INFLUENCE OF CuO, Ag AND Zn ADDITIVE MATERIALS ON THE PROPERTIES OF BARIUM STRONTIUM COBALT FERRITE CATHODE FOR SOLID OXIDE FUEL CELL APPLICATION: A SHORT REVIEW

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Abstract. Alternative energy sources are necessary to meet the world's energy needs. One of the most capable technologies for energy conversion in the future is solid oxide fuel cells (SOFCs). The development of SOFCs has attracted great attention worldwide, particularly for intermediate low-temperature applications. The commercialisation of SOFCs can be accomplished after satisfying a few system requirements and discovering suitable material candidates. Hence, this review was constructed to study the current approach of material development for intermediate-temperature SOFCs and the influence of additive materials on the properties of barium strontium cobalt ferrite (BSCF)-based cathodes. This article discussed three types of additive materials, i.e. copper oxide (CuO), silver (Ag) and zinc (Zn), focusing on their chemical, physical, thermal and electrochemical characteristics. The additive material was substituted in the B-site of the BSCF cathode to improve cell performance. In addition, the additive materials have remarkably improved the cathode performance and chemical stability. The addition of CuO was intended to increase chemical bonding and enhance the cathode's electrochemical reaction activity. On the one hand, the addition of Ag offers improved oxygen surface adsorption and dissociation of molecular oxygen into atomic oxygen. On the other hand, the addition of Zn can provide excellent thermal stability and good oxidation resistance and enhance the electrocatalytic activity of the oxygen reduction process. Therefore, this review focuses on identifying the influence and suitable additive material for BSCF perovskites regarding their chemical and physical properties for outstanding SOFC performance.

Keywords: BSCF, cathode, copper, SOFC, silver

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Introduction

The development of alternative power sources is necessary to reduce global energy consumption in a wide range of ways. Thus, an electrochemical system is a reliable fuel cell that can efficiently and cleanly transform chemical energy into electrical energy with less pollution [1]. The solid oxide fuel cell (SOFC) is one type of fuel cell that is dependable and adequate for modern problems. Moreover, it is a potentially clean and efficient power-generating device. The fact that SOFCs normally operate at a high temperature of approximately 1000 °C presents a considerable operating temperature problem for SOFC performance. The total efficiency of the system might be adversely affected by the operation of SOFCs at temperatures between 700 °C and 900 °C [2].

However, the essential operating temperature makes commercialisation difficult for SOFCs because it limits the usage of substances and increases the rate of the process. This concern can be avoided by decreasing the operating temperature to a moderate level at 600 °C–800 °C. Thus, SOFCs are attractive candidates for upcoming technological inventions for converting energy, and considerable work has already been expended on decreasing their working temperature for wider usage. This reduction can provide various benefits to the operation, including reduced material degradation, increased material structure stability, longer service life and reduced processing costs. However, lowering the temperature would compromise the cathode’s ability to catalyse the oxygen reduction reaction (ORR) process, which would in turn decrease the intermediate SOFC’s electrochemical performance. Therefore, searching for suitable cathode materials for SOFCs operating at intermediate temperatures is necessary. Furthermore, high catalytic activity and enhanced chemical stability should be taken as serious concerns when researching high-potential cathode materials [3].

Therefore, several investigations were performed to generate improved cathode material compounds and optimise their microstructure to increase the catalytic characteristics for the ORR process in intermediate-temperature SOFCs (IT-SOFCs). Amongst the various cathode substances investigated, perovskite oxide materials with mixed oxygen ionic and electronic conductivity (MIEC) have been given much attention [4]. The researcher suggested barium strontium cobalt ferrite (BSCF) as a potential cathode composition option for increased performance at intermediate temperatures, and this perovskite has high ion conductivity, higher catalytic performance in oxygen reduction and a cost-effective material profile. In addition, under fuel cell working circumstances, BSCF is well known for its good stability and compatibility with electrolyte materials, such as yttria-stabilised zirconia, gadolinium-doped ceria, lanthanum strontium gallium magnesium oxide (LSGM), samarium doped ceria (SDC) and many others [5]. However, BSCF exhibits chemical instability, which lowers the electrochemical efficiency and reduces the generation of oxygen ions [6]. The BSCF declines with time because of the transition from the cubic to the hexagonal phase, which impairs cell performance. Moreover, BCSF exhibits poor compatibility at the interfaces between the electrolyte and cathode due to its high thermal expansion coefficient (TEC) [6].

Therefore, this review examines and discusses recent studies on the effects of additive materials on cathode cell performance using BSCF-based cathodes that have transition metal elements added. ON the basis of this review, potential cathode materials for IT-SOFCs can be created and found. Additive materials, such as silver (Ag), copper oxide (CuO) and zinc (Zn), are highlighted in this review and are currently receiving much attention. They have been explored and was found to enhance oxygen flux substantially [7]. CuO is added to strengthen chemical bonds and improve the cathode’s ability to conduct electrochemical reactions. On the one hand, the inclusion of Ag provides improved molecular and atomic oxygen dissociation and surface adsorption. In addition, the inclusion of Zn can improve the electrocatalytic activity of the oxygen reduction process whilst delivering great thermal stability and good oxidation resistance. This study presents the findings and perspectives on the effects of these three added materials on the phase stability, microstructure and electrochemical parameters of the BSCF composite cathode acquired by previous research.

Phase Stability of BSCF Composite Cathode

Nanoscale materials may be useful in cathodes because of their substantial specific surface area. The number of ORR reactive sites restricted to the triple-phase boundary (TPB) is remarkably increased by using a two-dimensional (2D) nanostructure as the cathode material [8]. In addition, the utilisation of nanostructured materials generally increases access to reactive sites, thus improving the performance of cells [8].

Influence of Ag Addition

BSCF-SDC composite cathode powder was mixed by milling commercial BSCF and SDC powder through a wet milling process. BSCF-SDC and BSCF-SDC-Ag composite cathode powders underwent XRD examination, and the results are shown in Figure 1. Data showed that the intensity pattern of the basic material is produced at a common spectrum, and the material has an observable phase structure at JCPDS no: 00-005-0563 (cubic crystal) for BSCF, i.e. JCPDS no: 01-075-0157 (faced-centre cubic) for SDC and JCPDS no: 01-004-0783 (faced-centre cubic) for Ag [9].

However, impurities are observed on the XRD diffraction peaks. The secondary peaks of barium carbonate ($BaCO_3$) (JCPDS number: 01-071-2394) and iron carbonate ($Fe(CO_3)$) (JCPDS number: 01-083-1764) are shown in the graph. Based on Figure 1, secondary peaks are expected to arise when they move closer to the BSCF and SDC peaks. This rise is a result of the reaction between alkaline oxide and CO_2 during calcination process [9]. Based on past research, several possibilities remain for this secondary phase or impurity ions to exist in the BSCF system after the calcination process. Moreover, most of these impurities were effectively incorporated into the BSCF lattice. Additionally, this impurity production may occur if the composite powder was mixed via the high-energy ball milling method, which involves mixing at a high speed that causes the particle to shatter due to impact forces generated during the conversion of kinetic energy. Whilst SDC remains stable in the original crystallite phase, the BSCF material has a 28.83% crystallite percentage due to the presence of an impurity.

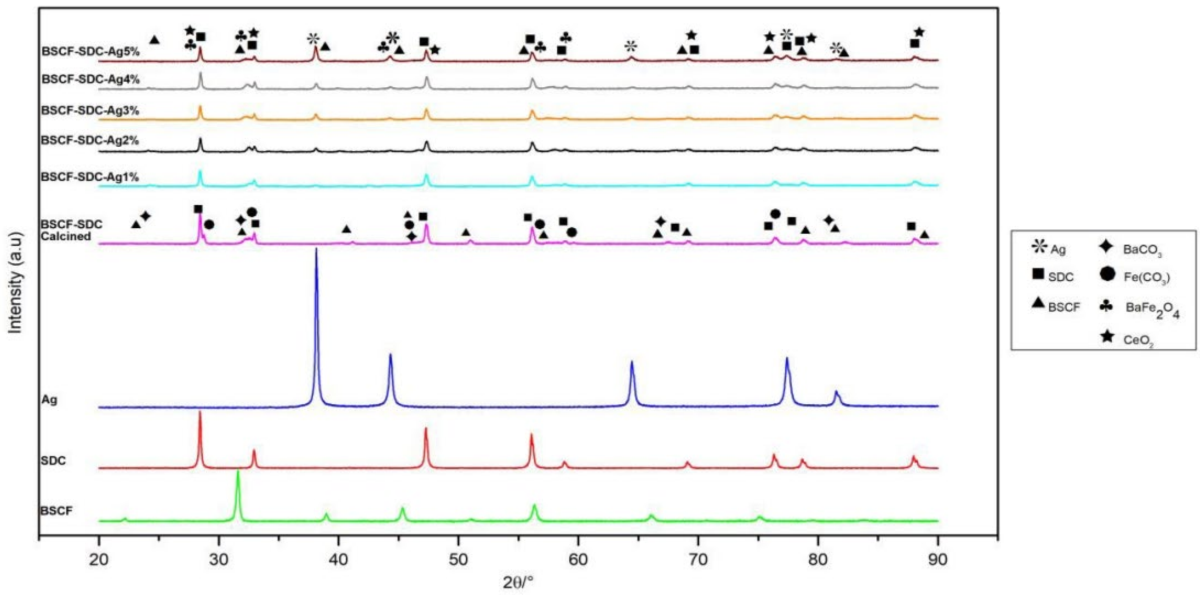

Figure 1: XRD pattern for the composite cathode powders of BSCF-SDC and BSCF-SDC-Ag [10]

Influence of CuO Addition

BSCF/SDC–CuO was prepared by compounding BSCF with SDC and 3 SDC–CuO at a mass ratio of 7:3. The XRD patterns of the BSCF/SDC and BSCF/SDC-CuO powders calcined at 900 °C, 950 °C and 1000 °C are shown in Figure 2 [11]. The XRD peaks of BSCF/SDC and BSCF/SDC-CuO were mostly caused by the BSCF and SDC phases. The impurity phases produced by the reaction of the BSCF phase and SDC phase correspond to the weak peaks depicted by the dotted oval in Figure 2. The two samples of BSCF/SDC and BSCF/SDC-CuO had similar XRD positions and diffraction peak numbers after calcination at different temperatures. This result demonstrated that the BSCF/SDC-CuO samples with CuO addition did not result in the formation of any new impurity phases, suggesting that the BSCF/SDC-CuO sample's chemical stability was improved by adding CuO as an additive material [12].

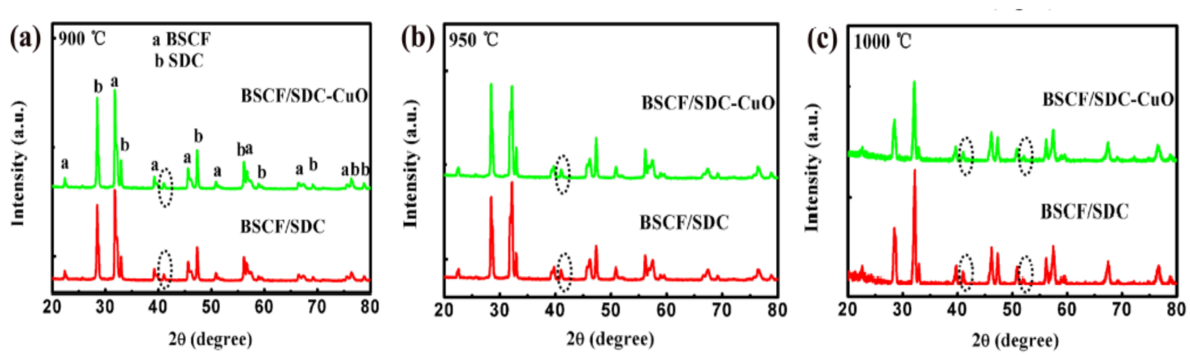

Figure 2: BSCF/SDC and BSCF/SDC-CuO powder XRD patterns after calcination at (a) 900 °C, (b) 950 °C and (c) 1000 °C [11].

Influence of Zn Addition

Zn-based BSCF (BZCY4) was synthesised by a combined EDTA-citrate complexing sol-gel process. Analysis of the Zn-additive element's influence on the BSCF-based cathode includes several tests. The outcome was recognised using XRD to define the chemical characteristics. According to Zhang et al. [13], all the elements involved exist at their peaks in the XRD pattern of the Zn-additive material, and no secondary phase peaks are observable. Furthermore, Zn has shown positive outcomes when combined with cobalt in the perovskite's B-site crystal structure [14]. Meanwhile, according to Park et al., lattice expansion caused the lattice parameters to increase and the peak location to shift slightly to the left [15]. Figure 3 shows the Zn-modified and untreated BZCY4 samples' room-temperature XRD patterns before and after the CO_2 treatment [16]. This figure shows that surface carbonate was most likely created when atmospheric CO_2 reacted with Zn-based BSCF (BZCY4) [17]. In addition, the carbonate phase peak intensity was lower in Zn-modified BZCY4 than in unmodified BZCY4. When compared with unmodified BZCY4, Zn modification reduced the peak intensity of the carbonate phase. This reduction indicates that the ZnO sintering assistance may have also increased the chemical stability of BZCY4 [18].

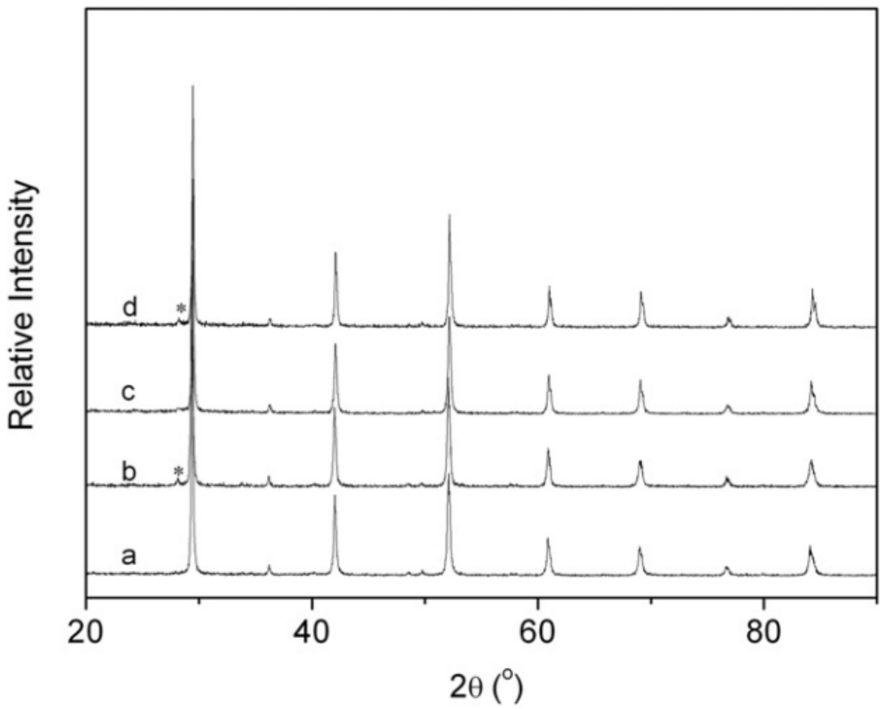

Figure 3: XRD peaks for samples before and after CO_2 treatment: (a) BZCY4 powder; (b) BZCY4 in CO_2; (c) Zn-modified BZCY4 powder; and (d) Zn modified in CO_2 [18]

Microstructure of BSCF Composite Cathode

The MIEC BSCF has a low polarisation resistance and excellent electrocatalytic activity for the ORR [19]. In addition, a fine microstructure of a material produces a large specific surface area, which raises the quantity of active sites at the cathode and electrolyte interface [5].

Influence of Ag Addition

Figure 4 shows the FESEM micrographs at 30 Kx magnification with a scale of 100 nm, and the particle shape may be seen clearly at this magnification [10]. In addition, the crystallinity percentage for the BSCF (40.65%) and SDC (59.93%) elements changing based on the growth of impurity is triggered by their element after 1%–5% of Ag was added to the BSCF-SDC composite cathode powder [9]. This circumstance is caused by the impurity ions that may have continued to exist as distinct phases after the sample was calcined at 950 °C. Moreover, the size of the material's crystallites affects the appearance of CeO_2. The crystallite size of the material also plays a role in CeO_2 appearance. Much extra heat is produced when the size is too large, which promotes the formation of crystallites and causes agglomeration [20]. The red circle indicates that particle agglomeration resulted from the increased amount of Ag. The removal of any leftover CeO_2 during the calcination process is most likely what caused agglomeration, which in turn generated strong bonding within each element.

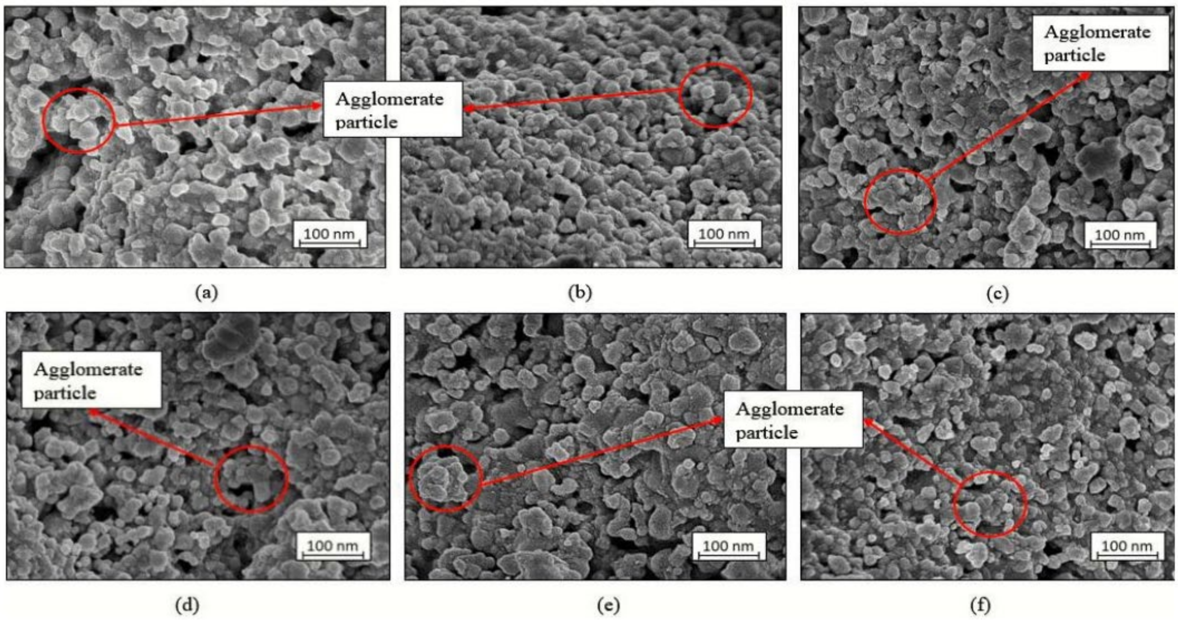

Figure 4: FESEM morphology of particles (a) BS-Ag0, (b) BS-Ag1, (c) BS-Ag2, (d) BS-Ag3, (e) BS-Ag4 and (f) BS-Ag5 [10]

Influence of CuO Addition

The SEM images of the BSCF, BSCF/SDC and BSCF/SDC-CuO cathodes are shown in Figure 5. All three sintering temperatures displayed a porous structure, which is necessary for the transportation of oxygen and the subsequent oxygen reduction reaction [21]. The BSCF particles in BSCF/SDC and BSCF/SDC-CuO were larger than the SDC and SDC-CuO particles and formed the cathode skeleton at a certain sintering temperature. Additionally, the BSCF skeleton was connected to the SDC and SDC-CuO particles. The SOFC application made good use of the layer combinations. More specific surface areas than BSCF were present in BSCF/SDC and BSCF/SDC-CuO, which is considerably helpful for boosting the cathode's electrochemical reaction activity and the quantity of oxygen reduction active sites [22].

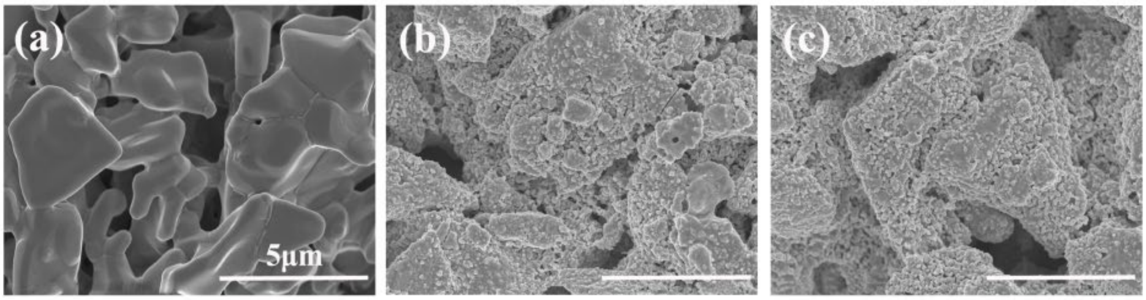

Figure 5: SEM images of BSCF, BSCF/SDC and BSCF/SDC-CuO cathodes calcined at (a) 900 °C, (b) 950 °C and (c) 1000 °C [23]

Influence of Zn Addition

Figure 6 shows the SEM image of the prepared quad-layer NiO D BZCY/BZCYZ/BCY/BSCF cell in cross-section. Although some enclosed pores remained in the BZCYZn electrolyte, especially close to the cathode substrate, their quantity was far lower than that in the BZCY4 layer of cell-3 [23]. Even though the BZCY4 (or BZCYZn) layers were made from the same amount of powder (0.02 g), BZCYZn layer thickness was approximately 27 mm as opposed to BZCY4 electrolyte layer thickness of 35 mm in the absence of the ZnO sintering promoter [24]. Hence, the addition of Zn substantially aided the sintering of the BZCY4 electrolyte. The sintering densities for the BCY and BZCYZn layers in cell-4 were discovered to be approximately 98% and 93%, respectively. Therefore, BZCYZn can be used as a fuel cell electrolyte because of its high sintering density [25].

In addition, ZnO worked well as a sintering promoter for various proton-conducting oxides, including BZY. For BZCY4 membranes, ZnO has previously been used as a sintering aid. The BZCY4 electrolyte sintering was discovered to be considerably affected by the ZnO modification process. Agglomeration is one structural change that can occur in the continuous oxidised state of Zn. Therefore, the sintering side effect can be minimised by the presence of Zn in Co-containing perovskites. The porous shape of the cathode enhances the cell performance through easier oxygen transport and more active ORR sites for SOFC applications. The carbonate formative rate for BZCYZn was only approximately half that of BZCY4, as shown in Table 1. This finding further indicates the suppressive effect of the ZnO sintering aid on surface carbonate production in comparison with BZCY4, which is consistent with the findings in the literature. The ZnO sintering aid probably accumulated on the BZCY4 surface during calcination and sintering, which not only helped the membrane sinter but also shielded it from CO_2 exposure.

Table 1: BET and carbonate formation rates of BZCY4 and BZCYZn powders [25]

Samples	**BET (m^2g^{-1})**	**Carbonate Formative Rate ($10^{-7}molm^{-2}min^{-1}$)**
BZCY4	13.3	5.27
BZCYZn	17.1	2.66

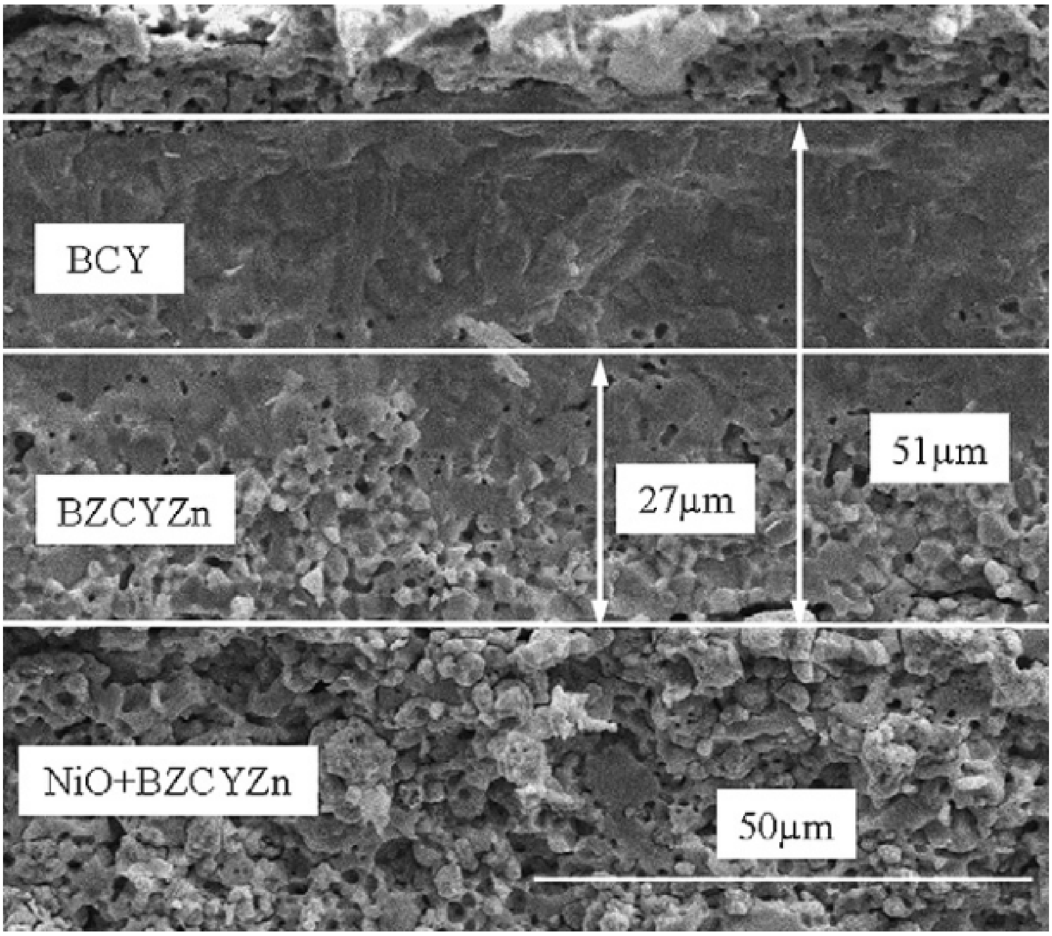

Figure 6: SEM images of Cross-sectional view of the constructed quad-layer NiO D BZCY/BZCYZ/BCY/BSCF cell [25]

Electrochemical Properties of the BSCF Composite Cathode

BSCF is known for being stable and compatible with electrolyte materials under fuel cell working conditions. However, the chemical instability of BSCF reduces the electrochemical performance, which conversely reduces the generation of oxygen ions. Moreover, its poor compatibility with the electrolyte and cathode interfaces is further reflected by BCSF's high TEC [17]. Additive materials, such as Ag, CuO and Zn, are highlighted in this review. These materials are constantly being explored and have excellent benefits for improving oxygen flow and cell performance.

Influence of Ag Addition

Electrochemical impedance spectroscopy (EIS) was used on a symmetric cell with a configuration of electrode|SDC|electrode to assess the ORR activity of the Ag-BSCF cathodes. Figure 7 displays the impedance spectra for the untreated BSCF and Ag-BSCF cathodes that were subjected to various microwave plasma treatment times. The electrode polarisation resistance is the difference between the real axis intercepts of the impedance plot (Rp). The Ag-BSCF cathode without treatment only slightly improves the ORR activity when compared with the pristine BSCF because Ag depletes the oxygen–silver oxide TPB, which is essential for ORR [24]. The transfer of Ag film into Ag particles that follow the development of TPB occurs during microwave plasma treatment [26]. The improvement in the ORR activity of the Ag-BSCF cathodes is indicated by the impedance arcs contracting as the treatment duration is increased [27].

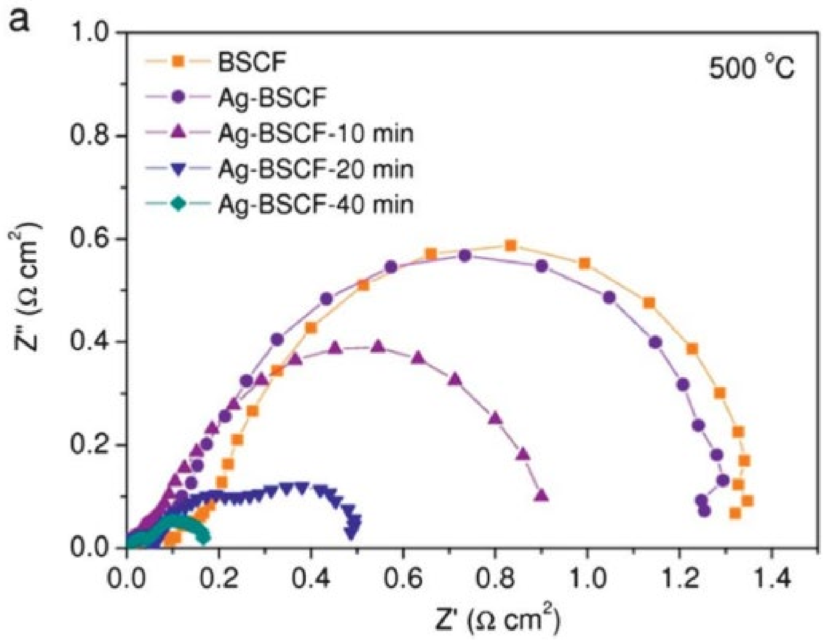

Figure 7: EIS diagram of BSCF, Ag-BSCF and Ag-BSCF treated for 10–40 minutes in microwave plasma [27].

Influence of CuO Addition

The EIS of the three cathode-based SOFCs is depicted in Figure 8. Regardless of the sintering temperature, the BSCF/SDC-CuO-based SOFC consistently demonstrated the lowest EPR amongst the three cathodes. Given that the same anode was used, any alterations in the cathode caused any variations in the EPR of the SOFCs. The BSCF/SDC-CuO cathode showed the best electrochemical performance amongst the three cathodes, regardless of the sintering temperature. The Rp values for the SOFCs based on the three cathodes are shown in Table 1, and they were calculated using equivalent circuit fitting of the EIS. The SOFC made from BSCF/SDC-CuO sintered at 950 °C has the lowest Rp value of all the SOFCs, which is 0.041 cm^2. The EIS of the three cathodes and SOFCs constructed utilising the three cathodes determined 950 °C to be the ideal sintering temperature for BSCF/SDC-CuO [28].

Table 2: For the SOFCs created by BSCF, BSCF/SDC and BSCF/SDC-CuO sintered at 900 °C, 950 °C and 1000 °C, respectively, Rp (cm^2) values were determined [29].

Sintering temperature	BSCF	BSCF/SDC	BSCF/SDC-CuO
900	0.124	0.051	0.048
950	0.103	0.073	0.041
1000	0.166	0.099	0.050

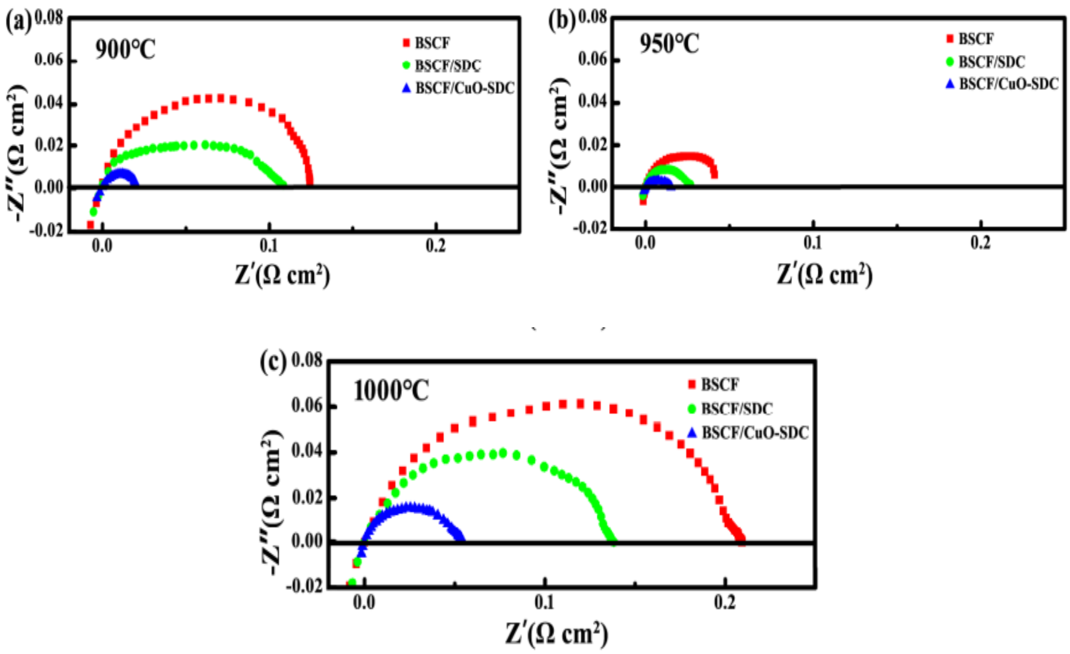

Figure 8: Three-electrode method at 800 °C with open-circuit voltage was used to determine the EIS values for the BSCF, BSCF/SDC and BSCF/SDC-CuO cathodes sintered at (a) 900 °C, (b) 950 °C and (c) 1000 °C [29].

Influence of Zn Addition

Figure 9 shows the EIS for cells 4 and 3 in Nyquist plots under OCV conditions. All the EIS measurements show a high-frequency induction tail and a depressed arc [30]. The intercepts in these spectra at low and high frequencies represent the total cell resistance and the electrolyte resistance, respectively. The variations in the intercepts at low and high frequencies are determined by the electrode polarisation resistances, which are the sum of the resistances at the cathode–electrolyte and anode–electrolyte interfaces, respectively.

Given the BZCY4 layer's higher sintering density in cell-4, the overall electrolyte resistance shown in Figure 9(a) was lower than that in Figure 9(b). The performance of the cells was affected by the Zn sintering assistance in two different ways. One of these was an improvement in sintering density, which benefited the performance of the cells. Nevertheless, doping with Zn might result in a drop in the internal conductivity of BZCY4, which affects cell performance. At higher temperatures, the positive effect of electrochemical performance was more remarkable. Nevertheless, the dual-layer electrolyte cell showed potential throughout the whole temperature range that was studied.

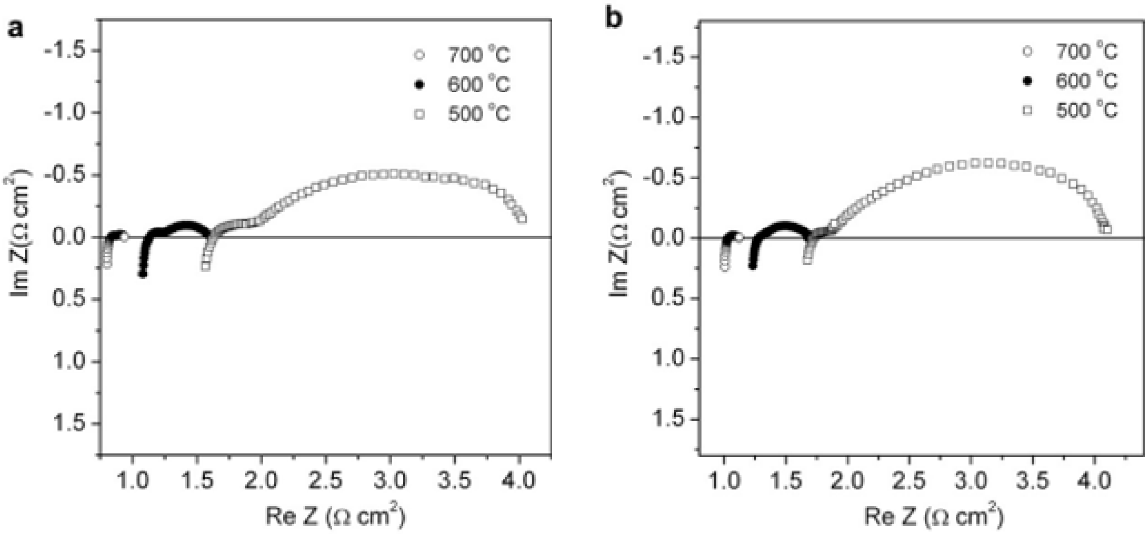

Figure 9: EIS results for (a) NiO D BZCY4/BZCYZn/BCY/BSCF and (b) NiO D BZCY4/BZCY4/BCY/BSCF cells at various temperatures under OCV conditions [30].

Conclusions

This study reviews recent results on substituting Ag, CuO and Zn as additive materials for BSCF-based cathodes. The B-site of BSCF-based cathode material for IT-SOFCs and the effect of transition element replacement were discussed. Both outcomes improved the cathode cell's physical and chemical properties based on the results of the doping of Ag, CuO and Zn with a BSCF-based cathode. The addition of CuO also balances the chemical stability along with increasing the ORR catalytic activity. The Copper Oxide (CuO) is the best additive material to improve the chemical and physical properties and achieve outstanding SOFC performance.

In addition, this study's findings show that future studies can investigate many other topics, such as how much additive material performed from the transition metal element group can act as a catalyst on BSCF-based cathode materials. A clear improvement in cell performance features is demonstrated by the theory on substitution elements at the B-site of BSCF-based cathodes. However, the contribution of transition metal element doping to BSCF-based cathodes or other perovskite materials needs further research.

The outcomes of this work can be used to identify the ideal IT-SOFC parameters and improve material qualities, including electrocatalytic activity and chemical and physical properties. Moreover, the improvement of the cathode material for IT-SOFCs can help overcome the SOFC's high-temperature limitation and advance the commercialisation of SOFCs. The findings of this review may be helpful and instructive in fulfilling these goals.

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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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