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Fabrication of Multilayers Electrodes and Electrolyte Via Screen Printing for Metal Supported Solid Oxide Fuel Cell

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Abstract: Metal-Supported Solid Oxide Fuel Cell (MS-SOFC) were produced using a manual screen-printing method on 430 stainless steel (SS430) substrates. Each of MS-SOFC sample was fabricated by using manual screen printing with two different mesh screens which are 305 and 355. The fabrication of NiO-GDC composite anode powder was done by mixing 60wt% NiO and 40wt% GDC. Meanwhile, 50wt% LSCF and 50wt% GDC was mixed to produce LSCF-GDC composite cathode powder. NiO-GDC, LSCF-GDC and GDC powders went through calcination in the furnace at 950°C for 2 hours. MS-SOFC samples with a different number of repetitions during the screen-printing process were sintered at 900°C for 90 minutes. In this study, the phase analysis was conducted via X-Ray Diffraction (XRD) method for commercial powder and composite powders. A good XRD pattern was obtained without the presence of any secondary peak in composite anode and cathode powder. The XRD data obtained were analysed to obtain the lattice structure and crystallise size for all the commercial and composite powder. 24.59 nm, 24.38 nm, 13.34 nm are the average crystallise size for NiO, GDC and LSCF, respectively. Scanning Electron Microscope (SEM) and Energy Dispersive Spectroscopy (EDS) were used to identify the thickness and distribution of elements on each MS-SOFC layer. As a result, the SOFC component layers fabricated by screen printed using 305-mesh screen at 10 times number of printings was selected as the ideal MS-SOFC sample. This is because the thickness of the layers obtained is lower compared to layers from mesh screens 305 and 355 at 15 and 20 times the number of printings which is 11.8 μm, 11.9 μm and 18.2 μm for anode, electrolyte and cathode, respectively. Thin electrode layer will produce low polarization resistance and can improve the SOFC performance itself.

Keywords: GDC, LSCF, NiO, Metal Supported, Screen Printing, Solid Oxide Fuel Cell

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1. Introduction

The configuration of a single cell unit of planar solid oxide fuel cell (SOFC) consists of anode, electrolyte, cathode, and interconnect. SOFC can be divided into several types namely as Electrolyte supported SOFC (ES-SOFC), Anode supported SOFC (AS-SOFC), Cathode supported SOFC (CS-SOFC) and Metal supported SOFC (MS-SOFC). Interconnect components serve to separate between cell units in the stack and for the transfer of electrical current between cell units to the external circuit. SOFC operating at low temperatures 400℃ until 600℃ can use metal materials such as stainless steel for interconnect components. This interconnection can also serve as a support for a single SOFC cell where this configuration is called MS-SOFC. 430 stainless steel metal is used as MS-SOC substrate [1]. MS-SOFC has many advantages over other SOFC types in terms of raw material cost and fabrication, mechanical strength, thermal shock tolerance and redox. SOFC has used two special designs, tubular and planar SOFC. The planar design shows a higher force density than the tubular design. Planar SOFC provides a much higher force density and potentially much lower than tubular cells [2].

The materials used in the fabrication of MS-SOFC are commercial NiO, GDC and LSCF powders to produce composite powders for electrodes NiO-GDC anode and LSCF-GDC cathodes. NiO-GDC, GDC, and LSCF-GDC powders were used to produce screen printing inks that was printed on the substrate using two different mesh screens. Screen printing is one of the layer fabrication methods for SOFC cell components. There are many factors that affect the layer properties of SOFC components such as mesh printing screen, ink viscosity etc. In this research, two different screen mesh values were used, 355 screen mesh and 305 screen mesh for the fabrication of multilayer electrodes and electrolyte for MS-SOFC.

The main objective of the research is to fabricate multilayers electrodes (cathode and anode) and electrolyte via screen printing MS-SOFC with free defect layers after the sintering process. Another objective is to determine the influence of mesh screen printing on the physical properties, thickness, and morphology of multilayers components (cathode, anode, and electrolyte) after the sintering process.

2. Materials and Methods

Before the fabrication of MS-SOFC single cell process can be carried out, it is important to study the types of materials to be used in this research and the best methods for the fabrication. Several important processes need to be carried out first such as preparation of composite powder, calcination process, preparation of screen-printing ink and the sample sintering process that has been printed using the manual screen-printing method.

2.1 Materials

Commercial powders such as nickel oxide (NiO), gadolinium doped ceria (GDC) and lanthanum strontium cobalt ferrite (LSCF) are the earliest materials to be used in the fabrication of MS-SOFC. The material used for the preparation of composite anode powder NiO-GDC was 60wt% NiO and 40wt% GDC. The mixture of these powders was mixed through ball milling method. 50wt% LSCF and 50wt% GDC were mixed for the fabrication of composites cathode powder LSCF-GDC. Composite anode NiO-GDC and composite cathode powder LSCF-GDC were used for the fabrication of screen-printing inks. Screen printing inks were prepared by adding dispersers, binders, and solvents with composite powders materials composition for the fabrication of anode, electrolyte and cathode electrode screen printing ink is shown in Table 1 until Table 3.

Table 1: Composition of Cathode Ink

Ingredient	Material	Density (g/cm^3)	Mass (g)	Volume (cm^3)	Volume (%)	Weightage (%)
Cathode Powder	LSCF	6.100	10.000	1.640	12.50	33.93
Electrolyte Powder	GDC	7.226	10.000	1.380	10.55	33.93
Dispersant	Oliec Acid	0.890	0.400	0.450	3.44	1.36
Binder	Ethycellulose	1.140	0.400	0.350	2.68	1.36
Solvent	Terpinol	0.934	8.674	9.290	70.83	29.43

Table 2: Composition of Electrolyte Ink

Ingredient	Material	Density (g/cm^3)	Mass (g)	Volume (cm^3)	Volume (%)	Weightage (%)
Electrolyte Powder	GDC	7.226	20.000	2.770	24.00	70.83
Dispersant	Oliec Acid	0.890	0.400	0.450	3.91	1.42
Binder	Ethycellulose	1.140	0.400	0.350	3.04	1.42
Solvent	Terpinol	0.934	7.435	7.960	69.04	26.33

Table 3: Composition of Anode Ink

Ingredient	Material	Density (g/cm^3)	Mass (g)	Volume (cm^3)	Volume (%)	Weightage (%)
Anode Powder	NiO	6.808	12.000	1.760	12.93	39.86
Electrolyte Powder	GDC	7.226	8.000	1.110	8.12	26.57
Dispersant	Oliec Acid	0.890	0.400	0.450	3.31	1.33
Binder	Ethycellulose	1.140	0.400	0.350	2.57	1.33
Solvent	Terpinol	0.934	9.304	9.960	73.07	30.91

For the screen-printing process, SS430 are used as substrate for this research. The ink was printed manually using screens with 305 mesh and 355 mesh in layers starting with the NiO-GDC layer which acts as an anode electrode on 430 stainless steel.

2.2 Methods

The method of preparation of composite powder was by mixing commercial powdera to be used either NiO with GDC or LSCF with GDC in a zirconia jar with zirconia balls. Ethanol was used as a grinding medium. The high energy ball milling (HEBM) planetary machine (Pulverisette 5 Fristch, Germany) was used to increase the homogeneity of the mixture. The process of HEBM at room temperature was done for 2 hours at a speed of 550 rpm with 10 minutes milling and 10 minutes rest. Once the HEBM process was completed, the solution was dried in an oven for 24 hours at a temperature 80℃. After the solution has been dried, the calcination process in the furnace was carried out for 2 hours at 950℃ before being crushed using agate mortar to form powders. Screen printing ink is prepared using a Triple Roll Mills machine (Exact 80, Germany) by adding dispersant, binder, and solvent into composite powders.

After the inks had been prepared, the inks were printed layer by layer starting with NiO-GDC as an anode component. The anode layer was then dried in an oven at 250℃ for 60 minutes to remove the solvent. The printing process is repeated for the electrolyte and cathode layers. Once all the layers are

printed on the SS430 substrate surface, the sintering process was carried out for 90 minutes at 900℃. For the MS-SOFC testing, the XRD, SEM and EDS machines used to identify the presence of phase in the powder, the thickness of each layer components (anode, cathode and electrolyte) and the distribution of elements in the MS-SOFC layer.

Each powder in this research has its own crystallize size. By using XRD data obtained from Eva Diffrac Plus Software, the crystallite size for each peak can be calculated by using the Scherrer equation (Eq. 1) before the average crystallite size can be obtained. XRD data for NiO, LSCF and GDC commercial powders can be used to determine Full Width at Half Maximum (FWHM) values and peak position values from analysis using OriginPro 2019 software before it can be applied into the Scherrer equation to calculate crystallize size.

$$D = \frac{K\lambda}{\beta \cos\theta} \qquad \text{Eq. 1}$$

3. Results and Discussion

Tests were conducted on each MS-SOFC layers in this research to explain its chemical and physical analysis. First by analysing using XRD machine to identify the presence of the desired phase in the composite powders that were used in the production of screen-printing inks. Then, SEM and EDS were used to observe the morphology and existence of elements of each MS-SOFC layer (cathode, anode, and electrolyte) after the sintering process.

3.1 Phase Identification for Composite Powder

The process of calcination using a furnace was carried out at a temperature 950℃ for 2 hours. 950℃ is a suitable calcination temperature in this research when a good XRD pattern has been obtained as shown in Figure 1 because there are no secondary peaks observed in it. Each peak resulting in NiO-GDC composite powder consists of a single phase for each commercial powder. According to Park, et al., (2014) the NiO and GDC phases can be maintained if the calcination process is carried out at temperature 800℃ to 1100℃. The increment and decrement of the peak intensity and width indicate the particle shrinkage because of heat treatment [3]. The JCPDS numbers for NiO and GDC are 00-047-1049 and 01-075-0161 respectively. This NiO-GDC powder has the same lattice structure which is face-centred cubic. The structure of this lattice can be determined by looking at the value of axial length and angle on the resulting XRD data which has the same axial length value 4.17710 (NiO), 5.41800 (GDC) and the same angle of 90°. The crystallite size produced in the NiO-GDC composite anode powder is 24.59 nm for NiO and 24.38 nm for GDC.

The peak of the GDC powder shows a higher intensity than the peak of the LSCF powder in the XRD pattern of the LSCF-GDC composite powder (Figure 2). According to Jamale & Bhosale (2016), the GDC peak will be higher due to the reflection of GDC particles in the composite powder LSCF-GDC. In addition, nanocrystalline at GDC has a high surface area and sinterability, which results in significant grain growth [4]. Each peak produced in this research on the LSCF-GDC composite powder consisted of only a single peak for each commercial powder. The JCPDS pattern number for LSCF is 01-089-5720 while for $Gd_{0.1}Ce_{0.9}O_{1.95}$ GDC is 01-075-0161. Each has the same lattice structure which is face-centred cubic where GDC powder has a longer axial length than LSCF powder which is 5.41800 compared to LSCF which is 3.87380. However, these two forms the same cubic lattice structure because they have the same axial length and angle at 90°. On average, the crystallite size produced in this LSCF-GDC composite powder is 13.34 nm for LSCF and 24.38 nm for GDC.

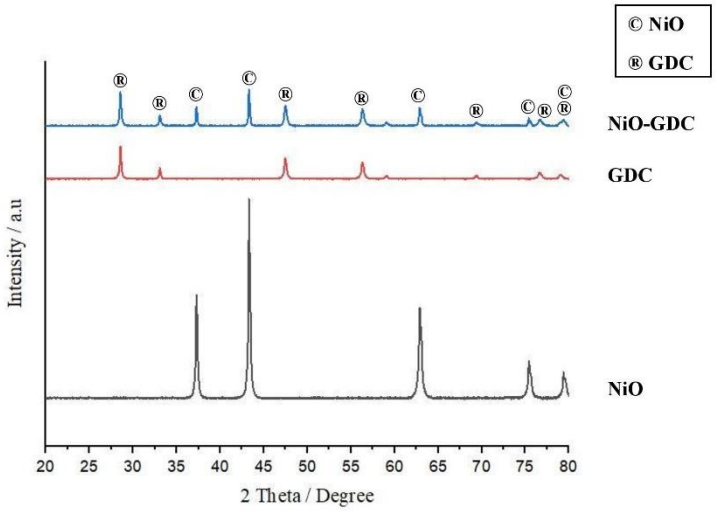


Figure 1: XRD Pattern of Composite Powder Anode NiO-GDC

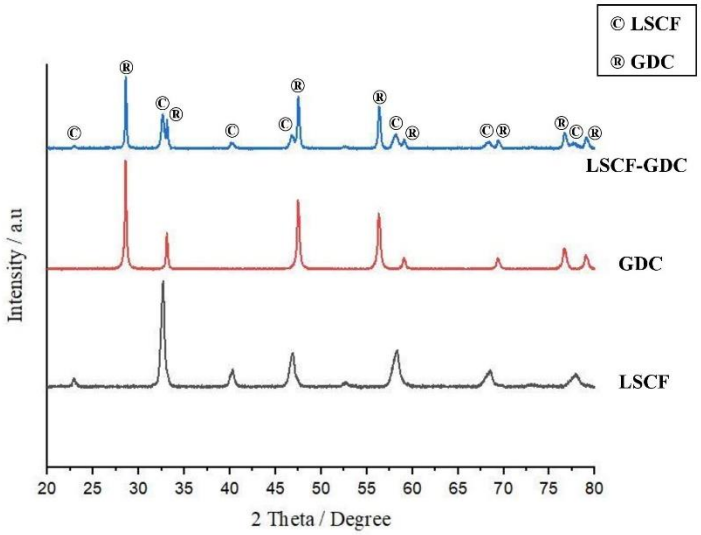


Figure 2: XRD Pattern of Composite Powder Cathode LSCF-GDC

3.2 Morphology of Thickness for Each MS-SOFC Layer

In this research, the ink produced for electrode NiO-GDC should have smaller physical properties than ink for electrode LSCF-GDC. The use of mesh screen 305 should be able to produce an ideal electrode and electrolyte for MS-SOFC compared to mesh screen 355. Table 4 shows the average thickness of the screen-printing layer. The results of the SEM test on all MS-SOFC samples show that the use of mesh screen 305 can produce more ideal electrode and electrolyte layers than mesh screen 355. According to Ning, et al., (2012), by controlling and reducing the thickness of the screen-printing layer will produce low polarization resistance and can improve SOFC performance [5]. Therefore, the 305-mesh screen at 10 times the number of printings is the best sample for MS-SOFC. This is because, the thickness of the MS-SOFC layer obtained is lower than other MS-SOFC samples which are 11.8 µm, 11.9 µm and 18.2 µm for anode, electrolyte, and cathode respectively (Table 5). However, according to Somalu, et al., (2011), the layer thickness resulting from this study is still in the proposed layer thickness range of 10µm to 200µm [6]. The use of different mesh screens significantly affects the thickness of SOFC component layer.

Table 4: Thickness of Screen-Printing Layer Via SEM Testing

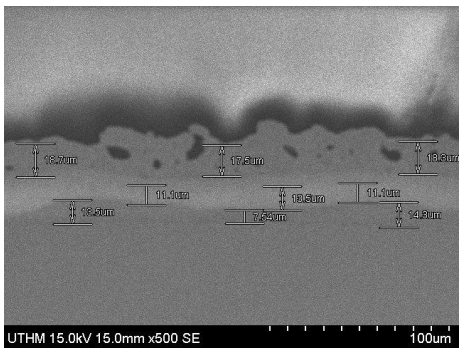
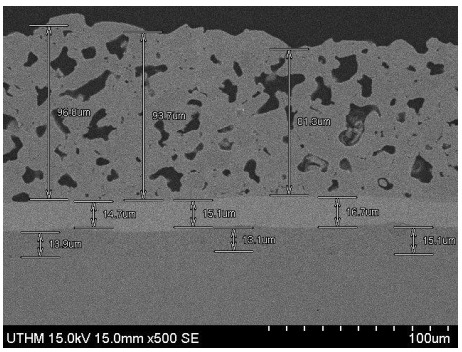
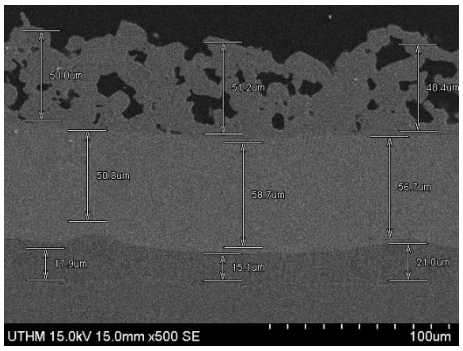
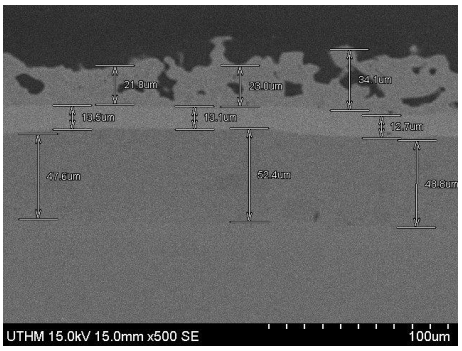
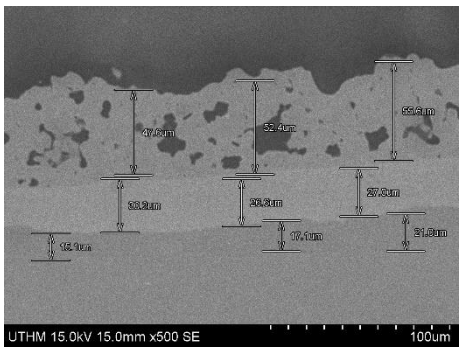
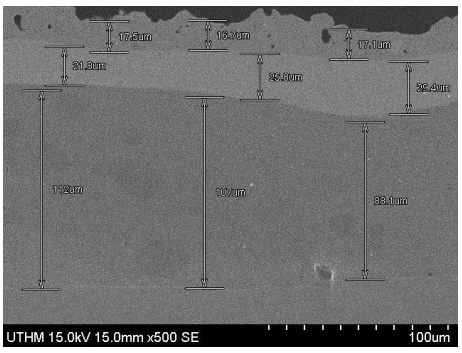
Number of Printing	Mesh Screen	
	305	355
10 times		
15 times		
20 times		

Table 5: Average Thickness of Screen-Printing Layer

Number of Printing	Mesh Screen	Layer Thickness (µm)		
		NiO-GDC	GDC	LSCF-GDC
10 times	305	13.9	11.9	18.2
	355	14.0	15.5	90.6
15 times	305	18.0	55.4	49.9
	355	49.6	13.1	26.3
20 times	305	17.7	27.9	51.9
	355	102.4	25.7	17.1

3.3 Elements Distribution for Each MS-SOFC Layer

Element distribution analysis is important to determine whether there is a mixture between each of these layers after the given treatment. Basically, the NiO-GDC layer will contain elements such as C, O, Ni, Ce and Gd while the LSCF-GDC layer is an element such as C, O, Fe, Co, Sr, La, Ce, Gd. Only elements such as C, O, Ce, Gd will be contained in the GDC electrolyte layer. The weight percentage for each element including elements such as Ni, La, Sr, Co, and Fe is also obtained. In this research, nanoparticle size for Ni smaller than LSCF will affect the percentage of weight obtained for Ni element that using 355 mesh screens at 10, 15 and 20 time number of prints. Ni element for 355 mesh screens will obtain a higher percentage of weight compared to Ni element that using 305 mesh screens. According to Moor (2004), there is a relationship between the screen-printing ink and the mesh screen used. He also stressed that there will be almost no ink that can be printed through the mesh screen if the use of thicker ink on the higher mesh screen [7]. The larger nanoparticle size for LSCF will reduce the percentage weight of the element in each MS-SOFC layer that using a 355-mesh screen at 10, 15, and 20 times the number of screen prints. Table 6 shows the weight percentage of element distribution for Ni and LSCF.

Table 6: Element Distribution for Ni, La, Sr, Co, Fe

Number of Printing	Mesh Screen	Weight (%)	
		Ni	La, Sr, Co, Fe
10 times	305	29.09	17.19
	355	32.23	21.21
15 times	305	27.82	17.36
	355	35.67	19.83
20 times	305	35.78	18.52
	355	32.75	20.87

4. Conclusion

In this study, fabrication of MS-SOFC multilayer has been obtained for anode, electrolyte, and cathode without defect after the sintering process. For all the samples produced, the anode and electrolyte electrode layers were successfully fabricated according to the suitability of the desired layer. A good XRD pattern was obtained without the presence of a secondary peaks. Cubic's lattice structure and average crystallite size are obtained from XRD data analysis. 24.59 nm, 24.38 nm, 13.34 nm are the average crystallise size for NiO, GDC and LSCF, respectively. Based on the thickness and distribution analysis of elements in each MS-SOFC layer, layers fabricated by using the 305-mesh screen at 10 times number of prints is the best MS-SOFC in this research. This is because, the obtained layers have lower thickness compared to layers from mesh screens 305 and 355 at 15 and 20 times number of prints which are 11.8 μm, 11.9 μm and 18.2 μm for the anode, electrolyte and cathode respectively. SOFC component layers with low thickness is expected to produce low polarization resistance and improve SOFC performance. In conclusion, the objectives for this study have been achieved, however, there are some improvements that can be made to get better results in future studies.

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