

## 固体氧化物燃料电池电解质与阴极界面研究进展及展望

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**摘 要:** 电解质与阴极界面是影响固体氧化物燃料电池(SOFC)性能的主要因素之一, 提升界面化学兼容性、强化荷电传导、减少界面间元素互扩散, 对提升 SOFC 单电池性能及长期运行稳定性至关重要。本文简述了近年来改善电解质与阴极界面问题的方法途径, 包括阴极组成与结构优化、多层复合电解质设计与构建、界面强化方法等, 为助力 SOFC 研发和应用提供参考。

**关键词:** 固体氧化物燃料电池; 电解质; 阴极; 界面; 研究进展

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固体氧化物燃料电池(SOFC)具有一次发电效率高(约 40%~60%)、燃料适用范围广等优势, 在热电联供、分布式发电、石化煤化及国防领域应用前景广泛。成本、性能和寿命是 SOFC 走向应用必须突破的瓶颈。SOFC 单电池是典型的多层陶瓷薄膜电化学器件, 主要包括致密电解质、多孔阳极和阴极。与电解质支撑单电池相比, 当前快速发展的阳极支撑、金属支撑等单电池实现了电解质薄膜化, 提升了单电池性能、降低了运行温度, 但在工业生产及应用中仍存在诸多难题<sup>[1-4]</sup>。

由于运行温度降低至 600~800 °C 的中温区间, 传统电子传导为主的  $\text{La}_{1-x}\text{Sr}_x\text{MnO}_{3-\delta}$  阴极的极化电阻显著增大, 因此, 开发了以  $\text{La}_{0.6}\text{Sr}_{0.4}\text{CoO}_{3-\delta}$  (LSC)、 $\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_{3-\delta}$  (LSCF)、 $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{Co}_{0.8}\text{Fe}_{0.2}\text{O}_{3-\delta}$ 、 $\text{PrBaCo}_2\text{O}_{5+\delta}$  等为代表的, 具有优异混合氧离子-电子电导率、快速氧还原反应速率的高活性阴极。但这些阴极材料本身以及阴极与电解质之间的界面仍存在的问题, 主要包括高温制备过程中的元素迁移、相结构变化和机械结构损伤, 阴极与电解质的热膨胀系数不匹配、化学不兼容、界面强度低, 高温运行后的元素扩散沉积形成高阻相,

以及空气中杂质和金属连接体 Cr 毒化等问题。

本文旨在对 SOFC 电解质与阴极界面相关研究进行总结, 从而为研发高性能稳定的 SOFC 单电池及其长寿命服役提供参考。由于固体氧化物电解池(SOEC)的材料与结构和 SOFC 相近, 对 SOEC 的研发也具有参考价值。

## 1 阴极组成与结构优化

### 1.1 消除宏观界面

传统电解质和阴极为“异质异构”设计, 即不同材料体系的致密电解质和多孔阴极, 存在宏观界面, 因此很容易在高温制备和极端工况下产生阴极剥离和脱落等问题。通过“同质异构”一体化设计, 构建致密电解质和多孔电解质骨架, 进一步采用溶液浸渍工艺在多孔电解质骨架中制备高活性纳米阴极, 能够消除电解质和阴极之间的宏观界面, 如图 1a 所示。杨志宾等<sup>[5]</sup>采用流延-等静压-共烧结工艺制备了阳极支撑三层一体化结构半电池, 并在多孔 YSZ 骨架中浸渍制备了 LSCF 阴极, 其多孔 YSZ 阴极骨架与致密 YSZ 电解质在高温下共烧制得, 阴极骨架的机械强度高、稳定性好; Jia 等<sup>[6]</sup>和 Tong

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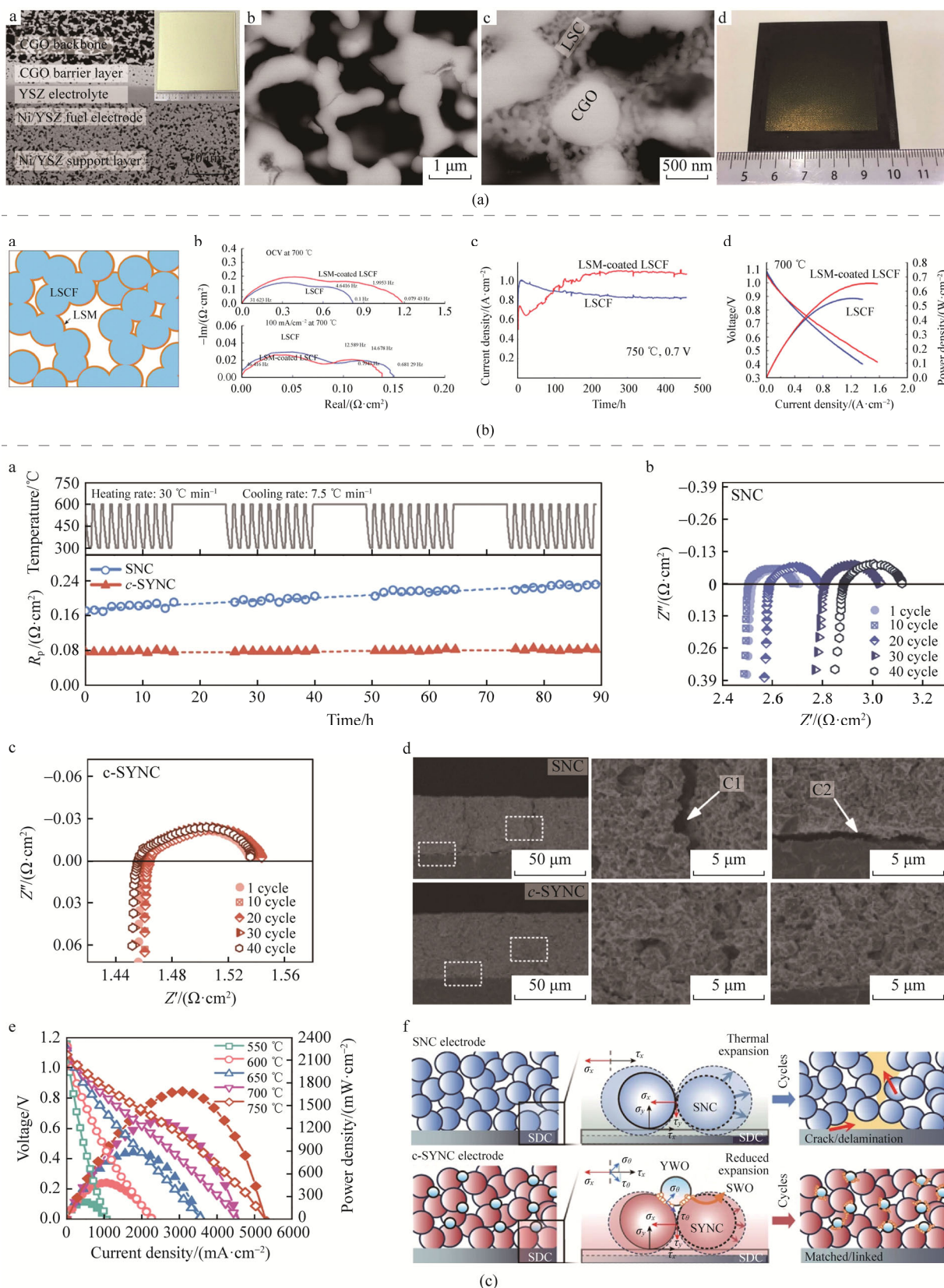


图 1 (a)同质异构一体化设计消除阴极电解质宏观界面: GDC 骨架浸渍 LSC 阴极<sup>[7]</sup>; (b)阴极表面修饰: LSCF 阴极表面浸渍 LSM 薄膜<sup>[8]</sup>; (c)抵消膨胀效应: 阴极中添加负热膨胀材料<sup>[12]</sup>

Fig. 1 (a) LSC catalyst was coated on the internal surfaces of the porous CGO backbone via the infiltration technique<sup>[7]</sup>; (b) Infiltration technique was utilized to deposit thin LSM coatings onto the LSCF<sup>[8]</sup>; (c) Thermal-expansion offset enhanced long-term electrode durability and ORR activity<sup>[12]</sup>

等<sup>[7]</sup>在放大尺寸单电池中分别评价了 LSCF 浸渍 YSZ、LSC 浸渍 GDC 一体化阴极, 获得了高性能输出和稳定运行, 展现出一定的应用前景。

## 1.2 阴极表面修饰

通过表面修饰不仅能够提升阴极电化学性能, 还能有效提高稳定性。图 1b 中, Lynch 等<sup>[8]</sup>采用浸渍工艺在 LSCF 阴极表面构建了  $\text{La}_{0.85}\text{Sr}_{0.15}\text{MnO}_{3\pm\delta}$  氧化物纳米薄膜, 同时实现了高性能输出和稳定运行; Gong 等<sup>[9]</sup>用 ALD 技术在 LSC 阴极表面制备了纳米  $\text{ZrO}_2$  包覆层, 在 700 °C 稳定运行 4000 h, 将阴极极化阻抗衰减速率降低了 20 倍; Li 等<sup>[10]</sup>利用压应力的作用, 设计了核壳结构 LSCF@GDC 复合阴极, 有效抑制了 LSCF 阴极表面 Sr 元素偏析; Chen 等<sup>[11]</sup>采用浸渍法在 LSCF 表面原位生长了  $\text{PrO}_x$  纳米颗粒和  $\text{PrNi}_{0.5}\text{Mn}_{0.5}\text{O}_3$  薄膜, 显著缓解了阴极 Cr 中毒现象, 单电池表现出优异的峰值功率密度 ( $P_{\max}=0.71 \text{ W/cm}^2$ ) 和显著提高的耐久性(0.7 V 条件下, 衰减率为  $0.043 \text{ 4\% h}^{-1}$ )。

## 1.3 热膨胀补偿策略

通过开发新型阴极材料、调控复相阴极中各组分比例, 以及添加负热膨胀材料能够有效改善阴极热膨胀特性。Zhang 等<sup>[12]</sup>采用热膨胀补偿策略在钴基电极中引入具有负热膨胀系数的钇钨氧化物获得  $\text{Sr}_x(\text{Y}_x(\text{Nb}_{0.1}\text{Co}_{0.9})_{1-y})\text{O}_{3-\delta}$  (SYNC) 复合电极, 在 750 °C 条件下单电池峰值功率密度达到  $1.69 \text{ W/cm}^2$ , 如图 1c 所示; Lu 等<sup>[13]</sup>开发了一种热膨胀偏移复合阴极  $\text{Ba}_{0.5}\text{Sr}_{0.5}\text{FeO}_{3-\delta}-x\text{NdMnO}_{3-\delta}$  ( $x=0\sim40\%$ , 质量分数), 采用热膨胀偏移策略对阴极进行晶粒细化, 显著提高电化学性能和运行稳定性; Hu 等<sup>[14]</sup>通过引入负热膨胀材料  $\text{Sm}_{0.85}\text{Zn}_{0.15}\text{MnO}_3$  (SZM) 和  $\text{SrNb}_{0.1}\text{Co}_{0.9}\text{O}_{3-\delta}$  (SCM) 构建复合阴极, 显著改善了阴极和电解质之间的热匹配性, SNC-20SZM 获得了最佳的综合电化学性能, 在 700 °C 条件下极化电阻为  $0.012 \Omega\cdot\text{cm}^2$ , 最大峰值功率密度为  $1.22 \text{ W/cm}^2$ , 在  $0.8 \text{ A/cm}^2$  的恒定电流下连续稳定运行 120 h。

# 2 多层复合电解质构建

## 2.1 多层复合电解质结构

在电解质层面, 构建多层电解质薄膜、优化单电池结构组成能够提升性能、减小衰减。大多数高活性阴极材料如 LSCF、LSC 等与常用的  $\text{ZrO}_2$  基电解质(如 YSZ 和 SSZ 等)在制备和运行过程中发生反应, 生成  $\text{SrZrO}_3$ 、 $\text{La}_2\text{Zr}_2\text{O}_7$  等高阻相物质<sup>[15–16]</sup>。最常用的方法是在阴极与电解质之间加入氧化铈基隔

离层(如  $\text{Ce}_{1-x}\text{Gd}_x\text{O}_{2-\delta}$  (GDC) 和  $\text{Ce}_{1-x}\text{Sm}_x\text{O}_{2-\delta}$  (SDC) 等)以提升化学兼容性<sup>[17–18]</sup>。Wilde 等<sup>[19]</sup>和 Szász 等<sup>[20]</sup>研究发现, 在 YSZ 表面丝印 GDC 并经过 1400 °C 高温烧结形成的  $(\text{Ce},\text{Zr})\text{O}_2$  互扩散层(IDL) 能有效阻挡 LSCF 阴极 Sr 元素扩散, 避免  $\text{SrZrO}_3$  生成。

Wachsman 等<sup>[21]</sup>提出了具有协同效应的  $\text{Er}_{0.4}\text{Bi}_{1.6}\text{O}_3$ (ESB)/GDC 快氧离子导体双层电解质结构, 将 GDC 置于燃料侧防止 ESB 被分解, 位于空气侧的 ESB 则有效阻挡了 GDC 电解质电流内短路, 在 650 °C 获得了约  $2 \text{ W/cm}^2$  峰值功率密度输出, 如图 2a 所示。Zhang 等<sup>[22]</sup>构建了 5  $\mu\text{m}$  厚 GDC/YSZ/GDC 3 层电解质, 虽然 YSZ 厚度仅为 0.8  $\mu\text{m}$ , 却有效阻挡了 GDC 内短路, 在 650 °C 开路电压达到了 1.04 V, 并在 0.7 V 实现了  $1.2 \text{ W/cm}^2$  的功率密度输出, 如图 2b 所示。

## 2.2 致密薄膜制备技术

多层复合结构提高了电解质可靠性和阴极界面化学兼容性, 然而其制备难度大, 主要依赖先进镀膜技术。Riegraf 等<sup>[23]</sup>通过物理气相沉积(PVD)技术制备了亚微米厚的 GDC 隔离层, 发现当致密 GDC 隔离层厚度达到 0.3~0.5  $\mu\text{m}$  以上时, 能够有效抑制由于阴极 Sr 元素扩散在电解质界面生成杂相; Fonseca 等<sup>[24]</sup>采用磁控溅射技术制备了约 1  $\mu\text{m}$  的 GDC 隔离层, 单电池在 800 °C 的输出性能达到  $1 \text{ W/cm}^2$ ; Wang 等<sup>[25]</sup>通过 PVD 法制备了约 1  $\mu\text{m}$  厚的 GDC 薄膜, 在低于 1100 °C 烧结获得了致密的 GDC 隔离层, 有效阻止了阴极中 Sr 的迁移扩散。

在工业生产中, 大多选用成本较低的流延、丝网印刷等湿法陶瓷工艺, 并结合高温烧结制备单电池, 但获得的氧化铈基隔离层致密度欠佳, 导致较大的欧姆电阻和较差的元素迁移阻隔效果。针对此, Ni 等<sup>[26]</sup>、Choi 等<sup>[27]</sup>和 Wang 等<sup>[28]</sup>通过多孔隔离层中浸渍前驱体溶液或助烧剂盐溶液, 有效提高了隔离层致密度, 降低了单电池欧姆阻抗; Lyu 等<sup>[29]</sup>通过在硝酸铈和硝酸钆溶液中水热处理, 也显著提升了多孔 GDC 隔离层致密度, 有效抑制了 Sr 元素扩散, 获得了高性能输出和稳定运行。

采用上述先进镀膜技术能够制备高质量的电解质薄膜, 然而存在成本高、放大均一性差等难题。Zhao 等<sup>[30]</sup>和 Li 等<sup>[31]</sup>提出了水热原位自结晶成膜技术, 具有制备成本低、工艺条件可控性好、放大均一性效果佳的特点, 如图 2c 所示。Lyu 等<sup>[32]</sup>研究并阐明了 GDC 薄膜水热原位自结晶过程机理, 构建

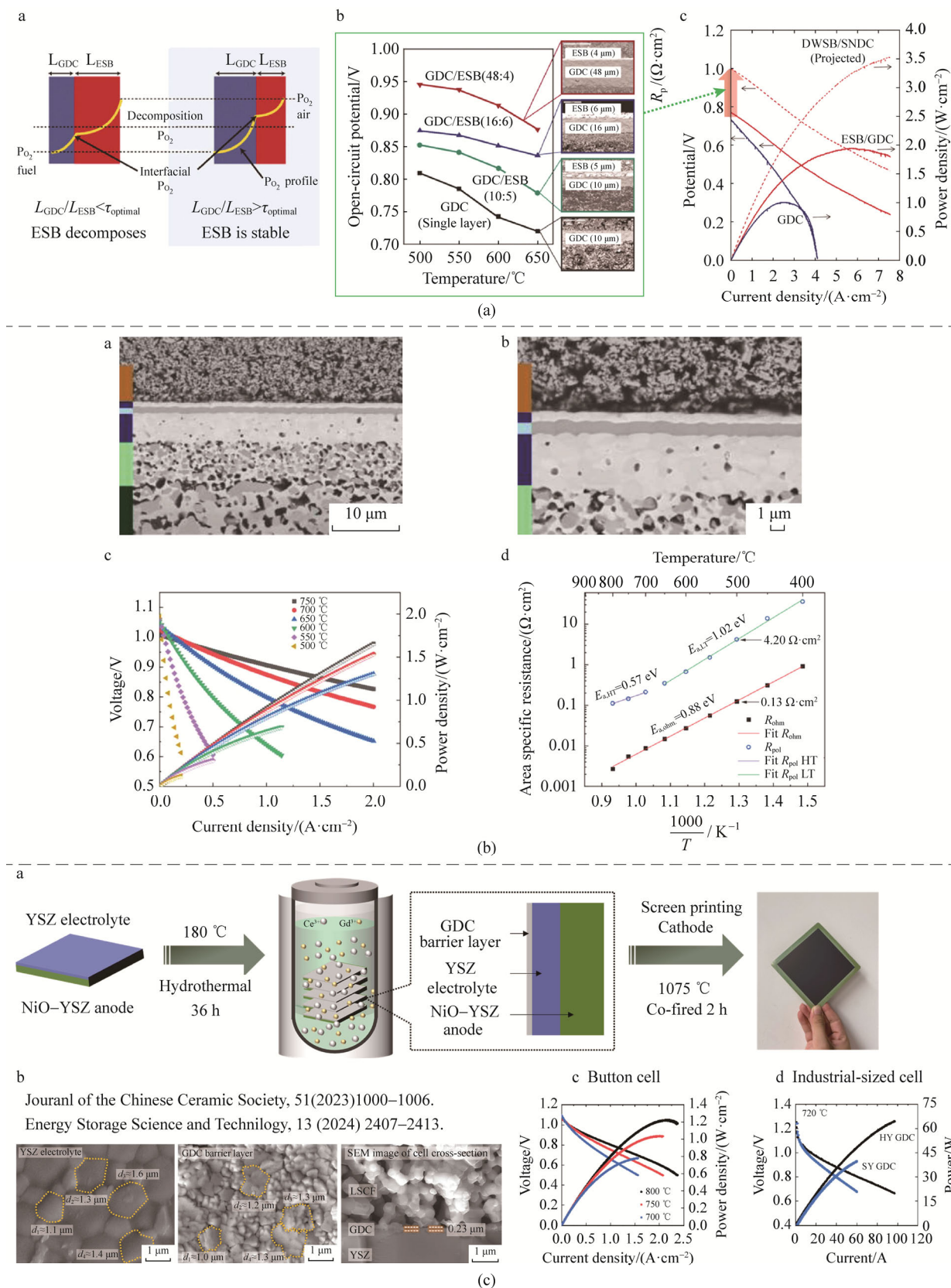


图 2 (a)采用脉冲激光沉积-喷涂工艺制备的 ESB/GDC 双层电解质结构<sup>[21]</sup>; (b)采用丝网印刷-物理气相沉积工艺制备的 GDC/YSZ/GDC 3 层电解质结构<sup>[22]</sup>; (c)采用水热原位自结晶工艺制备的 GDC/YSZ 双层电解质结构<sup>[30-31]</sup>

Fig. 2 (a) ESB/GDC bilayer electrolyte prepared using pulse laser deposition-spraying process<sup>[21]</sup>; (b) GDC/YSZ/GDC Tri-layer electrolyte fabricated using screen printing-physical vapor deposition process<sup>[22]</sup>; (c) GDC/YSZ bilayer electrolyte prepared using hydrothermal *in-situ* self-crystallization process<sup>[30-31]</sup>

表 1 各种镀膜技术成本及致密度评价表

Table 1 Cost and Density Evaluation Table of Various Coating Technologies

| Preparation method | Cost        | Production efficiency | Density  |
|--------------------|-------------|-----------------------|----------|
| Tape Casting       | Low         | Moderate              | Moderate |
| Screen printing    | Low         | High                  | Poor     |
| Spin coating       | High-street | Moderate              | Moderate |
| Thermal spraying   | High-street | High                  | Moderate |
| PVD/CVD            | Costly      | Poor                  | High     |

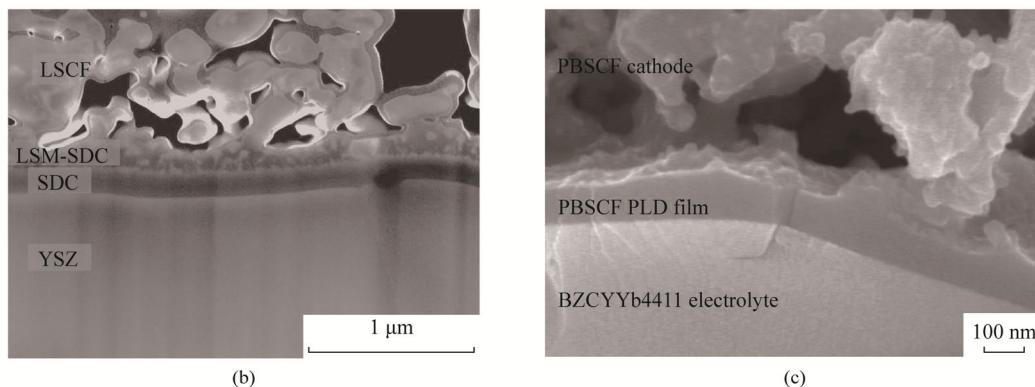
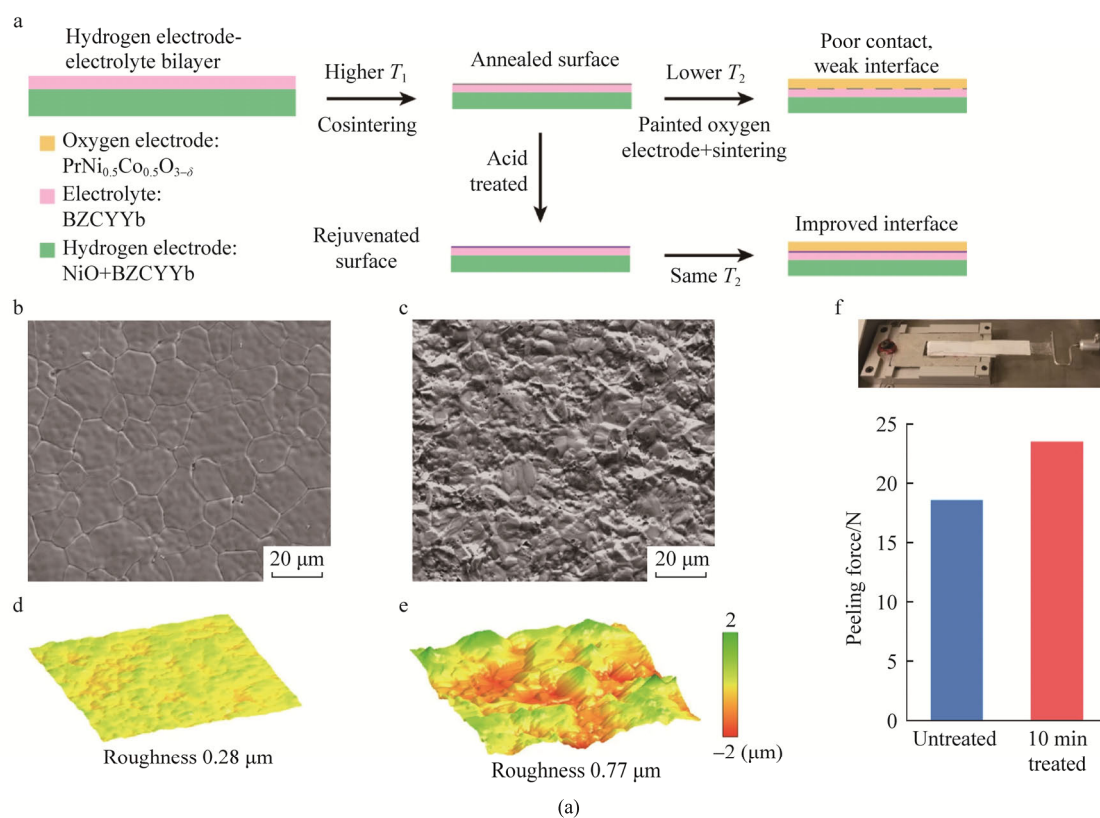
了 GDC-YSZ 双层电解质, 显著降低了单电池欧姆阻抗和极化阻抗, 有效提升了输出性能; Zhao 等<sup>[33]</sup>通过调节前驱液 pH 值, 显著提升了 GDC 薄膜生长

速率及 Gd 元素掺杂量, 提升了原子经济性, 降低了时间成本; Lyu 等<sup>[34]</sup>构建了亚微米连续致密 GDC-IDL 复合隔离层, 形成了 GDC-IDL-YSZ 三层电解质, 有效阻隔了阴极电解质界面元素互扩散。

### 3 电解质与阴极界面强化

#### 3.1 电解质表面改性

电解质和阴极界面接触直接影响了界面电阻、氧离子传导及界面结合强度, 因此, 通过界面强化能有效提升单电池能效和稳定性。图 3a 中, Bian 等<sup>[35]</sup>对高温烧结后的电解质表面进行简单的酸处

图 3 (a)电解质表面酸蚀改性<sup>[35]</sup>; (b-c)阴极/电解质之间构建致密纳米结构功能层<sup>[39, 41]</sup>Fig. 3 (a) Electrolyte surface of the cosintered hydrogen electrode–electrolyte bilayer with nitric acid<sup>[35]</sup>; (b-c) Construction of a dense nanostructured functional layer between the cathode and electrolyte<sup>[39, 41]</sup>

理,使其表面恢复烧结活性,从而在氧电极和电解质之间实现反应键合,提高电化学性能和稳定性;Chang 等<sup>[36]</sup>通过将 NaOH 和 YSZ 电解质共压,利用 NaOH 潮解有效提升 YSZ 表面的粗糙度,增大了电极与电解质之间的界面接触面积;He 等<sup>[37]</sup>优化了 GDC 隔离层的烧结温度,并经过水热原位生长的方法构建了活化 GDC 表面,提升了与 LSCF 阴极的界面接触。

### 3.2 致密纳米结构功能层

致密纳米结构功能层(NFL)突破了多孔阴极-致密电解质接触界面的物理极限,提升了单电池性能、界面结合及稳定性,如图 3b、图 3c 所示。Januschewsky 等<sup>[38]</sup>采用脉冲激光沉积 (PLD)制备了 300 nm 厚  $\text{La}_{0.6}\text{Sr}_{0.4}\text{CoO}_{3-\delta}$  作为 NFL,显著降低了阴极极化阻抗;Choi 等<sup>[39]</sup>采用 PLD 在  $\text{BaZr}_{0.4}\text{Ce}_{0.4}\text{Y}_{0.1}\text{Yb}_{0.1}\text{O}_3$  质子导体电解质表面制备了 100 nm 厚的致密  $\text{PrBa}_{0.5}\text{Sr}_{0.5}\text{Co}_{1.5}\text{Fe}_{0.5}\text{O}_{5+\delta}$  薄膜,显著降低了阴极电解质界面电阻,提升了单电池电化学性能输出和长期运行稳定性;Develos-Bagarinao 等<sup>[40]</sup>采用 PLD 制备了 300 nm 厚的  $\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_{3-\delta}-\text{Ce}_{0.9}\text{Gd}_{0.1}\text{O}_{2-\delta}$  作为 NFL, 700 °C, 0.7 V 下的输出功率密度达到了 3.3 W/cm<sup>2</sup>;Machado 等<sup>[41]</sup>构建了由  $\text{Sm}_{0.2}\text{Ce}_{0.8}\text{O}_{1.9}-\text{La}_{0.8}\text{Sr}_{0.2}\text{MnO}_{3-\delta}$  组成的多层异质外延阴极,有效增强了氧表面交换和氧离子的扩散能力,此外还发现,双层界面单电池的总电阻相比单层 NFL 界面结构单电池降低近 50%。

此外,通过直接组装法和闪烧等技术方法可以在较低的温度形成电极/电解质界面,避免了长时间高温烧结过程,能够直接用于无隔离层的  $\text{ZrO}_2$  基电解质单电池,为低熔点和纳米结构阴极的制备提供新途径<sup>[42-43]</sup>。

## 4 结论与展望

电解质和阴极之间的界面接触和荷电传导直接影响到单电池的性能和效率,是 SOFC 单电池基础研究和技术研发的重要关注领域。本文在阴极组成与结构优化、多层复合电解质结构,以及开发有效的界面强化方法方面进行了简述,以期为推动其发展提供见解。对该方向的未来发展做出以下展望。

1) 材料开发与结构设计。新型电解质和电极材料的研发是突破界面问题的有效途径,同时探索构建新的界面结构设计也能在现有材料体系基础上实现性能和稳定性的提升。未来研究工作应更加注重服务实际工业应用需求。

2) 制备工艺研发优化。电解质与阴极界面质量直接影响 SOFC 的整体性能,因此,优化制备工艺至关重要。目前,常用的镀膜技术如 PLD、CVD 等,可以实现特定组成和结构的精准制备。未来需要注重新的高性能、低成本并且具有规模化制造能力的薄膜制备技术研发,以进一步提高界面质量。

3) 多学科交叉研究。多种原位谱学表征方法、机器学习、大数据和人工智能技术也将助力 SOFC 研究,用于材料性能预测、参数优化设计、工况性能模拟等,能够促进理论模型与实际应用之间的联结,推动 SOFC 技术向更高效率、更低成本的方向发展。

4) 实际应用与产业化。尽管实验室研究取得了显著进展,但 SOFC 的商业化仍面临挑战。未来需要进一步优化电池的制备工艺和运行条件,如电解质结构、复合电极材料及烧结工艺等。降低成本,提高和延长电池的可靠性和寿命,以推动其在能源领域的广泛应用。

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## Research Progress of Electrolyte and Cathode Interface in Solid Oxide Fuel Cells

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### Extended Abstract

The interface between electrolyte and cathode is a primary factor affecting the performance of solid oxide fuel cells (SOFCs). Enhancing the chemical compatibility of the interface, fortifying the charge conduction, and mitigating the interdiffusion of elements between the interfaces are imperative to optimize the performance of SOFC single-cells and ensure a long-term operational stability. This review represents the methodologies used to enhance the interface between electrolyte and cathode in recent years. These methodologies encompass the optimization of cathode composition and structure, the design and construction of multilayer composite electrolyte, and interfacial reinforcement methods. These approaches serve as a valuable reference for SOFC development and application.

At the cathode layer, a dense electrolyte and a porous electrolyte backbone are constructed. A solution infiltration technique is utilized to prepare highly active nano-cathodes within the porous electrolyte backbone, thereby eliminating the macroscopic interface between the electrolyte and the cathode. Advanced coating techniques and infiltration technique are employed to modify the cathode surface, thereby enhancing cathode electrochemical performance and single-cell operational durability. The development of novel cathode materials, regulation of component ratios within composite cathodes, and the addition of negative thermal expansion materials prove effective in enhancing the thermal expansion compatibility of the cathode. The thermal-expansion offset enhances long-term electrode durability and ORR activity.

At the electrolyte layer, constructing multi-layer electrolyte films and optimizing the structure of single cell enhance performance and reduce degradation. Multi-layer composite structures improve an electrolyte reliability and a cathode interface chemical compatibility. However, the preparation becomes a challenge and primarily relies on advanced coating technologies. Low-cost, high-density coating technologies are urgently needed, and a hydrothermal *in-situ* growth technique emerges, thus reducing production costs, while enhancing single-cell performance and long-term durability.

At the electrolyte-cathode interface, the contact between the electrolyte and the cathode directly affects the interface resistance, oxygen ion conduction, and interface bonding strength. Therefore, interface reinforcement can effectively improve the energy efficiency and stability of single cells. The dense nanostructure functional layer (NFL) overcomes the physical limitations of the porous cathode-dense electrolyte interface, thus enhancing single-cell performance, interfacial bonding, and long-term durability.

This review summarizes some research work related to the interface between the electrolyte and cathode in SOFCs, thus providing a reference for the development of high-performance and long-term durability. This review also provides a reference for SOEC research and development since the materials and structure of solid oxide electrolysis cells (SOECs) are similar to those of SOFCs.

**Summary and Prospects** The interfacial contact and charge conduction between the electrolyte and cathode have a direct impact on the performance and efficiency of single cells, which is a crucial area in both fundamental research and technological development of SOFCs. The following perspectives for future development are provided.

The development of novel electrolytes and electrode materials represents an effective strategy to address interfacial issues. Concurrently, the exploration of innovative interfacial structural designs can enhance the performance and stability within existing material systems. Future research endeavors should prioritize meeting the practical demands of industrial applications. Furthermore, it is essential to optimize the preparation process. The quality of the interface between the electrolyte and the cathode directly affects the overall performance of SOFCs. Therefore, optimizing the preparation process is of crucial importance. Currently, coating technologies such as pulsed laser deposition (PLD) and chemical vapor deposition (CVD) can achieve a precise preparation of specific compositions and structures. In the future, it is necessary to focus on the research and development of new high-performance, low-cost thin film preparation technologies with large-scale manufacturing capabilities to further improve interface quality. In addition, there is a focus on interdisciplinary research. Various *in-situ* spectroscopic characterization methods, machine learning and artificial intelligence technologies will also contribute to SOFCs research, including material performance prediction, parameter optimization design, and operating condition performance simulation. This can promote the connection between theoretical models and practical applications, driving SOFCs technology toward higher efficiency and lower costs. Finally, despite significant progress in lab-scale research, the commercialization of SOFCs still faces some challenges. In the next phase, it is necessary to further optimize the cell preparation process and operating conditions, such as electrolyte structure, composite electrode materials, and sintering processes. Reducing costs, improving cell reliability and lifespan can promote SOFCs widespread application in the energy sector.

**Keywords** solid oxide fuel cells; electrolyte; cathode; interface; research progress