

Chapter 2

Literature Review

2.1 Introduction

The fuel cell performance is studied both experimentally and computationally in literature. The latter, in particular, can provide detailed insights into the inherent limiting mechanisms, transport phenomena, structural integrity, and electrochemistry on a local level throughout the cell. Moreover, mathematical modelling is an efficient approach for parametric studies, compared with repetitive and costly experimental techniques. This chapter conducts a literature review on conventional fuel cell modelling in view of five aspects: mathematical modelling of single cells (demonstrated with the P-SOFC), stack modelling, model simplification, numerical methods for computational simulation, and sensitivity analysis based on numerical simulation.

2.2 Single-Cell Modelling

A general target of fuel cell modelling is to calculate the cell polarization curve under various operating and design conditions, because the relationship between cell voltage and current density drawn from the cell constitutes the primary measure of a fuel cell performance [1]. Then three irreversible voltage losses, called the activation, ohmic, and concentration polarization, can be determined based on the conservation equations for the transport of total mass, each species, momentum, charges, and energy. The working fluid may be either in single gas phase (e.g., in the SOFC) or in two-phase (e.g., liquid water and gaseous flow in the PEMFC). According to objectives of studies, multi-dimensional models are developed in three-dimensional (3D), two-dimensional (2D), one-dimensional (1D), or zero-dimensional (0D) with certain simplifications and assumptions. The models can be assumed to be under a steady-state or transient condition according to the

research requirement [2]. When developing a model from the beginning or assessing the average cell performance, the steady-state condition can be selected for simplicity.

For example, a 1D model based on basic principles of gas phase transport was carried out by Bernardi to optimize boundary conditions in solid-polymer-electrolyte fuel cells so as to avoid flooding and dehydration of the membrane [3]. Weber and Newman presented a 1D non-isothermal single-cell model coupling both heat and mass transfer and taking the effects of ohmic loss and heat generation due to irreversible reactions into account [4]. A 2D model with various designs was proposed by Nguyen and White to control the humidification and limit its influence [5]. Yuan et al. conducted simulations for two-phase flow and heat transfer in 3D duct of the PEMFC [6]. For the SOFC, the first significant solid electrolyte fuel cell models were presented almost 30 years ago. In 1983, De Benedetti and Vayenas developed a mathematical model for the solid electrolyte cell as an electrochemical reactor in order to describe the cell performance under steady-state conditions [7]. Two years later, Vayenas et al. [8] developed a model with a cross-flow monolithic structure. After that, the works on SOFC modelling emerged increasingly in the literature. Nowadays, the modelling development of the SOFC has covered all the cell designs [9–12].

To demonstrate the mathematical models of planar fuel cells, Sects. 2.2.1–2.2.3 review the research works on the P-SOFC modelling regarding the polarizations, transport phenomena in electrodes, and spatial dimension. For other types of planar fuel cells (e.g., PEMFC, AFC, PAFC, MCFC), detailed modelling can be referred to the literature, such as Refs. [13–15].

2.2.1 Polarizations

A typical polarization curve for the fuel cell mainly consists of three regions for the activation, ohmic, and concentration polarizations [14], as shown schematically in Fig. 2.1. The first region represents the activation voltage drop called the activation polarization, η^{act} , due to the slowness of the chemical reactions taking place at electrode surfaces. Depending on the operating temperature and pressure as well as electrode structure, this region is more or less wide. The second region represents the resistive losses called the ohmic polarization, η^{ohm} , due to the internal resistance to the transport of electrons and ions in the solid structure of the cell. The last region represents the mass transport losses called the concentration polarization, η^{con} , resulting from the change in concentration of reactants as the reactants are consumed. In addition, there is another voltage drop called the leakage polarization, η^{leak} , that makes the actual measured open circuit voltage, E^{OCV} , often fall slightly below the reversible potential, E^{rev} [2, 14]. The real output voltage of the cell, V_{cell} , is then obtained by subtracting all the polarizations from the reversible potential, given by [1, 2, 14, 16]:

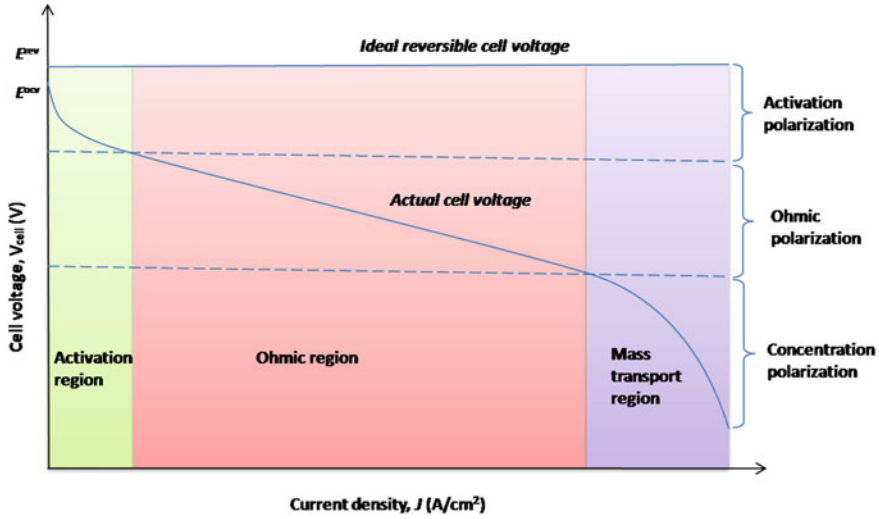


Fig. 2.1 Schematic of the polarization curve for a fuel cell

$$V_{\text{cell}} = E^{\text{rev}} - \eta^{\text{act}} - \eta^{\text{ohm}} - \eta^{\text{con}} - \eta^{\text{leak}}. \quad (2.1)$$

The reversible potential and the four kinds of polarizations on the right-hand side of Eq. (2.1) are explained in details below.

2.2.1.1 Reversible Potential

The reversible potential, E^{rev} , is usually determined by the Nernst equation for an electrochemical reaction with n electrons transferred at a temperature of T as demonstrated with the SOFC [1, 14, 16]:

$$E^{\text{rev}} = E^0 + \frac{RT}{nF} \ln \left(\frac{\left(\frac{p_{\text{H}_2}}{p^0} \right) \left(\frac{p_{\text{O}_2}}{p^0} \right)^{0.5}}{\left(\frac{p_{\text{H}_2\text{O}}}{p^0} \right)} \right), \quad (2.2)$$

in which F is the Faraday's constant. The reversible potential at standard conditions, E^0 , is 1.18 V for the SOFC fueled by hydrogen [16], according to $E^0 = -\Delta G^0/(nF)$, in which ΔG^0 is the standard Gibbs free energy change.

As shown in Eq. (2.2), the reversible Nernst potential depends on the partial pressure of the reactant and product species. The partial pressure of species is defined in different ways in the existing SOFC models. For example, Chan et al.

[17] used the inlet partial pressure of species to calculate the reversible Nernst potential in a 0D model, given by

$$E^{\text{rev}} = \frac{RT}{2F} \ln K - \frac{RT}{2nF} \ln \left(\frac{(p_{\text{H}_2\text{O}}^{\text{in}})^2 p^0}{p_{\text{H}_2}^{\text{in}} p_{\text{O}_2}^{\text{in}}} \right), \quad (2.3)$$

where K is the equilibrium constant from the JANAF table, the number of transfer elections $n = 2$, and p^{in} is the inlet pressure in Pa. The inlet partial pressure is applicable, because the cell was treated as a black box and only the performance of the entire cell was in interest.

Instead of using the inlet pressure, Yuan et al. [18] expressed E^{rev} in terms of the partial pressure of species in the bulk flow in the channel:

$$E^{\text{rev}} = E^0 + \frac{RT}{nF} \ln \left(\frac{p_{\text{H}_2, \text{fc}} p_{\text{O}_2, \text{fc}}^{0.5}}{p_{\text{H}_2\text{O}, \text{fc}}} \right), \quad (2.4)$$

where $E^0 = 1.2723\text{--}2.7645 \times 10^4$ T, $n = 2$, and $p_{i, \text{fc}}$ is pressure in bar for species i in the flow channel. The reaction sites were considered to be uniformly distributed in the electrode, which means that the electrochemical half-cell reaction takes place through the entire electrode rather than only at the electrode/electrolyte interface. On the other hand, if the reactions are assumed to occur merely at the electrode/electrolyte interface, the local partial pressures at this interface are used instead [19, 20]. Equation (2.4) was also employed by Ni et al. [21] with $E^0 = 1.253\text{--}2.4516 \times 10^4$ T.

In addition, Lazzareo et al. [22] evaluated the Nernst potential as an intermediate value between those at both cell inlet and outlet, given by

$$E^{\text{rev}, \text{in}} = -\frac{\Delta G^0}{2F} + \frac{RT^{\text{in}}}{2F} \ln \left(\frac{p_{\text{H}_2}^{\text{in}} (p_{\text{O}_2}^{\text{in}})^{0.5}}{p_{\text{H}_2\text{O}}^{\text{in}}} \right), \quad (2.5)$$

$$E^{\text{rev}, \text{out}} = -\frac{\Delta G^0}{2F} + \frac{RT^{\text{out}}}{2F} \ln \left(\frac{p_{\text{H}_2}^{\text{out}} (p_{\text{O}_2}^{\text{out}})^{0.5}}{p_{\text{H}_2\text{O}}^{\text{out}}} \right), \quad (2.6)$$

$$E^{\text{rev}} = k_v \frac{E^{\text{rev}, \text{in}} + E^{\text{rev}, \text{out}}}{2}. \quad (2.7)$$

The tuning constant, k_v , in Eq. (2.7) was between 0.97 and 1.02 to increase the accuracy of experimental data prediction.

2.2.1.2 Activation Polarization

The activation polarization derives from the activation barrier that the energy of reactants must exceed to trigger the electrochemical reactions [14]. The activation loss at the anode or cathode is evaluated according to the Butler-Volmer equation in terms of the asymmetry factor, $\mathfrak{R}$, which is usually between 0.4 and 0.6 experimentally [23],

$$i = i_0 \left[\exp\left(\frac{\mathfrak{R}nF}{RT} \eta^{\text{act}}\right) - \exp\left(-\frac{(1-\mathfrak{R})nF}{RT} \eta^{\text{act}}\right) \right], \quad (2.8)$$

or in terms of the transfer coefficient α as

$$i = i_0 \left[\exp\left(\frac{\alpha_a F}{RT} \eta^{\text{act}}\right) - \exp\left(-\frac{\alpha_c F}{RT} \eta^{\text{act}}\right) \right], \quad (2.9)$$

where α_a and α_c are the anodic and cathodic apparent transfer coefficient respectively, usually between 0.2 and 2 [24]. Both $\mathfrak{R}$ and α are related to how an applied potential favors one direction over the other in the half-cell reactions by Eqs. (1.4) and (1.5). The exchange current density, i_0 , represents the rate of exchange between the reactant and product states at equilibrium. Equations (2.8) and (2.9) assume a single rate determining step.

The exchange current density can be expressed in different equivalent forms. It is mainly affected by four factors including reactant concentration, activation barrier, temperature, and the number of possible reaction sites [14]. Moreover, some adjustable parameters may be involved in the expression of the exchange current density to fit the experimental data [25]. For example, the anodic exchange current density used by Shi et al. [25, 26] is

$$i_{0,a} = \frac{\mathcal{B}_a RT}{3F} \left(\frac{c_{\text{H}_2}}{c_{\text{H}_2}^{\text{ref}}} \right) \exp\left(-\frac{E_a^{\text{act}}}{RT}\right) (p_{\text{H}_2\text{O},a})^{0.133}, \quad (2.10)$$

where $c_{\text{H}_2}^{\text{ref}}$ is the hydrogen concentration in the reference case, and $p_{\text{H}_2\text{O},a}$ is the water vapor partial pressure at the anode. For the anode support layer, an activation energy $E_a^{\text{act}} = 1.2 \times 10^5 \text{ J mol}^{-1}$ and an adjustable parameter $\mathcal{B}_a = 6.17 \times 10^{11} \Omega^{-1} \text{ m}^{-2}$ were specified. For the anode active layer, $E_a^{\text{act}} = 1.2 \times 10^5 \text{ J mol}^{-1}$ and $\mathcal{B}_a = 6.8 \times 10^{12} \Omega^{-1} \text{ m}^{-2}$, which were tuned to fit the experimental data [25]. The cathodic exchange current density corresponding to both cathode support and active layers is determined by [27]

$$i_{0,c} = \frac{\mathcal{B}_c RT}{4F} \exp\left(-\frac{E_c^{\text{act}}}{RT}\right) (p_{\text{O}_2,c})^{0.25}, \quad (2.11)$$

where $i_{0,c}$ is the cathodic exchange current density, $p_{O_2,c}$ is the oxygen partial pressure at the cathode, $E_c^{\text{act}} = 1.3 \times 10^5 \text{ J mol}^{-1}$ and $\beta_c = 5.8 \times 10^{10} \Omega^{-1} \text{ m}^{-2}$ which are adjustable [25].

VanderSteen et al. [28] introduced the effects of gaseous species partial pressure, temperature, reaction order, and activation energy on the exchange current density,

$$i_{0,c} = i_{0,c}^{\text{ref}} \left(\frac{p_{O_2}}{p_{O_2}^0} \right)^\gamma \exp \left[\frac{-E_c^{\text{act}}}{R} \left(\frac{1}{T} - \frac{1}{T^0} \right) \right], \quad (2.12)$$

$$i_{0,a} = i_{0,a}^{\text{ref}} \left(\frac{p_{H_2}}{p_{H_2}^0} \right)^{\gamma_1} \left(\frac{p_{H_2O}}{p_{H_2O}^0} \right)^{\gamma_2} \exp \left[\frac{-E_a^{\text{act}}}{R} \left(\frac{1}{T} - \frac{1}{T^0} \right) \right], \quad (2.13)$$

where $i_{0,c}^{\text{ref}}$ and $i_{0,a}^{\text{ref}}$ are the reference exchange current densities, γ and $\gamma_1 + \gamma_2$ are the reaction order, T^0 is the reference temperature, and E^{act} is the activation energy. According to electrochemical kinetics, the reference exchange current density, the transfer coefficients, and the reaction order depend on the rate determining step(s) of the electrochemical reaction and the electrode microstructure [29].

The exchange current density is also affected by pore radius and porosity, which determine the TPB length in the electrode as reported by Ni et al. [30]:

$$\begin{aligned} i_{0,a} &= \beta_a \frac{1 - \varepsilon}{r} \exp \left(-\frac{E_a^{\text{act}}}{RT} \right), \\ i_{0,c} &= \beta_c \frac{1 - \varepsilon}{r} \exp \left(-\frac{E_c^{\text{act}}}{RT} \right), \end{aligned} \quad (2.14)$$

where $E_a^{\text{act}} = 1.0 \times 10^5 \text{ J mol}^{-1}$ and $E_c^{\text{act}} = 1.2 \times 10^5 \text{ J mol}^{-1}$ are the activation energy of anode and cathode respectively, ε is the porosity of electrode, r is the pore radius, β_a and β_c are the coefficients for exchange current density of the anode and cathode respectively. Increasing electrode porosity or pore size decrease the concentration polarization and increase the activation polarization of CH_4 fed SOFC [30]. At low current densities, low porosity and small pore size are desirable to reduce the electrode total polarizations, as concentration polarization is insignificant compared with activation polarization. At high current densities, the total polarizations can be minimized by optimizing the porosity and pore size.

The form of the Butler-Volmer equation for each half-cell reaction also depends on the assumption on where the reaction takes place. When the reaction is assumed to occur at the interface between the electrode and the electrolyte, the Butler-Volmer equation is expressed as a current density in A m^{-2} in terms of roughness factor a of the electrode, reference exchange current density i_0^{ref} , reference and local reactant mole concentration c_R^{ref} and c_R , and reaction order γ ,

$$i = ai_0^{\text{ref}} \left(\frac{c_R}{c_R^{\text{ref}}} \right)^{\gamma_{a/c}} \left[\exp \left(\frac{\Re n F}{RT} \eta^{\text{act}} \right) - \exp \left(- \frac{(1 - \Re) n F}{RT} \eta^{\text{act}} \right) \right]. \quad (2.15)$$

When the reaction takes place over a layer of finite volume, the Butler-Volmer equation is expressed as a volumetric current density in A m^{-3} involving the specific reactive surface area of electrode $\mathcal{A}$ [31–35]

$$J = \mathcal{A} i_0^{\text{ref}} \left(\frac{c_R}{c_R^{\text{ref}}} \right)^{\gamma_{a/c}} \left[\exp \left(\frac{\Re n F}{RT} \eta^{\text{act}} \right) - \exp \left(- \frac{(1 - \Re) n F}{RT} \eta^{\text{act}} \right) \right]. \quad (2.16)$$

Furthermore, the Butler-Volmer equation can be simplified depending on the desired accuracy and the expected activation polarization [36–40]. For a SOFC at 1073 K, under high activation polarization with $|\eta^{\text{act}}| > 0.2516$, Eq. (2.16) can be simplified to the Tafel equation,

$$J_c = -\mathcal{A} i_{0,\text{O}_2}^{\text{ref}} \left(\frac{c_{\text{O}_2}}{c_{\text{O}_2}^{\text{ref}}} \right)^{\gamma_{\text{O}_2}} \exp \left(- \frac{\Re n F}{RT} \eta_c^{\text{act}} \right), \quad (2.17)$$

for the cathode side with an error of 5 %. Under low activation polarization with $|\eta^{\text{act}}| < 0.1006$, a linear current-potential relation is applicable, such that Eq. (2.16) becomes

$$J_a = \mathcal{A} i_{0,\text{H}_2}^{\text{ref}} \left(\frac{c_{\text{H}_2}}{c_{\text{H}_2}^{\text{ref}}} \right)^{\gamma_{\text{H}_2}} \frac{\Re n F}{RT} \eta_a^{\text{act}}, \quad (2.18)$$

for the anode side.

2.2.1.3 Ohmic Polarization

Electricity in the SOFC is transported through the conducting solid porous backing layer by the flow of electrons, through the solid dense electrolyte by the flow of ions, and through the reaction zone layer of a finite volume by the flow of both electrons and ions. Therefore, the ohmic polarization in the cell arises due to electrical resistance in the cell components including [1, 14]:

- (1) Resistance to the flow of ions in the electrolyte (ionic resistance);
- (2) Resistance to the electrons and ions in the reaction zone layer (ionic and electronic resistance);
- (3) Resistance to the flow of electrons through the electrode-backing layer (or gas diffusion layer) (electronic resistance); and
- (4) Resistance to the flow of electrons through interconnects (electronic resistance).

The resistance to the electron or ion transport is described by the Ohm's law [2, 14],

$$\eta^{\text{ohm}} = I\mathcal{R} = (JA)\mathcal{R}, \quad (2.19)$$

where I is the current density, J is the volumetric current density, and A is the cross-sectional area of the conducting cell component normal to the conducting path, $\mathcal{R}$ is the electronic/ionic resistance in a cell component. The total cell ohmic loss, η^{ohm} , is the sum of electronic and/or ionic ohmic loss in anode/cathode interconnect, backing and reaction zone layers as well as the electrolyte, expressed as

$$\eta^{\text{ohm}} = \eta_{\text{ic,c}}^{\text{ohm}} + \eta_{\text{bl,c}}^{\text{ohm}} + \eta_{\text{rl,c}}^{\text{ohm}} + \eta_{\text{el}}^{\text{ohm}} + \eta_{\text{rl,a}}^{\text{ohm}} + \eta_{\text{bl,a}}^{\text{ohm}} + \eta_{\text{ic,a}}^{\text{ohm}}. \quad (2.20)$$

The ohmic polarization at the electrolyte usually dominates the total ohmic loss in fuel cell, because the electrolyte resistance is usually larger than the electrode resistance [14, 17].

2.2.1.4 Concentration Polarization

Concentration polarization is the voltage loss that is caused by non-uniform distributions of reactants, which result from processes hindering the transport of mass. In SOFC, these processes are the convective mass transfer of gaseous species from the flow channel to the porous electrode, and the slow diffusion of reactants and products through the porous electrodes. The chemical reactions are hindered due to the lack of reactants which transports slowly to reach the reaction sites. This is usually the case that the electrochemical reaction is too fast corresponding to a high current density. Wang et al. [41] suggested that the total concentration potential of the cell could be decreased by decreasing the thickness or increasing the porosity of the anode.

The concentration polarization is determined by

$$\eta^{\text{con}} = -\frac{RT}{nF} \ln \left(1 - \frac{i}{i_L} \right), \quad (2.21)$$

where i_L is the limiting current density, which is the maximum current density produced by the cell. At the limiting current density, the rate of reactants consumed is equal to the amount of reactants supplied, such that no extra reactants would accumulate at the reaction sites. η^{con} can also be formulated in the Nernst form in terms of species concentration as follows [42]:

$$\eta_a^{\text{con}} = \frac{RT}{2F} \left(\ln \frac{c_{\text{H}_2}^{\text{channel}}}{c_{\text{H}_2}^{\text{TPB}}} - \ln \frac{c_{\text{H}_2\text{O}}^{\text{channel}}}{c_{\text{H}_2\text{O}}^{\text{TPB}}} \right), \quad (2.22)$$

$$\eta_c^{\text{con}} = \frac{RT}{4F} \left(\ln \frac{c_{\text{O}_2}^{\text{channel}}}{c_{\text{O}_2}^{\text{TPB}}} \right). \quad (2.23)$$

Furthermore, the concentration polarizations can even be formulated in more complicated ways to show the dependence of η^{con} on diffusivities, electrode thickness, and mole fraction of species together, based on how the mass transport is modeled using the Fick's model, the Stefan-Maxwell model, or the Dusty-gas model [43]. All these three diffusion models are introduced in Sect. 2.2.2.2.

2.2.1.5 Leak Polarization

The maximum cell voltage is measured experimentally when there is no electric current drawn from the cell, thereby obtaining the open circuit, E^{OCV} . This open circuit cell voltage is always slightly less than the thermodynamic reversible cell potential, E^{rev} [2], as shown schematically in Fig. 2.1. The difference between E^{OCV} and E^{rev} is called leak polarization, η^{leak} , which may result from residual electronic conduction in the electrolyte, gases crossing over the electrolyte via micro cracks and fissures, and/or a leak current that is wasted by unwanted side reactions or non-electrochemical reactions at the electrodes [2, 14, 44]. To the best of my knowledge, there is no mathematical formula to evaluate the leak polarization involving all the causes above. The η^{leak} can be specified as a modelling parameter and its value is tuned to fit the numerically predicted polarization curve with experimental data [25, 26]. Typically, the leak polarization is considerable for low-temperature fuel cells such as PEMFCs, but less markedly for high-temperature fuel cells such as SOFCs, as reported in Ref. [44]. For example, it was estimated that $\eta_{\text{leak}} \sim 0.2 \text{ V}$ with respect to $E^{\text{rev}} \sim 1.2 \text{ V}$ for the PEMFC (i.e., $\eta_{\text{leak}}/E^{\text{rev}} \sim 17 \%$) [44], and $\eta_{\text{leak}} \sim 0.03 \text{ V}$ to $E^{\text{rev}} \sim 1.1 \text{ V}$ for the SOFC (i.e., $\eta_{\text{leak}}/E^{\text{rev}} \sim 3 \%$) [25].

2.2.2 Transport Phenomena in Electrodes

Before determining the various voltage and heat losses in the fuel cell, it is essential to understand the transport of mass, momentum, and energy within the cell components, especially in the porous electrodes [13]. The electrode can be subdivided into a backing (or gas diffusion) layer and a reaction zone layer, which are made of the same material but function differently [2]. The difference between them is that the half-cell electrochemical reactions by Eqs. (1.4) and (1.5) take place at the

reaction sites (namely, the triple phase boundaries (TPB)) in the reaction zone layer, but not in the backing layer. Both ions and electrons transfer in the reaction zone layer, while only electrons transfer in the backing layer.

2.2.2.1 Volume Averaging for Porous Media

In the porous electrode of the fuel cell, gas and solid phases coexist. A detailed description of every pore is not practical. In order to obtain the governing equations of the mass, momentum, and energy transport in porous media, a common approach, called the volume-averaging method [45], is implemented by taking an average of the governing equations originally describing the fluid in free flow fields over a representative elementary volume, whose length scale is considerably smaller than that of the macroscopic flow domain, but large enough to incorporate sufficient solid and void elements for an accurate average description [1, 45, 46].

It is worth mentioning a difference between an average taken over the whole representative elementary volume V , and that taken over a volume element $V^{(g)}$ consisting of pores filled by gaseous fluid. Let $\mathbf{B}$ be a quantity, either scalar, vector, or tensor, associated with the gas phase. Let $\langle \mathbf{B} \rangle$ represent the superficial volume average of $\mathbf{B}$:

$$\langle \mathbf{B} \rangle = \frac{1}{V} \int_{V^{(g)}} \mathbf{B} dV. \quad (2.24)$$

Let $\langle \mathbf{B} \rangle^{(g)}$ be the intrinsic volume average of $\mathbf{B}$ in the gas phase,

$$\langle \mathbf{B} \rangle^{(g)} = \frac{1}{V^{(g)}} \int_{V^{(g)}} \mathbf{B} dV. \quad (2.25)$$

The relation between the superficial and intrinsic volume average is

$$\langle \mathbf{B} \rangle = \varepsilon \langle \mathbf{B} \rangle^{(g)}, \quad (2.26)$$

in which ε is the porosity defined as the ratio of the pore space $V^{(g)}$ to the overall space V including both pores and solid, given by

$$\varepsilon = V^{(g)} / V. \quad (2.27)$$

The value of porosity can vary from zero for pure solid regions to unity for free fluid flow. The cell performance is strongly dependent on porosity as well as detailed porous microstructure distributions of electrodes [47].

Generally, the superficial average velocity is the preferred quantity for the macroscopic or volume averaged velocity field, while the intrinsic average

concentration and pressure are preferred in the representation of governing equations for porous media [45]. For simplicity, all angle brackets and superscript (g) are ignored in the following context.

2.2.2.2 Mass Transfer

The mass transfer of gaseous working fluid in the electrode is typically governed by the continuity equation [48],

$$\nabla \cdot (\rho \mathbf{v}) = S_{\text{mass}}, \quad (2.28)$$

where ρ is the density of the gaseous mixture, and $\mathbf{v}$ is the superficial velocity for the porous electrode. The mass source/sink term, S_{mass} , due to the electrochemical reactions at the reaction sites is calculated by the Faraday's law [24],

$$S_{\text{mass}} = - \sum_i \frac{s_i M_i |J|}{nF}, \quad (2.29)$$

where s_i is the stoichiometric coefficient, and n is the number of electrons consumed or produced by the cathode or anode half-cell reaction. Taking the reactions defined by Eqs. (1.4) and (1.5) for instance, S_{mass} is zero for the backing layer, but $M_{\text{O}_2} J_c / 4F$ for the cathode reaction zone layer and $-M_{\text{H}_2} J_a / 2F + M_{\text{H}_2\text{O}} J_a / 2F$ for the anode reaction zone layer in terms of the cathodic, J_c , and anodic volumetric current density, J_a . Here, J_c is inherently negative and J_a is positive.

The working fluid actually consists of various gaseous species. In theory, no molecule can go through the dense electrolyte from one electrode side to the other in the SOFC [49]. The mass transport of uncharged species involves both convection and diffusion in the electrode and is dominated by the latter [50]. Among these species, oxygen molecules are much larger in size than hydrogen molecules. Therefore, the transport of molecular oxygen to the electrode surface is much more difficult, and hence much slower, than the transport of the molecular hydrogen at the anode. The transport of gaseous species can be represented in terms of either the mass flux [28, 51, 52],

$$\nabla \cdot (\rho \omega_i \mathbf{v} + \mathbf{j}_i) = S_i, \quad (2.30)$$

$$S_i = \begin{cases} + \frac{M_{\text{O}_2} J_c}{4F} & (\text{O}_2, \text{ cathode rl}) \\ + \frac{M_{\text{H}_2\text{O}} J_a}{2F} & (\text{H}_2\text{O}, \text{ anode rl}) \\ - \frac{M_{\text{H}_2} J_a}{2F} & (\text{H}_2, \text{ anode rl}) \\ 0 & (\text{bl}) \end{cases}, \quad (2.31)$$

or the molar flux

$$\nabla \cdot (c_i \mathbf{v} + \mathbf{J}_i) = S_i, \quad (2.32)$$

$$S_i = \begin{cases} +\frac{J_c}{4F} & (\text{O}_2, \text{ cathode rl}) \\ +\frac{J_a}{2F} & (\text{H}_2\text{O}, \text{ anode rl}) \\ -\frac{J_a}{2F} & (\text{H}_2, \text{ anode rl}) \\ 0 & (\text{bl}) \end{cases}. \quad (2.33)$$

The first term on the left-hand side of Eqs. (2.30) and (2.32) corresponds to convection, and the second one stands for diffusion. In the literature, three diffusion models are available to describe the mass transfer of each species in porous media, called the Fick's model, the Stefan-Maxwell model, and the Dusty-Gas model [28].

The Fick's model (FM) is the simplest diffusion model [10, 28, 48, 51, 53], in which the mass diffusive flux of species i is defined as

$$\mathbf{j}_i = -\rho D_{ij} \nabla \omega_i, \quad (2.34)$$

and the molar diffusive flux of species i is

$$\mathbf{J}_i = -D_{ij} \nabla c_i = -c D_{ij} \nabla x_i, \quad (2.35)$$

where ω_i is the mass fraction, x_i is the mole fraction, c_i is the molar concentration of species i , c is the total molar concentration of the gas mixture, and D_{ij} is the binary diffusivity for a binary mixture of component i and j . The mass fraction formalism for chemical species concentration is better than the mole fraction, molar concentration or species partial pressure, in respect of the fact that reactions occur in a gas phase which is able to modify its local density due to variations in composition and temperature [54]. The FM is only used for dilute or binary systems, but not applicable when a large number of gaseous species is present [52].

The Stefan-Maxwell model (SMM) is a general alternative to calculate the diffusion in a multi-component system [28, 48]. The Stefan-Maxwell equations determining the diffusion flux for a multi-component system of n species are [14, 28, 43, 48, 55]

$$-c \nabla x_i = \sum_{j=1, i \neq j}^n \frac{1}{D_{ij}} (x_j \mathbf{N}_i - x_i \mathbf{N}_j), \quad (2.36)$$

where c is the concentration of the mixture, D_{ij} is the ordinary or binary diffusion coefficient of species i in j , x_i is the mole fraction of species i , and $\mathbf{N}_i$ is the diffusion flux of species i .

The Dusty-Gas model (DGM) or extended Stefan-Maxwell model combines the Stefan-Maxwell model with the Knudsen diffusion for small pore sizes [56].

Knudsen diffusion takes the frequent collisions between gaseous molecules and the solid porous material into account. In contrast, this molecule-wall collision effect is neglected in the FM and the SMM. A diffusion model accounting for both the ordinary and Knudsen diffusion are expressed as [10, 28, 43, 55]

$$-c\nabla x_i = \sum_{j=1, i \neq j}^n \frac{1}{D_{ij}} (x_j \mathbf{N}_i - x_i \mathbf{N}_j) + \frac{\mathbf{N}_i}{D_{\text{Kn},i}}, \quad (2.37)$$

where D_{ij} is the ordinary or binary diffusion coefficient and $D_{\text{Kn},i}$ is the Knudsen diffusion coefficient of species i . Mathematically, the two terms on the right-hand side of Eq. (2.37) can be combined to get modified Stefan-Maxwell equations for the multi-component diffusion in the porous electrodes,

$$-c\nabla x_i = \sum_{j=1, i \neq j}^n \frac{1}{\mathfrak{D}_{ij}} (x_j \mathbf{N}_i - x_i \mathbf{N}_j), \quad (2.38)$$

where $\mathfrak{D}_{ij}$ is the combined diffusion coefficient defined as [12, 28]

$$\mathfrak{D}_{ij} = \frac{D_{ij} D_{\text{Kn},i}}{D_{ij} + D_{\text{Kn},i}}. \quad (2.39)$$

Because both electrodes are porous, in order to account for the effect of the porous matrix on the diffusive transport, the intrinsic diffusion coefficient $\mathfrak{D}_{ij}$ is corrected with some geometric factors like porosity and tortuosity as follows [28]

$$D_{ij}^{\text{eff}} = \frac{\varepsilon}{\tau} \mathfrak{D}_{ij}. \quad (2.40)$$

Alternatively, the Bruggeman correction is also widely used in the estimation of the effective diffusivity in a porous medium [28],

$$D_{ij}^{\text{eff}} = \varepsilon^{1.5} \mathfrak{D}_{ij}, \quad (2.41)$$

where the exponent of 1.5 is an empirical constant. At high temperatures, Eq. (2.40) for the effective diffusivity is more accurate than Eq. (2.41) [14]. Consequently, the modified Stefan-Maxwell Eq. (2.38) becomes [48]

$$-c\nabla x_i = \sum_{j=1, i \neq j}^n \frac{1}{D_{ij}^{\text{eff}}} (x_j \mathbf{N}_i - x_i \mathbf{N}_j). \quad (2.42)$$

The diffusion process is characterized by the Knudsen number $\text{Kn} = \lambda_m/d$, where λ_m is the mean free path length in the gas, and d is a characteristic pore diameter [57]. When $\text{Kn} < 0.01$, bulk diffusion is dominant [57]. When $\text{Kn} > 1$, Knudsen diffusion is dominant, and interparticle collisions are less frequent than

collisions with the pore wall, such that the continuum theory is invalid for the diffusive mass transfer to occur through sub-micron size pores with a large Knudsen number [57]. When $0.01 < \text{Kn} < 1$, which is the typical situation in the SOFC electrode, both bulk diffusion and Knudsen diffusion are comparable, and thus should be considered together. The dusty-gas model (DGM) was developed to represent multicomponent transport in this situation [57].

All these three models are applicable to predict the concentration polarization inside the electrode. Cayan et al. [58] compared the distribution of the species mole fraction inside the porous anode between two models with the SMM and the FM, respectively. Both two models accounted for the Knudsen diffusion. These two models agreed well at low current densities, whereas an obvious difference appeared as the current increased. Yakabe et al. [40] and Lehnert et al. [59] developed mass transport models similar to the dusty-gas model considering the water-gas shift reaction. Suwanwarangkul et al. [43] employed all the three models to simulate two binary systems ($\text{H}_2\text{-H}_2\text{O}$ and CO-CO_2) and a ternary system ($\text{H}_2\text{-H}_2\text{O-Ar}$) without considering the shift reaction.

2.2.2.3 Momentum Transfer

The momentum transfer in the electrode of the SOFC can be described by the Darcy's law [1, 13], which is commonly applied with an assumption of a steady-state incompressible flow [60–62],

$$\nabla p = -\frac{\mu_{\text{mix}}}{\kappa} \mathbf{v}, \quad (2.43)$$

where p is the intrinsic pressure, $\mathbf{v}$ is the superficial velocity, μ_{mix} is the dynamic viscosity of the gas mixture, and κ is the permeability of porous media.

However, the Darcy equation cannot model a no-slip condition at a wall nor the resulting boundary layers, which means that it can only describe the flow within the porous structure well away from the walls and is problematic to define interfacial conditions at the interface between two domains [63]. Haberman and Yong [56] provided an estimation on the thickness the boundary layers formed over a solid surface and at the electrode/channel interface. They suggested to modify the Darcy's law by adding a so-called "Brinkman" term [64–66], such that another well-know equation, Brinkman equation, was obtained to allow their calculation at boundaries. The Brinkman equation for steady incompressible flow was developed by Brinkman in 1949 with an additional viscous term in the momentum balance [67, 68]:

$$\nabla p = -\frac{\mu_{\text{mix}}}{\kappa} \mathbf{v} + \nabla \cdot \left[\frac{\mu_{\text{mix}}}{\varepsilon} \left((\nabla \mathbf{v} + (\nabla \mathbf{v})^\dagger) \right) \right]. \quad (2.44)$$

Although the Darcy's law was also used to describe the weakly-compressible flow in some models [61, 69], many other models involved a non-zero rate of

expansion, $\nabla \cdot \mathbf{v}$, in Eq. (2.44) to govern the steady weakly-compressible flow, such that the Brinkman equation became [62, 70]

$$\frac{\mu_{\text{mix}}}{\kappa} \mathbf{v} + \frac{1}{\varepsilon^2} \nabla \cdot (\rho \mathbf{v}) \mathbf{v} = -\nabla p + \nabla \cdot \left[\frac{\mu_{\text{mix}}}{\varepsilon} \left((\nabla \mathbf{v} + (\nabla \mathbf{v})^\dagger) - \frac{2}{3} (\nabla \cdot \mathbf{v}) \mathbf{I} \right) \right]. \quad (2.45)$$

The second term on the left-hand side of Eq. (2.45) is derived from the convective term in Navier-Stokes equation, when applying the volume-averaging method to the electrode. For the transfer of gaseous species in porous electrodes, diffusion is dominant over convection [12, 50]. For the left-hand side of Eq. (2.45), in view of the order of magnitude, the second term is much smaller than the first term and thus can be dropped from the equation. Then, Eq. (2.45) becomes [71]

$$\frac{\mu_{\text{mix}}}{\kappa} \mathbf{v} = -\nabla p + \nabla \cdot \left[\frac{\mu_{\text{mix}}}{\varepsilon} \left((\nabla \mathbf{v} + (\nabla \mathbf{v})^\dagger) - \frac{2}{3} (\nabla \cdot \mathbf{v}) \mathbf{I} \right) \right], \quad (2.46)$$

Moreover, the Darcy's law was modified with a form-drag constant, c_F , to get the Forchheimer's equation in some other models [18, 70, 72],

$$\nabla p = -\frac{\mu_{\text{mix}}}{\kappa} \mathbf{v} - c_F \frac{\rho}{\sqrt{\kappa}} |\mathbf{v}| \mathbf{v}. \quad (2.47)$$

2.2.2.4 Energy Transfer

Although an isothermal state is often imposed on SOFC mathematical models [25, 30, 40, 73–76], the temperature may not distribute uniformly throughout the cell or stack [77]. Detailed non-isothermal numerical models began to emerge in the early 1990s, such as [78, 79]. In fact, the temperature distribution is related to the electrochemical kinetics of the cell: heat can be generated by the electrochemical reactions within the reaction zones and the current passing through the cell, and be consumed by the endothermic internal reforming reactions at the anode side [1]. Large temperature gradients can cause undesired large thermal-mechanical stresses and thermal expansion mismatches, since the cell generally consists of ceramic components with different thermal expansion coefficients [80]. This may thus result in the thermal-mechanical degradation and/or structure failure of a SOFC. Moreover, the material properties of the cell are strong functions of temperature, and reaction rates are temperature-dependent as well [28]. Therefore, accurate prediction of the heat transfer and temperature distribution is essential for designing and optimizing a SOFC single cell and stack.

In the electrode of the SOFC, the heat transfer in the solid structures occurs mainly through conduction, while convective heat transfer dominates in the gas channels and pores. Based on volume-averaging for porous media, the heat transfer

in the porous electrodes of the SOFC is generally modeled in two ways: local thermal equilibrium (LTE) and local thermal non-equilibrium (LTNE) [12]. LTE prescribes the same temperature, T , for the gas and solid phases associated with effective transport parameters. The heat flux is given by [28, 52, 56, 61, 71, 81, 82]

$$\mathbf{q} = -\nabla \cdot (k^{\text{eff}} \nabla T) + \rho c_{p, \text{mix}} \mathbf{v} \cdot \nabla T, \quad (2.48)$$

in which $c_{p, \text{mix}}$ is the specific heat of the gas mixture. The effective thermal conductivity $k^{\text{eff}} = \varepsilon k_{\text{mix}} + (1 - \varepsilon)k_s$ depending on the porosity of the electrode, ε , and thermal conductivities of the gas mixture and solid structure, k_{mix} and k_s . In contrast, LTNE predicts the temperatures of the solid and gas phases, T_s and T_g , separately. The heat flux is specified as [83, 84]

$$\mathbf{q}_s = -\nabla \cdot (k_s^{\text{eff}} \nabla T_s), \quad (2.49)$$

for the solid electrode, and

$$\mathbf{q}_g = -\nabla \cdot (k_g^{\text{eff}} \nabla T_g) + \rho c_{p, \text{mix}} \mathbf{v} \cdot \nabla T_g, \quad (2.50)$$

for the gas mixture. Here, the effective thermal conductivity $k_s^{\text{eff}} = (1 - \varepsilon)k_s$ for the solid phase and $k_g^{\text{eff}} = \varepsilon k_{\text{mix}}$ for the gas phase.

The LTE approach is applied when the difference between the temperatures of the fluid and the solid structure are small; otherwise, the LTNE approach is adopted. According to the results provided by Zheng et al. [85], the temperature difference between the solid and gas phases in the porous electrode remained on the order of 10^{-3} K under a wide range of operating conditions and structural parameters for H_2 -fueled SOFCs, and 1 K for NH_3 -fueled SOFCs at high operating temperatures such as 1200 K. The temperature difference is negligible in view of its order of magnitude. Therefore, it is safe to apply LTE to the electrode for the SOFC fueled by hydrogen or hydrocarbon.

In consideration of the high operating temperature of SOFC, thermal radiation was studied by some researchers for an accurate heat balance, [86, 87]. Nevertheless, radiation is generally neglected in the SOFC model, especial in the cell model [2, 9, 11, 28, 41].

2.2.2.5 Reaction Zone

For SOFC modelling, the reaction zones comprising the triple-phase boundaries for the electrochemical reactions to take place are generally modeled as a boundary or interface condition, rather than a spatial domain, when predicting the global cell performance [20, 40, 41, 52, 88–90] or the local distribution of mass fraction, current density, and/or temperature [91, 92]. Correspondingly, the source terms for the conservation of mass, species, energy, and charge appear as the boundary

conditions at the electrode/electrolyte interface. In fact, however, dependent variables undergo spatial changes in the reaction zones that are spread out some distance (10–50 μm) into the electrode from the electrode/electrolyte interface [39, 93–95].

Kim et al. [39] and Zhu and Kee [96] discussed the existence of the reaction zone layers, but still modeled them as boundary conditions rather than individual domains of finite volumes. Chung et al. assumed the triple phase boundaries (TPB) to distribute over the entire electrode of their model, because the thicknesses of the electrodes investigated were small and less than 50 μm [42]. Hussain et al. published a series of papers to predict the performance of the anode [55, 73] and the whole planar SOFC [31, 94, 97], in which the reaction zone was modeled as a separate layer of a finite volume adjacent to the backing layer. They also compared the approaches to model the anode reaction zone as a boundary condition and as a discrete volume [73]. Verda and Spakovsky [62] developed a continuous model for the electrochemistry, which allows one to examine different TPB geometries, instead of utilizing an equivalent resistive circuit approach or a homogeneous distribution of TPB.

2.2.3 *Spatial Dimension*

A practical problem of fuel cells can be solved by 0D, 1D, 2D, or 3D modelling [2, 10, 11]. A 2D grid in the plane of the cell can be used to model the P-SOFC with co- or counter-flow configurations [52, 80]. A third coordinate is at times considered to take into account concentration and temperature gradients along the perpendicular direction for the cross-flow configuration [60, 90].

2.2.3.1 **Zero-Dimensional Model**

The simplest approach for SOFC modelling is to consider system as a black box, resulting in a 0D model. Based on principles of thermodynamics and electrochemistry, the fuel cell is modeled into a group of algebra equations to solve for the cell output such as cell voltage, power output, and cell efficiency based on pre-defined operating conditions such as inlet gas composition, inlet temperature, fuel and air utilization ratio [2]. Therefore, the 0D models are fit to evaluate the performance of the whole cell or stack, rather than the local multiphysical phenomena inside the cell or stack. For example, Chan et al. [17] fully described the cell voltage and all three types of polarization in a 0D model, and conducted a sensitivity test to investigate the effect of the thickness of respective cell components on the drop in cell voltage. Akkaya et al. [98] developed a steady-state 0D model to simulate and evaluate the exergetic performance of the SOFC/gas turbine combined heat and

power (SOFC/GT CHP) system. Mueller et al. [99] designed a system control strategy for a bottoming SOFC/GT hybrid system. In recent years, 0D modelling is widely used in studies of the SOFC performance at the system level [100–105].

2.2.3.2 One-Dimensional Model

In the 1D model, the changes of gas and electrical properties in two directions are assumed to be uniform, such that two of the geometrical dimensions are neglected [2]. The retained dimension is usually along the flow direction, in which the variations of gas composition, temperature, and current density are more significant. Cheddie and Munroe [106] developed a 1D dynamic SOFC model for real time application to account for transport and polarization phenomena. Their results justified that the 1D model was an improvement over the zero-dimensional real time models, because it could provide more accurate predictions for the temperature and pressure variations along the cell and maintain real time capabilities with respect to computational time. Kang et al. [107] presented a 1D dynamic P-SOFC model based on mass and energy balances as well as electrochemical principles to predict some variables and their variation with time, including cell temperature, gas molar fractions, cell voltage, and current density along the cell length. The 1D modelling is applicable for the co-flow and counter-flow configurations, but not for the cross-flow design [2].

2.2.3.3 Two-Dimensional Model

For 2D modelling, the cell is represented by a 2D cross-sectional domain, and the changes of physics in the third dimension are ignored. The most representative co- or counter-flow cell configuration is with respect to the cell-length (streamwise) and cell-thickness (normal) directions, involving the interconnects, flow channels, and the positive electrode/electrolyte/negative electrode (PEN) structure. Based on such a 2D domain, Jin and Xue [76] developed a transient 2D isothermal model, which could be operated in both SOFC mode and SOEC (solid oxide electrolyzer cell) mode to investigate complicated multi-physics processes during the transient process of mode switching. Verda and Spakovsky [62] applied their CFD model to an anode-supported P-SOFC in 2D. Mahcene et al. [92] constructed a 2D P-SOFC single-cell model with co-flow pattern in Fortran language to investigate the distribution of the chemical species, temperature, current density, and power density. Their analysis was based on the fundamental conservation laws of momentum, energy, and species. In addition, when all the components are modeled in 2D, convective and radiative heat transfer become boundary conditions for the conservation of energy in gas channels and solid structures [77].

2.2.3.4 Three-Dimensional Model

Compared with 2D, 1D, and 0D modelling, 3D modelling can provide more detailed information of the SOFC behaviours [2]. Pasaogullari and Wang [108] proposed a 3D model to describe the electrochemical kinetics, multi-dimensional gas dynamics, and multi-component transport of species in the SOFC. Recknagle et al. [90] presented a 3D model for P-SOFC stacks to investigate the effect of the flow configuration on fuel utilization and the distribution of species mass fraction, temperature, and current density. All the three types of flow configurations, namely the co-, counter, and cross-flow configuration, can be modeled in 3D. The effects of the three flow configurations were compared and analysed not only at the single-cell level [19, 109] but also at the stack level [90, 110]. Yakabe et al. [89] analysed the distributions of chemical species concentrations, temperature, electric potential, and current density in two 3D single-cell models with co-flow and counter-flow configurations respectively. The effects of cell size, operating voltage, and thermal conductivities of the cell components on the simulation results were also investigated in their 3D models. Wang et al. [41] used two 3D P-SOFC single-cell models, with air and fuel channels in co-flow and counter-flow respectively, to predict the temperature distribution, molar concentration of gaseous species, current density, and over potential based on the fundamental conservation laws of mass, momentum, energy, and electrical charge.

Detailed 3D fuel cell models are usually very computationally expensive due to the highly coupled and nonlinear nature of their mathematical formulation as well as a large number of functional domains in the cell. In order to simplify the mathematical and computational complexity, the cell geometry is usually assumed to be 2D [18, 52, 80, 91], 1D [88, 106, 107], or 0D [17] instead of 3D. However, these assumptions are likely to lower the fidelity of model predictions. The flow fields of P-SOFCs in general are 3D entities comprising not only plain channels but also solid ribs. The ribs can affect the diffusion pathways of gaseous working fluid and electronic charges between the flow field and the backing layer [2]. An improper design for the geometry of ribs and electrodes may even cause the depletion of reactants to take place in the porous electrode at spanwise locations away from the channels at high current densities. These effects of ribs are generally not taken into account in existing 2D P-SOFC models, especially when modelling with respect to the streamwise and normal directions. This makes it difficult to achieve agreement between model predictions and experimental results [80].

Among the research works on SOFC modelling, single-cell models are generally the first consideration for researchers to embark on a new product design and conduct modelling, while stack modelling is necessary for the scale-up system design and often involves parametric studies. A single SOFC only produces a voltage of 0.7–0.5 V [93], such that a large amount of single cells must be stacked up to achieve a necessary high output voltage. The majority of computational studies focus on a SOFC single cell [36, 111–114], an anode [75, 115], or a cathode [74, 116], whereas relatively fewer can be found for stacks [17, 117] or the overall system [80, 93, 94]. The latter two in particular are challenging to solve with

detailed mechanistic models, because of the highly coupled and non-linear nature of the underlying mathematical formulation and the large number of functional domains in each and every cell in a SOFC stack.

2.3 Stack Modelling

Most research works on the fuel cells are about the development of single cells [118–120], which can only provide small power output. For power generation on a large scale, tens or hundreds of unit cells needs to be combined into high-performance cell stacks to increase the working voltage up to a necessary high value [2, 121]. For example, Forschungszentrum Jülich [122] built an anode-supported SOFC stack of 60 cells in 2004, which provided 13.3 kW with hydrogen as fuel and 11.9 kW with methane. It was the largest P-SOFC stack in the world at that time. However, in the simulation work, the majority of detailed mechanistic fuel cells models, which provide geometrical resolution and resolve the essential physics, still focused on the cell level [118–120, 123–127]. Only a few detailed mechanistic models were developed at the stack level, such as Refs. [128–137] for the PEMFCs and Refs. [138–140] for the P-SOFCs, which are limited to small stacks of up to around 5–10 cells.

Generally, the main concerns in stack design are pressure gradients and the uniformity of the distributions of current density, species, and temperature [90, 110]. Ideally, each unit cell in the stack should work under identical operating conditions to avoid non-uniformities between cells, which can weaken and reduce stack performance, reliability and/or durability. Typical non-uniformities between cells can occur in local performance, such as electric potential and current distributions between cells [141–146], the flow distribution at the fuel/air inlets due to external manifold design [147–150], and the temperature distribution [151, 152]. Aiming at reducing spatial variations in temperature and current density at the fuel source, Ayman et al. [153] proposed a novel stack design by introducing a mechanism of distributed fuel feed, in which the heat generation profile was directly influenced. Kulikovskiy [154] derived an approximate analytical expression for a rapid estimate on the temperature gradient along the air channel in a P-SOFC stack under real operating conditions.

Some models assumed that all the cells within the stack are identical, such that a unit cell was simulated to represent the distribution of dependent variables for the entire stack [2, 41, 155]. On the other hand, the variations in performance between the unit cells were investigated by some other authors such as Burt et al. [156]. The perturbations between cells may result from asymmetry properties in the cells, such as different inlet conditions from channel to channel in the unit cell and/or from cell to cell in the stack [156]. Although some authors used 3D models to predict the stack performance, they are confined to component level modelling, because detailed stack modelling in 3D is typically computationally expensive [2].

Mathematical modelling, which seeks to resolve the essential phenomena within a stack on a local level, is challenging, because it needs to consider coupled transport phenomena of mass, momentum, species, energy, and charge transfer in several or all of the length scales of the stack. As such, the largest stack model accounting for full geometrical resolution and a full set of conservation equations consisted of up to just 10 cells [137]. In order to model a stack of any size, reduced models were developed for the PEMFC stacks [148, 149, 157–165], direct methanol fuel cell stacks [166–169], and solid oxide fuel cell stacks [82, 139, 170–176]; however, they oversimplified the inherent physics and geometry. Modelling of larger stacks usually involves simplifications [82, 139, 148, 149, 163, 166–169, 171, 176–184] with a loss in the level of detail and resolution of the salient features of the electrochemical and transport phenomena.

2.4 Model Simplification

For fuel cell modelling, detailed 3D multiphysics models are computationally expensive in general, due to not only the highly coupled and nonlinear nature of the mathematical formulation but also a large number of functional domains, especially for stacks. Numerical simulation for fuel cell at the stack level usually accompanies with rigorous computational requirements.

In order to address this issue, two common assumptions to reduce the mathematical and computational complexity of the fuel cell modelling are discerned in literature. First, the geometry is simplified from 3D to 2D [14, 111, 185, 186], 1D [111, 185, 186], or 0D [17] counterparts. Second, the reaction zones comprising the TPB for the electrochemical reactions to take place are modeled as a boundary or interface condition, rather than a spatial domain, when predicting the global cell performance [14, 16, 44, 111, 112, 186, 187] or the local distribution of mass fraction, current density, and/or temperature [91, 92]. These assumptions are likely to lower the fidelity of model prediction for two reasons. Firstly, the flow fields of P-SOFC are generally 3D entities that consist of both plain channels and solid ribs. The ribs affect the diffusion pathways of gaseous working fluid, electronic charges, and heat between the flow field and the electrode. An improper design for the geometry of ribs and electrodes may cause the depletion of reactants to take place in the porous electrode underneath the ribs at high current densities. These effects of ribs are not captured by existing 2D P-SOFC models. This makes it difficult to achieve agreement between model predictions and experimental results at high current densities [80]. Secondly, dependent variables actually undergo spatial changes in the reaction zones that are spread out some distance (10–50 μm) into the electrode from the electrode/electrolyte interface [93, 94].

Instead of assuming the model dimensionality, researchers employed some methodologies to derived the reduced model. Roos et al. [69] applied the volume-averaging theory to omit the transverse direction in the flow field of a fuel cell and thus reduced a 3D flow field of channels and ribs to a 2D

counterpart. However, they did not formulate the effective parameters for the flow field counterpart or analyse the loss of information due to dimensional reduction. Billingham et al. [51] adopted the method of asymptotic expansions to construct solutions to a steady-state isothermal model for the combustion of methane in a loaded unit P-SOFC, such that the model was reduced from 3D to 2D. Only electrochemical flux and conservation of mass were taken into account, which means the model could not fully present the multiphysical phenomena such as the distribution of pressure, temperature, and mass fraction in the cell. Lately, Kulikovskiy [188] developed a simple model to predict heat and current transport in a stack element consisting of the fragment of interconnect with the single linear air channel, and constructed asymptotic solutions to the problem. The simplified model focused on heat and charge transfer only, such that the positive electrode-electrolyte-negative electrode (PEN) structure was assumed to be a thin interface generating heat, rather than detailed functional layers of the fuel cell.

2.5 Numerical Methods

The existing fuel cell models range from molecular/atomic level on half cell [74, 75, 115, 116] to single fuel cell level [19, 60, 91, 96, 117, 189, 190], stack-level [110, 117, 191, 192], and finally to system level [105, 193–195]. Micro-scale or nano-scale studies aim at the development of better electrodes through mathematical analysis considering detailed transport phenomena. Single-cell models (scale in 10^{-2} m) are often the first consideration for fuel cell manufacturers embarking on a new product design [2]. The results obtained from the single-cell models subsequently serve as the basis for stack-level and eventually for system-level simulations. Table 2.1 lists generally used computational methods according to the length scale of the model. For the macro-scale modelling, in particular for fuel cells, numerical solution methods like finite difference method (FDM), finite volume method (FVM), or finite element method (FEM) are proper approaches to discretize the governing equations.

The FEM is widely used in many engineering disciplines because of its advantages in the solution of multidisciplinary problems, which is crucial for fuel

Table 2.1 Computational methods for fuel cell modelling in different length scales [12]

Microscale ($\sim 10^{-9}$ m)	Meso-scale ($\sim 10^{-6}$ m)	Macro-scale ($>10^{-5}$ m)
Density functional theory (DFT)	Monte Carlo (MC)	Finite element method (FEM)
Quantum chemistry (QC)	Brownian dynamics (BD)	Finite volume method (FVM)
Lattice-Boltzman method (LBM)	Dissipative particle dynamics	Finite difference method (FDM)
Molecular dynamics (MD)	(DPD)	
Mechanistic models (MM)		

cell modelling. Chaisantikulwat et al. [61] implemented a 3D model in the FEM-based COMSOL (2005) to solve the coupled transport and electrochemical equations. Moreover, a combination of the thermo-fluid and structural models allows the thermo-mechanical modelling for fuel cells [89]. The FEM was also used by some researchers to solve meso-scale problems, although it was developed for phenomena occurring in the macro-scale. Huang et al. [196] used the COMSOL Multiphysics to investigate the multiphysics processes in the SOFC cathode-electrolyte interfaces at meso-scale. They took the detailed distribution and geometry of the ionic conducting phase, the electronic conducting phase, and the pores into account. Furthermore, some researchers applied both FVM and FEM in their works. Yakabe et al. [89] employed the FVM in the simulation of fluid flow phenomena including heat transfer, mass transfer, and chemical reactions in Star-CD, and the FEM for thermal stress analysis in a 3D unit P-SOFC model by the commercial code ABAQUS.

With the advancement of computer science and numerical techniques, it becomes possible to solve the equations for sophisticated geometrical design. Nevertheless, another problem in the development of numerical models is the validation of the simulation results against experimental data, due to the lack of such data in the available literature. The validation of existing detailed cell and stack models usually involve tuning or adapted parameters [12, 25, 26, 39, 61, 71, 76, 197], since these parameters are not available from the published experimental results. Furthermore, numerical simulation of fuel cells may come across a prohibitive requirement on computational source when solving a large 3D stack model which consists of tens or hundreds of cells.

2.6 Sensitivity Analysis

The performance of a fuel cell depends on both extrinsic operating conditions and intrinsic material properties and component geometries [2]. The effects of these factors on the global power output and local multiphysical mechanisms within the cell are generally studied via sensitivity analysis. The analysis is typically conducted by modelling with a few different values for every investigated modelling parameter and varying one parameter at a time [17, 77, 198–203]. The existing parametric studies generally aim at optimal values for the design parameters of fuel cells. However, optimal nominal solutions are not enough for a cell design to go into mass production, since the actual cell properties can deviate from the designed nominal value in high volume manufacturing processes. The influence of a combination of varying design parameters on the cell performance should be taken into account as well. Moreover, with just a few cases, it is difficult to perform deterministic analysis for a typically large population in manufacturing. Therefore, a method is required to not only find the input parameter showing the largest impact on results but also reveal what could possibly happen among a large population. Monte Carlo simulation (MCS) is one of such methods, based on generating a large

number of random samples. The MCS can provide insight on understanding the impact of each input factor on the variability of the output dependent variable, based on generating a large number of random samples for input factor.

The MCS can provide insight on understanding the impact of each input factor on the variability of the output dependent variable. Cornu and Wuillemin [204] performed the MCS of a computational fluid dynamics model to assess the impact of random geometric distortions on the fuel flow distribution in the anode channels of an SOFC. Their fluid dynamic model only solved for the fuel flow distribution in the channels under isothermal conditions without electrochemical reactions. The MCS is much more computationally expensive when solving a detailed electrochemical mechanistic model in 3D, in view of the highly coupled and nonlinear nature of the mathematical formulation and the large number of functional domains in the model as well as the large amount of runs. Pan et al. [205] conducted a MCS of a reduced SOFC model to analyse the sensitivity of output variables to operating conditions. Their MCS is based on a 1D reduced model that was developed by Lai et al. [179]. For the model reduction by Lai et al., the changes of physics along the width and through the thickness for each domain in the cell were assumed to be negligible. These assumptions were made according to simulation results predicted at a moderate cell voltage of 0.7 V [90]. However, these assumptions may become invalid at lower cell voltages or higher current densities. At high current densities, a large amount of reactants are consumed, resulting in large gradients in the distribution of dependent variables through the cell thickness and along the width [80, 206]. The depletion of species may even occur in the electrode at spanwise positions away from the channel due to the existence of the solid ribs that affect the pathways of transport processes. The impact of ribs should be taken into account when reducing the dimensionality of 3D SOFC models; otherwise, the fidelity of prediction will be weakened [16, 186, 187].

2.7 Remarks

This chapter has reviewed the existing research works on modelling and simulation of two typical kinds of the planar fuel cells: the PEMFC and P-SOFC. The planar fuel cells are typically equipped with parallel plain channels surrounded by solid ribs. The electrochemical and transport phenomena within the cell can be affected by geometrical structure and flow configuration, operating conditions, materials, and so on. According to the results of the present literature survey, four conclusions on the modelling of the planar fuel cells are drawn and highlighted below.

First, more mathematical models are developed in 2D than in 3D in recent years to predict the cell performance. Detailed 3D mechanistic models are challenging to solve numerically, because of not only the highly coupled and non-linear nature of the underlying mathematical formulation but also the large number of functional

domains, especially for stacks. In order to reduce the mathematical and computational complexity of modelling, the following two assumptions are generally imposed on the fuel cell models:

- The geometry is assumed to be in low dimensionality (such as 2D) instead of 3D. This assumption is likely to lower the fidelity of model predictions, because the influence of ribs on the pathways of transport processes is not taken into account. Spatial smoothing based on volume-averaging the flow field is a possible approach to handle the variation in the pathways for the fuel cells with parallel channels and slender sandwiched structure like the PEMFC and the P-SOFC. Nevertheless, existing spatially-smoothed models were only developed for single cells under isothermal conditions, but did not formulate the effective parameters for the flow field counterpart or analyse the loss of information due to dimensional reduction, and did not account for the coupling effects between the governing equations for transport phenomena.
- The reaction zones comprising the triple-phase boundaries for the electrochemical reactions to take place are modeled as a boundary or interface condition rather than a spatial domain, when predicting the global cell performance or the local distribution of mass fraction, current density, and/or temperature. However, dependent variables actually undergo spatial changes in the reaction zones that can spread out some distance (10–50 μm) into the electrode from the electrode/electrolyte interface.

Second, the existing 2D fuel cell models are generally developed with respect to the thickness, and streamwise direction along the cell length and the normal direction through the thickness, and neglect the physics in the spanwise direction along the cell width, in which dependent variables may undergo leading-order changes at high current densities. The information about the spanwise variability of dependent variables is necessary for cell design in order to avoid undesired cell performance such as depletion of reactants underneath the ribs.

Third, the majority of computational studies in the open literature focus on a single cell or cell components, whereas relatively fewer are found for stacks, although there is a trend to raise the study from the cell to stack level. When seeking for a detailed description of the transport phenomena and electrochemical characteristics in the stack, 3D stack models are computationally expensive to solve. Moreover, the perturbation or non-uniformity in the performance of the cells in the stack needs to be considered.

Last, parametric studies on cell performance are generally performed by varying a single modelling parameter with a few testing cases. In order to adopt statistical measures to analyse the sensitivity of cell and stack performance to varying parameters, efficient and accurate reduced models are needed for the simulation of a cell or stack with a large population of testing cases without a prohibitive computational cost.

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