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Characterizing the scouring efficiency of Granular Activated Carbon (GAC) particles in membrane fouling mitigation via wavelet decomposition of accelerometer signals

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Abstract

Particle fluidization is a promising unsteady-state shear means of mitigating membrane fouling, thereby potentially lower the energy requirement in membrane-based water 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, the scouring efficacy of three Granular Activated Carbon (GAC) particle diameters (namely, 1.20 mm, 1.85 mm, and 2.18 mm) has been investigated using an accelerometer to reveal the fluid dynamics. Specifically, because both liquid and solid phases contribute to the accelerometer signal, wavelet decomposition was used to extract information in the higher-frequency detail signals that reflect the particle dynamics. The energy contained in the accelerometer signal indicative of the solid phase dynamics correlated well with the extent of fouling mitigation in the filtration tests; the liquid contribution was less significant when the GAC particles were fully fluidized. Results indicate that the smallest particle diameter of 1.20 mm conferred the least scouring efficiency, while both the larger particle diameters of 1.85 mm and 2.18 mm provided similar scouring efficiency. Calculations of energy requirement indicate that the energy requirement of all three particle diameters (d_p) were similar at lower scouring efficiency, whereas the energy requirement of the smallest d_p of 1.20 mm was the greatest at higher scouring efficiency. Optimization of the particle diameter hinges on the balance between particle inertia, and the difference between superficial liquid velocity and minimum fluidization velocity.

Keywords:

Membrane fouling; liquid-solid fluidization; particle diameter; scouring efficiency; energy cost

1. Introduction

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. Except for very dilute feeds, tangential shear by liquid cross flow is used to mitigate fouling. However, for enhanced fouling control, unsteady-state shear means have been identified as potentially cost-effective [1-4]. Techniques that mitigate fouling by unsteady tangential shear forces on the membrane surface include 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 Granular Activated Carbon (GAC) [13, 15-24]. Specifically for activated carbon particles, besides the adsorption capacity, the fluidized 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, 25]. 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 (AFMBRs) 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) [26]. Unfortunately, the key obstacle in the implementation of MBRs is membrane fouling, which reduces the membrane permeability and increases the operation cost [27-30]. Kim et al. [13] proposed a two-stage anaerobic fluidized bed bioreactor for wastewater treatment, with granular activated carbon (GAC) 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 (AFBR), while the second stage is an anaerobic fluidized bed membrane bioreactor (AFMBR). 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 deposits on the membrane surface). Investigations revolving around the AFMBR 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 [31]. To date, the hydrodynamics of the liquid-solid fluidization of the GAC particles is not well 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.

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 low levels of discrimination are recognized [32-35]. 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 [35], in this case by the liquid flow, particles colliding with each other and with vessel walls, or structural vibrations [36]. Due to the complex nature of the signals, AE is largely used as a fingerprint in describing a specific set of conditions without necessarily knowing or understanding their cause [35]. Efforts harnessing such acoustic methods have been reported to determine the various facets of fluidization, for example, the dynamic behavior of the particles [37, 38], the onset of unstable fluidization [39], general bed dynamics and regime transition [36, 38, 40-43], average size of particles [44], and determining the minimum fluidization velocity (U_{mf}) [45, 46]. Because wavelet decomposition [47, 48] is capable of extracting the various frequency ranges, thereby well-acknowledged to enable classification of fluidized-bed measurement data into noise (micro-scale), flow structures like clusters or bubbles (meso-scale), and equipment (macro-scale) [49-54], we harness wavelet decomposition to extricate the dynamics of the GAC particles in this effort. Therefore, the sensitivity of AE signals to particle movement, coupled with the data analysis method of wavelet decomposition [47, 48], makes the accelerometer an ideal technique for the goal of understanding the scouring mechanism of the fluidized GAC particles in mitigating the fouling of membranes.

Despite particle fluidization becoming increasingly popular for membrane fouling mitigation [13, 15-24], gaps in the knowledgebase exist with regards to an in-depth understanding of (i) the hydrodynamics of the different particle diameters in enabling the scouring effect, and also (ii) the optimization of extent of fouling mitigation and energy requirement. Therefore the objectives of the

current study are threefold: (1) use an accelerometer, which can non-invasively characterize the fluidized bed dynamics due to flow-induced acoustic emissions [35], 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.

2. Materials and methods

2.1 Experimental setup

A schematic of the liquid-solid fluidization-membrane filtration setup used in this work is shown in Fig. 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. 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, which corresponds approximately to 60 % of the membrane module height to avoid any flow instabilities due to the distributor; the position of the accelerometer was fixed, because while horizontal variations are not detectable due to the non-intrusive nature of the probe, vertical variations were not detected presumably due to the limited membrane module height. 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 1. (a) Schematic of experimental setup, and (b) Distributor in the liquid-solid fluidization reactor to ensure uniform fluidization.

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 [55], 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 a 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 hours. The same concentration of GAC of 100 g/L was used for every test. Because of the short duration of fluidization (< 30 minutes), 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 minutes then agitated with a magnetic stirrer at 500 rpm for 20 minutes.

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 minutes 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.

2.3 Analysis of accelerometer signals

The accelerometer signals as a function of time are presented in Fig. 2 for the liquid-solid fluidization of GAC particles of various sizes (Fig. 2a - c) and for liquid flow alone (Fig. 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. 2a - d), Fig. 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 inability 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. Hence, another analysis method has to be implemented.

The frequencies of the signals for the various particle diameters at the same U_l are clearly different (Fig. 2), and consequently 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 [49-54]. More specifically, as summarized in Fig. 4, 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). 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. More details of the wavelet decomposition technique can be found in Mallat [47, 48]. All accelerometer signals were analyzed via wavelet decomposition using Matlab 2013. Fig. 5 displays the approximation and detail signals at various scales, illustrating (i) the decrease in signal frequency with the increase of scale, and (ii) negligible frequency information is present from scale 10 onwards, hence the maximum scale of decomposition in this work was capped at 13.

Figure 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. Average values of accelerometer signal for the three GAC diameters investigated

Figure 4. Wavelet decomposition analysis of experimental signal into various scales [57]

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 [50, 53, 57]:

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

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. The unit for E is m^2/s^4 , which is physically meaningless hence is omitted. 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 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

4. Results and discussion

4.1 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 (U_l) [58-60], as shown in Fig. 6 for each particle diameter (d_p) investigated. Specifically, U_{mf} is identified as the superficial liquid velocity (U_l) at which the upward fluid drag balances the bed weight, beyond 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. 6, the U_{mf} values for GAC particles with diameters of 2.18 mm, 1.85 mm and 1.20 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 (which is proportional to the liquid flow rate). On the other hand, the greater shear induced by the larger GAC particles leads to more effective mitigation of fouling [31]. Therefore, an optimal GAC particle diameter (d_p) should exist that balances increasing energy cost and increasing scouring efficiency with increasing d_p .

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

4.2 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 possible 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. 7 shows the energy associated with each detail signal obtained via wavelet decomposition (Eq. 1) for various data acquisition frequencies at incipient fluidization (i.e., $U_l = U_{mf,2.18mm}$) of the GAC particles with $d_p = 2.18$ mm. At the lower frequencies (i.e., 10 kHz and 20 kHz), only one peak is observed, implying that higher frequencies are needed to resolve 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 and 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. It should be noted that

the E_{Dj} values representing the liquid signals ($D_5 - D_7$) are larger than that of the solid ($D_1 - D_3$) in Fig. 7, because the GAC particles were only incipiently fluidized at $U_l = U_{mf,2.18mm}$.

Figure 7. Wavelet energy distribution of accelerometer sub-signals with various sampling frequency (namely, 10 kHz, 20 kHz, 30 kHz, and 40 kHz) for the fluidization of GAC particles with diameter of 2.18 mm at $U_l = 0.0085$ m/s (which equals to $U_{mf,2.18mm}$).

Figs. 8a, 8b and 8c 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. 8d shows the corresponding energy plots for liquid flow alone (i.e., in the absence of particles). It is worth noting that the accelerometer was non-intrusive, hence the signal was inevitably dampened somewhat through the vessel wall, but the non-disruption of the hydrodynamics is a key advantage of such non-intrusive probes. The areas under the plots increase as U_l increases due to the associated higher energy. For liquid flow alone (Fig. 8d), or in cases where GAC particles were present but the U_l values were lower than the characteristic U_{mf} of the particles (Fig. 8a - c), only one peak at $D_5 - D_7$ exists. On the other hand, when the superficial liquid velocity (U_l) was above the characteristic U_{mf} of the particles (Fig. 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. 8a, two peaks appear when U_l was 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. 8a - d 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. Regarding the relative shearing efficiency of the solid and liquid phases, the ratio $(E_{D1} + E_{D2} + E_{D3}) / (E_{D5} + E_{D6} + E_{D7})$ could provide an indication, as shown in Fig. 8e. For the system without GAC, the $(E_{D1} + E_{D2} + E_{D3}) / (E_{D5} + E_{D6} + E_{D7})$ values are expectedly negligible regardless of U_l . In cases wherein GAC particles were present, three trends with respect to U_l are interesting: (1) The intermediate particle diameter of $d_p = 1.85$ mm gave the highest $(E_{D1} + E_{D2} + E_{D3}) / (E_{D5} + E_{D6} + E_{D7})$ values at $0.0074 \leq U_l \leq 0.0095$ m/s, because the attendant inertia was greater than that of $d_p = 1.20$ mm on one hand, while $U_l - U_{mf}$ was greater than that of $d_p = 2.18$ mm on the other hand; (2) At U_l lower than 0.007 m/s, $(E_{D1} + E_{D2} + E_{D3}) / (E_{D5} + E_{D6} + E_{D7})$ is negligible for $d_p = 2.18$ mm, but non-negligible for $d_p = 1.20$ and 1.85 mm, presumably because (i) the smaller particles acted as a distributor causing the liquid to be more turbulent above the particle bed, and (ii) although the smallest GAC with $d_p = 1.20$ was fully fluidized beyond $U_l = 0.0055$ m/s, the associated inertia was weak; (3) The $(E_{D1} + E_{D2} + E_{D3}) / (E_{D5} + E_{D6} + E_{D7})$ values tend to increase then decrease with U_l - the initial increasing trend at lower U_l was due to increasing $U_l - U_{mf}$, while the decreasing trend at higher U_l was due to the increasing liquid turbulence.

Figure 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); (e) Relative shearing contribution by the solid ($E_{D1}+E_{D2}+E_{D3}$) and liquid ($E_{D5}+E_{D6}+E_{D7}$) with respect to U_l .

To further explore the impact of GAC particle diameter (d_p), each subplot in Fig. 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. 9a), while the other three are the U_{mf} values corresponding to each of the d_p investigated (Fig. 9b - d). Fig. 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 $U_l = 0.0042$ m/s. In Fig. 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. 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 dominated the acoustic effect. Finally for Fig. 9d, peaks at $D_1 - D_3$ are seen for all three GAC particle diameters but not for the liquid only system. In this case, the GAC particles with $d_p = 1.85$ 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) had the greatest $U_l - U_{mf}$ but least inertia, the largest GAC particles (i.e., $d_p = 2.18$ mm) had the least $U_l - U_{mf}$ but greatest inertia. In addition, it is interesting to note that, while the contributions of the liquid (i.e., $D_5 - D_7$) and solid (i.e., $D_1 - D_3$) phases were comparable for $d_p = 1.20$ and 1.85 mm in Figs. 9c and 9d, the contribution of the liquid phase was much greater than that of the solid phase for $d_p = 1.20$ in Fig. 9b and $d_p = 2.18$ mm in Fig. 9d. This re-emphasizes the interplay between particle inertia and particle mobility. In Fig. 9b, although the U_{mf} of the $d_p = 1.20$ mm particles has been exceeded, the liquid phase still dominated due to the lower particle inertia associated with the smaller particles. In Fig. 9d, although the U_{mf} of the $d_p = 2.18$ mm particles has been exceeded, the liquid phase still dominated due to the lower particle mobility associated with the larger particles.

Figure 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. 8 and Fig. 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. 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 [45, 46], 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 gave negligible values, and (ii) for the systems with GAC, the sum of E_{D1} , E_{D2} and E_{D3} for each d_p only started 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 $U_l - U_{mf}$ was the greatest, the inertia was the least, the latter of which presumably dominated 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 was countered by the lower $U_l - U_{mf}$ value; therefore, $d_p = 1.85$ mm seems to represent the optimal size of the particles tested 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 cost (which is proportional to liquid flow rate).

Figure 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).

4.3 Filtration results

The scouring effect of various GAC particle sizes was investigated in filtration experiments. Fig. 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. 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. 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. 10. Fig. 11b shows the relationship between $\Delta\text{TMP}/\Delta t$ and $E_{D1} + E_{D2} + E_{D3}$ for the six datasets in Fig. 11a. Recall that $E_{D1} + E_{D2} + E_{D3}$ has been shown to correlate well with particle dynamics in Fig. 9, hence Fig. 11b aims to further relate particle dynamics to the extent of fouling mitigation induced via scouring. Specifically, $\Delta\text{TMP}/\Delta t$ was obtained by dividing the TMP rise in the 60 minutes investigated by the 60 minutes duration. Fig. 11b indicates that a slight increase in $E_{D1} + E_{D2} + E_{D3}$ conferred by fluidizing the smallest GAC ($d_p = 1.20$ mm) at the corresponding U_{mf} results in a significant reduction in $\Delta\text{TMP}/\Delta t$, 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 U_l gave slight further improvement in $\Delta\text{TMP}/\Delta t$. It should be noted that this trend may be unique to Bentonite, which is relatively easier to remove than other foulants. For the practical MBR process wherein a mixture of foulants with different characteristics are present, the trend in Fig. 11b is expected to hold qualitatively but not so quantitatively.

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

4.4 Energy consumption

For a fluidized bed, the power requirements can be evaluated by the following equation [61]:

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

where P 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:

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

Accordingly, Fig. 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 according to Eq. 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 was insignificant with regards to energy cost incurred in mitigating fouling at lower scouring efficiency, but the larger particle diameters (d_p) demanded less energy at higher scouring efficiency. As noted earlier, the Bentonite foulant investigated here is relatively easier to remove, hence the scouring (Fig. 11b) and P_p required is presumably lower, in which case Fig. 12 shows that d_p exerted 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 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).

Conclusions

The scouring efficiency of three GAC 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, based on 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 smallest particle diameter of 1.20 mm conferred the least scouring efficiency, while both the larger particle 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 various GAC particle diameters was calculated. At lower scouring efficiency, the energy requirement of all three particle diameters (d_p) 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.

Different particle properties (e.g., density, shape), particle concentrations, reactor scales and foulant types are all expected to affect the extent of fouling mitigation possible via fluidization, hence more understanding on these parameters are needed. Also, the question of whether higher efficacy in the removal of reversible fouling by the larger particles, even at a higher energy expense, may be more beneficial for the long-term operation of MBRs needs to be addressed. Nonetheless, this study on the impact of particle diameter via the use of wavelet decomposition to analyze accelerometer signals serve as a useful basis for the correlation of particle dynamics with fouling mitigation via scouring.

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hihglights

- Scouring efficacy of three GAC sizes investigated using an accelerometer.
- Wavelet decomposition used to extract frequency ranges of the solid phase.
- Energy representing the solid phase consistent with fouling mitigation.
- Choice of particle size hinges on balance between particle inertia and energy cost.
- Smallest particles required highest energy cost at higher scouring efficiency.

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