## Lecture Note #9 (Spring, 2022)

# Fuel cell Fundamentals

- 1. Basic of fuel cells
- 2. Types of fuel cells
- 3. I-V characteristics and polarizations
- 4. Electrode structure
- 5. Proton-exchange membrane (PEM) fuel cells
- 6. Solid acid fuel cells

# Fuel cells (연료전지)

-Fuel cell: energy conversion device (cf. battery: energy storage device)  $\rightarrow$  to convert the chemical energy of a fuel directly into electricity

-Operation of fuel cell e.g. direct methanol fuel cell (DMFC)

 $\begin{array}{ll} \mathsf{CH}_3\mathsf{OH} + \mathsf{H}_2\mathsf{O} \to \mathsf{CO}_2 + 6\mathsf{H}^+ + 6\mathsf{e}^- & \text{anode} \\ 1.5\mathsf{O}_2 + 6\mathsf{H}^+ + 6\mathsf{e}^- \to 3\mathsf{H}_2\mathsf{O} & \text{cathode} \\ \mathsf{CH}_3\mathsf{OH} + 1.5\mathsf{O}_2 \to \mathsf{CO}_2 + 2\mathsf{H}_2\mathsf{O} & \text{overall} \end{array}$ 

-Fuels: H<sub>2</sub>, methanol, and so on

-Fuel cells are not thermal devices and are not limited by the Carnot efficiency of heat engines



Figure 9.1 Direct methanol fuel cell that uses an acid electrolyte.

Galvanic cell: negative, positive, anode, cathode

✓Fuel cell



#### Oh, ch.1



### Principle of Fuel Cell



Fuel cell : hydrogen + oxygen [air)  $\rightarrow$  water + <u>electric energy</u> Respiration : glucose + oxygen (air)  $\rightarrow$  carbon dioxide+ water + <u>energy</u>

## Types of fuel cell



# **Bio-fuel cell**

"An electro-chemical device in which energy derived from chemical reactions maintained by a continuous supply of chemical reactants is converted to electrical energy by means of the catalytic activity of **living cells and/or their enzymes**."



electrode: enzyme, bacteria...

## History of fuel cell

- 1839 Discovery of fuel cell (William Grove)
- -~1960 NASA, fuel cell in spacecraft
- -~1984 Development of fuel cell vehicle
- -~2000 Development of portable fuel cell







#### The Fuel Cell in Apollo 11

Size	111.8 X 55.9 cm
Weight	181.4kg
Cell output	2,300W max

- Each of the fuel cell power plants contain 31 separate cells connected in series

- Each cell produces 27 to 31 volts.

\* Source : PowerNational Air and Space Museum

### Hydrogen fuel cell vehicle 460 km driving distance

Power 113 kW Power density 3 kW/*I*, 2kW/kg FC weight 56 kg + (5 kg H<sub>2</sub> in tank (87 kg)) Efficiency 6 km/kWh



cf. Gasoline engine: ~800 km, 60~70 liter tank, 12 km/liter





Figure 7.7 Effect of rate on potential as a function of the fractional capacity for a lead–acid cell.

1.7 .....

*Electrochemical Engineering*, First Edition. Thomas F. Fuller and John N. Harb. © 2018 Thomas F. Fuller and John N. Harb. Published 2018 by John Wiley & Sons, Inc. Companion Website: www.wiley.com/go/fuller/electrochemicalengineering

0.0 0.1 0.2 0.3 0.4 0.5 0.6 0.7 0.8 0.9 1.0 Capacity

#### $\rm CH_3OH + 1.5O_2 \rightarrow \rm CO_2 + 2H_2O$

### ✓ Thermodynamics

-Equilibrium potential (E<sup>0</sup> or U<sup>0</sup>) for the methanol fuel cell using appendix C (thermodynamic data)  $\Delta G^{0} = \Delta G^{0}_{f,CO2} + 2\Delta G^{0}_{f,H2O} - \Delta G^{0}_{f,CH3OH} = -394.359 + (-2 \times 237.129) - (-166.6) = -702.3306 \text{ kJ/mol}$ 

 $E^{0} = -\Delta G^{0} / nF = (702,330.6 \text{ J/mol}) / (6 \times 96,485 \text{ C/mol}) = 1.21 \text{ V}$ 

-Change of enthalpy:  $\Delta H^0 = \Delta H^0_{f,CO2} + 2\Delta H^0_{f,H2O} - \Delta H^0_{f,CH3OH} = -726.5 \text{ kJ/mol} (large exothermic)$ 

 $\Delta H = Q - W$ 

Q: heat transfer to the system W: electrical work done by the system

 $\Delta G$  (or E): maximum electrical work



**Figure 9.2** Fuel cells are open systems with reactant streams entering and exiting the fuel cell.

Species	Molecular Formula		$\bar{g}_{f}^{\circ}$ (kJ/kmol)	$\bar{s}^{\circ}$ (kJ/kmol · K)	
Carbon	Cs	0	0	5.74	
Hydrogen	H <sub>2,g</sub>	0	0	130.57	
Nitrogen	N <sub>2,g</sub>	0	0	191.50	
Oxygen	O <sub>2g</sub>	0	0	205.03	
Carbon monoxide	COg	-110,530	-137,150	197.54	
Carbon dioxide	CO <sub>2,g</sub>	-393,520	-394,380	213.69	
Water vapor	$H_2O_g$	-241,820	-228,590	188.72	
Liquid water	$H_2O_1$	-285,830	-237,180	69.95	
Hydrogen peroxide	$H_2O_{2,g}$	-136,310	-105,600	232.63	
Ammonia	NH <sub>3,g</sub>	-46,190	-16,590	192.33	
Hydroxyl	OHg	39,460	34,280	183.75	
Methane	CH <sub>4,g</sub>	-74,850	-50,790	186.16	
Ethane	$C_2H_{6,g}$	-84,680	-32,890	229.49	
Propane	$C_3H_{8,g}$	-103,850	-23,490	269.91	
Octane vapor	C <sub>8</sub> H <sub>18,g</sub>	-208,450	17,320	463.67	
Octane liquid	C <sub>8</sub> H <sub>18,1</sub>	-249,910	6,610	360.79	
Benzene	$C_6H_{6,g}$	82,930	129,660	269.20	
Methanol vapor	CH <sub>3</sub> OH <sub>g</sub>	-200,890	-162,140	239.70	
Methanol liquid	CH <sub>3</sub> OH <sub>1</sub>	-238,810	-166,290	126.80	
Ethanol vapor	C <sub>2</sub> H <sub>5</sub> OH <sub>g</sub>	-235,310	-168.570	282.59	
Ethanol liquid	$C_2H_5OH_1$	-277,690	-174,890	160.70	

Table 3.3         Enthalpy of Formation, Gibbs Energy of Formation	ormation, and Entropy Values at 298 K, 1 atm
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Source: From [1].

#### Mathew M. Mench, Fuel Cell Engines, Wiley, 2008.

#### ✓ Efficiency of the fuel cell

= the electrical work produced / the energy available in the reactants

 $\rightarrow$  operate with the continuous flow of fuel and oxidant

 $\rightarrow$  rates of energy (power)

#### The voltage efficiency of a fuel cell

 $\eta_V^{fc}$  = rate of electrical energy from cell / maximum rate of electrical energy

- = electrical power from cell / theoretical power from cell
- = IV /  $[(\dot{m}_i / M_i)(-\Delta G_{rx})] = IV / [(I/nF)(nFE)] = V/E$

Assuming m<sub>i</sub> [kg·s<sup>-1</sup>] of reactants

 $\rightarrow$  the cell voltage to be as high as possible in order to maximize efficiency  $\rightarrow$  minimization of ohmic, kinetic, mass-transfer losses is desired

#### Illustration 9.1

Hydrogen fuel cell operating at 0.7 V. voltage efficiency?  $\rightarrow \eta_V^{fc} = 0.7 / 1.229 = 0.57$  (or 57%)



Figure 3.13 Comparison of maximum thermodynamic efficiency for heat engine (Carnot cycle) and fuel cell engine (hydrogen fuel cell, HHV assumed).

- At higher  $T \rightarrow$  heat engine theoretically more efficient (not always efficient for a fuel cell)
- FC shows a decreasing efficiency with T

Mathew M. Mench, Fuel Cell Engines, Wiley, 2008.

## Types of fuel cells

### -Types by the electrolyte used

#### Table 9.1Types of Fuel Cells

ation and design of the fuel cell depend dramatically on the nature of the electrolice. For example, while hydrogen always reacts at the anode and oxygen at the cathode, water can be orgitized at differ the Augde of the strigggen of the cathode, water

Fuel-cell type	el-cell type Main application temperature [°C]		Comments of the about of the here bounded of the
Direct methanol	Portable power	8.0 25–90	Uses same membrane as PEM FC
Proton-exchange membrane (PEMFC)	Automotive, buses portable	60–90	Tolerant to carbon dioxide in air Requires precious metal catalysts Rapid start-up and shutdown
Alkaline (AFC) $\rightarrow$ Anion-exchange I	Space membrane (AEM)	80–100	Requires pure hydrogen and oxygen Nonprecious metal catalysts possible
Phosphoric acid (PAFC)	Stationary, combined heat and power	180–220 <sub>abori</sub> ) ant da	Operates on reformed fuels Long life Some cogeneration possible
Molten carbonate (MCFC)	Stationary, combined heat and power	600–650	High efficiency Good cogeneration
Solid oxide (SOFC)	Stationary, combined heat and power	650–1000	High efficiency High temperature limits materials available and makes thermal cycles challenging

Table 9.2	Anode and Cathode	Reactions for	r Different	Types	of Fuel C	Cells	Operating	on Hydrogen	and	Oxygen
8 61 8 9 1 V / a feet	I Mode and Caulous	TTAAAAA ATATAA TAA					*			

Fuel-cell type	Anode	four silicate et :	Cathode	i budsv
Alkaline Acid Oxygen conducting ceramic, solid oxide	$H_2 + 2OH^- \rightarrow 2H_2O + H_2 \rightarrow 2H^+ + 2e^-  (0.7)$ $O^{2-} + H_2 \rightarrow H_2O + 2e^-$	- 2e <sup>-</sup> (-0.828 V) V)	$O_2 + 2H_2O + 4e^- \rightarrow 4OH^-$ $4H^+ + 4e^- + O_2 \rightarrow 2H_2O$ (1) $O_2 + 4e^- \rightarrow 2O^{2-}$	(0.401 V) 1.229 V)

## Pourbaix diagrams

 $E^0 = -\Delta G^0 / nF$ = (237,180 J/mol) / (2 x 96,485 C/mol) = 1.229 V

at 25°C, pH =  $-\log_{10}a$ 

 $2H^+ + 2e^- = H_2$ 

 $E = E^{0} - (RT/2F)ln(1/a_{H+}^{2})$ = 0 + (RT/F) ln(a\_{H+}) = -(RT/F)(2.303pH) = -0.0592pH

 $(1/2)O_2 + 2H^+ + 2e^- = H_2O$ 

 $E = E^{0} - (RT/2F)In(1/a_{H^{+}}^{2})$ = 1.229 + (RT/F) In(a\_{H^{+}}) = 1.229 - (RT/F)(2.303pH) = 1.229 - 0.0592pH



-Overall H<sub>2</sub>-O<sub>2</sub> fuel cell reaction:  $H_2 + 1/2O_2 \rightarrow H_2O$   $E_0 = 1.229 \text{ V}$ -In <u>acidic electrolyte</u>  $\rightarrow$  PEM, PAFC

anode:  $H_2 \rightarrow 2H^+ + 2e^ E_0 = 0.000 V$ cathode:  $1/2O_2 + 2H^+ + 2e^- \rightarrow H_2O$   $E_0 = 1.229 V$ Total:  $H_2 + 1/2O_2 \rightarrow H_2O$   $E_0 = 1.229 V$ 

-In <u>alkaline electrolyte</u>  $\rightarrow$  Alkaline, Anion-exchange membrane (AEM)

anode:  $H_2 + 2OH^- \rightarrow 2H_2O + 2e^- \quad E_0 = -0.828 \text{ V}$ cathode:  $1/2O_2 + H_2O + 2e^- \rightarrow 2OH^- \quad E_0 = 0.401 \text{ V}$ Total:  $H_2 + 1/2O_2 \rightarrow H_2O \qquad E_0 = 1.229 \text{ V}$ 

-Hydrogen oxidation reaction(HOR): not single step → elementary steps -Oxygen reduction reaction(ORR): reaction steps still unknown

### **ORR Electrocatalysis**



Qingyu Li et al, Electrochemical Energy Reviews 2(2019)518

Dong Young Chung *et al*, Adv. Mat. (2018)

## I-V and polarization

-Example polarization curves of 3 types of fuel cells

Galvanic cell:  $I\uparrow \rightarrow V\downarrow$ 

Understanding using thermodynamics (open circuit voltage), Kinetics (low-current behavior), ohmic losses (moderate-I behavior), mass transfer (high-I behavior)



**Figure 9.3** Example polarization curves for PEM, AFC, and SOFC.

#### Fuel cell polarization curve



M. M. Mench, Fuel Cell Engines, Wiley, Fig. 4.1

-5 regions

(i) Region I: activation (kinetic) overpotential at the electrodes
(ii) Region II: ohmic polarization. Electric & ionic conduction loss
(iii) Region III: concentration polarization. Mass transport limitations
(iv) Region IV. Departure from Nernst thermodynamic equilibrium potential; undesired species crossover through electrolyte, internal currents from electron leakage, other contamination or impurity

(v) Region V: the departure from the maximum thermal voltage; a result of entropy change which cannot be engineered

-Regions I, II, III are not discrete  $\rightarrow$  all modes of loss contribute throughout the entire current range

-Heat: current x (thermal voltage – FC voltage)

& electrical generation: cell current x FC voltage -FC voltage:

$$E_{\text{cell}} = E^{\circ}(T, P) - \eta_{a,a} - |\eta_{a,c}| - \eta_r - \eta_{m,a} - |\eta_{m,c}| - \eta_x$$
(4.1)







Seung Mo Oh, Electrochemistry(3<sup>rd</sup> ed.) (2019)

### (1) Thermodynamics

-Thermodynamics  $\rightarrow$  open circuit voltage (OCV at zero current density)  $\rightarrow$  In Fig.9.3, none of OCV reaches 1.229 V (E<sup>0</sup>) -Several factors lower voltage than OCV at zero current

(i) Temperature

 $\mathsf{E}^{0}(\mathsf{T}) \approx \mathsf{E}^{\theta}(\mathsf{T}_{\theta})(\mathsf{T}/\mathsf{T}_{\theta}) + (\Delta \mathsf{H}^{\theta}/\mathsf{n}\mathsf{F})(\mathsf{T}/\mathsf{T}_{\theta} - 1)$ 

for T(K), 1.171 V at 80°C, 0.9794 V at 800°C

(ii) Effect of gas composition

For H<sub>2</sub>-O<sub>2</sub> fuel cell E = E<sup>0</sup> + (RT/2F)ln[ $a_{H2}a_{O2}^{0.5} / a_{H2O}$ ]

-For SOFC in Fig. 9.3 94% hydrogen & 6% water, Cathode pure oxygen,  $800^{\circ}C$  $\rightarrow E = 0.9794 + (RT/2F)ln[(0.94)(1)/(0.06)]$ = 1.11 V (E ~ E<sup>o</sup> at high-T FC)



**Figure 9.4** Effect of temperature on the equilibrium potential of a hydrogen/oxygen fuel cell. Water is assumed to be a vapor.

-For the alkaline(and AEM) FC operating pure oxygen Mole fraction of water, hydrogen, and oxygen are all about 0.5 Standard pressure ( $p^0 = 100 \text{ kPa}$ ),  $a_i = p/p^0$ At 80°C and 414 kPa (Fig. 9.3)

→ E = 1.171 + (RT/2F)ln[(2.07)( $\sqrt{2.07}$ )/(2.07) = 1.172 V (lower OCV than thermodynamics in Fig. 9.3 → permeation of oxygen and hydrogen across the separator)

-For PEMFC, operating on air, humidified air and fuel with 47 mol% water vapor  $\rightarrow$  mole fractions y<sub>H2O</sub> = 0.47, y<sub>H2</sub> = 0.53, y<sub>O2</sub> = 0.21 x 0.53 = 0.11

→ E = 1.171 + (RT/2F)ln[(0.53)( $\sqrt{0.11}$ )/(0.47) = 1.156 V (well below OCV in Fig. 9.3 → permeation of reactants across the electrolyte ( a mixed-potential and depression of the OCV) and the sluggishness of the oxygen reduction reaction in acid at low temperatures (key reason!!) → the reaction is so slow that even minute impurities and contaminants can compete with the ORR → OCV↓

## (2) Kinetics

-Kinetics of electrochemical reactions depends strongly on overpotential, catalysts, and temperature

-In low-T FC in Fig. 9.3, small-current region  $\rightarrow$  cell potential to decrease rapidly  $\rightarrow$  then followed by a more gradual decline

-Tafel plot in small-current region

 $\rightarrow$  main differences between the polarization curve and Tafel plot (i) Cell potential is plotted as a function of the logarithm of the current density, (ii) the ohmic resistance is removed

IR-corrected curve  $\rightarrow$  kinetics  $\rightarrow$ Well described by Butler-Volmer equation

Fast hydrogen oxidation + slow ORR  $\rightarrow$  only slow ORR kinetics in Tafel Assume  $\alpha_c \approx 1 \rightarrow$ Tafel slope = ln(RT/ $\alpha_c$ F) = 2.303(RT/ $\alpha_c$ F) =2.303[(8.314x353) / (1 x 96485)] = 0.07 V/dec Illustration 9.2



**Figure 9.5** Tafel plot used for low-temperature PEM fuel cell from Figure 9.3.

## (3) Ohmic region

-At moderate current densities, the importance of ohmic polarization increases compared to the activation polarization

 $\rightarrow$  ohmic losses increase linearly with current (whereas kinetic losses are proportional to the logarithm of current density)

 $\rightarrow$  the absolute magnitude of the kinetic polarization is large for the PEMFC, but slowly at moderate current density

 $\rightarrow$  ohmic region at moderate current density  $\rightarrow$  slope = ohmic resistance

 $\rightarrow$  dependent on the conductivity and thickness of the electrolyte

slope =  $\Delta V / \Delta i \approx L/\kappa = R_{\Omega} [\Omega \cdot m^2]$ 

-In Fig. 9.3, the resistance of SOFC (0.04  $\Omega \cdot m^2$ ) is three times higher than the resistance of the PEMFC

### (4) Mass transfer

-As the current density increases further, mass-transfer effects become important

 $\rightarrow$  reactants and products are transported to and from the catalyst sites

-Summary of polarization of SOFC and PEMFC



Figure 9.6 Polarizations of the PEM and solid oxide fuel cells shown in Figure 9.3 at a cell potential of 0.7 V.

## Effect of operating conditions

 $V_{cell} = E - iR_{\Omega} - |\eta_{s, anode}| - |\eta_{s, cathode}| - |\eta_{conc, anode}| - |\eta_{conc, cathode}|$ 

#### Table 9.3 Effect of Temperature and Pressure on Fuel-Cell Performance

Temperature Factor Pressure  $\approx U^{\mathrm{o}} + \frac{RT}{2F} \ln \frac{\left(\frac{p_{\mathrm{H}_2}}{p^{\mathrm{o}}}\right) \left(\frac{p_{\mathrm{O}_2}}{p^{\mathrm{o}}}\right)^0}{2}$ U  $\Delta H < 0$  for fuel-cell reactions. Therefore U decreases with E increasing temperature; effect is not large, Equation 2.18 Resistance is strongly lowered as the temperature is increased.  $R_{\Omega}$ Negligible effect The conductivity is an activated process that increases exponentially with temperature:  $\kappa = \kappa_o \exp\left\{\frac{-E_{a,\kappa}}{RT}\right\}$ Exchange-current density is an activated process and increases Generally experimental data are needed to determine ns pressure dependence because reactions are not elementary exponentially with temperature:  $i_0 = A \exp\left\{\frac{-E_{a,i}}{RT}\right\}$ Diffusion coefficient in the gas phase depends roughly on the Affects gaseous reactants, for an ideal gas  $c_i = p_i/RT$  and  $\eta_{\rm conc}$ temperature to the 3/2 power, but concentration decreases with  $i_{\rm lim} \sim c_i$ . Thus, mass-transfer polarization decreases at higher temperature. Net result is a small reduction in mass-transfer pressure polarization with temperature

#### E = U in the textbook

**Illustration 9.3** 

## **Electrode structure**

-Contact between three phases is needed in FC electrode to carry out electrochemical reactions: (i) a solid phase that is electronically conductive to supply or remove electrons, (ii) an electrolyte phase (solid or liquid) that conducts ions, (iii) a gas phase that the reactants and products flow  $\rightarrow$  "triple-phase boundary (TPB)"

-example of SOFC :  $O^{2-} + H_2 \rightarrow H_2O + 2e^{-}$ electrolyte gas gas electronic conductor  $\rightarrow$  a mixed ionic-electronic conductor (MIEC)



Figure 9.7 Porous, mixed electronic ionic conductor used to increase the triple-phase boundary.

-FC with liquid electrolyte:  $H_2 + 2OH^- \rightarrow 2H_2O + 2e^$ gas electrolyte gas electronic conductor  $\rightarrow$  flooded-agglomerate model: taking the solubility of gases (oxygen..)



Figure 9.8 Flooded-agglomerate electrode, which is designed to increase the TPB.

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Electrode-electrolyte interface, pore structure



Figure 2.12 The simultaneous presence of reactant, catalyst, an ionic conductor with a continuous path to the main electrolyte, and a continuous path of electrical conductivity is needed or a reaction will not take place at a catalyst site. A catalyst rendered inactive by these situations is called an "orphan" catalyst.

Mathew M. Mench, Fuel Cell Engines, Wiley, 2008.

## Proton-exchange membrane(PEM) FCs

-Using solid polymer material (H<sup>+</sup> ionic conductor) : perfluorinated ionomers

 $\rightarrow$  Nafion: a copolymer of tetrafluoroethylene (Teflon) and sulfonyl fluoride vinyl ether

-Conducting mechanism: Fig. 9.10



Figure 9.9 Basic structure of Nafion<sup>®</sup> co-polymer.

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**Figure 9.10** Scheme of proton-transfer mechanisms (vehicle and Grotthuss mechanisms). *Source:* Ueki 2008. Reproduced with permission of American Chemical Society.

-The 2nd role of ionomers: water uptake,

 $\lambda = \#$  water molecules / # sulfonic acid group

-Conductivity vs. water content

$$\kappa = A + B\lambda$$

-other transport properties: electroosmotic drag coefficient ( $\xi$ ), Diffusion coefficient  $(D_0)$ 



Figure 9.12 Transport processes in PEM fuel cell.







Figure 9.11 Water content in membrane as a function of partial pressure of water vapor. Source: Adapted from Zawodzinski 1993.

-Current density is proportional to the molar flux of protons  $\rightarrow$  water movement across the membrane

-Membrane as a barrier between the fuel and oxidant

-Importance of ORR





Figure 9.13 Water content in ionomer membrane during current flow.

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#### **Illustration 9.5**

## Solid oxide fuel cells (SOFC)

### -Using solid ceramic material (O<sup>2-</sup> ionic conductor)

Table 9.4Types of Electrolytes Used in SOFCs

ZrO <sub>2</sub> , zirconia based	YSZ (yttria-stabilized zirconia), fluorite structure, most common electrolyte							700–1000 °C
CeO <sub>2</sub> , ceria based	Lower temperature, fluorite structure					vbsous	41511	600 °C
LaGaO <sub>3</sub>	Perovskite structure							800 °C

-Conductivity,  $\kappa = F^2 \sum z_i^2 u_i c_i$  $u_i$ : mobility of the ion,  $z_i$ : charge,  $c_i$ : concentration of the species



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**Figure 9.14** Creation of vacancies in ZrO<sub>2</sub>.

-YSZ, O<sup>2-</sup> ionic conductor + a very small electronic conductor ( $\sigma$ ) -transference number, t<sub>02-</sub> = ionic conductivity / electronic conductivity =  $\kappa / (\kappa + \sigma) \approx 1$ 

$$\begin{aligned} V_{cell} &= E - iR_{\Omega} - \left| \eta_{s, anode} \right| - \\ \left| \eta_{s, cathode} \right| - \left| \eta_{conc, anode} \right| - \\ \left| \eta_{conc, cathode} \right| \end{aligned}$$



Figure 9.16 Conductivity of common oxide materials used in SOFCs.



Figure 9.17 Anode-, electrolyte-, and cathode-supported designs used in SOFCs. Drawing is not to scale.



**Figure 9.18** Cross section of a SOFC showing the porous nature of the two electrodes and the dense separator. *Source:* Ormerod 2003. Reproduced with permission of Royal Society of Chemistry.

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Figure 9.19 Effect of temperature on SOFC performance. *Source:* Adapted from Zhao 2005.