even more effective scouring. To explore detailed particle behavioural characteristics for different particle diameters (d_p), at different superficial liquid velocities (U_l) and at different heights along the membrane, a high-speed video camera was employed in this study. ImageJ was used to analyse the images for particle velocity and particle concentration, following which particle momentum was also derived. To quantify membrane fouling, the trans-membrane pressure (TMP) was monitored.

The specific objectives of this study are:

2. The content in this chapter has been published as:

Jingwei Wang, Farhad Zamani, Andy Cahyadi, Jia Yuan Toh, Shiliang Yang, Bing Wu, Yu Liu, Anthony G Fane, Jia Wei Chew. Correlating the hydrodynamics of fluidized granular activated carbon (GAC) with membrane-fouling mitigation. *Journal of Membrane Science*, volume 510, 15 July 2016, Pages 38–49.

- i. Characterize the velocity and concentration distribution of GAC particle fluidization axially along the membrane;
- ii. Correlate the dynamics of particle behaviour with fouling mitigation efficacy;
- iii. Assess the relationship between membrane-fouling mitigation and energy consumption.

4.2 Materials and methods

4.2.1 Experimental setup

Fig. 4-1 shows a schematic of the setup used to determine the effectiveness of membrane-fouling mitigation via GAC fluidization. The water flow rate needed for GAC fluidization was provided by a magnetic-drive centrifugal pump (Cole-Parmer, 90-9000 rpm), the flow rate was measured by a variable-area flowmeter (Cole-Parmer, 1.5-20 LPM), while the permeate suction was provided by a peristaltic pump (MasterFlex, Cole-Parmer 7523-80, 0.1-600 RPM, USA). The increase in permeate mass was measured using a balance (Mettler-Toledo, PB3002-S, 0.5 - 3100 g, Switzerland).

Figure 4-1. Schematic of fluidized bed – membrane filtration experimental setup

Fig. 4-2a shows the schematic and dimensions of the fluidized bed – membrane filtration module, which has a rectangular cross-section and a distributor designed for uniform solid-liquid fluidization of the granular activated carbon (GAC) particles. Specifically, the setup was comprised of a lower section with a cross-sectional area of 89 mm by 33 mm and height of 194 mm, and an upper section with a wider cross-sectional area of 150 mm by 93 mm to reduce entrainment and height of 124 mm. A flat-sheet membrane module, with a cross-sectional area of 80 mm by 15 mm and height of 150 mm, holds a membrane with an active area of 46 mm by 87 mm. The membrane module was placed flush with the wall of the filtration setup to minimize disturbances to the particle fluidization. The distributor used was of the same design as that of the study in Chapter 3 and depicted in more detail in **Fig. 4-2b**. Specifically, the design of the

distributor was such that eight pairs of nozzles were projected downwards in an inverted-V configuration.

Figure 4-2. Schematic and dimensions of (a) fluidized bed – membrane filtration module, and (b) distributor. Dimensions are in units of mm.

For the filtration tests, the permeate flux was maintained at 30 L/(m² h), during which both the suction pressure and permeate mass were recorded every 20 s via LabView. To compare fouling results with GAC fluidization behaviour, the extent of fouling was investigated at three different heights of the membrane. Specifically, for each filtration test, only one-third of the membrane (i.e., an area of 46 mm by 30 mm) was exposed by adhering plastic sheets onto other parts of the membrane using double-sided adhesive tape.

A high-speed video camera (Olympus *i*-SPEED LT) was used to capture the velocity and concentration of GAC particles during fluidization. The videos were recorded about 10 minutes after setting the U_l at the desired value to ensure that steady-state was achieved, and each recording lasted for about 5 - 10 seconds at a frequency of 100 frames per second (fps). For each experimental condition investigated, the videos were taken at five heights along the setup, with the lowest and highest heights spanning the membrane at 15 mm and 75 mm, respectively, above the diffuser.

4.2.2 Materials

To investigate the effect of GAC particle diameter necessitates a narrower PSD to avoid the acknowledged influence of polydispersity [216, 217], hence the procured GAC particles were sieved into various size fractions using Cole-Parmer U.S. Standard Brass Test Sieves and the Ro-Tap sieve-shaker. The three particle diameters (d_p) investigated were 1.85 ± 0.15 mm, 1.55 ± 0.15 mm and 1.015 ± 0.165 mm. For each test, 35 g of GAC was used, which gave a particle concentration of around 90 g/L.

The flat-sheet membrane used was polyvinylidene fluoride (PVDF), hydrophilic, and had a nominal pore diameter of 0.22 μm (Merck Millipore). The feed solution investigated was a mixture of 2 g/L Bentonite (Sigma-Aldrich) and 2 g/L Yeast (Saf-instant) in DI water to simulate biomass, colloids, polysaccharides and inorganic particles in wastewater [218]. The mean particle size of the model solution was $5.35 \pm 0.11 \mu\text{m}$, as measured by a Mastersizer (Malvern Mastersizer 2000).

4.2.3 Image analysis

The videos captured by high-speed camera were analyzed using ImageJ. Each video clip was first converted into frames of images that were 0.01 s apart, after which the particles in focus were identified, and the particle concentration, and horizontal (x -directional) and vertical (y -directional) velocities analyzed. It should be noted that the focal plane of the camera was in the vertical plane near the wall of the fluidized bed – membrane filtration module (**Fig. 4-2a**). The reason for only taking particles in focus into account was because only these particles exhibited hydrodynamics representative of the region near the membrane surface, which exerted a similar influence on fluidization as the wall [216, 217, 219]. As shown in the image in **Fig. 4-3**, only ten particles in this example were in focus and were analyzed.

Particle velocity was computed by an adaptation of particle tracking velocimetry. Thus, to calculate particle velocity, the distance moved by the particles from one image to the next was divided by the time lapse between the two images (which in this case was 0.01 s). For example, the x - and y -coordinates

of the particles in focus on **Figs. 4-3a** and **4-3b** were first extracted via ImageJ, then the distances moved between **Figs. 4-3a** and **4-3b** in both the x - and y -directions were calculated respectively as Δx and Δy , following which the velocities in the x - and y -directions were calculated respectively as $\frac{\Delta x}{0.01}$ and $\frac{\Delta y}{0.01}$. **Table 4-1** lists the numerical values of these parameters. Regarding particle concentration, the number of in-focus particles were normalized with respect to the area evaluated. In the case of **Fig. 4-3a**, the number of particles was 10 while the area evaluated was 19.05 mm by 14.29 mm, hence the particle concentration was 0.0367 mm^{-2} . For each experimental condition assessed, between 200 and 1000 particles were evaluated.

Figure 4-3. Images extracted from a video clip, whereby the particle diameter (d_p) was 1.85 mm and the superficial liquid velocity (U_l) was 0.06 m/s. (a) and (b) are 0.01 s apart; only particles in focus as identified on the images were analyzed.

Table 4-1. Particle coordinates of the in-focus particles in **Fig. 4-3** and the corresponding particle velocities. (x_1, y_1) are particle coordinates obtained from **Fig. 4-3a**, and (x_2, y_2) are particle coordinates obtained from **Fig. 4-3b**. Distance moved ($\Delta x, \Delta y$) was derived by $(x_2 - x_1, y_2 - y_1)$, while particle velocities v_x and v_y were respectively calculated by $\Delta x / 0.01$ and $\Delta y / 0.01$.

Parti cles	Particle coordinate at t = 0 s (Fig. 4-3a)		Particle coordinate at t = 0.01 s (Fig. 4-3b)		Distance moved (mm)		Particle velocity (mm/s)	
	x_1	y_1	x_2	y_2	Δx	Δy	v_x	v_y
1	1.214	7.738	1.095	8.690	-0.119	-0.952	-11.9	-95.2
2	0.738	10.667	0.786	10.929	0.048	-0.262	4.80	-26.2
3	4.262	9.024	4.452	9.238	0.190	-0.214	19.0	-21.4
4	4.381	13.714	4.667	14.071	0.286	-0.357	28.6	-35.7
5	9.500	9.024	9.619	9.429	0.119	-0.405	11.9	-40.5
6	12.167	0.881	12.214	0.929	0.047	-0.048	4.70	-4.80
7	15.714	5.333	15.548	5.500	-0.166	-0.167	-16.6	-16.7
8	15.571	3.762	15.524	3.905	-0.047	-0.143	-4.70	-14.3
9	18.738	4.810	18.500	4.714	-0.238	0.096	-23.8	9.60
10	14.286	7.048	14.238	7.238	-0.048	-0.190	-4.80	-19.0

4.3 Results and discussion

4.3.1 Determination of minimum fluidization velocity (U_{mf})

The GAC particles have to be fluidized to enable the mechanical scouring capability to mitigate membrane fouling. To ensure the GAC particles were fully fluidized at the velocities investigated, the minimum fluidization velocity (U_{mf}), which is the minimum superficial liquid velocity at which the drag force balances the bed weight, was first determined via fluidization curves (i.e., dynamic pressure drop across the bed (ΔP_{bed}) versus superficial liquid velocity (U_l)) [220].

The U_{mf} values of GAC with particle diameters (d_p) of 1.85 mm, 1.55 mm and

1.015 mm were determined respectively as 0.0079 m/s, 0.0068 m/s and 0.0034 m/s, which agree with the work in Chapter 3. While the lowermost U_l used in the experiments was such that the fluidized bed height spanned the entire membrane, the uppermost U_l was such that entrainment was minimal. Accordingly, whereas the U_l values ranged from 0.04 m/s to 0.06 m/s for the GAC particles with $d_p = 1.85$ mm and 1.55 mm, the U_l values ranged from 0.02 to 0.03 m/s for the GAC particles with $d_p = 1.015$ mm.

4.3.2 Fouling results

The membrane fouling tendencies in the presence of the fluidization of various GAC particle diameters (d_p) at different superficial liquid velocities (U_l) were investigated. **Fig. 4-4** shows the rate of change of trans-membrane pressure (TMP) where the TMP was measured over 2 h, and normalized with respect to the duration to obtain $d\text{TMP}/dt$. Each sub-plot (i.e., **Figs. 4-4a, b and c**) represents each d_p investigated, and contains the effect of U_l at three heights (h/H , where h is the height assessed while H is the total module height of 194 mm (**Fig. 4-2a**)). The ranges of the y-axes are kept constant for all three sub-plots for easier comparison on the effect of d_p . Two observations are apparent. Firstly, $d\text{TMP}/dt$ tended to increase as h/H increased, which indicate that the lower parts of the membranes were more effectively scoured by the GAC particles. This may be construed as a size-segregation effect, whereby the larger particles segregated to the bottom and hence incurred more effective scouring, but it should be noted that the particles were sieved to as narrow a size range as experimentally possible; nonetheless, size segregation is inevitable, and studies of the corresponding effects in future studies will be useful. Secondly, the expected negative

correlation between $dTMP/dt$ and U_l was consistent across all three d_p investigated only at the middle h/H of 0.57.

Figure 4-4. Fouling rate values of GAC particle velocity with diameters (a) 1.85 mm, (b) 1.55 mm and (c) 1.015 mm at different heights and superficial velocities.

4.3.3 Influence of superficial liquid velocity (U_l) on GAC particle velocity (V_x and V_y)

The velocities of the particles are directly related to the efficiency of the mechanical scouring effects on membrane-fouling mitigation. **Fig. 4-5** provides a comprehensive overview of the probability distribution of the particle velocities in the horizontal (x) direction (V_x); the x - and y -axes ranges of all 15 plots are

kept the same for easier comparison. Specifically, each of the three columns represents a particle diameter (d_p) in the range of 1.015 mm to 1.85 mm, while each of the five rows represents a dimensionless height of the fluidized bed – membrane filtration module (h/H) in the range of 0.41 to 0.72, which correspond to the span of the active area of the membrane. In addition, each probability distribution in each sub-plot represents each U_l evaluated, hence the lowest U_l is absent in the sub-plots for the higher h/H because no particle was found higher in the module. Three observations are worth highlighting: (i) the V_x probability distributions are generally broadest at the lowest height ($h/H = 0.41$), presumably due to the effects of the distributor (i.e., entrance effects [221, 222]); (ii) at higher heights of $h/H \geq 0.64$, the V_x probability distributions were similar at different U_l values, which indicate the negligible impact of U_l at higher heights; and (iii) the V_x values tended to be dominantly negative (i.e., leftwards) or positive (i.e., rightwards) at the lower heights, but appeared to be more evenly distributed around $V_x = 0$ at the highest height ($h/H = 0.72$). Analogous to **Fig. 4-5**, **Fig. 4-6** provides a comprehensive overview of the probability distribution of the particle velocities in the vertical (y) direction (V_y). The three observations made for V_x , appear to still approximately hold for V_y . Such detailed information on particle velocities is expected to find further value in validating models, which would in turn provide more physical understanding to the particle hydrodynamics.

Figure 4-5. Particle velocity distribution in the horizontal (x) direction (V_x). Each of the three columns represents a particle diameter (d_p), while each of the five rows represents a dimensionless height of the fluidized bed - membrane module (h/H , where h is the height assessed while H is the total module height of 194 mm (**Fig. 4-2**))

Figure 4-6. Particle velocity distribution in the vertical (y) direction (V_y). Each of the three columns represents a particle diameter (d_p), while each of the five rows represents a dimensionless height of the fluidized bed - membrane module (h/H , where h is the height assessed while H is the total module height of 194 mm (**Fig. 4-2**)).

The V_x and V_y probability distributions respectively in **Figs. 4-5** and **4-6** are useful in understanding the trajectories of the GAC particles, but appear to be deficient in shedding any light on the varying scouring efficiency of the different d_p and U_i values shown in **Fig. 4-4**. To summarize the velocity trends, root-mean-square means (V_{rms}) rather than arithmetic means (V_{arith}) of the distributions were

obtained. Whereas V_{rms} (Eq. 4-1) accounts for both positive and negative V values which are both important in the mechanical scouring of the fouled membranes, V_{arith} (Eq. 4-2) is such that the positive and negative V values cancel out each other, hence V_{rms} is the more suitable mean here.

$$V_{rms} = \sqrt{\sum V^2 P(V)} \quad (4-1)$$

$$V_{arith} = \sum V P(V) \quad (4-2)$$

where $P(V)$ stands for the probability of a particle having a particle velocity of V (Figs. 4-5 and 4-6). Fig. 4-7 presents trends of $V_{rms,x}$ (the first column in Fig. 4-5) and $V_{rms,y}$ (the second column in Fig. 4-6) with respect to U_l ; each of the three rows in Fig. 4-7 represents each d_p investigated. The error bar represents one standard derivation (σ) calculated by:

$$\sigma = \sqrt{\sum (V - V_{rms})^2 P(V)} \quad (4-3)$$

Fig. 4-7 shows a lack of correlation between V_{rms} and U_l . Lower heights (i.e., $h/H \leq 0.49$) tended to give higher V_{rms} at the same U_l , which is analogous with the results in Fig. 4-5 and Fig. 4-6, and presumably due to the distributor effect. There is no obvious correlation between $V_{rms,x}$ and $V_{rms,y}$. However, both of them were beneficial for the scouring efficacy.

Figure 4-7. Root-mean-square particle velocities in the x -direction ($V_{rms,x}$; first column) and y -direction ($V_{rms,y}$; second column) at different U_l . Each of the three rows represents a d_p investigated.

Because both $V_{rms,x}$ and $V_{rms,y}$ contribute presumably equally to scouring, a means of combining both parameters as a single particle velocity value (V_p) will allow for better correlation with membrane-fouling mitigation (**Fig. 4-4**):

$$V_p = \sqrt{V_{rms,x}^2 + V_{rms,y}^2} \quad (4-4)$$

Analogous to **Fig. 4-7**, **Fig. 4-8** illustrates V_p versus U_l at various heights (h/H), with each subplot (i.e., **Figs. 4-8a, b and c**) representing each of the d_p

investigated and each error bar representing the span of V_p values. Again, **Fig. 4-8** shows that the characteristic particle velocity V_p did not correlate well with U_l or h/H . Collectively, **Figs. 4-5 to 4-8** indicate either the lack of or at most a weak correlation between V (either V_x or V_y or V_{rms} or V_p) and U_l or h/H , which hence imply surprisingly that particle velocity trends did not relate well to the fouling trends in **Fig. 4-4**.

Figure 4-8. V_p versus U_l trends at five heights (h/H) for three different d_p : (a) 1.85mm, (b) 1.55 mm and (c) 1.015 mm.

4.3.4 GAC particle concentration

Although the overall particle concentration was kept constant at 90 g/L for every experiment, the particle distribution throughout the module, which

depended on d_p and U_l among other factors, was not uniform. Apart from particle velocities, the number of particles in contact with the membrane also played an important role in the effectiveness of membrane-fouling mitigation. **Fig. 4-9** displays the trends of particle concentration versus U_l at various heights (h/H) for GAC particles with three different particle diameters (d_p). Three observations, the first of which appears to be deficient in explaining the fouling trends in **Fig. 4-4**, are noted. Firstly, **Fig. 4-9** shows that, regardless of d_p , the lowermost height ($h/H = 0.41$) gave the flattest particle concentration versus U_l profiles, which indicated that U_l had an insignificant impact on particle concentration at this height. On the contrary, **Fig. 4-4** shows that U_l had a negligible effect on $d\text{TMP}/dt$ for $d_p = 1.85$ mm (**Fig. 4-4a**) but affected $d\text{TMP}/dt$ for $d_p = 1.55$ mm (**Fig. 4-4b**) and 1.015 mm (**Fig. 4-4c**). Secondly, **Fig. 4-9** shows that, at the greater heights ($h/H \geq 0.49$), particle concentration tended to increase with U_l , implying that more energy (i.e., higher U_l) was needed to increase the number of particle contacts with the membrane. This is in line with the trends in **Fig. 4-4** where an increase in U_l tended to decrease $d\text{TMP}/dt$, which could stem from the higher particle concentration. Thirdly, the particle concentration trends appeared to be more dependent on U_l and h/H than the particle velocity trends (**Figs. 4-5 to 4-8**).

Figure 4-9. GAC particle concentration in terms of particle number per m² versus U_l trends at five heights (h/H) for three different d_p : (a) 1.85mm, (b) 1.55 mm and (c) 1.015 mm.

4.3.5 GAC particle momentum

Since neither particle velocity or particle concentration alone could explain completely the fouling trends in **Fig. 4-4**, although both effects are expected to contribute non-negligibly towards membrane-fouling mitigation, another parameter is needed. Particle momentum (p), which accounts for both

particle velocity (V_p) and particle concentration, can be used to quantify both effects:

$$p = mV_p = n \frac{1}{6} \pi d_p^3 \rho_p V_p \quad (4-5)$$

where m is the mass of the particle and ρ_p is the effective particle density (i.e., the density of the porous GAC particle saturated with water). **Fig. 4-10** displays p as a function of U_l at five heights corresponding to the active membrane area; each of the three sub-plots (i.e., **Figs. 4-10a, b and c**) represents a particle diameter (d_p). At greater heights of $h/H \geq 0.57$, p tended to increase with U_l regardless of d_p ; on the other hand, at lower heights of $h/H \leq 0.49$, the relationship between p and U_l tended to be non-monotonic presumably due to entrance effects [221, 222]. Furthermore, although the range of U_l investigated in both **Figs. 4-10a and b** are the same, the larger GAC particles with $d_p = 1.85$ mm (**Fig. 4-10a**) exhibited higher p values particularly at lower h/H .

Figure 4-10. Particle momentum (p) per m^2 versus U_l trends at five heights (h/H) for three different d_p : (a) 1.85mm, (b) 1.55 mm and (c) 1.015 mm.

4.3.6 Correlating GAC particle behavior with membrane-fouling mitigation

Figs. 4-11a - c summarize the relationship between membrane fouling and GAC particle behavior. Specifically Figs. 4-11a, b and c are plots of $dTMP/dt$ (Fig. 4-4) versus V_p (Fig. 4-8), particle concentration (Fig. 4-9), and

particle momentum (p ; **Fig. 4-10**). **Fig. 4-11a** illustrates that $dTMP/dt$ decreased with V_p for all three d_p investigated, especially for the largest d_p of 1.85 mm, which affirms expectedly that higher particle velocities (V_p) were more effective in reducing membrane fouling. Regarding **Fig. 4-11b**, $dTMP/dt$ decreased as particle concentration increased, particularly for the two larger d_p but less so for the smallest d_p , which indicates expectedly that increased particle contact with the membrane was beneficial in mitigating membrane fouling. As for **Fig. 4-11c**, the negative correlation between $dTMP/dt$ and p was most pronounced for all three d_p , compared to that between $dTMP/dt$ and V_p (**Fig. 4-11a**) or particle concentration (**Fig. 4-11b**), which suggests that momentum transfer between the GAC particles and the membrane represents a key mechanism effecting the scouring to diminish membrane fouling. This agrees with the finding in Chapter 3, whereby an increase in the particle contribution in the accelerometer signals diminished $dTMP/dt$.

Fig. 4-11d shows $dTMP/dt$ as a function of power required per unit permeate (P_p) to understand the relationship between the extents of membrane fouling and power requirement. The power required for fluidizing the GAC particles (P_r), which has unit of kW, is given by:

$$p_r = \Delta P_{bed} Q \quad (4-6)$$

where ΔP_{bed} is the pressure drop across the liquid-solid fluidized bed (kPa) and Q is the volumetric flow rate (m^3/s). For a given permeate flow rate Q_p (m^3/s), the power requirement per unit permeate (P_p), which has unit of kWh/m^3 , can be expressed as:

$$P_p = \frac{p_r}{3600Q_p} = \frac{\Delta P_{bed}Q}{3600Q_p} \quad (4-7)$$

Fig. 4-11d shows a weak and scattered correlation between $dTMP/dt$ and P_p , which indicates that a higher P_p did not guarantee a more effective reduction in membrane fouling. This agrees with the previous study in Chapter 3, where a trade-off was recognized between the increased efficacy of the larger particles in mitigating membrane fouling and the increased energy required to fluidize these larger particles.

Figure 4-11. $dTMP/dt$ versus (a) V_p , (b) particle concentration, (c) particle momentum (p), and (d) energy requirement per unit permeate (P_p).

4.4 Conclusions

The goal of this chapter's work was to understand the correlation between GAC particle behavior in a solid-liquid fluidized bed and the extent of membrane-fouling mitigation via mechanical scouring by the particles. The impact of GAC particle diameter (d_p), superficial liquid velocity (U_l) and axial position of the membrane (h/H) on particle behavior (namely, particle velocity, concentration and momentum) and membrane fouling was investigated. The characterization of the GAC particle velocity and concentration was enabled by a high-speed video camera and image analysis via ImageJ, while membrane fouling was characterized by the rate of rise of TMP (i.e., $d\text{TMP}/dt$).

The four trends obtained are summarized here. Firstly, the $d\text{TMP}/dt$ tended to increase as h/H increased, and the expected negative correlation between $d\text{TMP}/dt$ and U_l was consistent across all three d_p investigated only at the middle height of the membrane. Secondly, as for the particle velocity trends, despite the comprehensive landscape of vertical and lateral particle velocity distributions obtained, the particle velocity trends did not appear to relate well to the $d\text{TMP}/dt$ trends; nonetheless, the detailed velocity information is expected to find additional value in the validation of models for gaining further physical insights. Thirdly, with respect to the particle concentration trends, they exhibited more dependence on U_l and h/H than the particle velocity trends, but again a straightforward correlation with the fouling trends appeared elusive. Fourthly on the particle momentum (p) trends, p tended to increase with U_l at higher h/H but was non-monotonic with respect to U_l at lower h/H , and the larger GAC particles exhibited higher p values particularly at lower h/H .

Directly comparing particle behavior with membrane fouling trends show that higher particle velocities, particle concentration and particle momentum generally all showed a negative correlation with $dTMP/dt$. Of the three particle behavioral characteristics (namely, velocity, concentration, momentum), the negative correlation between $dTMP/dt$ and particle momentum was most pronounced, which suggests that momentum transfer between the GAC particles and the membrane represents a key mechanism effecting the scouring to diminish membrane fouling. Finally, a weak and scattered correlation between $dTMP/dt$ and power requirement was observed, which indicates that a higher energy input does not necessarily confer a more effective reduction in membrane fouling.

Chapter 5 Effect of fluidized granular activated carbon (GAC) on critical flux in the microfiltration of particulate foulants³

5.1 Introduction

As presented in previous chapters, the efficacy of fluidized GAC particles have flourished in recent years [13, 15-24]. Other than the capability to adsorb organic contaminants, the hydrodynamics of the fluidized GAC have been found to be directly relatable to the effectiveness of fouling mitigation in Chapter 3 and 4. This study is the first systematic study of the effect of mm-sized GAC particle fluidization on critical flux (J_c) of fine (micron-sized) particles in microfiltration.

The concept of critical flux (J_c), which is the minimum permeate flux at which fouling becomes observable, was first proposed by Field et al. [87]. In this regard, a direct observation technique, like the Direct Observation Through the Membrane (DOTM), is acknowledged to provide a more precise determination of critical flux compared to conventional methods like rate of change of the trans-membrane pressure (TMP) [223-225]. The unique feature of the DOTM technique is the use of a transparent membrane to allow for the focal plane of the observation to be exactly at the feed-membrane interface, in order for any deposition of foulant as small as 3 μm to be observed through the microscope.

3. The content in this chapter has been published as:

Jingwei Wang, Bing Wu, Yu Liu, Anthony G. Fane, Jia Wei Chew. Effect of fluidized granular activated carbon (GAC) on critical flux in the microfiltration of particulate foulants. *Journal of Membrane Science*, volume 523, 1 February 2017, Pages 409–417.

Other than for the measurement of critical flux, DOTM has also been proven to be useful for understanding the coalescence behaviour of a deposited oil emulsion [226], shear-induced diffusion behaviour of binary mixtures of polystyrene particles [227], and removal efficiency of algae via bubbling [225]. Clearly, the DOTM technique is advantageous for unravelling the various membrane fouling phenomenon. The assessment of the efficacy of particle fluidization in mitigating membrane fouling has not benefited from DOTM in part because DOTM has only been applied to horizontal cross-flow membrane modules. This study represents a novel attempt at adapting DOTM for a vertical fluidized bed - membrane module.

The overall goal of this study was to understand the enhancement of critical flux (J_c) of fine particles possible via the fluidization of GAC particles. The specific objectives were: (i) determine the critical flux (J_c) values of a fluidized bed – membrane filtration system using a novel adaptation of DOTM for a vertical membrane module; (ii) assess the impact of various operational parameters (namely, GAC particle diameter (d_p), superficial liquid velocity (U_l) and thereby power requirement (P_r)) on the critical flux (J_c) at various heights of the membrane; and (iii) evaluate the relationship between power requirement (P_r) and overall critical flux ($J_{c,overall}$), and thereby suggest optimal operating conditions.

5.2 Materials and methods

5.2.1 Experimental setup

The schematic of the vertical-DOTM setup used in this study for characterizing the critical flux in a solid-liquid fluidization membrane filtration cell is shown in **Fig. 5-1**. Notably, in contrast to all previous DOTM studies, this was a new configuration of the DOTM instrument in that the focal plane at the membrane-feed interface was vertical rather than horizontal. Specifically, the Zeiss microscope coupled with a camera (Axiocam 105 Color) was oriented 90° relative to previous setups, such that the focal axis was horizontal instead of vertical. An objective lens with 20× magnification was used. Because the fluidized GAC particles reduced the amount of light in the focal axis, an additional floodlight (Olympus, ILK-7A) was placed at the permeate side of the membrane cell. A digital gear pump (Cole Palmer, 75211-35) was used to control the flow rate required for the GAC particle fluidization, while a peristaltic pump (Cole Parmer, 7523-90) controlled the permeate flux. Three pressure transmitters (Cole Palmer; -14.7 – 15 psig) were used to measure the pressure signals at the feed inlet, feed outlet and permeate outlet. The permeate flux was quantified by a digital balance (Mettler-Toledo, PL4002). For each experiment, the pressure and permeate flux data were recorded every 60 s via LabView (National Instruments).

Fig. 5-2 depicts the detailed dimensions of the fluidized bed - membrane filtration cell, which had internal dimensions of 30 mm width by 15 mm depth by 146 mm height. The distributor at the bottom of the cell, made of multiple

layers of metal mesh, provided for more uniform fluidization. Another metal mesh was also positioned at the top of the cell to prevent GAC particle entrainment. The membrane used was an Anopore disk (Whatman, Germany), which was transparent when wetted, with a diameter of 47 mm and nominal pore size of 0.2 μm . A rectangular plastic sheet with dimensions of 54 mm width by 175 mm height was used to secure the membrane in position. Specifically, depending on which of the three heights (namely, $h/H = 0.29, 0.46$ and 0.62 , where h is the height assessed while H is the total membrane cell height of 146 mm) the critical flux was characterized at a square with dimensions of 26 mm by 26 mm which was cut out from the plastic sheet and the membrane was taped onto the plastic sheet. Thereby, the active membrane area used in the critical flux calculation was only the square area of 26 mm by 26 mm. To avoid fracturing the brittle Anopore membrane during the fluidization, the plastic sheet was further attached to a stainless steel spacer (**Fig. 5-2b**) with dimensions of 39 mm width by 161 mm height and uniformly distributed circles with diameters of 2 mm.

Figure 5-1. Schematic of the vertical-DOTM setup.

Figure 5-2. (a) Schematic of the fluidized bed - membrane filtration cell. All dimensions are in units of mm; (b) Photograph of the stainless steel spacer.

5.2.2 Materials

Polystyrene (also known as latex) particles (Sigma Aldrich, Singapore) with particle diameter of 5 μm were used as the model foulant. The volumetric ratio of the polystyrene particles to DI water in the feed was 0.01:100 (i.e., a concentration of 0.01 % v/v), and the feed solution was continuously stirred at 250 rpm using a magnetic stirrer (AS ONE; REXIM RS-1D).

The GAC particles (FILTRASORB 300, Calgon Carbon Corporation) obtained were first sieved using Cole-Parmer U.S. Standard Brass Test Sieves and the Ro-Tap sieve-shaker in order to investigate specific sizes with a narrower particle size distribution (PSD). Similar to previous Chapters 3-4, GAC particles of three diameters (d_p), namely, 1.85 ± 0.15 mm, 1.55 ± 0.15 mm and 1.015 ± 0.165 mm were studied. The concentration of GAC particles was 100 g/L for every experiment.

5.2.3 Determination of critical flux

Critical flux is defined as the flux above which fouling occurs significantly and below which negligible fouling occurs [87]. In the current study, the flux-stepping method was adopted to characterize critical flux. Specifically, each flux was held constant for 10 minutes, at the end of which the flux was increased by 5 $\text{L}/(\text{m}^2 \text{ h})$, and two DOTM images were captured for each flux at the end of the 1st and 10th minute. Care was taken to ensure the images were taken at the center of the membrane each time [228]. In prior work, the critical flux was determined as the flux at which the change in the percent foulant coverage between the two images (dC/dt) exceeded a specific threshold value [229-232].

However, this approach could not be implemented in this study because of the presence of the fluidized GAC particles.

Fig. 5-3 shows two examples of DOTM images of fouled membranes (i.e., at a flux higher than critical flux) in the presence (**Fig. 5-3a**) and absence (**Fig. 5-3b**) of fluidized GAC particles. It should be noted that the deposited polystyrene particles look different particularly in terms of color in **Figs. 5-3a and b**, because of the additional floodlight from the permeate side illuminating the particles in the case of **Fig. 5-3a**. In the absence of GAC particles (**Fig. 5-3b**), dC/dt can be easily derived by binarizing the image and counting the number of dark pixels. Unfortunately, **Fig. 5-3a** shows that the determination of dC/dt is not so straightforward when fluidized GAC particles are present, because the GAC particles appear as foulants (i.e., dark pixels) too during the binarization of the image. Fortunately, in both cases in **Fig. 5-3**, the polystyrene particles deposited on the membrane appeared as discrete particles, hence enabling them to be manually counted. Accordingly, it was possible to determine the percentage change in the number of deposited particles:

$$\frac{dN}{dt} = \frac{N_{10th\ minute} - N_{1st\ minute}}{9\ min} \quad (5-1)$$

Each test was repeated at least two times with new membranes. A plot of dN/dt versus flux identified J_c as the flux at which dN/dt started to rapidly increase.

Figure 5-3. DOTM images of polystyrene particles deposited on the membrane surfaces: (a) in the presence of fluidized GAC particles, and (b) in the absence of GAC particles.

5.2.4 Calculation of power requirement

The power required for fluidizing the GAC particles (P_r ; unit of kW) can be calculated by:

$$P_r = \Delta P_{bed} Q \quad (5-2)$$

where ΔP_{bed} is the pressure drop across the liquid-solid fluidized bed (kPa) and Q is the volumetric flow rate (m^3/s). The volumetric flow rate (Q) has to high enough to enable the particles to be fluidized. The minimum superficial liquid velocity (U_l ; unit of m/s) needed is termed the minimum fluidization velocity (U_{mf} , unit of m/s), at and above which the upward drag force balances the bed weight and the pressure drop across the bed (ΔP_{bed}) plateaus. Specifically, the superficial liquid velocity (U_l) is calculated by dividing the volumetric flow rate (Q) by the cross-sectional area of the fluidization section. ΔP_{bed} can be approximated by:

$$\frac{\Delta P_{bed}}{\Delta Z} = (1 - \varepsilon_{mf})(\rho_p - \rho)g \quad (5-3)$$

where ΔZ is the height of the bed (m), ρ_p is particle density (1480 kg/m³), ρ is water density (1000 kg/m³), g is gravitational acceleration (9.8 m/s²), and ε_{mf} is the voidage at minimum fluidization condition, which can be estimated from the empirical equation [147]:

$$\varepsilon_{mf} = \frac{0.415}{\varphi^{0.483}} \quad (5-4)$$

where φ is particle sphericity. In this study, the particle sphericity (φ) was derived via AutoCAD 2014 from images of 12 GAC particles of various sizes using the formula [233]:

$$\varphi = \frac{2\sqrt{A/\pi}}{d_{cmin}} \quad (5-5)$$

where A is the projected area of the particle and d_{cmin} is the diameter of the minimum circumscribed circle around the particle (d_{cmin}). The GAC particle sphericity (φ) was determined to be 0.834 ± 0.038 , where the value after ‘ $\pm$ ’ represents the standard deviation of φ values. Thereby, **Eq. (5-5)** gives ε_{mf} as 0.453, which can be substituted into **Eq. (5-3)** to obtain ΔP_{bed} then **Eq. (5-2)** to obtain P_r .

5.3 Results and discussion

5.3.1 Minimum fluidization velocity (U_{mf})

The minimum fluidization velocity (U_{mf}) was determined to ensure that sufficient liquid flow was supplied to fluidize the GAC particles. According to Yang [147], in liquid-solid fluidization, the minimum fluidization velocity (U_{mf}) can also be determined as the superficial liquid velocity (U_l) at which bed expansion becomes significant. Accordingly, bed height (Δz) versus superficial liquid velocity (U_l) was plotted in **Fig. 5-4** for the three particle diameters (d_p) investigated (namely, 1.015 mm, 1.55 mm and 1.85 mm). The U_{mf} values, as labeled in **Fig. 5-4**, were determined to be 0.0092 m/s, 0.0083 m/s and 0.0042 m/s for the particle diameters (d_p) of 1.85 mm, 1.55 mm and 1.015 mm, respectively, which agree with studies in previous chapters that determined U_{mf} from plots of pressure drop (ΔP_{bed}) versus superficial liquid velocity (U_l). Another method to estimate U_{mf} is to use the semi-empirical Ergun equation [148]:

$$\frac{\Delta P_{bed}}{\Delta z} = \frac{150U_{mf}\mu(1-\varepsilon_{mf})^2}{\phi^2 d_p^2 \varepsilon_{mf}^3} + \frac{1.75U_{mf}^2 \rho(1-\varepsilon_{mf})}{\phi d_p \varepsilon_{mf}^3} \quad (5-6)$$

where μ is liquid viscosity (0.001 Pa·s). **Eq. (5-5)** gives U_{mf} values of 0.0096 m/s, 0.0074 m/s and 0.0036 m/s for the particle diameters (d_p) of 1.85 mm, 1.55 mm and 1.015 mm, respectively, which also agree approximately (within ~ 15%) with the values obtained experimentally in **Fig. 5-4**.

Figure 5-4. Bed height (Δz) versus superficial liquid velocity (U_l) for GAC particles with diameters (d_p) of 1.85 mm, 1.55 mm and 1.015 mm

5.3.2 Critical flux

Critical flux can be determined from the trans-membrane pressure (TMP), although it is acknowledged to be not as sensitive as direct observation techniques [223, 224]. **Fig. 5-5** shows the trends of permeate flux and TMP with respect to time for $d_p = 1.85$ mm and $3.5 \times U_{mf}$. The TMP changed negligibly within each 10 min duration of constant flux at lower fluxes, but started increasing within the 10 min duration of constant flux when the permeate flux was at and above $45.8 \text{ L}/(\text{m}^2 \text{ h})$. Hence, the critical flux for this system was determined as $45.8 \text{ L}/(\text{m}^2 \text{ h})$ per the TMP method, which is similar to the DOTM value (see below). For other experiments, the critical flux values determined by

the TMP method was either equal to or marginally greater than that determined by DOTM.

Figure 5-5. Trends of permeate flux and trans-membrane pressure (TMP) with respect to time for the fluidized GAC particles with $d_p = 1.85$ mm at $U_l = 0.032$ m/s (3.5 times U_{mf}). The critical flux was determined to be 45.8 L/(m² h) by both the TMP and DOTM methods.

In this study, the more sensitive direct observation method of DOTM was used to investigate critical flux trends. **Fig. 5-6** shows examples of the DOTM images focused on the feed side of the membrane surface used in the determination of critical flux. It is clear that no polystyrene particle deposited below critical flux (**Fig. 5-6a**), polystyrene particles started to deposit at critical flux (**Fig. 5-6b**), while many polystyrene particles deposited above critical flux (**Fig. 5-6c**). Critical flux was determined as the onset of a significant change in the rate of the number of deposited polystyrene particles (dN/dt), as presented in

Eq. (5-1). Fig. 5-7 shows a plot of dN/dt versus flux at a superficial liquid velocity (U_l) of 0.032 m/s and dimensionless height (h/H) of 0.46 for conditions with and without GAC. It is evident that the critical fluxes were 45.8 L/(m² h) and below 5 L/(m² h) for the cases of fluidized GAC with $d_p = 1.85$ mm and when GAC was absent, respectively. It should be noted that the resolution of the flow rate controllable by the permeate pump was limited, thus the critical flux in the absence of GAC could not be precisely determined and was assumed to approximate 5 L/(m²h). These results highlight the dramatic effect of ‘large particle’ fluidization on the polarization of ‘fine particles’.

Figure 5-6. DOTM images at $U_l = 0.032$ m/s (3.5 times U_{mf}) and $h/H = 0.46$ for the fluidized GAC particles with $d_p = 1.85$ mm at the 10th minute of the flux of (a) 15.7 L/(m² h) (i.e., below critical flux), (b) 45.8 L/(m² h) (i.e., at critical flux), and (c) 60.4 L/(m² h) (i.e., above critical flux).

Figure 5-7. Rate of change of the number of deposited polystyrene particles (dN/dt) versus flux at $U_l = 0.032$ m/s and $h/H = 0.46$. Critical fluxes were determined as 45.8 L/(m² h) and 5 L/(m² h) for the cases of fluidized GAC with $d_p = 1.85$ mm and when GAC was absent, respectively.

5.3.3 Influence of various parameters (namely, superficial liquid velocity (U_l), height (h/H) and particle diameter (d_p)) on critical flux (J_c)

Fig. 5-8a presents critical flux (J_c) of the latex as a function of superficial liquid velocity (U_l) for the three GAC particle diameters (d_p) investigated, namely, 1.015, 1.55 and 1.85 mm, at the lowest dimensionless height (h/H) investigated of 0.29. It should be noted that the higher J_c is, the more effective is the fouling mitigation. The range of superficial liquid velocities (U_l) possible for GAC particles with each particle diameter (d_p) was bounded on the lower end by sufficient bed expansion such that the GAC particles were in contact with the membrane and on the higher end by the avoidance of entrainment (i.e., particles

exiting from the top of the membrane cell). **Fig. 5-8b** further shows the ratio of superficial liquid velocity to minimum fluidization velocity (U_l/U_{mf}), which indicate that higher multiples of U_{mf} were required for bed expansion for GAC particles with smaller particle diameters (d_p). Three observations should be noted from **Fig. 5-8a**. Firstly, the critical flux (J_c) values decreased as superficial liquid velocity (U_l) increased. This may appear counter-intuitive at first, based on the effect of crossflow velocity on J_c in the absence of fluidization [224, 232]. However, a closer inspection of the GAC concentration at $h/H = 0.29$ in **Fig. 5-9** indicates that the particle concentration decreased with U_l . As indicated in Chapter 4, the fouling mitigation effectiveness conferred by fluidized GAC is associated with both the particle velocity and particle concentration. Thereby, the negative correlation between J_c and U_l in **Fig. 5-8a** is because, although the particle velocity increased with U_l , particle concentration decreased with U_l at $h/H = 0.29$ (**Fig. 5-9**). Secondly, the highest critical flux (J_c) was surprisingly achieved with the smallest particle ($d_p = 1.015$ mm) at the lowest U_l . Since U_l is directly correlated with power requirement (P_r ; **Eq. (5-2)**), this implies that the lowest power input gave the highest critical flux (J_c) at the lowest portion of the membrane ($h/H = 0.29$). This is again a function of particle diameter (d_p), particle concentration and particle velocity. Thirdly, although the overlap of U_l values was limited among the different particle diameters (d_p), it is shown that, at the same U_l and thus P_r , the larger particle diameters (d_p) tended to give higher J_c values. This suggests the dominance of the particle inertial effect at these conditions. Armed with the surprising results on the relationship between U_l and J_c , the effect of the height of the membrane on fouling mitigation effectiveness

was explored in more detail.

Figure 5-8. Critical flux (J_c) with respect to superficial liquid velocity (U_l) at $h/H = 0.29$ for GAC particles with three particle diameters (d_p) investigated.

Figure 5-9. High-speed video images of fluidized GAC particles with $d_p = 1.85$ mm at the superficial liquid velocities (U_l) of (a) 0.027 m/s (i.e., $U_l/U_{mf} = 3$), and (b) 0.041 m/s (i.e., $U_l/U_{mf} = 4.5$). Images were taken at $h/H = 0.29$.

Fig. 5-10a illustrates the critical flux (J_c) values at various superficial liquid velocities (U_l) as a function of dimensionless height (h/H , where h is the height assessed and H is the total membrane cell height of 146 mm) for the GAC particle diameter (d_p) of 1.85 mm. **Figs. 5-10b and c** show the analogous plots for the GAC particle diameters (d_p) of 1.55 mm and 1.015 mm, respectively. It should be noted that, due to the limited resolution of the permeate pump, J_c was assigned a value of 5 L/(m² h) when it was too low to be accurately determined. Four observations are worth highlighting. Firstly, the J_c values generally decreased as height (h/H) increased, which is due to the decrease in particle concentration with height (h/H). Secondly, the lowest superficial liquid velocity (U_l) was only able to mitigate fouling at the lowest height of $h/H = 0.29$, while only the highest U_l was able to mitigate fouling at the highest height of $h/H = 0.62$. This implies expectedly that power requirement (P_r) was tied to the extent of overall fouling mitigation, specifically in that a higher power input is required to mitigate fouling throughout the entire height of the membrane cell. Thirdly, consistently for all three particle diameters (d_p), the lowest U_l gave the highest J_c

at $h/H = 0.29$. This was observed in **Fig. 5-8** and was attributed to the negative correlation between U_l and particle concentration. Fourthly, at the intermediate height of $h/H = 0.46$, the relationship between U_l and J_c was not clear for the two larger particle diameters (d_p), but J_c increased with U_l for the smallest particle diameter (d_p) of 1.015 mm. This again affirms that an interplay of factors (e.g., local particle concentration, local particle velocity) were at play in the mitigation of fouling by fluidized GAC.

Figure 5-10. Critical flux (J_c) at various heights (h/H) and superficial liquid velocities (U_l) for GAC particles with diameters (d_p) of (a) 1.85 mm, (b) 1.55 mm, and (c) 1.015 mm.

To have a representative critical flux value for the entire membrane cell, a $J_{c,overall}$ value can be calculated based on an arithmetic averaging of the J_c value at each of the three heights investigated. It should be noted that, despite the

bounds of the heights (h/H) characterized being limited by practical constraints of the lab-scale cell, it is clear in **Fig. 5-10** that difference of heights had a significant impact on the local J_c value, particularly in terms of the steep decrease of J_c with height. Therefore, to understand the overall performance of the filtration cell, a parameter to summarize all the local values is needed, which in this case is $J_{c,overall}$. **Fig. 5-11** describes $J_{c,overall}$ as a function of power requirement (P_r) for GAC particles with three particle diameters (d_p) investigated. For each particle diameter (d_p), a higher power input (P_r) expectedly gave a higher $J_{c,overall}$ value. Notably, at the same power input (P_r), the smaller particles gave higher $J_{c,overall}$ values, which agreed with our previous study on the effect of $U_l - U_{mf}$ in Chapter 3. The largest particle required at least 50% more power (P_r) than the smallest particle to confer the same $J_{c,overall}$. At the same power input (P_r), the magnitude of $U_l - U_{mf}$ is greater and the bed expansion is also greater for the smaller particles, thus the smaller particles have more mobility and more contacts with membrane, thereby confer greater scouring efficiency.

Figure 5-11. Overall critical flux ($J_{c,overall}$) versus power requirement with GAC particle diameters of 1.85 mm, 1.55mm, and 1.015 mm.

Since the aim of the fluidized bed membrane process is to control fouling there could be a problem if a significant (upper) region of the membrane was not subject to fluidized particle hydrodynamics. This region could rapidly foul and become ineffective. The only way to avoid this would be to increase U_l or increase bed mass. The result would be increased P_r . Therefore to further examine the contribution of the J_c value at each height (h/H) to the overall $J_{c,overall}$ value, **Fig. 5-12** plots the $J_c/J_{c,overall}$ ratio versus power requirement (P_r). Three observations should be noted. Firstly, the $J_c/J_{c,overall}$ trends are very similar for the three particle diameters (d_p) investigated, which indicate that the relationship between h/H , P_r and J_c are consistent across particle diameters (d_p). Secondly, as power input (P_r) increased, the $J_c/J_{c,overall}$ ratio at each height became more similar. This indicates that the fouling mitigation became more uniform across the membrane height as power input (P_r) increased. **Fig. 5-12c** suggests more

fouling mitigation can be achieved at a lower power (P_r) using the smaller GAC. Thirdly, the $J_c/J_{c,overall}$ ratio generally decreased with height (h/H), which implies that the lower heights (h/H) contributed more to the overall fouling mitigation and is expected since particle concentration decreased with height (h/H).

Figure 5-12. The $J_c/J_{c,overall}$ ratio at various heights (h/H) versus power requirement (P_r) for GAC particles with diameters (d_p) of (a) 1.85 mm, (b) 1.55mm, and (c) 1.015 mm.

Fig. 5-13 summarizes in a three-dimensional (3D) plot the influence of the power requirement (P_r) and GAC particle diameter (d_p) on the overall critical flux ($J_{c,overall}$). Two observations are apparent. Firstly, fouling mitigation was most enhanced (i.e., highest $J_{c,overall}$) when the power input (P_r) and particle diameter (d_p) were the highest. On one hand, a high power input (P_r) led to (i) a more expanded bed which caused more particles to be in contact with more of

the membrane surface, and (ii) greater mobility of the particles which enhanced the scouring effect. On the other hand, a larger particle diameter (d_p) had greater particle inertia in effecting the scouring mechanism when fluidized. Secondly, increasing the power input (P_r) had a greater effect on the overall critical flux ($J_{c,overall}$) of the larger particles, as evident in the steeper slope of $J_{c,overall}$ versus P_r .

Notably, **Fig. 5-13** does not point to an optimal condition. It is not desirable to have to increase P_r if reasonable performance can be obtained at a much lower P_r . The optimal fluidization scenario would give (i) good enhancement of $J_{c,overall}$ (**Fig. 5-11**), (ii) $J_c/J_{c,overall}$ close to unity at all membrane heights (**Fig. 5-12**), and (iii) a modest power requirement (P_r). At the same P_r , although the larger GAC particles were more effective locally (i.e., higher J_c) due to greater particle inertia (**Fig. 5-8**), the smaller GAC particles were more effective overall (i.e., higher $J_{c,overall}$) due to greater bed expansion enabling scouring of more membrane heights (**Fig. 5-11**). To identify the optimal fluidization for membrane fouling mitigation would involve a trade-off between capital cost (related to sustainable flux) and operating cost (related to power (P_r), fouling extent and cleaning).

Figure 5-13. Three-dimensional (3D) plot correlating power requirement (P_r) and GAC particle diameter (d_p) with the overall critical flux ($J_{c,overall}$).

5.4 Conclusions

This study represents the first systematic investigation on the influence of fluidized GAC particles (mm-sized) on critical flux (J_c) of micron-sized particles in microfiltration. The direct observation through the membrane (DOTM) technique was employed. The model foulant used was monodisperse polystyrene particle with particle diameter of 5 μm . The impact of GAC particle diameter (d_p), superficial liquid velocity (U_l) and axial position of the vertically aligned membrane (h/H) on critical flux (J_c) was investigated.

Consistent with previous chapters, both local particle concentration and local particle velocity play an important role in dictating the J_c values. It should also be mentioned that the continuous mechanical scouring action of the GAC particles has been reported to affect the integrity of the membrane, as has been observed by patterns of particle scraping on the membrane surface [139, 169,

234-237]. A recent study has further shown that the extent of damage varied greatly among commercially available membranes, indicating that the choice of membrane is important for the longer-term feasibility [172]. Four highlights are worth noting. Note that the results here may be unique to the foulant type and concentration investigated here. Firstly, the presence of fluidized GAC increased critical flux by an order-of-magnitude relative to that in the absence of GAC (i.e., only tangential liquid shear). Secondly, the overall critical flux ($J_{c,overall}$), based averaging the J_c values across the reactor height, increased with power input (P_r) for the same particle diameter (d_p), and decreased with particle diameter (d_p) at the same power input (P_r). The greatest $J_{c,overall}$ value was given by the combination of largest particle diameter (d_p) and highest power input (P_r), and increasing the power input (P_r) had a greater effect on the overall critical flux ($J_{c,overall}$) of the larger particles than the smaller ones. Thirdly, at the same P_r , although the larger GAC particles were more effective locally due to greater particle inertia, the smaller GAC particles were more effective overall due to greater bed expansion enabling scouring of more membrane heights. Fourthly, a higher power input (P_r) was required for sufficient bed expansion to allow for more uniform critical flux (J_c) values throughout the membrane height.

Chapter 6 Effect of bubble characteristics on critical flux in the microfiltration of particulate foulants⁴

6.1 Introduction

The beneficial impact of bubbles on membrane filtration processes has been reported as early as in 1989 by Imasaka et al. [77], who injected the produced methane of an anaerobic digester into the tubular ceramic membrane modules, and found that membrane fouling was reduced at a reasonable energy cost. Since then, significant research on the use of bubbly flow particularly on aerobic membrane bioreactors has flourished [6, 78-85]. Previous studies have indicated that bubbles are beneficial due to the disruption of the polarization layer [90], creation of wakes and secondary flows [91, 92], and the conferment of shear stress [93, 94], all of which contribute towards membrane fouling mitigation. Besides, it was also found that the presence of bubbles enhanced the back-wash efficiency [95, 96]. Bubble flow patterns, which are inevitably tied to the effectiveness of fouling mitigation have been categorized into the following according to gas flow rates [6, 85, 89, 98]: (i) bubble flow, whereby small discrete bubbles are uniformly distributed in the liquid phase; (ii) cap-bubble flow, whereby flattened and distorted bubbles exist due to the confinement by the walls; (iii) slug flow, whereby each bubble size spans more than 75% of the width of

4. The content in this chapter has been published as:

Jingwei Wang, Anthony G. Fane, Jia Wei Chew. Effect of bubble characteristics on critical flux in the microfiltration of particulate foulants. *Journal of Membrane Science*, volume 535, 1 August 2017, Pages 279-293.

the filtration module; (iv) churn flow, whereby the slugs are narrower and more distorted, and the flow is more chaotic, frothy and disordered; (v) annular flow, whereby the core and annulus are occupied by the gas and liquid phases, respectively. To optimize the fouling mitigation by bubbles, the influence of gas flow rates [78, 97, 99-101], bubble size and frequency [82, 102-105], and bubble shapes and orientations [93] have been investigated. Some studies found that larger bubbles were more effective at mitigating fouling [82, 238] due to the larger shear stress, larger wake regions, stronger secondary flow and the reduction in the risk of foaming. On the contrary, some other studies have found that at the same volumetric flowrate smaller bubbles were better [239, 240]. The contradictory conclusions suggest that more factors are at play other than bubble size alone.

The well-acknowledged advantages of introducing bubbles into membrane-based filtration processes necessitates a more mechanistic understanding of the relationship between the bubble characteristics and critical flux. This forms the goal of the current study. The direct observation through the membrane (DOTM) technique was used to determine the critical flux of a feed containing micron-sized polystyrene particles, while a high-speed video camera was used to characterize the bubble behavior. The specific questions addressed were: (i) how do the local critical flux values and local bubble characteristics (namely, velocity, bubble-membrane contact area, number of bubbles, bubble momentum) vary with height along the membrane, gas flow rate, liquid flow rate, and energy input; (ii) how do the local critical flux values correlate with the local bubble characteristics; (iii) how does the overall critical flux (average of the local

values) relate to the flow rates and energy input; and (iv) how do the extent of fouling mitigation and energy cost compare between unsteady-state achieved by the employment of bubbles versus fluidized granular activated carbon (GAC) particles in Chapter 5.

6.2. Materials and methods

6.2.1 Experimental setup

The schematic of the vertical DOTM setup used to measure the critical flux during microfiltration whereby bubbling was employed as an unsteady-state shear means of mitigating membrane fouling is shown in **Fig. 6-1**. Specifically, as opposed to the conventional DOTM setups [224] whereby the membrane and hence the focal plane is horizontal, the membrane was oriented vertically in this case to facilitate the bubbling. This is similar to that in Chapter 5's study on the improvement of the critical flux of micron-sized particulate foulants when millimeter-sized granular activated carbon (GAC) particles were fluidized. The key component of the DOTM setup is the Zeiss microscope coupled with a color video camera (Axiocam 105 color) to focus through the permeate and the membrane such that the focal plane is on the feed-membrane interface.

The feed suspension, which was continuously stirred at 250 rpm using a magnetic stirrer (AS ONE; REXIM RS-1D), was circulated by a gear pump (Cole-Parmer, 75211-15), and the liquid flow rate (Q_l) was measured by a variable-area flowmeter (Dwyer, 0.1-1.25 L/min). The liquid flow rate (Q_l) ranged between 0.3 and 1.0 L/min; the lower bound was limited by the sensitivity of the flowmeter while the upper bound was because higher pressures restricted

the injection of bubbles. Bubbles were generated by an air pump (Hiblow, HP-20, Japan), with the gas flow rate measured by a variable-area flowmeter (Dwyer, 0.2-2.5 L/min). Gas flow rate (Q_g) values were controlled between 0.2 - 1.0 L/min, whereby the lowest Q_g was such that sufficient bubbles were generated for characterization, while the highest was limited by the upper bound of the flow meter. A Y-branch fitting combined the liquid and gas phases upstream of the membrane filtration cell. A peristaltic pump (Cole-Parmer, 7523-90) was used to provide the suction for the permeate flow, with the permeate flux quantified by a digital balance (Mettler-Toledo, PL4002). Three pressure transmitters (Cole-Parmer) each in the range of -101.4-103.5 kPa were used to measure the pressure at the feed inlet, feed outlet and permeate outlet. For each of the experiments, the pressure and permeate flux values were recorded every 60 s via LabView (National Instrument).

The geometry of the membrane filtration cell was similar to that of our previous study in Chapter 5, specifically with a feed channel with dimensions of 30 mm width by 5 mm depth by 146 mm height (**Fig. 6-2a**) and a permeate channel with dimensions of 40 mm width by 2 mm depth by 162 mm height. In particular, for the feed chamber, an inlet section with dimensions of 6 mm height by 30 mm width by 120 mm depth allows for more uniform distribution of the bubbles upwards through the cell (**Fig. 6-2b**). The membrane used was an Anopore disk membrane (Whatman, Germany), with a diameter of 47 mm and nominal pore size of 0.2 μm ; the transparency of the membrane when wetted is key to the DOTM technique. To avoid fracturing the brittle membrane, a plastic sheet (with dimensions of 54 mm width by 175 mm height), on which the

membrane was attached, was further attached to a rectangular stainless steel support (with dimensions of 39 mm width by 161 mm height and uniformly distributed circles with diameters of 2 mm through which fouling was observed via the microscope), as shown in **Fig. 6-2c**. Specifically regarding the attachment of the membrane to the plastic sheet, depending on the height at which fouling was characterized, a square area of 26 mm by 26 mm was cut out from the plastic sheet and the membrane taped on to expose an active membrane area of 26 mm by 26 mm. Three heights along the membrane cell were characterized, namely, at h/H (whereby h is the height assessed and H is the total cell height of 146 mm) of 0.29, 0.5 and 0.71.

Finally, a high-speed camera (Olympus i-SPEED LT) was used to capture the dynamics of the bubbles in the lateral center of each height characterized. Each recording was at a frequency of 300 frames per second (fps) and lasted 5 - 10 s.

6.2.2 Particulate foulant

The model foulant used was polystyrene (latex) particles, with a particle diameter of 5 μm , purchased from Sigma Aldrich (Singapore). The concentration used was 0.01 % (i.e., volumetric ratio of the polystyrene particles to DI water was 0.01:100), which corresponds to the lower range of total suspended solids (TSS) in real wastewater [241].

Figure 6-1. Schematic of the vertical DOTM setup.

Figure 6-2. (a) Schematic of the membrane filtration cell, with the circled region depicted in the photo in (b); (b) Photograph of the inlet of the feed chamber to distribute the bubbles; (c) components of membrane filtration cell.

6.2.3 Determination of local critical flux ($J_{c,local}$)

Critical flux is defined as the flux below which negligible membrane fouling occurs and above which significant fouling occurs [87]. DOTM has been proven to be useful in the determination of critical flux in two-phase flow systems, namely, in the presence of fluidized granular activated carbon (GAC) particles (chapter 5). Since the study here also involved two-phase flow (in this case,

bubbles), DOTM was also employed. The flux-stepping protocol was similar to that adopted in previous chapter 5. Specifically, each permeate flux was held constant for 10 minutes, followed by the increment of the permeate flux by 10 L/(m²h), and the flux-stepping continued until fouling was clearly visible under the microscope. At each permeate flux, two sets of videos each lasting 5 s and at a frequency of 5 frames per second (fps) were taken at the end of the 1st and 10th minutes. Examples of sequential images taken 0.2 s apart of the membrane area assessed are presented in **Fig. 6-3** at a permeate flux below the local critical flux ($J_{c,local}$). **Fig. 6-3a** shows the approach of a bubble at the bottom of the image. In **Fig. 6-3b**, the bottom part of the image below the bubble (i.e., black line) has less deposits than that in the upper part of the image above the bubble. This provides evidence of the ability of the bubble to shear deposits off the membrane surface to mitigate membrane fouling. **Fig. 6-3c** shows a less distinct black line, which indicates the bubble is not in direct contact with the membrane.

The 25 images of each video clip were averaged to determine the number of polystyrene particles deposited (N). At each permeate flux, the rate of change of N between the 1st and 10th minutes was evaluated using **Eq. (6-1)**. Because dN/dt is positively correlated to the extent of fouling, a plot of dN/dt versus permeate flux can be used to determine the critical flux (J_c).

$$\frac{dN}{dt} (\text{min}^{-1}) = \frac{N_{10^{\text{th}} \text{ minute}} - N_{1^{\text{st}} \text{ minute}}}{9} \quad (6-1)$$

Figure 6-3. Sequential images 0.2 s apart of the membrane area assessed at $h/H = 0.29$. The black lines represent the contact areas between the bubble and the membrane. The permeate flux was $109 \text{ L}/(\text{m}^2 \text{ h})$ (which was below the local critical flux ($J_{c,local}$)), while the liquid (Q_l) and gas (Q_g) flow rates were respectively $0.6 \text{ L}/\text{min}$ and $0.3 \text{ L}/\text{min}$.

6.2.4 Characterization of the bubbles

In previous chapter 4, the hydrodynamics of the fluidized GAC was found to be directly related to the extent of membrane fouling mitigation. Along the same lines, bubble characteristics are expected to be tied to fouling mitigation. Therefore, bubble velocity (V_b), bubble area (A_b) and number of bubbles (N_b) were quantified in this study via high-speed video imaging (300 fps). Images were extracted from the videos and image analysis conducted using Image J. Examples of two sequential images 0.0033 s apart are displayed in **Fig. 6-4**. It is clear that the bubbles have non-uniform shapes, and appear flattened presumably due to the limited depth of the feed channel.

To compute bubble velocity (V_b), the center of each bubble in focus was approximated and designated with co-ordinates (x , y) via Image J, then the distance between the same bubble in two sequential images was normalized with respect to the time difference between the images. As an example, **Table 6-1** lists the coordinates obtained from the four bubbles in **Fig. 6-4a** (x_1 , y_1) and **Fig. 6-4b** (x_2 , y_2). Bubble velocity (V_b) was calculated using the following equation:

$$V_b = \frac{\sqrt{(x_1-x_2)^2+(y_1-y_2)^2}}{0.0033} \quad (6-2)$$

It should be noted that the direction of V_b is likely not crucial, since bubble movement in any direction presumably confers similar shearing effects. The mean bubble velocity ($V_{b,rms}$) can be calculated to summarize the bubble velocity values at a given position:

$$V_{b,rms} = \sqrt{\sum V_b^2 P(V)} \quad (6-3)$$

where $P(V)$ stands for the probability of a bubble having a bubble velocity of V_b . Furthermore, the width of the probability distribution of bubble velocity (V_b) can be represented by the standard deviation (σ):

$$\sigma = \sqrt{\sum (V_b - V_{b,rms})^2 P(V)} \quad (6-4)$$

The area of each bubble can be derived using Image J, while the number of bubbles can be manually counted. The normalized bubble area (A_b) and normalized number of bubbles (N_b) are defined here as the sum of the area occupied by the bubbles and the number of bubbles, respectively, in each image normalized with respect to the area evaluated (i.e., the image dimensions of 20.7 mm by 15.52 mm). To quantify each of these bubble parameters (i.e., V_b , A_b , and N_b), at least 300 bubbles were evaluated.

Figure 6-4. Sequential images taken 0.0033 s apart of the bubbles at $h/H = 0.71$. The liquid flow rate (Q_l) was 1.0 L/min, gas flow rate (Q_g) was 0.5 L/min, and permeate flux was 0 L/(m²h).

Table 6-1. Bubble parameters obtained from Fig. 6-4.

Bubbles	$t = 0$ s (Fig. 6-4a)				$t = 0.0033$ s (Fig. 6-4b)				
	Bubble coordinates		Area of each bubble	Bubble area per unit membrane area	Bubble coordinates		Area of each bubble	Bubble area per unit membrane area	Bubble velocity
	x_1	y_1	mm ²	A_b (mm ² /mm ²)	x_2	y_2	mm ²	A_b (mm ² /mm ²)	V_b (mm/s)
1	3.01	13.19	20.86	0.279	3.40	12.25	23.25	0.291	303.5
2	7.32	7.59	26.02		7.32	6.43	26.59		348.3
3	17.36	9.33	31.34		17.09	7.91	31.16		433.6
4	14.98	13.98	11.64		15.27	12.75	12.55		377.9
									0.01245

6.3. Results and Discussion

6.3.1. Local critical flux ($J_{c,local}$)

Examples of the images at permeate fluxes below and above the local critical flux at the operation condition of $Q_l = 0.6$ L/min and $Q_g = 0.3$ L/min are shown in **Fig. 6-5**. It is clear that the membrane at a permeate flux below the local critical flux (**Fig. 6-5a**) was relatively free of deposits compared to that above the

local critical flux (**Fig. 6-5c**). Furthermore, **Fig. 6-5b** shows an image at the determined local critical flux ($J_{c,local}$), which was ascertained by the rate of change of the number of deposited polystyrene particles on the membrane (dN/dt ; **Eq. (6-1)**). **Fig. 6-6a** presents the plot of dN/dt versus permeate flux. The local critical flux ($J_{c,local}$) was determined as the flux at the onset of a significant change in the dN/dt values, which in this case was 139.1 L/(m² h). Notably, the conventional means of measuring the trans-membrane pressure (TMP) to determine the $J_{c,local}$ value was not effective here, as shown in **Fig. 6-6b**, whereby the TMP remained approximately constant even at a permeate flux of 30 L/(m² h) above the $J_{c,local}$ value obtained via DOTM (**Fig. 6-6a**). This is probably because an increasing TMP profile requires a significant cake resistance to develop during the time increment. For the particle size and the low concentration used the resistance build up would be slow when flux is relatively close to the critical flux.

An interesting phenomenon was that, at permeate fluxes above $J_{c,local}$, small micron-sized bubbles also deposited on the membrane along with the foulant particles (i.e., polystyrene). **Fig. 6-7**, which portrays two sequential images (5 s apart) taken at a permeate flux above $J_{c,local}$, shows the deposition of small bubbles on the membrane. Similar to the deposited foulant particles, the deposited bubbles were not removed from the membrane as the larger bubbles sheared the membrane surface to mitigate fouling. Bubbles deposited on the lumen of hollow fiber membranes have been reported to cause fluctuations in the transmembrane pressure (TMP) [242]. The deposited bubbles presumably decreased the effective membrane area and increased membrane resistance. If the

bubbles coalesced the larger bubbles formed may get displaced; however, smaller bubbles could enter the pores in microfiltration applications and potentially become trapped in the membrane structure or even appear in the permeate. Further studies beyond the current scope on such effects are needed to clarify.

Figure 6-5. Images captured at the end of the 10th minute at the targeted permeate fluxes, which correspond to (a) below the local critical flux ($J_{c,local}$), (b) the local critical flux ($J_{c,local}$), and (c) above the local critical flux ($J_{c,local}$). The liquid flow rate (Q_l) was 0.6 L/min and the gas flow rate (Q_g) was 0.3 L/min.

Figure 6-6. (a) Plot of the rate of change of the number of deposited polystyrene particles (dN/dt) with respect to permeate flux to determine the local critical flux ($J_{c,local}$), which was 139.1 L/(m² h) in this case; (b) Trends of trans-membrane pressure (TMP) and permeate flux with respect to time. The liquid flow rate (Q_l) was 0.6 L/min, the gas flow rate (Q_g) was 0.3 L/min, and the height (h/H) was 0.29.

Figure 6-7. Two sequential images 5 s apart showing the deposition of bubbles on the membrane. The permeate flux was 197.8 L/(m² h) (i.e., above $J_{c,local}$), Q_l = 0.6 L/min, Q_g = 1.0 L/min and h/H = 0.29

6.3.2. Effect of operation conditions on local critical flux ($J_{c,local}$)

Fig. 6-8 illustrates the local critical flux ($J_{c,local}$) versus dimensionless heights along the membrane cell (h/H , where h is the assessed height and H is the total membrane cell height of 146 mm) at various gas flow rates (Q_g) and liquid flow rates (Q_l). Two observations are apparent. Firstly, the local critical flux ($J_{c,local}$) generally increased with height (h/H), which implies that the extent of fouling mitigation by the bubbles improved with height. This is contrary to that for the fluidized GAC system investigated in Chapter 5, in which $J_{c,local}$ generally decreased with h/H due to the reduced local GAC concentration with height. It should also be noted that the $J_{c,local}$ values here of above 100 L/(m² h) are much higher than that for the fluidized GAC system, whereby the highest $J_{c,local}$ value was 85.9 L/(m²h) (Chapter 5). Secondly, while an increase in the gas flow rate

(Q_g ; **Fig. 6-8a**) tended to increase the local critical flux ($J_{c,local}$), the increase in liquid flow rate (Q_l ; **Fig. 6-8b**) had an insignificant influence on $J_{c,local}$. This highlights that the enhancement of fouling mitigation by an increase in the number, size and velocity of the bubbles presumably associated with the increase of Q_g was more effective than the increase in liquid shear tied to the increase in Q_l . The dominance of the bubble flow is also interesting in the context of the submerged MBR, where liquid flow is not pumped per se but may experience a degree of circulation due to bubble-induced flow. The bubble characteristics investigated in the following sections will serve to shed light on this effect.

Figure 6-8. Local critical flux ($J_{c,local}$) versus dimensionless heights (h/H) at (a) a constant liquid flow rate (Q_l) of 0.6 L/min and different gas flow rates (Q_g); and (b) a constant gas flow rate (Q_g) of 0.5 L/min and different liquid flow rates (Q_l)

6.3.3. Influence of operating conditions on bubble behavior

Fig. 6-9 shows the probability distributions of the bubble velocity (V_b) at three dimensionless heights (h/H) and three gas flow rates (Q_g). The x - and y -axes ranges of all three sub-plots are kept the same for easier comparison. Two observations are worth highlighting. Firstly, the highest gas flow rate (i.e., $Q_g = 1.0$ L/min) generally gave the widest probability distribution of bubble velocity

(V_b), which implies the presence of a wider range of bubble velocities (V_b) compared to the lower gas flow rates (Q_g). However, the mean bubble velocity seems to be approximately similar at each height regardless of gas flow rate (Q_g). Secondly, higher bubble velocities (V_b) were present at higher heights (h/H), which widened the probability distribution with height (h/H). The mean bubble velocity also increased with height. This increased prevalence of higher bubble velocities (V_b) with height seems to correlate well with the increased local critical flux ($J_{c,local}$) values at higher heights (**Fig. 6-8**).

Figure 6-9. Probability distributions of bubble velocity (V_b) at three dimensionless heights (namely, $h/H = 0.29$, 0.50 and 0.71).

Fig. 6-10 summarizes the bubble characteristics with respect to height (h/H). The first column represents the effect of gas flow rate (Q_g) at a constant Q_l of 0.6 L/min, while the second column represents the influence of liquid flow rate (Q_l) at a constant Q_g of 0.5 L/min. The first row displays mean bubble velocity ($V_{b,rms}$; **Eq. (6-3)**), the second bubble area per unit membrane area (A_b) and the third the number of bubbles per unit membrane area (N_b). Each error bar represents the standard deviation of the corresponding distribution. Three observations are noted. Firstly, **Figs. 6-10a and e** shows that $V_{b,rms}$ generally increased with height (h/H). Although the distributions of bubble velocity (V_b) tended to overlap in the range of gas (Q_g) and liquid (Q_l) flow rates investigated, the highest gas flow rate (Q_g) of 1.0 L/min clearly gave the highest bubble velocities (V_b) at all heights, while the relationship between liquid flow rate (Q_l) and the highest bubbles velocities (V_b) was not clear. Secondly, the mean bubble area per unit membrane area (A_b) remained approximately constant with height, and increased with gas flow rate (Q_g ; **Fig. 6-10b**) but not with liquid flow rate (Q_l ; **Fig. 6-10f**). This indicates that the total bubble-membrane contact area was uniform along the membrane height, and that increasing the contact can be facilitated by increasing the gas flow rate (Q_g) but not the liquid flow rate (Q_l). Thirdly, regarding the number of bubbles per unit membrane area (N_b), the relationships are straightforward in **Fig. 6-10g** in that N_b decreased as height increased due to bubble coalescence with height (Appendix) and N_b increased as liquid flow rate (Q_l) increased presumably due to enhanced bubble coalescence. However, with respect to gas flow rate (Q_g ; **Fig. 6-10c**), (i) N_b decreased as height increased, except for the highest gas flow rate (Q_g) of 1.0 L/min; and (ii) N_b

decreased with gas flow rate (Q_g) at the lowest height (h/H), but increased with gas flow rate (Q_g) at the uppermost height (h/H). The reason for this peculiar trend is revealed in **Fig. 6-10d**, which shows that the mean area per bubble (A_b/N_b) increased with gas flow rate (Q_g) at all heights for the two lower gas flow rates (Q_g) but was comparatively constant with height for the highest gas flow rate (Q_g). Coupling **Figs. 6-10c and d** indicates that the highest gas flow rate (Q_g) led to a slug flow regime, whereby the bubbles are large enough to traverse the cross-section of the membrane cell, thereby A_b , N_b and A_b/N_b were relatively invariant with height (h/H).

Figure 6-10. Effect of gas flow rate (Q_g) with respect to height (h/H) at a constant Q_l of 0.6 L/min on: (a) mean bubble velocity ($V_{b,rms}$), (b) bubble area per unit area assessed (A_b), (c) number of bubbles per unit area assessed (N_b) and (d) mean area per bubble (A_b/N_b). Effect of liquid flow rate (Q_l) with respect to height (h/H) at a constant Q_g of 0.5 L/min on: (e) mean bubble velocity ($V_{b,rms}$), (f) bubble area per unit area assessed (A_b), (g) number of bubbles per unit area assessed (N_b), and (h) mean area per bubble (A_b/N_b).

6.3.4. Correlating bubble characteristics with membrane fouling

Since fluidized particle momentum was shown in Chapter 4 to be closely associated with the extent of fouling mitigation, the bubble momentum was similarly investigated here. Bubble momentum (P_b) can be calculated by:

$$P_b = m_b V_b = A_b d_c \rho_{air} V_b \quad (6-5)$$

where m_b is bubble mass, d_c is the depth of the cell (0.005 m) and ρ_{air} is air density (1.225 kg/m³). Since gas flow rate (Q_g) has a greater influence on the bubble parameters (**Fig. 6-10**) and local critical flux ($J_{c,local}$; **Fig. 6-8**), the relationship between gas flow rate (Q_g) and bubble momentum (P_b) is evaluated here. **Fig. 6-11** presents the plot of bubble momentum (P_b) as a function of gas flow rate (Q_g) at a constant liquid flow rate (Q_l) of 0.6 L/min at three heights along the membrane (h/H). It is clear that bubble momentum (P_b) increased with both gas flow rate (Q_g) and height (h/H), which indicates a positive correlation between bubble momentum (P_b) and local critical flux ($J_{c,local}$; **Fig. 6-8**).

Figure 6-11. Bubble momentum (P_b) versus gas flow rate (Q_g) at various dimensionless heights (h/H) at a constant liquid flow rate (Q_l) of 0.6 L/min.

Previous studies have found links between flux enhancement and shear stresses, particularly in terms of standard deviation, caused by bubbles [105, 243]. To further such understanding, **Fig. 6-12** describes the relationship between the local critical flux ($J_{c,local}$) values and the bubble behavioral parameters, namely, mean bubble velocity ($V_{b,rms}$; **Fig. 6-12a**), bubble area per unit membrane area (A_b ; **Fig. 6-12b**), number of bubbles per unit membrane area (N_b ; **Fig. 6-12c**), mean area per bubble (A_b/N_b ; **Fig. 6-12d**) and bubble momentum (P_b ; **Fig. 6-12e**). Each horizontal error bar represents the standard deviation of the distribution, while each vertical error bar represents the span of the $J_{c,local}$ values measured. Bubble momentum (P_b ; **Fig. 6-12e**) gave a clear positive correlation with respect to the local critical flux ($J_{c,local}$) values, which agrees with the trends for particle momentum in Chapter 4 on fluidized GAC and indicates that P_b is a key mechanism underlying the fouling mitigation. In addition, the mean area per

bubble (A_b/N_b ; **Fig. 6-12d**) displayed also a positive correlation with respect to the local critical flux ($J_{c,local}$) values, which suggests that larger bubbles provided more effective fouling mitigation. Both mean bubble velocity ($V_{b,rms}$; **Fig. 6-12a**) and bubble area per unit membrane area (A_b ; **Fig. 6-12b**) exhibited weaker but generally positive relationships with the local critical flux ($J_{c,local}$) values. However, the number of bubbles per unit membrane area (N_b ; **Fig. 6-12b**) had a slight negative correlation with respect to local critical flux ($J_{c,local}$). This is presumably tied to bubble coalescence, specifically in that a smaller number of bubbles implies coalesced, larger bubbles, which had larger $V_{b,rms}$ and A_b values, which in turn are shown to be positively related to the local critical flux ($J_{c,local}$). It should be noted that a higher frequency of smaller bubbles (i.e., higher N_b coupled with lower A_b/N_b) enabled by either baffles [244] or nozzle design [105] has been reported to contribute towards better fouling mitigation (i.e., higher $J_{c,local}$), although the effect of each parameter in **Figs. 6-12c and d** did not quite agree. This suggests an interplay of various bubble characteristics that warrant further study.

Figure 6-12. Correlating the local critical flux ($J_{c,local}$) with (a) mean bubble velocity ($V_{b,rms}$), (b) bubble area per unit membrane area (A_b), (c) number of bubbles per unit membrane area (N_b), (d) mean area per bubble (A_b/N_b), and (e) bubble momentum (P_b).

6.3.5. Bubble flow patterns

The flow patterns of the bubbles, which are known to vary depending on the ratio of the gas (Q_g) and liquid (Q_l) flow rates and reactor design, inevitably affects the mitigation of membrane fouling [6, 85, 98]. **Fig. 6-13** displays the bubble flow patterns at a constant liquid flow rate (Q_l) of 0.6 L/min. The top and bottom rows present the upper ($h/H = 0.53 - 1$) and lower sections ($h/H = 0 -$

0.45), respectively, of the membrane cell. The three columns reflect the three gas flow rates (Q_g) of 0.3, 0.5 and 1.0 L/min. It is clear that the bubbles became larger as the height (h/H) or gas flow rate (Q_g) increased. Secondly, the bubbles were most spherical at the lowest gas flow rate (Q_g) of 0.3 L/min, became more distorted at the intermediate gas flow rate and then slugs at the highest gas flow rate (Q_g) of 1.0 L/min. This suggests the dominance of bubble coalescence at higher gas flow rates (Q_g). The trajectories of bubbles leading to coalescence and sequential images of bubble coalescence are provided in **Fig. 6-14 and 6-15**, which indicates the well-reported migration of bubbles towards the center [244, 245]. The motions of three bubbles (labelled Bubble 1, 2 and 3) at the bottom of the membrane cell were tracked using Image J, and the trajectories leading up to their coalescence into a larger bubble are presented in **Fig. 6-14**. It is clear that the bubbles do not move vertically upwards, but swerve around in the cell. The coalescence behavior was such that Bubbles 1 and 2 coalesce first lower in the cell, before the coalesced bubble (Bubble 1+2) further coalesced with Bubble 3. **Fig. 6-15** further illustrates the progressive coalescence of two bubbles through a sequence of images taken 0.0033 s apart. Two smaller, approximately spherical bubbles met (**Fig. 6-15a**) initially, then coalesced into a larger, ellipsoidal bubble (**Figs. 6-15b and c**), which in turn became a spherical bubble over time (**Figs. 6-15d - h**). Higher gas flow rates (Q_g) are acknowledged to be associated with better fouling mitigation [6, 238], which implies that bubble coalescence and larger bubbles are beneficial. Larger bubbles have a greater mass and a higher bubble velocity, hence greater momentum (P_b ; **Eq. (6-5)**) and enhanced fouling mitigation capability, as reflected in **Fig. 6-12d**.

Figure 6-13. Bubble flow patterns in the microfiltration cell at $Q_l = 0.6$ L/min. The top and bottom rows present the upper ($h/H = 0.53 - 1$) and lower sections ($h/H = 0 - 0.45$), respectively, of the membrane cell.

Figure 6-14. Example of the trajectory of three bubbles in the cell at $Q_g = 0.6$ L/min and $Q_g = 0.3$ L/min. h/H is the dimensionless height, whereby h is the height assessed and H is the total cell height of 146 mm; and w/W is the dimensionless width whereby w is the width assessed and W is the total cell width of 54 mm.

Figure 6-15. Sequential images 0.0033 s apart of the bubble coalescence phenomenon.

In gas-liquid systems, a ratio representing the dominance of the gas flow rate (Q_g) with respect to the total flow rates (i.e., sum of Q_g and Q_l) is typically expressed as:

$$\theta = \frac{Q_g}{Q_g + Q_l} \quad (6-6)$$

To understand how the gas flow rate (Q_g) impacts critical flux, **Fig. 6-16** shows a plot of overall critical flux ($J_{c,overall}$), which is an arithmetic average of the $J_{c,local}$ values at each of the three heights (h/H) investigated, versus θ (**Eq. (6-6)**). The approximately linear increase of $J_{c,overall}$ with θ highlights the dominance of the gas phase in promoting fouling mitigation.

Figure 6-16. Overall critical flux ($J_{c,overall}$) versus gas to total flow rates ratio (θ ; **Eq. (6-6)**).

6.3.6. Energy consumption

The energy cost for inducing bubbles in such systems for mitigating membrane fouling is non-negligible, hence it is necessary to quantify if the benefits justify the energy cost. The power required can be calculated by [246]:

$$P_r = \frac{\alpha}{\alpha-1} p_{atm} Q_g \left[\left(\frac{p_i}{p_{atm}} \right)^{(\alpha-1)/\gamma} - 1 \right] \quad (6-7)$$

where P_r is the power required (kW), α is 1.4 for air, p_i is the pressure at inlet (kPa), and p_{atm} is atmospheric pressure (101 kPa).

Fig. 6-17 plots the ratio of local to overall critical flux ($J_{c,local}/J_{c,overall}$) with respect to power required (P_r) to understand the uniformity of fouling mitigation along the membrane. The closer the $J_{c,local}/J_{c,overall}$ value is to unity, the more uniform is the extent of fouling mitigation. Three interesting observations in **Fig. 6-17** which contrast the results for the fluidized GAC system in Chapter 5 are that (i) the $J_{c,local}/J_{c,overall}$ values are generally closer to unity than that for fluidized GAC, which indicates the fouling mitigation was more uniform across the membrane for the bubbling system; (ii) the greatest height (h/H) investigated of 0.71 had the largest $J_{c,local}/J_{c,overall}$ values, which indicates that fouling mitigation was most effective at this height and is contrary to the results for the fluidized GAC system (wherein the greatest $J_{c,local}/J_{c,overall}$ values were at the lowest height investigated); and (iii) the increase in power requirement (P_r) had limited impact on the $J_{c,local}/J_{c,overall}$ values, which is contrary to the results for the fluidized GAC system, wherein the $J_{c,local}/J_{c,overall}$ became closer to unity as P_r increased.

Fig. 6-18 displays the overall critical flux ($J_{c,overall}$) versus power requirement (P_r) for both the bubbling and fluidized GAC systems (Chapter 5). The inset figure magnifies the trends of the fluidized GAC particles, which shows that smaller GAC particles gave higher overall critical fluxes ($J_{c,overall}$) at the same P_r . Two observations are noteworthy. Firstly, it is clear that the $J_{c,overall}$ and P_r values for the two systems do not overlap, which agree with an earlier review on unsteady-state shear means for fouling mitigation [1] in that the various methods offer different ranges of enhancement and shear values. While it was not possible to operate the bubbling system studied here at lower power (P_r) to compare directly with the fluidized GAC system due to the limited range of the flowmeters in the current setup (**Fig. 6-1**), it was not possible to increase the power (P_r) for the fluidized GAC setup due to entrainment. Secondly, increasing power (P_r) monotonically increased the overall critical flux ($J_{c,overall}$) values for the bubbling system, but the GAC particle diameter used was an additional factor that dictated the interplay between $J_{c,overall}$ and P_r , such that at a given power input higher $J_{c,overall}$ values were achieved by fluidizing smaller GAC particles.

Figure 6-17. Ratio of local to overall critical flux critical flux ($J_{c,local}/J_{c,overall}$) versus power requirement (P_r) at the three dimensionless heights (h/H) investigated.

Figure 6-18. Overall critical flux ($J_{c,overall}$) versus power requirement (P_r) for microfiltration systems employing bubbling and fluidized GAC particles as means of membrane fouling mitigation.

Fouling control by two-phase flow is widely implemented in membrane bioreactors (MBRs). Such unsteady-state shear means are acknowledged to be more energy-efficient than the steady-state ones [1], hence are more promising as higher fluxes are made possible as membrane permeability improves. Gas bubbling is one of the most popularly employed methods of membrane fouling mitigation, and fluidized GAC particles has more recently been adopted. A comparison is carried out here between the gas-liquid (i.e., bubbling) and solid-liquid (i.e., fluidized GAC) systems:

- (i) Whereas the local critical flux ($J_{c,local}$) increased with height for the bubbling system, it decreased with height for the fluidized GAC system (Chapter 5).
- (ii) The hydrodynamics of both the bubbles and GAC particles (Chapter 4 and 5) in terms of velocity, concentration and momentum were shown to be positively correlated to critical flux (J_c). Of the parameters characterized, momentum was shown in both cases to have the strongest relationship to the local critical flux ($J_{c,local}$) values.
- (iii) An increase in energy input generally enhanced the overall critical flux ($J_{c,overall}$) values for both systems.
- (iv) For the bubbling system, increasing the critical flux seemed to involve more simply an increase in the gas flow rate (Q_g). However, for the fluidized GAC system, optimization of the particle size, particle concentration and liquid flow rate were necessary (Chapter 3 - 5).

- (v) For the bubbling system, the ratio of local to overall critical flux ($J_{c,local}/J_{c,overall}$) was generally close to unity, which suggests that the extent of fouling mitigation was quite uniform. On the contrary, for the fluidized GAC system, a higher energy cost was needed to increase the height of the fluidized bed in order to enable the $J_{c,local}/J_{c,overall}$ value to be closer to unity (Chapter 5).
- (vi) Both the power requirement (P_r) and overall critical flux ($J_{c,overall}$) values were higher for the bubbling system vis-à-vis the fluidized GAC system.

6.4. Conclusions

This study systematically correlated the hydrodynamics of bubbles with the fouling mitigation effectiveness in a microfiltration system which employed bubbling as a means of fouling mitigation.

Five highlights are noted here:

Firstly, the DOTM results showed that the local critical flux ($J_{c,local}$) values increased with height along the membrane (h/H) and with gas flow rate (Q_g), but was relatively invariant with liquid flow rate (Q_l). The $J_{c,local}$ values were generally found to be quite uniform along the membrane height, which contrasted to that for the fluidized GAC particles in Chapter 5 whereby the $J_{c,local}$ values varied more significantly with height. Furthermore, other than the deposition of the particulate foulant (in this case, polystyrene), micron-sized bubbles were observed to be deposited on the membrane too.

Secondly, the high-speed video images indicated that the mean bubble velocity ($V_{b,rms}$) and mean bubble area per bubble (A_b/N_b) increased with h/H , number of bubbles per unit membrane area (N_b) decreased with h/H , while the area of bubbles per unit membrane area (A_b) was relatively invariant with h/H . Changing the gas flow rate (Q_g) impacted all the bubble parameters characterized; on the other hand, changing the liquid flow rate (Q_l) only affected N_b , but had negligible impact on $V_{b,rms}$ and A_b .

Thirdly, regarding the relationship between the local bubble hydrodynamics and local critical flux ($J_{c,local}$), bubble momentum (P_b) and mean area per bubble (A_b/N_b) had the most positive correlation with $J_{c,local}$, both $V_{b,rms}$ and A_b had slightly positive correlations with $J_{c,local}$, while N_b and $J_{c,local}$ were slightly negatively correlated. That the bubble momentum (P_b) had the strongest relationship with $J_{c,local}$ agrees with our results in Chapter 4, which showed that the momentum of the fluidized granular activated carbon (GAC) particles and the extent of fouling mitigation was the most positively correlated compared to other hydrodynamics parameters. In addition, the decrease in N_b increased $J_{c,local}$ because of bubble coalescence and the greater bubble velocities (V_b) associated larger bubbles.

Fourthly, the overall critical flux ($J_{c,overall}$) increased linearly with respect to the ratio of gas to total flow rates (θ), which underscores the dominance of the gas flow rate (Q_g) in fouling mitigation. With respect to energy cost, $J_{c,overall}$ increased steeply with power required (P_r) at the lower P_r range, then more gradually at the higher P_r range.

Fifthly, between the two unsteady-state shear means of fouling mitigation via two-phase flows, increasing the critical flux seemed to involve more simply an increase in the gas flow rate (Q_g) for the bubbling system, but optimization of the particle size, particle concentration and liquid flow rate were necessary for the fluidized GAC system in Chapter 3 - 5.

Chapter 7 Relationship between scouring efficiency and overall concentration of fluidized granular activated carbon (GAC) in microfiltration⁵

7.1 Introduction

To improve the operation of the anaerobic fluidized bed membrane bioreactor (AnFMBR), efforts have focused on the design of the process [15, 20, 23, 51, 131, 136, 155, 157, 158, 160, 162, 173, 177, 247], membrane materials and module configurations [157, 158, 168, 177], feed suspension [15, 20, 152, 156, 163, 164], operating conditions [156, 157] and fluidized media properties [14, 131, 158, 159, 247-249]. For existing AnFMBRs, changes to operating conditions to improve the fouling mitigation is more tractable than more drastic changes involving design, feed, membrane or the type of fluidized media. Works in previous chapters have ascertained that the effectiveness of the solid-liquid fluidization of the typically used granular activated carbon (GAC) in fouling mitigation is dependent on an interplay of factors including superficial liquid velocity (U_l), which is tied to power requirement (P_r), foulant type, GAC particle diameter (d_p), position along the membrane (i.e., over different heights of the reactor). One particularly counter-intuitive observation was that the critical flux decreased as the increase of superficial liquid velocity in microfiltration of

5. The content in this chapter has been published as:

Jingwei Wang, Anthony G. Fane, Jia Wei Chew. Relationship between scouring efficiency and overall concentration of fluidized granular activated carbon (GAC) in microfiltration. *Chemical Engineering Research and Design*, volume 132, April 2018, Pages 28-39.

micron-sized foulant, which was attributed to the varying local concentration of fluidized GAC media along the membrane. Specifically, despite the similar initial bed height (i.e., packed bed of GAC particles) in these studies, it was noted unsurprisingly that the expanded bed height (i.e., bed height when the GAC particles are fluidized) differed for different GAC particle diameters (d_p) and superficial liquid velocities (U_l). The expanded bed height tends to be higher when a higher U_l is implemented, and higher for the smaller particles when the same U_l is used. This has important implications for the mitigation of membrane fouling, because the mechanical scouring effect is pivoted on contact between the fluidized GAC particles and the membrane, which has been found as a positive correlation between accelerometer signals and extent of fouling mitigation.

The benefits of the larger GAC particles on the extent of membrane fouling migration have been extensively studied [13, 130, 131, 157, 171, 188, 247], however, investigation regarding the effects of overall GAC concentrations on fouling mitigation is very limited. Both Kim et al. [13] and Wu et al. [157] found that the higher GAC dosages achieved better fouling mitigation stemming from the increased contact between the fluidized GAC and the membrane. A more recent investigation reported by Charfi et al. [248] using modelling method suggested that the increase of fluidized media (i.e., polyethylene terephthalate beads and silica particles) concentration increased the scouring effect on the mitigation of cake layer and pore blocking, and decreased the compressibility of the cake layer. However, the benefits of higher overall GAC concentration have to be weighed against the higher energy requirement due to the greater pressure

drop across the fluidized bed. Moreover, Aslam et al. [131] found that concentrations that are too high became ineffective due to the restricted movement of the fluidized GAC media. Similar results were found for the fluidization of PET (namely, polyethylene terephthalate) beads and silica media by Charfi et al. [248] that an optimal concentration of 50 % (v/v) was suggested. Therefore, tradeoffs involved in higher overall GAC concentration include higher fouling mitigation effectiveness, higher local GAC concentration, higher power requirement (P_r), but lower superficial liquid velocity required (U_l) hence lower particle velocity (V_p). Furthermore, because the local concentration along the expanded bed varied along the membrane (Chapter 4), the local critical flux ($J_{c,local}$) varied significantly along the membrane height (Chapter 5). This led to the conclusion that, for effective fouling mitigation, the ratio of the local critical flux to the overall critical flux ($J_{c,local}/J_{c,overall}$) should be as close to unity as possible along the membrane to ensure uniform fouling mitigation. In view of the importance of the concentration of the fluidized media in the operation of the AnFMBRs, a closer investigation and understanding is warranted.

Therefore, the objectives of this chapter's study were twofold: to assess the impact of overall GAC concentration on (i) critical flux vis-à-vis power requirement locally and overall; (ii) uniformity of fouling mitigation. The effect of varying overall GAC concentration was also compared to that of varying GAC particle diameter to assess the relative advantages. Both the direct observation through the membrane (DOTM) technique and trans-membrane pressure (TMP) were used to characterize the extent of fouling mitigation. The two foulants studied were polystyrene and bentonite.

7.2 Materials and methods

7.2.1 Experimental setup

A Direct Observation Through the Membrane (DOTM) setup was used to determine critical flux (J_c), which was similar to that in Chapter 5 and 6, as depicted in **Fig. 7-1**. Specifically, the membrane module was oriented vertically to allow for the fluidization of the GAC media, and a Zeiss microscope coupled with a color camera was oriented such that the focal plane was at the membrane-feed interface. An objective lens with 20× magnification was used. As described in Chapter 5, the presence of fluidized GAC media, which are opaque, reduced the amount of light in the focal axis, therefore an additional floodlight (Olympus, ILK-7A) was placed at the permeate side of the membrane cell. A digital gear pump (Cole Palmer, 75211-35) was used to control the flow rate required for the GAC particle fluidization, while a peristaltic pump (Cole Parmer, 7523-90) controlled the permeate flux. Three pressure transmitters (Cole Palmer; -101.4 – 103.5 kPa) were used to measure the pressure signals at the feed inlet, feed outlet and permeate outlet. The permeate flux was quantified by a digital balance (Mettler-Toledo, PL4002). For each experiment, the pressure and permeate flux data were recorded every 60 s via LabView (National Instruments).

Figure 7-1. Schematic of fluidized bed membrane filtration cell - DOTM setup.

The details of the solid-liquid fluidization membrane filtration cell are shown in **Fig 7-2a**, which consists of (i) a feed channel, (ii) a plastic sheet to expose only the targeted portion of the membrane, (iii) a membrane taped onto a plastic sheet, (iv) a metal mesh to provide mechanical support for the membrane, and (v) a permeate channel. The feed channel had internal dimensions of 30 mm width by 15 mm depth by 146 mm height, and had multiple layers of metal mesh at the bottom to ensure more uniform fluidization, and another metal mesh at the top to prevent the entrainment of GAC media (i.e., carry-over of the GAC media out of the top of the module). For the DOTM tests, the membrane was an Anopore disk (Whatman, Germany) with a diameter of 47 mm and nominal pore size of 0.2 μm , and was transparent when wetted; the transparency is necessary for the use of the DOTM technique, which requires focus through the permeate channel and the membrane such that the focal plane is the membrane-feed interface. The

plastic sheet was first cut into the dimensions of the cell (namely, 54 mm width by 175 mm height), then a square with dimensions of 26 mm by 26 mm was cut out from the plastic sheet in order to characterize specific heights of the membrane (**Fig. 7-2b**). Thereby, the active area of the membrane characterized at each height was 0.000676 m^2 , and the three heights characterized were $h/H = 0.29, 0.46$ and 0.62 (where h is the height assessed and H is the height of the membrane filtration cell of 146 mm). The lower bound of 0.29 was such to enable sufficient light to penetrate through the fluidized GAC particle bed especially for the highest overall concentration, while the upper bound of 0.62 was such to sufficient mechanical scouring effects especially for the lowest overall concentration. As for the $d\text{TMP}/dt$ (i.e., rate of change of transmembrane pressure) tests, the membrane was a polyvinylidene fluoride (PVDF) flat-sheet membrane from Merck Millipore Ltd with a nominal pore size of $0.22 \text{ }\mu\text{m}$. The membrane was cut into 45 mm by 100 mm and pasted onto the plastic sheet with a cut-out of 26 mm width by 78 mm height (**Fig. 7-2b**), thereby the active area of the membrane was 0.002028 m^2 .

Figure 7-2. (a) Components of the solid-liquid fluidization membrane filtration cell; and (b) Schematic of the four different plastic sheets used for the experiments: the first three plastic sheets had Anopore membranes (diameter of 47 mm) attached for the DOTM tests (h/H is the ratio of the height assessed to the total membrane cell height of 146 mm), while the fourth plastic sheet had a PVDF membrane attached for the dTMP/dt test.

7.2.2 Materials

GAC media (FILTRASORB 300, Calgon Carbon Corporation) with a particle diameter (d_p) of 1.55 ± 0.15 mm, which was approximately the mean of the un-sieved GAC media, were used for the study. This narrower particle size distribution (PSD) used in this study was obtained by sieving using the Cole-Parmer U.S. Standard Brass Test Sieves and Ro-Tap sieve-shaker. Three overall concentrations of GAC were investigated in the DOTM study, namely, 50 g/L, 100 g/L, and 150 g/L, which corresponded respectively to initial packed bed heights of 0.019 m, 0.035 m and 0.057 m. Higher concentrations were not possible for DOTM because the considerable blocking of the light impeded observations. For the trans-membrane pressure (TMP) tests, concentrations of up to 200 g/L was used.

Two particulate foulants were used in this study. Polystyrene particles (Sigma Aldrich, Singapore) with a characteristic particle diameter of 5 μm and density of 1.05 g/cm³ were used for the determination of critical flux (J_c) via DOTM. The concentration of the polystyrene particles used in this study was 0.01 % by volume in DI water. For the dTMP/dt tests, 1 – 5 g/L of bentonite (Sigma Aldrich, Singapore) in DI water was used as the feed solution. The feed solution was continuously stirred at 250 rpm by using a magnetic stirrer (AS ONE; REXIM RS-1D) throughout the experiments. As shown in **Fig. 7-3**, although the mean particle diameter measured by a Mastersizer (Malvern Mastersizer 2000) was 5.4 μm for both bentonite and polystyrene particles, bentonite had a wider particle size distribution (PSD) than the polystyrene particles.

Figure 7-3. Particle size distributions (PSDs) of polystyrene and bentonite particles

7.3 Results and Discussion

7.3.1 Minimum fluidization velocity (U_{mf})

The minimum fluidization velocity (U_{mf}) was first determined to ensure the GAC media were fully fluidized. It is well acknowledged that the U_{mf} value can be determined experimentally by plotting the pressure drop (ΔP_{bed}) across the bed or particle bed height (z) or bed density as a function of the superficial liquid velocity (U_l) [147]. **Fig. 7-4** shows a plot of bed height (z) versus superficial liquid velocity (U_l) for the three overall concentrations investigated in the range of 50 – 150 g/L; the vertical dotted line represents the minimum fluidization velocity (U_{mf}) of the particles. Two observations are worth noting.

Firstly, it is clear that, regardless of overall concentration, the bed height was constant at the lower range of U_l values, and then started increasing beyond a critical U_l . This critical U_l is the U_{mf} of the GAC media whose particle diameter (d_p) was 1.55 mm. The U_{mf} in this case was 0.0083 m/s, which agrees with the studies in previous Chapters 3-5. Secondly, at the same U_l , the bed heights were naturally taller for the higher overall concentrations, which implies increased contact between the fluidized GAC and the membrane, which in turn has been reported in Chapter 5 to improve the overall fouling mitigation. However, it should be noted that the same U_l value does not directly translate to the same power requirement (P_r) when the overall concentration is different. More explicitly, as the overall concentration increases, the bed height (z) increases and so does the pressure drop across the bed (ΔP_{bed}), which hence implies that the power required (P_r) is greater for a higher overall GAC concentration for the same U_l :

$$P_r = \Delta P_{bed} U_l A \quad (7-1)$$

where A is the cross-sectional area of the reactor. Therefore, understanding the tradeoff between overall concentration and power required (P_r) is necessary. **Fig. 7-4** shows that, while an overall concentration of 150 g/L at a U_l of 0.010 m/s gave an expanded bed height of 0.061 m, an overall concentration of 100 g/L required a U_l of approximately 0.026 m/s to achieve the same bed height. Recall that GAC-membrane contact caused the critical flux (J_c) of the micron-sized particles to be an order-of-magnitude greater than that without. **Eq. (7-1)** hence indicates that, for the same bed height to ensure the same area of GAC-membrane

contact, the power required (P_r) for the overall concentration of 150 g/L is 0.63 times that for the lower overall concentration of 100 g/L; this suggests that the taller bed associated with a higher overall concentration may be more energy-efficient.

Figure 7-4. Bed height (z) versus superficial liquid velocity (U_l) for overall GAC ($d_p = 1.55$ mm) concentrations of 50 g/L, 100 g/L and 150 g/L, which corresponded respectively to initial packed bed heights of 0.019 m, 0.035 m and 0.057 m. The vertical dotted line represents the minimum fluidization velocity (U_{mf}).

7.3.2 Determination of critical flux (J_c) via DOTM

Critical flux (J_c) is the permeate flux above which fouling occurs significantly and below which negligible fouling occurs [87]. As with our studies in Chapter 5 and 6, DOTM was used to characterize the local critical flux ($J_{c,local}$) during the microfiltration of the micron-sized particulate foulants in the presence

of fluidized millimeter-sized GAC media by the flux-stepping method. Specifically, each permeate flux was held constant for 10 minutes, during which images were taken at the end of 1st and 10th minute. At the end of every 10 minutes, the flux was increased 5 L/(m² h). The initial image was taken at the end of 1st minute not 0 minute, which was to stabilize the sudden change of permeate flux. Examples of images at the end of the 1st and 10th minute at various fluxes, and at $h/H = 0.29$, $U_l = 0.021$ m/s and overall GAC ($d_p = 1.55$ mm) concentration of 150 g/L, are shown in **Fig. 7-5a**. The darker background is due to the opaque GAC media. It is clear that negligible deposition of the polystyrene particles was observed at the permeate flux of 0 L/(m² h), but significant number of polystyrene particles had deposited at the higher fluxes of 86 and 91 L/(m² h). Therefore, the critical flux can be determined by accounting for the rate of change of the number of polystyrene particles (dN/dt) deposited at each flux:

$$\frac{dN}{dt} = \frac{N_{10th\ minute} - N_{1st\ minute}}{9\ min} \quad (7-2)$$

Each test was repeated at least two times with new membranes. To avoid any possible change of the GAC particles due to particle attrition, fresh GAC and polystyrene particles were used for every test. **Fig. 7-5b** presents an example of a plot of dN/dt versus flux at $h/H = 0.29$, $U_l = 0.021$ m/s and overall GAC ($d_p = 1.55$ mm) concentration of 150 g/L. The dN/dt values were zero at the lower fluxes then started increasing at the higher fluxes. In this case, the $J_{c,local}$ was determined as 81 L/(m² h), which was the flux beyond which the dN/dt values became non-zero.

Figure 7-5. (a) DOTM images at the end of the 1st and 10th minute interval for four permeate fluxes at $h/H = 0.29$, $U_l = 0.021$ m/s and overall GAC ($d_p = 1.55$ mm) concentration of 150 g/L; (b) the rate of the change of the number of deposited polystyrene particles (dN/dt) versus flux at $h/H = 0.29$, $U_l = 0.021$ m/s and overall GAC ($d_p = 1.55$ mm) concentration of 150 g/L, which indicates the local critical flux ($J_{c,local}$) was 81 L/(m² h).

7.3.3. Local critical flux ($J_{c,local}$)

Fig. 7-6 depicts the local critical flux ($J_{c,local}$) at various heights (h/H) across the membrane as a function of U_l for three overall concentrations (i.e., 50 g/L, 100 g/L and 150 g/L). It should be noted that the higher the $J_{c,local}$ value is, the more effective the fouling mitigation is. The ranges of the x - and y -axes are kept the same for easier comparison. The lower limit of the superficial liquid velocity (U_l) for each overall concentration was such that it enabled the fluidized GAC to be in contact with at least the lowermost portion of the membrane, and upper limit such that entrainment of the fluidized GAC was avoided. Four observations are worth highlighting. Firstly, when the fluidized GAC was in

contact with the membrane, the $J_{c,local}$ values were an order-of-magnitude greater than that in the absence of fluidized GAC. This agrees with the general acknowledgement of the efficacy of fluidized GAC in mitigating membrane fouling. It should be noted that $J_{c,local}$ was assumed to be 5 L/(m² h) for the cases whereby $J_{c,local}$ was too low to be accurately determined, because of the limited resolution of the permeate pump. Secondly, at the same U_l , the $J_{c,local}$ values were greater for the higher overall concentrations. This is presumably because of the increased local GAC concentration, which has been shown in our previous studies Chapter 4 and 5 to improve fouling mitigation, associated with the higher overall concentrations. Thirdly, values of $J_{c,local}$ decreased as height (h/H) increased for all three overall concentrations, which is due to the decrease in local GAC concentration with height in Chapter 4. Fourthly, although $J_{c,local}$ was expected to increase with U_l , it was not consistently the case. While the J_c values for the two higher overall concentrations decreased with U_l at the lower U_l range at the lowest height ($h/H = 0.29$), the $J_{c,local}$ values were approximately invariant with U_l at the middle height ($h/H = 0.46$). This contrasts with the results based on the effect of crossflow velocity on membrane critical flux (J_c) in the absence of fluidized GAC media, and has been affirmed causing by the effect of local GAC concentration changes via high speed video method in Chapter 4.

Fig. 7-7 displays the $J_{c,local}$ along the membrane height (h/H) at different U_l values for three overall GAC concentrations (i.e., 50 g/L, 100 g/L and 150 g/L). The ranges of the x - and y -axes are kept the same for easier comparison. Four observations are worth noting: (i) the lowest overall concentration (**Fig. 7-7a**) generally gave lower $J_{c,local}$ values even at higher U_l values, which indicate

that overall concentration is also a significant parameter to consider in the optimization of such systems, on top of GAC size (d_p), U_l and height along the membrane;. (ii), $J_{c,local}$ decreased with height (h/H) because of the reduced GAC media concentration with height (Chapter 4); (iii) an increase of U_l increased the $J_{c,local}$ values only if the increased U_l brought about greater bed expansion, otherwise the $J_{c,local}$ were not much affected by U_l at the same height (h/H); and (iv) higher U_l gave more similar $J_{c,local}$ values along the membrane height, which indicates more uniform extents of fouling mitigation along the membrane height.

Figure 7-6. Local critical flux ($J_{c,local}$) of three overall GAC ($d_p = 1.55 \text{ mm}$) concentrations with respect to superficial liquid velocities (U_l) at (a) $h/H = 0.29$, (b) $h/H = 0.46$, and (c) $h/H = 0.62$. The foulant was polystyrene particles. The hollow symbols represent the cases whereby the fluidized GAC was not in contact with the membrane, hence the critical flux was similar to that in the system without GAC.

Figure 7-7. Dimensionless height (h/H) versus local critical flux ($J_{c,local}$) for fluidized GAC ($d_p = 1.55$ mm) media with overall concentrations of (a) 50 g/L, (b) 100 g/L and (c) 150 g/L at different superficial liquid velocities (U_l).

7.3.4. Power requirement (P_r)

While the local critical flux ($J_{c,local}$) sheds light on the extent of fouling mitigation at each local position of the membrane, the overall critical flux ($J_{c,overall}$), which is an arithmetic average of the three $J_{c,local}$ values (Fig. 7-6), is useful for understanding the overall performance of the membrane module. **Fig. 7-8a** presents the overall critical flux ($J_{c,overall}$) values versus superficial liquid velocity (U_l) for the various overall concentrations. The $J_{c,overall}$ values increased with both U_l and overall concentration. This is because a higher U_l leads to higher local particle velocities and higher expanded bed heights, while a higher overall concentration leads to higher local particle concentration and higher bed heights. Because the power requirement (P_r) is higher at the same U_l for the higher overall concentrations (Eq. (7-1)), the relationship between $J_{c,overall}$ values versus power requirement (P_r) for the various overall concentrations are displayed in **Fig. 7-8b**. It should be noted that the relationship between overall concentration and power requirement (P_r) is such that, while a higher overall concentration gives a

taller bed that in turn enables more particle-membrane contact to enhance the fouling mitigation, it also requires more power (P_r) to fluidize the bed due to the higher pressure drop across the taller bed (**Eq. (7-1)**). Two key observations are noted. Firstly, the positive correlation between $J_{c,overall}$ and P_r is clear. This indicates expectedly that a higher power input is needed for a higher extent of fouling mitigation. Secondly, at the same power requirement (P_r), while the two lower concentrations (i.e., 50 and 100 g/L) behaved similarly, the highest concentration of 150 g/L gave a significantly higher $J_{c,overall}$ than the intermediate concentration of 100 g/L. This indicates that the overall concentration has to be increased beyond a certain value to confer benefits, and suggests that overall concentrations beyond 150 g/L may give even higher $J_{c,overall}$ values. Because **Fig. 7-8a** shows that the higher overall concentrations can allow for operation at higher permeate fluxes due to the higher $J_{c,overall}$ values, it is worth assessing the system in terms of power requirement per unit permeate. Interestingly, **Fig. 7-8c** presents quite different trends from **Fig. 7-8b**. While the y-axes are the same, the x-axis in **Fig. 7-8c** represents power per unit permeate (P_p), which is the power requirement (P_r) normalized by the product of the overall critical flux ($J_{c,overall}$) and the membrane area. Two observations are worth noting from **Fig. 7-8c**. Firstly, a general negative correlation is shown between $J_{c,overall}$ and P_p , which indicates that a lower P_p gave more effective fouling mitigation. Secondly, at the lower range of P_p investigated, the higher overall concentrations gave higher $J_{c,overall}$ values and the highest concentration showed positive correlation with P_p opposite to lower overall GAC concentrations. As P_p was derived by the division of power requirement (P_r) by the product of the overall critical flux ($J_{c,overall}$) and

the membrane area. This positive correlation for 150 g/L GAC was caused by the ratio of P_r increased over the ratio of $J_{c,overall}$ increased. This indicates that values of $J_{c,overall}$ for lower GAC concentrations were more sensitive to changes in the power requirement (P_r). In terms of power needed per unit production of permeate (P_p), the higher overall concentrations were better in mitigating fouling. **Fig. 7-8c** affirms that the taller beds associated with higher overall concentrations have a clear advantage both in terms of higher $J_{c,overall}$ and also lower P_p . The agrees with **Fig. 7-4** and **Eq. 7-1**, which indicate that, for the same bed height to ensure the same area of GAC-membrane contact, the power required (P_r) for the overall concentration of 150 g/L is only 0.63 times that for the lower overall concentration of 100 g/L.

Figure 7-8. Overall critical flux ($J_{c,overall}$) versus (a) superficial liquid velocity (U_l), (b) power requirement (P_r), and (c) power requirement per unit permeate (P_p) for the three overall concentrations of GAC ($d_p = 1.55$ mm) investigated using polystyrene particles as foulants.

It has been indicated in Chapter 5 that the optimal fluidization scenario should not only give a good enhancement of $J_{c,overall}$ at a modest P_r , but also ratios of $J_{c,local}/J_{c,overall}$ close to unity at all membrane heights to enable uniform fouling mitigation across the membrane. Non-uniform fouling across the membrane is undesirable, since it causes elevated flux through the lesser-fouled parts in a constant flux mode, which thereby accelerates the fouling in these regions. **Figs. 7-9a - c** depict the ratios of $J_{c,local}/J_{c,overall}$ versus power requirement (P_r) at different heights along the membrane for each of the three concentrations investigated. The ranges of the y-axes are kept the same for easier comparison. Three observations are apparent. Firstly, consistent with our results in Chapter 5, the $J_{c,local}/J_{c,overall}$ values approached unity as P_r increased, which indicates more uniform fouling mitigation across the membrane at a higher power input. On one hand, a lower P_r gave lower bed expansion (**Fig. 7-4**), which led to higher $J_{c,local}$ values at lower heights (e.g., $h/H = 0.29$) due to higher local particle concentrations, and lower $J_{c,local}$ values at higher heights (e.g., $h/H = 0.62$) due to the lack of particle-membrane contact. On the other hand, a higher P_r gave greater bed expansion (**Fig. 7-4**), which led to a more uniform local particle concentration across the membrane and thereby more uniform $J_{c,local}$ values. Secondly, among the three overall GAC concentrations, the highest overall concentration of 150 g/L (**Fig. 7-9c**) gave the most similar $J_{c,local}$ values among the three heights in the range of P_r investigated. This is due to the tallest bed heights associated with the highest overall concentration (**Fig. 7-4**), which correspond to more uniform local particle concentrations and thus $J_{c,local}$ values along the membrane. Thirdly, the $J_{c,local}/J_{c,overall}$ values at all the three heights

were the closest to unity at the highest P_r for the intermediate overall concentration of 100 g/L. This is presumably due to the interplay between local particle velocity and local particle concentration for the different overall concentrations.

Figure 7-9. Ratios of $J_{c,local} / J_{c,overall}$ versus power requirement (P_r) at various heights (h/H) along the membrane for overall GAC ($d_p = 1.55$ mm) concentrations of (a) 50 g/L, (b) 100 g/L, and (c) 150 g/L

Since an increase of either superficial liquid velocity (U_l) or overall concentration can increase bed height, it is worth evaluating the power required to maintain the expanded bed height constant at the total membrane height of 0.104 m (**Fig. 7-2b**) to ensure scouring of the whole membrane. **Fig. 7-10** shows the power required (P_r) as a function of overall concentration (in the range of 50 – 200 g/L). Clearly, the power required (P_r) was non-monotonic with respect to the overall GAC concentration, because of the two opposing trends to achieve the same expanded bed height. As the overall concentration increased, while the superficial liquid velocity (U_l) required to achieve the targeted bed height was reduced due to the initially taller packed bed height on one hand, the pressure drop across the bed (ΔP_{bed}) was increased on the other hand. Therefore, the maximum power required (P_r) was found at an overall concentration of 143 g/L, whereby the initial packed bed height was about half of the targeted expanded bed height.

Figure 7-10. Superficial liquid velocity (U_l), pressure drop (ΔP_{bed}), and power requirement (P_r) versus overall GAC concentrations for a constant expanded bed height of 0.104 m.

7.3.5. Foulant type

An issue worth highlighting regarding the efficacy of fluidized GAC in fouling mitigation is the seemingly contradictory results obtained in our previous studies. Whereas one indicated that the smallest GAC media were the most effective at the same power input when the foulants were 5 μm polystyrene particles in Chapter 5, another indicated that the smallest GAC media were the least effective at the same expanded bed height when the foulant was bentonite in Chapter 4. As **Fig. 7-10** shows, the different bases (i.e., power input versus bed height) could cause the contradiction. Another different basis is the foulant type.

To assess if foulant type could lead to contrary trends, the bentonite foulant was used in the same experimental setup (**Fig. 7-1**). Because the critical flux (J_c) for bentonite could not be determined with confidence via DOTM due to the cloudiness of the feed suspension, the dTMP/dt (i.e., rate of change of transmembrane pressure) method, which provides a measure of the extent of fouling, was used. The higher the dTMP/dt value is, the less effective is the fouling mitigation. Analogous to **Fig. 7-8**, **Figs. 7-11a, b and c** show dTMP/dt, whereby dt was 1 h, versus superficial liquid velocity (U_l), power requirement (P_r), and power requirement per unit permeate (P_p), respectively, for the three overall concentrations at a constant permeate flux of 30 L/(m² h). **Fig. 7-11a** shows that (i) at the same overall concentration, dTMP/dt decreased with U_l , which indicates expectedly that a higher U_l was more effective in mitigating fouling due to the higher local particle velocities and higher expanded bed; and

(ii) at the same U_l , $dTMP/dt$ decreased with concentration, which indicates that a higher overall concentration was more effective in mitigating fouling due to the higher expanded bed. Since **Eq. (7-1)** indicates that the power requirement (P_r) for the different overall concentrations is different at the same U_l , it is necessary to also assess the relationship between $dTMP/dt$ and P_r . **Fig. 7-11b** shows that (i) $dTMP/dt$ expectedly decreased with P_r , and (ii) the effect of concentration was negligible in the lower P_r range but the higher concentration was more effective in the higher P_r range, which is similar to the observation in **Fig. 7-8b**. It should be noted that, unlike between **Figs. 7-8b and c**, the trends for P_r (**Fig. 7-11b**) and P_p (**Fig. 7-11c**) were similar here because a constant flux was used. The consistency in trends between **Figs. 7-8a and 7-11a**, along with that between **Figs. 7-8b and 7-11b**, indicates that the effect of overall concentration was similar regardless of whether polystyrene or bentonite was the foulant. Therefore, the seeming contradiction between the two studies in Chapter 4 and 5 on the effect of GAC particle size was likely not because of the different foulants used, since both polystyrene and bentonite foulants gave similar trends. It is unexpected that an abnormal increase then decrease trend was found in **Fig. 7-11abc** for 100 g/L GAC concentration. This was presumably due to the varying local GAC concentration and particle velocity along the membrane with respect to the extents of bed expansion, which highlights again the interplay between local particle velocity and local particle concentration for fouling mitigation effectiveness.

Figure 7-11. $dTMP/dt$ versus (a) superficial liquid velocity (U_l), (b) power requirement (P_r), and (c) power requirement per unit permeate (P_p) for the three overall concentrations of GAC ($d_p = 1.55$ mm) investigated using 2 g/L of bentonite feed at a constant permeate flux 30 L/(m² h).

The effect of the ratio of bentonite concentration to overall GAC concentration ($C_{\text{bentonite}}/C_{\text{GAC}}$) was also evaluated to understand how it affects fouling mitigation. **Fig. 7-12** presents $d\text{TMP}/dt$ versus $C_{\text{bentonite}}/C_{\text{GAC}}$. Interestingly, $d\text{TMP}/dt$ was not affected by changes in $C_{\text{bentonite}}/C_{\text{GAC}}$ at the lower and upper ends, but increased approximately linearly with $C_{\text{bentonite}}/C_{\text{GAC}}$ at the intermediate $C_{\text{bentonite}}/C_{\text{GAC}}$ values. At the lowermost $C_{\text{bentonite}}/C_{\text{GAC}}$ values of less than 0.01, the fluidized GAC was in sufficient amounts to effectively mitigate membrane fouling even as the bentonite concentration increased. As $C_{\text{bentonite}}/C_{\text{GAC}}$ increased to between 0.01 and 0.05, the linear increase of $d\text{TMP}/dt$ with $C_{\text{bentonite}}/C_{\text{GAC}}$ was because of insufficient GAC, which indicates that increasing the GAC concentration in this region would be most advantageous for improving fouling mitigation. However, as $C_{\text{bentonite}}/C_{\text{GAC}}$ increased beyond 0.05, the extent of membrane fouling had maximized under the high bentonite concentration such that further increases in bentonite concentration did not cause further fouling. This highlights the important contribution of overall GAC concentration at the intermediate $C_{\text{bentonite}}/C_{\text{GAC}}$ values.

Figure 7-12. dTMP/dt versus the ratio of feed suspension to GAC concentration for GAC media with diameter of 1.55 mm at constant fluidization bed height of 0.104 m.

Particularly for the bentonite foulant, due to the sticky cake formed, it was observed that the GAC media were deposited on the cake formed, which agrees with a previous report that the GAC media deposited and increased the cake resistance [130]. The deposition of the GAC media not only increased fouling but also reduced the fouling mitigation effectiveness of the fluidized GAC due to the reduced amounts of GAC. **Fig. 7-13** shows the number of GAC media deposited per unit area as a function of the ratio of superficial liquid velocity to minimum fluidization velocity (U_l/U_{mf}) at an overall GAC concentration of 100 g/L and TMP of 30 kPa. This indicates that the deposition of GAC media expectedly decreased as U_l/U_{mf} increased, which informs that sufficient U_l and thereby P_r is necessary to ensure sufficient movement of the

fluidized GAC media in order to avoid deposition of these fouling mitigation agents.

Figure 7-13. GAC deposition amount per unit membrane versus the ratio of U_l to U_{mf} at an overall GAC concentration of 100 g/L and TMP of 30 kPa.

7.3.6. Comparative analysis of the overall GAC concentration and GAC particle diameter with respect to membrane fouling mitigation

Either adjustments to the overall GAC concentration as studied here or the particle diameter of the GAC media as reported in Chapter 3-5 can improve the extent of membrane fouling mitigation. A comparison was carried out to assess the relative advantages of these two factors:

- (1) Greater overall GAC concentration and larger GAC media generally gave lower dTMP/dt (Chapter 3-5) and higher critical flux values (Chapter 5). The higher overall concentration increased the contact

with the membrane, while the larger GAC media gave greater particle momentum and thereby more effective scouring.

- (2) At the same U_t , a greater GAC concentration was more effective in mitigating membrane fouling due to the increased bed heights, while smaller GAC media was better due to the relatively larger $(U_t - U_{mf})$ and taller expanded beds (Chapter 3). It should be noted that a greater power requirement (P_r) is associated with the greater GAC concentration, while the same P_r was needed for the various sizes at the same U_t .
- (3) At the same expanded bed height, power requirement (P_r) increased then decreased as the overall GAC concentration increased, while P_r increased monotonically with GAC particle diameter. Fouling mitigation improved with either an increase in overall GAC concentration or particle diameter, as stated in (1).
- (4) At the same P_r , higher overall GAC concentration improved fouling mitigation only at the higher P_r range but not at lower P_r , while smaller particles gave higher $J_{c,overall}$ values (Chapter 5).
- (5) For similar $J_{c,overall}$ values, the various GAC concentrations did not affect the P_r values much, while small GAC media required less power (P_r) (Chapter 5).
- (6) The uniformity of fouling mitigation (defined by how close the ratio of local to overall critical flux ($J_{c,local}/J_{c,overall}$) was to 1) increased with P_r regardless of whether it was by virtue of overall GAC concentration or GAC particle diameter.

7.4 Conclusions

The influence of the overall concentration of fluidized millimeter-sized GAC media on the fouling of micron-sized particulate foulants during microfiltration is systematically investigated. The Direct Observation Through the Membrane (DOTM) technique was used to determine the critical flux (J_c) of the polystyrene foulants at three overall concentrations of GAC (namely, 50, 100 and 150 g/L, which corresponded to initial packed bed heights respectively of 0.019, 0.035 and 0.057 m). Trans-membrane pressure (TMP) was used to characterize the extent of fouling mitigation of the 1 – 5 g/L bentonite foulant at overall GAC concentrations of between 50 – 200 g/L. The relative advantages of adjusting overall GAC concentration was assessed versus that of adjusting the particle diameter of the GAC media. Four key conclusions are here summarized:

- (i) At the same superficial liquid velocity (U_l), the increase of overall GAC concentration increased the overall critical flux ($J_{c,overall}$) values and reduced the power required per unit production of permeate (P_p). A taller beds associated with higher overall are clearly advantageous in terms of both higher $J_{c,overall}$ and lower P_p . However, a greater pressure drop across the bed (ΔP_{bed}) required and thereby increased the power required (P_r).
- (ii) Higher overall GAC concentration generally gave more similar $J_{c,local}$ values among the three heights in the range of P_r investigated. Furthermore, the $J_{c,local}/J_{c,overall}$ values approached unity as P_r increased, which indicates more uniform fouling mitigation across the membrane at a higher power input regardless

of whether it was by virtue of overall GAC concentration or GAC particle diameter (Chapter 5).

- (iii) The benefits of higher overall GAC concentration were negligible at the lower P_r values but significantly advantageous at the higher P_r values regardless of whether polystyrene or bentonite was the foulant.
- (iv) Although a higher minimum P_r was necessary to initiate the mechanical scouring effects for higher overall concentration or larger d_p , they tended to give greater $J_{c,overall}$ values at the same P_p , which means that higher overall concentration or larger d_p conditions were more energy-efficient in mitigating membrane fouling.

Chapter 8 Characteristics of non-spherical fluidized media in a fluidized bed – membrane bioreactor: Effect of particle sphericity on critical flux⁶

8.1. Introduction

Interestingly, although GAC particle is presumably superior due to its adsorption of organics and ability to function as a biocarrier in addition to the mechanical scouring capacity [51], the use of fluidized glass beads, which were larger and denser than GAC, has been reported to reduce membrane fouling by an impressive 95% in a tubular ceramic membrane [158], and non-porous fluidized media such as polyethylene terephthalate (PET), which were larger and less dense than GAC, was reported to mitigate membrane fouling better than fluidized GAC [131]. In a recent review regarding fouling mitigation by scouring agents in MBRs [51], it was concluded that the effectiveness of mechanical scouring depends not on a single parameter but the interplay of several key factors including hydrodynamics, scouring media, membrane type, bulk composition and cake layer properties. This suggests that more mechanistic studies on various aspects are needed. Regarding hydrodynamics, our studies in Chapters 3-7 have served to shed light on the correlation between the behavior of the fluidized GAC and extent of membrane fouling mitigation. Higher particle

6. The content in this chapter has been published as:

Jingwei Wang, Anthony G. Fane, Jia Wei Chew. Characteristics of non-spherical fluidized media in a fluidized bed-membrane reactor: Effect of particle sphericity on critical flux. *Separation and Purification Technology*, volume 202, 31 August 2018, Pages 185-199.

velocities, particle concentration and particle momentum were generally positively correlated with the extent of fouling mitigation. With respect to the particle properties of the fluidized media, the larger GAC conferred more effective scouring due to greater inertia but at a higher energy cost, thereby the smaller GAC was found to be more energy-efficient at mitigating fouling at a given power input; **Table 8-1** summarizes the pros and cons of fluidizing the smaller GAC relative to the larger ones for mitigating membrane fouling. However, the optimal fluidizing media properties are yet to be identified. Towards this end, a question that remains unanswered is the effect of particle sphericity (ϕ), which is known to confer different hydrodynamics in fluidized bed systems [250-253] and may possibly be harnessed to improve the energy efficiency of such scouring agents.

Table 8-1. Pros and cons of fluidizing smaller GAC relative to larger ones for mitigating membrane fouling.

Pros	Cons
<ul style="list-style-type: none"> • Higher adsorption capacity [131] • Greater particle mobility at the same superficial liquid velocity (U_l) due to the smaller minimum fluidization velocity (U_{mf}) (Chapter 3 and 4) • Greater bed expansion at the same U_l and overall GAC concentration, thereby more membrane area scoured (Chapter 5) • Lower energy required to achieve the same water shear stress and 	<ul style="list-style-type: none"> • Greater fouling rate at the same packing ratio [131] • Particle entrainment issues due to lower terminal velocity (U_t), which may hinder fouling mitigation if higher superficial liquid velocity (U_l) is necessary (Chapter 5) • GAC media sized below 0.5 mm may act as foulants and have negative impact on fouling mitigation [157, 188]

particle momentum [247]	
<ul style="list-style-type: none"> • Lower tendency for damage to membrane integrity [157, 165, 172] • Lower energy required to achieve the same overall critical flux, water shear stress and particle momentum [247] 	<ul style="list-style-type: none"> • Lower critical flux at the same power input due to lower particle inertia and thereby lower mechanical scouring (Chapter 3 and 5) • Lower extent of fouling mitigation at the same packing ratio and expansion bed height [131, 157, 165]

Compared to spherical particles, non-spherical ones behave differently. For example, for the same particle volume, they have higher drag coefficients [254, 255], exhibit different segregation tendencies [256] and different pressure drops across the fluidized beds [148, 252], tend to channel at low superficial liquid velocities [252], give poorer fluidizing quality [257], have different contact force and particle velocity [258], and rotate differently [259]. Correspondingly, armed with the knowledge accumulated to date on the trade-offs between particle scouring efficiency and energy consumption for the fluidization of the rather spherical GAC media [247], the current study aimed to investigate the efficacy of non-spherical particles (namely, rice) in mitigating membrane fouling.

Relative to GAC media, the three types of rice media investigated had greater aspect ratios, similar particle densities and were similarly millimeter-sized. A lab-scale AnFMBR setup was employed with micron-sized polystyrene particles as the particulate foulants, along with the direct observation through the membrane (DOTM) instrument and a high-speed video camera to characterize respectively critical flux and dynamics of the fluidized rice media.

The specific objectives were to (i) correlate particle sphericity (ϕ) with critical flux (J_c) and particle fluidization characteristics (namely, particle velocity, particle rotation velocity, particle-membrane contact area, and particle momentum); and (ii) assess the benefits or lack thereof in the use of non-spherical particles in view of overall critical flux and energy requirement.

8.2. Materials and methods

8.2.1 Experimental setup

The schematic of the experimental setup used to characterize critical flux is shown in **Fig. 8-1**, which is similar to that in Chapters 5 and 6 on fouling mitigation by granular activated carbon (GAC) particles and bubbles. A digital gear pump (Cole-Parmer, 75211-35) was used to control the feed flow rate required for the fluidization, while a peristaltic pump (Cole-Parmer, 7523-90) the suction for the permeate flow. Three pressure transmitters (Cole-Parmer), each in range of -101.4 – 103.5 kPa, were used to measure the pressure in the feed inlet, feed outlet and permeate outlet. A balance (Mettler-Toledo, PL4002) was used to measure the permeate flux. Both the pressure and permeate flux values were averaged and recorded every 60 s via Labview (National Instrument).

The membrane module was of the same design as that in Chapter 5 study on the fluidization of the millimeter-sized GAC media. Specifically, the feed channel had dimensions of 30 mm width by 15 mm depth by 146 mm height, and multiple layers of metal mesh were secured at the bottom to enable uniform upflow, while another metal mesh was at the top to prevent the entrainment of the fluidized media. The membrane used for every experiment was the Anopore

disk membrane (Whatman, Germany), which was 47 mm in diameter, had nominal pore size of 0.2 μm , and was transparent when wetted to allow for visualization through the membrane (which is key to the DOTM technique). A plastic sheet with dimensions of 54 mm width by 175 mm height was used to secure the position of the membrane at three different positions along the height of the membrane module. Specifically, a square of 26 mm by 26 mm was cut out from the plastic sheet, and the membrane was taped onto this area, which gave an active membrane area of 0.000676 m^2 . The three heights along the membrane module assessed were namely h/H values of 0.29, 0.46 and 0.62, where h is the height assessed and H is the total membrane module height of 146 mm. While the lower h/H bound ensured that the packed bed of fluidized media was expanded enough to allow for adequate light to pass through for observation of the membrane, the upper h/H bound was such that sufficient fluidized media were present. Because the membranes were brittle, a stainless-steel sheet provided mechanical support for the plastic sheet with the adhered membrane. The stainless-steel support had dimensions of 39 mm width by 161 mm height, and uniformly distributed circles each with diameter of 2 mm to allow for observation by the microscope.

To study the hydrodynamics of the fluidized media, a high-speed camera (Olympus *i*-SPEED LT) was employed. Each recording lasted 5 - 10 s at a frequency of 100 frame per second (fps).

Figure 8-1. Schematic of experimental setup.

8.2.2 Materials

The media fluidized for mitigating membrane fouling were three types of rice media (bought at a local supermarket), as presented in **Fig. 8-2a**, which were approximately ellipsoidal and differed in their dimensions. Accordingly, the three characteristic dimensions of an ellipsoid given in **Fig. 8-2b** were quantified by a Vernier caliper (KERN, Germany) for the three types of rice media. The particle sphericity (φ) of each particle type was further derived from the images of the saturated rice media via the AutoCAD 2014 software and using the equation [233]:

$$\varphi = \frac{2\sqrt{A/\pi}}{d_{min}} \quad (8-1)$$

where A is the projected area of the particle and d_{min} is the diameter of the minimum circumscribed circle around the particles. The greater the deviation

of the ϕ value from 1, the less spherical the particle is. At least 20 particles were characterized and the averaged values along with the standard deviations are given in **Table 8-2**. Clearly, Type 1 had the largest a value and lowest ϕ value, whereas Type 3 had the largest b , c and ϕ values. The average volumes of the ellipsoids (i.e., $4\pi abc/3$) and the volume-based equivalent spherical diameters (d_s) are presented in **Table 8-2** too. While the d_s values of Types 1 and 3 were similar, that of Type 2 was smaller. Therefore, comparing Types 1 and 3 gives the effect of sphericity (ϕ), whereas comparing with Type 2 gives the combined effects of sphericity and size of the fluidized rice media; note that size effects have been unveiled in previous studies (**Table 8-1**).

Before each experiment, the rice media were fluidized in the membrane module with DI water for at least 1 hour in order to (i) get rid of the fines released initially to maintain the particle size distribution during the tests, and (ii) saturate the porous rice media to maintain the apparent particle density during the tests. The saturated particle density of the rice media was approximately 1450 kg/m^3 for all three particle types. For each experiment, only one type of rice media was used (**Fig. 8-2a**; **Table 8-2**) at an overall concentration of 150 g/L. The model foulant particles used were monodisperse, spherical ($\phi \approx 1$) polystyrene (also known as latex) particles (Sigma Aldrich) each with a particle diameter of $5 \mu\text{m}$. The concentration used of the polystyrene particles was 0.005% (v/v) or 50 mg/L, and 0.02% sodium azide (Sigma Aldrich) was added as a bactericide. The feed suspension was continuously stirred at 250 rpm using a magnetic stirrer (AS ONE; REXIM RS-1D) during the experiments. Fresh rice and polystyrene particles were used for each experiment.

Figure 8-2. (a) Photograph of the three types of rice media used in the experiments; and (b) characteristic dimensions of an ellipsoid.

Table 8-2. Rice media parameters

Rice media	a (mm)	b (mm)	c (mm)	ϕ (-)	Volume (mm ³)	d_s (mm)
Type 1	4.19 ± 0.13	1.14 ± 0.04	0.90 ± 0.04	0.54 ± 0.03	18.0	3.25
Type 2	2.82 ± 0.09	1.01 ± 0.06	0.83 ± 0.03	0.60 ± 0.02	9.9	2.66
Type 3	2.66 ± 0.19	1.70 ± 0.20	1.11 ± 0.03	0.77 ± 0.04	21.0	3.42

8.2.3 Determination of local critical flux ($J_{c,local}$)

Critical flux is defined as the permeate flux below which negligible fouling occurs and above which foulants can be observed to deposit on the membrane [223]. In this study, the commonly used flux-stepping method was adopted to determine the local critical flux ($J_{c,local}$). Specifically, each permeate flux was held constant for 10 min, followed by the progressive increment by 10 L/(m² h) every 10 minutes until the polystyrene particles were seen to clearly

deposit on the membrane via DOTM. **Fig. 8-3** displays the images obtained at above the local critical flux ($J_{c,local}$) of membranes with deposited polystyrene particles. 10 images were analyzed at the end of the 1st and 10th minute of each permeate flux to calculate the average number of deposited polystyrene particles (N), after which the rate of change of N was computed:

$$\frac{dN}{dt} (\text{min}^{-1}) = \frac{N_{10^{\text{th}} \text{ minute}} - N_{1^{\text{st}} \text{ minute}}}{9 \text{ min}} \quad (8-2)$$

The local critical flux ($J_{c,local}$) was identified as the flux at which dN/dt starts to increase dramatically. Each experiment was repeated at least twice with new membranes.

Figure 8-3. DOTM images of the deposited polystyrene particles (pink circles) after 10 minutes at 86.9 L/(m²h), which was above the critical flux of 48.4 L/(m²h). Particle sphericity (ϕ) and equivalent spherical diameter (d_s) of the fluidized rice were 0.54 and 3.25 mm, respectively (Type 1). The images are 1 s apart.

8.2.4 Characterization of fluidization hydrodynamics

In previous chapters, the hydrodynamics of the fluidized GAC media and bubbles were correlated with the extent of fouling mitigation. This study focused on elucidating the effect of the sphericity of the fluidized media. Accordingly, the characteristics measured in the previous studies, namely, particle velocity

(V_p), particle number (N_p) and particle momentum (P_p), were also quantified. Furthermore, in view of the different rotation and contact with the membrane due to the lower sphericity (ϕ) of the fluidized rice media, two additional parameters were quantified, namely, rate of rotation (V_θ) and particle-membrane contact area (A_p). At least 600 particles were characterized for each parameter.

Because the fouling mitigation by fluidization necessitates that the rice media become fluidized, the determination of the minimum fluidization velocity (U_{mf}) of each particle type was warranted. This was enabled by a plot of fluidized bed height (L) with respect to superficial liquid velocity (U_l), as shown in **Fig. 8-4**. The minimum fluidization velocity (U_{mf}) is defined as the superficial liquid velocity (U_l) at which the fluidized bed height starts increasing [260], hence the U_{mf} values were determined as 0.020 m/s for the rice media type with the intermediate sphericity (ϕ) of 0.60 and smallest d_s (Type 2), and the same at 0.022 m/s for the other two rice media types with similar d_s values (Types 1 and 3). The U_{mf} values agree well with correlations developed for non-spherical particles [145, 261, 262]. **Fig. 8-4** also indicates that the bed heights differ among the fluidized rice media of different sphericities (ϕ), which agree with past reports on non-spherical particles [251, 252].

Figure 8-4. Fluidized bed height (L) versus superficial liquid velocity (U_l) for the three types of rice media for the determination of minimum fluidization velocities (U_{mf}).

High-speed videos capturing the motion of the fluidized rice media were taken at above the minimum fluidization velocities (U_{mf}) of each particle type, and first converted into frames 0.01 s apart using Image J. **Fig. 8-5** displays representative images 0.01 s apart. The centers of the particles in focus were designated with the co-ordinates (x, y), which are listed in **Table 8-3**. Only the particles in focus were accounted for since only those were in contact with the membrane to confer the scouring effects, and only the coordinates of the particles whose centers within the image were determined. In **Fig. 8-5**, 12 rice media were in focus in each of these images, and the coordinates and angles with respect to

the x -axis (θ) obtained via Image J are listed in **Table 8-3**. Armed with the data, the particle velocity (V_p) was calculated using the following equation:

$$V_p = \frac{\sqrt{(x_1 - x_2)^2 + (y_1 - y_2)^2}}{\Delta t} \quad (8-3)$$

where the subscripts 1 and 2 represent the same particle in the two images 0.01 s apart, and Δt represents the time lapse between the images which is 0.01 s in this case. The mean particle velocity ($V_{p,rms}$) was calculated from:

$$V_{p,rms} = \sqrt{\sum V_p^2 f(V)} \quad (8-4)$$

where $f(V)$ is the number-based fraction of particles characterized with a velocity of V_p . The standard deviation was also quantified:

$$\sigma_{V_p} = \sqrt{\sum (V_p - V_{p,rms})^2 f(V)} \quad (8-5)$$

In addition, the rotation speed (V_θ) was quantified using:

$$V_\theta = \frac{|\theta_1 - \theta_2|}{\Delta t} \quad (8-6)$$

and the mean ($V_{\theta,rms}$) and standard deviation (σ_{V_θ}) of the rotation speed calculated similarly to **Eqs. (8-4) and (8-5)**, respectively.

Figure 8-5. Images taken 0.01 s apart of the fluidized rice media ($\phi = 0.54$; $d_s = 3.25$ mm; Type 1) at h/H of 0.62 and superficial liquid velocity (U_l) of 0.061 m/s ($U_l/U_{mf} = 2.8$).

The contact area between the fluidized rice media and the membrane was measured using AutoCAD 2014. **Fig. 8-6** shows the processed images from **Fig. 8-5**, where the areas in contact with the membrane were colored green. The contact area (A_p) values, expressed as the green area normalized with respect to the image area assessed, are also listed in **Table 8-3**. At least 20 images at each condition were analyzed and averaged.

Figure 8-6. Processed images from Fig. 5, with the green color denoting the areas of the fluidized rice media that were in contact with the membrane.

Table 8-3. Particle behavioral characteristics obtained from **Figs. 8-5 and 8-6**.

Particles	$t = 0$ s (Figs. 8-5a and 8-6a)				$t = 0.01$ s (Figs. 8-5b and 8-6b)				Velocities	
	Particle coordinates		Angle	Contact area per unit membrane area	Particle coordinates		Angle	Contact area per unit membrane area	Particle velocity	Rotation velocity
	x_I	y_I	θ_I (°)	A_p (-)	x_2	y_2	θ_2 (°)	A_p (-)	V_p (mm/s)	V_θ (°/s)
1	8.46	7.00	27.95		8.22	6.59	23.10		47.1	484.8
2	-	-	58.98		-	-	58.73		-	25.0
3	-	-	9.03		-	-	3.32		-	571.5
4	13.62	4.81	33.17		13.69	4.66	33.24		16.0	7.3
5	-	-	63.74		-	-	61.06		-	268.1
6	13.18	1.29	19.29		12.60	0.97	25.22		65.9	593.4
7	-	-	48.58	0.42	-	-	45.73	0.41	-	285.1
8	5.68	4.69	69.68		5.57	4.33	69.94		36.7	25.5
9	8.18	4.06	49.12		8.42	3.53	48.21		58.3	91.3
10	3.00	3.58	26.71		3.06	3.47	27.02		12.5	30.2
11	-	-	33.18		-	-	31.19		-	198.7
12	-	-	80.12		-	-	74.55		-	556.2

The particle momentum per unit membrane area (P_p) was calculated by:

$$P_p = \frac{mV_p}{\text{membrane area assessed}} = \frac{n \frac{4\pi abc}{3} \rho_p V_p}{\text{membrane area assessed}} \quad (8-7)$$

where m is the mass of the particle (kg), n is the number of particles, $\frac{4\pi abc}{3}$ is the volume of an ellipsoid, and ρ_p is the saturated rice media density (kg/m³). Furthermore, the power required for fluidizing the rice media (W ; unit of kW) was quantified by:

$$W = \Delta P_{bed} Q \quad (8-8)$$

where ΔP_{bed} is the pressure drop across the fluidized bed (kPa) and Q is the volumetric flowrate (m³/s). The ΔP_{bed} could not be quantified due to scale and sensitivity limitations of the pressure transmitters, thus was calculated by:

$$\Delta P_{bed} = L(1 - \varepsilon_{mf})(\rho_p - \rho)g \quad (8-9)$$

where L is the height of the fluidized bed (m), ρ is the water density of 1000 kg/m³, g is gravitational acceleration of 9.8 m/s², and ε_{mf} is the voidage estimated from the empirical equation using the sphericity (φ) values obtained (Table 8-2) [147]:

$$\varepsilon_{mf} = \frac{0.415}{\varphi^{0.483}} \quad (8-10)$$

8.3. Results and Discussion

8.3.1. Local critical flux ($J_{c,local}$)

Fig. 8-7 shows the change in the trans-membrane pressure (TMP) and rate of change of the number of deposited polystyrene particles (dN/dt) during the flux-stepping experiment for the Type 1 rice media ($\varphi = 0.54$; $d_s = 3.25$ mm) at $U_l = 0.053$ m/s ($U_l/U_{mf} = 2.4$) and $h/H = 0.46$. The dN/dt versus flux plot in **Fig. 8-7a** indicates that the dN/dt value started increasing drastically beyond 48.4 L/(m²h), which implies that the local critical flux ($J_{c,local}$) value was 48.4 L/(m²h) in this case. Specifically, **Fig. 8-8** shows the DOTM images from which the dN/dt values were analyzed. **Fig. 8-8a**, an image taken at below the local critical flux ($J_{c,local}$), shows that the polystyrene particles were not in focus which implies they were not yet deposited on the membrane. Right at the local critical flux ($J_{c,local}$) of 48.4 L/(m²h), **Fig. 8-8b** shows that some polystyrene particles had deposited.

At above the local critical flux ($J_{c,local}$), **Fig. 8-8c** shows that many polystyrene particles had deposited and were clearly in focus. Regarding the conventional method of using TMP to monitor fouling, **Fig. 8-7b** shows that the TMP values fluctuated quite significantly even when the flux was constant, and no clear steep increase in TMP was observed even beyond the critical flux value determined via DOTM (**Fig. 8-7a**). Two noteworthy differences between our previous study on fluidized GAC in Chapter 5 and the current on the less spherical rice are (i) both the TMP and DOTM methods gave the same local critical flux ($J_{c,local}$) value in the case of GAC, but DOTM gave a lower $J_{c,local}$ than TMP (which agrees with previous reports [228, 230]) in this study; and (ii) the TMP fluctuations were lesser in the case of GAC. These disagreements are attributable to the different sphericities (ϕ) of the fluidized media, and underscores the non-negligible impact of sphericity (ϕ).

Figure 8-7. (a) Rate of change of the number of deposited polystyrene particles (dN/dt) with respect to permeate flux to determine the local critical flux ($J_{c,local}$), which was $48.4 \text{ L}/(\text{m}^2 \text{ h})$ in this case; and (b) Evolution of trans-membrane pressure (TMP) during the flux-stepping experiment. The superficial liquid velocity was 0.053 m/s (i.e., $U/U_{mf} = 2.4$), and ϕ and d_s of the fluidized rice were respectively 0.54 and 3.25 mm (Type 1), and h/H was 0.46 .

Figure 8-8. DOTM images at the end of the 10th minute at (a) below the local critical flux ($J_{c,local}$), (b) the local critical flux ($J_{c,local}$), and (c) above the local critical flux ($J_{c,local}$). The superficial liquid velocity was 0.053 m/s (i.e., $U_l/U_{mf} = 2.4$), and ϕ and d_s of the fluidized rice were respectively 0.54 and 3.25 mm (Type 1), and h/H was 0.46.

8.3.2. Effect of particle sphericity (ϕ) of fluidized rice media on local critical flux ($J_{c,local}$)

Fig. 8-9 shows the influence of the particle sphericity (ϕ) of the fluidized rice media on critical flux (J_c). The y-axes denote the local critical flux ($J_{c,local}$), while the x-axes are designated the superficial liquid velocity (U_l), with the ratio of superficial liquid velocity to minimum fluidization velocity (U_l/U_{mf}) varying between 2 and 2.8. Each row represents each height along the membrane module, namely, $h/H = 0.29, 0.46$ and 0.62 . The first column plots the local critical flux ($J_{c,local}$). The second column plots the ratio of the local critical flux to the overall critical flux ($J_{c,local}/J_{c,overall}$), whereby $J_{c,overall}$ is the arithmetic average of the local critical flux ($J_{c,local}$) at the three heights (h/H), to reflect the uniformity of fouling mitigation along the membrane module; the closer the critical flux ratio ($J_{c,local}/J_{c,overall}$) is to 1, the more uniform it is. Observations similar to previous chapters on fluidized GAC media include (i) the local critical flux ($J_{c,local}$) varied with height (h/H) and superficial liquid velocity (U_l) due to the varying local

hydrodynamics, and (ii) the uniformity of fouling mitigation along the membrane module improved (i.e., $J_{c,local}/J_{c,overall}$ values became closer to 1) as the superficial liquid velocity (U_l) increased. To understand the effect of particle sphericity (ϕ), comparison was first made between the similar-sized Types 1 and 3. Clearly, particle sphericity (ϕ) affected the local critical flux ($J_{c,local}$) values more significantly at the highest height ($h/H = 0.62$) evaluated, at which the fluidized rice with the lower ϕ (Type 1) gave lower $J_{c,local}$ values and thereby lesser fouling mitigation. Interestingly, although the smaller Type 2 was expected to give greater $J_{c,local}$ values because of the greater mobility stemming from the smaller U_{mf} (Chapter 3), Type 2 did not out-perform the larger Types 1 and 3, which indicates too that the lower sphericity (ϕ) of the fluidized media hampers fouling mitigation at the same U_l values. Type 2 performed similarly or worse than the larger media, which contrasts with the results in Chapter 5 and implies the adverse influence of lower sphericity due to the different fluidization behaviour of the media with different sphericities. The hydrodynamics of the fluidized rice media are examined in the next section to understand the underlying mechanisms.

Figure 8-9. Plots of local critical flux ($J_{c,local}$) and critical flux ratio ($J_{c,local}/J_{c,overall}$) versus superficial liquid velocity (U_l) for fluidized rice media with sphericity (ϕ) values of 0.54, 0.60 and 0.77 at various heights (h/H).

8.3.3. Effect of particle sphericity (ϕ) of fluidized rice media on hydrodynamics

The probability distributions of the particle velocity (**Eq. (8-3)**) of the three types of fluidized rice media at three heights (h/H) and three superficial liquid velocities (U_l) are displayed in **Fig. 8-10**. Each row represents a height along the membrane module, while each column represents a superficial liquid velocity. Only Type 2 is plotted in the top-left sub-figure ($h/H = 0.62$, $U_l = 0.049$ m/s) because the fluidized bed heights of the other two larger rice media types were lower, and Type 2 is missing from the rightmost column ($U_l = 0.049$ m/s).

due to entrainment issues at this highest velocity. The impact of particle sphericity (ϕ) on the distribution of particle velocity (V_p) is evident in two observations. Firstly, the particle velocity (V_p) distributions generally were broader at higher heights (h/H) and higher superficial liquid velocities (U_l), which contrasts somewhat with our previous study on the more spherical GAC media ($\phi = 0.83$) (Chapter 4), in which the distributions narrowed with height. This highlights an important effect of particle sphericity (ϕ). Secondly, comparing Types 1 and 3 which had more similar d_s values, the lower-sphericity Type 1 behaved such that the particle velocity (V_p) distributions were (i) shifted leftwards at the lowest superficial liquid velocity ($U_l = 0.049$ m/s; leftmost column in **Fig. 8-10**) due to the increased drag of this particle type which had the highest aspect ratio (**Fig. 8-2a**), and (ii) broadened at the highest height ($h/H = 0.62$; topmost row in **Fig. 8-10**).

Figure 8-10. Probability distributions of particle velocity (V_p).

In view of the rotational and preferred orientation effects associated with non-spherical particles [250, 263], the distributions of the rotation velocity (V_θ ; Eq. (8-6)) were also examined. Analogous to Fig. 8-10, Fig. 8-11 presents the probability distributions of the rotation velocity (V_θ) of the three types of fluidized rice media at three heights (h/H) and three superficial liquid velocities (U_l). Generally, the lowest-sphericity (ϕ) Type 1 gave the narrowest rotational velocity (V_θ) distributions that are most leftwards, which implies lowest rotation velocities (V_θ). This agrees with a simulation study which indicated that the particle orientation was more random with increased sphericity [250].

Figure 8-11. Probability distributions of particle rotation velocity (V_θ).

Fig. 8-12 summarizes Figs. 8-10 and 8-11 by presenting the average particle velocity ($V_{p,rms}$; Eq. (8-4)) and average particle rotation velocity ($V_{\theta,rms}$), respectively, as functions of superficial liquid velocities (U_l). The error bars represent the standard deviations of the distributions. With respect to average

particle velocity ($V_{p,rms}$; left column of **Fig. 8-12**), (i) the effect of particle sphericity (ϕ) was more significant at higher heights (h/H), at which the $V_{p,rms}$ of the lower-sphericity (ϕ) Type 1 was greater than Type 3, although the former gave relatively lower $J_{c,local}$ (**Fig. 8-9**); (ii) the $V_{p,rms}$ of the most-spherical Type 3 was most invariant with superficial liquid velocity (U_l), which agrees with our previous study in which the fluidized GAC media ($\phi = 0.83$) also exhibited a weak correlation between V_p and U_l (Chapter 4); and (iii) the smaller Type 2 fluidized media gave similar $V_{p,rms}$ values, which agrees with the previous study on the negligible effect of size on $V_{p,rms}$ (Chapter 4). As for average particle rotation velocity ($V_{\theta,rms}$; right column of **Fig. 8-12**), (i) between Types 1 and 3, the lower-sphericity (ϕ) Type 1 gave lower $V_{\theta,rms}$ values because the orientation of each rice media was most stable and thereby least subject to any rotational effects [259], and (ii) Type 2 displayed greater $V_{\theta,rms}$ values than the higher-sphericity Type 3 at the higher heights ($h/H \geq 0.46$), which is evident in the overlapping V_{θ} distributions in **Fig. 8-11** and indicates that the smaller sizes enhanced V_{θ} .

Figure 8-12. Plots of average particle velocity ($V_{p,rms}$) and average particle rotation velocity ($V_{\theta,rms}$) versus superficial liquid velocity (U_l) for fluidized rice media with sphericity (ϕ) values of 0.54, 0.60 and 0.77 at various heights (h/H).

Fig. 8-13 further shows the impact of particle sphericity (ϕ) on contact area per unit membrane area (A_p ; **Fig. 8-6**) and particle momentum per unit membrane area (P_p ; **Eq. (8-7)**) as functions of superficial liquid velocities (U_l) at various heights (h/H). Regarding contact area (A_p), (i) it tended to decrease with superficial liquid velocity (U_l) for all three particle types at the lower heights ($h/H \leq 0.46$), and (ii) the effect of sphericity (ϕ) was only straightforward at the intermediate height ($h/H = 0.46$) whereby the lower-sphericity Type 1 consistently gave higher A_p values than Type 3, and that of the intermediate-sphericity Type 2 was in between the other two types, which suggests the dominance of sphericity effects over size effects. As for particle momentum (P_p),

it should be noted that this parameter has been reported to be most closely related to the extent of fouling mitigation (Chapters 4 and 6). Two observations are highlighted. Firstly, between similar-sized Types 1 and 3, the lower-sphericity Type 1 consistently gave lower P_p values than Type 3 at the lower heights ($h/H \leq 0.46$), although **Fig. 8-9** indicates similar $J_{c,local}$ at these positions. This appears to contradict earlier results in Chapter 4 on fluidized GAC media in which higher P_p values are tied to better fouling mitigation (i.e., higher $J_{c,local}$), and also another study in Chapter 6 on bubbles, which indicated that both the bubble momentum (P_p) and contact area with the membrane (A_p) were closely tied to fouling mitigation. This underscores the need to better understand sphericity (ϕ) effects. Secondly, although the smaller GAC media was shown to exhibit greater momentum (P_p) (Chapter 4), the smaller Type 2 gave similar or lower momentum (P_p) values than the larger Types 1 and 3. This again indicates that previous knowledge on the more spherical fluidized media cannot be directly applied to less spherical media like those in this study.

Figure 8-13. Plots of contact area per unit membrane area (A_p) and particle momentum per unit membrane area (P_p) versus superficial liquid velocity (U_l) for fluidized rice media with sphericity (ϕ) values of 0.54, 0.60 and 0.77 at various heights (h/H).

8.3.4. Correlating membrane fouling mitigation with hydrodynamics

Fig. 8-14 relates the extent of fouling mitigation to the various hydrodynamics parameters for the three types of fluidized rice media of various particle sphericities (ϕ). All the y-axes represent the local critical flux ($J_{c,local}$), with each error bar denoting the span of $J_{c,local}$ values obtained from repeated experiments. Each column represents each rice type. Each row represents a hydrodynamics characteristic, namely, average particle velocity ($V_{p,rms}$), average

particle rotation velocity ($V_{\theta,rms}$), contact area per unit membrane area (A_p), and particle momentum per unit membrane area (P_p). The axes ranges are the same for all 12 sub-figures to ease comparison. Two contrasts with our studies in Chapters 4 and 6 are notable, which emphasizes that particle sphericity (ϕ) is an important particle property of the fluidized media that affects the effectiveness of fouling mitigation. Firstly, in contrast to our previous study in which fouling mitigation improved with particle or bubble velocities, the extent of fouling mitigation decreased as the particle velocities ($V_{p,rms}$) increased. At first sight this appears counter-intuitive because higher velocities presumably conferred greater mechanical scouring effects, but the underlying reason may be that high particle velocities were ineffectual when particle concentrations were too low in Chapter 4, as evident in the decrease of A_p with respect to U_l in **Fig. 8-13**. Whereas the local critical flux ($J_{c,local}$) was negatively correlated with particle velocity ($V_{p,rms}$), it was clearly positively correlated with the contact area per unit membrane area (A_p), which includes both the effects of particle concentration and the increased contact area due to the lower particle sphericity, thereby resulted in a surprising weak relationship between $J_{c,local}$ and the particle momentum per unit membrane area (P_p). Secondly, among the four hydrodynamics parameters, the local critical flux ($J_{c,local}$) was surprisingly most weakly correlated with the particle momentum per unit membrane area (P_p), which contrasts with our studies in Chapters 4 and 6 on fluidized GAC media ($\phi = 0.83$) and bubbles, in which momentum had the most positive relationship with critical flux. The results here hence imply that the non-spherical particles mitigated fouling not by virtue of particle velocity (V_p) or momentum (P_p), but instead by the contact area between

the particles and the membrane (A_p). This is in stark contrast with the more spherical GAC media by which particle momentum was the main contributor in Chapter 4 .

To scrutinize more into the effect of particle sphericity (ϕ), the regression slopes of Types 1 and 3 were compared. It is observed that particle sphericity (ϕ) had (i) negligible impact on the relationship between the local critical flux ($J_{c,local}$) and contact area per unit membrane area (A_p) or particle momentum per unit membrane area (P_p), as evident in the similar gradients of the trendlines, and (ii) significant impact on the relationships between the local critical flux ($J_{c,local}$) and both the particle velocity ($V_{p,rms}$) and particle rotation velocity ($V_{\theta,rms}$). More specifically on (ii), as particle sphericity decreased, the relationship between $J_{c,local}$ and $V_{p,rms}$ became less negative, while the relationship between $J_{c,local}$ and $V_{\theta,rms}$ became more negative.

Figure 8-14. Correlating the local critical flux ($J_{c,local}$) with mean particle velocity ($V_{p,rms}$), mean particle rotation velocity ($V_{\theta,rms}$), contact area per unit membrane area (A_p), and particle momentum per unit membrane area (P_p) for the fluidized rice media of different sphericities (ϕ).

8.3.5. Overall critical flux ($J_{c,overall}$) with respect to hydrodynamics parameters and energy requirement

Fig. 8-15 attempts to summarize the results to conclude on the benefits or lack thereof in the fluidization of non-spherical particles for membrane fouling mitigation. The overall critical flux ($J_{c,overall}$) was correlated with the overall particle velocity ($V_{p,overall}$), overall particle rotation velocity ($V_{\theta,overall}$), overall

contact area per unit membrane ($A_{p,overall}$) and overall particle momentum per unit membrane ($P_{p,overall}$). Two interesting observations are noted in **Fig. 8-15**. Firstly, between the similar-sized Types 1 and 3, the lower-sphericity Type 1 generally gave lower $J_{c,overall}$ values for the same hydrodynamic parameter (namely, $V_{p,overall}$, $V_{\theta,overall}$, $A_{p,overall}$), except $P_{p,overall}$, which indicates that the less spherical media were less effective in mitigating fouling given the same hydrodynamics behavior. Since the fluidized rice media were typically oriented with the longest dimension (a in **Fig. 8-2b**) approximately horizontal, the least spherical media experienced the greatest drag [254, 255], which diminished the mechanical scouring. Secondly, regarding the smaller Type 2, the generally lower $J_{c,overall}$ are consistent with the effect of size in Chapter 4, which indicates the dominance of the size effect over the sphericity effects. Collectively, the results indicate that, for the same hydrodynamic parameter, increased sphericity (ϕ) of the fluidized media improved fouling mitigation when the media were of similar sizes, but the effect of size was more dominant than that of sphericity when the media were of different sizes.

Figure 8-15. Correlating overall critical flux ($J_{c, overall}$) with (a) overall particle velocity ($V_{p, overall}$), (b) overall particle rotation velocity ($V_{\theta, overall}$), (c) overall particle contact area per unit membrane area ($A_{p, overall}$), and (d) overall particle momentum per unit membrane area ($P_{p, overall}$).

Finally, **Fig. 8-16** relates the overall critical flux ($J_{c, overall}$) with power requirement (P_r ; **Eq. (8-8)**) for the three rice media types investigated in this study, along with that of the more spherical GAC media ($\phi = 0.83$) reported in Chapter 5 at a lower concentration of 100 g/L. The power required were greater for the rice media compared to the GAC media, because the former had larger d_s values and were at a higher overall concentration. Somewhat surprisingly, although the smaller GAC media gave significantly greater $J_{c, overall}$ values at the same power (P_r), the smaller Type 2 rice media instead gave lower $J_{c, overall}$ values than the larger Type 1 at the lower P_r range. This indicates that the lower

sphericity (ϕ) of the Type 1 rice media aided to enhance the fouling mitigation so much that the size effects were overcome. On the other hand, at the higher P_r range, the effect of sphericity (ϕ) was diminished. This is presumably because the change in drag coefficient with respect to increased U_l became lesser at higher U_l [264, 265]. The results here therefore conclude that (i) the effect of sphericity (ϕ) of the fluidized media on $J_{c,overall}$ was such that, sphericity effects dominated over size effects at the lower P_r range, with the lower-sphericity, larger Type 1 media giving greater $J_{c,overall}$ values, but sphericity effects were inconsequential at the higher P_r range, and (ii) an increase in power input (P_r) was more beneficial for the higher-sphericity GAC media than the lower-sphericity rice media, as evident in the steeper slope for the former.

Figure 8-16. Correlating overall critical flux ($J_{c,overall}$) with power requirement (P_r) for the fluidized rice media and GAC media (higher sphericity (ϕ) of 0.83; lower overall concentration of 100 g/L in Chapter 5).

8.4. Conclusions

This current study systematically studied the influence of the particle sphericity (ϕ) of millimeter-sized fluidized media on the critical flux (J_c) of micron-sized particulate foulant (namely, polystyrene particles) in microfiltration using Anopore membranes. Specifically, of the three rice media types, two (namely, Types 1 and 3) were of similar sizes but different sphericity, while one (namely, Type 2) differed in both size and sphericity. The direct observation through membrane (DOTM) method was used to measure the local critical flux ($J_{c,local}$) at three heights along the membrane module, while a high-

speed video camera was used to capture the local fluidization hydrodynamics (namely, particle velocity (V_p), particle rotation velocity (V_θ), particle contact area (A_p) and particle momentum (P_p)). The relationships between critical flux and the hydrodynamic parameters were determined.

The results here indicate clearly that the sphericity (ϕ) of the fluidized media impacted fouling mitigation under some conditions. However, the correlation between sphericity and the extent of fouling mitigation (and the attendant hydrodynamics) was not straightforward, likely tied to the influence of drag on the countering effects of media-membrane contact and particle rotation. It should be noted that the fluidized rice media were typically oriented with the longest dimension (a in **Fig. 8-2b**) approximately horizontal. The effect of sphericity on the hydrodynamics are summarized here. Firstly, where sphericity had an impact, lower particle sphericity (ϕ) gave lower local critical flux ($J_{c,local}$) values at the same U_l values, which indicates lesser fouling mitigation. Comparing the similar-sized Types 1 and 3, Type 1 with the lower ϕ (Type 1) gave lower $J_{c,local}$ values at the highest height ($h/H = 0.62$) evaluated. Also, although the smaller Type 2 was expected to give greater $J_{c,local}$ values because of the greater mobility stemming from the smaller U_{mf} (Chapter 3), Type 2 did not out-perform the larger Types 1 and 3. Secondly, in contrast to our results in Chapters 4 and 6, the local critical flux ($J_{c,local}$) decreased as the particle velocities ($V_{p,rms}$) increased, and was surprisingly most weakly correlated with the particle momentum per unit membrane area (P_p). The results here hence imply that the non-spherical particles mitigated fouling not by virtue of particle velocity (V_p) or momentum (P_p), but instead by the contact area between the

particles and the membrane (A_p). Thirdly, particle sphericity (ϕ) had negligible impact on the relationship between the local critical flux ($J_{c,local}$) and contact area per unit membrane area (A_p) or particle momentum per unit membrane area (P_p), but significant impact on the relationships between the local critical flux ($J_{c,local}$) and both the particle velocity ($V_{p,rms}$) and particle rotation velocity ($V_{\theta,rms}$). Fourthly, for the same hydrodynamic parameter, increased sphericity (ϕ) of the fluidized media improved fouling mitigation when the media were of similar sizes, but the effect of size was more dominant than that of sphericity when the media were of different sizes.

Two key results pertaining to membrane fouling mitigation are worth highlighting. Firstly, at the same power input (P_r), the lower-sphericity but larger Type 1 media gave greater $J_{c,overall}$ values than Type 2 at the lower P_r range, indicating that the beneficial lower-sphericity effects dominated over the beneficial smaller-size effects reported in Chapter 5, but sphericity effects were inconsequential at the higher P_r range. Secondly, an increase in power input (P_r) was more beneficial for the higher-sphericity GAC media than the lower-sphericity rice media, as evident in the steeper $J_{c,overall}$ versus P_r slope for the former. The findings here are expected to be valuable in the optimization of the properties of the fluidized media towards more energy-efficient membrane fouling mitigation.

Chapter 9 Monitoring local membrane fouling mitigation by fluidized GAC in lab-scale and pilot-scale AnFMBRs⁷

9.1 Introduction

For fluidized bed systems, experimental measurement of the behavior relies on a host of instruments that can be largely characterized into invasive and non-invasive probes [189, 266-268]. Because the more common invasive probes tend to disrupt the hydrodynamics that the probe is supposed to be measuring, non-invasive ones are preferred, although issues with lower resolution are acknowledged. Thereby, non-intrusive measurements of vibration signals can provide more reliable information for hydrodynamics characterization. Abbasi et al. [269] employed vibration signal analysis method to characterize the hydrodynamic of gas–solid fluidized beds (i.e., formation of large bubbles, small bubbles and clusters using frequency components of vibration signal). The energy of accelerometer signals via wavelet decomposition distribution helped to show which sub-signal was dominant at different conditions. Sheikhi et al. [270] shown that vibration inspection can also be used as a reliable method for the hydrodynamic characterization of liquid-solid fluidized beds, i.e., measurement of minimum fluidization velocity (U_{mf}) and solid regime transition. A more recent

7. The content in this chapter has been published as:

Jingwei Wang, Bing Wu, Yu Liu, Anthony G. Fane, Jia Wei Chew. Monitoring local membrane fouling mitigation by fluidized GAC in lab-scale and pilot-scale AnFMBRs. *Separation and Purification Technology*, volume 199, 30 June 2018, Pages 331-345.

study by Sheikhi et al. [271] examined the due effect of solid particles on the local hydrodynamics of a three phase gas-solid-liquid fluidized bed using the energy of accelerometer signals, which had a good consistency with the visual observations. These provide the ability of accelerometer to characterize the hydrodynamics of fluidized media. Regarding the characterization of the hydrodynamics of the fluidized GAC within the AnFMBR, the non-invasive accelerometer has been shown in Chapter 3. Specifically, the accelerometer signals had to be wavelet-decomposed into various frequency ranges, which contains information on the various aspects of the fluidization behavior [272-277], including the energy components corresponding to solid and liquid shear in this case. Thereby, the extraction of the frequency range corresponding to the fluidized GAC dynamics allowed for understanding of the relationship between the mechanical scouring efficiency and the mitigation of membrane fouling.

Correspondingly, the current study was targeted at evaluating the feasibility of using the accelerometer for characterizing and differentiating the hydrodynamics of the fluidized GAC (i) at different positions (i.e., various heights and sides) along the reactor and under different conditions (i.e., various overall GAC concentrations and superficial liquid velocities), (ii) for an idealized pilot-scale reactor with DI water and new membranes, and (iii) for a pilot-scale reactor treating wastewater.

9.2 Materials and methods

9.2.1 Lab-scale setup

The schematic of the lab-scale setup is shown in **Fig. 9-1**, which was similar to that in Chapter 4. Specifically, the anaerobic fluidized bed – membrane bioreactor (AnFMBR) module had two distinct portions: (i) a smaller cross-section (89 mm width by 33 mm depth) with a height of 194 mm, where the membrane cell was immersed vertically aligned and the GAC particles were fluidized, and (ii) an upper section with larger cross-sectional area (150 mm width by 93 mm depth) with a height of 124 mm, whose purpose was to reduce the entrainment of the fluidized GAC particles. The flat-sheet membrane cell, which had a cross-section of 78 mm by 15 mm and height of 146 mm, was positioned flush with the wall and held the membrane to expose an active area of 46 mm by 87 mm. The distributor used was of the same design as studies in Chapter 3 and 4 and as the pilot-scale AnFMBR (**Fig. 9-2**). The design of the distributor was such that eight pairs of nozzles were projected downwards in an inverted-V configuration, and spread evenly along a central pipe traversing the reactor bottom. The water flow rate needed to fluidize the GAC particles was provided by a peristaltic pump (Cole-Parmer, 7528-10), and measured by a variable-area flowmeter (Cole-Parmer, 1.5-20 L/min).

A piezoelectric accelerometer sensor (National Instrument; sensitivity of 100.6 mV/g) was employed to capture the characteristic hydrodynamics non-invasively to avoid any disruption to the fluidization. The same accelerometer was used for both lab- and pilot-scales. Specifically, the accelerometer signals

were taken at 9 positions, namely, at three different heights (i.e., corresponding to the top, bottom and center of the membrane) of three sides of the reactor (i.e., west, south, and east, as marked in **Fig. 9-1**). The three heights characterized were at h/H (where h is the height assessed and H is the total lab-scale reactor height of 194 mm) values of 0.28, 0.57 and 0.85, and the north side was not characterized as it was the side the membrane cell was flush with.

Figure 9-1. Schematic of the lab-scale AnFMBR reactor. The black dots on the reactor represent positions where accelerometer signals were taken; signals at different sides of the reactor (namely, West, South and East) were taken to determine any mal-distribution of the fluidized GAC particles.

9.2.2 Pilot-scale setup

Figs. 9-2 and 9-3 show respectively the schematic and photograph of the pilot-scale two-stage AnFMBR studied. The raw sewage wastewater from the Ulu Pandan Water Reclamation Plant (Singapore) first entered the clarifier to remove the larger solids, then the first-stage anaerobic fluidized bed reactor

(AnFBR; i.e., no membrane), followed by the second-stage AnFMBR with either flat-sheet or hollow-fiber membranes. The unit of interest in this study is the second-stage AnFMBR with flat-sheet membranes, which is geometrically similar to that in the lab-scale setup (**Fig. 9-1**), as schematically shown in **Fig. 9-4a**. Specifically, the reactor also had two parts, with the lower having a smaller cross-sectional area (1450 mm by width and 664 mm by depth) and height of 1590 mm, and the upper having a larger cross-sectional area (1800 mm by width and 1070 mm by depth) and height of 503 mm. The unit had 70 sheets of double-sided flat-sheet membranes vertically aligned in parallel, as photographed in **Fig. 9-4b**. As with the lab-scale AnFMBR, the same accelerator probe was used to non-invasively monitor the hydrodynamics of the particle fluidization at four different heights and four different sides (i.e., west, south, east and north) of the reactor. The four heights characterized were at h/H (where h is the height assessed and H is the total pilot-scale reactor height of 1590 mm) values of 0.12, 0.33, 0.57, and 0.81, respectively.

Figure 9-2. Schematic of the pilot-scale two-stage AnFMBR [13], comprising of the first-stage AnFBR and two parallel second-stage AnFMBRs, with one using flat-sheet membranes (FS-AnFMBR) and the other hollow-fiber membranes (HF-AnFMBR).

Figure 9-3. Photograph of the pilot-scale two-stage wastewater treatment [13] facility housed in the Ulu Pandan Water Reclamation Plant (Singapore), comprising of the first-stage AnFBR and two parallel second-stage AnFMBRs, with one using flat-sheet membranes and the other hollow-fiber membranes.

Figure 9-4. (a) Dimensions of the pilot-scale AnFMBR with flat-sheet membranes (FS-AnFMBR); and (b) photograph of the flat-sheet membrane cassette inside the FS-AnFMBR (view from the south side).

Table 9-1. Operating conditions for the pilot-scale two-stage wastewater treatment.

Parameter	AFBR	HF-AnFMBR	FS-AnFMBR
Reactor volume (m ³)	2	1	1
Membrane module	-	PVDF (0.03 μm)	PVDF (0.08 μm)
GAC packing amount (kg)	250	150	150
Inoculated anaerobic sludge (g, dry weight)	850	0	0
Recirculation flowrate (m ³ /h)	12.8	30	42
HRT (h)	2	2	2
Feed flowrate (m ³ /day)	24	12	12
Designed permeate flux (L/(m ² h))	-	5.5	5.5

9.2.3 Materials

For the lab-scale setup (**Fig. 9-1**), the GAC particles (FILTRASORB 300, Calgon corporation, Pittsburgh, PA USA) obtained, which had a wide particle size distribution, were first sieved into various size fractions using Cole-Parmer U.S. Standard Brass Test Sieves. The size range of 2.18 ± 0.18 mm was used in this study since it had the highest mass fraction and thereby was the most representative size. As for the pilot-scale setup (**Figs. 9-2 and 9-3**), the GAC particles were used as obtained without sieving.

For the lab-scale setup, PVDF flat-sheet membranes with a nominal pore size of $0.22\ \mu\text{m}$ (Merck Millipore) were used. As for the pilot-scale, PVDF hollow fiber membranes with a nominal pore size of $0.03\ \mu\text{m}$ and flat-sheet membranes with a nominal pore size of $0.08\ \mu\text{m}$ were used in the AnFMBRs. The vendor is not mentioned here due to confidentially agreements.

The feed solution was DI water for the lab-scale setup, and either DI water or raw sewage wastewater from the Ulu Pandan Water Reclamation Plant (Singapore) for the pilot-scale setup.

9.2.4 Accelerometer signals

Accelerometer signals were recorded on a computer via the Labview 2013 software for 30 s at each position investigated. The data acquisition frequency was 40,000 Hz, which was consistent with results in Chapter 3 and re-affirmed in this study to be adequate for distinguishing between the solid and liquid phases.

The accelerometer signals as a function of time for the lab-scale AnFMBR and pilot-scale FS-AnFMBR are shown in **Figs. 9-5 and 9-6**, respectively, at approximately mid-height of the reactors. Both contained DI water. In each figure, each row represents each side of the reactor where the signals were collected, while the left and right columns represent respectively the absence and presence of fluidized GAC. Three observations in **Figs. 9-5 and 9-6** are worth highlighting: (i) the means of the signals were similar whether or not GAC was present, which agrees with the study in Chapter 3 and necessitates an alternative method of analyzing the fluidization hydrodynamics rather than a simple averaging; (ii) particularly in the presence of fluidized GAC, the amplitudes of the acceleration values were much higher for the lab-scale than pilot-scale reactor, presumably because of the different material (acrylic and stainless steel for the lab-scale and pilot-scale reactors, respectively) and thickness of the reactor walls, and the different arrangements of the flat-sheet membranes (**Fig. 9-4b**); and (iii) despite the similar means of approximately zero for all signals in **Figs. 9-5 and 9-6**, the amplitudes of the fluctuations were different in the absence and presence of fluidized GAC, and among the different sides of the reactors.

Figure 9-5. Accelerometer signals obtained in lab-scale reactor as a function of time at h/H of 0.57, superficial liquid velocity (U_l) of 0.033 m/s, overall GAC concentration of 150 g/L, and permeate flux of 0 L/(m² h). Each row represents each side of the reactor that the signals were collected at, while the left and right columns represent respectively the absence and presence of fluidized GAC.

Figure 9-6. Accelerometer signals obtained in pilot-scale FS-AnFMBR (with a new membrane cassette and DI water) as a function of time at h/H of 0.57, inlet flow rate of $67 \text{ m}^3/\text{h}$, 50 kg of GAC particles, and permeate flux of $0 \text{ L}/(\text{m}^2 \text{ h})$. Each row represents each side of the reactor that the signals were collected at, while the left and right columns represent respectively the absence and presence of fluidized GAC.

In order to unveil the fluidization hydrodynamics to correlate with the extent of membrane fouling mitigation, wavelet decomposition was employed in this study, as was also the case in the study of Chapter 3. Specifically, wavelet decomposition is a mathematical method for extracting the information in various frequency ranges so as to understand the dominance of the various constituent

dynamics in fluidized bed systems [273-275, 277]. As described in **Fig. 9-7**, the raw signal is broken down into higher-frequency detail (D_1) and lower-frequency approximation (A_1) signals at the first scale, then A_1 is further broken down into higher-frequency detail (D_2) and lower-frequency approximation (A_2) signals at the second scale, and so on. Therefore, as the scale increases, the frequency represented decreases. To quantify the dominance of each constituent frequency range in the original signal, the energy of the detail signal at each scale (D_j) can be calculated as follows [272, 276]:

$$E_{D_j} = \sum_{t=1}^N |D_j(t)|^2 \quad (9-1)$$

where E is the energy of the signal, D_j is the detail signal at the j^{th} scale of decomposition, N is the total number of data points and t is the time index of the signal.

Figure 9-7. Wavelet decomposition analysis of accelerometer signal into various scales [272].

9.3. Results and Discussion

9.3.1. Wavelet decomposition of accelerometer signals

Figs. 9-8 and 9-9 display the energy of the detail signal (E_{D_j} ; **Eq. (9-1)**) obtained via wavelet decomposition with respect to scale for the lab-scale AnFMBR and pilot-scale FS-AnFMBR (with a new membrane cassette and DI water), respectively. Notably, each sub-figure in **Figs. 9-8 and 9-9** corresponds to those of the accelerometer signals in **Figs. 9-5 and 9-6**, respectively. The x - and y -axes are of the same ranges for easier comparison. Two noteworthy observations in **Fig. 9-8** are described as follows. Firstly, a dominant peak is clear at the scales of 9 – 12 in the absence of GAC (first column of **Fig. 9-8**), while the dominant peak is at the lower scales of 1 – 3 in the presence of fluidized GAC (second column of **Fig. 9-8**). The peak at the higher scale corresponds to the liquid dynamics (since GAC is absent), while that at the lower scale to the fluidized GAC, whose signals drown out that of the liquid. This is consistent with the study in Chapter 3, and the physical understanding that the liquid and solid phases contribute to the higher and lower frequency ranges, respectively. Secondly, different sides of the reactor gave different E_{D_j} values, which suggests non-uniformity of the fluidization and agrees with an earlier simulation study [247]. In particular, the profiles at the west side had the highest peak E_{D_j} values, while that at the east lowest. The non-uniform fluidization hydrodynamics likely stems from the distributor, which caused the most turbulent liquid upflow and extensive fluidization closer to the liquid inlet at the west side (**Fig. 9-1**), and relatively poorer upflow further from the liquid inlet at the east side. On the one

hand, a secondary peak corresponding to the liquid is clear even in the presence of fluidized GAC at the west side (**Fig. 9-8d**), because of the robust liquid upflow (**Fig. 9-8a**). On the other hand, a secondary peak is clear in the presence of fluidized GAC at the east side (**Fig. 9-8f**), but in this case was due instead to poor liquid upflow (**Fig. 9-8c**).

Figure 9-8. Energy of the detail signal at each scale (E_{Dj} ; Eq. (9-1)) obtained via wavelet decomposition with respect to scale (j) for the lab-scale accelerometer signals in Fig. 9-5. Specifically, the accelerometer signals were obtained in lab-scale reactor as a function of time at h/H of 0.57, superficial liquid velocity (U_l) of 0.033 m/s, overall GAC concentration of 150 g/L, and permeate flux of 0 L/(m² h). Each row represents each side of the reactor that the signals were collected at, while the left and right columns represent respectively the absence and presence of fluidized GAC.

The E_{D_j} versus scale plots in **Fig. 9-9** for the pilot-scale reactor shows four interesting differences compared to the lab-scale reactor (**Fig. 9-8**). It should be noted that the two reactors were geometrically similar and had the same distributor design, but the reactor walls and arrangement of the flat-sheet membranes were different, which caused the amplitudes of the accelerometer signals to be different (**Figs. 9-5 and 9-6**). It should also be noted that the similarity between **Figs. 9-8 and 9-9** is that the liquid and solid phases correspond to the higher and lower frequency ranges, respectively. Firstly, the scale ranges corresponding to the liquid phases were different, but that for the solid was the same. Specifically, the scale range representing the liquid phase was 9 – 12 for the lab-scale reactor (first column of **Fig. 9-8**), but 5 – 10 for the pilot-scale reactor (first column of **Fig. 9-9**), while the range was 1 – 3 for the solid phase in both cases. Secondly, despite the liquid inlet being on the west sides for both the lab-scale (**Fig. 9-1**) and pilot-scale reactors (**Fig. 9-4a**), the east side had the poorest fluidization in terms of lowest E_{D_j} values for the lab-scale reactor (**Fig. 9-8f**), but the east had better fluidization than the south for the pilot-scale reactor (**Figs. 9-9g and f**). Thirdly, the mal-distribution was such that liquid signals were almost absent for the north side (**Fig. 9-9d**), while the solid signals were so instead for the east side (**Fig. 9-9g**), which disagrees with that for the lab-scale reactor (**Fig. 9-8**), whereby higher peak E_{D_j} values in the absence of GAC (first column of **Fig. 9-8**) gave higher peak E_{D_j} values in the presence of GAC (second column of **Fig. 9-8**). This suggests that the presence of the fluidized GAC changed the trajectory of the liquid flow too in the pilot-scale reactor, which

was likely due to the arrangement of the membranes (**Fig. 9-4b**). Fourthly, in the presence of fluidized GAC (second column of **Fig. 9-9**), the energies of the detail signals corresponding to the liquid phase were higher than that of the solid phase at the west and south sides (**Figs. 9-9e and f**). This disagrees with the lab-scale reactor, whereby the contribution of the solid phase was distinctly higher in the presence of fluidized GAC (second column of **Fig. 9-8**). As noted earlier, the differences in the trends between the lab-scale and pilot-scale are presumably due to the different arrangements of the flat-sheet membranes (**Fig. 9-1** versus **Fig. 9-4b**) and the different scales, despite the two reactors being geometrically similar.

Figure 9-9. Energy of the detail signal at each scale (E_{Dj} ; Eq. (9-1)) obtained via wavelet decomposition with respect to scale (j) for the pilot-scale accelerometer signals in Fig. 9-6. Specifically, the accelerometer signals were obtained in pilot-scale FS-AnFMBR (with a new membrane cassette and DI water) as a function of time at h/H of 0.57, inlet flow rate of $67 \text{ m}^3/\text{h}$, 50 kg of GAC particles, and permeate flux of $0 \text{ L}/(\text{m}^2 \text{ h})$. Each row represents each side of the reactor that the signals were collected at, while the left and right columns represent respectively the absence and presence of fluidized GAC.

9.3.2. Using the accelerometer signals from a lab-scale reactor to study the effect of superficial liquid velocity (U_l) and overall GAC concentration

Since **Figs. 9-8 and 9-9** indicate that the solid phases corresponded to the scales of 1 – 3, the sum of the energies of the details at the three scales (E_{D1} , E_{D2} and E_{D3}) was assessed to understand the effectiveness of the GAC fluidization in mitigating fouling. Higher $E_{D1} + E_{D2} + E_{D3}$ values have been reported to correlate well with the extent of fouling mitigation in Chapter 3, but the different efficacy at different sides of the reactor and the ability of the accelerometer signals to provide such information for a pilot-scale reactor has not been evaluated to date.

Fig. 9-10 displays the dimensionless height (h/H) versus the sum of the E_{D1} , E_{D2} and E_{D3} values for a constant overall GAC concentration of 200 g/L for the three sides of the lab-scale reactor investigated (**Fig. 9-1**). The three sub-plots represent three superficial liquid velocities (U_l), with the range adopted such that the lowermost ensured that the fluidized GAC contacted the membrane, while the uppermost was to ensure negligible entrainment. The ranges of the x - and y -axes of the sub-plots are the same for easier comparison. Expectedly, the $E_{D1} + E_{D2} + E_{D3}$ values increased as U_l increased or as h/H decreased, both of which have been linked to increased particle momentum and thereby increased extent of fouling mitigation, as studied in Chapters 3-8, although by different extents for the different sides. At the lowest U_l of 0.024 m/s (**Fig. 9-10a**), the $E_{D1} + E_{D2} + E_{D3}$ values were consistently largest on the south side and approximately similar on the other two sides, which indicates the best scouring efficiency on the south side presumably due to proximity to the distributor nozzles (**Fig. 9-1**). However, at the higher U_l values (**Figs. 9-10b and c**), the south side had the

largest $E_{D1} + E_{D2} + E_{D3}$ values only at the highest height (h/H), whereas the west side had the largest $E_{D1} + E_{D2} + E_{D3}$ values at the lowest height (h/H). This suggests that the expanded fluidized bed height may be different at different sides resulting from the non-uniform fluidization, with the expanded bed height taller for the south side than the west side at the higher U_l values (**Figs. 9-10b and c**). Analogous to **Fig. 9-10**, **Fig. 9-11** shows the effect of overall GAC concentration on the fluidization behavior at different sides of the lab-scale reactor. The trends are similar to that in **Fig. 9-10**, specifically, (i) the $E_{D1} + E_{D2} + E_{D3}$ values decreased with h/H , and (ii) the relative $E_{D1} + E_{D2} + E_{D3}$ values among the different sides were similar. In particular, **Fig. 9-11** indicates that the $E_{D1} + E_{D2} + E_{D3}$ values increased as overall GAC concentrations increased, which has been shown to better mitigate fouling in Chapter 7.

Regarding the relationship between the $E_{D1} + E_{D2} + E_{D3}$ values and expanded bed height, both were generally positively correlated, as observed in both **Figs. 9-10** and **9-11**. This indicates that the extent of mechanical scouring by the fluidized GAC increased as the fluidized bed height expanded. Even at positions above the expanded bed, the much diminished GAC media were still effective in conferring scouring, as seen in the non-negligible $E_{D1} + E_{D2} + E_{D3}$ values in **Fig. 9-10a** and **Fig. 9-11a**. The results in **Figs. 9-10** and **9-11** underscore the ability of the accelerometer to detect the quality of fluidization, which is tied to the extent of fouling mitigation (Chapter 3), at various positions of the lab-scale reactor under different conditions.

Figure 9-10. Dimensionless height (h/H) versus the sum of the E_{D1} , E_{D2} and E_{D3} values at different sides of the lab-scale reactor at a constant overall GAC concentration of 200 g/L and (a) $U_l = 0.024$ m/s, (b) $U_l = 0.028$ m/s, and (c) $U_l = 0.033$ m/s. The greyed areas represent the corresponding expanded bed heights.

Figure 9-11. Dimensionless height (h/H) versus the sum of the E_{D1} , E_{D2} and E_{D3} values at different sides of the lab-scale reactor at a constant U_l value of 0.033 m/s and overall GAC concentrations of (a) 100 g/L, (b) 150 g/L, and (c) 200 g/L in the lab-scale reactor. The greyed areas represent the corresponding expanded bed heights.

9.3.3. Using the accelerometer signals from a pilot-scale reactor to study the effect of inlet liquid flow rate (Q) and overall GAC mass

Armed with the knowledge that the accelerometer can detect the fluidization quality, which is linked to fouling mitigation effectiveness, locally at different parts of the lab-scale reactor, the applicability to the pilot-scale AnFMBR with flat-sheet membranes (FS-AnFMBR) was investigated. **Fig. 9-12** contains plots of dimensionless height (h/H) versus $E_{D1}+E_{D2}+E_{D3}$ for the pilot-scale reactor. Each column represents a different inlet liquid flow rate (Q ; namely,

23, 46 and 67 m³/h), while each row represents a different overall GAC mass (namely, no GAC, and 25, 50 and 75 kg of GAC). It should be noted that (i) the pilot-scale FS-AnFMBR is an idealized one with a new membrane cassette (**Fig. 9-4b**) and DI water, and (ii) the volumetric flow rate (Q) instead of superficial liquid velocity (U_l) and GAC mass instead of overall GAC concentration are given here, because the membrane cassette used (**Fig. 9-4b**) hampers an accurate quantification of the cross-sectional area and liquid volume of the pilot-scale reactor (**Fig. 9-4a**). All the x -axes are in the same ranges for easy comparison, as are the y -axes. Three observations are worth noting. Firstly, the $E_{D1}+E_{D2}+E_{D3}$ values were insignificant for all the cases without GAC particles (first row of **Fig. 9-12**), which re-affirms that the $E_{D1}+E_{D2}+E_{D3}$ values represent the dynamics of the fluidized GAC. Secondly, the west side consistently gave the largest $E_{D1}+E_{D2}+E_{D3}$ values, which is presumably because it was the side at which the liquid enters the distributor (**Fig. 9-4**). This contrasts somewhat with the results for the lab-scale reactor (**Figs. 9-10 and 9-11**), whereby the south side gave the largest $E_{D1}+E_{D2}+E_{D3}$ values particularly at lower superficial liquid velocities (U_l) and higher heights (h/H). Although the distributor and reactor designs were similar for both the lab-scale and pilot-scale reactors, the effect of the different scales on the fluidization behavior appears to be significant. The asymmetric flow behavior in both cases highlights the need for instruments like the accelerometer used here to troubleshoot such processes. Thirdly, similar to the lab-scale reactor (**Figs. 9-10 and 9-11**), the $E_{D1}+E_{D2}+E_{D3}$ values increased as h/H decreased or overall GAC concentration increased or U_l increased between 23 – 46 m³/h. Interestingly, as U_l increased between 46 and 67 m³/h, the $E_{D1}+E_{D2}+E_{D3}$ values

decreased, which could be due to liquid channeling at higher velocities [260, 278] leading to poor fluidization of GAC or particle entrainment reducing the overall concentration of GAC. This further highlights the need for real-time monitoring of the process. Collectively, these observations underscore the efficacy of the accelerometer in monitoring local fluidization quality even at the pilot-scale.

Figure 9-12. Dimensionless height (h/H) versus the sum of the E_{D1} , E_{D2} and E_{D3} values at different sides of the pilot-scale FS-AnFMBR with a new membrane cassette (Fig. 9-4b) and DI water. Each column represents a different inlet liquid flow rate (Q), while each row represents a different overall GAC mass.

9.3.4. Monitoring the pilot-scale AnFMBRs

The conventional method of using the trans-membrane pressure (TMP) to monitor membrane-based filtration processes is popular, but is not able to give local information (e.g., more extensive fouling at certain sides of the membrane). The accelerometer addresses this and is as simple to operate primarily due to the non-invasive nature.

Fig. 9-13 displays the TMP and flux trends over almost a year for the pilot-scale AnFMBRs (**Figs. 9-2 and 9-3**) with flat-sheet (FS-AnFMBR; **Fig. 9-13a**) and hollow fiber membranes (HF-AnFMBR; **Fig. 9-13b**). For each reactor, the amount of GAC present was 150 kg and the targeted permeate flux was 5.5 L/(m² h), and the inlet liquid flow rates from the first-stage AnFBR (**Figs. 9-2 and 9-3**) were 42 and 30 m³/h, respectively, for the FS-AnFMBR and HF-AnFMBR. Regarding TMP, both **Figs. 9-13a and b** surprisingly show steep increases in the TMP values within the first few days, after which the TMP values were relatively constant. As for flux, it continuously decreased for the FS-AnFMBR, and fluctuated more for the HF-AnFMBR reactor due to the 2 min/day of backwash with permeate. No information regarding the hydrodynamics of the fluidized GAC, which is key to mitigating membrane fouling, is available from **Fig. 9-13**.

Figure 9-13. Evolution of TMP and flux for the (a) FS-AnFMBR reactor with $Q = 42 \text{ m}^3/\text{h}$; and (b) HF-AnFMBR reactor with $Q = 30 \text{ m}^3/\text{h}$. For each reactor, the amount of GAC present was 150 kg and the targeted permeate flux was $5.5 \text{ L}/(\text{m}^2 \text{ h})$.

Accelerometer signals were obtained from the west side of the AnFMBRs and the results are plotted in **Fig. 9-14**, with dimensionless height (h/H) versus $E_{D1}+E_{D2}+E_{D3}$. In **Fig. 9-14a**, one of the trends represent the data from the FS-AnFMBR with new membranes, DI water, $Q = 46 \text{ m}^3/\text{h}$ and 75 kg of GAC, while the other two represent the data obtained towards the end of the campaign (i.e., at 291 days for the FS-AnFMBR and 350 days for the HF-AnFMBR; **Fig. 9-13**). Clearly, the FS-AnFMBR with new membranes and DI water had significantly larger $E_{D1}+E_{D2}+E_{D3}$ values throughout the reactor, which signifies the presence of fluidized GAC and the attendant fouling mitigation effect. As for the other two trends reflecting AnFMBRs at the end of the campaigns, the $E_{D1}+E_{D2}+E_{D3}$ values were almost zero throughout the reactors, which indicates that the GAC were not fluidized. Inspection of the flat-sheet membrane cassette revealed that the fouling and clogging was so severe that GAC fluidization was impossible. **Fig. 9-14b** further displays the h/H versus $E_{D1}+E_{D2}+E_{D3}$ trends for the HF-AnFMBR when the GAC were defluidized due to inadvertent diminished flow rate to the inlet of the distributor, and also when the inlet flow was re-established. Clearly, the

$E_{D1}+E_{D2}+E_{D3}$ values were significantly higher in the latter case, which provides more confidence for the deductions for **Fig. 9-14a**.

Figure 9-14. Dimensionless height (h/H) versus the sum of the E_{D1} , E_{D2} and E_{D3} values obtained at the west side of the pilot-scale AnFMBRs at a targeted permeate flux of $5.5 \text{ L}/(\text{m}^2 \text{ h})$ for each reactor: (a) One of the trends represent the data from the FS-AnFMBR with new membranes, DI water, $Q = 46 \text{ m}^3/\text{h}$ and 75 kg of GAC, while the other two represent the data obtained towards the end of the campaign (i.e., at 291 days for the FS-AnFMBR and 350 days for the HF-AnFMBR; Fig. 9-13); (b) One trend represents the data from the FS-AnFMBR when defluidization occurred due to diminished inlet flow, while the other when fluidization was re-established when the targeted inlet flow rate was enabled; in both cases, the HF-AnFMBR has been operated for more about 350 days at $Q = 30 \text{ m}^3/\text{h}$ and 150 kg of GAC.

Furthermore, the evolution of the $E_{D1}+E_{D2}+E_{D3}$ values were monitored.

Fig. 9-15a shows that for the FS-AnFMBR with new membranes, 75 kg of GAC, $Q = 46 \text{ m}^3/\text{h}$, and a DI water feed, while **Fig. 9-15b** the HF-AnFMBR with 150 kg GAC, $Q = 30 \text{ m}^3/\text{h}$, and wastewater at about 350 days of operation. Both acceleration signals were collected at the west side of the reactors. The $E_{D1}+E_{D2}+E_{D3}$ values in **Fig. 9-15a** are much larger than that in **Fig. 9-15b** due to the much diminished GAC fluidization in the latter case, as seen also in **Fig. 9-14**. **Fig. 9-15a** indicates that the $E_{D1}+E_{D2}+E_{D3}$ values (i) decreased with h/H , which agrees with earlier figures; and (ii) were constant within seven days but

decreased slightly subsequently presumably signifying the onset of some fouling or clogging, since the HF-AnFMBR contained some residual foulants (which can be observed visually) despite having a new membrane cassette installed. As for **Fig. 9-15b**, the $E_{D1}+E_{D2}+E_{D3}$ values were relatively similar (i.e., within the span of error bars seen in **Figs. 9-10 and 9-11**) at the heights (h/H) and through the duration characterized for the severely clogged HF-AnFMBR. Nonetheless, the drastic decline in the $E_{D1}+E_{D2}+E_{D3}$ values affirms the ability of the accelerometer to shed light on the fluidization behavior within the reactor, which thereby assists operators in the trouble-shooting of such reactors.

Figure 9-15. Evolution of $E_{D1}+E_{D2}+E_{D3}$ values for the (a) FS-AnFMBR with new membranes, 75 kg of GAC, $Q = 46 \text{ m}^3/\text{h}$, and a DI water feed, and (b) HF-AnFMBR with 150 kg GAC, $Q = 30 \text{ m}^3/\text{h}$, and wastewater at about 350 days of operation. Both acceleration signals were collected at the west side of the reactors.

9.4. Conclusions

The feasibility of using an accelerometer to non-invasively characterize the hydrodynamics of fluidized GAC, which has been shown to directly impact the extent of fouling mitigation, was studied systematically. Because the means of the accelerometer signals failed to provide any insights, wavelet

decomposition of the accelerometer signals was implemented to enable the extraction of the higher-frequency information corresponding only to the hydrodynamics of the fluidized GAC in Chapter 3, denoted as $E_{D1}+E_{D2}+E_{D3}$.

Firstly, a lab-scale AnFMBR was investigated. Specifically, the effect of height along the reactor (h/H), sides of the reactor (relative to the distributor), superficial liquid velocity (U_l) and overall GAC concentration on the fluidized GAC hydrodynamics in a lab-scale AnFMBR was studied. The $E_{D1}+E_{D2}+E_{D3}$ values, which has been positively correlated to the extent of fouling mitigation in Chapter 3, expectedly increased as h/H decreased, U_l increased or overall GAC concentration increased. Interestingly, the $E_{D1}+E_{D2}+E_{D3}$ values were different at different sides of the reactor, which suggests non-uniform fluidization within the AnFMBR. Armed with the success of the accelerometer in the lab-scale reactor, an idealized pilot-scale FS-AnFMBR (with a new membrane cassette and DI water) was assessed. Similarly, the effect of height along the reactor (h/H), sides of the reactor (relative to the distributor), inlet liquid flow rate (Q) and overall GAC mass was investigated. In contrast to the lab-scale AnFMBR, it was found that the pilot-scale exhibited (i) the largest $E_{D1}+E_{D2}+E_{D3}$ values at a different reactor side, despite having similar distributor and reactor design; and (ii) a decrease in $E_{D1}+E_{D2}+E_{D3}$ values when increasing U_l between the two higher U_l values, due possibly to liquid channeling or GAC entrainment.

The conventional means of monitoring the trans-membrane pressure (TMP) and flux were not able to provide much insights in the understanding or trouble-shooting the pilot-scale AnFMBRs, but the accelerometer was able to shed significant insight. The $E_{D1}+E_{D2}+E_{D3}$ values for the pilot-scale AnFMBRs

used for almost a year were negligible, which suggests poor GAC fluidization, which agrees with the visual observation of an extremely clogged membrane cassette. Furthermore, the evolution of the $E_{D1}+E_{D2}+E_{D3}$ values indicates the possibility of earlier knowledge of clogging issues, which would enable measures to avert exacerbating the problem.

Collectively, the results in this study affirm that the accelerometer is promising as a non-invasive, real-time means in revealing the hydrodynamics of the fluidized GAC in AnFMBRs, the lack of which results in severe fouling and clogging issues.

Chapter 10 Conclusions and Recommendations

10.1. Conclusions

This thesis focuses on the correlation between fluidized media behavior in a solid-liquid fluidized bed and the extent of membrane-fouling mitigation via mechanical scouring by the particles. The characteristics of the local fluidization hydrodynamics (namely, particle velocity (V_p), particle rotation velocity (V_θ), particle contact area (A_p) and particle momentum (P_p)) was enabled by a high-speed video camera and image analysis via ImageJ, while membrane fouling was characterized either by the rate of rise of TMP (i.e., $dTMP/dt$) or by the critical flux (J_c) of micron-sized particles via the direct observation through the membrane (DOTM) technique. The impact factors include GAC particle diameter (d_p), particle concentration, particle sphericity (ϕ), superficial liquid velocity (U_l) and axial position of the vertically aligned membrane (h/H). The main conclusions are summarized here:

1. The scouring efficacy of GAC particles with three particle diameters (namely, 1.20 mm, 1.85 mm, and 2.18 mm) has been investigated via wavelet decomposition of accelerometer signals. Results indicate that the smallest GAC particle with $d_p = 1.2$ mm conferred the least scouring efficiency (around 50% lower at $U_l = 0.013$ m/s), compared to larger GAC particles, which is due to the balance between particle inertia, and the difference between superficial liquid velocity and minimum fluidization velocity ($U_l - U_{mf}$). At the same U_l , the ($U_l - U_{mf}$) values of the $d_p = 1.20$ mm can be 0.003 m/s higher than that of $d_p = 2.18$ mm. However, particle inertia is lower due to the lower d_p .

2. Directly comparing particle behavior with membrane fouling trends show that higher particle velocities (V_p), particle concentration, and particle momentum (P) generally all showed a negative correlation with $dTMP/dt$. The negative correlation between $dTMP/dt$ and particle momentum was most pronounced, which suggests that momentum transfer between the GAC particles and the membrane represents a key mechanism effecting the scouring to diminish membrane fouling. Larger GAC particles exhibited higher particle momentum values are more beneficial for the mitigation of membrane fouling.

3. The presence of fluidized GAC increased critical flux by an order-of-magnitude relative to that in the absence of GAC (i.e., only liquid shear) that the J_c values increased from 5 L/(m² h) to around 60 L/(m² h) at h/H of 0.29 due to the presence of fluidized GAC. The overall membrane critical flux increased with power input (P_r) for the same particle diameter (d_p), and decreased with particle diameter (d_p) at the same power input (P_r). Increasing the power input (P_r) had a greater effect on the overall critical flux of the larger particles than the smaller ones.

4. At the same superficial liquid velocity (U_l), the increase of overall GAC concentration increased the overall critical flux values and reduced the power required per unit production of permeate (P_p). It was found that an increase of GAC concentration from 50 g/L to 150 g/L could increase the $J_{c,overall}$ values from 18.19 to 59.36 L/(m² h), but reduced the P_p from 0.0198 to 0.014 kWh/m³. However, the power requirement (P_r) increased, due to the greater pressure drop across the bed (ΔP_{bed}). The benefits of higher overall GAC concentration were negligible at the lower power input (P_r) values but significantly advantageous at

the higher P_r values. Higher overall GAC concentration generally gave more similar local critical flux values among the three heights in the range of energy consumption investigated.

5. Lower particle sphericity (ϕ) was found to give lower local critical flux ($J_{c,local}$) values at the same U_l values, which indicates lesser fouling mitigation. Particle sphericity (ϕ) had negligible impact on the relationship between the local critical flux ($J_{c,local}$) and contact area per unit membrane area (A_p) or P_p , but significant impact on the relationships between $J_{c,local}$ and both the particle velocity ($V_{p,rms}$) and particle rotation velocity ($V_{\theta,rms}$). An increase in power input was more beneficial for the higher-sphericity GAC media than the lower-sphericity media.

6. The feasibility of using an accelerometer to non-invasively characterize the hydrodynamics of fluidized GAC was tested in both of a lab-scale and pilot-scale AnFMBR setups. The conventional means of monitoring the trans-membrane pressure (TMP) and flux were not able to provide much insights in the understanding or trouble-shooting the pilot-scale AnFMBRs, but the accelerometer was able to shed significant insight. The accelerometer is promising as a non-invasive, real-time means in revealing the hydrodynamics of the fluidized GAC in AnFMBRs, the lack of which results in severe fouling and clogging issues.

10.2 Recommendations

The findings emanating from this study are expected to be valuable for optimizing the energy-efficiency and control of the fluidized bed–membrane reactors, and also in validating models for solid-liquid fluidization. To optimize the AnFMBRs towards energy efficient wastewater treatment, the followings should be considered:

1. At lower scouring efficiency, the energy requirement (P_r) of GAC particles with various diameters were similar. At higher scouring efficiency, the P_r of smaller GAC particles was the greatest. Thereby, larger GAC particles are more beneficial for the fouling mitigation efficiency if higher mechanical scouring needed in the operating variables of AnFMBRs.

2. For the monodisperse foulant (i.e., polystyrene particles), the larger GAC particles were more effective locally due to greater particle inertia, while smaller GAC particles were more effective overall due to greater bed expansion enabling scouring of more membrane heights at the same P_r . From this point, an energy efficient membrane fouling control is tied to the height of the membrane in AnFMBRs.

3. At the same superficial liquid velocity (U_l), the increase of overall GAC concentration increased the overall critical flux values and reduced the power required per unit production of permeate (P_p). Thereby, it would be possible to operate the AnFMBR at higher permeate flux values at higher overall GAC concentrations, since reduced energy consumption.

4. The uniformity of fouling mitigation increased with P_r regardless of whether it was by virtue of overall GAC concentration or GAC particle diameter. Thereby, the increase of overall GAC concentration or GAC particle diameter are always positive for the fouling mitigation, while is linked to operational problems like GAC particle clogging, and the membrane integrity issues. The accelerometer is promising as a non-invasive, real-time means in revealing the hydrodynamics of the fluidized GAC, and the operational problems like fouling and clogging issues in pilot-scale AnFMBRs.

5. It is not desirable to have to increase the power input (P_r) if reasonable performance can be obtained at a much lower P_r . This seems to indicate the smaller GAC particles may hold the key, but a lower limit to the GAC particle diameter inevitably exist to have sufficient inertia for the mechanical scouring effects, which underlies the effectiveness of the millimeter-sized GAC vis-à-vis PAC (powdered activated carbon, which is an order-of-magnitude smaller). To avoid the deposition of GAC particles and prevent the particle clogging, an optimal value of U/U_{mf} should be practised to ensure sufficient movement of the fluidized GAC media.

In summary, the optimal fluidization scenario in the operation of AnFMBRs would give (i) good enhancement of $J_{c,overall}$, (ii) $J_{c,local}/J_{c,overall}$ close to unity at all membrane heights, and (iii) a modest power requirement (P_r). To identify the optimal fluidization for membrane fouling mitigation would involve a trade-off between capital cost (related to sustainable flux) and operating cost (related to power (P_r), fouling extent and cleaning). The related question remained is the effect of the ratio of the sizes of the foulant and scouring particles.

Studies on the shear-induced diffusion of binary-size mixtures of micron-sized particles have been reported [227, 232], but the corresponding effects on the inertial lift associated with the millimeter-sized GAC is yet unknown.

REFERENCES

- [1] F. Zamani, J.W. Chew, E. Akhondi, W.B. Krantz, A.G. Fane, Unsteady-state shear strategies to enhance mass-transfer for the implementation of ultrapermeable membranes in reverse osmosis: A review, *Desalination*, 356 (2015) 328-348.
- [2] M.Y. Jaffrin, Hydrodynamic Techniques to Enhance Membrane Filtration, *Annu Rev Fluid Mech*, 44 (2012) 77-96.
- [3] N. Al-Bastaki, A. Abbas, Use of fluid instabilities to enhance membrane performance: a review, *Desalination*, 136 (2001) 255-262.
- [4] B.B. Gupta, Use of hydrodynamic methods for pressure driven membrane operations, *Indian J Chem Techn*, 3 (1996) 156-163.
- [5] Z.F. Cui, S. Chang, A.G. Fane, The use of gas bubbling to enhance membrane processes, *Journal of Membrane Science*, 221 (2003) 1-35.
- [6] Y. Wibisono, E.R. Cornelissen, A.J.B. Kemperman, W.G.J. van der Meer, K. Nijmeijer, Two-phase flow in membrane processes: A technology with a future, *Journal of Membrane Science*, 453 (2014) 566-602.
- [7] M.Y. Jaffrin, Dynamic shear-enhanced membrane filtration: A review of rotating disks, rotating membranes and vibrating systems, *Journal of Membrane Science*, 324 (2008) 7-25.
- [8] A. Armando, B. Culkin, D. Purchas, New separation system extends the use of membranes, in: *Proceedings of the Euromembrane*, 1992, pp. 459-462.
- [9] V.G.J. Rodgers, R.E. Sparks, Effect of transmembrane pressure pulsing on concentration polarization, *Journal of Membrane Science*, 68 (1992) 149-168.
- [10] R. Benamar, B.B. Gupta, M.Y. Jaffrin, Apple Juice Clarification Using Mineral Membranes - Fouling Control by Backwashing and Pulsating Flow, *J Food Sci*, 55 (1990) 1620-1625.

- [11] S. Ilias, R. Govind, Potential Applications of Pulsed Flow for Minimizing Concentration Polarization in Ultrafiltration, *Separation Science and Technology*, 25 (1990) 1307-1324.
- [12] J. Meier, Mechanical influence of PAC particles on membrane processes, *J Membrane Sci*, 360 (2010) 404-409.
- [13] J. Kim, K. Kim, H. Ye, E. Lee, C. Shin, P.L. McCarty, J. Bae, Anaerobic Fluidized Bed Membrane Bioreactor for Wastewater Treatment, *Environ Sci Technol*, 45 (2011) 576-581.
- [14] H.J. Bixler, G.C. Rappe, Ultrafiltration Process, United States Patent Office, 3,541,006 (1970).
- [15] J. Bae, C. Shin, E. Lee, J. Kim, P.L. McCarty, Anaerobic treatment of low-strength wastewater: A comparison between single and staged anaerobic fluidized bed membrane bioreactors, *Bioresource Technol*, 165 (2014) 75-80.
- [16] J. Bae, R. Yoo, E. Lee, P. McCarty, Two-stage anaerobic fluidized-bed membrane bioreactor treatment of settled domestic wastewater, *Water Sci. Technol*, 68 (2013) 394-399.
- [17] A.Y. Hu, D.C. Stuckey, Activated carbon addition to a submerged anaerobic membrane bioreactor: Effect on performance, transmembrane pressure, and flux, *J Environ Eng-Asce*, 133 (2007) 73-80.
- [18] R. Yoo, J. Kim, P. McCarty, J. Bae, Effect of temperature on the treatment of domestic wastewater with a staged anaerobic fluidized membrane bioreactor, *Water Science & Technology*, 69 (2014) 1145-1150.
- [19] R. Yoo, J. Kim, P.L. McCarty, J. Bae, Anaerobic treatment of municipal wastewater with a staged anaerobic fluidized membrane bioreactor (SAF-MBR) system, *Bioresource technology*, 120 (2012) 133-139.

- [20] C. Shin, P.L. McCarty, J. Kim, J. Bae, Pilot-scale temperate-climate treatment of domestic wastewater with a staged anaerobic fluidized membrane bioreactor (SAF-MBR), *Bioresource Technol*, 159 (2014) 95-103.
- [21] D.W. Gao, Q. Hu, C. Yao, N.Q. Ren, Treatment of domestic wastewater by an integrated anaerobic fluidized-bed membrane bioreactor under moderate to low temperature conditions, *Bioresource Technol*, 159 (2014) 193-198.
- [22] D.W. Gao, Q. Hu, C. Yao, N.Q. Ren, W.M. Wu, Integrated anaerobic fluidized-bed membrane bioreactor for domestic wastewater treatment, *Chem Eng J*, 240 (2014) 362-368.
- [23] J. Li, Z. Ge, Z. He, A fluidized bed membrane bioelectrochemical reactor for energy-efficient wastewater treatment, *Bioresource Technology*, 167 (2014) 310-315.
- [24] L.J. Ren, Y. Ahn, B.E. Logan, A Two-Stage Microbial Fuel Cell and Anaerobic Fluidized Bed Membrane Bioreactor (MFC-AFMBR) System for Effective Domestic Wastewater Treatment, *Environ Sci Technol*, 48 (2014) 4199-4206.
- [25] C.Y. Gomec, High-rate anaerobic treatment of domestic wastewater at ambient operating temperatures: A review on benefits and drawbacks, *Journal of Environmental Science and Health Part A*, 45 (2010) 1169-1184.
- [26] A.Y. Hu, D.C. Stuckey, Treatment of dilute wastewaters using a novel submerged anaerobic membrane bioreactor, *Journal of environmental engineering*, 132 (2006) 190-198.
- [27] P. Berube, E. Hall, P. Sutton, Parameters governing permeate flux in an anaerobic membrane bioreactor treating low-strength municipal wastewaters: a literature review, *Water environment research*, 78 (2006) 887-896.
- [28] Z. Huang, S.L. Ong, H.Y. Ng, Submerged anaerobic membrane bioreactor for low-strength wastewater treatment: effect of HRT and SRT on treatment performance and membrane fouling, *Water research*, 45 (2011) 705-713.

- [29] H.J. Lin, W. Peng, M.J. Zhang, J.R. Chen, H.C. Hong, Y. Zhang, A review on anaerobic membrane bioreactors: Applications, membrane fouling and future perspectives, *Desalination*, 314 (2013) 169-188.
- [30] M. Aslam, P.L. McCarty, J. Bae, J. Kim, The effect of fluidized media characteristics on membrane fouling and energy consumption in anaerobic fluidized membrane bioreactors, *Separation and Purification Technology*, 132 (2014) 10-15.
- [31] P. Le-Clech, V. Chen, T.A.G. Fane, Fouling in membrane bioreactors used in wastewater treatment, *Journal of Membrane Science*, 284 (2006) 17-53.
- [32] F. Meng, S.-R. Chae, A. Drews, M. Kraume, H.-S. Shin, F. Yang, Recent advances in membrane bioreactors (MBRs): Membrane fouling and membrane material, *Water Research*, 43 (2009) 1489-1512.
- [33] F.G. Meng, S.Q. Zhang, Y. Oh, Z.B. Zhou, H.S. Shin, S.R. Chae, Fouling in membrane bioreactors: An updated review, *Water Res*, 114 (2017) 151-180.
- [34] J.C.T. Lin, D.J. Lee, C.P. Huang, Membrane fouling mitigation: membrane cleaning, *Separ Sci Technol*, 45 (2010) 858-872.
- [35] Z.W. Wang, J.X. Ma, C.Y.Y. Tang, K. Kimura, Q.Y. Wang, X.M. Han, Membrane cleaning in membrane bioreactors: A review, *J Membrane Sci*, 468 (2014) 276-307.
- [36] X. Shi, G. Tal, N.P. Hankins, V. Gitis, Fouling and cleaning of ultrafiltration membranes: A review, *Journal of Water Process Engineering*, 1 (2014) 121-138.
- [37] N. Hilal, O.O. Ogunbiyi, N.J. Miles, R. Nigmatullin, Methods employed for control of fouling in MF and UF membranes: A comprehensive review, *Separ Sci Technol*, 40 (2005) 1957-2005.

- [38] T. Nguyen, F. Roddick, L. Fan, Biofouling of water treatment membranes: A review of the underlying causes, monitoring techniques and control measures, *Membranes*, 2 (2012) 804.
- [39] L.D. Tijing, Y.C. Woo, J.S. Choi, S. Lee, S.H. Kim, H.K. Shon, Fouling and its control in membrane distillation-A review, *J Membrane Sci*, 475 (2015) 215-244.
- [40] F.G. Meng, S.R. Chae, A. Drews, M. Kraume, H.S. Shin, F.L. Yang, Recent advances in membrane bioreactors (MBRs): Membrane fouling and membrane material, *Water Res*, 43 (2009) 1489-1512.
- [41] A. Drews, Membrane fouling in membrane bioreactors-Characterisation, contradictions, cause and cures, *J Membrane Sci*, 363 (2010) 1-28.
- [42] P. Le-Clech, V. Chen, T.A.G. Fane, Fouling in membrane bioreactors used in wastewater treatment, *J Membrane Sci*, 284 (2006) 17-53.
- [43] W. Gao, H. Liang, J. Ma, M. Han, Z.L. Chen, Z.S. Han, G.B. Li, Membrane fouling control in ultrafiltration technology for drinking water production: a review, *Desalination*, 272 (2011) 1-8.
- [44] T. Leiknes, The effect of coupling coagulation and flocculation with membrane filtration in water treatment: A review, *J Environ Sci-China*, 21 (2009) 8-12.
- [45] J.H. Jhaveri, Z.V.P. Murthy, A comprehensive review on anti-fouling nanocomposite membranes for pressure driven membrane separation processes, *Desalination*, 379 (2016) 137-154.
- [46] S.W. Leong, A. Razmjou, K. Wang, K. Hapgood, X.W. Zhang, H.T. Wang, TiO₂ based photocatalytic membranes: A review, *J Membrane Sci*, 472 (2014) 167-184.
- [47] V. Kochkodan, N. Hilal, A comprehensive review on surface modified polymer membranes for biofouling mitigation, *Desalination*, 356 (2015) 187-207.

- [48] N.M. D'Souza, A.J. Mawson, Membrane cleaning in the dairy industry: A review, *Crit Rev Food Sci*, 45 (2005) 125-134.
- [49] C. Regula, E. Carretier, Y. Wyart, G. Gesan-Guiziou, A. Vincent, D. Boudot, P. Moulin, Chemical cleaning/disinfection and ageing of organic UF membranes: A review, *Water Res*, 56 (2014) 325-365.
- [50] N. Porcelli, S. Judd, Chemical cleaning of potable water membranes: A review, *Sep Purif Technol*, 71 (2010) 137-143.
- [51] M. Aslam, A. Charfi, G. Lesage, M. Heran, J. Kim, Membrane bioreactors for wastewater treatment: a review of mechanical cleaning by scouring agents to control membrane fouling, *Chem Eng J*, 307 (2017) 897-913.
- [52] H.M. Kyllonen, P. Pirkonen, M. Nystrom, Membrane filtration enhanced by ultrasound: a review, *Desalination*, 181 (2005) 319-335.
- [53] H.M. Huotari, G. Tragardh, I.H. Huisman, Crossflow membrane filtration enhanced by an external DC electric field: A review, *Chem Eng Res Des*, 77 (1999) 461-468.
- [54] Z. Wang, Z. Wu, A Review of Membrane Fouling in MBRs: Characteristics and Role of Sludge Cake Formed on Membrane Surfaces, *Separation Science and Technology*, 44 (2009) 3571-3596.
- [55] Z. Wang, J. Ma, C.Y. Tang, K. Kimura, Q. Wang, X. Han, Membrane cleaning in membrane bioreactors: A review, *Journal of Membrane Science*, 468 (2014) 276-307.
- [56] L. Song, M. Elimelech, Theory of concentration polarization in crossflow filtration, *Journal of the Chemical Society, Faraday Transactions*, 91 (1995) 3389-3398.
- [57] M.C. Porter, Concentration Polarization with Membrane Ultrafiltration, *Product R&D*, 11 (1972) 234-248.

- [58] S. Bhattacharjee, A.S. Kim, M. Elimelech, Concentration Polarization of Interacting Solute Particles in Cross-Flow Membrane Filtration, *Journal of Colloid and Interface Science*, 212 (1999) 81-99.
- [59] X.-m. Wang, T.D. Waite, Role of Gelling Soluble and Colloidal Microbial Products in Membrane Fouling, *Environ Sci Technol*, 43 (2009) 9341-9347.
- [60] Q. Wang, Z. Wang, Z. Wu, J. Ma, Z. Jiang, Insights into membrane fouling of submerged membrane bioreactors by characterizing different fouling layers formed on membrane surfaces, *Chem Eng J*, 179 (2012) 169-177.
- [61] B. Lesjean, S. Rosenberger, C. Laabs, M. Jekel, R. Gnirss, G. Amy, Correlation between membrane fouling and soluble/colloidal organic substances in membrane bioreactors for municipal wastewater treatment, *Water Science & Technology*, 51 (2005) 1-8.
- [62] P. Le-Clech, B. Jefferson, S. Judd, Impact of aeration, solids concentration and membrane characteristics on the hydraulic performance of a membrane bioreactor, *Journal of Membrane Science*, 218 (2003) 117-129.
- [63] I.-S. Chang, P. Le Clech, B. Jefferson, S. Judd, Membrane fouling in membrane bioreactors for wastewater treatment, *Journal of environmental engineering*, 128 (2002) 1018-1029.
- [64] S. Shirazi, C.-J. Lin, D. Chen, Inorganic fouling of pressure-driven membrane processes — A critical review, *Desalination*, 250 (2010) 236-248.
- [65] D.C. Stuckey, Recent developments in anaerobic membrane reactors, *Bioresource Technol*, 122 (2012) 137-148.
- [66] R. Field, D. Wu, J. Howell, B. Gupta, Critical flux concept for microfiltration fouling, *Journal of Membrane Science*, 100 (1995) 259-272.
- [67] N. Ratkovich, W. Horn, F.P. Helmus, S. Rosenberger, W. Naessens, I. Nopens, T.R. Bentzen, Activated sludge rheology: A critical review on data collection and modelling, *Water Res*, 47 (2013) 463-482.

- [68] J.W. Wang, F. Zamani, A. Cahyadi, J.Y. Toh, S.L. Yang, B. Wu, Y. Liu, A.G. Fane, J.W. Chew, Correlating the hydrodynamics of fluidized granular activated carbon (GAC) with membrane-fouling mitigation, *J Membrane Sci*, 510 (2016) 38-49.
- [69] J.W. Wang, B. Wu, S.L. Yang, Y. Liu, A.G. Fane, J.W. Chew, Characterizing the scouring efficiency of Granular Activated Carbon (GAC) particles in membrane fouling mitigation via wavelet decomposition of accelerometer signals, *J Membrane Sci*, 498 (2016) 105-115.
- [70] A. Cahyadi, S.L. Yang, J.W. Chew, CFD study on the hydrodynamics of fluidized granular activated carbon in AnFMBR applications, *Sep Purif Technol*, 178 (2017) 75-89.
- [71] R. Sengur, G. Deveci, R. Kaya, T. Turken, S. Guclu, D.Y. Imer, I. Koyuncu, CFD modeling of submerged membrane bioreactors (sMBRs): a review, *Desalin Water Treat*, 55 (2015) 1747-1761.
- [72] X.F. Liu, Y. Wang, T.D. Waite, G. Leslie, Numerical simulations of impact of membrane module design variables on aeration patterns in membrane bioreactors, *J Membrane Sci*, 520 (2016) 201-213.
- [73] X.F. Liu, Y. Wang, T.D. Waite, G. Leslie, Numerical simulation of bubble induced shear in membrane bioreactors: Effects of mixed liquor rheology and membrane configuration, *Water Res*, 75 (2015) 131-145.
- [74] N. Al-Bastaki, A. Abbas, Use of fluid instabilities to enhance membrane performance: a review, *Desalination*, 136 (2001) 255-262.
- [75] M.Y. Jaffrin, Hydrodynamic Techniques to Enhance Membrane Filtration, *Annual Review of Fluid Mechanics*, 44 (2011) 77-96.
- [76] B. Culkin, A.D. Armando, New separation system extends the use of membranes, *Filtration & Separation*, 29 (1992) 376-378.

- [77] T. Imasaka, N. Kanekuni, H. So, S. Yoshino, Cross-flow filtration of methane fermentation broth by ceramic membranes, *Journal of Fermentation and Bioengineering*, 68 (1989) 200-206.
- [78] J.-y. Tian, Y.-p. Xu, Z.-l. Chen, J. Nan, G.-b. Li, Air bubbling for alleviating membrane fouling of immersed hollow-fiber membrane for ultrafiltration of river water, *Desalination*, 260 (2010) 225-230.
- [79] G. Ducom, H. Matamoros, C. Cabassud, Air sparging for flux enhancement in nanofiltration membranes: application to O/W stabilised and non-stabilised emulsions, *Journal of Membrane Science*, 204 (2002) 221-236.
- [80] M.M. Mosallem, Tud, Tud, Bubble dynamics and cavitation inception in a turbulent separating flow, 1999.
- [81] T.Y. Chiu, A.E. James, Critical flux enhancement in gas assisted microfiltration, *Journal of Membrane Science*, 281 (2006) 274-280.
- [82] Q.Y. Li, Z.F. Cui, D.S. Pepper, Effect of bubble size and frequency on the permeate flux of gas sparged ultrafiltration with tubular membranes, *Chem Eng J*, 67 (1997) 71-75.
- [83] S. Laborie, C. Cabassud, L. Durand-Bourlier, J.M. Lainé, Fouling control by air sparging inside hollow fibre membranes—effects on energy consumption, *Desalination*, 118 (1998) 189-196.
- [84] M. Mercier-Bonin, C. Lagane, C. Fonade, Influence of a gas/liquid two-phase flow on the ultrafiltration and microfiltration performances: case of a ceramic flat sheet membrane, *Journal of Membrane Science*, 180 (2000) 93-102.
- [85] T. Wilmarth, M. Ishii, Two-phase flow regimes in narrow rectangular vertical and horizontal channels, *International Journal of Heat and Mass Transfer*, 37 (1994) 1749-1758.
- [86] I.-S. Chang, S.J. Judd, Air sparging of a submerged MBR for municipal wastewater treatment, *Process Biochemistry*, 37 (2002) 915-920.

- [87] R.W. Field, D. Wu, J.A. Howell, B.B. Gupta, Critical flux concept for microfiltration fouling, *Journal of Membrane Science*, 100 (1995) 259-272.
- [88] E. Braak, M. Alliet, S. Schetrite, C. Albasi, Aeration and hydrodynamics in submerged membrane bioreactors, *Journal of Membrane Science*, 379 (2011) 1-18.
- [89] Z.F. Cui, S. Chang, A.G. Fane, The use of gas bubbling to enhance membrane processes, *Journal of Membrane Science*, 221 (2003) 1-35.
- [90] T.W. Cheng, H.M. Yeh, C.T. Gau, Enhancement of Permeate Flux by Gas Slugs for Crossflow Ultrafiltration in Tubular Membrane Module, *Separation Science and Technology*, 33 (1998) 2295-2309.
- [91] T. Miyahara, K. Tsuchiya, L.S. Fan, Wake properties of a single gas bubble in three-dimensional liquid-solid fluidized bed, *International Journal of Multiphase Flow*, 14 (1988) 749-763.
- [92] F. Wicaksana, A.G. Fane, V. Chen, Fibre movement induced by bubbling using submerged hollow fibre membranes, *Journal of Membrane Science*, 271 (2006) 186-195.
- [93] A.C. Rust, M. Manga, K.V. Cashman, Determining flow type, shear rate and shear stress in magmas from bubble shapes and orientations, *Journal of Volcanology and Geothermal Research*, 122 (2003) 111-132.
- [94] C.C.V. Chan, P.R. Bérubé, E.R. Hall, Relationship between types of surface shear stress profiles and membrane fouling, *Water Research*, 45 (2011) 6403-6416.
- [95] C. Serra, L. Durand-Bourlier, M.J. Clifton, P. Moulin, J.-C. Rouch, P. Aptel, Use of air sparging to improve backwash efficiency in hollow-fiber modules, *Journal of Membrane Science*, 161 (1999) 95-113.
- [96] T.M. Qaisrani, W.M. Samhaber, Impact of gas bubbling and backflushing on fouling control and membrane cleaning, *Desalination*, 266 (2011) 154-161.

- [97] S. Delgado, R. Villarroel, E. González, Effect of the shear intensity on fouling in submerged membrane bioreactor for wastewater treatment, *Journal of Membrane Science*, 311 (2008) 173-181.
- [98] J. Xu, Experimental study on gas–liquid two-phase flow regimes in rectangular channels with mini gaps, *International Journal of Heat and Fluid Flow*, 20 (1999) 422-428.
- [99] H.C. Chua, T.C. Arnot, J.A. Howell, Controlling fouling in membrane bioreactors operated with a variable throughput, *Desalination*, 149 (2002) 225-229.
- [100] T. Ueda, K. Hata, Y. Kikuoka, O. Seino, Effects of aeration on suction pressure in a submerged membrane bioreactor, *Water Research*, 31 (1997) 489-494.
- [101] L. De Temmerman, T. Maere, H. Temmink, A. Zwijnenburg, I. Nopens, The effect of fine bubble aeration intensity on membrane bioreactor sludge characteristics and fouling, *Water Research*, 76 (2015) 99-109.
- [102] K. Zhang, Z. Cui, R.W. Field, Effect of bubble size and frequency on mass transfer in flat sheet MBR, *Journal of Membrane Science*, 332 (2009) 30-37.
- [103] S. Jankhah, P.R. Bérubé, Power induced by bubbles of different sizes and frequencies on to hollow fibers in submerged membrane systems, *Water Research*, 47 (2013) 6516-6526.
- [104] A.P.S. Yeo, A.W.K. Law, A.G. Fane, Factors affecting the performance of a submerged hollow fiber bundle, *Journal of Membrane Science*, 280 (2006) 969-982.
- [105] A.P.S. Yeo, A.W.K. Law, A.G. Fane, The relationship between performance of submerged hollow fibers and bubble-induced phenomena examined by particle image velocimetry, *Journal of Membrane Science*, 304 (2007) 125-137.

- [106] C. Chan, P. Berube, E. Hall, An investigation of the hydrodynamic conditions inside a submerged membrane module under aeration, in: British Columbia Water and Wastewater Association Annual General Meeting, Whistler, British Columbia, 2006.
- [107] E. Braak, M. Alliet, S. Schetrite, C. Albasi, Aeration and hydrodynamics in submerged membrane bioreactors, *Journal of Membrane Science*, 379 (2011) 1-18.
- [108] Z.F. Cui, K.I.T. Wright, Flux enhancements with gas sparging in downwards crossflow ultrafiltration: performance and mechanism, *Journal of Membrane Science*, 117 (1996) 109-116.
- [109] New separation system extends the use of membranes, *Filtration & Separation*, 29 (1992) 376-378.
- [110] A.I. Zouboulis, M.D. Petala, Performance of VSEP vibratory membrane filtration system during the treatment of landfill leachates, *Desalination*, 222 (2008) 165-175.
- [111] S. Ahmed, M.G. Rasul, M.A. Hasib, Y. Watanabe, Performance of nanofiltration membrane in a vibrating module (VSEP-NF) for arsenic removal, *Desalination*, 252 (2010) 127-134.
- [112] W. Shi, M.M. Benjamin, Effect of shear rate on fouling in a Vibratory Shear Enhanced Processing (VSEP) RO system, *Journal of Membrane Science*, 366 (2011) 148-157.
- [113] W. Shi, M.M. Benjamin, Fouling of RO membranes in a vibratory shear enhanced filtration process (VSEP) system, *Journal of Membrane Science*, 331 (2009) 11-20.
- [114] F. Zamani, A.W.K. Law, A.G. Fane, Hydrodynamic analysis of vibrating hollow fibre membranes, *Journal of Membrane Science*, 429 (2013) 304-312.

- [115] G. Genkin, T. Waite, A. Fane, S. Chang, The effect of vibration and coagulant addition on the filtration performance of submerged hollow fibre membranes, *Journal of membrane science*, 281 (2006) 726-734.
- [116] S.P. Beier, M. Guerra, A. Garde, G. Jonsson, Dynamic microfiltration with a vibrating hollow fiber membrane module: Filtration of yeast suspensions, *Journal of Membrane Science*, 281 (2006) 281-287.
- [117] T. Li, A.W.-K. Law, M. Cetin, A. Fane, Fouling control of submerged hollow fibre membranes by vibrations, *Journal of Membrane Science*, 427 (2013) 230-239.
- [118] G. Genkin, T.D. Waite, A.G. Fane, S. Chang, The effect of vibration and coagulant addition on the filtration performance of submerged hollow fibre membranes, *Journal of Membrane Science*, 281 (2006) 726-734.
- [119] O.A. Akoum, M.Y. Jaffrin, L. Ding, P. Paullier, C. Vanhoutte, An hydrodynamic investigation of microfiltration and ultrafiltration in a vibrating membrane module, *Journal of Membrane Science*, 197 (2002) 37-52.
- [120] S. Rosenblat, Flow between torsionally oscillating disks, *Journal of Fluid Mechanics*, 8 (2006) 388-399.
- [121] T. Li, A.W.-K. Law, A.G. Fane, Submerged hollow fibre membrane filtration with transverse and longitudinal vibrations, *Journal of Membrane Science*, 455 (2014) 83-91.
- [122] S.C. Low, H.H. Juan, L.K. Siong, A combined VSEP and membrane bioreactor system, *Desalination*, 183 (2005) 353-362.
- [123] A. Kola, Y. Ye, A. Ho, P. Le-Clech, V. Chen, Application of low frequency transverse vibration on fouling limitation in submerged hollow fibre membranes, *Journal of Membrane Science*, 409-410 (2012) 54-65.

- [124] A. Kola, Y. Ye, P. Le-Clech, V. Chen, Transverse vibration as novel membrane fouling mitigation strategy in anaerobic membrane bioreactor applications, *Journal of Membrane Science*, 455 (2014) 320-329.
- [125] H.-y. Li, C.D. Bertram, D.E. Wiley, Mechanisms by which pulsatile flow affects cross-flow microfiltration, *AIChE Journal*, 44 (1998) 1950-1961.
- [126] V.G.J. Rodgers, R.E. Sparks, Reduction of membrane fouling in the ultrafiltration of binary protein mixtures, *AIChE Journal*, 37 (1991) 1517-1528.
- [127] H.B. Winzeler, G. Belfort, Enhanced performance for pressure-driven membrane processes: the argument for fluid instabilities, *Journal of Membrane Science*, 80 (1993) 35-47.
- [128] C.D. Bertram, M.R. Hoogland, H. Li, R.A. Odell, A.G. Fane, Flux enhancement in crossflow microfiltration using a collapsible-tube pulsation generator, *Journal of Membrane Science*, 84 (1993) 279-292.
- [129] N.M. Al-Bastak, A. Abbas, Periodic Operation of a Reverse Osmosis Water Desalination Unit, *Separation Science and Technology*, 33 (1998) 2531-2540.
- [130] M. Aslam, J. Kim, Investigating membrane fouling associated with GAC fluidization on membrane with effluent from anaerobic fluidized bed bioreactor in domestic wastewater treatment, *Environmental Science and Pollution Research*, (2017).
- [131] M. Aslam, P.L. McCarty, J. Bae, J. Kim, The effect of fluidized media characteristics on membrane fouling and energy consumption in anaerobic fluidized membrane bioreactors, *Sep Purif Technol*, 132 (2014) 10-15.
- [132] M. Aslam, A. Charfi, J. Kim, Membrane scouring to control fouling under fluidization of non-adsorbing media for wastewater treatment, *Environmental Science and Pollution Research*, (2017).

- [133] T. Kurita, K. Kimura, Y. Watanabe, Energy saving in the operation of submerged MBRs by the insertion of baffles and the introduction of granular materials, *Sep Purif Technol*, 141 (2015) 207-213.
- [134] M. Pradhan, S. Vigneswaran, J. Kandasamy, R.B. Aim, Combined effect of air and mechanical scouring of membranes for fouling reduction in submerged membrane reactor, *Desalination*, 288 (2012) 58-65.
- [135] A. Fenu, J. Roels, T. Wambecq, K. De Gussem, C. Thoeye, G. De Gueldre, B. Van De Steene, Energy audit of a full scale MBR system, *Desalination*, 262 (2010) 121-128.
- [136] S. Chaiprapat, A. Thongsai, B. Charnnok, W. Khongnakorn, J. Bae, Influences of liquid, solid, and gas media circulation in anaerobic membrane bioreactor (AnMBR) as a post treatment alternative of aerobic system in seafood industry, *Journal of Membrane Science*, 509 (2016) 116-124.
- [137] B.D. Shoener, C. Zhong, A.D. Greiner, W. O. Khunjar, P.-Y. Hong, J.S. Guest, Design of anaerobic membrane bioreactors for the valorization of dilute organic carbon waste streams, *Energy & Environmental Science*, 9 (2016) 1102-1112.
- [138] X. Huang, C.-H. Wei, K.-C. Yu, Mechanism of membrane fouling control by suspended carriers in a submerged membrane bioreactor, *Journal of Membrane Science*, 309 (2008) 7-16.
- [139] B. Siembida, P. Cornel, S. Krause, B. Zimmermann, Effect of mechanical cleaning with granular material on the permeability of submerged membranes in the MBR process, *Water Research*, 44 (2010) 4037-4046.
- [140] F. Chen, X. Bi, H.Y. Ng, Effects of bio-carriers on membrane fouling mitigation in moving bed membrane bioreactor, *Journal of Membrane Science*, 499 (2016) 134-142.

- [141] T. Kurita, K. Kimura, Y. Watanabe, The influence of granular materials on the operation and membrane fouling characteristics of submerged MBRs, *Journal of Membrane Science*, 469 (2014) 292-299.
- [142] B. Düppenbecker, M. Engelhart, P. Cornel, Fouling mitigation in Anaerobic Membrane Bioreactor using fluidized glass beads: Evaluation fitness for purpose of ceramic membranes, *Journal of Membrane Science*, 537 (2017) 69-82.
- [143] C.Y. Wen, Mechanics of fluidization, *Chem. Eng. Prog. Symp. Ser.*, 6 (1966) 100-101.
- [144] K. Tannous, M. Hemati, C. Laguerie, Caractéristiques au minimum de fluidisation et expansion des couches fluidisées de particules de la catégorie D de Geldart, *Powder Technology*, 80 (1994) 55-72.
- [145] A. Lucas, J. Arnaldos, J. Casal, L. Puigjaner, Improved equation for the calculation of minimum fluidization velocity, *Industrial & Engineering Chemistry Process Design and Development*, 25 (1986) 426-429.
- [146] S. Babu, B. Shah, A. Talwalkar, Fluidization correlations for coal gasification materials-minimum fluidization velocity and fluidized bed expansion ratio, in: *AIChE Symp. Ser.*, 1978, pp. 176-186.
- [147] W.-C. Yang, Chapter 26 Liquid-solids fluidization, in: *Handbook of fluidization and fluid-particle systems*, CRC press, 2003.
- [148] S. Ergun, Fluid flow through packed columns, *Chem. Eng. Prog.*, 48 (1952) 89-94.
- [149] M.J. Van Der Waal, P.M. Van Der Velden, J. Koning, C.A. Smolders, W.P.M. Van Swaay, Use of fluidised beds as turbulence promoters in tubular membrane systems, *Desalination*, 22 (1977) 465-483.
- [150] B.E. Logan, K. Rabaey, Conversion of Wastes into Bioelectricity and Chemicals by Using Microbial Electrochemical Technologies, *Science*, 337 (2012) 686-690.

- [151] B.D. Shoener, C. Zhong, A.D. Greiner, W.O. Khunjar, P.Y. Hong, J.S. Guest, Design of anaerobic membrane bioreactors for the valorization of dilute organic carbon waste streams, *Energy & Environmental Science*, 9 (2016) 1102-1112.
- [152] P.L. McCarty, J. Bae, J. Kim, Domestic Wastewater Treatment as a Net Energy Producer—Can This be Achieved?, *Environ Sci Technol*, 45 (2011) 7100-7106.
- [153] J. Kim, K. Kim, H. Ye, E. Lee, C. Shin, P.L. McCarty, J. Bae, Anaerobic fluidized bed membrane bioreactor for wastewater treatment, *Environmental science & technology*, 45 (2010) 576-581.
- [154] B. Wu, Y. Li, W. Lim, S.L. Lee, Q. Guo, A.G. Fane, Y. Liu, Single-stage versus two-stage anaerobic fluidized bed bioreactors in treating municipal wastewater: Performance, foulant characteristics, and microbial community, *Chemosphere*, 171 (2017) 158-167.
- [155] J. Li, S. Luo, Z. He, Cathodic fluidized granular activated carbon assisted-membrane bioelectrochemical reactor for wastewater treatment, *Sep Purif Technol*, 169 (2016) 241-246.
- [156] R.H. Yoo, J.H. Kim, P.L. McCarty, J.H. Bae, Effect of temperature on the treatment of domestic wastewater with a staged anaerobic fluidized membrane bioreactor, *Water Sci Technol*, 69 (2014) 1145-1150.
- [157] B. Wu, Y. Wang, W. Lim, J.W. Chew, A.G. Fane, Y. Liu, Enhanced performance of submerged hollow fibre microfiltration by fluidized granular activated carbon, *Journal of Membrane Science*, 499 (2016) 47-55.
- [158] B. Düppenbecker, M. Engelhart, P. Cornel, Fouling mitigation in Anaerobic Membrane Bioreactor using fluidized glass beads: Evaluation fitness for purpose of ceramic membranes, *Journal of Membrane Science*, 537 (2017) 69-82.

- [159] M. Aslam, A. Charfi, J. Kim, Membrane scouring to control fouling under fluidization of non-adsorbing media for wastewater treatment, *Environmental Science and Pollution Research*, (2017) 1-11.
- [160] D.-W. Gao, Q. Hu, C. Yao, N.-Q. Ren, Treatment of domestic wastewater by an integrated anaerobic fluidized-bed membrane bioreactor under moderate to low temperature conditions, *Bioresource Technol*, 159 (2014) 193-198.
- [161] D.-W. Gao, Q. Hu, C. Yao, N.-Q. Ren, W.-M. Wu, Integrated anaerobic fluidized-bed membrane bioreactor for domestic wastewater treatment, *Chem Eng J*, 240 (2014) 362-368.
- [162] J. Bae, R. Yoo, E. Lee, P.L. McCarty, Two-stage anaerobic fluidized-bed membrane bioreactor treatment of settled domestic wastewater, *Water Sci Technol*, 68 (2013) 394-399.
- [163] K. Dutta, M.-Y. Lee, W.W.-P. Lai, C.H. Lee, A.Y.-C. Lin, C.-F. Lin, J.-G. Lin, Removal of pharmaceuticals and organic matter from municipal wastewater using two-stage anaerobic fluidized membrane bioreactor, *Bioresource Technol*, 165 (2014) 42-49.
- [164] D.L. McCurry, S.E. Bear, J. Bae, D.L. Sedlak, P.L. McCarty, W.A. Mitch, Superior Removal of Disinfection Byproduct Precursors and Pharmaceuticals from Wastewater in a Staged Anaerobic Fluidized Membrane Bioreactor Compared to Activated Sludge, *Environmental Science & Technology Letters*, 1 (2014) 459-464.
- [165] B. Wu, P.C.Y. Wong, A.G. Fane, The potential roles of granular activated carbon in anaerobic fluidized membrane bioreactors: effect on membrane fouling and membrane integrity, *Desalin Water Treat*, 53 (2015) 1450-1459.
- [166] J. Wang, B. Wu, S. Yang, Y. Liu, A.G. Fane, J.W. Chew, Characterizing the scouring efficiency of Granular Activated Carbon (GAC) particles in

- membrane fouling mitigation via wavelet decomposition of accelerometer signals, *Journal of Membrane Science*, 498 (2016) 105-115.
- [167] J. Wang, F. Zamani, A. Cahyadi, J.Y. Toh, S. Yang, B. Wu, Y. Liu, A.G. Fane, J.W. Chew, Correlating the hydrodynamics of fluidized granular activated carbon (GAC) with membrane-fouling mitigation, *Journal of Membrane Science*, 510 (2016) 38-49.
- [168] M. Aslam, P.L. McCarty, C. Shin, J. Bae, J. Kim, Low energy single-staged anaerobic fluidized bed ceramic membrane bioreactor (AFCMBR) for wastewater treatment, *Bioresource Technol.*
- [169] C. Shin, K. Kim, P.L. McCarty, J. Kim, J. Bae, Integrity of hollow-fiber membranes in a pilot-scale anaerobic fluidized membrane bioreactor (AFMBR) after two-years of operation, *Sep Purif Technol*, 162 (2016) 101-105.
- [170] C. Shin, P.L. McCarty, J. Kim, J. Bae, Pilot-scale temperate-climate treatment of domestic wastewater with a staged anaerobic fluidized membrane bioreactor (SAF-MBR), *Bioresource Technology*, 159 (2014) 95-103.
- [171] B. Wu, F. Zamani, W. Lim, D. Liao, Y. Wang, Y. Liu, J.W. Chew, A.G. Fane, Effect of mechanical scouring by granular activated carbon (GAC) on membrane fouling mitigation, *Desalination*, 403 (2017) 80-87.
- [172] C. Shin, K. Kim, P.L. McCarty, J. Kim, J. Bae, Development and application of a procedure for evaluating the long-term integrity of membranes for the anaerobic fluidized membrane bioreactor (AFMBR), *Water Sci Technol*, 74 (2016) 457-465.
- [173] R. Lee, P.L. McCarty, J. Bae, J. Kim, Anaerobic fluidized membrane bioreactor polishing of baffled reactor effluent during treatment of dilute wastewater, *Journal of Chemical Technology & Biotechnology*, 90 (2015) 391-397.

- [174] K.-Y. Kim, H.-S. Kim, J. Kim, J.-W. Nam, J.-M. Kim, S. Son, A hybrid microfiltration–granular activated carbon system for water purification and wastewater reclamation/reuse, *Desalination*, 243 (2009) 132-144.
- [175] A.Y. Hu, D.C. Stuckey, Activated Carbon Addition to a Submerged Anaerobic Membrane Bioreactor: Effect on Performance, Transmembrane Pressure, and Flux, *Journal of Environmental Engineering*, 133 (2007) 73-80.
- [176] J.J. Heijnen, A. Mulder, W. Enger, F. Hoeks, Review on the application of anaerobic fluidized bed reactors in waste-water treatment, *The Chemical Engineering Journal*, 41 (1989) B37-B50.
- [177] B. Düppenbecker, S. Kale, M. Engelhart, P. Cornel, Fluidized glass beads reduce fouling in a novel anaerobic membrane bioreactor, *Water Sci Technol*, (2017).
- [178] B. Düppenbecker, J. Behnisch, M. Engelhart, P. Cornel, Fouling mitigation in anaerobic membrane bioreactors using fluidized resin beads, *Water Sci Technol*, (2017).
- [179] H. Prieske, L. Böhm, A. Drews, M. Kraume, Optimised hydrodynamics for membrane bioreactors with immersed flat sheet membrane modules, *Desalin Water Treat*, 18 (2010) 270-276.
- [180] Q. Yang, J. Chen, F. Zhang, Membrane fouling control in a submerged membrane bioreactor with porous, flexible suspended carriers, *Desalination*, 189 (2006) 292-302.
- [181] T. Kurita, K. Kimura, Y. Watanabe, The influence of granular materials on the operation and membrane fouling characteristics of submerged MBRs, *Journal of Membrane Science*, 469 (2014) 292-299.
- [182] L. Jin, S.L. Ong, H.Y. Ng, Fouling control mechanism by suspended biofilm carriers addition in submerged ceramic membrane bioreactors, *Journal of Membrane Science*, 427 (2013) 250-258.

- [183] D. Kwon, H. Chang, H. Seo, J. Kim, Fouling behavior and system performance in membrane bioreactor introduced by granular media as a mechanical cleaning effect on membranes, *Desalin Water Treat*, 57 (2016) 9018-9026.
- [184] T. Kurita, K. Kimura, Y. Watanabe, Energy saving in the operation of submerged MBRs by the insertion of baffles and the introduction of granular materials, *Sep Purif Technol*, 141 (2015) 207-213.
- [185] F. Chen, X. Bi, H.Y. Ng, Effects of bio-carriers on membrane fouling mitigation in moving bed membrane bioreactor, *Journal of Membrane Science*, 499 (2016) 134-142.
- [186] J. Hu, H. Ren, K. Xu, J. Geng, L. Ding, X. Yan, K. Li, Effect of carriers on sludge characteristics and mitigation of membrane fouling in attached-growth membrane bioreactor, *Bioresource Technol*, 122 (2012) 35-41.
- [187] C.-H. Wei, X. Huang, C.-W. Wang, X.-H. Wen, Effect of a suspended carrier on membrane fouling in a submerged membrane bioreactor, *Water Sci Technol*, 53 (2006) 211-220.
- [188] A. Charfi, M. Aslam, G. Lesage, M. Heran, J. Kim, Macroscopic approach to develop fouling model under GAC fluidization in anaerobic fluidized bed membrane bioreactor, *Journal of Industrial and Engineering Chemistry*, 49 (2017) 219-229.
- [189] J.G. Yates, S.J.R. Simons, Experimental Methods in Fluidization Research, *Int J Multiphas Flow*, 20 (1994) 297-330.
- [190] C. Boyer, A.M. Duquenne, G. Wild, Measuring techniques in gas-liquid and gas-liquid-solid reactors, *Chemical Engineering Science*, 57 (2002) 3185-3215.
- [191] J.R. van Ommen, R.F. Mudde, Measuring the Gas-Solids Distribution in Fluidized Beds-A Review, *Int J Chem React Eng*, 6 (2008).

- [192] J.W.R. Boyd, J. Varley, The uses of passive measurement of acoustic emissions from chemical engineering processes, *Chemical Engineering Science*, 56 (2001) 1749-1767.
- [193] G.D. Cody, D.J. Goldfarb, G.V. Storch, A.N. Norris, Particle granular temperature in gas fluidized beds, *Powder Technology*, 87 (1996) 211-232.
- [194] J.D. Wang, C.J. Ren, Y.R. Yang, Characterization of Flow Regime Transition and Particle Motion Using Acoustic Emission Measurement in a Gas-Solid Fluidized Bed, *Aiche Journal*, 56 (2010) 1173-1183.
- [195] H. Tsujimoto, T. Yokoyama, C.C. Huang, I. Sekiguchi, Monitoring particle fluidization in a fluidized bed granulator with an acoustic emission sensor, *Powder Technology*, 113 (2000) 88-96.
- [196] L. de Martín, J.V. Briongos, J.M. Aragón, M.C. Palancar, Can low frequency accelerometry replace pressure measurements for monitoring gas-solid fluidized beds?, *Chemical Engineering Science*, 65 (2010) 4055-4064.
- [197] J.V. Briongos, C. Sobrino, J. Gomez-Hernandez, D. Santana, Characterization of flow-induced vibrations in gas-solid fluidized beds: Elements of the theory, *Chemical Engineering Science*, 93 (2013) 181-196.
- [198] H. Azizpour, R. Sotudeh-Gharebagh, R. Zarghami, M. Abbasi, N. Mostoufi, M.J. Mahjoob, Characterization of gas-solid fluidized bed hydrodynamics by vibration signature analysis, *Int J Multiphas Flow*, 37 (2011) 788-793.
- [199] G. Book, K. Albion, L. Briens, C. Briens, F. Berruti, On-line detection of bed fluidity in gas-solid fluidized beds with liquid injection by passive acoustic and vibrometric methods, *Powder Technology*, 205 (2011) 126-136.
- [200] A. Abbasi, R. Sotudeh-Gharebagh, N. Mostoufi, R. Zarghami, M.J. Mahjoob, Nonintrusive Characterization of Fluidized Bed Hydrodynamics Using Vibration Signature Analysis, *Aiche Journal*, 56 (2010) 597-603.

- [201] X.J. Jiang, J.D. Wang, B.B. Jiang, Y.R. Yang, L.X. Hou, Study of the power spectrum of acoustic emission (AE) by accelerometers in fluidized beds, *Ind Eng Chem Res*, 46 (2007) 6904-6909.
- [202] Y.F. Zhou, Q. Shi, Z.L. Huang, J.D. Wang, Y.R. Yang, Z.W. Liao, J. Yang, Effects of Interparticle Forces on Fluidization Characteristics in Liquid-Containing and High-Temperature Fluidized Beds, *Ind Eng Chem Res*, 52 (2013) 16666-16674.
- [203] A. Sheikhi, R. Sotudeh-Gharebagh, N. Mostoufi, R. Zarghami, Experimental investigation on the hydrodynamics of a gas-liquid-solid fluidized bed using vibration signature and pressure fluctuation analyses, *Int J Heat Fluid Fl*, 42 (2013) 190-199.
- [204] C.M. Hrenya, Kinetic theory for granular materials: Polydispersity, in: S. Pannala, M. Syamlal, T. O'Brien (Eds.) *Computational Gas-Solids Flows and Reacting Systems: Theory, Methods and Practice*, IGI Global, Hershey, PA, 2011, pp. 475.
- [205] N. Ellis, L.A. Briens, J.R. Grace, H.T. Bi, C.J. Lim, Characterization of dynamic behaviour in gas-solid turbulent fluidized bed using chaos and wavelet analyses, *Chemical Engineering Journal*, 96 (2003) 105-116.
- [206] G.B. Zhao, Y.R. Yang, Multiscale resolution of fluidized-bed pressure fluctuations, *Aiche Journal*, 49 (2003) 869-882.
- [207] X.S. Lu, H.Z. Li, Wavelet analysis of pressure fluctuation signals in a bubbling fluidized bed, *Chemical Engineering Journal*, 75 (1999) 113-119.
- [208] J.Q. Ren, Q.M. Mao, J.H. Li, W.G. Lin, Wavelet analysis of dynamic behavior in fluidized beds, *Chemical Engineering Science*, 56 (2001) 981-988.
- [209] L.A. Briens, N. Ellis, Hydrodynamics of three-phase fluidized bed systems examined by statistical, fractal, chaos and wavelet analysis methods, *Chemical Engineering Science*, 60 (2005) 6094-6106.

- [210] J.H. Li, Compromise and resolution - Exploring the multi-scale nature of gas-solid fluidization, *Powder Technology*, 111 (2000) 50-59.
- [211] J.W. Chew, R. Hays, J.G. Findlay, T.M. Knowlton, S.R. Karri, R.A. Cocco, C.M. Hrenya, Cluster characteristics of Geldart Group B particles in a pilot-scale CFB riser. I. Monodisperse systems, *Chemical Engineering Science*, 68 (2012) 72-81.
- [212] L.-S. Fan, C. Zhu, Chapter 9 Dense-phase fluidized beds, in: *Principles of gas-solid flows*, Cambridge University Press, Cambridge ; New York, 1998, pp. 371-415.
- [213] D. Kunii, O. Levenspiel, Chapter 3 - Fluidization and Mapping of Regimes, in: *Fluidization Engineering*, Butterworth-Heinemann, Newton, MA, 1991, pp. 61-92.
- [214] W.-C. Yang, Chapter 26 Liquid-solids fluidization, in: *Handbook of fluidization and fluid-particle systems*, CRC Press, 2003.
- [215] D. Kunii, O. Levenspiel, Chapter 4 - The Dense Bed: Distributor, Gas Jets, and Pumping Power, in: *Fluidization Engineering*, Butterworth-Heinemann, Newton, MA, 1991, pp. 95-114.
- [216] J.T. Jenkins, M.Y. Louge, On the flux of fluctuation energy in a collisional grain flow at a flat, frictional wall, *Phys Fluids*, 9 (1997) 2835-2840.
- [217] T.W. Li, J. Grace, X.T. Bi, Study of wall boundary condition in numerical simulations of bubbling fluidized beds, *Powder Technol*, 203 (2010) 447-457.
- [218] H.W. Sur, Z.F. Cui, Enhancement of microfiltration of yeast suspensions using gas sparging - effect of feed conditions, *Sep Purif Technol*, 41 (2005) 313-319.
- [219] P.N. Loezos, P. Costamagna, S. Sundaresan, The role of contact stresses and wall friction on fluidization, *Chem Eng Sci*, 57 (2002) 5123-5141.

- [220] W.C. Yang, Chapter 26 Liquid-solids fluidization, in: Handbook of fluidization and fluid-particle systems, CRC press, 2003.
- [221] A. Yan, J.H. Parssinen, J.X. Zhu, Flow properties in the entrance and exit regions of a high-flux circulating fluidized bed riser, Powder Technol, 131 (2003) 256-263.
- [222] D. Geldart, J. Baeyens, The Design of Distributors for Gas-Fluidized Beds, Powder Technol, 42 (1985) 67-78.
- [223] P. Bacchin, P. Aimar, R.W. Field, Critical and sustainable fluxes: Theory, experiments and applications, Journal of Membrane Science, 281 (2006) 42-69.
- [224] H. Li, A.G. Fane, H.G.L. Coster, S. Vigneswaran, Direct observation of particle deposition on the membrane surface during crossflow microfiltration, Journal of Membrane Science, 149 (1998) 83-97.
- [225] F. Wicaksana, A.G. Fane, P. Pongpairroj, R. Field, Microfiltration of algae (*Chlorella sorokiniana*): Critical flux, fouling and transmission, Journal of Membrane Science, 387 (2012) 83-92.
- [226] E.N. Tummons, V.V. Tarabara, J.W. Chew, A.G. Fane, Behavior of oil droplets at the membrane surface during crossflow microfiltration of oil-water emulsions, Journal of Membrane Science, 500 (2016) 211-224.
- [227] H.J. Tanudjaja, W. Pee, A.G. Fane, J.W. Chew, Effect of spacer and crossflow velocity on the critical flux of bidisperse suspensions in microfiltration, Journal of Membrane Science, 513 (2016) 101-107.
- [228] F. Zamani, F. Wicaksana, A.H. Taheri, A.W.K. Law, A.G. Fane, W.B. Krantz, Generalized criterion for the onset of particle deposition in crossflow microfiltration via DOTM – Modeling and experimental validation, Journal of Membrane Science, 457 (2014) 128-138.
- [229] S.-T. Kang, A. Subramani, E.M.V. Hoek, M.A. Deshusses, M.R. Matsumoto, Direct observation of biofouling in cross-flow microfiltration:

- mechanisms of deposition and release, *Journal of Membrane Science*, 244 (2004) 151-165.
- [230] F. Wicaksana, A.G. Fane, P. Pongpairroj, R. Field, Microfiltration of algae (*Chlorella sorokiniana*): Critical flux, fouling and transmission, *Journal of Membrane Science*, 387–388 (2012) 83-92.
- [231] F. Zamani, A. Ullah, E. Akhondi, H.J. Tanudjaja, E.R. Cornelissen, A. Honciuc, A.G. Fane, J.W. Chew, Impact of the surface energy of particulate foulants on membrane fouling, *Journal of Membrane Science*, 510 (2016) 101-111.
- [232] Y.P. Zhang, A.G. Fane, A.W.K. Law, Critical flux and particle deposition of bidisperse suspensions during crossflow microfiltration, *Journal of Membrane Science*, 282 (2006) 189-197.
- [233] H. Wadell, Sphericity and Roundness of Rock Particles, *The Journal of Geology*, 41 (1933) 310-331.
- [234] B. Bethune, Surface Cracking of Glassy Polymers under a Sliding Spherical Indenter, *J Mater Sci*, 11 (1976) 199-205.
- [235] B. Lawn, R. Wilshaw, Indentation Fracture - Principles and Applications, *J Mater Sci*, 10 (1975) 1049-1081.
- [236] M.J. van der Waal, P.M. van der Velden, J. Koning, C.A. Smolders, W.P.M. Vanswaay, Use of Fluidized-Beds as Turbulence Promoters in Tubular Membrane Systems, *Desalination*, 22 (1977) 465-483.
- [237] R. de Boer, J.J. Zomerman, J. Hiddink, J. Aufderheyde, W.P.M. Vanswaay, C.A. Smolders, Fluidized-Beds as Turbulence Promoters in the Concentration of Food Liquids by Reverse-Osmosis, *J Food Sci*, 45 (1980) 1522-1528.
- [238] I. Yamanoi, K. Kageyama, Evaluation of bubble flow properties between flat sheet membranes in membrane bioreactor, *Journal of Membrane Science*, 360 (2010) 102-108.

- [239] A. Sofia, W.J. Ng, S.L. Ong, Engineering design approaches for minimum fouling in submerged MBR, *Desalination*, 160 (2004) 67-74.
- [240] A.G. Fane, A. Yeo, A. Law, K. Parameshwaran, F. Wicaksana, V. Chen, Low pressure membrane processes ~ doing more with less energy, *Desalination*, 185 (2005) 159-165.
- [241] L. Metcalf, H.P. Eddy, G. Tchobanoglous, *Wastewater engineering : treatment, disposal, and reuse*, McGraw-Hill, New York 2004.
- [242] S. Chang, A. Yeo, A. Fane, M. Cholewa, Y. Ping, H. Moser, Observation of flow characteristics in a hollow fiber lumen using non-invasive X-ray microimaging (XMI), *Journal of Membrane Science*, 304 (2007) 181-189.
- [243] G. Ducom, F.P. Puech, C. Cabassud, Air sparging with flat sheet nanofiltration: a link between wall shear stresses and flux enhancement, *Desalination*, 145 (2002) 97-102.
- [244] N.V. Ndinisa, A.G. Fane, D.E. Wiley, Fouling control in a submerged flat sheet membrane system: Part I - Bubbling and hydrodynamic effects, *Separation Science and Technology*, 41 (2006) 1383-1409.
- [245] V.V. Buwa, V.V. Ranade, Dynamics of gas-liquid flow in a rectangular bubble column: experiments and single/multi-group CFD simulations, *Chem Eng Sci*, 57 (2002) 4715-4736.
- [246] D. Kunii, O. Levenspiel, *Fluidization engineering* (Chapter 4), 2nd ed., Butterworth-Heinemann, Boston, 1991.
- [247] A. Cahyadi, S. Yang, J.W. Chew, CFD study on the hydrodynamics of fluidized granular activated carbon in AnFMBR applications, *Sep Purif Technol*, 178 (2017) 75-89.
- [248] A. Charfi, M. Aslam, J. Kim, Modelling approach to better control biofouling in fluidized bed membrane bioreactor for wastewater treatment, *Chemosphere*, 191 (2018) 136-144.

- [249] A. Cahyadi, A.G. Fane, J.W. Chew, Correlating the hydrodynamics of fluidized media with the extent of membrane fouling mitigation: Effect of bidisperse GAC mixtures, *Sep Purif Technol*, 192 (2018) 309-321.
- [250] Z.Y. Zhou, D. Pinson, R.P. Zou, A.B. Yu, Discrete particle simulation of gas fluidization of ellipsoidal particles, *Chem Eng Sci*, 66 (2011) 6128-6145.
- [251] T. Oschmann, J. Hold, H. Kruggel-Emden, Numerical investigation of mixing and orientation of non-spherical particles in a model type fluidized bed, *Powder Technology*, 258 (2014) 304-323.
- [252] K. Vollmari, R. Jasevičius, H. Kruggel-Emden, Experimental and numerical study of fluidization and pressure drop of spherical and non-spherical particles in a model scale fluidized bed, *Powder Technology*, 291 (2016) 506-521.
- [253] K. Papadikis, S. Gu, A.V. Bridgwater, 3D simulation of the effects of sphericity on char entrainment in fluidised beds, *Fuel Processing Technology*, 91 (2010) 749-758.
- [254] E.S. Pettyjohn, Effect of particle shape on free settling rates of isometric particles, *Chem. Eng. Prog.*, 44 (1948) 157-172.
- [255] P.D. Komar, Settling Velocities of Circular Cylinders at Low Reynolds Numbers, *The Journal of Geology*, 88 (1980) 327-336.
- [256] R. Escudié, N. Epstein, J.R. Grace, H.T. Bi, Effect of particle shape on liquid-fluidized beds of binary (and ternary) solids mixtures: segregation vs. mixing, *Chem Eng Sci*, 61 (2006) 1528-1539.
- [257] B. Liu, X. Zhang, L. Wang, H. Hong, Fluidization of non-spherical particles: Sphericity, Zingg factor and other fluidization parameters, *Particuology*, 6 (2008) 125-129.

- [258] W. Nan, Y. Wang, J. Wang, Numerical analysis on the fluidization dynamics of rodlike particles, *Advanced Powder Technology*, 27 (2016) 2265-2276.
- [259] X. Wu, K. Cen, Z. Luo, Q. Wang, M. Fang, Measurement on particle rotation speed in gas–solid flow based on identification of particle rotation axis, *Experiments in Fluids*, 45 (2008) 1117-1128.
- [260] N. Epstein, Chapter 26 Liquid-solids fluidization, in: W.C. Yang (Ed.) *Handbook of fluidization and fluid-particle systems*, CRC press, New York, 2003.
- [261] J.J.J. Chen, Improved Equation for the Calculation of Minimum Fluidization Velocity - Comment, *Ind Eng Chem Res*, 26 (1987) 633-634.
- [262] S.C. Saxena, G.J. Vogel, Measurement of Incipient Fluidization Velocities in a Bed of Coarse Dolomite at Temperature and Pressure, *T I Chem Eng-Lond*, 55 (1977) 184-189.
- [263] A. Dziugys, B. Peters, An approach to simulate the motion of spherical and non-spherical fuel particles in combustion chambers, *Granul Matter*, 3 (2001) 231-265.
- [264] A. Holzer, M. Sommerfeld, New simple correlation formula for the drag coefficient of non-spherical particles, *Powder Technology*, 184 (2008) 361-365.
- [265] E. Loth, Drag of non-spherical solid particles of regular and irregular shape, *Powder Technology*, 182 (2008) 342-353.
- [266] J. Werther, Measurement techniques in fluidized beds, *Powder Technol*, 102 (1999) 15-36.
- [267] N.P. Cheremisinoff, Review of Experimental Methods for Studying the Hydrodynamics of Gas Solid Fluidized-Beds, *Ind Eng Chem Proc Dd*, 25 (1986) 329-351.

- [268] J. Chaouki, F. Larachi, M.P. Dudukovic, Noninvasive tomographic and velocimetric monitoring of multiphase flows, *Ind Eng Chem Res*, 36 (1997) 4476-4503.
- [269] M. Abbasi, R. Sotudeh-Gharebagh, N. Mostoufi, M.J. Mahjoob, Non-intrusive monitoring of bubbles in a gas–solid fluidized bed using vibration signature analysis, *Powder Technology*, 196 (2009) 278-285.
- [270] A. Sheikhi, R. Sotudeh-Gharebagh, M. Alfi, N. Mostoufi, R. Zarghami, Hydrodynamic characterisation of liquid–solid two–phase fluidised beds: Vibration signature and pressure fluctuations analyses, *The Canadian Journal of Chemical Engineering*, 90 (2012) 1646-1653.
- [271] A. Sheikhi, R. Sotudeh-Gharebagh, N. Mostoufi, R. Zarghami, Experimental investigation on the hydrodynamics of a gas–liquid–solid fluidized bed using vibration signature and pressure fluctuation analyses, *International Journal of Heat and Fluid Flow*, 42 (2013) 190-199.
- [272] J.W. Chew, R. Hays, J.G. Findlay, T.M. Knowlton, S.B. Reddy Karri, R.A. Cocco, C.M. Hrenya, Cluster characteristics of Geldart Group B particles in a pilot-scale CFB riser. I. Monodisperse systems, *Chem Eng Sci*, 68 (2012) 72-81.
- [273] N. Ellis, L.A. Briens, J.R. Grace, H.T. Bi, C.J. Lim, Characterization of dynamic behaviour in gas–solid turbulent fluidized bed using chaos and wavelet analyses, *Chem Eng J*, 96 (2003) 105-116.
- [274] X. Lu, H. Li, Wavelet analysis of pressure fluctuation signals in a bubbling fluidized bed, *Chem Eng J*, 75 (1999) 113-119.
- [275] J. Ren, Q. Mao, J. Li, W. Lin, Wavelet analysis of dynamic behavior in fluidized beds, *Chem Eng Sci*, 56 (2001) 981-988.
- [276] G.-B. Zhao, Y.-R. Yang, Multiscale resolution of fluidized-bed pressure fluctuations, *AIChE Journal*, 49 (2003) 869-882.

- [277] J.W. Chew, R. Hays, J.G. Findlay, S.B.R. Karri, T.M. Knowlton, R.A. Cocco, C.M. Hrenya, Reverse Core-Annular Flow of Geldart Group B Particles in Risers, *Powder Technology*, 221 (2012) 1-12.
- [278] L.A. Briens, C.L. Briens, A. Margaritis, S.L. Cooke, M.A. Bergougnou, Characterization of channelling in multiphase systems. Application to a liquid fluidized bed of angular Biobone particles, *Powder Technology*, 91 (1997) 1-9.