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Gas Transport in Solid Oxide Fuel Cells



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Preface

The ultimate goal of this book is to provide an integrated view of the basic theory, materials science, and engineering of gas transport in solid oxide fuel cells (SOFCs). Further, this book will provide an invaluable, contemporary reference for the development of fundamental theory and experiment, advanced experimental measurement techniques, and industrial applications of gas diffusivity in solid oxide fuel cells.

Interest in fuel cell technologies has been motivated by their function: directly converting stored chemical energy into electrical energy without combustion and emission of pollutants, such as nitrogen oxides (N_xO_y). These devices can overcome combustion efficiency limitations since the operation of fuel cells does not necessarily involve the Carnot cycle, thus reducing the emission of pollutants. Compared with other types of fuel cells, solid oxide fuel cells have shown clear advantages over other systems, since hydrogen, hydrocarbons, carbon monoxide, and carbon can be utilized as constituent fuels. The major disadvantage of SOFCs is their high operation temperature, which can reach 1000 °C. At such high temperatures, few materials can function effectively as electrolytes or electrodes. This feature of SOFCs increases their operation and fabrication costs, and hinders their application in rapidly developing areas of application, such as in portable power and automobile power device applications. The impedance of SOFCs, including the activation and concentration polarizations of electrodes and the Ohmic loss of electrolytes, increases sharply with decreasing operating temperatures. To reduce the impedance, fundamental comprehension of the mechanism of gas diffusion through the electrode and that of gas transport between the electrode and the electrolyte is necessary. Mechanisms and mathematical models of gas diffusion are discussed in detail in the first chapter of this book.

Several techniques for directly measuring gaseous diffusivity have been developed in recent years. These techniques allow gas transport coefficients to be accurately evaluated. The results of these measurements help to optimize the configuration of solid oxide fuel cells, including the surface properties of electrodes and the structure of electrodes and electrolytes, as well as the techniques for preparing electrolytes. Recent theoretical and experimental advancements in these measurement techniques are discussed in the middle chapters of this book.

Gas diffusivity of electrodes in solid oxide fuel cells drops rapidly with reducing operation temperatures. This loss of diffusivity cannot be compensated through the optimization of the configuration of the fuel cell. Therefore, the key to lowering the operation temperature of solid oxide fuel cells is the development of high-efficiency electrodes. The role of gas diffusivity measurement techniques in the exploration of novel electrode materials are also explored in the middle chapters of this book. Then, the book focuses on the strategies of realizing advanced solid oxide fuel cells with improved gas transport. This chapter presents an overview of novel porous electrode materials, and the techniques allowing for the rational design of electrode microstructure with highly efficient gas transport parameters, including porosity, tortuosity, etc. Finally, an outlook on research and development of low-temperature solid oxide fuel cells is presented.

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Nomenclature

E	Potential (V)
E^o	Equilibrium redox potential (V)
E_{ocp}	Open circuit potential (V)
T	Absolute temperature (K)
η_{ohm}	Voltage drops due to ohmic polarization losses (V)
η_{act}	Voltage drops due to activation, losses (V)
η_{con}	Voltage drops due to concentration polarization losses (V)
K_n	Knudsen number
d_p	Diameter of the pore (cm)
d_g	Effective diameter of a gas molecule (cm)
λ	Gas mean free path (cm)
k_B	Boltzmann constant (1.3807×10^{-23} J/K)
P	Gas pressure (Pa)
P_t	Total gas pressure (Pa)
D_i	Bulk diffusivity of gas species i (cm^2/s)
D_{ij}	Binary diffusivity of gas i and j (cm^2/s)
D_i^t	Total diffusivity of species i (cm^2/s)
D_{ij}^{eff}	Effective binary diffusivity of gas i and j (cm^2/s)
D_{iK}	Knudsen diffusivity of gas i (cm^2/s)
$D_i^{t,eff}$	Total effective diffusivity (cm^2/s)
ϕ	Porosity
T	Tortuosity
R	Gas constant (8.314 J/(mol K))
c	Total gas molar concentration (mol/L)
c_i	Molar concentration of gas i (mol/L)
μ_i	Chemical potential of species i (J/mol)
μ_0	Standard chemical potential of gas i under 1 atm and 1 mol/L (J/mol)
M_i	Gas molecular weight (g/mol)
Ω	Collision integral
σ_{12}	Collision diameter (angstrom)
k_i	Effective permeability (m^2)
k_0	Absolute permeability (m^2)

μ_g	Gas viscosity (kg/(m·s))
ρ_g	Gas density (kg/m ³)
ω_i	Mass fraction of gas species i
X_i	The mole fraction of gas species i
C	Total concentration (mol/L)
J	Total net gas transport (mol/(m ² s))
J_i	Molar flux of gas species i (mol/(m ² s))
J_j^D	Diffusive molar flux of gas species j (mol/(m ² s))
J_j^T	Total diffusive and advective molar flux (mol/(m ² s))
\bar{D}_2	Simplified diffusivity, (cm ² /s)
\bar{c}_2	Simplified molar concentration (mol/L)
\bar{k}_2	Simplified permeability (m ²)
F	Faraday constant (96485.3 C/mol)
l_a	Anode thickness (m)
L_c	Cathode thickness (m)
$p_{H_2}^i$	H ₂ Pressure inside YSZ tube (Pa)
$p_{H_2}^o$	H ₂ Pressure outside YSZ tube (Pa)
$p_{O_2}^i$	O ₂ Pressure inside YSZ tube (Pa)
$p_{O_2}^o$	O ₂ Pressure outside YSZ tube (Pa)
$p_{H_2O}^i$	H ₂ O Pressure inside YSZ tube (Pa)
$p_{H_2O}^o$	H ₂ O Pressure outside YSZ tube (Pa)
D_x	Diffusivities along x electrode direction (cm ² /s)
D_y	Diffusivities along y electrode direction (cm ² /s)
D_z	Diffusivities along z electrode direction (cm ² /s)
D_s	Summed 3D diffusivity (cm ² /s)
R_i	Ohmic resistance (Ω /cm ²)
i_o	Exchange current density (A/m ²)
i_a	Anode limiting current density (A/m ²)
i_c	Cathode current density (A/m ²)
η_a	Anode concentration polarization (V)
η_c	Cathode concentration polarization (V)
τ_a	Anode tortuosity
τ_c	Cathode tortuosity
ϕ_a	Anode porosity
ϕ_c	Cathode porosity
Δi	Current error (A/m ²)
ΔD	Diffusivity error (cm ² /s)
Δi_a	Anode limiting current density error (A/m ²)
ΔT	Temperature error (K)
Δi_c	Cathode current density error (A/m ²)
η_a	Anode concentration polarization error (V)
η_c	Cathode concentration polarization error (V)
D_{A-mix}^{eff}	Effective diffusivity of A in a multicomponent gas mixture (cm ² /s)
A	Electrode area (m ²)
R_D	Gas diffusion resistance (Ω /cm ²)

Abbreviations

SOFC	Solid oxide fuel cell
YSZ	Ytria-stabilized zirconia
PEMFC	Proton exchange membrane fuel cell
MCFC	Molten carbonate fuel cell
SHE	Standard hydrogen electrode
AL	Activation loss
OL	Ohmic loss
CP	Concentration polarization
OCP	Open circuit potential
EMF	Electromotive force
FL	Fick's law
ADM	Advective–diffusive model
MSM	Maxwell–Stefan model
DGM	Dusty gas model
BFM	Binary friction model
LCD	Limiting current density
Ni-YSZ	Porous composites of Ni and YSZ
LSM	Sr-doped LaMnO ₃
GDC	Ce _{0.9} Gd _{0.1} O _{1.95}
SDC	Ce _{0.9} Sm _{0.1} O _{1.95}
ESB	Er _{0.4} Bi _{1.6} O ₃
LSGM	La _{0.9} Sr _{0.1} Ga _{0.8} Mg _{0.2} O ₃
TPB	Triple phase boundary
MIEC	Mixed ionic and electron conductivity
LSM	La _{0.8} Sr _{0.2} MnO _{3-δ}
LSCF	La _{0.6} Sr _{0.4} Co _{0.2} Fe _{0.8} O _{3-δ}
SSC	Sm _{0.5} Sr _{0.5} CoO _{3-δ}
BSCF	BSCF (Ba _{0.5} Sr _{0.5} Co _{0.8} Fe _{0.2} O _{3-δ})
LT-SOFC	Low temperature Solid oxide fuel cell
TEC	Thermal expansion coefficient
SEM	Scanning electron microscopy
TEM	Transmission electron microscopy

BET	Brunauer–Emmett–Teller method
FIB	Focused ion beam
FIB-SEM	Focused ion beam—scanning electron microscopy
TXM	Transmission X-ray microscopy
RVE	Representative volume element
MPD	Maximum power density
EIS	Electrochemical impedance spectroscopy
STN	Nb-doped SrTO ₃
ScYSZ	Sc doped YSZ

Chapter 1

Introduction to Gas Transport in Solid Oxide Fuel Cells

Solid oxide fuel cells (SOFCs) produce electricity by oxidizing fuel gases. The biggest characteristic of SOFCs is their high energy conversion efficiency, up to 60–80 % in theory, since the conversion efficiency is not limited by the Carnot cycle due to the absence of combustion in SOFC devices. Other advantages of SOFCs include fuel flexibility, low emission, long-term stability, and relatively low cost. The major challenge associated with SOFCs is their high operating temperatures, typically above 500 °C. For SOFCs to find a large range of applications for electricity generation in the twenty-first century, numerous efforts are needed to lower the operating temperatures and to enhance the practical conversion efficiency at moderate operating temperatures. The key is to improve mass transfer involved in SOFCs. Since SOFCs operate with gaseous fuels and oxidants, gas transport in the porous electrodes largely influences their performance. In this chapter, a brief introduction to SOFCs will be first given. The main issues to be solved, gas transport phenomenon, as well as the scientific problems in this field will then be depicted in later chapters.

1.1 Introduction to SOFCs

1.1.1 Brief History of SOFC Development

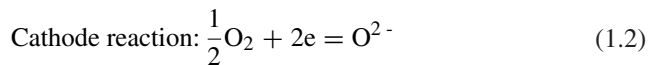
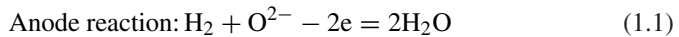
Solid oxide fuel cells were originally realized by Nernst for use as a commercial light source, in an effort to replace carbon filament lamps in the 1900s [1]. The device used Nernst mass, which was made of yttria-stabilized zirconia (YSZ), a conductor of oxide ions in the air. This system operated at temperatures from 600 to 1,000 °C. The electrolyte composition, found in Nernst mass, is still the basis of the commonly used electrolyte. In 1937, Baur et al. [2] developed the first solid oxide fuel cells that used materials, such as zirconium, yttrium, cerium, lanthanum, and tungsten; since then, solid O^{2-} conductor-based SOFCs have attracted increasing attention. In the 1940s, a Russian scientist, O.K. Davtyan, added monazite sand to the mixture of sodium carbonate, tungsten trioxide, and soda glass to increase the conductivity and mechanical strength. However, Davtyan's designs could solve

the problems including unwanted chemical reactions and short life ratings. In the late 1950s, research on solid oxide technology began to accelerate at the Central Technical Institute in the Hague, Netherlands, Consolidation Coal Company, in Pennsylvania, and General Electric, in Schenectady, New York. A 1959 discussion of fuel cells noted that problems with solid electrolytes included relatively high internal electrical resistance, melting, and short-circuiting due to semiconductivity. Apparently, many researchers began to believe that molten carbonate fuel cells showed more pronounced short-term promise. Nevertheless, the excellent CO tolerance and the long-term stability still draw the attention of researchers whose focus is on the improvement of SOFC performance for space, submarine, and other military applications. More recently, the emerging energy crisis (climbing energy prices, environmental problems, and advances in materials technology) has reinvigorated work on SOFCs. Numerous companies, universities, and research agencies all over the world are now working in this field.

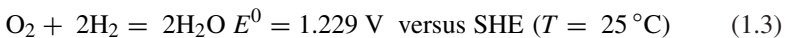
1.1.2 Principles of SOFCs

As the global population and economy continues to expand, much attention is focused on improving the performance of existing energy systems as well as exploring new forms of sustainable energy sources [3–5]. One of the main proposed strategies toward sustainable energy sources is hydrogen-based fuel cells, such as proton exchange membrane fuel cells (PEMFCs), molten carbonate fuel cells (MCFCs), and solid oxide fuel cells (SOFCs). Fuel cells directly convert stored chemical energy of fuels into electrical energy without combustion and, thus, are capable of overcoming the combustion efficiency limitations as imposed by the Carnot cycle. Moreover, fuel cells reduce the emission of pollutants like nitrogen oxides (N_xO_y) and are environmentally friendly. Compared with other types of fuel cells, solid oxide fuel cells exhibit excellent fuel flexibility, since many chemicals, including hydrogen, hydrocarbons, carbon monoxide, and carbon can be utilized as fuels [6–9].

In an SOFC, as shown by the scheme in Fig. 1.1, oxygen molecules diffuse through the cathode and are reduced to oxygen ions at the cathode active layer. These oxygen ions then transport through a solid ion-conductive electrolyte and react with the fuel gas at the anode/electrolyte interface. The driving force of a fuel cell is provided by the anode/cathode reaction as shown in Eq. 1.1 and Eq. 1.2.



The overall reaction of an SOFC is shown in Eq. 1.3



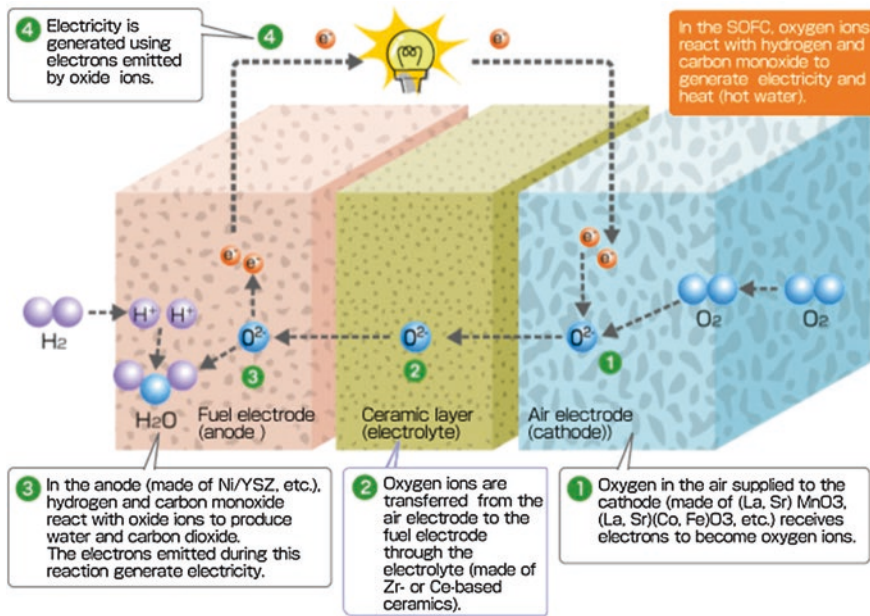


Fig. 1.1 Scheme of the operation principle of an SOFC. SOFCs use a solid oxide electrolyte to conduct negative oxygen ions from the cathode to the anode. The solid oxide electrolyte, usually made of doped ZrO_2 or CeO_2 ceramics, is adequately ionicly conductive for O^{2-} only at high temperatures, typically between 500 and 1,000 °C. Adopted from Web site: <http://www.osakagas.co.jp/en/rd/fuelcell/sofc/sofc/index.html>

The equilibrium potential of the overall reaction E^0 is 1.229 V *versus* SHE at the room temperature (25 °C), which decreases linearly as the temperature increases with a rate of 23 mV per 100 K. For SOFCs operated at 800 °C, the theoretical open potential is ~1.050 V *versus* SHE [10].

Practical SOFCs are usually fabricated in either planar or tubular structures as shown in Fig. 1.2. Both cell setups exhibit their merits and drawbacks. The advantages of planar cells include construction simplicity, lower fabrication cost, lower ohmic resistance, and higher power density compared with tubular cells. The advantages of tubular cells include no need for high-temperature sealing (the cell can be sealed in the cold ends), and long-term operation stability without obvious decay (the decay rate is only ~0.1 % per 1,000 h). Both designs of SOFCs have been developed extensively by research agencies and industrial communities all over the world.

Solid oxide fuel cells have a large variety of applications ranging from portable power and transport to stationary power supply with outputs from 100 W to 2 MW. Research has led to remarkable success in reducing the energy losses of SOFCs well below 30 %. The major drawback of SOFCs is their high operating temperatures, which are typically above 500 °C. At these temperatures, only a few

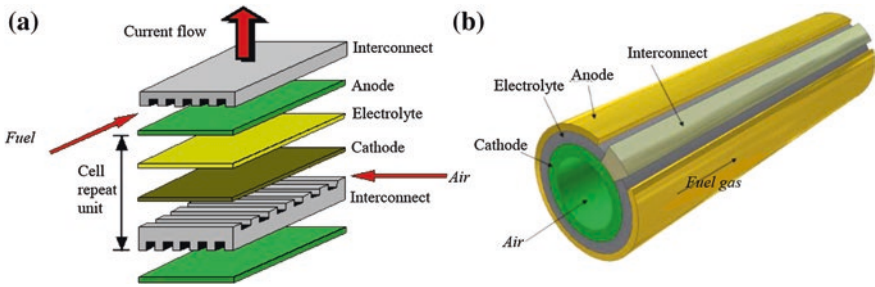


Fig. 1.2 SOFCs with structures of **a** planar cell and **b** tubular cell. Adopted from Web site: <http://www.csa.com/discoveryguides/fuecel/overview.php>, and <http://de.wikipedia.org/wiki/Festoxidbrunnstoffzelle>

materials can be employed as electrolytes and electrodes. This leads to an increase in cost and hinders potential applications in portable and automobile power sources. Lowering the operation temperatures of SOFCs while maintaining low energy losses has become the key focus in SOFC research [11]. Development of new electrolytes with high ionic conductivities and new electrode materials with high activities at low temperatures (300–600 °C) are paving the way for this direction. In addition to materials development, the rational design of the structure with existing SOFC materials also can greatly decrease various energy polarizations, especially the energy loss caused by gas transport in the operation of SOFCs, and facilitate the operating-temperature-lowering efforts of SOFCs.

1.1.3 Energy Losses in SOFCs

For a gas-based SOFC to operate, the activation energy of the anode/cathode reaction must be overcome [12]. In the process of producing power, the pressure gradient of anode/cathode gas and the concentration gradient of oxygen ions across the anode/cathode and the electrolyte are determined by the transport rates of these gaseous and ionic species. While providing electricity, electrical resistance also is present in all operating components of the fuel cell [13]. These energy losses associated with a fuel cell are divided into three types: activation loss (AL), concentration polarization (CP), and ohmic loss (OL). Figure 1.3 demonstrates the correlation between concentration polarization loss, ohmic loss, and activation loss with different components of a fuel cell. Ohmic loss, which results from electrical resistance, is present across all fuel cell components, whereas activation loss is induced by offsetting energy barrier for catalytic reactions at electrode/electrolyte interfaces. Concentration polarization is induced by the pressure gradient due to limited transport rates of gaseous reactant and/or product species through SOFC electrodes [14].

The cell voltage, E , can be calculated by Eq. 1.4,

$$E = E_{\text{ocp}} - \eta_{\text{ohm}} - \eta_{\text{act}} - \eta_{\text{con}} \quad (1.4)$$

Fig. 1.3 Energy losses associated with SOFC components. Reprinted from Ref. [15]. Copyright (2013), with permission from Elsevier

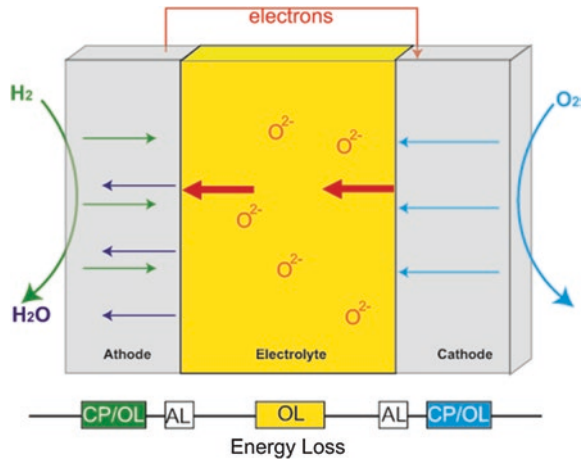
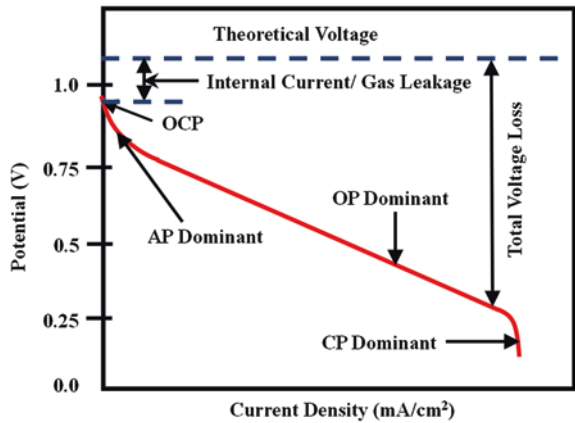


Fig. 1.4 Ideal and actual fuel cell current–voltage characteristics and polarization losses at each region



where E_{ocp} is the open circuit potential (OCP) and η_{ohm} , η_{act} , and η_{con} are the voltage drops due to ohmic, activation, and concentration polarization losses, respectively. In principle, E_{ocp} should be the same as the reversible Nernst potential of the reactants, but in many practical cases, the measured OCP is less than the ideal voltage due to leaky gas seals and open pores/micro-cracks in the electrolyte. As shown in Fig. 1.4, for SOFCs working at low output current densities, the activation loss dominates the overall potential drop. The ohmic energy loss increases nearly linearly as the current density increases. At a relatively large output current density, the potential of the cell decreases more rapidly, which indicates the dominance of the concentration polarization. A large output of the current density requires fast consumption of reactant gases and exhaustance of products, exceeding the gas transport rate in the electrodes. Therefore, concentration energy loss should be minimized to increase the energy conversion efficiency and to improve the performance of SOFCs under operation modes of large output current densities.

Porous electrodes are usually employed in SOFCs to improve the gas transport. Factors such as the porosity, pore size, and tortuosity of the electrodes should be optimized to improve the performance of gas transport.

1.2 Gas Transport in SOFCs

1.2.1 General Consideration

In an SOFC system, to produce electricity, continuous supply of fuels and oxidants is required. In the meanwhile, the reaction product must be exhausted outward. The motion of reactant and product species is called mass transport in SOFCs. Although the transport of the oxygen ions in the electrolyte is also a major portion of mass transport, we focus primarily on the mass transport of the uncharged species. The uncharged species are typically gaseous fuels and oxidants, and their transport is gas transport. The motion of the uncharged gases is not affected by the potential gradient and must rely on the diffusion and convection. The gas transport occurs in two areas of SOFC systems—the flow field and the electrode as shown in Fig. 1.5a. The flow field is channels of the size on millimeter-to-centimeter scale, and the gas transport in this area is controlled by convection as driven by the external pressure of the gas sources. On the contrary, the electrodes of SOFCs are porous structures with the pore size on nanometer-to-micrometer scale, and

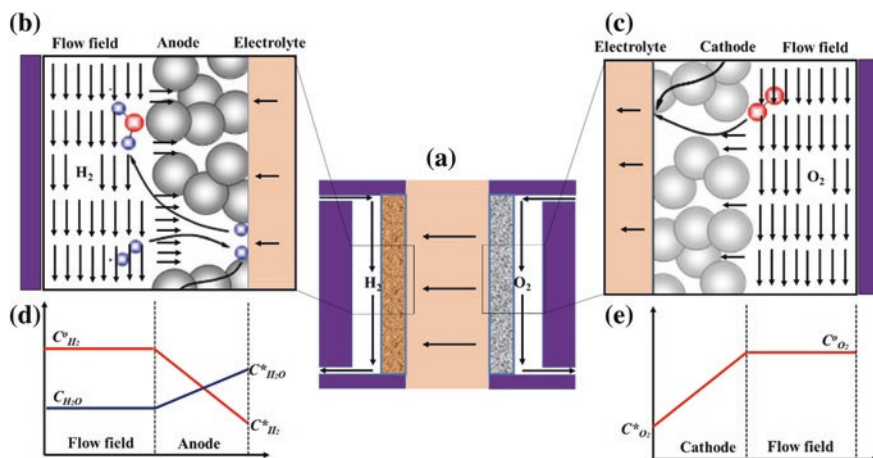


Fig. 1.5 The scheme of gas transport in **a** the H₂–O₂ SOFC system and the diffusion layer, **b** on the anode surface, and **c** on the cathode surface. The consumption of H₂(O₂) at the anode (cathode)–electrolyte interfaces results in the depletion of H₂(O₂) in the anode (cathode). The concentration of H₂(O₂) decreases from the bulk concentration to a lower concentration, as shown in **d–e**. The exhaust of the product of H₂O at the anode is also shown in **d**. At the flow field/anode interface (cathode), the flow rate of H₂(O₂) decreases to zero, which indicates the start point of the diffusion layer