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Effect of strontium doping on structural and electrical conductivity of $BaTi_{0.8}Zr_{0.2}O_{3-\delta}$ cathode materials for solid oxide fuel cell

Abul Kalam Azad*[1], Lukman Ahmed Omeiza[1]

[1]*Faculty of Integrated Technologies, Universiti Brunei Darussalam, Jalan Tungku Link, Gadong BE1410, Brunei Darussalam*

** Correspondence: lukahmed378@gmail.com*

Abstract. Cathode materials are crucial for the efficient operation of solid oxide fuel cell (SOFC), serving as a key component for the oxygen reduction reaction (ORR). Cobalt-containing cathode have shown good performance in terms of electronic conductivity and ORR kinetics. However, they suffer from significant drawbacks, including high cost, and instability at high operating temperatures of SOFC. Thus, there is an increasing need to investigate cobalt-free alternatives that can overcome these challenges. In this study, cobalt-free $Ba_{1-x}Sr_xTi_{0.8}Zr_{0.2}O_{3-\delta}$ (BSTZ0, BSTZ25, and BSTZ50; x = 0, 0.25, 0.5) cathode materials were synthesized via traditional solid-state reaction techniques. The synthesized samples were characterized using X-ray diffraction (XRD), scanning electron microscopy (SEM), to analyse their structural properties, and surface morphology. Electrical conductivity of the synthesised materials measured in air revealed promising results, with BSTZ0, BSTZ25, and BSTZ50 exhibiting activation energy (E_a) of 0.25 eV, 0.45 eV, and 0.48 eV, respectively. X-ray diffraction analysis confirmed that all compositions crystallize in the tetragonal P4/mmm space group. Findings from this investigation indicate that the synthesized Sr-doped $Ba_{1-x}Sr_xTi_{0.8}Zr_{0.2}O_{3-\delta}$ materials exhibit favourable electrical conductivity, and are promising candidates for SOFC cathode.

Keywords: electrical conductivity; SOFC; cathode materials; cobalt-free; ORR

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[1] *the corresponding author

1. Introduction

The development of solid oxide fuel cell (SOFC) represents a significant advancement in energy conversion technologies. This is primarily due to their high efficiency and flexibility in using various fuels. SOFC is a clean and sustainable means of energy generation when hydrogen is used as the fuel source [1]. Thus, addressing the environmental concerns associated with fossil fuel resources. Among the critical components of SOFC, the cathode plays a vital role in determining the overall cell performance. Cathode material must exhibit high electrical conductivity, chemical stability, and favourable reaction kinetics for the oxygen reduction reaction (ORR) [2–4]. Recent studies focused on enhancing the properties of cathode materials through various doping strategies. The incorporation of strontium (Sr) into barium titanate ($BaTiO_3$) has gained a lot of attention. Strontium doping has been shown to improve the dielectric properties and electrical conductivity of these materials, making them more suitable for high-temperature applications [5-9]. Barium titanate ($BaTiO_3$) is a well-known perovskite oxide that exhibits excellent ferroelectric and dielectric properties, which can be further optimized through the substitution of Sr ions. The introduction of Sr into the $BaTiO_3$ lattice modifies the structural characteristics, electrical conductivity, and thermal stability [10,11]. Several studies have revealed that Sr-doped barium titanate (BST) ceramics can achieve a high dielectric constant with a low dissipation factor [11–14]. Furthermore, the incorporation of Zr into $BaTiO_3$ matrix, as in $BaTi_{0.8}Zr_{0.2}O_{3-\delta}$, has been reported to enhance the ionic conductivity, which is crucial for the efficient operation of SOFC [15,16]. Figure 1 illustrates a typical SOFC, and its various components.

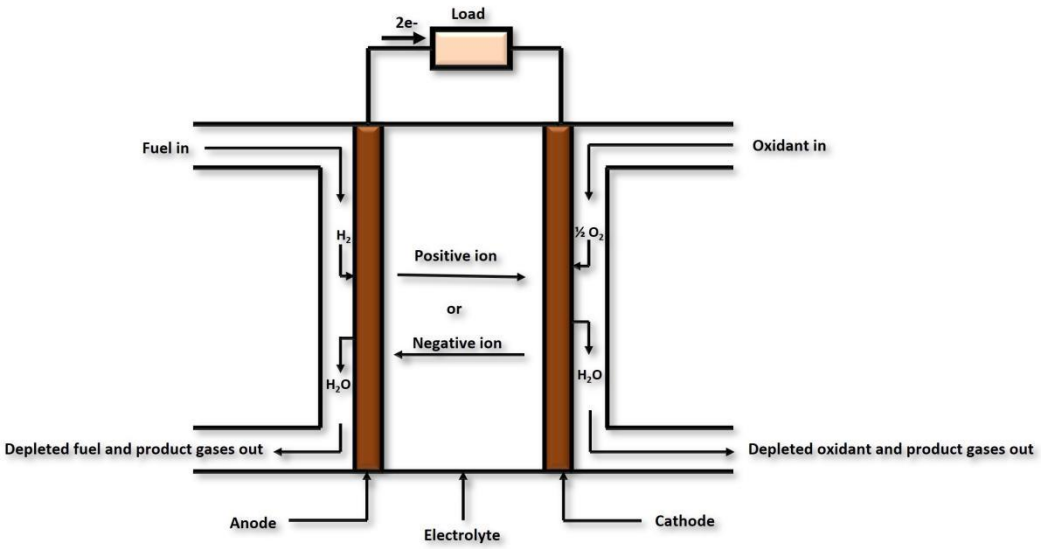

Figure 1. Typical SOFC and its various components [17].

In this study, structural and electrical properties of cobalt-free $Ba_{1-x}SrxTi_{0.8}Zr_{0.2}O_{3-\delta}$ (BSTZ0, BSTZ25, and BSTZ50; x = 0, 0.25, 0.5) were investigated. These materials can help overcome

the high costs associated with cobalt-containing cathode materials in SOFC [18,19]. Optimizing its properties through controlled doping can lead to improved performance metrics, such as enhanced electrical conductivity and thermal stability [20,21]. Additionally, the ratio of doping and the resultant microstructural characteristics can significantly affect the electrochemical performance of the cathode, necessitating a thorough investigation [22–24]. Cobalt-containing cathode materials have been reported to have poor linear thermal elongation, thus resulting in compatibility issues with commonly used electrolytes (yttria-stabilized zirconia and gadolinium-doped ceria). Cobalt-free BSTZ0, BSTZ25, and BSTZ50 were synthesized and investigated as potential cathode materials for SOFC in the present study. The influence of Sr doping on $BaTi_{0.8}Zr_{0.2}O_{3-\delta}$ series was studied. Key parameters, such as electrical conductivity and activation energy, were analyzed and compared with existing cobalt-free cathode materials.

2. Experimental

Cobalt-free $Ba_{1-x}Sr_xTi_{0.8}Zr_{0.2}O_{3-\delta}$ (BSTZ0, BSTZ25, and BSTZ50; x = 0, 0.25, 0.5) was synthesised via solid state reaction techniques, and analysed as potential cathode materials for SOFC. Stoichiometric amounts of the precursors of $SrCO_3$, $BaCO_3$, ZrO_2, and Ti_2O_3 were weighed and thoroughly mixed using an agate mortar and pestle for 2 hours under ethanol as a grinding medium to ensure homogeneity. The resulting mixture was dried and calcined at 1000 ℃ for 10 hours in air, followed by intermediate grinding. This was carried out to ensure complete reaction and phase formation. The calcined powders were pressed into pellets using a uniaxial press under a pressure of 5 MPa, with polyvinyl alcohol (PVA) as a binder. BSTZ0, BSTZ25, and BSTZ50 ellets were sintered at 1200 ℃ for 10 hours in air, followed by natural cooling to room temperature.

The phase purity and crystal structure of the synthesized BSTZ0, BSTZ25, and BSTZ50 samples were analysed using X-ray diffraction (Bruker D8 Advance) at a 2θ range of 20 to 80° with a step size of 0.02°. Chekcell was used to confirm the phase composition, and lattice cell parameters [25,26]. Microstructure of the sintered samples was examined using a scanning electron microscope (SEM). Energy-dispersive X-ray spectroscopy (EDS) was performed to verify the elemental distribution within the samples. The electrical conductivity of BSTZ0, BSTZ25, and BSTZ50 samples was measured using a DC four-probe technique. Platinum paste was coated on both sides of the sintered pellets to act as current collectors, followed by heating at 900 ℃ for 1 hour to ensure good contact. The electrical conductivity measurements were conducted in air over a temperature range of 400 to 1000 ℃.

3. Results and discussion

3.1. Structural investigations of $Ba_{1-x}Sr_xTi_{0.8}Zr_{0.2}O_{3-\delta}$ (BSTZ0, BSTZ25, and BSTZ50; x = 0, 0.25, 0.5)

Figure 2 shows the XRD analysis of BSTZ0, BSTZ25, and BSTZ50 powders at room temperature. XRD pattern of the synthesized cathode material confirms its crystalline nature

and phase purity. Diffraction peaks were indexed via chekcell to a tetragonal structure, with the P4/mm-123 space group. Obtained peak positions confirm with reference pattern PDF-01-089-821 (P4/mmm-123). The sharp and intense peaks observed in the diffraction pattern indicate high crystallinity in the material. This property is desirable for solid oxide SOFC applications, as it enhances ionic and electronic conductivity. Table 1 gives the lattice cell parameters for all compositions. The absence of additional peaks indicates the successful doping of Sr into the crystal lattice of $BaTi_{0.8}Zr_{0.2}O_{3-\delta}$, maintaining the structural integrity of the parent lattice [27].

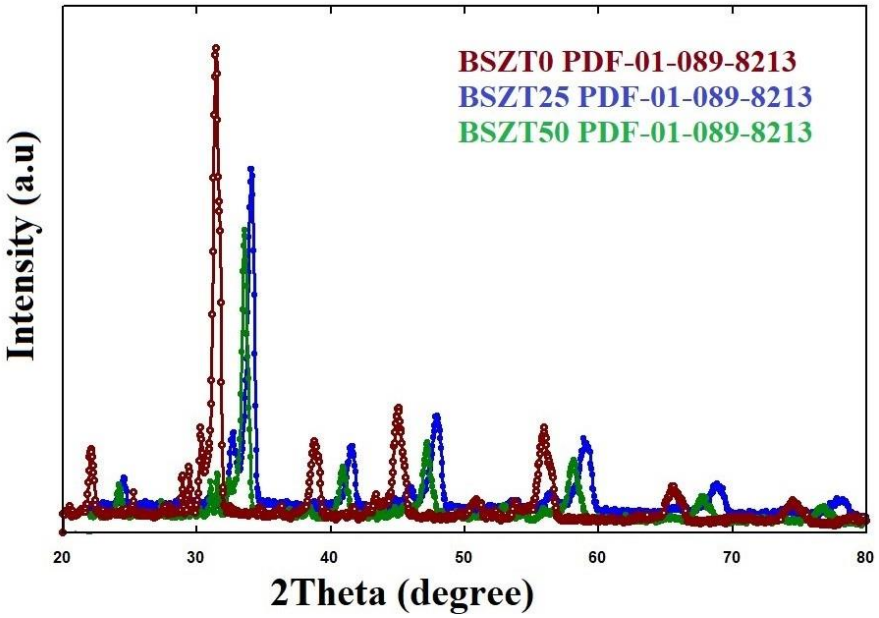

Figure 2. XRD diffraction pattern of BSTZ0, BSTZ25, and BSTZ50.

Table 1. Lattice cell parameters for all compositions

Composition	Space group	Cell parameters	Cell Volume
BSZT0	Tetragonal-P4/mmm (123)	a=4.00186 Å, c=3.99347 Å	63.96 Å3
BSZT25	Tetragonal-P4/mmm (123)	a=4.00678 Å, c=3.99347 Å	64.39 Å3
BSZT50	Tetragonal-P4/mmm (123)	a=3.94470 Å, c=3.96985 Å	61.77 Å3

3.2. SEM-EDS Analysis

Surface morphology and elemental composition of BSTZ0, BSTZ25, and BSTZ50 cathode materials were examined using scanning electron microscopy (SEM) coupled with energy-dispersive X-ray spectroscopy (EDS). SEM micrographs of all compositions revealed a porous microstructure with

uniformly distributed particles. Figure 3 shows the SEM analysis of the synthesised samples. BSTZ0 exhibited slightly larger grain sizes compared to BSTZ25, and BSTZ50, suggesting that the incorporation of Sr influenced the sintering behaviour. All the samples showed good porosity, ensuring efficient charge transport [28,29]. EDS spectra confirmed the presence of all the expected elements (Ba, Sr, Ti, Zr, and O) in each composition. The SEM-EDS analysis confirms that BSTZ0, BSTZ25, and BSTZ50 cathode materials exhibit the desired morphological and compositional characteristics, making them suitable candidates for SOFC applications. O-KA, Ba-LA, and Sr-L in the EDS profile refer to specific characteristic X-ray emissions from the elements present in the sample. Figure 4 illustrates the EDS investigation of the synthesised cathode materials.

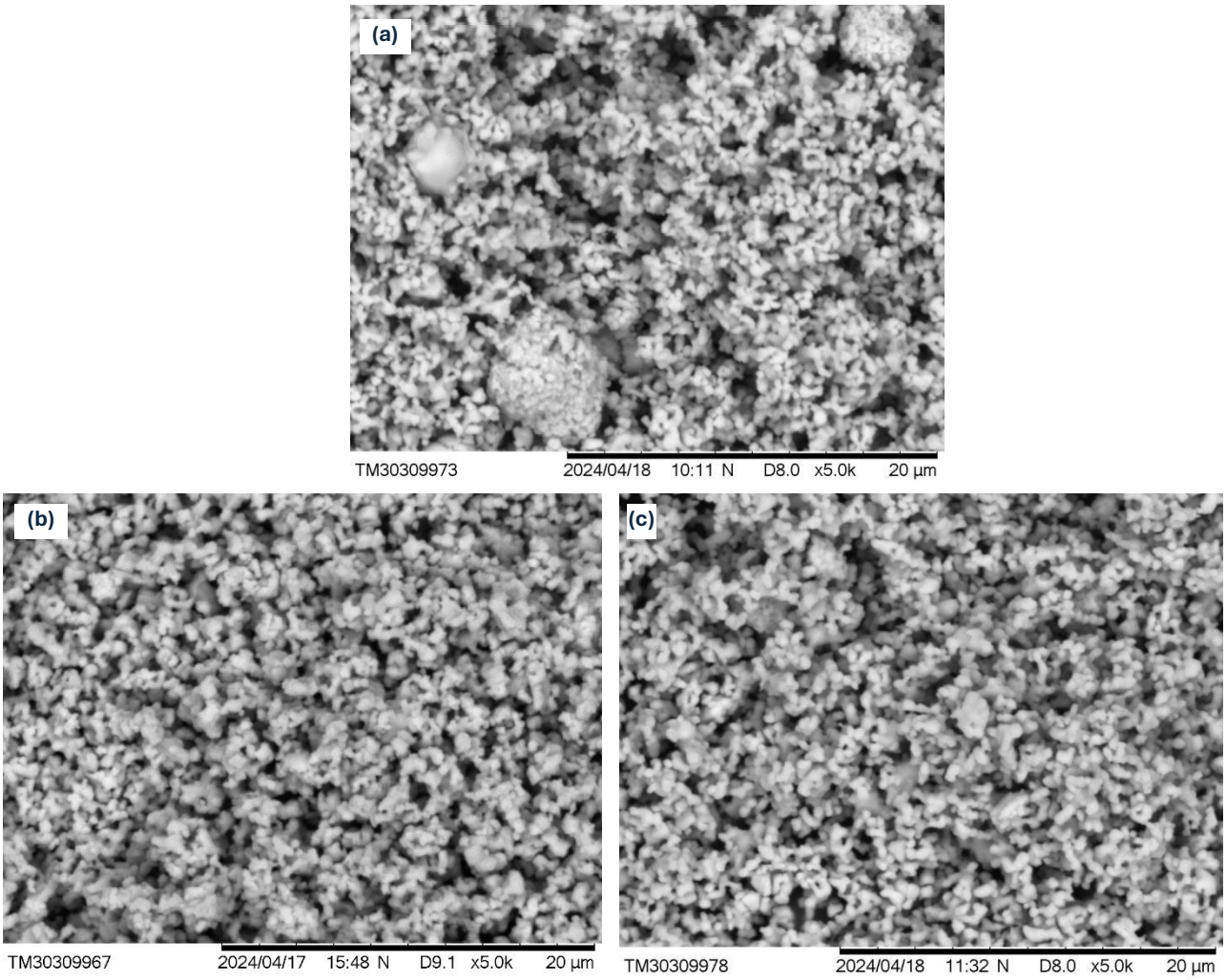

Figure 3. SEM analysis for: (a) BSTZ0, (b) BSTZ25, and (c) BSTZ50.

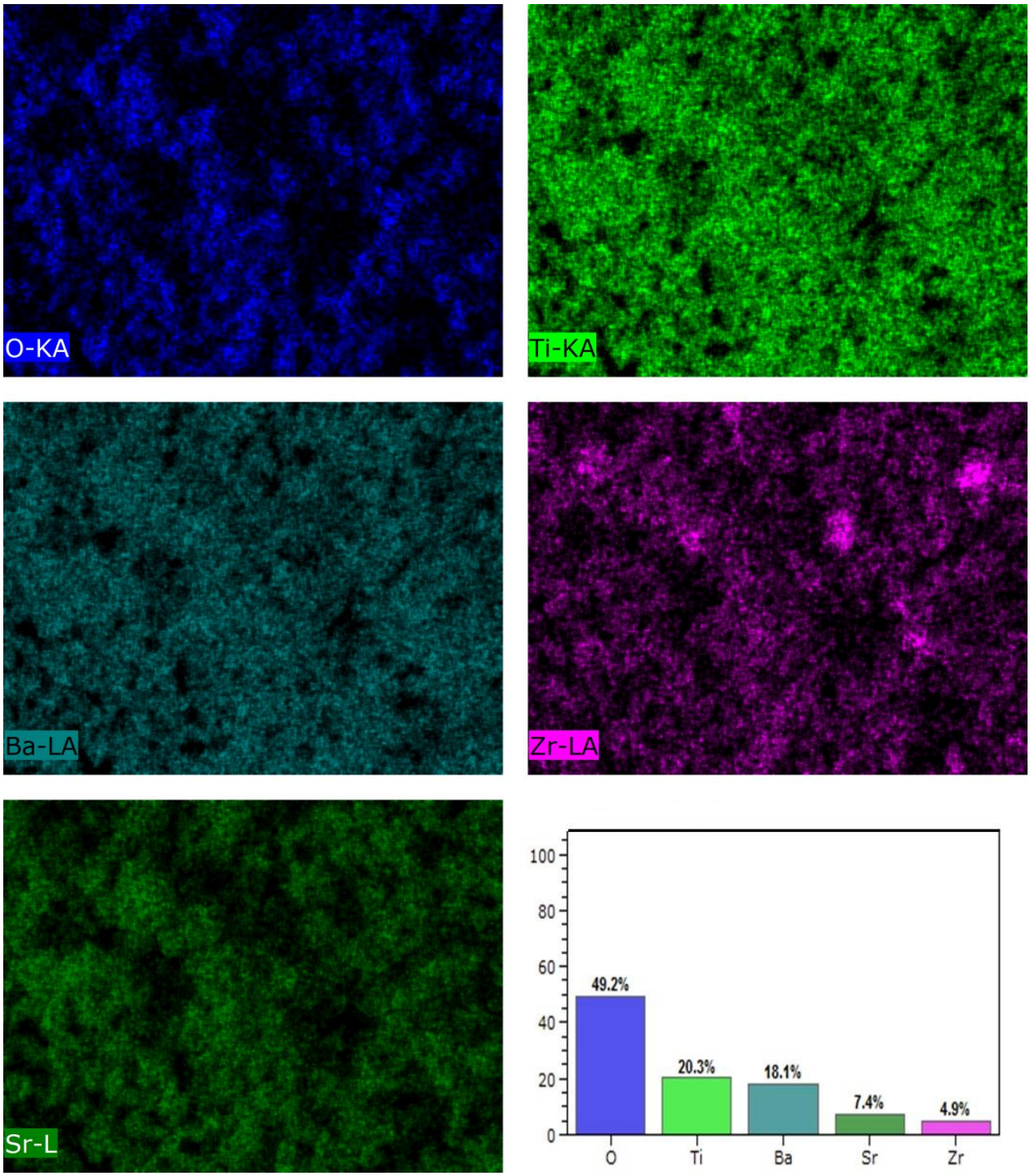

Figure 4. EDS investigation showing elemental distribution of BSZT25.

Electrical Conductivity

The electrical conductivity of BSZT0, BSZT25, and BSZT50 was investigated in air, at a temperature of 600 to 1000 ℃. Temperature dependence of the conductivity can be calculated in accordance with Equation 1 [18,28]:

The symbol "σ" represent the electrical conductivity in Equation 1, σ_0 is the pre-exponential factor, E_a is the activation energy, K_B is the Boltzmann constant, and T is the absolute temperature. Arrhenius plots (Figure 5) for BSZT0, BSZT25, and BSZT50 revealed distinct activation energy values of 0.25 eV, 0.45 eV, and 0.48 eV, respectively. These values indicate the influence of strontium doping on the electrical conduction mechanism [31-33]. BSZT0 exhibited the lowest activation energy (E_a*=0.2eV*), suggesting superior electronic conductivity compared to the doped samples. This could be attributed to the higher intrinsic mobility of charge carriers in the undoped composition. For BSZT25 *(E_a=0.45eV),* and BSZT50 *(E_a=0.48 eV)* the increased activation energies imply a stronger dependence of conductivity on temperature. This behaviour is likely associated with the enhanced ionic conduction pathways and defect chemistry introduced by strontium doping [34–36]. Doped samples exhibited a balance between ionic and electronic conduction, which is desirable for SOFC cathode materials. At high temperatures (> 700 °C), all compositions demonstrated sufficient electrical conductivity to meet the requirements for SOFC cathodes (σ > 0.1 S/cm). However, BSZT0 showed the highest conductivity over the entire temperature range, while BSZT50 exhibited the lowest, reflecting the trade-off between electronic and ionic conduction mechanisms. Findings from this study suggest that strontium doping modifies the conduction pathways and energy barriers for charge transport. Thus, the optimal composition may depend on the operating temperature and specific SOFC design requirements. Further studies on long-term stability and compatibility with electrolytes are needed to fully assess their applicability. Table 2 gives an overview of the E_a obtained in this study with those reported in related literature on cobalt-free cathode materials development for SOFC.

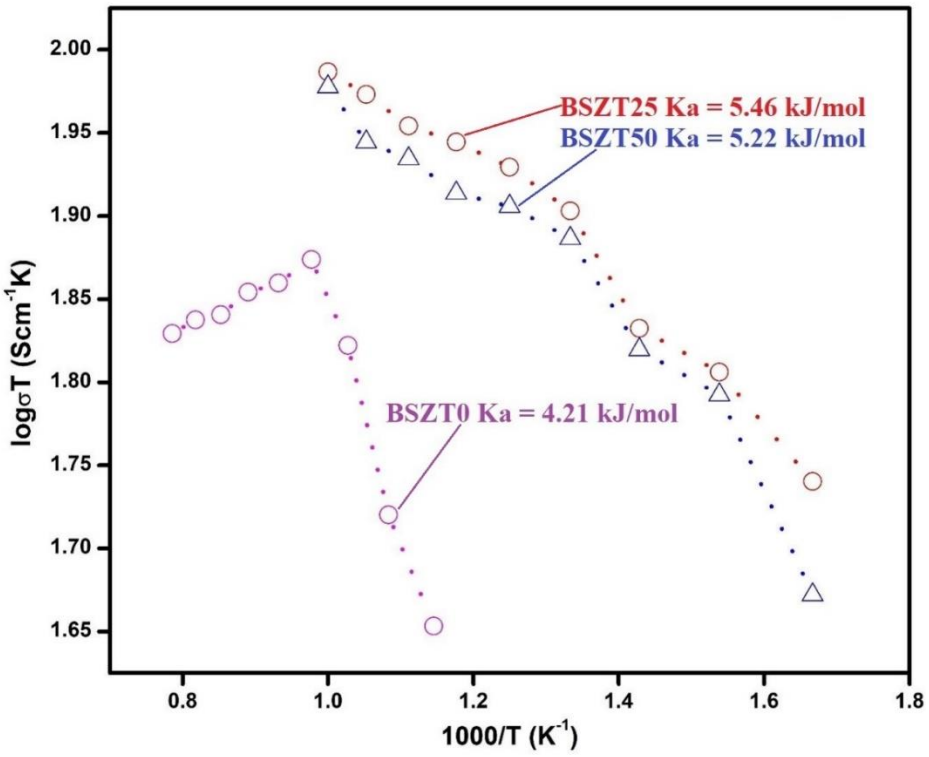

Figure 5. Arrhenius plots of electrical conductivity of BSZT0, BSZT25, and BSZT50

Table 2. Comparison of the activation energy obtained in this study with related literature.

Composition	Temperature (°C)	E_a (eV)	Ref.
BSZT0	600 – 1000	0.25	This study
BSZT25	600 – 1000	0.45	This study
BSZT50	600 – 1000	0.48	This study
$(Nd_{0.9}La_{0.1})_2(Ni_{0.74}Cu_{0.21}Al_{0.05})O_{4+\delta}$	100 – 400	0.12	[37]
$Nd_{0.5}Sr0.5Mn_{0.5}Cr_{0.5}O_{3-\delta}$	400 – 600	0.21	[20]

4. Conclusion

Effect of strontium doping on the structural and electrical properties of $BaTi_{0.8}Zr_{0.2}O_{3-\delta}$ cobalt-free cathode materials was investigated in this study. Strontium substitution into the $BaTi_{0.8}Zr_{0.2}O_{3-\delta}$ lattice was confirmed through structural analysis, revealing slight lattice distortions without compromising phase stability. These modifications played a crucial role in enhancing the materials' electrical properties. Eelectrical conductivity of undoped BSZT0 and strontium-doped BSZT25 and BSZT50 was investigated in air, with activation energies of 0.25 eV, 0.45 eV, and 0.48 eV, respectively. While BSZT0 exhibited the highest conductivity due to superior electronic conduction, the doped compositions demonstrated a favourable balance between ionic and electronic conduction, crucial for SOFC performance. All samples surpassed the conductivity threshold required for SOFC cathodes at high operating temperatures. Results of this study highlight Sr-doped $BaTi_{0.8}Zr_{0.2}O_{3-\delta}$ as potential cobalt-free cathode materials for SOFC applications. Thus, offering an environmentally and economically viable alternative. Future studies are needed to understand the long-term stability and electrochemical performance in practical SOFC configurations.

Authors' contributions

Abul Kalam Azad – original draft writing, investigation

Lukman Ahmed Omeiza – conceptualization, reviewing, investigation

Conflict of interest statement

The authors have no conflict of interest to declare, as all sources used were duly cited and acknowledged.

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Абул Калам Азад[1]*, Лукман Ахмед Омейза[1]

[1]Біріктірілген технологиялар факультеті, Брунея-Даруссалама университеті, Джалан Тунгку Линк, Гадонг BE1410, Бруней-Даруссалам;

** Хат алмасуға арналған автор: lukahmed378@gmail.com*

Қатты оксидті отын элементтеріне арналған $BaTi_{0.8}Zr_{0.2}O_{3-\delta}$ катодты материалдардың құрылымы мен электр өткізгіштігіне стронций қоспасының әсері

Аңдатпа. Катодты материалдар оттегінің тотықсыздану реакциясының (ORR) негізгі компоненті ретінде қызмет ететін қатты оксидті отын ұяшығының (SOFC) тиімді жұмыс

істеуі үшін өте маңызды. Құрамында кобальт бар катод электрондық өткізгіштік пен ORR кинетикасы бойынша жақсы өнімділік көрсетті. Дегенмен, олардың айтарлықтай кемшіліктері бар, соның ішінде жоғары құны және жоғары SOFC жұмыс температурасында тұрақсыздық. Осылайша, осы мәселелерді шеше алатын кобальтсыз баламаларды зерттеу қажеттілігі артып келеді. Бұл жұмыста кобальтсыз катодты материалдар $Ba_{1-x}Sr_xTi_{0.8}Zr_{0.2}O_{3-\delta}$ (BSTZ0, BSTZ25 және BSTZ50; x = 0, 0.25, 0.5) дәстүрлі қатты фазалық реакция әдістерімен синтезделді. Синтезделген үлгілер олардың құрылымдық қасиеттері мен бетінің морфологиясын талдау үшін рентгендік дифракция (XRD) және сканерлеуші электронды микроскопиямен (SEM) сипатталды. Ауада өлшенген синтезделген материалдардың электр өткізгіштігі перспективалы нәтижелер көрсетті: BSTZ0, BSTZ25 және BSTZ50 сәйкесінше 0,25 эВ, 0,45 эВ және 0,48 эВ активтену энергияларын (Ea) көрсетті. Рентгендік дифракциялық талдау барлық композициялардың P4/mmmm тетрагональды кеңістік тобында кристалданатынын растады. Бұл зерттеудің нәтижелері синтезделген Sr-қоспаланған $BaTi_{0.8}Zr_{0.2}O_{3-\delta}$ материалдарының жақсы электрөткізгіштікке ие және SOFC катодына перспективалы үміткерлер екенін көрсетеді.

Түйінді сөздер: электр өткізгіштік; SOFC; катодты материалдар; кобальтсыз; ORR

Абул Калам Азад*[1], Лукман Ахмед Омейза[1]

[1]Факультет интегрированных технологий, Университет Брунея-Даруссалама, Джалан Тунгку Линк, Гадонг BE1410, Бруней-Даруссалам;

** Автор для корреспонденции: lukahmed378@gmail.com*

Влияние легирования стронцием на структуру и электропроводность катодных материалов $BaTi_{0.8}Zr_{0.2}O_{3-\delta}$ для твердооксидных топливных элементов

Аннотация. Катодные материалы имеют решающее значение для эффективной работы твердооксидного топливного элемента (SOFC), служа ключевым компонентом реакции восстановления кислорода (ORR). Кобальтсодержащий катод показал хорошие характеристики с точки зрения электронной проводимости и кинетики ORR. Однако они имеют существенные недостатки, в том числе высокую стоимость и нестабильность при высоких рабочих температурах SOFC. Таким образом, существует растущая потребность в исследовании альтернатив, не содержащих кобальт, которые могут решить эти проблемы. В данной работе безкобальтовые катодные материалы $Ba_{1-x}Sr_xTi_{0,8}Zr_{0,2}O_{3-\delta}$ (BSTZ0, BSTZ25 и BSTZ50; x = 0, 0,25, 0,5) были синтезированы традиционными методами твердофазных реакций. Синтезированные образцы были охарактеризованы методами рентгеновской дифракции (РФА), сканирующей электронной микроскопии (СЭМ) для анализа их структурных свойств и морфологии поверхности. Электропроводность синтезированных материалов, измеренная на воздухе, показала многообещающие результаты: BSTZ0, BSTZ25 и BSTZ50 показали энергию активации (Ea) 0,25 эВ, 0,45 эВ и 0,48 эВ соответственно. Рентгеноструктурный анализ подтвердил, что все составы кристаллизуются в тетрагональной пространственной группе P4/mmm. Результаты этого исследования показывают, что синтезированные материалы $BaTi_{0,8}Zr_{0,2}O_{3-\delta}$, легированные Sr, обладают хорошей электропроводностью и являются перспективными кандидатами для катода SOFC.

Ключевые слова: электропроводность; SOFC; катодные материалы; не содержащий кобальт; ORR

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Information about the authors:

Abul Kalam Azad – corresponding author, PhD – senior lecturer in the Faculty of Integrated Technologies, Universiti Brunei Darussalam, Jalan Tungku Link, Gadong BE1410, Brunei Darussalam

Lukman Ahmed Omeizaa – PhD – researcher of the Faculty of Integrated Technologies, Universiti Brunei Darussalam, Jalan Tungku Link, Gadong BE1410, Brunei Darussalam

Абул Калам Азад **–** автор для корреспонденции, PhD – старший преподаватель факультета интегрированных технологий, Университет Брунея-Даруссалама, Джалан Тунгку Линк, Гадонг BE1410, Бруней-Даруссалам
Лукман Ахмед Омейза **–** PhD – исследователь факультета интегрированных технологий, Университет Брунея-Даруссалама, Джалан Тунгку Линк, Гадонг BE1410, Бруней-Даруссалам

Абул Калам Азад **–** хат-хабар авторы, PhD – Брунея-Даруссалама университетінің біріктірілген технологиялар факультетінің аға оқытушысы, Джалан Тунгку Линк, Гадонг BE1410, Бруней-Даруссалам
Лукман Ахмед Омейза **–** PhD, Брунея-Даруссалама университетінің біріктірілген техноло-гиялар факультетінің ғылыми қызметкері, Джалан Тунгку Линк, Гадонг BE1410, Бруней-Даруссалам