

Article

Exsolved Nanoparticles Decorated Double Perovskites as High-Performance Anodes for Direct-Ammonia Solid Oxide Fuel Cells

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Abstract: Due to the high energy density, mature production technology, ease of storage and transportation, and the no carbon/sulfur nature of ammonia fuel, direct-ammonia solid oxide fuel cells (DA-SOFCs) have received rapidly increasing attention, showing distinct advantages over H₂-fueled SOFCs and low-temperature fuel cells. However, DA-SOFCs with conventional Ni-based cermet anodes still suffer from several drawbacks, including serious sintering and inferior activity for ammonia decomposition, strongly limiting the large-scale applications. To tackle the above-mentioned issues, exsolved NiCo nanoparticles decorated double perovskite oxides are fabricated and employed as high-performance anodes for DA-SOFCs in this work. By optimizing the Ni doping amount in Sr₂CoMo_{1-x}Ni_xO_{6-δ} (x = 0.1, 0.2 and 0.3), the reduced Sr₂CoMo_{0.8}Ni_{0.2}O_{6-δ} (r-SCMN2) anode exhibits superb catalytic activity for ammonia cracking reaction and high anti-sintering capability. More specifically, the electrolyte-supported single cell with r-SCMN2 nanocomposite anode delivers superior power outputs and operational durability in ammonia fuel as compared with other r-SCMN anodes owing to the significantly promoted nanoparticle exsolution and stronger interaction between alloy nanoparticles and the support. In summary, this study presents an effective strategy for the design of efficient and stable nanocomposite anodes for DA-SOFCs.

Keywords: solid oxide fuel cell; anode; perovskite oxide; ammonia fuel; nanoparticle exsolution



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1. Introduction

In recent years, due to ever-increasing environmental pollution and energy crisis caused by the excessive utilization of fossil fuels such as global warming and climate change, the development of advanced technologies for the utilization of alternative clean and sustainable energies has attracted increasing attention [1–7]. Fuel cells are considered as highly promising energy conversion devices due to low emissions and high efficiencies [8–14]. Particularly, solid oxide fuel cells (SOFCs) exhibit distinct merits over other fuel cells, such as all-solid structure and fuel flexibility [15–19]. In theory, all the combustible fuels can be used for SOFCs, such as hydrogen (H₂), ammonia (NH₃), and hydrocarbons [20–24]. However, the H₂ fuel for SOFCs suffers from several drawbacks, such as no natural sources and limited infrastructures for the transport and storage [25–28]. Compared to H₂, hydrocarbons, including methane and natural gas, are more promising as fuels for SOFCs owing to the ease in transportation and abundant resources. However, the conventional nickel (Ni)-based anodes in SOFCs suffer from severe sulfur poisoning and carbon deposition when using hydrocarbon fuels, greatly restricting the long-term stability of SOFCs [29–31]. Therefore, the utilization of carbon/sulfur-free fuels is highly crucial

for high-performance and durable SOFCs. Ammonia (NH_3) is a carbon/sulfur-free fuel with the advantages of high volumetric energy density, ease in transportation/storage, and matured synthesis technology. It has attracted particular interests as a promising fuel for SOFCs recently [32–35].

Based on the potential reactions in the anode chamber of direct-ammonia (DA)-SOFCs [36], the ideal anodes should show high (electro)catalytic activity for NH_3 decomposition reactions (NDR) and H_2 oxidation reactions (HOR). Although the conventional Ni-based anodes exhibited superior HOR activity, they still suffered from serious particle agglomeration and low NDR activity at lower temperatures, leading to inferior power outputs and operational durability of DA-SOFCs [37]. Therefore, the design and development of alternative anodes to replace Ni-based cermetes are highly crucial to achieve highly efficient and durable DA-SOFCs. Perovskite oxides have been regarded as promising anodes for DA-SOFCs due to their excellent structural and compositional flexibility and high chemical and thermal stability [38–42]. However, pure perovskite-based anodes suffer from low catalytic activity for NDR and HOR [43,44]. Thus, perovskite oxides have been employed as promising substrates to support in situ exsolved metal nanoparticles (NPs), exhibiting superior (electro)catalytic activity/stability for both NDR and HOR by stabilizing the surface NPs [36,45–47]. For instance, we have fabricated $\text{La}_{0.55}\text{Sr}_{0.30}\text{TiO}_{3-\delta}$ -based anode with exsolved NiCo alloy NPs for DA-SOFCs, showing considerable (electro)catalytic activity and stability owing to the strong interaction between the exsolved NPs and substrate, outstanding anti-sintering capability, and the balance of NH_3 adsorption and N_2 desorption processes. The single cell with exsolved NPs composite anode exhibited a high PPD of 361 mW cm^{-2} at 800°C and favorable operational stability of 120 h at 700°C operated on NH_3 fuel [45]. Nevertheless, such nanocomposite suffered from limited NDR catalytic activity, which was attributed to the inferior catalytic activity of $\text{La}_{0.55}\text{Sr}_{0.30}\text{TiO}_{3-\delta}$ substrate for NDR, restricting the further enhancement in the performance of DA-SOFCs. Therefore, it is important to design efficient perovskite oxide-based support with the exsolved NPs to realize highly efficient and durable DA-SOFCs. Very recently, we have reported a slightly Ru substitution strategy to facilitate the exsolution of alloy NPs in active $\text{Pr}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_{3-\delta}$ (PSCF)-based matrix. The slightly Ru doping effectively increased the amount of exsolved NPs with remarkably reduced particle sizes, and enhanced the nanoparticle/substrate interaction as compared with the Ru-free counterpart, remarkably boosting the (electro)catalytic performance of the anode for NDR and HOR [46]. DA-SOFCs with the Ru-doped anode exhibited much higher PPDs than DA-SOFCs, with PSCF anode at 800°C and better operational durability.

Double perovskite oxides ($\text{A}'\text{A}''\text{B}'\text{B}''\text{O}_{6-\delta}$), as an important class of perovskite oxides, have also been reported as efficient anodes for SOFCs due to the higher electrical conductivity and better chemical/thermal stability than those of single perovskites (ABO_3) [48–53]. However, there are few investigations of double perovskite oxides as anodes for DA-SOFCs due to the inferior NDR catalytic activity of pure-phase double perovskites. Thus, the combination of NPs exsolution and double perovskite-based substrates may be a potential and useful strategy to enhance the performance of perovskite-based anodes for DA-SOFCs. Herein, $\text{Sr}_2\text{CoMo}_{1-x}\text{Ni}_x\text{O}_{6-\delta}$ (SCMN, $x = 0.1, 0.2$, and 0.3 ; denoted as SCMN1, SCMN2, and SCMN3, respectively) double perovskites are employed as efficient substrates to anchor alloy NPs through exsolution for the application as anodes for DA-SOFCs. Upon optimizing the Ni doping amount in SCMN, SCMN2 nanocomposite anode exhibited the highest (electro)chemical activity for NDR and HOR due to the promoted NPs exsolution in terms of smaller particle size and higher amounts of exsolved NiCo alloy NPs, much improved anti-sintering capability, and enhanced interaction between NPs and double perovskite matrix. Consequently, SCMN2-based DA-SOFCs delivered higher power outputs and better durability than those with SCMN1, SCMN3, and conventional Ni-based cermet anodes. This work provides an effective and simple strategy for designing highly anti-sintering, active, and stable nanocomposite anodes for DA-SOFCs, promoting the practical applications of this technology.

2. Results and Discussion

The crystal structural information of various SCMNs and reduced $\text{Sr}_2\text{CoMo}_{1-x}\text{Ni}_x\text{O}_{6-\delta}$ (r-SCMN, $x = 0.1, 0.2$, and 0.3 ; denoted as r-SCMN1, r-SCMN2, and r-SCMN3, respectively) perovskite oxides were analyzed by X-ray diffraction (XRD), with results displayed in Figure 1a,b and Figure S1. Similar to $\text{Sr}_2\text{CoMoO}_{6-\delta}$ (SCM), the as-synthesized SCMN samples exhibited a tetragonal structure and no NiO phases were detected [54], demonstrating the successful doping of Ni into the SCMN perovskite lattice (Figure 1a). As depicted in Figure 1b and Figure S1, after the treatment at 800°C in pure H_2 for 1 h, three SCMN samples with different Ni doping contents exhibited phase transitions accompanied by the exsolution of NiCo NPs and/or the impurity phases [45]. It should be noted that excessive exsolution of B-site cations in SCMN with higher Ni doping concentration may lead to the destruction of the perovskite structure [55]. The main phase of SCMN1 and SCMN2 was changed from $\text{Sr}_2\text{CoMoO}_{6-\delta}$ to Sr_3MoO_6 (JCPDS No.00-027-1441) [56] while the main phases of SCMN3 were $\text{Sr}_3\text{Mo}_2\text{O}_7$ (JCPDS No.00-052-1252) [57] and SrMoO_4 (JCPDS No.01-085-0809) [58] after the reduction. H_2 temperature-programmed reduction (H_2 -TPR) was used to investigate the reducibility and NPs/substrate interaction in various SCMN samples as depicted in Figure 1c. As can be seen, the SCMN samples with larger Ni doping amounts exhibited much higher NPs/substrate interaction as evidenced by the higher temperatures on H_2 consumption peaks. SEM was employed to obtain the morphological information of various SCMN perovskites before and after the reduction treatment as depicted in Figure S2 and Figure 1d–f. The as-prepared SCMN samples showed smooth surface and micron-sized particles (Figure S1). After reduction, numerous NPs were exsolved from the double perovskite lattice, which were uniformly distributed on the surface of SCMN substrate. Furthermore, r-SCMN2 displayed a much larger amount of exsolved NiCo NPs with remarkably reduced particle sizes as compared with the r-SCMN1 and r-SCMN3 (Figure 1d–f), leading to the increased Brunauer–Emmett–Teller (BET) specific surface area of r-SCMN2, as depicted in Figure S3.

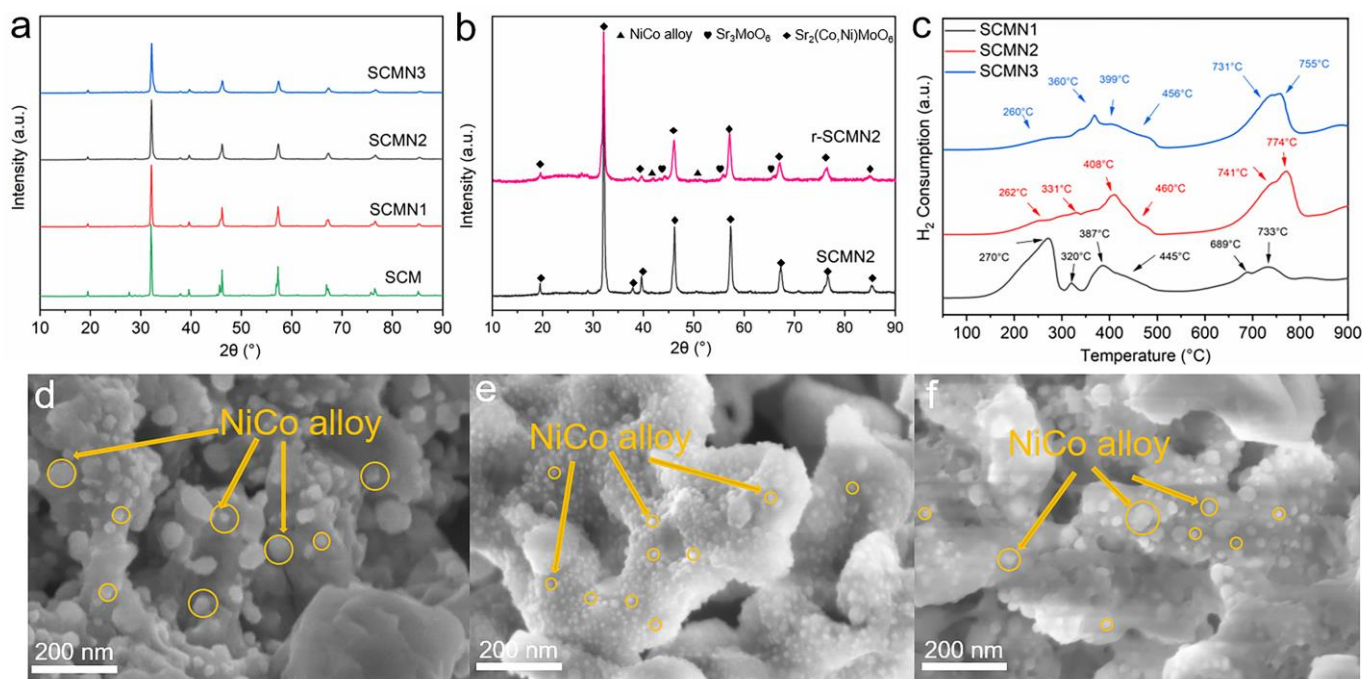


Figure 1. (a) XRD patterns of various SCMN samples. (b) XRD patterns of SCM2 and r-SCMN2 samples. (c) H_2 -TPR profiles of various SCMN samples. SEM images of (d) r-SCMN1, (e) r-SCMN2, and (f) r-SCMN3 samples.

To further investigate the exsolution behavior of alloy NPs in r-SCMN1, r-SCMN2, and r-SCMN3 samples, scanning transmission electron microscope (STEM)-energy-dispersive X-ray (EDX) and high-resolution (HR)-transmission electron microscopy (TEM) were employed as displayed in Figures 2, S5 and S6. Numerous NPs with an average particle size of ~ 20 nm were observed in r-SCMN2 anode, which were well distributed and partially anchored on the substrate surface, indicating a strong interaction between the NPs and the substrate (Figure 2a) [59]. Based on the HR-TEM images in Figure 2b, lattice spacings of around 0.291, 0.197, and 0.205 nm were observed in r-SCMN2 anode, corresponding to the (440), (002), and (111) diffraction planes of Sr_3MoO_6 , $\text{Sr}_2(\text{CoNi})\text{MoO}_6$, and NiCo alloy, respectively. On the contrary, r-SCMN1 and r-SCMN3 samples exhibited a much smaller amount of exsolved NiCo alloy NPs with a much larger average particle size of ~ 50 nm than those of r-SCMN2 (Figures S4 and S5), demonstrating that the proper Ni doping amount in SCM was beneficial for the exsolution of NiCo alloy NPs. The STEM-EDX results also confirmed the successful exsolution of NiCo alloy NPs in all the three reduced SCM samples (Figures 2c,d, S4 and S5).

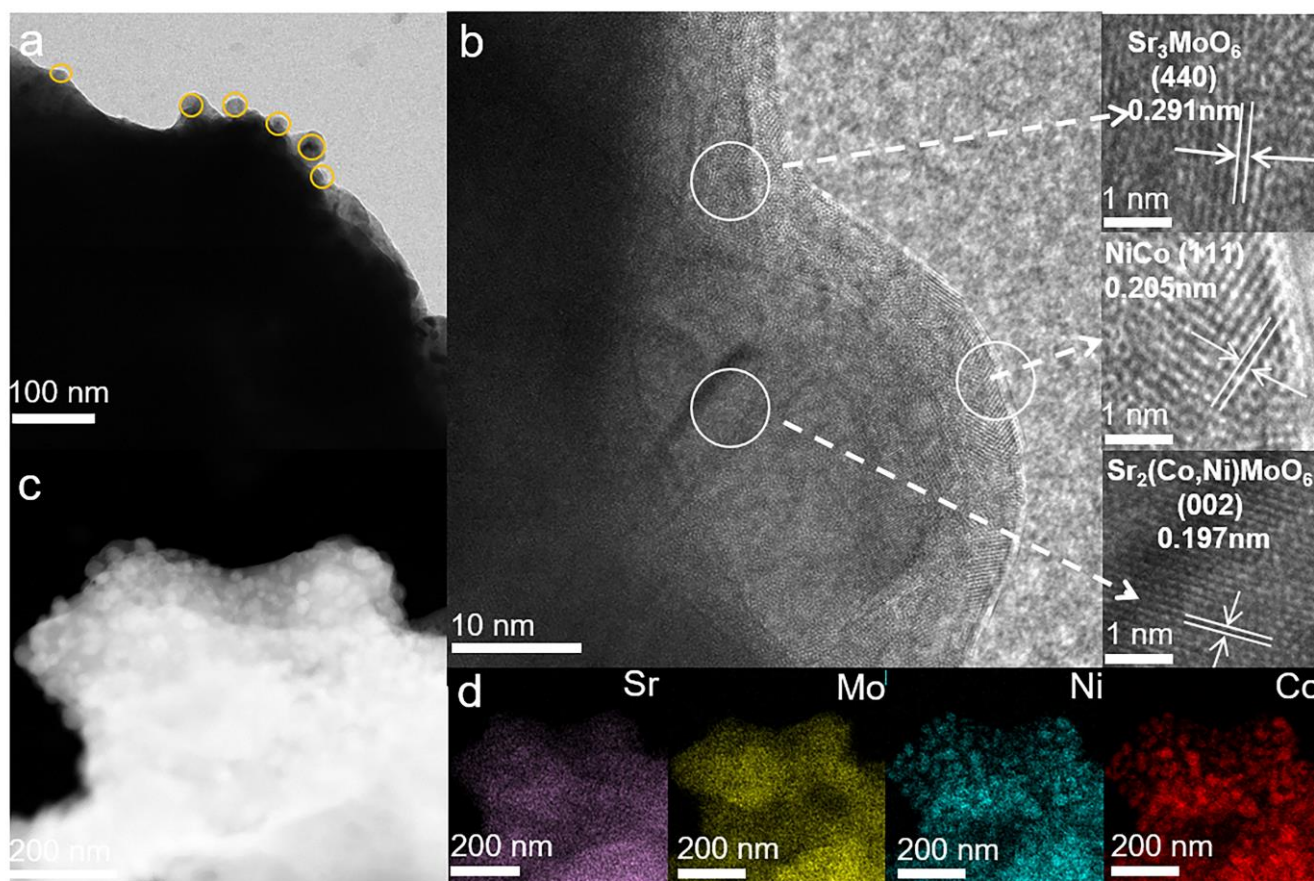


Figure 2. (a) TEM, (b) HR-TEM, (c) STEM, and (d) EDX mapping images of r-SCMN2 sample.

X-ray photoelectron spectroscopy (XPS) technique was used to analyze the oxidation states of various SCM and r-SCMN anodes, as depicted in Figures 3 and S6. As can be seen, no obvious changes were observed in the Sr 3d spectra of SCM2 and r-SCMN2 samples (Figure 3b), while the $\text{Mo}^{5+}/\text{Mo}^{6+}$ ratio of SCM2 sample was slightly increased after reduction as depicted in Figure 3c. In addition, the Co 2p spectra of unreduced SCM samples showed two broad peaks (Figure 3d), which were fitted to two subpeaks of Co^{3+} (796.9 and 780.0 eV) and Co^{2+} (795.1 and 781.7 eV), respectively. After reduction, XPS spectra for Co cations of r-SCMN2 were fitted into three subpeaks of Co^{3+} (795.5 and 780.0 eV), Co^{2+} (797.2 and 781.5 eV), and Co^0 (794.4 and 779.2 eV), indicating the successful Co cation exsolution from the perovskite lattice [55,60]. Similar to Co 2p, the Ni 2p in

SCMN2 were mainly expressed as Ni^{2+} species (855.6 eV), while Ni^0 species were observed at 853.8 eV in Ni 2p XPS spectra of r-SCMN2 [47,61]. Furthermore, the amount of Co^0 species in r-SCMN2 was much higher than those of r-SCMN1 and r-SCMN3, demonstrating the promoted NiCo alloy NPs exsolution induced by proper Ni doping concentration.

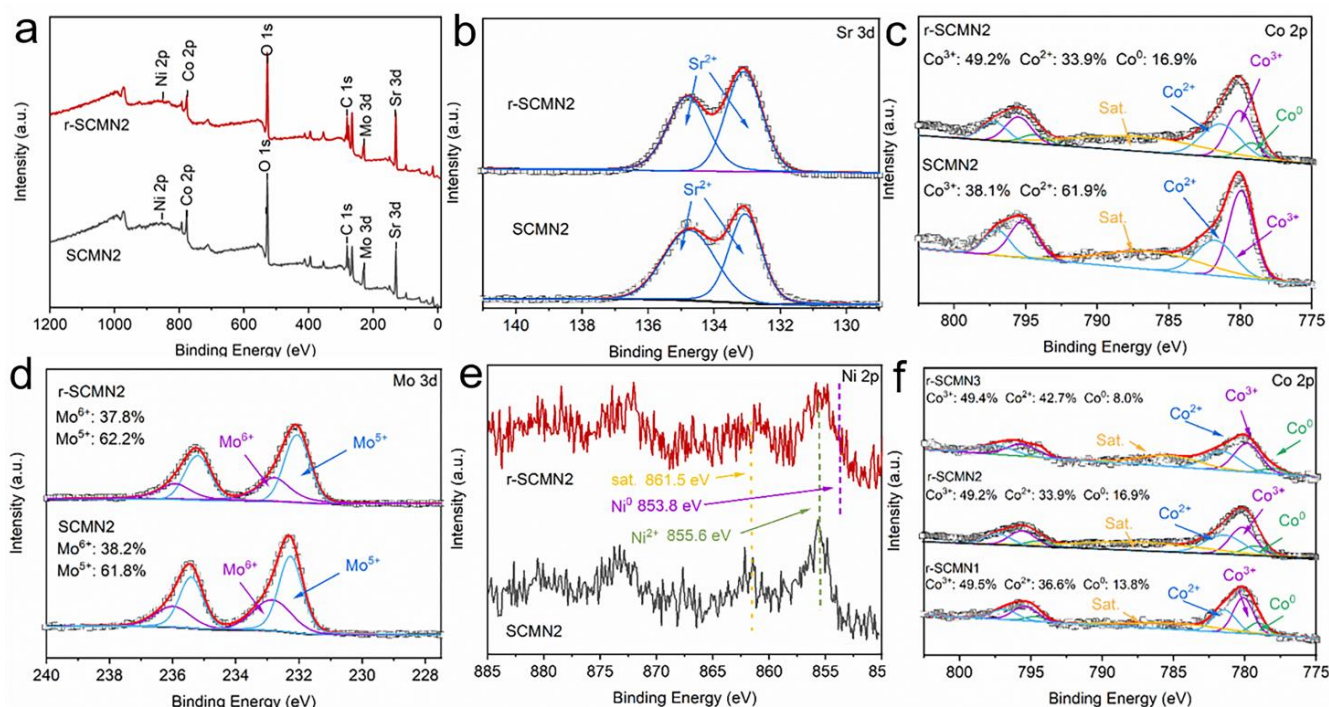


Figure 3. XPS spectra of SCMN2 and r-SCMN2 samples: (a) XPS survey, (b) Sr 3d, (c) Co 2p, (d) Mo 3d, and (e) Ni 2p. (f) Co 2p spectra of r-SCMN1, r-SCMN2, and r-SCMN3 samples.

It is well known that ideal anodes for DA-SOFCs should exhibit excellent catalytic activity and durability for NDR. Thus, the catalytic activities of various r-SCMN anodes for NDR were evaluated as displayed in Figure 4a. Three r-SCMN anodes all exhibited high NH_3 conversion rates of around 100% at 750–800 °C. In addition, the r-SCMN2 showed higher catalytic activity than those of r-SCMN1 and r-SCMN3 at reduced temperatures due to the higher amount and smaller particle sizes of exsolved NiCo alloy NPs. For instance, the r-SCMN2 anode exhibited a much higher NH_3 conversion rate of 89.4% at 650 °C than those of r-SCMN1 (79.3%) and r-SCMN3 (71.6%). As depicted in Figure 4b, the durability of r-SCMN2 sample for NDR at 800 °C was also tested. As can be seen, the r-SCMN2 anode showed a highly durable NH_3 conversion rate at around 98% during 242 h for NDR test, highlighting the superior stability of r-SCMN2 anode for DA-SOFCs.

To evaluate the performance of various r-SCMN anodes in practical DA-SOFCs, electrolyte-supported SOFCs with a configuration of r-SCMN anode/ $\text{Sm}_{0.2}\text{Ce}_{0.8}\text{O}_{1.9}$ (SDC) electrolyte/ $\text{Sm}_{0.5}\text{Sr}_{0.5}\text{CoO}_{3-\delta}$ (SSC) cathode were prepared and tested. The thicknesses of the anode, electrolyte, and cathode were around 15, 350, and 15 μm , respectively (Figure S7). To estimate the potential phase reaction between SDC electrolyte and r-SCMN anodes, SDC and SCMN2 samples were mixed by physical mixing at a weight ratio of 1:1, which was then annealed in air at 1000 °C for 2 h. As displayed in Figure S8, no obvious phase reactions between the SDC electrolyte and r-SCMN2 anode were observed based on the XRD patterns. As depicted in Figure S9a,e, the cell with r-SCMN2 anode delivered high peak power densities (PPDs) of 465, 380, 314, and 239 mW cm^{-2} at 800, 750, 700, and 650 °C in H_2 and 350, 266, 194, 141 mW cm^{-2} at 800, 750, 700, and 650 °C in NH_3 . For comparison, the single cell with Ni-SDC anode was also tested in H_2 and NH_3 fuels. The cells with r-SCMN1, r-SCMN2, r-SCMN3, and Ni-SDC anodes generated PPDs of 437, 465, 409, and 281 mW cm^{-2} at 800 °C in H_2 , respectively (Figure 5a). The cells with r-SCMN2 exhibited

a high PPD of 437 mW cm^{-2} at 800°C in H_2 , which was 1.06, 1.13, and 1.65 times more than those of the cells with r-SCMN1, r-SCMN3, and Ni-SDC anodes, respectively. After switching from H_2 to NH_3 fuel, the cells with r-SCMN1, r-SCMN2, r-SCMN3, and Ni-SDC anodes produced PPDs of 327, 350, 296, and 219 mW cm^{-2} at 800°C in NH_3 , respectively (Figure 5b). The r-SCMN2-based cell shows the high PPD of 350 mW cm^{-2} at 800°C in NH_3 , which was 1.07, 1.18, and 1.59 times higher than those of the cells with r-SCMN1, r-SCMN3, and Ni-SDC anodes, respectively. The higher power outputs of r-SCMN2 anode-based single cell in H_2 and NH_3 fuels were attributed to the higher amount and smaller particle sizes of exsolved NiCo NPs, providing more active centers for NDR and HOR. However, the cells with r-SCMN1, r-SCMN2, r-SCMN3, and Ni-SDC anodes exhibited lower PPDs in NH_3 fuel than those of the cells that operated on H_2 fuel (Figure S9 and Table S1), which may be assigned to the unreacted NH_3 and/or the dilution effect of N_2 products.

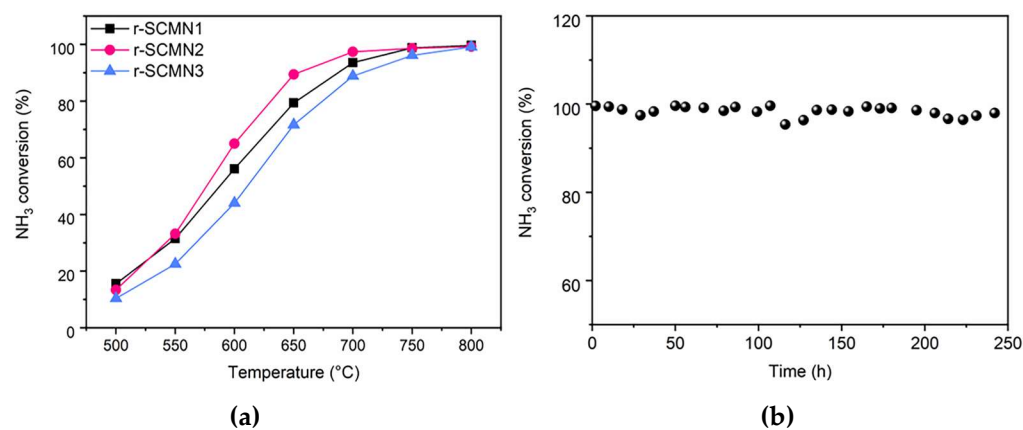


Figure 4. (a) Catalytic activity of various r-SCMN anodes for NDR at $500\text{--}800^\circ\text{C}$. (b) Catalytic stability test of the r-SCMN2 anode for NDR at 800°C .

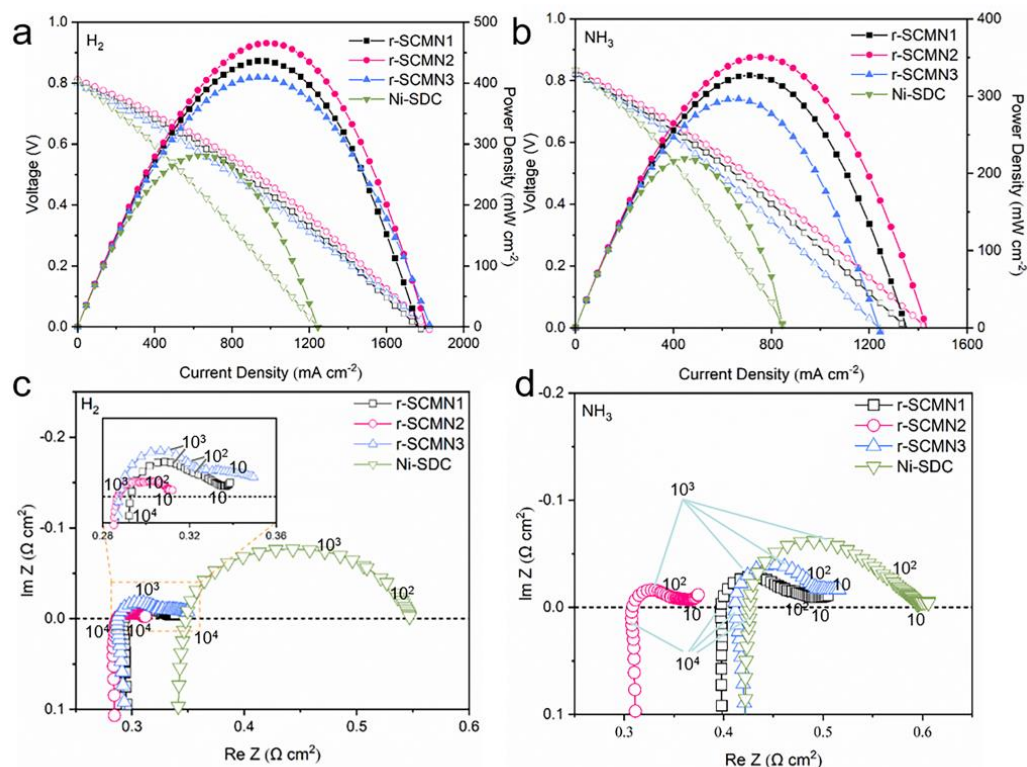


Figure 5. I - V and I - P curves of single cell with various r-SCMN and Ni-SDC anodes in (a) H_2 and (b) NH_3 fuels at 800°C with corresponding EIS spectra in (c) H_2 and (d) NH_3 fuel.

Based on the electrochemical impedance spectroscopy (EIS) profiles of single cells with various anodes in Figure 5c,d and Table S2, the cell with r-SCMN2 exhibited much lower polarization resistance (R_p) compared to the r-SCMN1, r-SCMN3, and Ni-SDC anodes in both H_2 and NH_3 fuels, agreeing well with the I - V and I - P results, which was assigned to the higher (electro)catalytic activity for NDR and HOR. The r-SCMN2-based cell exhibited low R_p of 0.027, 0.041, 0.063, and 0.198 $\Omega\text{ cm}^2$ in H_2 at 800 to 650 $^\circ\text{C}$ and 0.041, 0.054, 0.158, 0.410 $\Omega\text{ cm}^2$ in NH_3 at 800 to 650 $^\circ\text{C}$. More specifically, the r-SCMN2-based cell exhibited a much lower R_p of 0.063 $\Omega\text{ cm}^2$ than those of the cells with r-SCMN1 (0.095 $\Omega\text{ cm}^2$), r-SCMN3 (0.203 $\Omega\text{ cm}^2$), and Ni-SDC (0.437 $\Omega\text{ cm}^2$) as anodes in NH_3 fuel at 800 $^\circ\text{C}$. It should be noted that both the ohmic resistance (R_o) and R_p values of the cells in NH_3 fuel were obviously larger than those of H_2 -fueled SOFCs with various anodes, as shown in Figure S10, due to the endothermic nature of NDR and the incomplete decomposition of NH_3 fuel, leading to decreased real operational temperatures and increased R_o/R_p values. In addition, the power output of DA-SOFCs with r-SCMN2 anode was higher than most of the DA-SOFCs with reported anodes in the literature (Table S3) [31,45,46,62–64], further demonstrating the superiority of r-SCMN2 nanocomposite anode for DA-SOFCs.

It has been reported that the conventional Ni-based anodes suffered from severe sintering operated on NH_3 fuel at elevated temperatures, strongly restricting the long-term operational durability of DA-SOFCs [65]. Therefore, it is important to evaluate the operational durability of DA-SOFCs with alternative perovskite-based anodes. The stability tests of the cells with various r-SCMN and Ni-SDC anodes were conducted under the conditions of NH_3 fuel, 100 mA cm^{-2} , and 800 $^\circ\text{C}$, with the results displayed in Figure S11. The SOFCs with r-SCMN1, r-SCMN2, r-SCMN3, and Ni-SDC anodes exhibited voltage degradation rates of 0.91, 0.74, 0.98, and 14.6 mV h^{-1} , respectively. It is clearly observed that DA-SOFCs with r-SCMN anodes showed superior operational stability to that of the cell with Ni-SDC anode, which was assigned to the better anti-sintering capability of r-SCMN anodes than that of Ni-SDC anode in NH_3 fuel (Figure S12). Furthermore, r-SCMN2-based cell displayed better durability for 250 h than those of r-SCMN1 and r-SCMN3-based cells in NH_3 fuel, which was attributed to the stronger interaction between the double perovskite substrate and exsolved NPs, which effectively inhibited the NPs aggregation.

3. Materials and Methods

3.1. Synthesis and Fabrication

The powders of SCMN anodes, SSC cathode, and SDC electrolyte were synthesized via a sol–gel route [66,67]. For a typical synthesis, stoichiometric amounts of $\text{Sr}(\text{NO}_3)_2$, $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, $(\text{NH}_3)_4\text{Mo}_7\text{O}_{24} \cdot 6\text{H}_2\text{O}$, and $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ were heated at 80 $^\circ\text{C}$ under stirring. Ethylenediaminetetraacetic acid (EDTA) and citric acid (CA) were added to act as complexing agents at a molar ratio of 1:1:2 for total metal ions, EDTA and CA, in order to mix the metal ions evenly. $\text{NH}_3 \cdot \text{H}_2\text{O}$ was used to adjust the pH value of the aqueous solution to around 7. An atropurpureous gel was obtained after the removal of water in the solution by evaporation, which was pre-decomposed at 200 $^\circ\text{C}$ to obtain the precursors. The as-synthesized precursors were then annealed at 1100 $^\circ\text{C}$ for 6 h in air to obtain the final products. The SSC cathode and SDC electrolyte were also prepared via the same method, and the samples were annealed at 1000 $^\circ\text{C}$ for 5 h and 800 $^\circ\text{C}$ for 5 h, respectively. The r-SCMN samples were prepared by treating the SCMN powders in pure H_2 atmosphere at 800 $^\circ\text{C}$ for 1 h. The SDC pellets were obtained by dry-pressing and were annealed at 1400 $^\circ\text{C}$ for 10 h. The anode and cathode slurry were prepared by mixing the powders in isopropanol, glycol and glycerol with a ball milling process (30 min, 400 rpm), spray-painted on the different sides of the electrolyte with an active area of 0.45 cm^2 , which were annealed at 1000 $^\circ\text{C}$ for 2 h in air. Finally, silver pastes and silver wires were used as collector and current lead for single cell, respectively. The operation temperature range of single cell ranged from 650 to 800 $^\circ\text{C}$.

3.2. Characterization

XRD (D8 advance Bruker, Germany) with Cu-K α radiation was employed to analyze the crystal structures of the samples by using a receiving slit of approximately 0.2–0.4 mm. The data were tested in a step-scan model between $2\theta = 10\text{--}90^\circ$ with intervals of 0.02° . TEM (FEI Tecnai G2 F30), STEM, and field emission scanning electron microscopy (FE-SEM, JEOL-S4800) were used to examine the microstructures of the powders and single cells. EDX analyzer (FEI Tecnai G2 F30 STWIN) was employed to obtain EDX mapping images operated at 300 kV. Thermo ESCALAB 250 was used to acquire XPS spectra. BET-specific surface areas of various samples were investigated by obtaining the nitrogen adsorption–desorption isotherms on a Quantachrome AutoSorb-iQ3 instrument at liquid nitrogen temperature. H₂-TPR was investigated by the BELCAT-A equipment with a thermal conductivity detector (TCD). The catalytic activities of various samples for NDR were tested in a quartz tube reactor at 500–800 °C [68]. About 0.2 g of anode particles were placed in the reactor. The flow rates of NH₃ and Ar (diluting gas) were fixed at 10 and 80 mL min^{−1} [STP], respectively. A gas chromatograph (GC-9860) with a chromatographic column (PLOT Q) and a TCD was used to analyze the gas compositions after NDR. The Keithley 2420 source meter was used to measure *I*-*V* and *I*-*P* curves of single cells through the 4-probe mode. The H₂ and NH₃ fuels were fixed at 80 mL min^{−1} [STP] at the anode side of the cell, while the cathode side was exposed to the air. EIS profiles of single cells were acquired by a Solartron 1260 frequency response analyzer and Solartron 1287 potentiostat under open circuit voltage conditions, with frequencies ranging from 0.1 to 1 MHz.

4. Conclusions

In this work, SCM double perovskites were employed as efficient substrates to construct nanocomposite anodes for DA-SOFCs by NPs exsolution strategy. By tailoring the Ni doping concentration in SCM, the r-SCMN2 nanocomposite exhibited superior (electro)catalytic activity for NDR and HOR, which was attributed to the promoted alloy NPs exsolution and enhanced interaction between substrate and NPs as compared with r-SCMN1 and r-SCMN3 anodes. The single cell with r-SCMN2 anode displayed a high PPD of 350 mW cm^{−2} and a reasonable operational stability of 250 h with a voltage degradation rate of 0.74 mV h^{−1} in NH₃ fuel at 800 °C, outperforming the cells with the r-SCMN1, r-SCMN3, and Ni-SDC anodes because of the better anti-sintering capability, stronger interfacial interaction, and higher amount of active centers for NDR. This work reports a highly active and durable double perovskite-based nanocomposite anode for DA-SOFCs, accelerating the practical applications of DA-SOFCs.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/catal13060996/s1>, Figure S1: XRD patterns of reduced SCM and SCM samples; Figure S2: SEM images of (a) SCM1, (b) SCM2 and (c) SCM3 samples; Figure S3: Nitrogen adsorption/desorption isotherms of SCM1, SCM2 and SCM3 samples (a) before and (b) after the reduction; Figure S4: (a) TEM, (b) HR-TEM, (c) STEM and (d) EDX mapping images of r-SCMN1 sample; Figure S5: (a) TEM, (b) HR-TEM, (c) STEM and (d) EDX mapping images of r-SCMN3 sample; Figure S6: XPS spectra of SCM1 and SCM3 samples before and after the reduction: (a,d) XPS survey, (b,e) Co 2p and (c,f) Mo 3d; Figure S7: Typical cross-sectional SEM images of a fuel cell with r-SCMN2 anode; Figure S8: XRD patterns of SDC, SCM2 and SDC+SCMN2 composite (1:1, weight ratio) prepared by physical mixing after a calcination at 1000 °C for 2 h in air; Figure S9: *I*-*V* and *I*-*P* curves of SOFCs with (a,d) r-SCMN1, (b,e) r-SCMN2 and (c,f) r-SCMN3 anodes operated on H₂ (a–c) and NH₃ (d–f) fuels at 650–800 °C; Figure S10: EIS spectra of SOFCs with (a,d) r-SCMN1, (b,e) r-SCMN2 and (c,f) r-SCMN3 anodes operated on H₂ (a–c) and NH₃ (d–f) fuels at 650–800 °C; Figure S11: Operational stability of single cells with r-SCMN1, r-SCMN2, r-SCMN3 and Ni-SDC anodes operated on NH₃ fuel under a certain current density of 100 mA cm^{−2} at 800 °C. Figure S12: SEM images of (a,b) Ni-SDC, (c,d) r-SCMN1, (e,f) r-SCMN2 and (g,h) r-SCMN3 anodes before (a,c,e,g) and after (b,d,f,h) the treatment in NH₃ fuel at 800 °C for 30 h; Table S1: PPDs comparison of the single cells with r-SCMN anodes at different temperatures in H₂ and NH₃ fuels; Table S2. R_p of the single cells with r-SCMN anodes at different temperatures in H₂ and NH₃ fuels;

Table S3: PPDs comparison of DA-SOFCs with r-SCMN1, r-SCMN2 and r-SCMN3 anodes developed in this work and other representative anodes reported in the literature.

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