# INFLUENCE OF DIFFERENT RATIOS OF GDC-SDC ON SSC/GDC–SDC DUAL COMPOSITE CATHODE FOR INTERMEDIATE-TEMPERATURE SOLID OXIDE FUEL CELLS

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**Abstract**. Solid oxide fuel cells (SOFCs) have emerged as highly promising contenders in the field of energy conversion, offering enhanced efficiency and environmental sustainability. A considerable amount of effort has been devoted to developing a version of SOFC that operates at intermediate temperatures. Thus, new material development is necessary to enhance cell performance due to limited operating temperatures. The primary objective of this study is to identify the potential of samarium strontium cobaltite (SSC), gadolinium-doped ceria (GDC) and samarium-doped ceria (SDC), i.e. SSC/GDC–SDC dual composite, as a cathode material for intermediate-temperature SOFC (IT-SOFC). SSC/GDC–SDC dual composite cathode powders were developed through high-energy ball milling and mixing at a fixed weight ratio of 50% SSC and 50% GDC–SDC. Meanwhile, the weight ratio of the GDC–SDC electrolyte powders used in this study was within the range of 50%–70% GDC. The dual composite cathode powders were calcined at 750 °C, and pellets were sintered at 600 °C. The chemical compatibility and microstructure properties of the composite powders were examined via X-ray diffraction and scanning electron microscope, respectively. The porosity and density of the composite cathode pellets were also measured. A good chemical compatibility between the SSC cathode and GDC–SDC electrolyte powders was observed when no secondary phase existed after the mixing of the dual composite cathode powders. The dual composite cathodes produced an acceptable porosity (40%–50%) of the cathode component for SOFC application. Finally, the findings of this study significantly contribute to the investigation of SSC/GDC–SDC as a potential dual composite cathode material for IT-SOFCs.

**Keywords:** Cathode, dual composite, gadolinium-doped ceria (GDC), samarium-doped ceria (SDC), samarium strontium cobaltite (SSC)

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## 1. INTRODUCTION

Solid oxide fuel cells (SOFCs) are cutting-edge energy conversion and storage technologies renowned for their fuel versatility, high efficiency, and environmental friendliness [1]. These cells, like reversible solid oxide fuel cells (RSOCs), have the potential to be very efficient in multi-energy distributed systems [2]. Recent advances in SOFC technology have addressed obstacles and investigated potential future directions [3]. SOFCs excel in converting gaseous fuels' chemical energy into electrical energy via electrochemical reactions, making them extremely efficient energy converters [4]. However, one major problem in the field of intermediate-temperature SOFCs (IT-SOFCs) is the use of conventional cathode components. This problem limits the operating temperatures of cells. The demand for new materials to boost cell performance at intermediate temperatures has driven research efforts towards the development of revolutionary cathode materials [5].

Solid oxide fuel cells (SOFCs) create energy by a sequence of electrochemical processes at the cathode, anode, and electrolyte interfaces. The cathode undergoes oxygen reduction processes, whereas the anode undergoes hydrogen oxidation reactions. These reactions are critical to the overall performance and efficiency of SOFCs. Electrode/electrolyte interfaces are critical for promoting electrochemical reactions and understanding the link between electrochemical processes and microstructure. In SOFCs, the primary chemical reactions, such as oxygen reduction and oxygen ion diffusion, take place at the cathode/electrolyte interface, which has a substantial impact on their performance.

The study of dual composite cathodes is critical for improving battery performance. There are numerous advantages to adjusting the electrolyte composition in dual composite cathodes. For example, using dual-phase electrolytes can increase anode stability and cathode kinetics. Incorporating improved electrolytes, such as double-layer composite gel polymer electrolytes, can also limit the dissolution of organic cathode materials, increasing cyclic stability.

SOFCs are classified according to their working temperatures, with IT-SOFCs operating at intermediate temperatures versus high-temperature SOFCs. The discovery of cathode materials suited for IT-SOFCs is critical for increasing the utility and efficiency of these energy conversion devices. One strategy to overcome this difficulty is through the notion of embedded modification, wherein twin composite cathodes are developed to increase the performance of SOFCs at intermediate temperatures [6]. Although the potential for dual composite cathodes, such as samarium strontium cobaltite (SSC)/gadolinium-doped ceria (GDC)–samarium-doped ceria (SDC), in IT-SOFC applications is considerable, this study's issue statement should be clarified. This work intends to evaluate the chemical compatibility and microstructure features of SSC/GDC–SDC dual composite cathodes, focusing on porosity and density analyses to assess their suitability for SOFC applications at intermediate temperatures.

Recently, researchers have investigated composite cathodes as a viable method for improving the performance of IT-SOFCs. These cathodes include the combination of several materials and have demonstrated potential in this regard. The composite cathode known as SSC/GDC–SDC, which comprised $Sm_{0.5}Sr_{0.5}CoO_3$ (SSC) as the cathode material, $Gd_{0.1}Ce_{0.9}O_2$ (GDC) as the electrolyte and $Sm_{0.2}Ce_{0.8}O_2$ (SDC) as an additional cathode material, achieved encouraging outcomes in one study [7]. According to [7], the SSC material exhibited favourable electronic conductivity and catalytic activity for oxygen reduction, whilst the GDC and SDC materials had notable ionic conductivity.

At present, the utilisation of SSC perovskite oxide as a cathode material has elicited attention due to its promising characteristics in terms of combined ionic and electronic conductivity. Solid-state electrolytes, such as SDC, have a unique mix of ionic and electrical conductivity, making them interesting materials for various energy storage and conversion applications. SSC has high oxygen ion conductivity because of the existence of oxygen vacancies, allowing for efficient oxygen ion transport inside a material. Previous studies have indicated that the utilisation of an SSC cathode frequently involves the combination of an ionic conducting ceria-based electrolyte, such as GDC or SDC [8].

The use of SOFCs with composite electrolytes has resulted in the integration of alkaline salts into SDC. These materials offer the possibility of improving the electrical efficiency and stability of microstructural and physical concerns. They exhibit a notable divergence from pure SDC, particularly when considering the influence of reduced temperature [9]. This investigation focused on the integration of GDC composite electrolytes into SSC and SDC materials. The phase stability features and material preparation process considerably influenced the chemical and microstructural stability of the SSC cathode, as stated by a previous author [10].

The study on the dual composite cathode of SSC/GDC–SDC suggests a novel strategy for improving the performance of SOFCs. This cathode design combines SDC, GDC and SSC to form a triple-conductive material [7]. The dual-phase composite cathode has demonstrated outstanding performance, indicating its potential for high-efficiency SOFC operation.

The current study investigates a novel dual composite cathode material that consists of SSC, GDC and SDC. It differs from earlier research in three major ways. Firstly, it investigates a novel material composition by employing a ternary combination of elements that have not yet been researched together for this application. Secondly, this study examines how the GDC-to-SDC ratio in the composite material affects its attributes and performance. Finally, this study focuses on optimising cathode materials for IT-SOFCs, offering tremendous potential for increasing total system efficiency. By integrating these distinct features, our study helps the development of improved cathode materials tailored specifically for next-generation IT-SOFCs.

Firstly, the material has a unique composition that combines SSC (which improves electronic conductivity and reduces oxygen), GDC and SDC (which is known for its strong ionic conductivity). This synergy is predicted to increase overall cathode conductivity. Secondly, this study investigates the possibility of improved electrochemical performance in IT-SOFCs due to optimal material design. Thirdly, this study examines the chemical compatibility of the cathode and electrolyte components, which is critical for maintaining phase stability and reducing undesired by-products. Finally, by analysing the microstructure and properties of the composite at different ratios, researchers obtain crucial information on how to optimise a cathode's physical attributes for achieving optimal IT-SOFC performance.

In summary, this research endeavours to enhance comprehension regarding the effect of varying GDC-SDC ratios on the efficacy of the SSC/GDC–SDC composite cathode in IT-SOFCs. The results of this study will not only contribute to the advancement of our understanding in the area of SOFC materials but will also hold practical significance for the development and enhancement of high-performance SOFCs. The development of dual composite cathodes, such as SSC/GDC–SDC, represents a cutting-edge approach in SOFC research, offering enhanced conductivity, stability, and performance. By exploring novel material combinations and structures, researchers are paving the way for more efficient and reliable SOFC technologies.

## 2. MATERIALS AND METHODS

This section explains the procedure for the preparation of dual composite cathode powders. The powder was prepared via the wet ball milling method by using a Fritsch Planetary Monomill (Pulveristte, Germany) high-energy ball milling machine (HEBM). Composite powder for GDC–SDC was initially prepared. Composite powder of GDC-SDC was mixed with SSC powder to form the dual composite powder known as SSC/GDC–SDC.

The dual composite powder of SSC/GDC–SDC was developed using SSC, GDC and SDC commercial powders (Kceracell Co., Ltd., Korea). The powder underwent ball milling with ethanol as the solvent for 2 hours, followed by oven drying. Then, it was subjected to calcination at a temperature of 750 °C for one hour in air environment, resulting in the formation of the GDC–SDC composite electrolyte. Three distinct weight ratios, namely, 50, 60 and 70 wt.%, of GDC powder (obtained from Kceracell Co., Ltd., Korea) and SDC powder were subjected to ball milling for 2 hours at a rotational speed of 550 rpm.

SSC (Kceracell Co., Ltd., Korea) powder was mixed with GDC–SDC at a ratio of 1:1 to produce SSC/GDC–SDC dual composite powder. Hence, the total composition would be SSC/GDC–SDC (50:50:50), SSC/GDC–SDC (50:60:40) and SSC/GDC–SDC (50:70:30). SSC/GDC–SDC dual composite powders were produced using the HEBM machine and the wet ball milling technique. Zirconia balls with a diameter of 1 mm and 100 mL of ethanol were employed. The zirconia jar was sealed tightly with masking tape before the grinding operation began to ensure that no solution would leak during the high-speed milling procedure. The dual composite powders underwent 2 hours mixing process at 550 rpm and cooled down at room temperature.

Subsequently, the resulting mixture was dried in an oven for 16 hours to obtain the composite cathode powder of SSC–SDC. The phase formation analysis and stability of the SSC–SDC composite cathode powder were observed using a Bruker D8 Advanced X-ray diffractometer. The apparatus produced X-rays with a wavelength of 1.5406 Å by using Cu Kα sources. X-ray diffraction (XRD) data were collected within an angular range of 20° to 80°, as measured in 2θ.

The mixture of SSC/GDC–SDC and SSC was filtered. Both dual composite powders were dried overnight at 100 °C in an oven (Obsnap, Malaysia). The powders were then pulverised with an agate mortar until they could pass through a sieve. SSC/GDC–SDC was heated for 60 minutes at 750 °C in an electric furnace (Protherm, Turkey) to produce a perovskite material. Figure 1 depicts the calcination template for the dual composite SSC/GDC–SDC cathode powders. The choice of calcination temperature and soaking time for SSC/GDC–SDC was provided by [11]. The scanning electron microscope SEM (Hitachi, SU1510, Japan) was used to analyze the cross-sectional morphology of the composites.

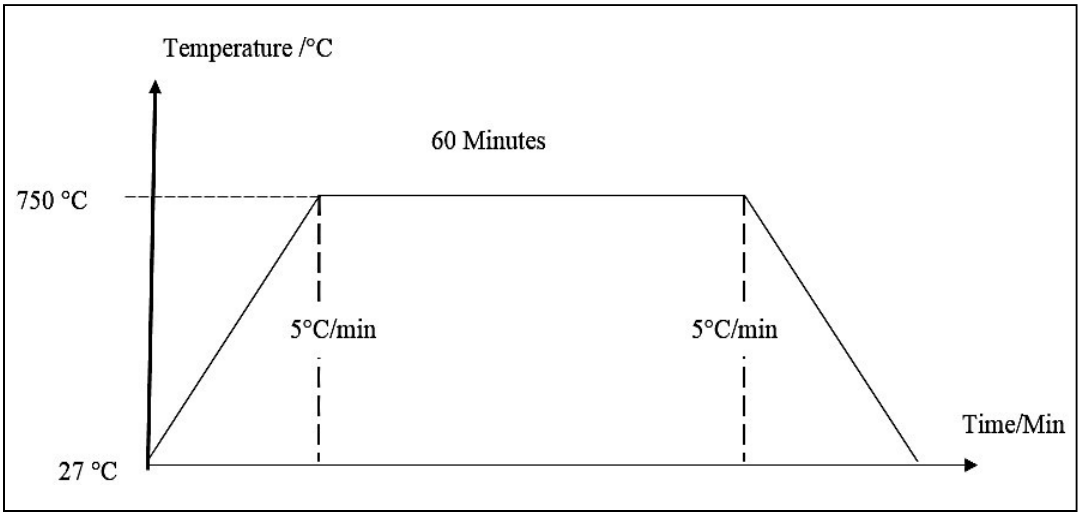

**Figure 1:** Calcination profile for the dual composite cathode SSC/GDC-SDC

## 3. RESULTS AND DISCUSSION

### *3.1 Phase Formation Analysis*

The XRD examination reveals important information regarding the crystal structure, phase composition and purity of the cathode material. The absence of secondary peaks in the XRD spectra suggests that the dual composite powders are synthesised successfully with a high degree of purity. This purity is critical for preserving the integrity and stability of the cathode material, exerting a direct effect on its electrochemical performance in SOFCs. XRD patterns are used as an important tool for confirming the production of appropriate phases and evaluating the quality of a cathode material, eventually influencing the efficiency and reliability of SOFCs.

The fabricated GDC–SDC composite powders were then mixed with the SSC powder to produce dual composite cathode powders. Figure 2 presents the XRD pattern of the SSC/GDC–SDC dual composite powder compared with the SSC, GDC and SDC powders. From the observation of the XRD spectra of the SSC/GDC–SDC calcined powders in Figure 2, excellent purity was indicated when no secondary peak was detected during the analysis. The Joint Committee on Powder Diffraction Standards pattern numbers for SSC, GDC and SDC were 00-053-0112, 00-043-1014 and 01-75-0157 respectively. SSC has a perovskite crystal structure, which is typical in SOFC components. The perovskite structure of SSC adds to its electrochemical characteristics, making it an ideal material for SOFC cathodes. The crystal structure phase of SDC in SOFCs is characterised by a cubic fluorite-type structure, which is critical for its high oxygen ion conductivity as an electrolyte. Substantial research on SDC crystal structure emphasises its importance in the development of high-performance SOFCs. Furthermore, GDC has a fluorite-type crystal structure, which is required for its strong oxygen ion conductivity, making it an ideal material for SOFC electrolytes.

The percentage of purity can be computed using the composition of the desired materials and any impurities present. The purity percentage is normally calculated by dividing the weight of the target component (in this case, the dual composite powders) by the overall weight of the sample, including any impurities. Therefore, the purity of the resulting dual composite powders for all three compositions, i.e. SSC/GDC–SDC (50:50:50), SSC/GDC–SDC (50:60:40) and SSC/GDC–SDC (50:70:30), was 85.74%, 86.91% and 90.17%, respectively. Addressing whether purity levels below 90% are acceptable for laboratory-scale production is essential. Although reaching purity levels above 90% for laboratory-scale powder manufacturing is desirable, evaluating the application's specific requirements is critical. In some circumstances, purity levels slightly lower than 90% may be acceptable, depending on material qualities and manufacturing tolerances.

Factors to consider include the existence of acceptable contaminants, the planned application of the synthesised powders and the feasibility of reaching higher purity levels within the given limits. However, achieving perfect purity (100%) is difficult in laboratory-scale powder production, as experienced by other researchers [12].

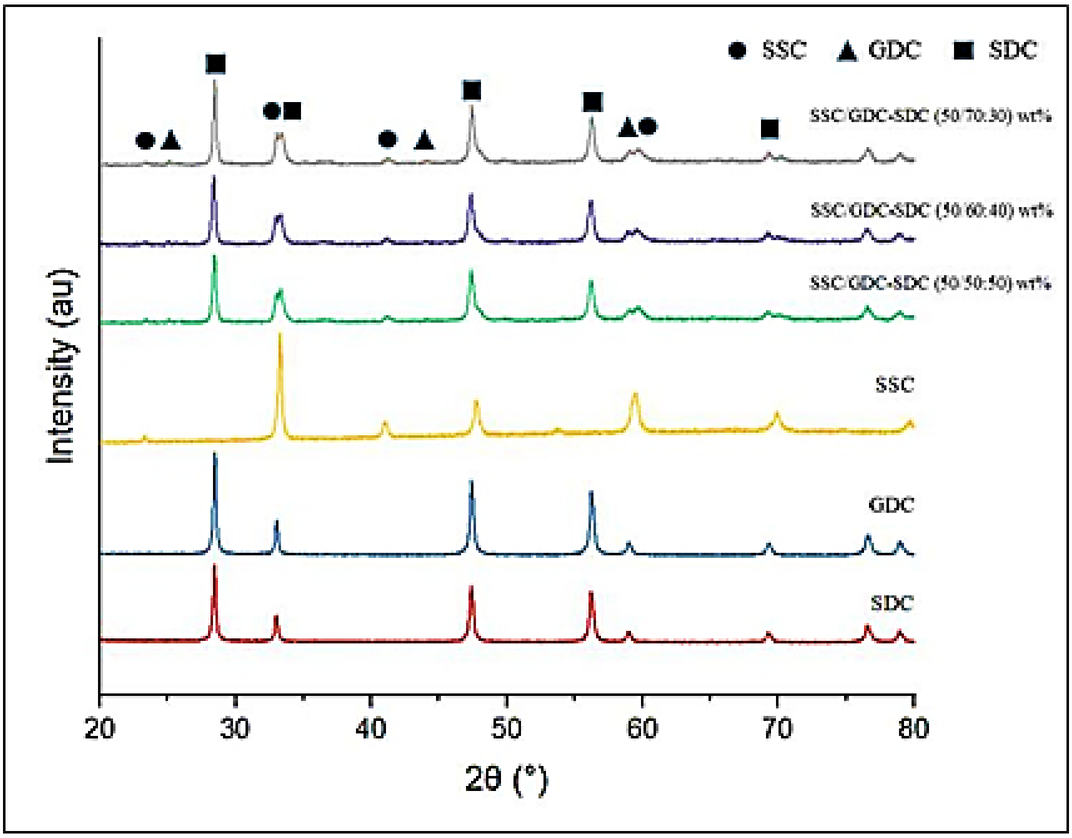

**Figure 2:** XRD pattern of the calcined SSC/GDC–SDC (50:50:50), SSC/GDC–SDC (50:60:40) and SSC/GDC–SDC (50:70:30) composite powders

Overall, the XRD results showed that the calcined SSC/GDC–SDC dual composite powders were highly pure, with no secondary phases observed, efficiently maintaining the crystallite structure across all compositions. Such purity is critical for retaining the integrity and stability of the cathode material, which eventually influences its electrochemical performance in SOFCs.

### *3.2 Microstructure Analysis*

In this section, the characteristics of the SSC/GDC–SDC composite cathode powder in particle form were systematically investigated from the perspective of their porous and dense structures. Figure 3 depicts all the associated images. As described in this section, the maximum porosity percentage of all SSC/GDC–SDC composite cathodes was maintained within an acceptable range.

The SEM micrographs of SSC/GDC–SDC (50:50:50), SSC/GDC–SDC (50:60:40) and SSC/GDC–SDC (50:70:30) are displayed in Figure 3. For SSC/GDC–SDC (50:50:50), the grains are less porous and denser. Scanning electron microscopy (SEM) images exhibit a composite microstructure with a porous surface structure where it is depicted by darker images. Porosity can be seen more significantly on the surface of the pellets on the cross section of the cathode composite pellets. This statement was discussed in [13], which reported that larger particle sizes had lower surface areas and provided a weak driving force for densification.

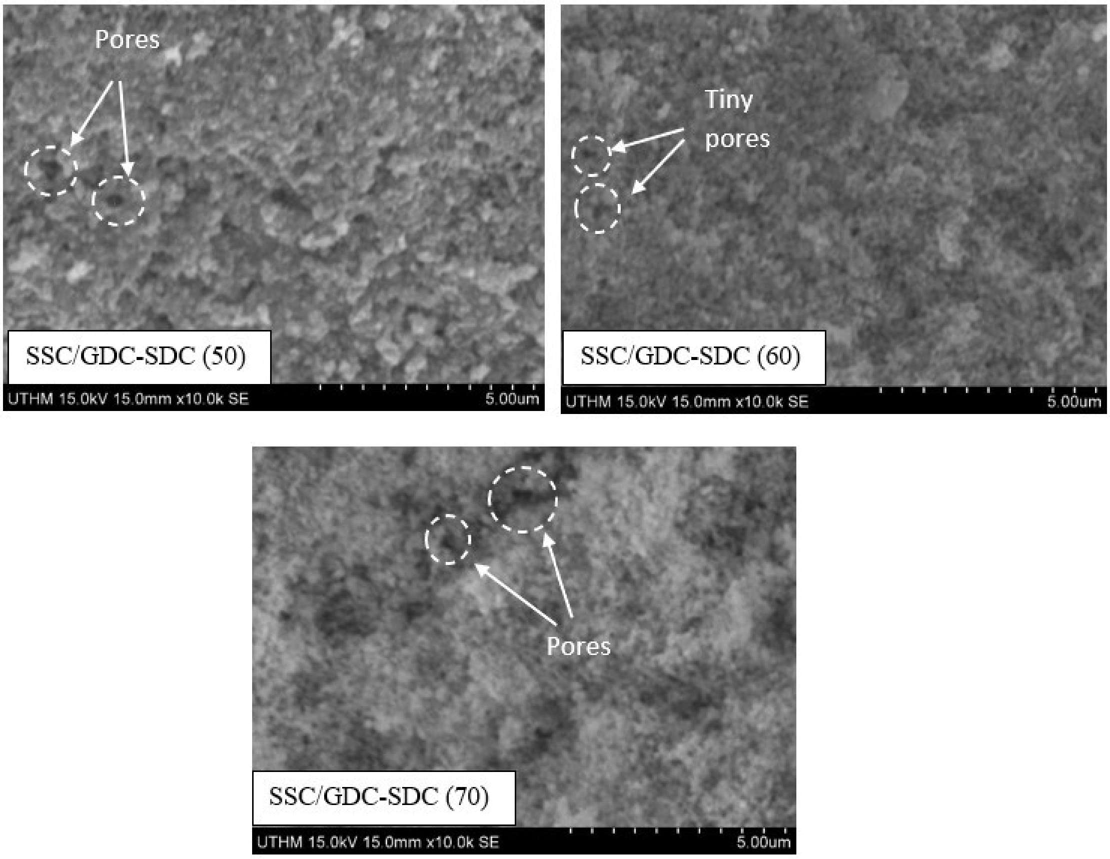

**Figure 3:** SEM image of the cross-sectioned area of the SSC/GDC–SDC (50:50:50), SSC/GDC–SDC (50:60:40) and SSC/GDC–SDC (50:70:30) pellets with compositions of 50, 60 and 70, respectively

The SEM micrographs revealed that composite cathodes with varied GDC–SDC ratios had different morphologies, with certain compositions exhibiting porous surface structures whilst others were denser. Understanding and refining the structure and porosity of cathode materials are critical for improving SOFC performance and stability. The morphology and porosity study results provide essential insights into the structural features of dual composite cathodes, guiding the design and development of high-performance cathode materials for IT-SOFC applications.

### *3.3 Density and Porosity of Sample*

Measuring the density and porosity of different compositions, such as SSC/GDC–SDC cathodes with varying ratios, enables a comparison of their attributes. Understanding how composition changes affect density and porosity is useful in developing material formulations for specific purposes. Table 1 presents the result of the porosity and density obtained from the SSC/GDC–SDC composite cathode.

**Table 1:** Average values of the porosity and density of the calcined SSC/GDC–SDC (50:50:50), SSC/GDC–SDC (50:60:40) and SSC/GDC–SDC (50:70:30) composite cathode pellets

| Calcined composite cathode | Porosity | Density |
|---|---|---|
| SSC/GDC-SDC (50:50:50) | 45.23 ± 0.21 | 3.03 ± 0.29 |
| SSC/GDC-SDC (50:60:40) | 52.25 ± 0.11 | 2.90 ± 0.71 |
| SSC/GDC-SDC (50:70:30) | 52.31 ± 0.17 | 2.91 ± 0.84 |

For the SSC/GDC–SDC 50 (50/50:50 wt.%) cathode, porosity was 45.23% and density was 3.03 g/cm$^3$. For the SSC/GDC–SDC (50:60:40) cathode, porosity was 52.25% and density was 2.90 g/cm$^3$. For the SSC/GDC–SDC (50:70:30) cathode, porosity was 52.31% and density was 2.91 g/cm$^3$. Figure 4 shows average values of the porosity and density of the SSC–SDC composite cathode pellets. The porosity was lowest at the SSC/GDC–SDC (50:50:50) composite. Meanwhile, density was maximal at the composition of SSC/GDC–SDC (50:60:40).

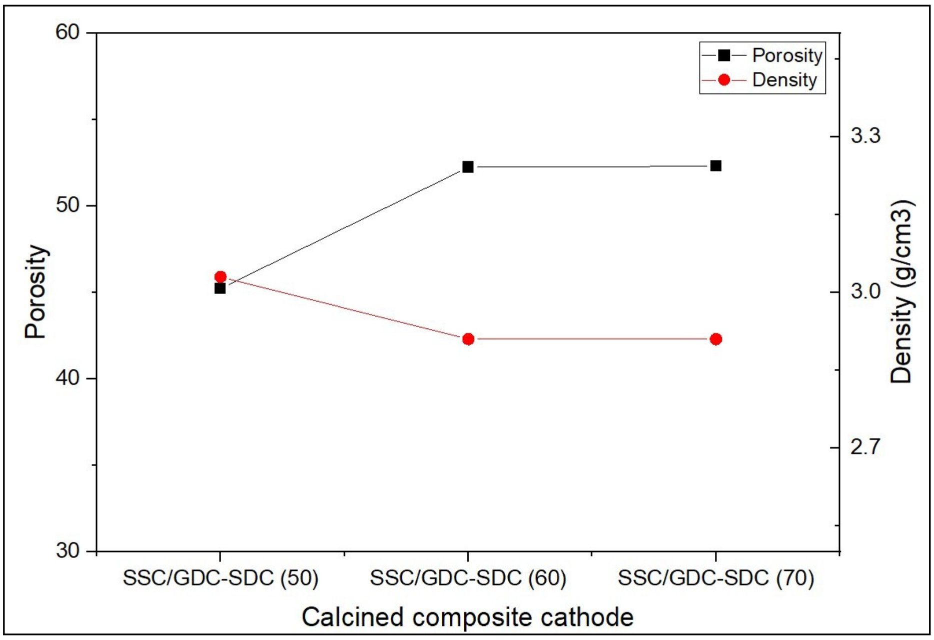

**Figure 4:** Average values of the porosity and density of the SSC–SDC composite cathode pellets

According to numerous researchers, porosity value should be around 30% to achieve excellent ionic conductivity [13]. In addition, according to [14], a cathode's porosity should range from 30% to 50%. Therefore, all the calcined SSC/GDC–SDC dual composite cathodes must have appropriate porosity (31%–44%) to be employed as prospective cathode materials. The selection range for the calcination temperature and sintering temperature (600 °C) was suitable and remained relevant by this finding.

The ideal porosity range for a cathode in SOFCs is typically between 30% and 50%. This range is essential for various reasons. Firstly, a sufficient level of porosity within this range allows for the effective passage of reactant gases, such as oxygen, to the reaction sites at the cathode–electrolyte interface. This condition is critical for sustaining the electrochemical processes inside SOFCs. Furthermore, a porosity range of 30% to 50% enables the effective removal of products, such as water vapour, from the cathode, limiting flooding and preserving a cell's efficiency. Furthermore, the linked pores in this porosity range provide a wide interfacial surface for electrochemical processes to occur, resulting in improved cell performance.

When the porosity of a cathode goes below 30%, the performance of SOFCs may be compromised. Insufficient porosity can impede gas diffusion and restrict access to reaction sites, resulting in increased mass transport losses and poor cell performance. When porosity exceeds 50%, a cathode's electronic and ionic conductivities decline, whilst gas permeability increases. This condition can result in reduced triple phase boundary density, leading to fewer electrochemically active sites and greater concentration polarization, affecting the overall performance of SOFCs.

## 4. CONCLUSIONS

The XRD spectra analysis of the calcined SSC/GDC–SDC powders demonstrated excellent purity, with no secondary peaks detected. The purity of the resulting dual composite powders with three different compositions (50SSC/50GDC–50SDC, 50SSC/60GDC–40SDC and 50SSC/70GDC–30SDC) was 85.74%, 86.91% and 90.17%, respectively. Addressing whether purity levels below 90% are acceptable for laboratory-scale production is essential.

Although reaching purity levels above 90% for laboratory-scale powder manufacturing is desirable, evaluating an application's specific requirements is critical. In some circumstances, purity levels slightly lower than 90% may be acceptable, depending on material qualities and manufacturing tolerances. In conclusion, the developed SSC/GDC–SDC dual composite cathodes exhibit a lack of secondary phases post-processing, maintaining their crystallite structure effectively across all compositions.

` However, more comprehensive electrochemical impedance spectroscopy testing of calcination temperatures and secondary phase must be further explored because calcination temperature can exert effects on the conductivity and performance of dual composite cathodes.

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## Author Contributions

All authors contributed toward data analysis, drafting and critically revising the paper and agree to be accountable for all aspects of the work.

## Disclosure of Conflict of Interest

The authors have no disclosures to declare.

## Compliance with Ethical Standards

The work is compliant with ethical standards.

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