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Graphical Abstracts

Impact of oxygen

Impact of salt

Effects of salt- and oxygen-coupled stimuli on the reactive behaviors of hemoglobin-loaded polymeric membranes

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Abstract: A high-performance polymeric membrane is usually associated with excellent electrochemical and mechanical behaviors. Thereby, this paper examines the effects of salt- and oxygen-coupled stimuli on reactive behaviors of hemoglobin-loaded polymeric membranes with varying initial fixed charge densities. For capturing the coupled chemo-electro-mechanical responses of the membrane, a multiphysics model is mathematically formulated and then experimentally validated. The numerical finding unveils that the Donnan potential strength of polyacidic membrane decreases with increase of ambient oxygen level, whereas the Donnan potential of present polyampholytic membrane, at neutral pH conditions, is almost invariant towards changes of environmental salt concentration. When the environmental salt concentration is smaller than the initial fixed charge concentration of the membrane, the surface conductivity of the system is enhanced bi-linearly with increase of the salt concentration due to weakened Donnan potential strength acting over the polymer-solution, while the ion transport in the system is dominantly diffusion-governed if environmental salt concentration is greater than the initial fixed charge concentration of the membrane. Ultimately, these findings can be employed to systematically design and optimize the dual salt-oxygen reactive hemoglobin-loaded polymeric membrane.

Keywords: Dual oxygen-pH-sensitive polymeric membrane; Hydration-induced deformation; Donnan potential; Surface conductivity

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1. Introduction

Dissolved oxygen O_2 and salt are two emerging biofuels in the field of bioenergy, especially in seawater battery cell system [1]. As such, there is a necessity to explore hemoglobin-loaded polymeric membrane as an innovative coupled oxygen-salt biofuel material platform. Unfortunately, the impacts of coupled salt- and oxygen-stimuli on responsive characteristics in hemoglobin-loaded polyelectrolyte membrane remain unclear [2]. Therefore, this paper examines the electrochemical and mechanical behaviors of the membrane, especially its surface conductivity [3, 4], Donnan potential [5, 6] and hydration-induced swelling deformation [7]. Moreover, the effects of initial fixed charge density on the responsive performance of hemoglobin-loaded polyacidic, polybasic and polyampholytic membranes are also investigated as a function of ambient oxygen O_2 coupled with environmental salt.

We now ask the question of how environmental salt concentration modifies the reactive electrochemical behaviors of the polymeric membrane in particular its surface conductivity and Donnan potential. For this aim, literature reviews are conducted, where it is found that the surface conductivity of charged polymeric system enhanced bi-linearly by increasing environmental salt concentration [3, 8, 9], where the conductivity was increased sharply at low salt concentrations. However, Kamcev *et al.* recently reported that the conductivity level of charged polymeric system was almost invariant with change of low environmental salt concentration [10]. As such, there are discrepancies of experimental observations in literature for the salt-induced surface conductivity of charged polymeric system. Furthermore, the relationship between the polymeric system surface conductivity and Donnan potential strength acting over the polymer-solution interface also remains unclear currently in the literature, especially when subjected to couple salt- and oxygen-stimuli [11].

Besides the surface conductivity and Donnan potential, the literature surveys also unveiled that the understanding of salt-induced hydration behaviors of polyelectrolyte membranes remains incomplete [12], especially regarding the tendency of the membrane to swell at greater salt concentration, or so-called the anti-polyelectrolyte behaviors [13, 14]. The current understanding is that polyacidic/basic membrane experiences a sigmoid-shaped like de-swelling behaviors by increasing environmental salt concentration [15, 16]. Furthermore, the previously developed chemo-electro-mechanical models for investigating polymeric membrane also revealed that salt-induced volumetric behaviors of the membrane is divided into two parts [17-19]: (1) the membrane significantly dehydrated with increase of environmental salt concentration and (2) the membrane remained at a collapse state, when the environmental salt concentration was further increased. Interestingly, recent works by de la Cruz *et al.* via a chemo-electro-mechanical model revealed that membrane swelled when the environmental salt concentration is significantly increased [20]. (For chemo-electro-mechanical models which applied the poroelasticity theory for examining polymeric membrane, please refer to publications by Sou's group [21, 22].) As such, several contradictory hydration-induced swelling deformation patterns were reported in the literature, where polyelectrolyte membranes undergone either swelling-to-collapse [17], collapse-to-swelling-collapse [23] or swelling-to-collapse-to-swelling [20] volumetric transition behaviors with increase of environmental salt concentration. However, recent investigations pointed to effects of initial fixed charge density governing the membrane hydration behaviors, especially its anti-polyelectrolyte mechanical and electrochemical performances. Thereby, it is worthwhile and necessary to investigate the effects of salt concentration on transport and hydration behaviors of hemoglobin polyelectrolyte membranes with varying initial fixed charge densities.

In terms of impacts of oxygen, to the best of our knowledge, no effort was conducted to examine effects of ambient oxygen O_2 level on responsive surface conductivity and Donnan potential behaviors of hemoglobin-loaded polyelectrolyte membrane especially when subjected to variation of environmental salt solutions, resulting in incomplete understanding of such materials in literature. It is commonly known that despite the attractive capability of the hemoglobin for sensing and capturing oxygen O_2 in salt solution [24], a few crucial electrochemical reactions influence performance of free hemoglobin in biological fluids including: (1) the ionization and denaturation of the hemoglobin, responding to the environmental physiological conditions [25] and (2) the oxidization of oxygen-binding group (heme) in the hemoglobin [26]. Therefore, researchers began immobilizing hemoglobin into polymeric membranes as a plausible strategy to enhance the performance of the hemoglobin for sensing and storing of oxygen O_2 in salt solution [25-31]. Unfortunately, due to lack of literatures on the hemoglobin-mediated oxygen transport in polyelectrolyte membranes, it is also worthwhile and necessary to elucidate impacts of ambient oxygen level on the transport performances in the membranes especially the surface conductivity [32, 33] and Donnan potential of polyelectrolyte membranes [34, 35], when operating in salt-loaded environmental solutions.

The aim of this work is to investigate effects of salt-oxygen coupled stimuli on responsive surface conductivity, Donnan potential and swelling deformation of polyelectrolyte membranes via a [steady-state](#) multiphysics model, where the model includes mass transport equations coupled with a non-linear mechanical equation [36]. In order to explain the multiphysics interactions within the membrane, two types of mass transport equations are integrated into the multiphysics model, capturing oxygen- and salt-induced behaviors of the membrane: (1) the

diffusion equation for hemoglobin-mediated oxygen transport in the membrane and (2) the Poisson-Nernst-Planck equation for fixed charge group-governed counter- and co-ion transport in the polymeric system [37]. The model is deemed validated as a good agreement is achieved between the numerical results and published experimental data in open literature. Subsequently, the model is executed to elucidate effects of coupled salt- and oxygen-concentrations on surface conductivity and Donnan potential of oxygen-sensitive hemoglobin-loaded polyelectrolyte membranes.

2. Methodology

In order to investigate the responsive chemo-electro-mechanical behaviors of hemoglobin-loaded polyelectrolyte membranes, a multiphysics model is developed to characterize [38]: (i) the diffusion- and/or migration-governed ions and molecules transport between the polymer-solution [39]; (ii) the bio-electrochemical interaction between the mobile solutes/ligands (oxygen and ions) and the fixed functional components (hemoglobin and charge group); (iii) the electrical interactions between mobile ions in the membrane and fixed charge groups in the membrane; (iv) the formation of Donnan potential due to imbalance ionic concentration over the polymer-solution interface; (v) the hydration-induced swelling deformation of the membrane, arising partly from the Donnan potential, and (vi) the equilibrium surface conductivity of the membrane.

The present multiphysics model is formulated based on the following assumptions:

- (i) The oxygen O_2 concentration in environmental solution is a function of ambient oxygen O_2 level, such that the namely $[O_2] = \alpha pO_2$, where α is the solubility of oxygen O_2 in the solution and pO_2 is the ambient oxygen O_2 level (mmHg).

- (ii) The net charge density of the hemoglobin-loaded polyelectrolyte membranes is contributed by both the fixed (acidic and/or basic groups) and mobile (co and counter ions) charges, where the fixed charge groups are decorated uniformly onto the polymeric network chains [40].
- (iii) The membrane size is significantly larger than its Debye length, such that electroneutrality condition in the entire membrane can be assumed.
- (iv) Due to the macro-porous nature of the present polymeric membrane, the diffusivity of mobile species in the membrane is equal to its counterpart in the environmental solution, resulting in the independence between diffusivity of mobile species in the polymeric membrane and hydration degree of the membrane.
- (v) The oxygen O_2 reversibly binds to the immobilized hemoglobin in the membrane, and thus the oxygen O_2 concentration in the membrane is summation of mobile (dissolved in microenvironment) and fixed (oxygen-hemoglobin complex) oxygen O_2 concentrations.

2.1 Hemoglobin-oxygen reaction

It is well-established that the formation of oxyhemoglobin on the polymeric network chains is governed by equilibrium reactions between hemoglobin and oxygen O_2 [41, 42], as shown below

where Hb / HbH^+ and O_2Hb / O_2HbH^+ refer to reduced/protonated reduced and oxygenated/protonated oxygenated hemoglobins, while K_{O_2} denotes association constant of oxygen O_2 from the hemoglobin.

The immobile hemoglobin consists of ionizable functional components, where the ionization of the hemoglobin is determined by the equilibrium electrochemical reactions for the binding/unbinding of hydrogen H^+ ion from the membrane [41]

where K_1 and K_2 represent association constants for reduced and oxygenated hemoglobins, and they can be written as $K_1 = [HbH^+]/[H^+][Hb]$ and $K_2 = [O_2HbH^+]/[H^+][O_2Hb]$. We assume that these reactions are assumed to be in equilibrium locally due to faster reaction rate, in comparison with progression of membrane hydration.

The total hemoglobin concentration $[Hb]^{Total}$ in the membrane is can written as

$$[Hb]^{Total} = [Hb] + [O_2Hb] + [HbH^+] + [O_2HbH^+] \quad (3)$$

where it consists of reduced and protonated reduced hemoglobins, and oxygenated and protonated oxygenated hemoglobins.

The saturation of hemoglobin S with the oxygen O_2 is given as

$$S = \frac{[HbO_2]}{[Hb]^{Total}} \quad (4)$$

where the oxygenated hemoglobin concentration $[HbO_2]$ can be described as follows

$$\begin{aligned} [HbO_2] &= [O_2Hb] + [O_2HbH^+] \\ &= K_{O_2} [O_2][Hb] (1 + K_2 [H^+]) \end{aligned} \quad (5)$$

while the total hemoglobin concentration $[Hb]^{Total}$ can be written as

$$\begin{aligned}
 [\text{Hb}]^{\text{Total}} &= [\text{Hb}] + [\text{HbH}^+] + [\text{HbO}_2] \\
 &= [\text{Hb}] \left(1 + K_1 [\text{H}^+] + K_{\text{O}_2} [\text{O}_2] (1 + K_2 [\text{H}^+]) \right)
 \end{aligned}
 \tag{6}$$

The rate of oxygenation of membrane can be characterized mathematically by

$$r_{\text{O}_2} = k_{\text{OxyHb}} (1-S) [\text{O}_2] [\text{Hb}]^{\text{Total}} + k_{\text{Oxy}} S [\text{Hb}]^{\text{Total}} \tag{7}$$

where k_{OxyHb} and k_{Oxy} refer to forward and backward biochemical rates for association of oxygen with hemoglobin. The first term on the right characterizes the association of oxygen-hemoglobin complexes, while the second term on the right describes the dissociation of oxygen O_2 from hemoglobin. There is no consumption of oxygen O_2 in the system, which explains the absent of oxygen O_2 sink term in the [polymeric membrane](#).

2.2 Acid and base reactions

It is commonly known that the ionization of weakly acidic and basic groups bounded onto polymeric network chains of the membrane is determined by the electrochemical reactions for association/dissociation of hydrogen ion with the ionizable functional components [\[17, 43\]](#)

where A and B are fixed acidic and basic groups, and K_a and K_b are dissociation constants for the equation 8, in which they can be written as $K_a = \frac{[\text{A}^-][\text{H}^+]}{[\text{HA}]}$ and

$$K_b = \frac{[\text{B}][\text{H}^+]}{[\text{HB}^+]}$$

The initial ionizable dry fixed acidic $C_A^{o,D}$ and basic group $C_B^{o,D}$ concentrations can be written as

$$C_A^{o,D} = [\text{A}^-] + [\text{HA}] \quad \text{and} \quad C_B^{o,D} = [\text{HB}^+] + [\text{B}] \tag{9}$$

1 where by employing the dissociation constants described above, we can re-write equation 9 into
 2 equation 10, in which the initial ionizable dry fixed acidic $C_A^{o,D}$ and basic group $C_B^{o,D}$
 3 concentrations can be transformed into its counterpart hydrated state by $C_A^{o,D} = \frac{C_A^o}{H}$ and
 4 $C_B^{o,D} = \frac{C_B^o}{H}$, where H is the hydration of the membrane and is defined as ratio of final volume V_f
 5 to initial volume V_o of the membrane [44].

$$\left[A^- / C_A \right] = C_A^{o,D} \frac{K_a}{K_a + [H^+]} \quad \text{and} \quad \left[HB^+ / C_B \right] = C_B^{o,D} \left(\frac{[H^+]}{K_b + [H^+]} \right) \quad (10)$$

6 2.3 Formulation of the multiphysics model

7 For the development of the model, at least five mobile species are considered in the system,
 8 namely oxygen (O_2), hydrogen ion (H^+), hydroxide ion (OH^-), a cation and an anion solutes.
 9 For this, the mass conservation law is employed to described the mass transport between
 10 polymer-solution [18]

$$\nabla \cdot \mathbf{N}_k + r_k = 0 \quad (k = O_2, H^+, OH^-, \text{anion}, \text{cation}) \quad (11)$$

11 where $\mathbf{N}_k$ indicates molar flux (mM/s) and r_k refers to rate of chemical reaction for kth species.
 12 For charged polymeric system, under the influence of chemical concentration gradient coupled
 13 with electrical field, the mass transport between the membrane and its environmental solution
 14 can be described mathematically by a Nernst-Planck equation [45-47]

$$\nabla \cdot C^{-1} D_k [\nabla C_k + F \mu_k z_k C_k \nabla \Phi] + r_k = 0 \quad (k = O_2, H^+, OH^-, \text{anion}, \text{cation}) \quad (12)$$

in which D_k , C_k , z_k , μ_k and r_k refer to diffusion coefficient (m^2/s), mobile solutes concentration (mM), valence number, ion mobility, and rate of chemical reaction. R , T , F , and Φ denote the universal gas constant ($8.314 \text{ J mol}^{-1} \text{ K}^{-1}$), absolute temperature (K), Faraday constant ($96,487 \text{ Cmol}^{-1}$), and electrical potential of the membrane. $\mathbf{C}^{-1}$ is inverse of the right Cauchy-Green tensor.

The ion mobility for k th species can be defined by Nernst-Einstein relationship, establishing a linkage between the ion mobility and its diffusion coefficient, given as [36, 40, 44]

$$\mu_k = \frac{D_k}{RT} \quad (13)$$

In order to capture impacts of electrical potential strength on the mobile ions concentrations in the membrane, Eq 14 [48, 49], is incorporated into the model which gives the relationship between the electrical potential of the membrane and the net charge density of the system [46, 47]

$$\nabla \cdot (\varepsilon_r \varepsilon_0 \mathbf{C}^{-1} \nabla \Phi) = -\frac{F}{(1 + C\nu)} \left(\sum_k z_k C_k - C_A + C_B \right) \quad (14)$$

where ε_r is relative dielectric constant of environmental solution and ε_0 is permittivity of vacuum, and C is number of moles of water molecule and ν is volume of a mole of fluid molecules. C_A and C_B are concentration of the fixed acidic and basic charge groups, respectively.

Following the electroneutrality assumption in the polymeric membrane, the additional condition to support the Eq. 14 is given as follows

$$\sum_k z_k C_k - C_A + C_B = 0 \quad (15)$$

Surface conductivity characterizes the ionic transport of the membrane as function transferable counter- and co-ion concentrations coupled with their diffusion kinetics [50], where the surface conductivity of the membrane can be written as [10, 51]

$$K^\sigma = \frac{F^2}{RT} \left(\sum_k D_k C_k \right) \quad (16)$$

where K^σ is surface conductivity (S/m) of the membrane, where it is a function of counter- or co-ion concentration coupled with its respective diffusion coefficients.

In terms of hydration-induced swelling deformation of the membrane, the mechanical swelling pressure acting over the polymer-solution interface arises due to: (i) the elastic nature of the polymeric network chains, (ii) the non-ionic and ionic concentration difference between the membrane and environmental solution [52], (iii) the mixing between the polymeric system and the environmental solution, and (iv) the electrical interactions between the fixed charge groups and the mobile ions, which accounts for the anti-polyelectrolyte behaviors [53]. At swelling equilibrium, the hydrostatic pressure p over the polymer-solution interface is defined as the sum of the osmotic pressure, given as [46]

$$p = RT \sum_k \left(\frac{C_k}{H} - \frac{\kappa^3}{24\pi} \sigma \right) - RT \sum_k \left(\bar{C}_k - \frac{\bar{\kappa}^3}{24\pi} \bar{\sigma} \right) \quad (17)$$

$$- N_k k_B T \left[\frac{1}{(1+Cv)^{1/3}} \left(\frac{1}{3} \text{tr}(\mathbf{C}_e) - \frac{1}{(1+Cv)^{2/3}} \right) \right] - \frac{RT}{v} \left(\ln \frac{Cv}{(1+Cv)} + \frac{1}{(1+Cv)} + \frac{\chi}{(1+Cv)^2} \right)$$

where κ is inverse screening length, N_k is the number of polymer chains per volume of the dry polymer ($2.43 \times 10^{25} \text{ m}^{-3}$), k_B is the Boltzmann constant, T is the absolute temperature, χ is polymer-solvent interaction parameter which indicates the dis-affinity between membrane-solution and $\text{tr}(\mathbf{C}_e)$ is the trace of the elastic Cauchy-Green tensor $\mathbf{C}_e$, where it can be solved by

summing the tensor diagonal components [54]. The first term on the right-hand side is due to imbalance of mobile species concentration between membrane-solution which accounts for the anti-polyelectrolyte behaviors, the second term arises due to contractive force exhibited by the polymeric network chains to prevent dissolution of the membrane during hydration and the last term is due to the mixing between the polymeric network chains and environmental solution.

The inverse screening length κ can be written as [53]

$$\kappa = \sqrt{\frac{F^2}{\epsilon_r \epsilon_0 RT} \sum (C_k z_k^2 + C_f z^2)} \quad (18)$$

and σ is given as [55]

$$\sigma_k(\kappa a) = 1 - \frac{3}{2}\kappa a + \frac{9}{5}(\kappa a)^2 \quad (19)$$

where κ and a refer to inverse screening length and ion radius

It is well-established that, at swelling equilibrium condition, the hydration-induced deformation of the membrane progresses, until it is balanced by the elastic forces exerted by the polymeric network chains to achieve an equilibrium hydration state, where the swelling deformation λ_s of the free surface between membrane-solution interface can be given as [46]

$$\lambda_s = \frac{N_k k_B T}{p} = \frac{L_{gel}}{L_{gel}^0} \quad (20)$$

where $N_k k_B T$ is ground shear modulus, p is hydrostatic pressure, and L_{gel} and L_{gel}^0 are final and initial lengths of the membrane, respectively.

In this paper, only one-dimensional steady-state simulations of the multiphysics model are performed via the commercial software COMSOL for the responsive characteristics of a cylindrical hemoglobin polyelectrolyte membrane in response to environmental stimuli. The

deformation of the is taken at radial direction and restricted in axial direction, where Neumann boundary conditions are set at the center of the membrane $\frac{\partial C_k}{\partial X} = 0, \frac{\partial \Phi}{\partial X} = 0$ ($k = H^+, OH^-, \dots, N$) while Dirichlet boundary conditions are assumed at the environmental solution edges $C_k = \bar{C}_k, \Phi = 0$.

3. Result and discussions

3.1 Model validation

For model validation, the examination between present numerical results and published experimental observation is performed for: (i) the swelling deformation behaviors of polybasic and polyampholytic membranes as a function of environmental salt concentration (ii) the surface conductivity of polyacidic and polybasic polymeric systems a function of environmental salt concentration.

Fig. 1(a) compares the deformation of polybasic membrane as a function of environmental salt concentration C_{NaCl} for system with different initial fixed charge densities. The material parameters required by the model are given as $L = 15$ mm, $L_{gel}^0 = 1.5$ mm, $K_A = 10^{-10}$, $\chi = 0.3$, $C_B^{0,A} = 175$ mM/ $C_B^{0,B} = 810$ mM (for A and B simulation inputs, respectively) [17, 56], while the remaining inputs are provided in Table. 1. For the present simulation, the membrane is immersed in environmental pH of 7.0 and ambient oxygen O_2 level 160 mmHg, with temperature 37 °C. As observed in Fig. 1(a), the results show that the swelling deformation of the membrane decreases in a sigmoid-like pattern with the enlargement of environmental salt concentration, due to weakening of osmotic swelling pressure by increasing salt concentration. On the other hand, Fig. 1(b) probes the deformation of polyampholytic membrane as a function of environmental salt concentration for system with different initial fixed charge densities. The material

parameters required by the model are given as $L=1000 \mu\text{m}$, $L_{gel}^0=252.5 \mu\text{m}$, $K_A=10^{-1.5}$, $K_B=10^{-11.5}$, $\chi=0.3$, $C_A^0+C_B^0=700 \text{ mM}$, where $C_B^{0,C}-C_A^{0,C}=-2 \text{ mM}$ or $C_B^{0,D}-C_A^{0,D}=-14 \text{ mM}$ [57], while the remaining inputs are provided in Table.1. The polyampholytic membrane undergoes collapse-to-swelling behaviors, or so-called the anti-polyelectrolyte phenomena, due to high diffusion of mobile species into the membrane which promotes its swelling behaviors. Overall, the present numerical results are in a good agreement with the published experimental observations, consequently validating the model.

We now examine the surface conductivity of anion- and cation-charged polymers via present numerical results and published experimental data [9] via Fig. 2. The material parameters required by the model are given as $L=900 \mu\text{m}$, $L_{gel}^0=90 \mu\text{m}$ [9], $\chi=0.1$ (highly hydrated system) [58], fully ionized initial fixed charge group density $C_A^{0,CMX}=7620 \text{ mM}$ / $C_B^{0,AMX}=7930 \text{ mM}$ [10] (for CMX and AMX simulation inputs, respectively), while the remaining inputs are provided in Table. 1. As seen in Fig. 2, the surface conductivity of the membrane increases monotonously by increasing the environmental salt concentration where the surface conductivity reaches an almost convergence state when the salt concentration is increased larger than $\sim 0.1 \text{ M}$, due to the diminishing electrical potential gradient between membrane-solution interface. The higher membrane solubility (*i.e.* smaller χ values) causes the greater stretching of the polymeric network chains, diluting the counter- and co-concentrations in the polymer and reducing the fixed charge group density in the membrane which decreases the Donnan potential strength acting over the membrane-solution interface. These coupled effects lead to the plateauing of the polymeric system surface conductivity with increase of salt concentration, as seen in Fig. 2. As such, the polymeric system associated with greater hydrophilic interaction with environmental solution leads to decrease of its surface conductivity values, given that the

environmental salt concentration and initial fixed charge group density of the polymeric systems are the same.

After comparison with the experimental data, it is concluded that the multiphysics model can characterize well the coupled bio-chemo-electro-mechanical responses of hemoglobin-loaded membrane, and thus the model is validated.

3.2 Effects of salt coupled with oxygen on the pNIPAM-based membranes

In order to investigate the effects of environmental salt concentration coupled with ambient oxygen level on surface conductivity and Donnan potential of hemoglobin-loaded pNIPAM-based polyelectrolyte membranes, the multiphysics is necessarily performed, in which the material parameters required by the model are given as $L=100 \mu\text{m}$, $L_{gel}^0=50 \mu\text{m}$, $K_A=10^{-5}$ / $K_B=10^{-9}$, $\chi=0.91$, $C_A^0/C_B^0=100 \text{ mM}$ [59], and the remaining inputs are provided in Table. 1. For the present numerical simulation, unless specified otherwise, the membrane is immersed in an environmental solution of pH 7.40 with ambient temperature of 37 °C.

Fig. 3(a) examines the effects of salt concentration C_{NaCl} on oxygen-induced deformation of hemoglobin-loaded polyacidic membrane with initial fixed charge density $C_f^0=100 \text{ mM}$ [60]. Fig. 3(a) illustrates that the membrane hydrates in a sigmoid-shaped fashion with increase of ambient oxygen O_2 level, where the membrane linear swelling deformation response ranges from oxygen O_2 level of 1 to 100 mmHg. When $C_f^0 \geq C_{NaCl}$, the increase of environmental salt concentration C_{NaCl} enlarges screening effects on the fixed charge group which reduces the ionic osmotic swelling pressure in the membrane, leading to its shrinkage. Surprisingly, when $C_{NaCl} > C_f^0$, the increase of environmental salt concentration C_{NaCl} causes the membrane to

significantly hydrate itself. For example, at ambient oxygen O_2 level of 100 mmHg, the membrane de-hydrates by 0.2 % if salt concentration C_{NaCl} increases from 10 to 100 mM, but the hydration of the membrane increases by 2.7 % when salt concentration C_{NaCl} increases from 100 to 1000 mM. However, the tendency of the membrane to swell at higher salt concentration (*i.e.* when $C_{NaCl} > C_f^o$) known as the anti-polyelectrolyte behavior, requires further investigation as this phenomenon remains incompletely understood in current literature, as visualized in Figs. 3-6.

Fig. 3(b) illustrates the influence of salt concentration C_{NaCl} on oxygen O_2 loaded in the polyacidic membrane as function ambient oxygen O_2 level. It appears that oxygen O_2 concentration in the membrane enlarges in a sigmoid-shaped characteristic by increasing ambient oxygen O_2 level, where the loading of oxygen O_2 in the membrane linearly increases only from ~1 to 100 mmHg. This is because the active-sites of immobile hemoglobin are saturated with oxygen O_2 when ambient level is increased beyond 100 mmHg, showing independence between oxygen O_2 loading and ambient level. In addition, Fig. 3(b) also reveals that the oxygen O_2 loading in the membrane decreases with increase of environmental salt concentration C_{NaCl} . This result is consistent with published experimental data where hemoglobin oxygen O_2 affinity decreases with increase of salt concentration C_{NaCl} [61].

The loading of counter-ion (*i.e.* sodium Na^+ ion) and co-ion (*i.e.* chloride Cl^- ion) in the present polyacidic membrane is visualized in Fig. 3(c) and Fig. 3(d), respectively, as a function of ambient oxygen O_2 level when subjected to variation of environmental salt concentrations C_{NaCl} . From Figs. 3(c) and 3(a), it is seen that the increase of hydration-induced swelling

deformation of the membrane, by enlarging ambient oxygen O_2 level, decreases the counter-ion concentration in the membrane. This is because the increase of hydration reduces fixed charge group density in the membrane. Consequently, its ability to electrically attract counter-ion into the polymeric system is weakened. For example, when at constant ambient oxygen O_2 level of 100 mmHg, the ratio of counter-ion concentration in the membrane to its environmental solution decreases with increase of environmental salt concentration C_{NaCl} , such that the ratio changes from 1.85 to 1.06 when salt concentration C_{NaCl} is enlarged from 100 to 1000 mM, demonstrating the decrease of membrane Donnan potential strength [62]. On the other hand, Fig. 3(d) shows that the loading of co-ion is enlarged in a sigmoid-shaped manner with decrease of oxygen-induced Donnan potential strength. But when subjected to lower environmental salt concentration C_{NaCl} of 10 mM, the co-ion concentration in the membrane appears to be unchanged with increase of ambient oxygen O_2 level due to relatively stronger Donnan potential strength.

Fig. 3(e) examines the influences of environmental salt concentration C_{NaCl} on Donnan potential of hemoglobin-loaded polyacidic membrane as a function of ambient oxygen O_2 level. Fig. 3(e) shows that the Donnan potential strength of the membrane decreases in a sigmoid-like shaped by increasing ambient oxygen O_2 level, in which the Donnan potential strength is further weakened with increase of environmental salt concentration C_{NaCl} . The increase of ambient oxygen O_2 level enlarges swelling deformation of the membrane which reduces fixed charge group concentration, consequently weakening Donnan potential strength of the membrane. Moreover, the increase of environmental salt concentration C_{NaCl} also reduces Donnan potential strength of the membrane, in which the greater number of mobile ions enhances the screening of

fixed charge group in the membrane, electrically neutralizing the membrane. Although, it appears that the ambient oxygen O_2 level influences the Donnan potential strength of the membrane, however, how ambient oxygen O_2 impacts the surface conductivity of the membrane remains unclear in the open literature.

Fig. 3(f) is plotted to investigate the impacts of ambient oxygen O_2 level on surface conductivity of the membrane when subjected to variation of environmental salt concentrations C_{NaCl} . A comparison between Fig. 3(e) and Fig. 3(f) shows that although the Donnan potential strength of the membrane decreases in a sigmoid-like shaped by increasing ambient oxygen O_2 level. However, the surface conductivity of the membrane remains almost unchanged with enlargement of ambient oxygen O_2 level. This is because, by increasing ambient oxygen O_2 level, the increase of hydration-induced swelling deformation promotes counter- and co-ions transport in the membrane [63], weakening the membrane Donnan potential strength. These coupled effects cause the surface conductivity of the membrane to remain almost the same with increasing ambient oxygen O_2 level. From Fig. 3(e), it is established that the increase environmental salt concentration C_{NaCl} weakens Donnan potential strength of the membrane which then promotes both counter- and co-ions transport between the interfaces over membrane-solution, enhancing the surface conductivity of the system. For example, when at ambient oxygen O_2 level of 100mmHg, the increase of environmental salt concentration C_{NaCl} from 10 to 100 mM or from 100 to 1000 mM enhances surface conductivity of the membrane by 2.23- or 9.33-fold, respectively. As seen in Fig. 3(a), the increase of environmental salt concentration C_{NaCl} from 100 to 1000 mM leads to increase hydration-induced swelling deformation coupled with decrease of Donnan potential strength, explaining the improvement of surface conductivity

by 9.33, instead of 2.23, times when environmental salt concentration C_{NaCl} is further increased by 10-fold.

Fig. 4(a) investigates the salt-induced volumetric behaviors of hemoglobin-loaded polyelectrolyte membrane with initial fixed charge density $C_f^o=100$ mM when subjected to variation of ambient oxygen O_2 levels, such as 1, 10 and 100 mmHg. These critical levels are chosen because hemoglobin-mediated oxygen transport in the membrane is only reactive from ambient oxygen O_2 level of 1 to 100 mmHg, as shown in Figs. 3. As observed in Fig. 4(a), the salt-induced volumetric behaviors of the membrane are divided into three stages: (i) when $C_{NaCl} < 0.1 C_f^o$, the initial increase of salt concentration C_{NaCl} provokes the ionization of the fixed charge group, leading to a collapse-to-swelling volumetric transitional behavior [23], (ii) when $0.1 C_f^o \leq C_{NaCl} < C_f^o$, the following increase of salt concentration C_{NaCl} causes the membrane to undergo a swelling-to-collapse transitional characteristics due to increase of screening effects by mobile ions on the fixed charge group, limiting the swelling capability of the membrane, and (iii) when $C_{NaCl} \geq C_f^o$, the membrane significantly rehydrates itself with the further increase of environmental salt concentration C_{NaCl} due to a large electrochemical potential difference between the membrane and its environmental solution which overwhelms the fixed charge group-mediated migration of ion transport, consequently driving the diffusion of environmental mobile ions down its concentration gradient into the membrane. The in-depth examination of Fig. 4 (a) reveals that the initial fixed charge density C_f^o of the polymeric system plays a pivotal role in its hydration-induced swelling deformation characteristics, which warrants further

investigation of initial fixed charge group effects on the membrane in the later part, as plotted from Figs. 5.

Fig. 4(b) visualizes the oxygen O_2 loading in the membrane as a function environmental salt concentration C_{NaCl} at different ambient oxygen O_2 levels. As shown in Fig. 4(b), it appears that a reciprocal relationship is observed between oxygen O_2 concentration and hydration-induced deformation of the membrane. For example, the collapse-to-swelling volume transition behaviors causes the decrease of oxygen O_2 concentration in the system, while the swelling-to-collapse volume transition characteristics leads to increase of oxygen O_2 concentration in the membrane. Furthermore, the increase ambient oxygen O_2 level enlarges oxygen-hemoglobin saturation which promotes the oxygen O_2 loading in the membrane. For example, when subjected to environmental salt concentration C_{NaCl} of 100 mM, the increase of ambient oxygen level O_2 from 1 to 10 mmHg or from 10 to 100 mmHg enlarges the oxygen O_2 concentration in the membrane by 3.70- or 1.34-fold, respectively. It is well-established that the oxygen-hemoglobin saturation increases bi-linearly with increase of ambient oxygen level O_2 [64], in which the linear hemoglobin saturation curve slope is relatively larger if the ambient level is increased from 1 to 10 mmHg as compared to the slope when ambient level is increased from 10 to 100 mmHg. This explains the greater increase of oxygen O_2 concentration in the membrane associated with the increment of lower ambient level, such as from 1 to 10 mmHg.

The counter-ion (*i.e.* sodium ion) and co-ion (*i.e.* chloride ion) loaded in the present hemoglobin-enriched polyacidic membrane is illustrated in Fig. 4(c) and Fig. 4(d), respectively, as a function of environmental salt concentration C_{NaCl} when subjected to variation of ambient oxygen O_2 levels. From Fig. 4(c), it is demonstrated that the counter-ion concentration increases

bi-linearly with increase of environmental salt concentration C_{NaCl} : (1) when $C_{NaCl} \leq C_f^o$ (*i.e.* 100mM) the initial increase of salt concentration C_{NaCl} promotes counter-ion migration, where the counter-ion concentration in the membrane is much greater than that of environmental solution and (2) when $C_{NaCl} > C_f^o$, the further increase of environmental salt concentration C_{NaCl} significantly weakens the Donnan potential strength of the membrane in which the dominant driving force for ionic transport in the system arises due to concentration gradient, almost diminishing the imbalance of counter-ion concentration between the membrane and its environmental solution. On the other hand, Fig. 4(d) shows that the co-ion concentration is tri-linearly enlarged by increasing environmental salt concentration C_{NaCl} : (1) when $C_{NaCl} \leq 0.01 C_f^o$ (*i.e.* 1mM), the co-ion concentration is apparently invariant with initial increase of salt concentration due to strong Donnan potential in the membrane, (ii) when $0.01 C_f^o < C_{NaCl} \leq C_f^o$, the subsequent increase of environmental salt concentration C_{NaCl} starts to weaken the Donnan potential strength in the membrane, however, its co-ion concentration is still much lesser than that of environmental solution and (iii) when $C_{NaCl} > C_f^o$, the Donnan potential strength in the membrane is depleted, in which the difference of counter-ion concentration between membrane and environmental solution is minuscule.

Fig. 4(e) inspects the Donnan potential of hemoglobin-loaded membrane as a function of environmental salt concentration, when subjected to variation of ambient oxygen levels. As visualized in Fig. 4(e), the Donnan potential strength of the membrane monotonously decreases with increase of salt concentration, where it approaches ~ 0 mV if $C_{NaCl} \geq C_f^o$. It is well established that Donnan potential measures capability of membrane to exclude co-ions into its

polymeric system, where the co/counter-ions concentration in the membrane is decreased/increased with stronger electrical potential strength in the membrane.

Fig. 4(f) investigates the surface conductivity of the membrane as a function of environmental salt concentration C_{NaCl} when subjected to variation of ambient oxygen O_2 levels.

Fig. 4(f) shows that the surface conductivity of the membrane is enhanced bi-linearly by

increasing the salt concentration C_{NaCl} , in which, when $C_{NaCl} \geq C_f^o$, the surface conductivity

begins to converge into a single surface conductivity value at ambient oxygen level of 1, 10 or

100 mmHg, given that the membrane is operating at a constant environmental salt concentration

C_{NaCl} . In addition, the surface conductivity of the membrane is almost independent of ambient

oxygen O_2 level, especially at two salt concentration C_{NaCl} regions, low ($C_{NaCl} \leq \sim 1$) and high ($C_{NaCl} \geq \sim 1000 \text{ mM}$) environmental salt concentrations. On the one hand, at high environmental

salt concentration, the dominantly diffusion-governed ion transport into the membrane cancels

the effects of increasing ambient oxygen O_2 level on surface conductivity of the membrane. On

the other hand, at low environmental salt concentration C_{NaCl} , the principally migration-

administered counter-ion transport into the membrane, due to relatively strong Donnan potential

strength, leads to the independence between ambient oxygen O_2 level and surface conductivity of

the membrane.

We now answer the question of how initial fixed charge group density C_f^o influences salt-

induced behaviors of hemoglobin-loaded polyelectrolyte membranes [65]. Fig. 5(a) examines the

impacts of initial fixed charge group density C_f^o on deformation behaviors of the membrane as a

function of environmental salt concentration C_{NaCl} . Fig. 5(a) demonstrates that the salt-induced

volumetric behaviors of the membrane are divided into three stages: (1) when $C_{NaCl} < 0.1 C_f^o$, the initial increment of environmental salt concentration C_{NaCl} promotes ionization of fixed charge group, resulting in the swelling of the membrane, where this observation agrees well with previously reported findings [23], (ii) when $0.1 C_f^o \leq C_{NaCl} \leq C_f^o$, the subsequent increment of salt concentration C_{NaCl} causes the enlargement fixed charge group screening by the mobile ions, making the membrane to exhibit a swelling-to-collapse behaviors and this qualitatively with previous experimental results [7, 15, 17] and (iii) when $C_{NaCl} > C_f^o$, the membrane significantly imbibes environmental solvent due to entrance of counter- and co-ions with the further increase of environmental salt concentration C_{NaCl} , where the mobile ions diffuse down its concentration gradient into the polymeric system. This observation is consistent with the findings reported by de la Cruz *et al.*, in which that polymeric membrane swelled when the environmental salt concentration is significantly increased [20].

Fig. 5(b) visualizes the impacts of initial fixed charge group density on oxygen O_2 loading in the membrane as a function environmental salt concentration C_{NaCl} . As shown in Figs. 5(a) and 5(b), an inverse relationship is observed between oxygen O_2 concentration in the membrane and hydration-induced deformation of the membrane, where the increase of swelling deformation of the membrane decreases oxygen O_2 concentration in the polymeric system. Furthermore, the increase initial fixed charge group density C_f^o enlarges swelling deformation of the membrane by diluting oxygen O_2 concentration in the membrane. For example, when subjected to environmental salt concentration C_{NaCl} of 100 mM, the increase of initial fixed charge group

density C_f^o from 10 to 100 mM or from 100 to 1000 mM enlarges the hydration of the membrane which decreases oxygen O_2 concentration in the membrane by 3.54 or 17.5 %, respectively.

The counter-ion (*i.e.* sodium ion) and co-ion (*i.e.* chloride ion) concentration in the present hemoglobin-enriched polyacidic membrane is illustrated in Fig. 5(c) and Fig. 5(d), respectively, as a function of environmental salt concentration C_{NaCl} , when operating at varying densities of initial fixed charge group C_f^o . As seen in Fig. 5(c), the imbalance of counter-ion concentration between the membrane and environmental solution diminishes with increasing environmental salt concentration C_{NaCl} , in which: (i) when $C_{NaCl} \leq C_f^o$, the initial increase of salt concentration C_{NaCl} decreases the ratio of counter-ion concentration in the membrane to its environmental solution due to the greater screening effects of the fixed charge group by mobile ions, weakening the Donnan inclusion of counter-ions into the membrane and (ii) when $C_{NaCl} > C_f^o$, the further increase of environmental salt concentration significantly weakens the Donnan potential strength in which the ratio of counter-ion concentration in the membrane to its environmental solution is ~ 1 . On the other hand, Fig. 5(d) shows that the co-ion concentration is generally enlarged by increasing environmental salt concentration. When $C_{NaCl} \leq 0.1 C_f^o$, the initial increase of salt concentration C_{NaCl} decreases the ratio of co-ion concentration in the membrane to environmental solution, in which the stronger Donnan potential causes co-ion concentration in the membrane to remain unchanged with the initial increase of salt concentration C_{NaCl} . When $C_{NaCl} > C_f^o$, the further increase of environmental salt concentration C_{NaCl} completely vanishes

Donnan potential in membrane, in which the ratio of co-ion concentration in the membrane to its environmental solution is ~ 1 .

Fig. 5(e) evaluates the Donnan potential of hemoglobin-loaded membrane as a function of environmental salt concentration C_{NaCl} for membrane loaded with different initial fixed charge densities C_f^o . As visualized in Fig. 5(e), the Donnan potential strength of the membrane monotonously decreases with increase of salt concentration C_{NaCl} , where this observation agrees qualitatively with previously published findings [66]. In addition, the Donnan potential strength approaches ~ 0 mV when $C_{NaCl} \geq 0.1C_f^o$. On the other hand, Fig. 5(f) visualizes the effects initial fixed charge density C_f^o on surface conductivity of the membrane as a function of environmental salt concentration C_{NaCl} . When operating at a constant environmental salt concentration C_{NaCl} , the increase of fixed charge group density from 10 to 100 mM enhances the migration of counter-ions into the membrane, resultantly improving the surface conductivity of the membrane. When environmental salt concentration C_{NaCl} is greater than 100 mM, the surface conductivity of the membrane appears to remain unchanged with increase of fixed charge group density C_f^o from 10 to 100 mM, due to weakened Donnan potential strength of the membrane in which ion transport is dominantly diffusion-governed, suggesting a strong dependence between surface conductivity of the membrane and salt-rich environmental solution.

We now examine the salt-induced volumetric behaviors of hemoglobin-loaded polyacidic, polybasic and polyampholytic membranes where the total initial fixed charge density C_f^o of respective membranes is set at 100 mM. As visualized in Fig. 6(a), the polyacidic or polybasic

experiences a collapse-to-swelling-collapse-swelling volumetric behaviors by increasing environmental salt concentration C_{NaCl} , where the swelling deformation of polyacidic or polybasic almost coincides with each other. It is established by English *et al.* that when operating at zero excess charges (Donnan potential strength of ~ 0 mV) in the membrane, the decrease of environmental salt concentration C_{NaCl} increases the electrical attraction between immobile acidic and basic charge groups in the polyampholytic membrane which collapses its polymeric system [57]. This observation is consistent with our present numerical result, such that when $C_{NaCl} < C_A^o / C_B^o$, the present polyampholytic membrane undergoes a swelling-to-collapse volumetric transitional behaviors by decreasing environmental salt concentration C_{NaCl} , and it remains at a collapse-state with further decrease of the environmental salt concentration C_{NaCl} due to maximum electrical attraction between acidic-basic charge group.

On the one hand, Fig. 6(b) visualizes the oxygen O_2 loading in hemoglobin-loaded polyacidic, polybasic and polyampholytic membranes as a function environmental salt concentration C_{NaCl} . As seen in Fig. 6(b), the oxygen O_2 concentration is decreased in the membrane by increasing environmental salt concentration C_{NaCl} , consistent with Figs. 3(b) and 4(b). As expected, the relatively smaller degree hydration of polyampholytic membrane enlarges the oxygen concentration in the polymeric system. On the other hand, Figs. 6(c) and 6(d) illustrate the sodium Na^+ and chloride Cl^- ion concentration, respectively, in the membranes as a function of environmental salt concentration C_{NaCl} . In general, sodium Na^+ and chloride Cl^- ion concentration enlarges bi-linearly with increase of environmental salt concentration, where the ratio of sodium/chloride-ion concentration in the polyampholytic membrane to environmental

solution is constantly ~ 1 . This is because when the membrane is operating at charge balanced conditions, where the Donnan potential strength of the polyampholytic membrane is extremely weakened to approximately ~ 0 mV, which causes the ion transport to be dominantly diffusion-governed. As such, it is imperative to investigate the Donnan potential strength to elucidate this assumption, as shown in Fig. 6(e).

Fig. 6(e) inspects the Donnan potential of hemoglobin-loaded polyacidic, polybasic and polyampholytic membranes as a function of environmental salt concentration C_{NaCl} when operating at pH neutral conditions. As visualized in Fig. 6(e), the Donnan electrical potential strength of polyacidic/basic membrane bi-linearly decreases with increase of environmental salt concentration C_{NaCl} , where the Donnan potential approaches ~ 0 mV if salt concentration $C_{NaCl} \geq$ initial fixed charge density C_f^o . On the other hand, for present polyampholytic membrane at neutral pH conditions, its Donnan potential is almost independent of environmental salt concentration C_{NaCl} . When the polyampholytic membrane is operating at environmental pH coinciding with its isoelectric point—average value of dissociation constants for acidic and basic charge groups, such as $(pK_a + pK_b)/2 =$ isoelectric point—the Donnan potential of the membrane remains a constant value of ~ 0 mV, as illustrated from Fig. 6(e), suggesting maximum electrical attraction between the fixed acidic-basic groups which causes the Donnan potential of the membrane to be unresponsive toward changes of environmental salt concentration C_{NaCl} .

Fig. 6(f) investigates the surface conductivity of hemoglobin-loaded polyacidic, polybasic and polyampholytic membranes as a function of environmental salt concentration C_{NaCl} . The surface conductivity pattern of the polyacidic and polybasic membrane almost coincide with

each other when operating at environmental salt concentration C_{NaCl} ranging from 0.1 to 1000 mM, as illustrated in Fig. 6(f). However, the polybasic membrane is observed to have greater surface conductivity level due to higher ion mobility associated with its counter-ion [67], whereas the surface conductivity of present polyampholytic membrane increases linearly by increasing environmental salt concentration C_{NaCl} . Since the Donnan potential of present polyampholytic membrane is insignificant when operating at pH neutral condition, as shown in Fig. 6(f), the increase of environmental salt concentration C_{NaCl} enlarges its concentration gradient between membrane-solution, promoting the ion transport in the system via diffusion-governed mechanism.

4. Conclusions

In this work, a multiphysics model is developed to investigate effects of salt- and oxygen-coupled stimuli on coupled chemo-electro-mechanical responses of hemoglobin-loaded polyelectrolyte membranes, accounting for the multi-physical interactions between immobile functional groups and environmental solution. The model includes a Poisson-Nernst-Planck (PNP) equation for the transport of mobile species acting over the membrane-solution interface, which is coupled with a nonlinear mechanical equation to describe the transformation of environmental biochemical cues into membrane electromechanical-based responses. For model validation, a direct comparison between published experimental observation and current numerical result is performed, in which a good agreement is achieved. As described in section 3, the numerical finding unveils that Donnan electrical potential strength of polyacidic/basic membrane decreases bi-linearly with increasing environmental salt concentration C_{NaCl} where the Donnan potential approaches ~ 0 mV if salt concentration $C_{NaCl} \geq$ initial fixed charge

density C_f^o , while for present polyampholytic membrane at neutral pH conditions, its Donnan potential is independent of environmental salt concentration. The result also shows that the decrease of Donnan potential strength promotes ionic transport over the membrane-solution interface, improving the surface conductivity of the polymeric system.

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Table. 1. Input data for the numerical simulation of the multiphysics model.

Parameter	Value
Diffusion coefficients D_{H^+} [68]	$9.3 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$
Diffusion coefficients D_{OH^-} [68]	$5.2 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$
Diffusion coefficients D_{Na^+} [68]	$1.3 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$
Diffusion coefficients D_{Cl^-} [68]	$2.0 \times 10^{-9} \text{ m}^2 \text{ s}^{-1}$
Flory interaction parameter χ for pNIPAM [59]	0.906
Permittivity ϵ_r, ϵ_0 (water)	$7.1 \times 10^{-10} \text{ A s V}^{-1} \text{ m}^{-1}$
Forward rate k_{OxyHb} ; Backward rate k_{Oxy} [69]	$3.3 \times 10^7 \text{ M}^{-1} \text{ s}^{-1}$; 50 s^{-1}
Association constant: K_1 ; K_2 [41]	$\frac{1}{10^{-7.58}}$; $\frac{1}{10^{-7.72}}$
Ion radius a [53]	$3 \text{ \AA}$
Solubility of oxygen O_2 α	$1.34 \mu\text{M/mm Hg}$

Fig. 1. Comparison of membrane swelling deformation as a function of environmental salt concentration via present numerical simulation and published experimental observation for: (a) polybasic [17, 56] and (b) polyampholytic membranes [57].

Fig. 2. Comparison of membrane surface conductivity as a function of environmental salt concentration via present numerical simulation and published experimental observation for AMX and CMX charged polymeric system [9].

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Fig. 3. The (a) swelling deformation, (b) oxygen loading, (c) counter- concentration, (d) co-ion concentration, (e) electrical potential and (f) surface conductivity of the hemoglobin-loaded polyacidic membrane with initial fixed charge density of 100 mM as a function of ambient oxygen level, when operating at environmental pH of 7.40 and temperature of 37 °C.

Fig. 4. The (a) swelling deformation, (b) oxygen loading, (c) counter- concentration, (d) co-ion concentration, (e) electrical potential and (f) surface conductivity of the hemoglobin-loaded polyacidic membrane with initial fixed charge density of 100 mM as a function of environmental salt concentration, when operating at environmental pH of 7.40 and temperature of 37 °C.

Fig. 5. The effect of initial fixed charge density on (a) swelling deformation, (b) oxygen loading, (c) counter- concentration, (d) co-ion concentration, (e) electrical potential and (f) surface conductivity of the hemoglobin-loaded polyacidic membrane with initial fixed charge density of 100 mM as a function of environmental salt concentration, when operating at environmental pH of 7.40 and temperature of 37 °C.

Fig. 6. The (a) swelling deformation, (b) oxygen loading, (c) counter- concentration, (d) co-ion concentration, (e) electrical potential and (f) surface conductivity of the hemoglobin-loaded polyacrylic, polybasic and polyampholytic membranes with initial fixed charge density of 100 mM as a function of environmental salt concentration, when operating at environmental pH of 7.40 and temperature of 37 °C.