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# Effective schemes to control the dynamic behavior of the water transport in the membrane of PEM fuel cell

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**Abstract**

We have investigated the transient behavior of the water transport across the membrane of the PEM fuel cell to seek for effective control schemes so that the best dynamic performance of the fuel cell can be obtained. It is found that both a larger starting operational current density $i_0$ and a smaller operational current density $i$ can lead to a smaller dynamic response time $t_{ss}$, the time for the water distribution across the membrane to reach the steady state. Present results nevertheless point out that the most powerful as well as the most feasible control scheme is to control the humidification parameter $k$, i.e. to adjust the water content of the feeding fuel, so that the $t_{ss}$ would remain steadily in a reasonably low value in a wide range of water flux fraction $\beta$, another control parameter of the membrane. The present conclusion can be useful for the design of the PEM fuel cell when its application on the dynamic mobile system is concerned.


*Keywords:* PEM fuel cell; Membrane; Water management; Transient behavior; Dynamic response time

## 1. Introduction

Among various designated applications of PEM fuel cell, the power system of mobile vehicles is one of the most important objects being rooted in the hearts of fuel cell researchers [1,2]. In such a mobile power system, the transient behavior and the corresponding characteristic times play various crucial roles in determining the dynamic performance of the fuel cell. For example, the time interval from the start of the operation to the steady state of the fuel cell is one of the most important characteristic times, which depends largely on the many dynamic response time constants of each component of the fuel cell, among them is the dynamic response time of the water transportation of the membrane. To examine the dynamics response of the whole fuel cell, the transient behavior of each component shall be investigated [3] and then the dynamics characteristics of each component shall be integrated and considered as a whole according to the interaction between the components. To seek this dynamic response time and other potential characteristics times, we start from the unsteady water transportation of the PEM fuel, which in fact had become a crucial issue in the fuel cell design in the last decade because the electric conductivity depends significantly on the water content in the membrane [4–8].

In the membrane of the PEM fuel cell, see Fig. 1, the water is generated at the cathode and transported through the membrane to the anode by diffusion effect. On the other hand, the water is also transported from the anode to cathode due to the electro-osmosis effect. When electro-osmosis effect is

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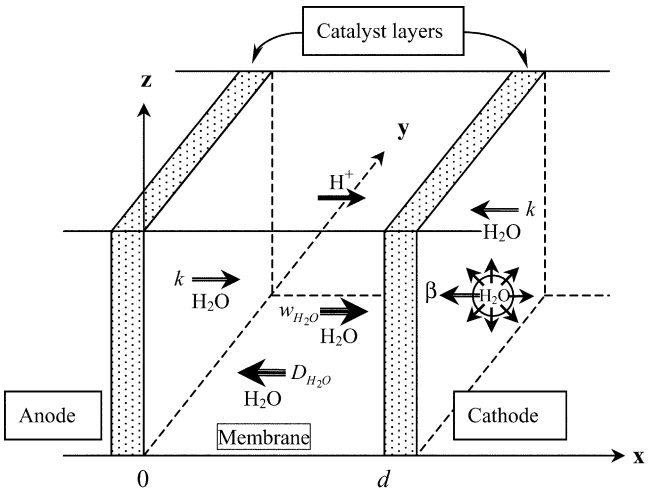


Fig. 1. A schematic description of the physical domain considered.

more significant than diffusion effect, the membrane at the anode side will be dehydrated; when the opposite occurs, the membrane at the cathode side will be over flooding. Both of these cases lead to enhance the Ohmic loss accounting for a large part of the voltage loss of the fuel cell [9]. To consider the water management of the fuel cell, many of the studies investigated the water transport of the two-phase flow system in which relevant heat transfer problems were also taken into account. Some studies considered the physical domain including the gas diffusion layer, the catalyst layer and the membrane [10–15], while some focused on the membrane [16,17]. A common conclusion from these studies was that the operational current density, the humidification parameter and the membrane thickness had a significant influence on the water transportation in the fuel cell. Recently, Okada [18,19] pointed out that the water transportation is also influenced significantly by the presence of the contaminants, and that at the cathode is more serious than that at anode.

In most of previous studies mentioned above, only the steady state water distributions were investigated. In the present paper, we investigate the transient behavior of the water transportation across the membrane to examine the dynamic response time of the water transportation of the membrane and, based on the present results, we propose effective schemes to control the dynamic response time at a reasonably low value under various operational situations. The paper is organized as follows. In Section 2 the mathematical model and relevant initial and boundary conditions regarding the unsteady water transport across the membrane is presented. In Section 3 the effects of the two control physical parameters on the unsteady features of the water transport are discussed, special attention is paid to the time needed for the water distribution to reach the steady state. In Section 4, the application of the present results in terms of practical feasibility is discussed so that the effective control schemes to shorten the dynamic response time can be proposed. And, finally, in Section 5 concluding remarks are drawn.

## 2. Theoretical model of unsteady water transport in membrane

Consider a membrane of PEM fuel cell of thickness $d$ (Fig. 1). We investigate the unsteady water transport across the membrane. We assume that the interfaces between the membrane and the two electrodes are flat plane and all the physical properties are uniform on the plane [16]. The volume of membrane is assumed to be not changing with the variation of hydration, so that the thickness $d$ remains constant. The reactions in electrodes are assumed to be much faster than the water transport in the membrane. Since the combination of the diffusion and the electro-osmosis effects accounts for the flux of water, namely,

$$J = -D\frac{\partial c}{\partial x} + \frac{i}{F}\mathrm{w}, \tag{1}$$

the water transport equation in the membrane can be written as

$$\frac{\partial c}{\partial t} = -\frac{\partial J}{\partial x} = \frac{\partial}{\partial x}\left(D\frac{\partial c}{\partial x} - \frac{i}{F}\mathrm{w}\right), \tag{2}$$

in which $J$ is the flux of water across the membrane, $D$ is the diffusion coefficient of the water in the membrane, $c$ is the water concentration in the membrane, $i$ is the operational current density across the membrane, and $F$ is the Faraday constant.

To examine the unsteady features of the water transport in the membrane, we assume initially the water concentration is,

$$c(x, 0) = c_0(x) \quad (0 < x < \mathrm{d}t = 0) \tag{3}$$

in which $c_0(x)$ is the water concentration corresponding to the initial current density $i_0$, when $i_0 = 0$ the initial water concentration is uniform, i.e. $c_0(x) = c_0$. Besides, there are two boundaries in the physical domain shown in Fig. 1. On the left, the interface between the anode and the membrane ($x = 0$), the continuity of water flux shall be satisfied; namely,

$$\frac{\beta_1 i}{F} + k[c_\mathrm{a} - c(0, t)] = -D\frac{\partial c(0, t)}{\partial x} + \frac{\mathrm{w}i}{F}, \tag{4}$$

in which $k$ is the rate constant of Henry's law accounting for the water flux entering into or getting out of the membrane driven by the water concentration gradient in the membrane, or called the humidification parameter [18], $c_\mathrm{a}$ is the water concentration of the membrane at $x = 0$ being equilibrium with the saturated water vapor of the anode gas, and $\beta_1$ is the coefficient accounting for the ratio between the water entering the membrane to the water generated at the cathode [19], or called the water flux fraction. On the right, the interface between the membrane and the cathode ($x = d$), the water flux across the membrane is also conserved, namely,

$$\frac{\beta_2 i}{2F} + k[c_\mathrm{c} - c(d, t)] = D\frac{\partial c(d, t)}{\partial x} - \frac{\mathrm{w}i}{F}. \tag{5}$$

Eq. (5) differs from Eq. (4) by the first term on the left hand side, which accounts for the amount of water generated at the cathode and enters the membrane. In Eq. (5) $c_c$ is the water concentration of the membrane at the interface $x = d$ being equilibrium with the saturated water vapor of the cathode gas, and we assume $\beta_2 = \beta_1$. This condition is justified because during the operation of fuel cell the water is constantly generated from the reaction at the cathode so that the water concentration at the cathode can be maintained constant.

In the present paper, we consider the membrane made of Nafion 117. The water diffusion coefficient is $D = 1.25 \times 10^{-5}\,\mathrm{cm^2\,s^{-1}}$ (at $\lambda = 14$, $T = 80\,^\circ\mathrm{C}$) [20]. The water concentration in the membrane can be expressed by $c = \lambda d_{\mathrm{dry}}/MV_{\mathrm{ex}}$, where $M$ is the molar weight of membrane taken as $1100\,g\,\mathrm{mol^{-1}}$, $V_{\mathrm{ex}}$ is the volume expansion rate of the membrane from dry to wet and is taken as 1.62, and $d_{\mathrm{dry}}$ is the density of the dry membrane and is taken to be $2.02\,\mathrm{g\,cm^{-3}}$ [21]. By assuming that the initial water concentration $c_0$ is equilibrium with the water vapor of the membrane, or the vapor entering into the cathode is saturated, one shall have $\lambda = 14$ [22,23]. There are some other routine parameters used in the present analyses but not shown here. For the details of their values the reader is referred to the Table 1 of Chen et al. [3].

Note please that in the following analyses special attention will be paid to the dynamic response time, or $t_{\mathrm{ss}}$, which is defined as the time for the water distribution across the membrane to reach the steady state from the initial distribution. Mathematically, we define that the steady state is reached when the following relation is satisfied

$$\mathrm{abs}\left\{\mathrm{Max}\frac{[y_{n+1}(i,j) - y_n(i,j)]}{y_{n+1}(i,j)}\right\} < 10^{-4} \tag{6}$$

in which $y_n\,(i, j)$ is any physical variable at the $n$th time step on node $(i, j)$. In the present paper, the water concentration is taken as the physical variable. Fig. 2 illustrates a typical example to show the transient variation of the water distribution across the membrane while, for more easily showing the transient behavior, the water concentration at the cathode is fixed to be the initial level $C_0$. It is seen that the water concentration decreases with time in the whole membrane, and due to the electro-osmosis effect the water depletion at the anode becomes more intense as time goes on. The steady state is reached at about $t_{\mathrm{ss}} = 9.78\,\mathrm{s}$, at which the electro-osmosis effect dragging water from anode to cathode is balanced by the diffusion effect pumping water from cathode to anode.

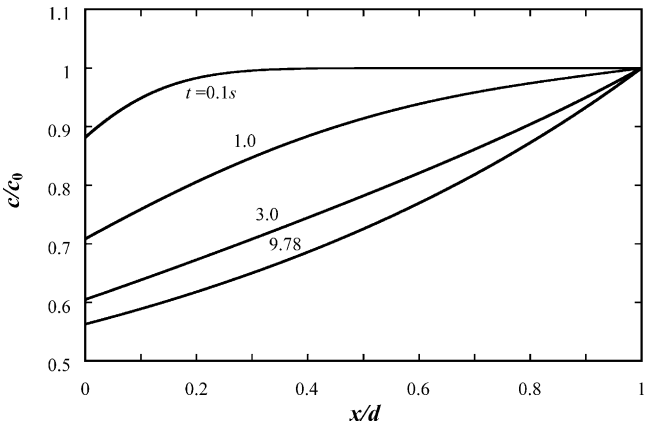


Fig. 2. The water distribution across the membrane at different times: an example of the change of the water distribution with time, starting from a uniform distribution $c = C_0$ to the steady state occurring at $t = 9.78\,\mathrm{s}$. The analysis is carried out under the condition that $i_0 = 0.0\,\mathrm{A\,cm^{-2}}$, $T = 80\,^\circ\mathrm{C}$, $p = 1\,\mathrm{atm}$, $d = 100\,\mu\mathrm{m}$, and $k = 0.001\,\mathrm{cm\,s^{-1}}$.

## 3. The effects of the two control parameters

As stated previously, during the fuel cell operation, there are two mechanisms driving the transportation of the water across the membrane. One is the electro-osmosis effect dragging water from anode to cathode and the other is the diffusion effect pumping water from cathode to anode. Usually, the water depletion at the anode is more serious than that at the cathode because the electro-osmosis effect is usually higher than the diffusion effect. But this may be changed when the two physical parameters $\beta$ and $k$ vary in a way to fit the desirable water balance control. In the present paper, based on the theoretical model and the boundary and initial conditions shown previously, we seek for the effective scheme to control these two parameters so that the water balance can be reached in a shortest time interval. By examining the transient behavior of the water concentration across the membrane under various operational current densities $i$, we investigate the effects of these two control parameters on the variation of the so-called dynamic response time $t_{\mathrm{ss}}$. A small $t_{\mathrm{ss}}$ is one of the few prerequisites of the modern mobile system requiring a high power performance.

Physically, according to Eqs. (4) and (5), the humidification parameter $k$ accounts for the capability of the water concentration of the membrane to balance with the saturated water vapor of the fuel channels at either the anode or cathode. Namely, a larger $k$ means the water balance between the membrane and the fuel channel can be achieved in a shorter time, and vice versa. Similarly, according to Eq. (5), the water flux fraction $\beta$ accounts for the portion of the water generated at the cathode can enter the membrane. A larger $\beta$ accordingly means that more water will enter into the membrane from the cathode during operation. By changing these two parameters, the diffusion effect of the membrane will also change so that the water balance in the membrane is changed under the desired way.

In the following analyses, we consider the base case $T = 80\,^\circ\mathrm{C}$, $p = 1\,\mathrm{atm}$, $d = 100\,\mu\mathrm{m}$, $i_0 = 0\,\mathrm{A\,cm^{-2}}$, $i_0 = 1\,\mathrm{A\,cm^{-2}}$, $\beta = 0$ and $k = 0.001\,\mathrm{cm\,s^{-1}}$. As one considers the effect of one parameter, the values of the other parameters are fixed as above. We first examine the effect of the control parameter $\beta$ on the dynamic response time $t_{\mathrm{ss}}$ and a typical result in terms of the variation of $t_{\mathrm{ss}}$ with respect to $\beta$ under various $i$ is shown in Fig. 3. For a large operational current

density $i = 1\,\mathrm{A\,cm^{-2}}$, the need of water in the membrane to carry protons is large. So that when the water flux from the cathode is small, i.e. a case of small $\beta$, the dehydration in the membrane becomes significant so that the time for the water in the membrane to reach the steady state is longer, i.e. a larger $t_{ss}$ is resulted. On the other hand, when $\beta$ is large, the water added from the cathode is large so that the flooding problem in the membrane becomes serious, leading again to another postpone of the water in the membrane to reach the steady state, i.e. a larger $t_{ss}$ is required. Consequently, for such a case of large $i$, there leaves a small range of $\beta$, or $0.35 < \beta < 0.62$, in which the $t_{ss}$ remains at approximately 4 s. In such a region, the water concentration across the membrane reaches to a steady state within a reasonably short period of time. This is the most desirable range of $\beta$ which the operational condition of fuel cell shall fit in. For convenience of the following discussion, we will call this range of $\beta$ the short time range, or STR. And the range of $\beta$ smaller than STR is called the dehydration range (DR) and that larger than STR is called the flooding range (FR). In both DR and FR, the $t_{ss}$ increases monotonically with either decreasing or increasing $\beta$, respectively, and generally has a larger value than that of STR.

For a smaller operational current density $i = 0.75\,\mathrm{A\,cm^{-2}}$, as also shown in Fig. 3, due to the smaller amount of water generated in the membrane under a lower $i$, the dehydration of the membrane is appeased so that a smaller water flux added from the cathode is sufficient for the water of the membrane to reach the steady state in a reasonably shorter time. Accordingly, the DR disappears while both the STR and the FR are enlarged and the STR is shifted to the lower side of $\beta$, i.e. $0.19 < \beta < 0.52$. As the operational current density is lowered further to $i = 0.5\,\mathrm{A\,cm^{-2}}$, the STR is enlarged to $0 \leq \beta < 0.62$ while the FR is reduced further. Similar results are obtained for an even lower $i$, see for example the case of $i = 0.25\,\mathrm{A\,cm^{-2}}$, the STR remains approximately the same with the previous case of $i = 0.5\,\mathrm{A\,cm^{-2}}$ while the flooding problem becomes less serious.

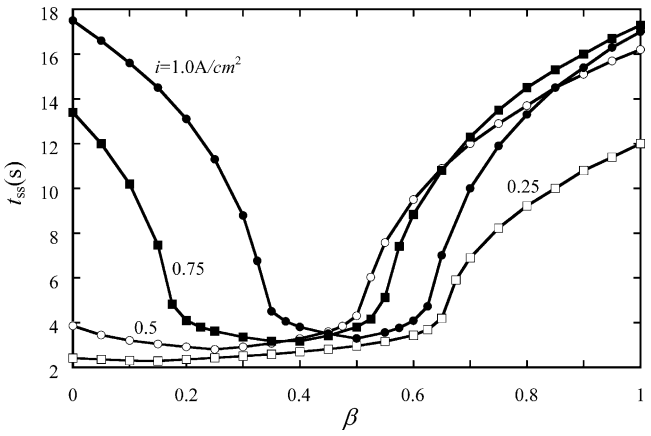


Fig. 3. The variations of $t_{ss}$ with $\beta$ for various current density $i$ under the condition that $i_0 = 0.0\,\mathrm{A\,cm^{-2}}$, $T = 80\,^{\circ}\mathrm{C}$, $p = 1\,\mathrm{atm}$, $d = 100\,\mu\mathrm{m}$, and $k = 0.001\,\mathrm{cm\,s^{-1}}$.

We selected three values of $\beta$, i.e. $\beta = 0$, 0.45 and 1.0, to examine the steady state distribution of the water concentration across the membrane under four different operational current densities, and the results are shown in respectively Fig. 4(a)–(c). It is seen that for all the cases considered, the water concentration increases monotonically from the anode to cathode, confirming that the dehydration usually occurs at the anode side while the flooding occurs at the cathode side. This is obviously due to the predominance of the osmosis effect over the diffusion effect so that the water dragged from the anode to cathode is more than the water diffused from the cathode to anode, and which becomes more obvious when a larger operational current density is applied. It is also seen that, for a larger $\beta$, the water entering from the cathode increases so that the water concentration at the cathode side increases, which also leads to an increase of the overall water concentration across the membrane.

The humidification parameter $k$ is another important control parameter to the dynamic response characteristic of the

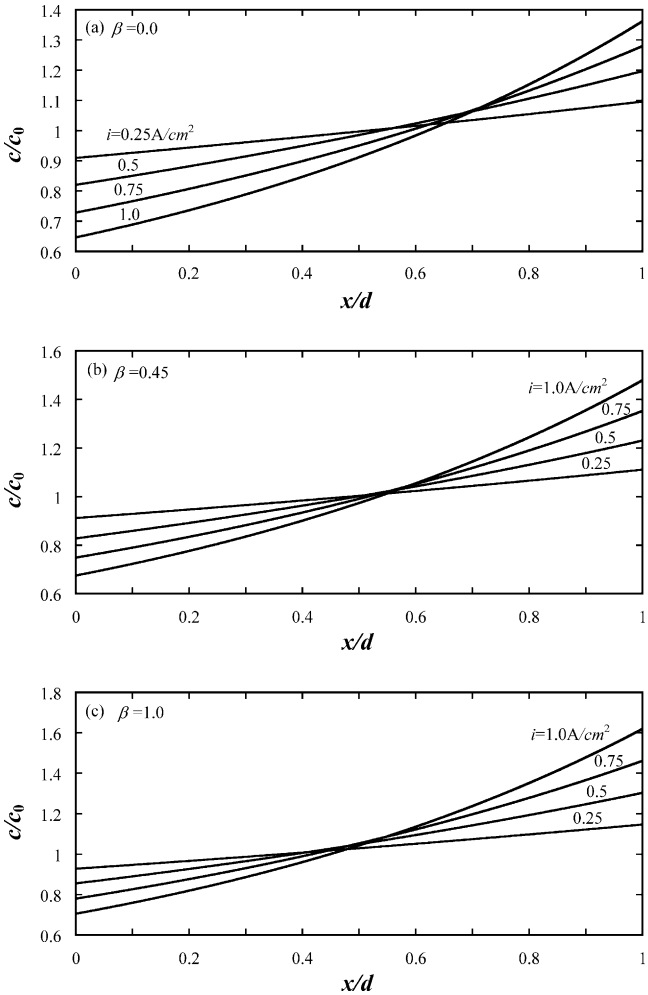


Fig. 4. The distributions of water concentration across the membrane for various current density $i$ under the condition that $i_0 = 0.0\,\mathrm{A\,cm^{-2}}$, $T = 80\,^{\circ}\mathrm{C}$, $p = 1\,\mathrm{atm}$, $d = 100\,\mu\mathrm{m}$, and $k = 0.001\,\mathrm{cm\,s^{-1}}$. (a) $\beta = 0$; (b) $\beta = 0.45$ and (c) $\beta = 1$.

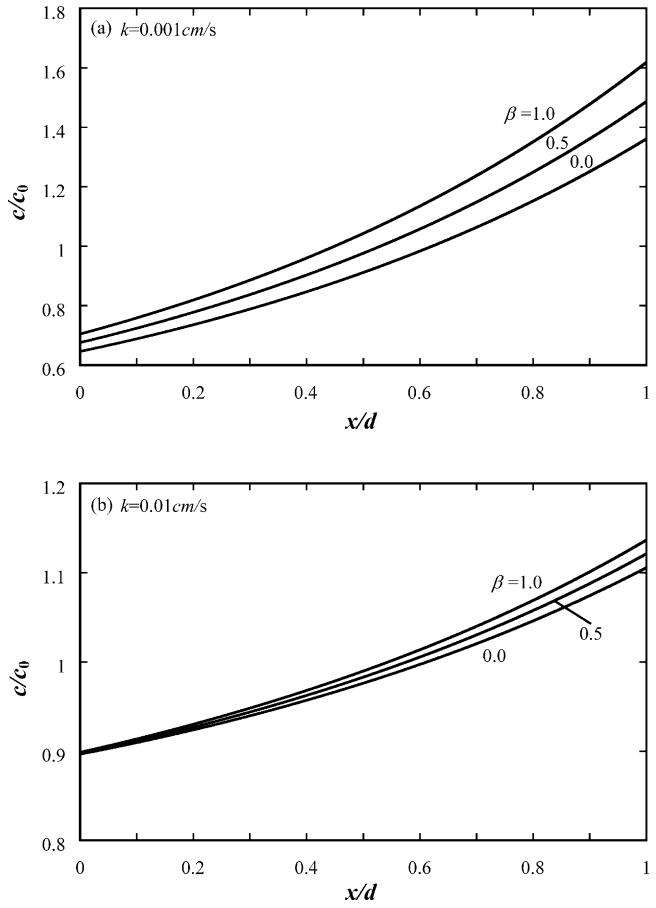


Fig. 5. The distributions of water concentration across the membrane under a operational current density $i = 1.0\ \mathrm{A\,cm^{-2}}$ for two different $k$; (a) $k = 0.001\ \mathrm{cm\,s^{-1}}$ and (b) $k = 0.01\ \mathrm{cm\,s^{-1}}$. The results are calculated under the conditions $T = 80\ ^\circ\mathrm{C}$, $p = 1$ atm, and $d = 100\ \mu\mathrm{m}$.

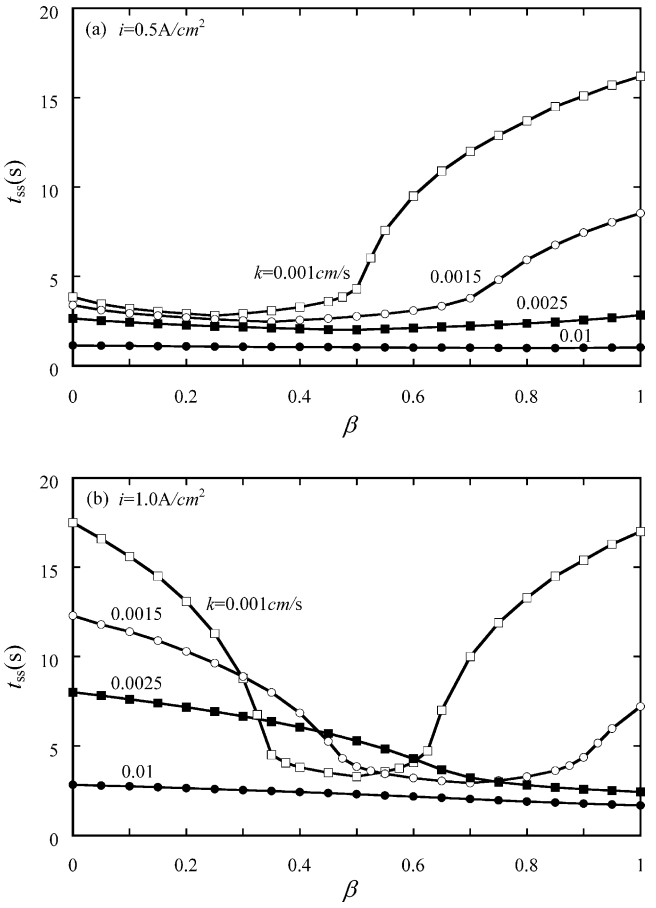


Fig. 6. The variations of $t_{ss}$ with $\beta$ for two different operational current densities (a) $i = 0.5\ \mathrm{A\,cm^{-2}}$; (b) $i = 1.0\ \mathrm{A\,cm^{-2}}$. The results are calculated under the conditions $T = 80\ ^\circ\mathrm{C}$, $p = 1$ atm, and $k = 0.001\ \mathrm{cm\,s^{-1}}$.

water concentration across the membrane. To illustrate this, we show in Fig. 5 the steady state water concentration distribution across the membrane of two different $k$, and for each $k$ we consider three different $\beta$. For $k = 0.001\ \mathrm{cm\,s^{-1}}$ (Fig. 5(a)), the water concentration increases monotonically from anode to cathode and a larger $\beta$ results in an overall increase of the water concentration across the membrane because more water enters into the membrane from the cathode. As $k$ increases by 10 times to $0.001\ \mathrm{cm\,s^{-1}}$ (Fig. 5(b)), the influence of $\beta$ remains, the water concentration at the cathode decreases and that at the anode increases, while the overall water concentration across the membrane is lowered. This is because a larger $k$ enhances the water penetration (or removal) across the anode/membrane or the cathode/membrane interface, so that at the cathode more water diffuses into the fuel channel due to both a larger water concentration difference between the membrane and the channel and a larger $k$ accounting for a greater diffusion capability of the membrane. At the anode, on the other hand, a larger $k$ diffuses more water of the membrane towards the anode and therefore lifts up the water concentration level of this dehydration region.

The results of Fig. 5 indicate that a larger $k$ means that water can more easily leave from or enter into the membrane through diffusion so that the water balance can be more easily reached within a shorter time. This scenario is also confirmed by the results shown in Fig. 6, in which the effect of the humidification parameter $k$ on the variation of $t_{ss}$ is illustrated. For a lower operational current density $i = 0.5\ \mathrm{A\,cm^{-2}}$ (Fig. 6(a)), a larger $k$ results in a smaller $t_{ss}$ for all the $\beta$ considered because the diffusion of water between the membrane and the fuel channels at both the cathode and anode sides predominates over the pumping of the water entering from the cathode. For the case $k = 0.001\ \mathrm{cm\,s^{-1}}$, the STR is about $0 \le \beta < 0.5$ within which the $t_{ss}$ remains approximately at 4 s and the FR is about $\beta \ge 0.5$ in which the $t_{ss}$ increases with $\beta$. As $k$ increases to say $0.0015\ \mathrm{cm\,s^{-1}}$, the STR is enlarged to $0 \le \beta < 0.7$ and the $t_{ss}$ is lowered slightly, the range of FR is reduced and the $t_{ss}$ is lowered more significantly. As $k$ increases further to $0.0025\ \mathrm{cm\,s^{-1}}$ and $0.01\ \mathrm{cm\,s^{-1}}$, the FR disappears and the STR covers the whole range of $\beta$, the $t_{ss}$ is lowered to respectively 2.5 and 1.5 s.

For a larger operational current density $i = 1.0\ \mathrm{A\,cm^{-2}}$ (Fig. 6(b)), the dehydration at the anode and the flooding at the cathode become more serious so that for a case of small

$k$, say $k = 0.001\ \mathrm{cm\,s^{-1}}$, the STR is small and the DR and FR are predominant. As $k$ increases, say $k = 0.0015\ \mathrm{cm\,s^{-1}}$, the FR disappears and the DR and STR share evenly the whole range of $\beta$, suggesting that the flooding at the cathode side is improved significantly by the increase of $k$, i.e. more water is diffused from the membrane to the fuel channel at the cathode, while the improvement at the anode due to the increase of water diffusion is less significant. As $k$ increases further to say 0.0025 and $0.01\ \mathrm{cm\,s^{-1}}$, this phenomenon becomes more obvious and the $t_{ss}$ is reduced further to a lower value.

## 4. The control schemes

Above results show that there exists a range of $\beta$, or STR, in which the $t_{ss}$ remains at a reasonably low value under various operational conditions, and, more importantly, this $t_{ss}$ can be lowered either by increasing $k$ or by decreasing $i$. This outcome is superior in terms of the feasibility of an effective control scheme to apply on the dynamic fuel cell system. Although all the three parameters $i$, $k$ and $\beta$ influence the range of STR and the value of $t_{ss}$, but in real situations the operational current density $i$ is usually set to be one of the characteristics parameters in the specification, and in most of the situations a higher operational current density is desired for a more powerful motor system. So that, it is rare that $i$ is employed as a control parameter in real situations. But, nevertheless, if the use of $i$ as a control parameter is considered, according to the present results, a lower $i$ is always desirable since it corresponds to a smaller $t_{ss}$. A similar but opposite conclusion also applies for the starting operational current density $i_0$ that a larger $i_0$ corresponds to a smaller $t_{ss}$ because a larger $i_0$ is closer to the $i$ at operation [3], while in practical sense it cannot be used as a control parameter either.

The water flux fraction $\beta$ is an intrinsic parameter of the membrane and cannot be changed, according to the extrinsic control conditions, so that in the present study we take $\beta$ as an index of goal. Namely, under a prescribed operational condition of designated $i$ and $i_0$, we control the value of other parameter, such as $k$ so that a lowest stable value of $t_{ss}$ can be available in a larger range of $\beta$, or a larger STR. A larger STR means a larger range of choice of the membrane to be used in the fuel cell, a more convenient condition for the researchers in the design of fuel cell dynamic systems.

The humidification parameter $k$, on the other hand, can be a useful parameter to control the dynamic response time. Although the value of $k$ is also an intrinsic property of the membrane, but, nevertheless, as shown in Eqs. (4) and (5), it can to some extent account for the amount of water entering or leaving the membrane, which in fact depends not only on the value of $k$, but also on the difference between the water concentration of the membrane and that of the fuel channel. Accordingly, by changing this difference of water concentration can account for the effect of changing $k$. According to the present results, a larger $k$ gives a smaller $t_{ss}$ in a larger range of $\beta$ (a larger STR), being a most desirable situation for a powerful fuel cell system of fast-response dynamic characteristics. This can be resulted by humidifying the fuel gas at the anode side and dehumidifying the fuel gas at the cathode side. Both these schemes enhance the concentration difference between the fuel channel and the membrane, leading to a better dynamic performance in terms of a smaller $t_{ss}$.

## 5. Concluding remarks

We have investigated the unsteady water transport in the membrane of the PEM fuel cell to seek for effective schemes with which the water transport in the membrane can reach a steady state in a reasonable short time under a wide range of control condition, or more specifically a smallest $t_{ss}$ in a widest range of a control parameter such as $\beta$. It is found that there are essentially four parameters relevant to the unsteady features of the water transport of the membrane, they are the starting operational current density $i_0$, the operational current density $i$, the water flux fraction $\beta$ at the cathode, and the humidification parameter $k$ at both the anode and cathode. The first two parameters $i_0$ and $i$ are used to be taken as the specification data and cannot be used as the control parameters during the fuel cell operation. But, if one needs to use them to control the dynamic system, then both a larger $i_0$ and a smaller $i$ can result in a smaller $t_{ss}$. Otherwise, present results indicate that the parameter $k$ is the most powerful and useful control parameter that a larger $k$ lead to a smaller $t_{ss}$ in a wider range of $\beta$, where $\beta$ is used as an index of goal instead of a control parameter. The change of $k$ can be accounted for by changing the water concentration difference between the membrane and the fuel channel at either the cathode side or the anode side. More precisely in practical sense, both the humidification in the fuel gas of the anode and the dehumidification in the fuel gas of the cathode can lead to a smaller $t_{ss}$ in a larger range of $\beta$, which represents a better dynamic performance of the fuel cell as well as a wider choice of the membrane. In brief, as indicated previously by Wood et al. [9] as well as by the present results, the humidification at the anode not only can improve the fuel cell performance, but also is an important scheme to control the dynamic behavior of the water transportation in the membrane. Namely, under appropriate humidification schemes, the water balance in the membrane can be reached in a reasonably short time, which is an important feature for the fuel cell to have a better dynamic performance.

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