Electrophoretic Deposition of Ba$_{0.5}$Sr$_{0.5}$Co$_{0.8}$Fe$_{0.2}$O$_{3-\delta}$-SDC Carbonate Composite Coating on Solid Oxide Fuel Cell Interconnect

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Abstract

Application of protective coating on ferritic stainless steel (FSS) as interconnect is crucially essential to restrict degradation of cell performance. In this study, barium strontium cobalt ferrite-samarium doped ceria (Ba$_{0.5}$Sr$_{0.5}$Co$_{0.8}$Fe$_{0.2}$O$_{3-\delta}$-Sm$_{0.2}$Ce$_{0.8}$O$_{1.9}$) carbonate (BSCF-SDCC) composite perovskite powder are fabricated by low speed wet milling method. The BSCF-SDCC is then applied as protective coating on ferritic stainless steel via electrophoretic deposition (EPD) technique. The BSCF-SDCC perovskite powder is examined by X-ray diffraction (XRD) and elemental energy dispersive X-ray spectroscopy (EDS). The characterisation of deposition is examined by scanning electron microscope (SEM). XRD displayed distinct crystalline phases of BSCF and SDCC and EDX results displayed a uniform element distribution in powder after milling. The EPD of BSCF-SDCC perovskite powder on ferritic stainless steel is conducted at 5 V to 12 V with duration of coating from 5 min to 20 min in an organic solvent. EPD at 10V for 10 min and 12V for 5min are found to produce the best coating parameter through the observation of the deposition morphology and thickness. Crack free and uniform coating are observed. The highest deposition weight and thickness obtained is 7.2mg and 81.6µm respectively. Hence EPD method has been successfully applied in depositing perovskite BSCF-SDCC on stainless steel with remarkable coating properties.

Keywords: Perovskite, Interconnect, Electrophoretic deposition.

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Introduction

Solid Oxide Fuel Cell (SOFC) is attracting huge consideration in recent years due to its high energy conversion efficiency with low pollutant emission [1]. Interconnects detach anode of a cell and cathode of another cell to prevent consolidation of oxidant and fuel. Interconnects are connected to external electric circuit to deliver electricity generated from single cell to external application [2]. Ferritic stainless steels are preferable choice for interconnect material in application of low to intermediate temperature (400°C to 800°C) SOFC due to low cost, good manufacturability, good conductivity and good thermal expansion coefficient match with the adjacent electrode [3-5]. However, one compelling disadvantage of ferritic stainless steel is formation of chromium oxide (Cr$_2$O$_3$) or chromia scale at elevated temperature. Chromia scale forms in vapor stage migrates and eventually deposits on triple phase boundary of electrolyte and thus, leading crucial degradation of cell performance [6].
An effective way to overcome chromia poisoning issue is by application of deposition layer on interconnect to reduce outward diffusion of chromium reaction with oxygen. Perovskite oxide coating such as lanthanum strontium manganese, LaSrMn (LSM), lanthanum strontium cobalt ferrite, LaSrCoFe (LSCF), lanthanum calcium chromite, LaCaCr (LCC) and lanthanum strontium cobalt, LaSrCo (LSC) have been employed as protective layer on interconnect [7-10]. These perovskite oxide deposition were found to exhibit better deposition characteristics, i.e uniform coating and remarkable area specific resistance (ASR) of lower than 0.1 Ωcm\(^{-1}\) which are indeed the essential requirement of interconnect protective layer [6]. Substitution of barium ions into lanthanum ions in LSCF barium, thus yielding barium strontium cobalt ferrite, BaSrCoFe (BSCF) is found to be a promising perovskite materials. BSCF has been proven to exhibit high electric conductivity and low ASR at high temperature [11,12]. However, BSCF demonstrates poor electric conductivity at low operating temperature. Thus, in recent years, BSCF-SDCC perovskite composite has been developed by incorporating electrolyte of samarium doped ceria carbonate (SDCC) into BSCF for low temperature SOFC [13]. However, BSCF-SDCC perovskite composite developed at high speed milling was found to be inadequate in phase stabilisation. Therefore, the development of BSCF-SDCC and appropriate characterisation of BSCF-SDCC deposition onto interconnect is of high novelty.

There are various deposition techniques available, such as sol gel dip coating, magnetron sputtering, screen printing and electrophoretic deposition [14]. Electrophoretic deposition (EPD) is particularly a simple, low cost and effective method producing precise coating thickness and robust morphology adjustment [15]. In this study, EPD was employed to deposit BSCF-SDCC perovskite composite powder on ferritic stainless steel interconnect.

The influence of EPD parameters such as voltage supplied and duration on the BSCF-SDCC coating is indisputable. Thus, BSCF-SDCC coating deposited through EPD by using the voltage supplies of 5V to 12V and the deposition duration of 5 to 20 mins were investigated.

**Materials and Methods**

**Sample Preparation**

Commercial powder SDC (Sigma Aldrich) was mixed with binary carbonates (67 mol%Li\(_2\)CO\(_3\) and 33 mol% Na\(_2\)CO\(_3\)) (Sigma Aldrich) in a weight ratio of 4:1 by milling at 150rpm in ethanol for 24 hours. Then, the slurry was dried and calcined at 680°C for two hours. SDCC was mixed with commercial powder BSCF (Sigma Aldrich) by ratio of 1:1 at 150 rpm for two hours in ethanol and was dried.

Square samples of 1cm x 1.5cm stainless steels were cut from commercial SUS430 stainless steel sheet. The samples were grounded with SiC sand paper to remove surface defects. Afterwards, the residue from grounding process was cleaned with acetone in ultrasonic bath for 10 min followed by rinsing with deionised water.

**EPD set up**

Cathodic electrophoretic deposition was performed in 100ml glass beaker cell. The counter electrode was set up by stainless steel sheet with 3cm x 3cm dimension. Counter electrodes were arranged in parallel with the substrate and separated by 1cm distance. The suspension applied for EPD was 10g/l BSCF-SDCC in ethanol. Polydiallyldimethylammonium chloride (PDADMAC) was dropped into suspension by 10μl as dispersing agent. The suspension was homogenised by a sonicator (Elma, German) for 5 min. The EPD was conducted in suspension of pH 8, as prior determined as stable suspension, analysed by zeta potential analysis. A DC voltage power supply from 5 to 12V
was applied. The deposition time was varied from 5 to 20 mins. After completion of EPD, the samples were air dried at room temperature.

**Characterization**

Phases of as-prepared BSCF-SDCC powder by high speed milling were identified by X-ray diffraction (XRD, Bruker D8 Advance, Germany). Energy Dispersive Spectroscopy (EDS, JSM 6380LA-JEOL, Japan) was performed to identify elemental distribution of the BSCF-SDCC powder. Deposition morphology and thickness were examined by field emission scanning electron microscope, FESEM (Hitachi, SU1510, Japan).

**Result and Discussion**

The XRD pattern of BSCF composites after 150 rpm milling in ethanol is as presented in Figure 1. Both cubic BSCF phase (JCPDS No. 01-079-5253) and cubic SDC phase (JCPDS No. 01-075-0158) were detected in the BSCF composites. Carbonate was present in amorphous phase after calcination of SDCC at 680°C. Slight decrease in peak intensity of BSCF was detected in BSCF composite which may be due to impact caused by BSCF particle collision during milling process. The collision energy had slightly destructed BSCF phase which was also reported by Ng et. al. on pure BSCF milling at 150 rpm [16]. Despite of the slight decrement of BSCF crystalline phase, milling at 150 rpm in ethanol did not any inflict significant chemical reaction thus still maintaining crystalline phase of BSCF composites.

![Figure 1: XRD pattern of composite BSCF-SDCC composite powder](image)

Figure 2 shows the elemental distribution of developed BSCF-SDCC. All elements in BSCF were found to be homogeneously distributed. The analysis had detected the carbon element in the BSCF-SDCC composites, thus proving the carbonate presence.
Figure 2: Elemental distribution of BSCF composites

Figure 3 presents the morphology of stainless steels SUS430 coated BSCF-SDCC by EPD, ranged from 5 to 12V for 5 to 20 mins duration. EPD at 5V and 7V indicated less particle deposition on the stainless steel surface. This phenomena might occur due to the electrolysis of water and poor particle distribution [17]. EPD at 10V shows successful deposition of more particles on the stainless steel substrate and thus yielding a uniform layer of deposition. The deposition uniformity was in agreement with weight of deposition as depicted in Table 1. Increased weight of deposition were observed with increased uniformity of the deposition layer. A turbulent flow were vividly observed during the 12V deposition process. The deposition performed at 12V for 5 mins coating was uniform and the depositions were less uniform compared to deposition conducted at 10 to 20 mins. This suggested that long exposure of over strong electric field strength led to deposition of lesser uniform.

<table>
<thead>
<tr>
<th>Elements</th>
<th>Atomic (%)</th>
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<tbody>
<tr>
<td>C</td>
<td>20.370</td>
</tr>
<tr>
<td>Fe</td>
<td>5.233</td>
</tr>
<tr>
<td>Co</td>
<td>19.850</td>
</tr>
<tr>
<td>Sr</td>
<td>11.463</td>
</tr>
<tr>
<td>Ba</td>
<td>11.552</td>
</tr>
<tr>
<td>Ce</td>
<td>22.216</td>
</tr>
<tr>
<td>Sm</td>
<td>4.357</td>
</tr>
<tr>
<td>Total</td>
<td>100%</td>
</tr>
</tbody>
</table>

Figure 3: SEM micrographs of BSCF-SDCC deposited on SUS430
Table 1: Deposition weight of BSCF-SDCC

<table>
<thead>
<tr>
<th></th>
<th>5V</th>
<th>7V</th>
<th>10V</th>
<th>12V</th>
</tr>
</thead>
<tbody>
<tr>
<td>5 min</td>
<td>0.90±0.45</td>
<td>1.07±0.31</td>
<td>3.30±0.26</td>
<td>6.60±0.14</td>
</tr>
<tr>
<td>10 min</td>
<td>1.11±0.52</td>
<td>2.07±0.31</td>
<td>7.00±0.16</td>
<td>5.41±0.32</td>
</tr>
<tr>
<td>15 min</td>
<td>1.94±0.35</td>
<td>5.27±0.25</td>
<td>6.40±0.23</td>
<td>2.57±0.48</td>
</tr>
<tr>
<td>20 min</td>
<td>1.14±0.36</td>
<td>2.03±0.26</td>
<td>5.43±0.40</td>
<td>1.02±0.44</td>
</tr>
</tbody>
</table>

The cross section of the coated stainless steel was observed to identify the deposition thickness as shown in Figure 4. The maximum thickness of deposition (46.3 µm) of 5V was produced at 15 mins duration and maximum deposition thickness (58.5 µm) of 7V was also produced at 15 mins duration. However, morphology of the deposition at 5V for 15mins and 7V for 15 mins deposition were not uniform. The maximum deposition thickness (81.6 µm) was obtained at 10V for 10 mins. A stronger electrical field applied at 12V for 5 mins yielded maximum thickness of 71.9 µm. The deposition thickness was decreasing followed the longer duration. Comparing deposition morphology and thickness, 10V 10min and 12V 5 min developed homogenous, dense, uniform deposition film and achieved EPD coating thickness requirement which is below 100 µm [18]. This also contributed to well-developed BSCF-SDCC powders with no agglomeration by wet-milling method.

![Figure 4: Deposition thickness of BSCF-SDCC coating on SUS 430](image-url)
Conclusion

High speed milling process of obtaining BSCF-SDCC composite, the XRD and EDS manifest excellent crystalline perovskite phase and well distributed elemental distribution. Electrophoretic deposition is successfully applied for BSCF-SDCC composite powder deposition on SUS430 stainless steel. Depositions at 10V for 10 mins and 12V for 5 mins were observed to yield deposition with excellent morphology and deposition thickness. Thus, the current study confirms the possibility of obtaining BSCF-SDCC deposition on SOFC interconnect by cathodic EPD using organic suspension.

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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.

References


