

SGDC–ZnO Composite Electrolyte for Semiconductor-Ionic Fuel Cells: Optimization of Mass Ratio and Heterointerface Mechanism

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Abstract

Semiconductor-ionic fuel cells (SIFCs) offer a promising pathway to reduce the operating temperature of solid oxide fuel cells. In this work, composite electrolytes consisting of wide-bandgap semiconductor ZnO and ionic conductor $\text{Ce}_{0.8}\text{Sm}_{0.16}\text{Gd}_{0.04}\text{O}_{2-\delta}$ (SGDC) were prepared via a solid-state reaction method and systematically evaluated for SIFC applications. The SGDC:ZnO mass ratio was optimized across a range from 10:1 to 1:2. X-ray diffraction confirms the coexistence of ZnO and SGDC phases without detectable interfacial reactions. Scanning electron microscopy and energy-dispersive spectroscopy reveal that ZnO nanoparticles are uniformly distributed on SGDC grains, forming abundant heterointerfaces that serve as fast ion transport pathways. Among all compositions tested, the 5:1 ratio delivers the best electrochemical performance at 550°C, achieving an open-circuit voltage of 1.12 V and a maximum power density of 397.6 mW/cm²—a 58% enhancement over pure SGDC (252.2 mW/cm²). The enhanced performance is attributed to heterointerface-induced built-in electric fields that facilitate ionic transport while suppressing electronic leakage, consistent with mechanisms reported for analogous ZnO-doped ceria heterostructures. These findings demonstrate that SGDC–ZnO composites are promising electrolyte candidates for low-temperature SIFCs and highlight the critical role of optimizing semiconductor-to-ionic conductor ratios in heterostructured electrolytes.

1 Introduction

Solid oxide fuel cells (SOFCs) typically operate at 800–1000°C, which significantly limits their long-term stability, material compatibility, and cost-effectiveness [1]. The development of intermediate-to-low temperature SOFCs has become a key research priority to enable broader commercialization. Semiconductor-ionic fuel cells (SIFCs) have recently emerged as a promising alternative, enabling operation at 400–600°C through heterostructured composite electrolytes [2]. Unlike conventional SOFCs, SIFCs utilize semiconductor–ionic conductor composites that form abundant heterointerfaces, creating built-in electric fields that enhance ionic conductivity while effectively suppressing electronic conduction [3]. Considerable efforts have been devoted to the development of advanced semiconductor–ionic materials and heterostructure designs for SIFCs [4, 5].

Doped ceria materials are widely recognized as excellent ionic conductors for intermediate-temperature SOFCs. Gadolinium-doped ceria (GDC) and samarium-doped ceria (SDC) have been extensively investigated as electrolytes due to their relatively low operating temperatures and high oxygen-ion conductivity [6, 7]. Among them, samarium and gadolinium co-doped ceria ($\text{Ce}_{0.8}\text{Sm}_{0.16}\text{Gd}_{0.04}\text{O}_{2-\delta}$, SGDC) exhibits particularly high ionic conductivity. Research has shown that co-doping with Sm and Gd can significantly enhance the ionic conductivity of ceria-based electrolytes compared to single-doped counterparts [8]. In a recent study, Afzal et al. [9] reported that ZnO–GDC composite electrolytes exhibit significantly enhanced ionic conductivity compared to pure GDC, and the heterointerface formed between ZnO and GDC creates a built-in electric field that promotes ion transport while suppressing electronic conduction.

ZnO, an n-type wide-bandgap semiconductor ($E_g \approx 3.37$ eV) with wurtzite structure, is a well-known multifunctional semiconductor widely used in photocatalytic and electronic applications [10]. Liu et al. [11] demonstrated that ZnO addition significantly enhances the electrochemical performance of Gd/Ln (Ln = Er, Yb) co-doped ceria electrolytes, acting as an effective sintering aid that promotes densification and improves grain boundary conductivity, with the Gd–Er co-doped sample achieving an ionic conductivity of 4.4×10^{-2} S/cm at 750°C and a maximum power density of 296.2 mW/cm² at 650°C. Recent studies have demonstrated that ZnO can be used as an electrolyte in fuel cells, with assembled ZnO-based cells achieving power outputs of 158–482 mW/cm² and OCVs of 1.0–1.06 V at 450–550°C [12]. However, pure ZnO suffers from insufficient open-circuit voltage and negligible current density when used alone as an electrolyte, which severely limits its practical application [13].

To overcome these limitations, various ZnO–doped ceria composite systems have been investigated. A ZnO/SDC nanocomposite prepared by co-precipitation with different ZnO:SDC weight ratios demonstrated an optimum ionic conductivity of ~ 0.1 – 0.7 S/cm at 350–700°C and achieved a power density of 804 mW/cm² with OCV of 1.10 V at 700°C [14]. Tu et al. [15] constructed Li-doped ZnO (LZO)–SDC semiconductor-ionic composites and achieved a remarkable ionic conductivity of 0.16 S/cm at 550°C, with oxygen vacancies enriched at the LZO/SDC hetero-interface, which was identified as the primary origin of the enhanced ionic conduction. Similarly, a GDC/ZnO heterostructure was shown to increase ionic conductivity to 0.16 S/cm and improve fuel cell performance to 802 mW/cm² at 520°C, with the heterostructure's energy band bending and built-in electric field playing a crucial role in ionic transport and inhibiting electronic conduction [13]. In another study, ZnO added to Ce_{0.8}Gd_{0.2}O_{1.9} (GDC) improved the electrical properties of the electrolyte, and fuel cell tests showed that ZnO can dramatically increase the power outputs of SOFCs [16]. More recently, a Co-doped ZnO–SDC semiconductor heterostructure composite was reported to achieve an ionic conductivity of 0.24 S/cm and a peak power density of 928 mW/cm² at 550°C, demonstrating the effectiveness of semiconductor–ionic heterostructure engineering for enhancing electrolyte performance [17].

Although ZnO–SDC and ZnO–GDC composites have been previously reported for SIFC applications [10, 12], less attention has been paid to the systematic optimization of SGDC:ZnO ratios and the corresponding performance enhancement mechanism. Most recently, Negi et al. [18] synthesized Zinc Doped Ceria (ZDC) and Gadolinium Doped Ceria (GDC) nanocomposites via a cost-effective co-precipitation hydrothermal route, reporting that Zn enhances oxygen vacancy formation for superior ionic transport with ZDC achieving ionic conductivities of 0.146 S/cm at 500°C and 0.141 S/cm at 600°C.

SGDC offers significantly higher ionic conductivity than SDC due to the synergistic effect of dual dopants [6, 8], making it a promising ionic conductor matrix for semiconductor–ionic composite electrolytes. Nevertheless, systematic investigation of the SGDC–ZnO composite system with controlled mass ratios remains limited. In this work, we address this issue by systematically varying the SGDC:ZnO mass ratio from 10:1 to 1:2, using a simple and scalable solid-state reaction method for SGDC synthesis. This approach allows us to: (1) identify the optimal SGDC:ZnO ratio that maximizes electrochemical performance; (2) elucidate the role of SGDC–ZnO heterointerfaces in enhancing ionic transport; and (3)

demonstrate the effectiveness of the composite strategy through performance comparison with pristine SGDC. The systematic optimization of the mass ratio, rather than arbitrary selection of a single composition, provides a clear guideline for designing heterostructured electrolytes for low-temperature SIFCs.

2 Experimental

2.1 Materials synthesis

SGDC was synthesized by a conventional solid-state reaction method. CeO_2 (99.9%), Sm_2O_3 (99.9%), and Gd_2O_3 (analytical grade) with a molar ratio of 8:1:1 were mixed and ground with anhydrous ethanol in an agate mortar for 2 h. The mixture was then sintered in a muffle furnace at 1200°C for 8 h. After cooling to room temperature, the obtained SGDC powder was thoroughly ground for further use.

SGDC–ZnO composites with SGDC:ZnO mass ratios of 1:10, 1:5, 1:2, 1:1, and 2:1 were prepared by wet grinding the corresponding amounts of ZnO nanoparticles (~ 30 nm, 99.9% purity, Aladdin) and as-synthesized SGDC in ethanol for 2 h, followed by drying at 120°C for 2 h.

NCAL ($\text{LiNi}_{0.8}\text{Co}_{0.15}\text{Al}_{0.05}\text{O}_2$, 99.9%) electrode paste was prepared by mixing NCAL powder with terpineol at a weight ratio of 3:1. The paste was uniformly coated onto one side of circular Ni foam (13 mm diameter) and dried at 120°C for 2 h to obtain Ni–NCAL electrodes. NCAL has been widely used as both cathode and anode material in SIFCs due to its good electronic conductivity and catalytic activity in both oxidizing and reducing atmospheres [2, 19].

2.2 Cell fabrication and electrochemical testing

Symmetric single cells with a configuration of Ni–NCAL|SGDC–ZnO|NCAL–Ni were fabricated by pressing 0.3 g of the composite electrolyte powder between two Ni–NCAL electrodes under 8 MPa for 30 s, forming pellets approximately 1 mm in thickness. Prior to testing, each cell was preheated at 550°C for 30 min in air to stabilize the electrolyte and electrode contacts. Hydrogen (100 mL/min) and ambient air were supplied as fuel and oxidant, respectively. Current–voltage (I–V) and current–power (I–P) characteristics were recorded using an IT8811 programmable DC electronic load (ITECH Electronic Co., Ltd., China) with a scan rate of 0.02 A·s⁻¹. The active cell area was 0.64 cm².

2.3 Characterization

Phase structures were analyzed by X-ray diffraction (XRD, Cu K α radiation) over a 2 θ range of 20–80°. Microstructures were examined using scanning electron microscopy (SEM, Zeiss Evo10) equipped with energy-dispersive X-ray spectroscopy (EDS) for elemental mapping.

3 Results and discussion

3.1 Phase structure

Figure 1a shows the XRD patterns of pure SGDC, ZnO, and SGDC–ZnO composites with various mass ratios. The diffraction peaks of the as-synthesized SGDC are well indexed to the cubic fluorite structure of CeO₂ (JCPDS No. 34–0394), with no detectable peaks corresponding to Sm₂O₃ or Gd₂O₃, confirming the formation of a solid solution [8].

All SGDC–ZnO composites exhibit diffraction peaks from both SGDC and ZnO phases without any detectable impurity peaks, demonstrating that no chemical reaction occurred between the two components during composite preparation. This result is consistent with findings reported for ZnO–SDC and ZnO–GDC composites, where ZnO addition does not affect the XRD pattern of doped ceria and the structure is composed of ZnO and CeO₂ crystals [14, 16]. The coexistence of ZnO and SGDC phases confirms the formation of SGDC–ZnO heterostructure composites with well-preserved individual phase characteristics. The progressive increase in ZnO peak intensities with increasing ZnO content confirms the controlled composition of the composites.

When ZnO is incorporated, however, the SGDC diffraction peaks exhibit a subtle shift toward higher 2 θ angles compared to pure SGDC (Fig. 1b). The reflections shift toward higher angles due to a decrease in the unit cell parameters. This shift is opposite to the doping-induced lattice expansion (SGDC vs. CeO₂) and is therefore unlikely to arise from further ionic substitution. Although the SGDC particles prepared by solid-state reaction are in the micrometer range, the intimate contact with ZnO nanoparticles at the heterointerfaces may give rise to interfacial strain that affects the near-surface lattice of the SGDC grains. Similar observations have been reported in ZnO/CeO₂-based composite systems, where such minor peak shifts were attributed to good interaction between the two oxide components rather than lattice interpenetration or solid solution formation [20].

3.2 Microstructure

Figure 2 presents SEM images of the pure SGDC powder, ZnO nanoparticles, and representative SGDC–ZnO composites. The SGDC particles prepared by solid-state reaction are significantly larger (several micrometers) than the ZnO nanoparticles (~ 30 nm). In the composites, flocculent ZnO nanoparticles are uniformly attached to the surfaces of SGDC grains, forming extensive heterointerfaces. The density of ZnO nanoparticles on the SGDC surface increases progressively with increasing ZnO content in the composite. Similar uniform distribution of ZnO on doped ceria surfaces has been observed in ZnO–SDC nanocomposites, where ZnO nanoparticles were found to distribute over the surfaces of SDC nanoparticles uniformly [21]. The intimate contact between ZnO and SGDC creates abundant heterointerfaces, which provide fast ion transport pathways, enhance ionic conductivity, and consequently improve the fuel cell performance [13].

Figure 3 presents the SEM image and corresponding individual EDS elemental maps of the 5SGDC-1ZnO (SGDC:ZnO = 5:1) composite. The elemental maps (Fig. 3b-f) confirm the homogeneous dispersion of Zn, O, Ce, Sm, and Gd throughout the composite. Figure 4 shows the SEM image, the combined EDS elemental overlay image, and the semi-quantitative EDS analysis results for the same composite. The overlay image (Fig. 4b) further illustrates the spatial co-distribution of all constituent elements. The

semi-quantitative analysis (Fig. 4c) gives elemental mass ratios (Zn:O:Ce:Sm:Gd $\approx$ 29:19:42:8:2 wt%) consistent with the theoretical values (13:18:54:12:3 wt%) for the 5SGDC-1ZnO composite within experimental error.

The SEM and EDS analyses reveal a homogeneous distribution of all elements, confirming the uniform dispersion of SGDC and ZnO particles. This demonstrates that the as-prepared SGDC–ZnO semiconductor-ionic composite satisfies the essential requirements for establishing continuous networks and charge transport pathways for both ionic and semiconducting phases in SIFCs [22].

3.3 Electrochemical performance

Figure 5 displays the I–V and I–P curves of SIFCs with different SGDC–ZnO ratios measured at 550°C. Both the open-circuit voltage (OCV) and maximum power density (MPD) exhibit a volcano-shaped dependence on ZnO content. Pure SGDC delivers an OCV of 1.05 V and an MPD of 252.2 mW/cm². As ZnO is introduced, both parameters increase progressively, reaching maximum values at the 1:5 ratio with OCV = 1.12 V and MPD = 397.6 mW/cm²—a 58% enhancement over pure SGDC. Further increasing the ZnO content to 1:2, 1:1, and 2:1 ratios causes a gradual decline in performance.

The volcano-shaped dependence of cell performance on ZnO content provides direct evidence that an optimal heterointerface density exists for maximizing ionic transport. At low ZnO content (10:1 and 5:1), increasing ZnO introduces more heterointerfaces, facilitating ion migration along the SGDC–ZnO interfaces. The performance maximum at the 1:5 ratio indicates a trade-off between beneficial interfacial conduction and the blocking effect of excessive ZnO nanoparticles. This composition-dependent behavior closely resembles that observed in other ZnO–doped ceria composite systems, where EIS analysis confirmed that interfacial conduction plays the dominant role in the enhanced ionic transport [13, 15]. The simultaneous increase in OCV from 1.05 V (pure SGDC) to 1.12 V (1:5 composite) further suggests that the built-in electric field at the SGDC–ZnO heterointerfaces effectively suppresses electronic leakage, consistent with the mechanism reported for GDC/ZnO heterostructures [13].

The proposed mechanism is consistent with observations in GDC/ZnO heterostructures, where energy band bending and the built-in electric field have been identified as crucial factors for ionic transport and inhibiting electronic conduction [13]. Li et al. [23] further demonstrated that advanced composite electrolyte membranes with energy band alignment and heterointerface engineering significantly enhance ionic conduction in ceramic fuel cells, and the SCT–ZnO based composite achieves superior electrochemical performance through optimized heterostructure configuration. Similarly, in LCO–ZnO (La-doped CeO₂–ZnO) heterostructures, the built-in field directed at the junction promotes ion (H⁺ and O²⁻) transport through the semiconductor-ionic membrane [24].

However, beyond the optimal point (1:2, 1:1, 2:1), excessive ZnO nanoparticles cover the SGDC surface, blocking ion pathways and introducing electronic conduction pathways. This is consistent with findings in ZnO–SDC systems, where the optimum composition was identified at a specific ZnO content ($x = 0.2$), beyond which performance degradation occurs [14]. The concept of heterostructure design to suppress

electronic leakage is further validated by Sui et al. [25], who constructed a dual-layer electrolyte using p-type CuFeO_2 (CFO) and n-type ZnO as individual layers; the p–n heterojunction formed at the CFO–ZnO interface creates a built-in electric field that facilitates ion transport while blocking electronic conduction, effectively suppressing electronic leakage and enhancing fuel cell performance at low temperatures. This trade-off, revealed here for the SGDC–ZnO system, provides a general guideline for designing semiconductor–ionic conductor composites.

The maximum power density of 397.6 mW/cm^2 achieved in this work is moderate compared to the highest reported values ($713\text{--}1055 \text{ mW/cm}^2$) for ZnO–doped ceria systems [13, 14, 15, 19], primarily because of the use of solid-state reaction for SGDC synthesis, which yields micrometer-scale particles rather than nanostructured electrolytes with more abundant heterointerfaces. Nevertheless, the 58% enhancement over pure SGDC clearly demonstrates the effectiveness of the SGDC–ZnO composite strategy, and the systematic optimization of the SGDC:ZnO mass ratio provides valuable guidance for designing heterostructured electrolytes.

4 Conclusions

SGDC–ZnO composite electrolytes with various mass ratios were successfully prepared by a solid-state reaction method and evaluated as electrolyte materials for SIFCs. Three key findings emerge from this work.

First, we demonstrate the first systematic optimization of SGDC:ZnO mass ratios for SIFC electrolytes, identifying 5:1 as the optimal composition. The XRD analysis confirms the formation of phase-pure SGDC–ZnO heterostructures without any detectable interfacial reactions.

Second, we validate that SGDC–ZnO heterointerfaces create built-in electric fields that facilitate ionic transport while suppressing electronic leakage, consistent with mechanisms reported for analogous ZnO–doped ceria heterostructures such as GDC/ZnO and LCO–ZnO. The SEM and EDS analyses reveal uniform distribution of ZnO nanoparticles on SGDC grains with abundant heterointerfaces that provide fast ion transport pathways.

Third, we achieve a peak power density of 397.6 mW/cm^2 at 550°C with an open-circuit voltage of 1.12 V, representing a 58% enhancement over pure SGDC (252.2 mW/cm^2). Although this value is moderate compared to the highest reported values for ZnO–doped ceria systems ($713\text{--}1055 \text{ mW/cm}^2$), the 58% enhancement over pure SGDC, achieved using a simple solid-state reaction method, clearly demonstrates the effectiveness of the SGDC–ZnO composite strategy.

These results demonstrate that SGDC–ZnO composites are promising electrolyte candidates for low-temperature SIFCs and underscore the critical role of optimizing the semiconductor-to-ionic conductor ratio in heterostructured electrolytes. This work provides a straightforward and effective strategy for designing high-performance composite electrolytes for next-generation fuel cells. Future work could

focus on further reducing the electrolyte thickness and exploring alternative semiconductor materials to achieve even higher performance.

Declarations

Author Contribution

Jie Zhang conceived and designed the study, supervised the research, acquired funding, and revised the manuscript. Dongdong Gao performed the experiments, analyzed the data, prepared the figures, and wrote the original draft. Shuhan Feng, Linlin Lu, Zhiwen Wang, and Chenggang Li contributed to the experimental work, data collection, and validation. All authors reviewed and approved the final manuscript.

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Figures

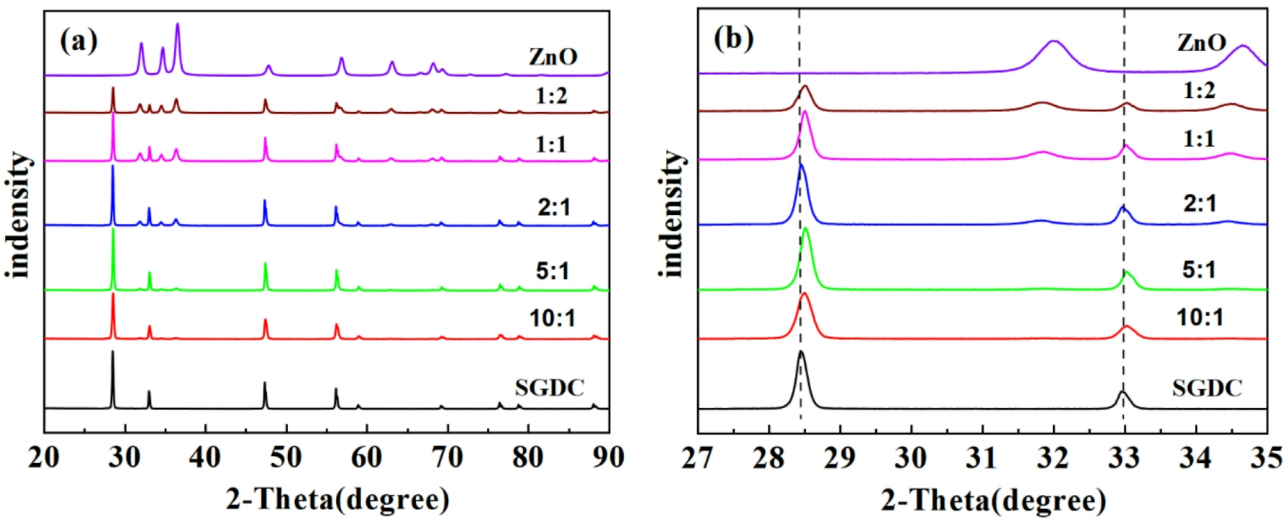


Figure 1
XRD patterns of SGDC, ZnO, and SGDC–ZnO composites (a); the displacement of the main reflections of SGDC in the composites with mass ratios of 10:1, 5:1, 2:1, 1:1, and 1:2 (b).

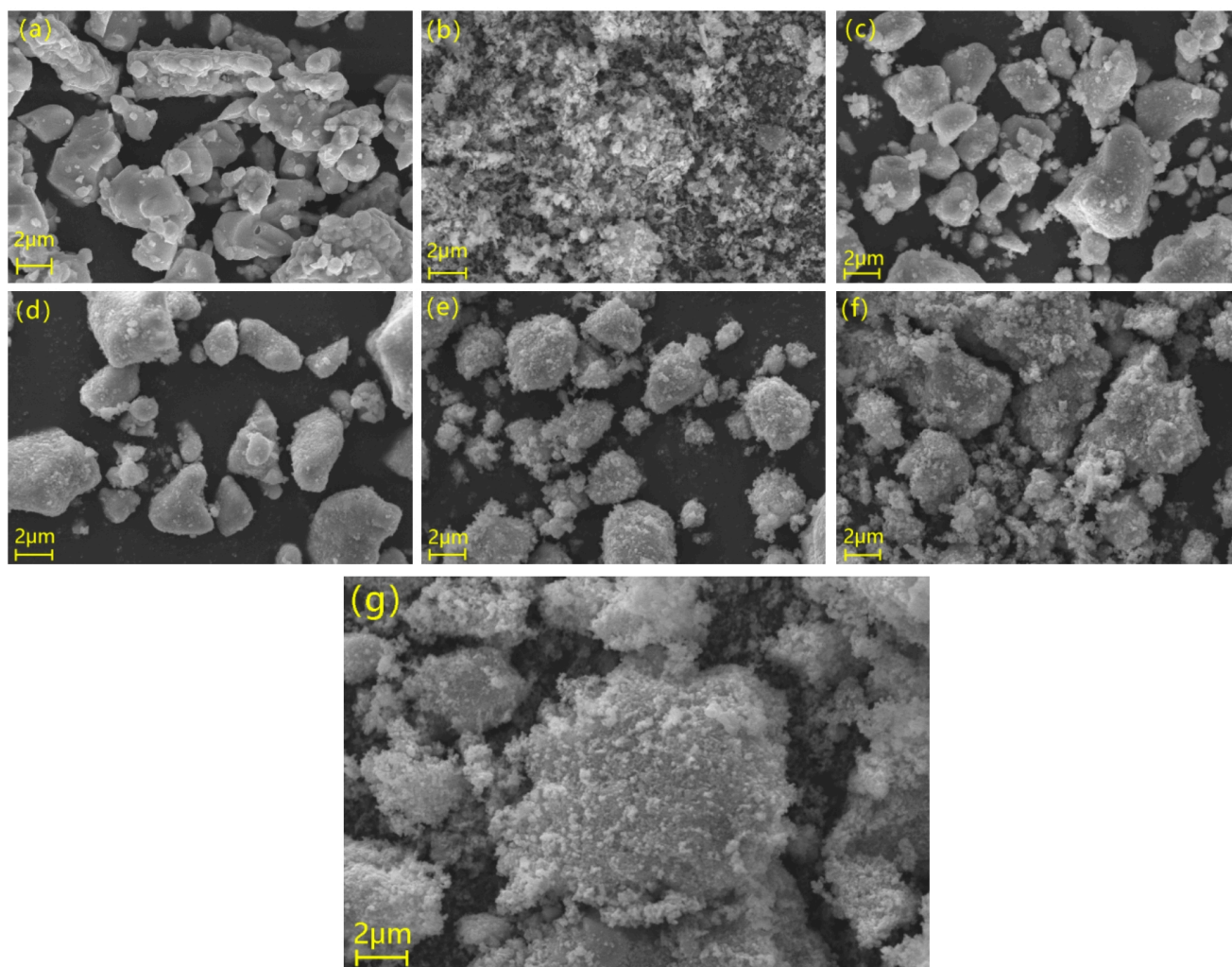


Figure 2

SEM images of (a) pure SGDC powder, (b) ZnO nanoparticles, and (c-g) SGDC-ZnO composite powders with SGDC:ZnO mass ratios of (c) 10:1, (d) 5:1, (e) 2:1, (f) 1:1, and (g) 1:2.

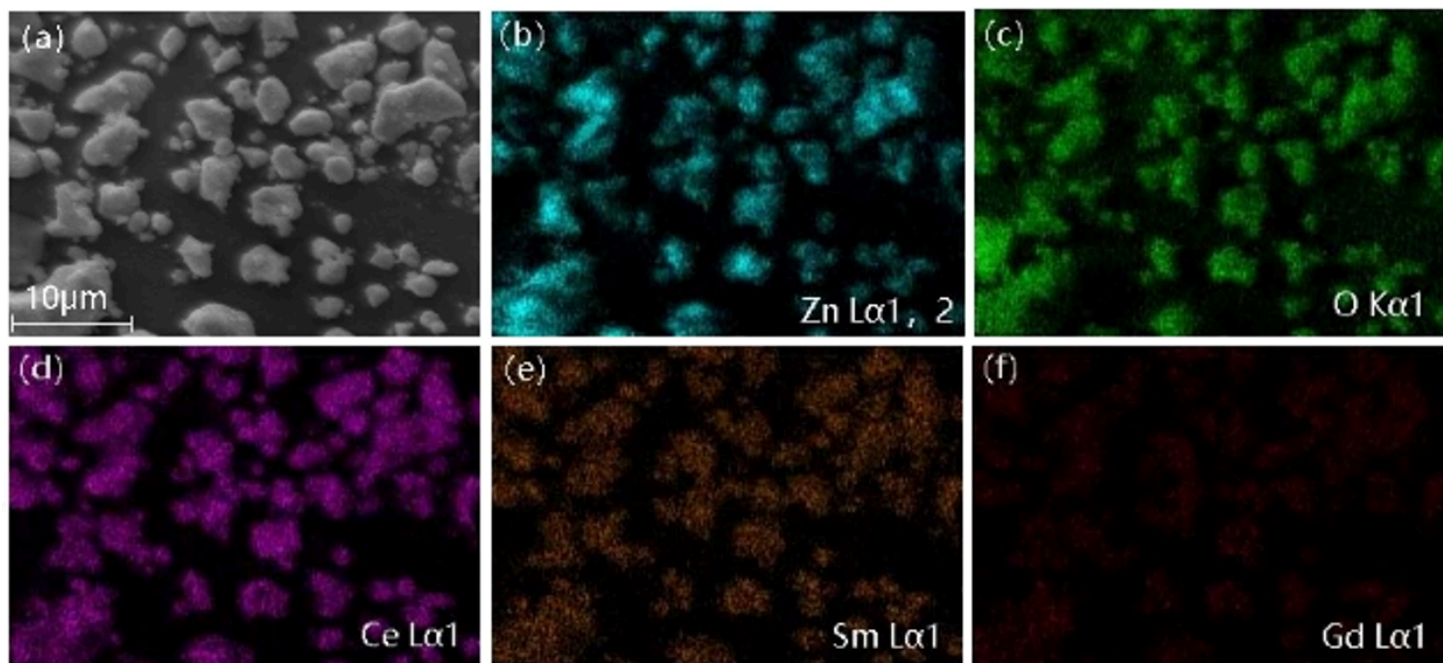


Figure 3

(a) SEM image of the 5SGDC-1ZnO (SGDC:ZnO = 5:1) composite showing the EDS mapping area, and (b-f) individual elemental distribution maps of Zn, O, Ce, Sm, and Gd.

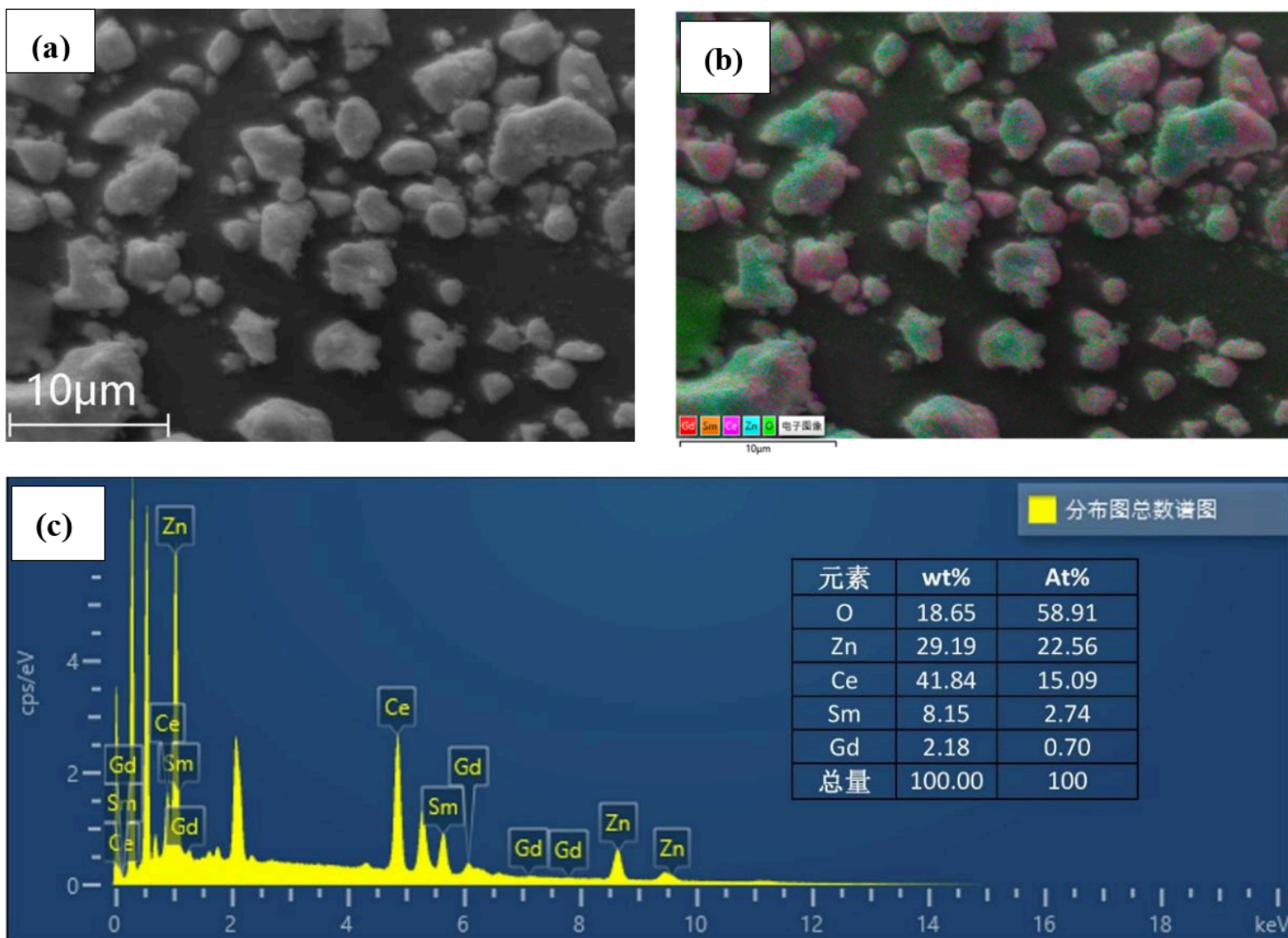


Figure 4

(a) SEM image of the 5SGDC-1ZnO (SGDC:ZnO = 5:1) composite, (b) combined EDS elemental overlay image showing the distribution of all detected elements (Zn, O, Ce, Sm, and Gd), and (c) semi-quantitative EDS analysis results.

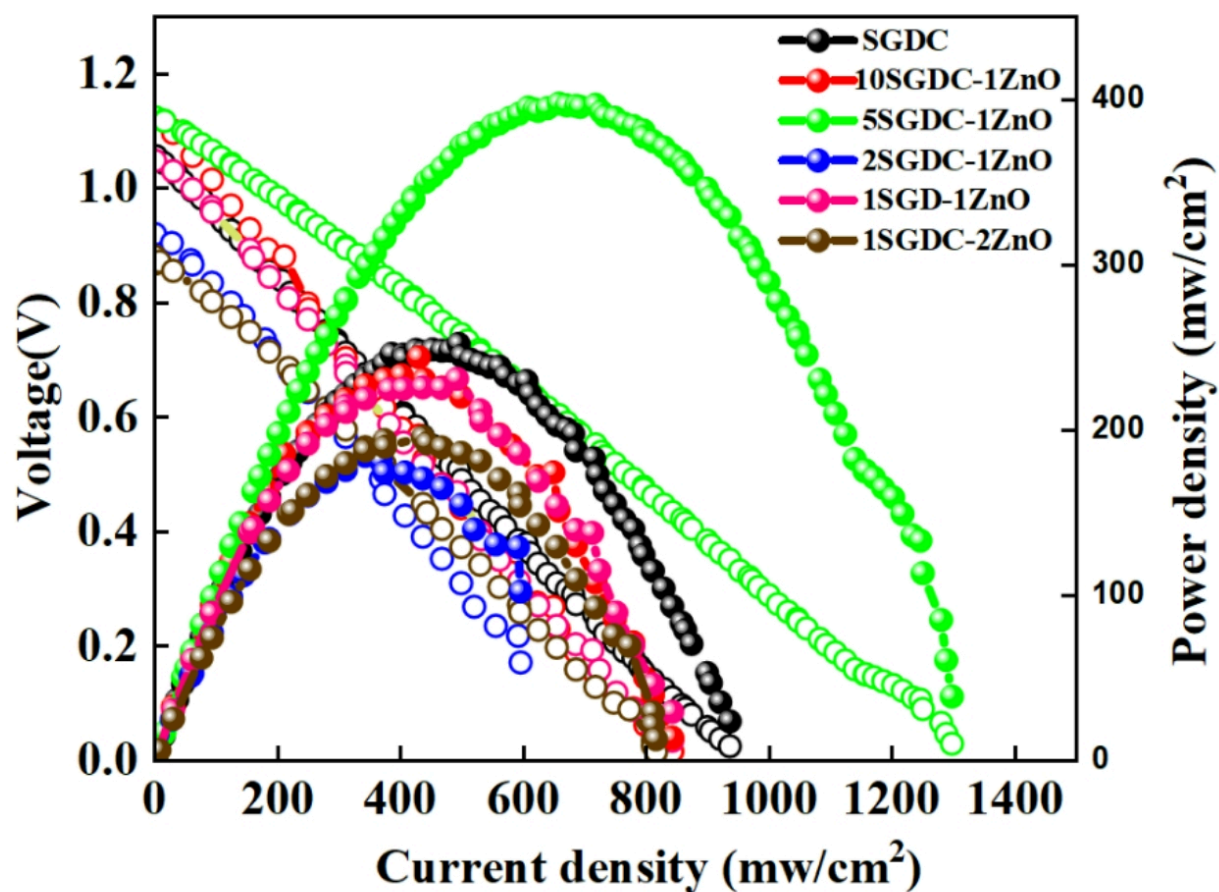


Figure 5

I-V and I-P curves of single cells with different SGDC:ZnO mass ratios (pure SGDC, 10:1, 5:1, 2:1, 1:1, and 1:2) measured at 550 °C. The solid and hollow symbols represent I-P and I-V curves, respectively.