REVIEW ARTICLE



Limitations and trends on cobalt-free cathode materials development for intermediate-temperature solid oxide fuel cell- an updated technical review

Lukman Ahmed Omeiza^{1,2} · Kairat A. Kuterbekov² · Asset Kabyshev² · Kenzhebatyr Bekmyrza² · Marzhan Kubenova² · Shammya Afroze² · Saifullah Abu Bakar¹ · Abul Kalam Azad¹

Received: 15 March 2024 / Accepted: 28 April 2024 © Qatar University and Springer Nature Switzerland AG 2024

Abstract

Intermediate-temperature solid oxide fuel cell (IT-SOFC) work at moderate temperature range (600-800 °C), thereby eliminating the issue of thermal degradation of electrode materials, reduce operational cost, increase flexibility of material selections, and enhance electrochemical stability of cell components. At intermediate-temperature range, there exists sluggish cathodic reaction, high activation energy and slow oxygen reduction reaction (ORR) at the cathode. Several cobaltcontaining cathode perovskite materials with mixed ionic and electronic properties have been developed, which has helped in resolving sluggish ORR and enhances cathodic reaction, thereby increasing the overall performance of IT-SOFC. The expensive nature of cobalt, high evaporation rate and poor thermal expansion coefficient (TEC) means cobalt-free cathode materials need to be investigated. The present study gives an insight into the current trends of cobalt-free cathode materials development in IT-SOFC. Literature reviewed showed composite La_{0.65}Ca_{0.35}FeO₃₋₈-Gd_{0.2}Ce_{0.8}O₂₋₈ (LCF-GDC), and La_{0.7}Sr_{0.3}Cu_{0.15}Fe_{0.85}O_{3-δ} cathode materials has good polarisation resistance of 0.28 Ωcm² at 750 °C, and 0.0153 Ωcm² at 700 °C, respectively. Limitations, challenges, gaps were identified, and possible future research direction was recommended. The study also analysed the use of symmetrical electrodes, as it will help resolve the complexity of developing different electrode materials for cathode and anode in IT-SOFC. Holistic efforts were devoted to ensuring that the literature reviewed was recent (within the last 4yrs), and relevant to the current constraints impeding cathode materials use in IT-SOFC. This review study is meant to serve as a reference material to related researchers, and industry experts looking for the most recent accomplishments in cobalt-free cathode materials development.

Keywords Fuel cell · Cathode materials · Cobalt-free · Oxygen reduction reaction · Activation energy

Lukman Ahmed Omeiza lukahmed378@gmail.com

Asset Kabyshev assetenu@gmail.com

Abul Kalam Azad abul.azad@ubd.edu.bn

- ¹ Faculty of Integrated Technologies, Universiti Brunei Darussalam, Jalan Tungku Link, Gadong, BE1410 Bandar Seri Begawan, Brunei
- ² Faculty of Physics and Technical Sciences, L.N. Gumilyov, Eurasian National University, Astana 010008, Kazakhstan

1 Introduction

Global warming, depletion of the ozone layers and environmental concerns have led to the continued search for a more clean, sustainable, and affordable means of energy generation. Solid oxide fuel cells (SOFCs) are electrochemical devices that convert the chemical energy of fuels directly into electricity [1]. The absence of a combustion chamber in SOFCs means that there are no toxic or harmful emissions of gases, and thus are reliable and clean sources of energy. In comparison to other fuel cell technologies, SOFC has high energy and power densities (around 60%), fuel flexibility, long-term stability, and relatively low cost [2]. SOFCs work at high temperatures (800 - 1000 °C), which causes some challenges such as degradation of electrode materials, high maintenance cost, limited materials selection, reactivity with



cell interconnects and carbon dioxide poisoning [3, 4]. To mitigate some of these constraints, researchers have been able to invent intermediate-temperature solid oxide fuel cells (IT-SOFCs), which work in intermediate-temperature range $(600 - 800 \degree C)$ [5, 6]. At intermediate-temperatures range, there exist several demerits such as slow oxygen reduction reaction (ORR) rate, material stability, high activation energy requirements of the electrochemical process, sluggish cathodic reactions, incomplete fuel conversion, and redox stability. However, cobalt-containing perovskite cathode materials with mixed ionic and electronic properties have been developed, which has helped in resolving some of these challenges and enhances cathodic reaction, thereby increasing the overall performance of IT-SOFCs [7, 8]. Figure 1 shows the conventional SOFC and its various components.

The primary role of cathode in IT-SOFC is to facilitate ORR by catalysing the conversion of oxygen molecules (O_2) into oxygen ions (O^{2-}) at the cathode-electrolyte interface. Typically, cathode consists of mixed ionic-electronic conducting materials, which promote the migration of both electrons and oxygen ions [9, 10]. This redox process allows the release of energy and the generation of electrical current. Thus, cathode materials must ensure smooth transport of oxygen ions from the cathode-electrolyte interface to the anode-electrolyte interface through the electrolyte. This transport process is essential for the continuous flow of ions within the IT-SOFC, ensuring a steady electrochemical reaction. Efficient functioning of the cathode relies on maximizing the triple phase boundary (TPB) length, which is the region where the cathode, electrolyte, and fuel (usually hydrogen) meet [11]. Key to ensuring TPB effectiveness include optimized electrode design, high catalyst activity, adequate ionic-electronic conductivity, efficient thermal management, optimized gas composition, and electrode stability [12, 13]. Hence, cathode materials with a large TPB surface area are desirable to promote ORR and improve overall cell efficiency.

Fig. 1 Conventional SOFC and its various components

Conventionally, lanthanum strontium cobalt ferrite (LSCF) has been widely employed as the cathode material in IT-SOFCs [14-16]. While LSCF exhibits excellent electrochemical activity and conductivity, it suffers from some inherent limitations, such as high material cost, limited stability under reducing atmospheres, and dependency on scarce cobalt resources. These challenges have motivated researchers to explore alternative cathode materials that offer comparable or improved performance and are devoid of cobalt, thus addressing sustainability concerns. SrFeO3-6 perovskite oxide series are among the first group of cobalt-free cathode materials developed [17, 18]. With its mixed ionic and electronic properties, this group of perovskite oxides showed excellent and promising prospects for IT-SOCFs, oxygen permeation membranes, and oxygen sensor applications. However, its crystal structure and defect properties can vary depending on the oxygen vacancy content (δ), which can lead to a structural phase shift from cubic to brownmillerite at temperature $< 870 \,^{\circ}C$ [19, 20]. The resulting long-range ordering in oxygen vacancies reduces the electrical conductivity of this material, highlighting the urgent need to repress orthorhombic to brownmillerite phase at low temperatures to retain the superior electrical conductivity of the cubic phase. In order to address this limitation, researchers have attempted to stabilize cubic perovskite structure through fractional replacement of Fe sites with other elements such as La, Pr, Sm, Cr, Mo, Sb, Ce, and Nb [21–23]. Several perovskite oxides, which include Sr₂Ti_{0.9}FeNi_{0.1}O₆, Pr_{1.90-x}Ce_{0.1}CuO₄, YBaCo₂O₅₊₆, and $La_{0.54}Sr_{0.46}Zn_{0.20}Fe_{0.80}O_{3-\delta}$, have been studied as possible cathode materials for IT-SOFCs [24-26]. Other series of cobalt-free cathode materials have also been investigated for IT-SOFC application [27, 28]. Figure 2 gives an overview of the topics treated in the present study.

The main goal of this study is to provide an overview of the recent progress in cobalt-free cathode materials development for IT-SOFCs. Efforts were dedicated to ensuring that the literature reviewed is recent (within the last 4yrs), and







Fig. 2 Overview of the discussed topics in the present study

relevant to the current challenges limiting the wide use of cobalt-free cathode materials. Figure 3 shows the methodology adopted in this study, and the credibility of sources used. A systematic literature review was carried out to highlight the gaps, challenges, and limitations facing the practical application of cobalt-free cathode materials. Future research directions and possible solutions were recommended in addressing the key challenges identified by the present study. A vast number of review studies related to IT-SOFCs have been published in the last five years, but none was dedicated to addressing the constraints faced by cobalt-free cathode materials and cobalt-free symmetrical electrodes. Most of the existing studies are dedicated to addressing the challenges of the traditional solid oxide fuel cell, which operates at 800 to 1000 °C. Thus, the present study will serve as a reference to energy experts and related researchers looking for recent accomplishments in the field of IT-SOFCs cobalt-free cathode material development. Table 1 provides an overview of previous related review studies on cobaltfree cathode material development for solid oxide fuel cells. Recent review studies on cobalt-free cathode materials for traditional SOFC have highlighted advancements in nickel, manganese, and iron-based compositions, emphasizing their potential for mitigating cobalt's cost and toxicity concerns based on the information in Table 1. However, the need to lower the operation temperature of SOFC to the intermediate-temperature range necessitates a fresh review to assess emerging cobalt-free cathode materials and performance.

2 Overview of cathode reaction mechanism in IT-SOFC

Cathode in IT-SOFC is responsible for facilitating ORR, where oxygen molecules from air are electrochemically reduced to oxide ions. This reaction is crucial as it balances the charges produced during fuel oxidation at the anode, thereby enabling the flow of electrical current through the external circuit. Oxygen reduction reaction is typically the slowest step in the electrochemical process, imposing limitations on cell performance [37, 38]. Figure 4 shows the schematic region between the electrolyte and cathode where TPB exist. Equation (1) gives the overall cathode reaction in IT-SOFC [39]. The cathode reaction mechanism involves multiple steps, which include adsorption of oxygen molecules, dissociation into oxygen ions at TPB, diffusion of ions through the electrolyte, and incorporation into electrolyte



Fig. 3 Screening methodology adopted in the present study



Table 1 Overview of related review studies on cobalt-free cathode material development for SOFC

Case studied	Ref
A review on new cobalt-free cathode materials for reversible solid oxide fuel cells	[29]
Perovskite-structured cobalt-free cathode materials for solid oxide fuel cells	[<mark>30</mark>]
Short review on cobalt-free cathodes for solid oxide fuel cells	[31]
Lanthanide nickelates for their application on solid oxide cells	[32]
Cobalt-free cathode materials: families and their prospects	[33]
Recent advances, practical challenges, and perspectives of intermediate temperature solid oxide fuel cell cathodes	[21]
Mitigating thermal expansion effects in solid oxide fuel cell cathodes: a critical review	[34]
Cobalt-free perovskite cathodes for solid oxide fuel cells	[35]
Advancements in composite cathodes for intermediate-temperature solid oxide fuel cells: A comprehensive review	[<mark>36</mark>]



Fig. 4 Schematic of cathode triple-phase boundary

lattice [40]. The rate-limiting step often occurs at the TPB, where the oxygen molecules need to overcome activation barriers for adsorption and dissociation. Therefore, catalytic materials that can lower the activation barriers are essential for enhancing cathode kinetics. The $\frac{1}{2}O_2$ in Fig. 4 represents half a molecule of oxygen, and the $2e^{-}$ represents two electrons involved in the reduction process. The two oxygen vacancies on the surface of the cathode are denoted as $2VO^{**}$, and the oxygen vacancy left behind after the oxygen reduction VO_0 .

$$O_2 + 4e^- \to 20^{2-} \tag{1}$$

The microstructure of cathode materials is crucial for promoting effective diffusion of oxygen and surface interactions. Nanostructuring and controlling the grain boundaries can enhance TPB density and facilitate faster oxygen incorporation into electrolytes [41, 42]. Moreover, proper electrode-electrolyte contact is essential for minimizing polarization losses. Techniques such as infiltration, co-firing, and advanced deposition methods are currently being investigated to engineer well-defined microstructures in cathode materials [43, 44]. Figure 5 shows the schematics of possible oxygen incorporation pathways for a mixed ionic and electronic conductor. Thus, the process involved in oxygen surface exchange can be described by the following steps:

Step 1: Oxygen adsorption and dissociation

$$O_2 + 4e^- + 2vacancies \rightarrow 2O^{2-}$$
⁽²⁾



O-O dissociation with oxygen vacancy Incorporation of vacancy toward O*



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Step 2: Oxygen incorporation

 O^{2-} + vacancies $\rightarrow O^{2-}$ vacancycomplex $\rightarrow O^{2-}$ incorporation into cathodelattice (3)

Perovskite oxides, particularly those with mixed ionicelectronic conducting properties, have shown great potential as cathode materials in IT-SOFCs. Materials such as $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\delta}$ (LSCF) and $La_{0.8}Sr_{0.2}MnO_3$ (LSM) exhibit favourable oxygen transport properties and offer efficient pathways for charge transfer [45, 46]. These materials also exhibit excellent stability under operating conditions and possess reasonable catalytic activity for ORR. Advanced characterization techniques such as in-situ spectroscopy, electron microscopy, and computational modelling have provided more valuable insights into cathode reaction mechanism [47, 48]. These techniques allow researchers to directly observe the oxygen exchange processes, surface interactions, and defect chemistry, aiding in the design of improved cathode materials and structures.

3 IT-SOFC cathode materials

Several cathode materials have been investigated for IT-SOFC applications, each with its own advantages and limitations [49, 50]. Cathode materials should exhibit rapid and efficient ORR kinetics to minimize polarization losses and enhance overall cell efficiency. Operating at intermediate temperatures implies that cathode materials must have good thermal stability to prevent degradation and ensure prolonged cell lifespan. This involves resistance to sintering, phase transitions, and chemical interactions with other cell components. Lanthanum strontium manganite (LSM) is a widely studied cathode material due to its high electronic conductivity and good catalytic activity [51, 52]. However, it may suffer from strontium segregation at the cathode-electrolyte interface under certain conditions, leading to performance degradation. Li et al. [53] synthesized novel $La_{0.7}Sr_{0.3}Co_{0.25}Fe_{0.25}Ni_{0.25}Mn_{0.25}O_{3-\delta}$ (LSCFNM73) perovskite oxide via a one-pot combustion method. Findings from their investigations showed that the substitution of Sr²⁺ with La³⁺ increased the electrical conductivity, thermal expansion coefficient, oxygen vacancy concentration, and oxygen adsorption. The LSCFNM73 cathode material exhibited a high conductivity of 464 S/cm at 800 °C. The NiO-YSZ|YSZ|GDC|LSCFNM73-GDC fuel cell achieved a maximum power density of 1088 mWcm² at 800 °C, and demonstrated good thermal stability. Though LSCFNM73 appears to have excellent cathode potential, the effect of cobalt on the B-site was not investigated. A comprehensive study is needed to determine the effect of cobalt poisoning, stability, and degradation over a long period of time. Klyndyuk et al. [54] investigated NdBa(Fe,Co,Cu)₂O_{5+ δ}

(NBCC2) as cathode material for SOFC application. Their result showed that Cu- and Fe-doped variants show compatibility with commonly used electrolytes (GDC and YSZ), low thermal expansion coefficient (TEC), and high electrical conductivity. They reported that NBCC2 with slight Cu substitution exhibits the best performance, offering enhanced conductivity, stability, and electrochemical benefits, making it a promising cathode material for IT-SOFCs. While the results of their findings are exciting, further investigations are needed to understand the activation energy (E_a) of the system. The E_a is an important factor in determining how fast the electrochemical reduction of oxygen will be achieved. As large E_a values will mean sluggish cathodic reaction, thereby reducing the overall cell performance. Costilla-Aguilar et al. [55] synthesized $La_{1.8}Sr_{0.2}FeCoO_{5+\delta}$ (DP-LSCF) double perovskite oxide via modified sol-gel technique and evaluated its performance as cathode material for IT-SOFCs. The new compound displayed a stable rhombohedral structure (space group R3c) and good compatibility with GDC electrolyte. Raman analysis confirmed the double perovskite structure, while XPS showed mixed Co²⁺/Co³⁺ and Fe³⁺/Fe⁴⁺ states. Substituting Fe for Co lowered the thermal expansion coefficient value to 19.4×10^{-6} K⁻¹ and reduced the electrical conductivity from 9 to 50 S/cm at 300 - 900 °C. They obtained a favorable area-specific resistance (ASR) value of $0.42 \,\Omega \,\mathrm{cm}^2$ at 800 °C. Their electrochemical impedance spectrometer (EIS) analysis highlighted DP-LSCF as an excellent cathode material for IT-SOFCs. Figure 6 illustrates the electrical conductivity and TEC profile of their experiment.

Based on the profile analysis of DP-LSCF in Fig. 6(a), it's obvious that an increase in temperature results in a corresponding increase in electrical conductivity. This is very true for cathode materials when their electrical conductivity is measured in air. Perovskite oxides often exhibit mixed ionic and electronic conductivity. At lower temperatures, ionic mobility might dominate, which can impede electronic conduction. As the temperature increases, ionic mobility becomes more significant, allowing ions to move more freely within the material. The increase in ionic mobility can lead to higher electronic conductivity by facilitating fast charge movement. However, in the study, the electrical conductivity of DP-LSCF under an oxidizing environment (5%H₂/95%Ar) wasn't investigated. Exposing DP-LSCF to an oxidizing environment will trigger various degradation mechanisms, such as chemical reactions, phase changes, and microstructural alterations. These mechanisms can negatively impact the cathode's electrical conductivity over time. Further study should be conducted to understand the influence of oxidizing environment on DP-LSCF, so that more useful insights can be gained on the degradation mechanisms, and work towards mitigating it.

Liu et al. [56] investigated Ca-doped layered double perovskite $\text{Sm}_{1-x}\text{Ca}_x\text{BaCo}_2\text{O}_{5+\delta}$ (x=0-0.4; SCBC) oxides as





Fig. 6 (a) Electrical conductivity of DP-LSCF in air at 300 – 900 °C; (b) Thermal elongation measurement of DP-LSCF [55]. (This result was reproduced with copyright permission from Elsevier)

cathodes for IT-SOFCs. The SCBC cathodes demonstrated good chemical compatibility with CeO₂-based electrolytes at high temperatures, with Co³⁺ and Co⁴⁺ coexisting in the lattice due to Ca doping, promoting oxygen vacancies. They also reported that SCBC2 showed superior conductivity, catalytic activity, and CO₂ durability, suggesting potential use as cathode in IT-SOFC. The authors demonstrated the influence of calcium doping in enhancing the catalytic performance of SCBC cathode materials, but little was known about the thermal compatibility of these materials with the two commonly used electrolytes (YSZ and GDC). Further studies are needed to determine the appropriate thermal elongation of these materials under the influence of varying temperatures. Table 2 gives an overview of some recent findings in cathode materials development for IT-SOFC.

A critical and comparative analysis of Table 2 shows that composite cathode material $Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3-8}-Sm_{0.2}Ce_{0.8}O_{1.9}$ has good thermal elongation of 12.4×10^{-6} K⁻¹ at 30 to 600 °C. The good thermal expansion coefficient noticed in the composite cathode can be attributed to synergistic effects that lead to desirable properties. The interactions between the different constituents can result in composite cathode material with a more controlled and balanced thermal expansion behaviour. It's important to note that while composite cathode materials generally offer advantages in terms of thermal expansion coefficient, they also need to fulfil other requirements such as electrical conductivity, oxygen reduction kinetics, and chemical compatibility with the fuel cell environment. Another obvious fact is that most of the studies in Table 2 contain cobalt, this can be attributed to the fact that cobalt-containing materials are less prone to sintering at intermediate temperatures compared to other cathode materials. Despite all the unique properties of cobalt-containing

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جامعة قطر QATAR UNIVERSITY cathode materials used in IT-SOFC, its effectiveness is being hampered by the diffusive nature of cobalt which results in cell losses. The expensive nature of cobalt, high evaporation rate and poor thermal expansion coefficient (TEC) means other cobalt-free cathode materials need to be investigated.

4 IT-SOFC cobalt-free cathode materials

Traditional cathode materials often contain cobalt, and pose challenges due to their limited availability, high cost, and susceptibility to performance degradation. Thus, investigating alternative cobalt-free cathode materials will address these limitations and enhance the overall viability of IT-SOFC technology. Materials belonging to the Ruddlesden-Popper phase, such as La₂NiO₄ (LNO) are being investigated as potential substitutes for cobalt-containing perovskite oxides [65, 66]. This is due to their mixed ionic-electronic conductivity and good chemical compatibility, making them potential candidates for advanced IT-SOFC cathodes. Recent studies also showed that Ruddlesden-Popper phase containing cobalt has better thermal stability under increasing temperature than its perovskite oxide counterpart [67, 68]. Ghorbani-Moghadam et al. [69] successfully synthesized $La_{0.7}Sr_{1.3}Co_{1.x}Fe_{x}O_{4}$ Ruddlesden-Popper oxides (x = 0, 0.1, 0.3, 0.5) via modified sol-gel method for IT-SOFC cathodes. Structural analysis displayed a tetragonal phase with expanded lattice parameters due to Fe substitution in Co sites. Electrical conductivity decreased with Fe substitution, yet oxygen reduction reaction activity improved, leading to enhanced electrochemical performance with lower areaspecific resistance, making La_{0.7}Sr_{1.3}Co_{1-x}Fe_xO₄ a promising cathode materials. Challenges associated with some Table 2 Insight into recent findings on cathode materials development for IT-SOFC, with a comparative overview of the thermal expansion coefficient (TEC), electrical conductivity (σ_i) and power density (P)

Composition	TEC (*10 ⁻⁶ K ⁻¹)	$\sigma_i^{}\left(\text{S/cm}\right)$	$P(mW/cm^2)$	Ref
$\overline{X_{0.5}Sr_{0.5}Co_{0.8}Mn_{0.2}O_3} (X = La/Ba)$	-	At 600 °C X = La $\sigma_i = 3.51$ X = Ba $\sigma_i = 2.26$	At 600 °C X = La P = 277 X = Ba P = 186	[57]
$SmBa_{0.5}Sr_{0.5}Fe_2O_{5+\delta} (SBSF)$	At 50 – 950 °C TEC = 16.2	At 400 °C $\sigma_i = 36.4$	At 800 $^{\circ}$ C P=874	[58]
Co ₃ O ₄ -GDC	-	At 600 °C $\sigma_i = 2.54$	At 800 °C $P = 1012$	[59]
$Y_{1-x}Bi_xBaCo_2O_{5+\delta}$ (YBBCx, x=0.0 - 0.2)	At $100 - 935$ °C x=0 TEC = 14.1 x=0.1 TEC = 15.3 x=0.2 TEC = 14.1	At 300 °C x=0 $\sigma_i=4.5$ x=0.1 $\sigma_i=1.5$ x=0.2 $\sigma_i=1.49$	At 700 °C x=0 P=537	[60]
$Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3-\delta}-Sm_{0.2}Ce_{0.8}O_{1.9}$	At $30 - 600$ °C TEC = 12. 4	At 600 °C $\sigma_i = 67.06$	-	[<mark>61</mark>]
CuBi ₂ O ₄ (CBO)	-	At 800 °C $\sigma_i = 0.14$	At 750 °C $P = 507$	[<mark>62</mark>]
$Sr_2CoTaO_{6-\delta}$ (SCT)	At $30 - 1000$ °C TEC = 15.0	At 800 °C σ _i =7.5	At 800 °C P=448	[63]
$BaCo_{0.8}Zr_{0.1}Y_{0.1}O_{3\text{-}\delta}\left(BCZY\right)$	At 30 – 1000 °C TEC = 18.8	At 800 °C σ _i =4.55	At 800 °C P=460	[64]

Ruddlesden-Popper oxides are phase transitions, chemical degradation, or structural changes at intermediate-temperature. These issues can lead to cathode instability, reduced performance, and a shorter operational lifespan. Also, synthesizing Ruddlesden-Popper oxides can involve complex procedures, potentially leading to higher production costs. Developing scalable and cost-effective synthesis routes is important for practical implementation.

Qi et al. [70] investigated self-assembled cobalt-free $Ba_{0.5}A_{0.5}Fe_{0.8}Zr_{0.2}O_{3-\delta}$ (A = Sr²⁺/Sm³⁺, BSrFZ/BSmFZ) iron-based composite oxides, with Sr²⁺ and Sm³⁺ ions compared for enhanced ORR activity. Both composites showed uniform nanoparticle size, benefiting from multi-phase interactions during self-assembly. The BSrFZ composite exhibited superior oxygen vacancy content, adsorption, and redox activity, resulting in a high-power density of 1.64 mW/cm² at 750 °C. Their work showed BSrFZ as a potential cathode for IT-SOFC, with improved electrochemical performance. Wang et al. [71] synthesized $BaFe_{0.9}Nb_{0.1}O_{3-\delta}$ (BFNb) with 10 mol.% Nb⁵⁺ doped BF oxide as cathode material for IT-SOFCs application. The cubic phase in BFNb synthesized at 1000 °C facilitated disordered oxygen vacancies, and thus enhancing oxygen vacancy concentration. Favourable electrochemical performance and stability showed BFNb as a potential cathode in IT-SOFC. La_{0.8}Sr_{0.2}Fe_{1-x}Cu_xO_{3-δ} (LSF-Cux, x = 0.0 - 0.3) nanofiber cathodes doped with Cu at the B site were synthesized via electrospinning [72]. The calcination of the samples at 800 °C yielded perovskite-based structures with high crystallinity. Doping with copper increased the oxygen vacancy concentration and shifted the rate-limiting step to cathode charge transfer. Thus, demonstrating outstanding electrochemical properties. The study reported that LSFCu2 achieved a polarization resistance of 0.674 Ωcm² at 650 °C, while Ni-SDC/SDC/LSFCu2 single cell exhibited a stable power density of 362 mW/cm² at 600 °C for 50 h. Figure 7 shows the result profiles of their investigation.

Based on the result profiles in Fig. 7 (a - d), it's evident that LSFCux is a highly porous cathode material, with what appears to be an even distribution of nanofibers. Porous LSFCux will aid in providing pathways for reactant gases (oxygen and fuel) to diffuse within the cathode material. This will promote efficient gas-phase reactions at the electrode-electrolyte interface, enhancing overall cell performance. The excellent interchange at electrode-electrolyte interface is collaborated by the result profiles in Fig. 7 (e, f). Belotti et al. [73] explored A-site deficiency effects in $(Ba_{0.95}La_{0.05})_{1-x}$ FeO₃₋₈ perovskite oxides. Density functional theory (DFT) computations revealed charge effects enhancing p-type conductivity via lowered oxygen vacancy and increased ligand holes. Experimental validation showed improved electrical conductivity and catalytic performance at intermediate temperatures. Moderate deficiency of up to 10% yields stable power output, while excess deficiency of more than 20% results in the formation of secondary phases. The insights from their work can be used as a template for





Fig.7 SEM images of $La_{0.8}Sr_{0.2}Fe_{1-x}Cu_xO_{3-\delta}$ (LSFCux, x=0.0–0.3) nanofibers fired at 800 °C and distributions of fibre diameters. (a) x=0.0, (b)=0.1, (c)=0.2, (d)=0.3; (e) Electrochemical impedance spectroscopies of the single cell with LSFCu2 cathodes at 500–650

analyzing the limitation and effects of A-site deficiency in other cobalt-free oxides, and can also be used as a tool in the rational design of IT-SOFC's cathodes. Table 3 gives an overview of some recent findings on cobalt-free cathode materials for IT-SOFC applications.

Comparative analysis of Table 3 shows that cobalt-free cathode materials have good polarization resistance, with $La_{0.7}Sr_{0.3}Cu_{0.15}Fe_{0.85}O_{3-\delta}$ having the least value of 0.0153 Ω cm² at 700 °C. Low polarization resistance is vital for cathode materials in IT-SOFCs because it directly impacts the cell's efficiency, power output, voltage losses, stability, and compatibility at intermediate-temperatures. Materials

°C, (f) performance output curve for NiO-SDC/SDC/LSFCu2 anodesupported single cell at 500–650 °C [72]. (This result was reproduced with copyright permission from Elsevier)

with low polarization resistance facilitate faster electrochemical reactions, reduce overpotential, and contribute to overall improved cell performance. Though, while $La_{0.7}Sr_{0.3}Cu_{0.15}Fe_{0.85}O_{3-\delta}$ has low polarization resistance at 700 °C, it's also obvious that this material has low electrical conductivity of 1.54 S/cm in air at 700 °C compared to composite $La_{0.65}Ca_{0.35}FeO_{3-\delta}$ -Gd_{0.2}Ce_{0.8}O_{2- δ} (LCF-GDC), which has a good electrical conductivity of 100 S/cm in air at a lower temperature of 650 °C. Higher electrical conductivity reduces cathodic overpotential, allowing the cell to operate at lower temperatures with reduced energy losses. This is particularly important for IT-SOFCs that operate at

Table 3	Insight into cobalt-free
cathode	materials development
for IT-S	OFC, and comparative
overviev	w of their electrical
conduct	ivity (σ_i), polarization
resistan	$ce(R_p)$, and activation
energy ((E _a)

Cobalt-free composition	σ_i (S/cm)	$R_p (\Omega cm^2)$	$E_a (eV)$	Ref
LaBa _{0.5} Sr _{0.5} Fe ₂ O _{5.875-8} F _{0.125}	At 80	00 °C At	t 750 °C	At 600 – 750 °C [74]
	$\sigma_i = 45.45$	$R_{p} = 0.089$	$E_{a} = 1.0$	5
La _{0.65} Ca _{0.35} FeO ₃₋₆ -	At 65	50 °C At	t 750 °C	At 650 – 750 °C [75]
$Gd_{0.2}Ce_{0.8}O_{2-\delta}$ (LCF-GDC)	$\sigma_i = 100$	$R_{p} = 0.28$	$E_{a} = 1.1$	2
$BaCe_{0.5}Fe_{0.5}O_{3-\delta}$ (BCF)	At 80	00 °C At	t 800 °C	At 300 – 800 °C [76]
	$\sigma_i = 0.94$	$R_p = 0.1877$	$E_{a} = 0.3$	75
La _{0.7} Sr _{0.3} Cu _{0.15} Fe _{0.85} O _{3-δ}	At 70	00 °C At	t 700 °C	At 47 – 650 °C [77]
	$\sigma_i = 1.54$	$R_p = 0.0153$	$E_a = 0.1$	59
BaFe _{0.75} Ni _{0.25} O ₃₋₆ (BFNi25)	At 75	50 °C At	t 700 °C	At 500 – 700 °C [78]
	$\sigma_i = 4.5$	$R_{p} = 0.095$	$E_a = 1.1$	5
$Bi_{0.5}Sr_{0.5}Fe_{0.95}P_{0.05}O_{3-\delta}$ (BSFP)	At 70	00 °C At	t 700 °C	At 550 – 700 °C [79]
	$\sigma_i = 2.55$	$R_{p} = 0.18$	$E_a = 1.4$	7
Bi _{0.5} Sr _{0.5} FeO _{3-δ}	At 80	00 °C At	t 700 °C	At 550 – 700 °C [80]
	$\sigma_i = 1.8$	$R_{p} = 0.124$	$E_a = 2.6$	1



intermediate temperatures (600 - 800 °C) compared to traditional high-temperature SOFCs. Thus, there is a need to enhance the electrical conductivity of cobalt-free cathode materials to be able to compete fairly with those of cobaltcontaining counterparts. Most recent studies on cobalt-free cathode materials showed good activation energy, which is the minimum energy required to kickstart an electrochemical reaction occurring at the cathode-electrolyte interface [81, 82]. This reaction is essential for the overall operation of the fuel cell, as it generates electrons that flow through the external circuit to produce electric power. The activation energy of the cathode materials plays a significant role in determining the speed at which this electrochemical reaction occurs.

Lach et al. [82] synthesized $La_{1-x}Sr_xNi_{0.75}Cu_{0.25}O_{3-\delta}$ and La_{1-x}Sr_xNi_{0.5}Cu_{0.5}O_{3-δ} perovskites via the soft chemistry technique. They observed a transition from R-3c to Pm-3 m crystal structure at high temperatures. The doping with Sr reduced the phase transition temperatures, with $La_{1,v}Sr_{v}Ni_{0,5}Cu_{0,5}O_{3,8}$ -series showing the lowest at 450 °C. The study concludes that Sr-doping increased oxygen nonstoichiometry and thermal expansion coefficients, similar to solid electrolytes. Both samples showed good stability and chemical compatibility with GDC-electrolyte. However, the samples were not compatible with YSZ electrolyte. La_{1-x}Sr_xNi_{0.5}Cu_{0.5}O₃₋₈-series demonstrated promising performance in anode-supported IT-SOFCs, achieving a power density of 445 mWcm⁻² at 650 °C in a humidified H₂ environment, indicating its potential as a high-performance cathode material for intermediate-temperature SOFCs. Figure 8 shows the excellent electrochemical performance of $La_{0.95}Sr_{0.05}Ni_{0.5}Cu_{0.5}O_{3.8}$ -based cathode. Based on the result profile, we can conclude that the $La_{0.95}Sr_{0.05}Ni_{0.5}Cu_{0.5}O_{3-\delta}$ -based cathode has good power density at 600 and 650 °C. While the electrochemical impedance analysis was impressive, the values of nearly 0.7 and 1.6 Ω cm² seen on the EIS plot for polarization resistance at 650 and 600 °C are a bit high compared to those reported in the literature [83–86]. Thus, there is a need to further enhance the performance of this material with the addition of an electrocatalyst.

Dengie Chen et al. [87] investigated the influence of rare earth metals on various properties of composite oxides $LnBaFe_2O_{5+\delta}$. The study found that rare earth metals influence phase structure, oxygen content, conductivity, thermal expansion behaviour, microstructure, and electrochemical performance of the oxides. Electronic conductivity decreased with decreasing ionic radius of Ln³⁺, while oxygen desorption temperature and thermal expansion coefficient showed opposing trends. $SmBaFe_2O_{5+\delta}$ exhibited the best electrocatalytic activity for oxygen reduction reaction (ORR), achieving peak power densities at different temperatures. Additionally, cathodic polarization was found to enhance the ORR performance of $LnBaFe_2O_{5+\delta}$ cathodes. Metallic nanoparticle-decorated with ceramic anodes were synthesized via in-situ reduction of Sr₂FeMo_{0.65}Ni_{0.35}O₆₋₈ (SFMNi) in H₂ environment at 850 °C [88]. The reduction induced a phase transformation, resulting in the formation of mixed phases, including the Ruddlesden–Popper Sr₃FeMoO_{7.8}, perovskite Sr(FeMo)O₃₋₈, and FeNi₃ bimetallic alloy nanoparticle catalysts. It was observed that the resulting materials exhibited a unique structure with a homogeneous distribution of metallic nanoparticle catalysts on the ceramic surface. Furthermore, incorporating metallic nanoparticle catalysts onto the SFMNi ceramic anode led to a significant enhancement in electrochemical performance. This improvement can be attributed to the catalytic activity of the exsolved Fe-Ni alloy nanoparticle catalysts, which facilitated the electrochemical reactions at the anode-electrolyte interface. While most of the studies reviewed on cobalt-free cathode materials development for IT-SOFC provided valuable insight on strontium-doping and in-situ reduction in H₂ environment for enhanced ORR rate and linear thermal elongation, more investigation is needed to understand the chemical compatibility of these materials with YSZ and GDC electrolytes. Mixed ionic and electronic conduction (MIEC) in composite cathodes also show promise for enhancing ORR at lower temperatures. Fe-based cobalt-free perovskite emerges as a favourable cathode material owing to its catalytic activity, stability, and cost-effectiveness [89, 90]. Table 4 gives an overview of recently developed Fe-based perovskite cathode materials for IT-SOFCs.

Fig. 8 (a) Voltage and power density as a function of current density and (b) impedance spectra for anode-supported SOFC with La0.95Sr0.05Ni0.5Cu0.5O3-8-based cathode [82]. (This result was reproduced from MDPI open access journal, no copyright permission is required)



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Table 4 Cobalt-free iron-based cathode materials for IT-SOFCs

Composition	TEC (*10 ⁻⁶ K ⁻¹)	$R_p (\Omega cm^2)$	Ref
LaBaFe _{1.8} Zn _{0.2} O _{5+δ}	At 30 − 750 °C TEC = 11.4	At 800 °C $R_{p} = 0.014$	[91]
$Bi_{0.5}Sr_{0.5}Fe_{0.95}Nd_{0.05}O_{3\text{-}\delta}$	At $25 - 800 \degree C$ TEC = 14.1	At 700 °C $R_{p} = 0.062$	[<mark>92</mark>]
$Bi_{0.5}Sr_{0.5}Fe_{0.90}Zr_{0.10}O_{3-\delta}$	At $30 - 800$ °C TEC = 12.89	At 600 °C $R_{p} = 0.52$	[<mark>93</mark>]
$PrBaFe_{1.9}Mo_{0.1}O_{5+\delta}$	$At = 30 - 750 \degree C$ TEC = 18.8	At = 800 °C $R_p = 0.092$	[<mark>94</mark>]
$Pr_{0.93}BaFe_2O_{5+\delta}$	At $25 - 800 \degree$ C TEC = 15.95	$At = 800 \degree C$ $R_p = 0.059$	[<mark>95</mark>]
$La_{0.35}Ca_{0.15}Ba_{0.5}FeO_{3\text{-}\delta}$	At $30 - 750 ^{\circ}C$ TEC = 13.56	$At = 800 \degree C$ $R_p = 0.059$	[<mark>90</mark>]
$SmBa_{0.5}Sr_{0.5}Fe_2O_{5+\delta}$	At $50 - 800$ TEC = 15.2	$At = 800 \ ^{\circ}C$ $R_p = 0.023$	[<mark>96</mark>]

Comparative analysis of Table 4 shows that cobalt-free iron-based LaBaFe_{1.8}Zn_{0.2}O_{5+ δ} has the best electrochemical performance with polarization resistance of 0.014 Ω cm² at 800 °C, and an average linear thermal expansion coefficient of 11.4×10⁻⁶ K⁻¹ at 30 to 750 °C. Despite the good electrochemical properties of LaBaFe_{1.8}Zn_{0.2}O_{5+ δ}, Fe-based cathode materials often suffer from chemical instability under operating conditions, leading to degradation and reduced cell performance. This instability can result from reactions with the electrolyte or other cell components.

Traditionally, the intermediate-temperature solid oxide fuel cell (IT-SOFC) has distinct electrode materials for the cathode and anode. However, efforts have been directed toward the development of electrodes for symmetrical configuration. This will help reduce material costs and effectively ensure homogeneity in electrode material development investigations. Nevertheless, there remains inadequate information regarding which materials can effectively serve as symmetrical electrodes. Novel Srdoped PrBa_{0.5}Sr_{0.5}Fe₂O_{5+δ} (PBSF) symmetrical electrode was synthesized via a conventional sol-gel technique [97]. A cubic structure was reported for PBSF after calcination in air for 1000 °C. The PBSF demonstrated robust structural integrity when exposed to a CO₂ atmosphere and showed compatibility with the LSGM electrolyte. In comparison to the original PBF, the polarization resistance of PBSF decreased significantly by 75% in air and 61% in H₂. Furthermore, PBSF electrode showed resilience against CO₂ poisoning and maintained short-term stability. The study reported that in a single cell configuration with a 300 µm-thick LSGM electrolyte supporting PBSF/ LSGM/PBSF, the maximum power output (Pmax) reached 1067 mWcm⁻² at 850 °C using humidified hydrogen fuel and 535 mWcm⁻² using propane fuel at 800 °C. Additionally, effective electrolysis of CO2 was accomplished using PBSF as a symmetrical electrode. The PBSF electrode demonstrated consistent stability in both fuel cell and electrolytic cell operations. Their findings strongly indicate that PBSF is a promising symmetrical electrode material for IT-SOFC applications. Figure 9 shows the structural integrity of Sr-doped PrBa_{0.5}Sr_{0.5}Fe₂O_{5+ δ} (PBSF) and its chemical compatibility with commonly used electrolyte. Table 5 illustrates recent findings on electrode materials for symmetrical solid oxide fuel cell operating at intermediate temperatures.

A critical overview of the recently developed electrode materials for symmetrical solid oxide fuel cell in Table 5 shows that the polarization resistance values in air are higher compared to traditional intermediate-temperature solid oxide fuel cells cathode materials. Realistically, it's difficult to synthesize electrode materials with excellent electrochemical properties for both fuel oxidation and oxygen reduction reactions. Thus, there is a need to control the microstructure of this type of electrode material to minimize grain boundaries and maximize the percolation pathways for charge carriers. This can be achieved through careful optimization of sintering conditions, grain size control, and the addition of pore formers. The study on identical electrodes for symmetrical configuration is mainly restricted to polarization resistance measurement in air, because our concern is mainly on cobalt-free cathode materials development for IT-SOFC applications.

5 Challenges and future research recommendations on cobalt-free cathode materials for IT-SOFC

Despite advancements in cobalt-free cathode materials, many still face challenges related to performance and stability. Lower electrical conductivity, sluggish oxygen reduction kinetics, and susceptibility to chemical and thermal degradation can hinder the overall efficiency and long-term stability of IT-SOFC as evident from the reviewed literature. A significant limitation lies in the incomplete understanding of the fundamental mechanisms governing the performance of cobalt-free cathode materials [103]. This has hampered the ability to design and optimize materials with targeted properties, hindering progress in achieving high-efficiency IT-SOFC. The integration of cobalt-free cathode materials with other cell components, such as electrolytes and anodes, can lead to compatibility issues and performance degradation. Thus, interactions at the interfaces between different materials need further investigation to ensure overall system stability. Moving from laboratory-scale synthesis to largescale manufacturing of cobalt-free cathode materials presents challenges related to cost-effectiveness, reproducibility and maintaining consistent performance [104]. Hence,

Fig. 9 (a) XRD profile of PBF and PBSF samples and PBSF sample was reduced at 5%H₂/Ar at 800 °C for 10 h and then oxidized at 800 °C for 10 h in air; (**b**) An enlarged view of (110) peak of PBF and PBSF samples; (c) Chemical compatibility between PBSF and LSGM electrolyte; (d) XRD patterns of PBSF fired at 800 °C for 10 h at 6% CO₂ and annealed at 800 °C for 10 h at 100% CO₂ [97]. (This result was reproduced with copyright permission from Elsevier)



innovative approaches are needed to address these hurdles and enable practical commercialization. The pool of suitable cobalt-free cathode materials is still limited compared to traditional cobalt-containing counterparts. Exploring and identifying new materials with improved properties is crucial to broadening the range of options for IT-SOFC development. Below are the specific future research recommendations on cobalt-free cathode materials development for IT-SFOC applications:

- There is a need to conduct a comprehensive study to understand the oxygen reduction kinetics and mechanisms in cobalt-free cathode materials. Advanced computational modelling and experimental techniques can provide insights into surface reactions, charge transfer processes, and diffusion mechanisms, aiding in material design.
- Research efforts should focus on developing scalable synthesis routes for cobalt-free cathode materials that ensure consistent properties. Comprehensive characterization techniques should be employed to correlate material structure, composition, and performance, enabling targeted improvements.
- Investigating the interactions between cobalt-free cathode materials and other cell components is vital. Interface engineering strategies can be explored to enhance compatibility and mitigate performance degradation in multi-component IT-SOFC systems.

- Rigorous electrochemical testing, including impedance spectroscopy and electrochemical performance under relevant operating conditions, can provide valuable insights into the limitations of cobalt-free cathode materials and guide their optimization.
- Long-term stability assessments under realistic operating conditions are essential to evaluate the performance and durability of cobalt-free cathode materials. These studies can guide improvements and provide insights into degradation mechanisms.
- More research effort is needed on the development of cobalt-free electrode materials for symmetrical solid

 Table 5
 Various investigations on electrode materials for symmetrical solid oxide fuel cell and its polarization resistance in air

Composition	$R_p (\Omega cm^2)$	Ref
SrFeO _{3-δ}	At 800 °C	[<mark>98</mark>]
$La_{0.00}Cr_{0.75}Mn_{0.25}O_{2.55}$	$R_p = 0.082$ At 750 °C	[9 9]
$Ce_{0.9}Gd_{0.1}O_{1.95}$	$R_{p} = 0.29$	[]
NdBaFe ₂ O _{5+δ}	At 750 °C R.=0.27	[100]
$Sr_{1.9}La_{0.1}Fe_{1.5}Mo_{0.5}O_{6\text{-}\delta}$	م At 800 °C	[101]
	$R_{p} = 0.075$	F1001
$PrBaFe_{1.8}Ta_{0.2}O_{5+\delta}$	$R_{p} = 0.171$	[102]
$Nd_{0.5}Sr_{0.5}Mn_{0.5}Cr_{0.5}O_{3\text{-}\delta}$	At 800 °C	[103]
	$R_{p} = 0.12$	

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oxide fuel cells working at intermediate-temperature range, as existing studies are not sufficient.

• Future studies should consider investigating agricultural waste, such as rice-husk crop residues, weeds, leaf litter, sawdust, forest waste, and livestock waste, as possible cathode materials in IT-SOFC. This will go a long way in addressing the high cost associated with traditional cobalt-containing cathode materials, and any environmental concerns.

Cobalt-free cathode materials hold great prospects for enhancing the efficiency and sustainability of IT-SOFCs, several limitations must be addressed through tailored research efforts. A deep understanding of material properties, interfaces, and performance mechanisms will pave the way for the development of advanced cobalt-free cathode materials that enable practical implementation.

6 Conclusion

In the present study, we have discussed a range of cobalt-free cathode materials, which include perovskites, pyrochlores, Ruddlesden-Popper, spinels, and double perovskites. Literature reviewed showed that these materials exhibit promising electrochemical performance, thermal stability, and compatibility with IT-SOFC operating conditions. However, it is evident that the development of cobalt-free cathode materials is not without its challenges. Issues such as chemical stability, conductivity, and thermal compatibility with commonly used electrolytes (YSZ and GDC) in IT-SOFC must be addressed to ensure long-term reliability and efficiency. Mixed ionic conducting cobalt-free LaBa_{0.5}Sr_{0.5}Fe₂O_{5.875-δ}F_{0.125} showed good electrical conductivity of 45 S/cm at 800 °C, and a favourable polarisation resistance of 0.089 Ω cm² at 750 °C. Hence, optimizing, and enhancing this group of materials could yield more desirable electrochemical properties for oxygen reduction reaction. Furthermore, the transition from laboratory-scale studies to practical-based IT-SOFC applications requires careful consideration of several factors such as stability, scalability, and long-term performance. The potential benefits of reduced degradation, cost affordability and improved durability offered by some cobalt-free cathode materials need continued investigation under realistic operating conditions. Despite the significant progress made in the field, cobalt-free cathode materials are not the only solution to the challenges associated with cobalt-containing counterparts. Different operating conditions, desired performance metrics, and cell configurations will dictate the optimal choice of materials. Thus, continued research in the field to explore new materials, optimize existing ones, and bridge the gap between fundamental understanding and practical implementation is necessary.

Authors contribution Lukman Ahmed Omeiza and Asset Kabyshev: Formal analysis, Writing – original draft. Lukman Ahmed Omieza: Writing – review & editing, Conceptualization, Investigation, Resources, Supervision. Abul Kalam Azad: Formal analysis, Writing – original draft. Kenzhebatyr Bekmyrza: Validation, Investigation, Methodology, Writing – review & editing. Marzhan Kubenova and Shammya Afroze: Formal analysis, Writing – original draft. Kenzhebatyr Bekmyrza and Asset Kabyshev: Writing – review & editing. Saifullah Abu Bakar: Writing – original draft. Lukman Ahmed Omieza: Writing – review & editing. Kairat A Kuterbekov and Abul Kalam Azad: Resources.

Funding This study was supported by the grant with reference number BR21882359, provided by the Ministry of Science and Higher Education of Kazakhstan.

Data availability The data and materials that have been used are embedded in the body of the manuscript.

Code availability Not applicable.

Declarations

Ethics approval Not applicable.

Consent to participate Not applicable.

Content for publication Not applicable.

Conflict of interest The authors affirm that there is no conflict of interest to declare, given that all sources of information used have been appropriately cited and attributed.

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