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# A Study Review On Spinel as Interconnect Coating for Solid Oxide Fuel Cell (Sofc)

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Abstract: A solid oxide fuel cell (SOFC) is a green and reliable alternative energy source that under development which generates electricity. In order to understand the spinel as interconnect coating of Solid Oxide Fuel Cell (SOFC), a review has been conducted. The review was about understanding on how Mn-Co spinel coating that has been successfully fabricated, so that the technique of Electrophoretic Deposition (EPD) was used to produce the desired coating effectively. The (Mn,Co)<sub>3</sub>O<sub>4</sub> spinel coatings on the surface of Cr-containing steel through electrophoretic deposition (EPD) are followed by reduced-atmosphere sintering for solid oxide fuel cell (SOFC) interconnect application. The review was about the effects of EPD voltages and sintering atmospheres on the microstructure, electrical conductivity and long-term stability of the coated interconnects are examined by means of scanning electron microscopy (SEM), energy dispersion spectrometry (EDS) and Electro Impedance Spectroscopy (EIS). SOFC performance with different voltages and coating durations parameter are analysed. Therefore, the EPD technique has successfully provided that better coating thickness of spinel as interconnect for SOFC.

Keywords: SOFC, Spinel, Interconnect, Coating, EPD, XRD, SEM, EDS, ASR

## 1. Introduction

A solid oxide fuel cell (SOFC) is a green and reliable alternative energy source that under development which generates electricity through an ionic conducting oxide electrolyte by electrochemically combining a fuel and an oxidant [1]. SOFC comprises of the anode, cathode, and electrolyte. It is operated in the form of an interconnected array to enhance the power output. [2]. Interconnects or also known as substrate typically deliver electrical conduction between single cells and act as a physical barrier to separate anode fuel from cathode air.

Over the recent years, efforts have been concentrated mainly on the compound ceramic oxides family with crystalline structure. Only a few such oxide systems can meet the strict specifications for substrate materials in SOFCs [3]. The advancement of the (IT-SOFC), an intermediate temperature solid oxide fuel cell, has led to a reduction in the ambient temperature of the fuel cell from 1000 °C to 800 °C. In the high temperature period of SOFC (1000°C), the only efficient interconnects were made of electrically conductive ceramic materials. Due to their complex geometries, these interconnects were very complicated to manufactured as a result the cost are very high.

### 2. Methodology

This chapter explains all the working procedure for this project. In establishing a comprehensive overview, selection of journal articles that focused on the parameter of spinel coating substrate were systematically analyses. Data base search was performed on two well established data bases; ScienceDirect and ResearchGate. Keywords such as "solid oxide fuel cell (SOFC)", "spinel\*", "interconnect" and "substrate" were included in the search options and the scope was restricted to academic papers in English, including recent publications from year 2001 to 2020.

#### 2.1 Methods

In the first step, the terms "solid oxide fuel cell (SOFC)", "spinel\*", "interconnect" and "substrate" were being searched. In the second step, the title, abstract and keywords were manually checked, and selections were refined by removing all non-related papers. In the third step, the related papers were searched through exploration of references and citations of already selected papers. In the fourth step, in covering all experiments and analysis regarding interconnect coating with various experiment, the searched paper was included with terms such as "Electrophoretic Deposition", "Xray Diffraction Analysis (XRD)", "Scanning Electron Microscopic (SEM)" and "Electro Impedance Spectroscopy (EIS)". Lastly, a final refinement was made predicated on full texts and sorting of all papers were also predicated on full texts.

#### 3. Results and Discussion

#### 3.1 Characterization Analysis Review of Coating Interconnect for SOFC.

The following analysis has been reviewed from the previous research studies to determine the characterization of manganese cobalt powder for solid oxide fuel cell.

3.3.1 Phase Analysis Review



Figure 1: The Changed of XRD pattern of Co-33Mn-17Cu [4]



Figure 2: XRD Configuration of MnCu<sub>0.5</sub>Co<sub>1.5</sub>O<sub>4</sub> coated with SUS430 alloy [5]



Figure 3: : XRD patterns collected from Mn1.5Co1.5O4 coated Crofer22APU samples sintered [6]

The first study involved is the uses of SS430 as substrate whereas Co-38M-2La and Co-33Mn-17Cu alloy as a coating material. Based on the analysis review, 110 V voltage and 1.2 min time for deposition was set up as the parameter for the experiment. The XRD pattern for alloy coating of Co-38Mn-2La and Co-33Mn-17Cu was shown in Figure 1. The figure depicts the primarily Cu characteristic peak prior to oxidation was observed for Co-33Mn-17Cu alloy coating. For Co-38Mn-2La coating, the tetragonal (MnCo)<sub>3</sub>O<sub>4</sub> spinel diffraction peak is intensified after it oxidised.

Next, for the second reference of analysis review, using  $MnCu_{0.5}Co_{1.5}O_4$  and SUS430 as a coated. Figure 2 illustrates the XRD spectrum of the SUS430 as the coating sample after cyclic oxidation in air at 750 °C for 1000 hours. It can be observed that the main phase in the oxide scale in all samples is  $MnCu_{0.5}Co_{1.5}O_4$ , and a weak diffraction peak from the matrix is also detected. At the same time, due to the thick coating, no diffraction peaks of  $Cr_2O_3$  can be seen. However, as the oxidation time increases, a slight shift of the diffraction peak of  $MnCu_{0.5}Co_{1.5}O_4$  to a lower angle can be observed, which may be anticipated to the dispersion of Fe or Mn from the interconnect to the spinel coating. [7]

For the third reference, (Smeacetto et al. 2015) was using  $Mn_{1.5}Co_{1.5}O_4$  powder deposited on Crofer22APU by EPD at 50 V, and the deposition time is 20 seconds. Related to the  $Mn_{1.5}Co_{1.5}O_4$ 

spinel reference powder, no phase change was observed in the EPD deposited coatings sintered in air at 800 °C for 2 hours; they all show that they correspond to  $MnCo_2O_4$  and XRD peak of Mn2CoO4 phase. As reported by other authors, spinel with a nominal composition of Mn1.5Co1.5O4 is known to have a two-phase structure, including cubic  $MnCo_2O_4$  and tetragonal ( $MnCo_2O_4$ ) phases like shown in Figure 3.

According to the results of previous studies, most of the detected phases consist of two the types of  $(Mn,Co)_3O_4$  spinel phases are cubic and tetragonal. XRD pattern. When the parameter is increases, it also shows the increase of  $(Mn,Co)_3O_4$  spinel strength. This increase means a better coating microstructure to prevent poisoning. Therefore, it indicates that spinel is suitable as a coating of steel interconnect for SOFC applications.

3.3.2 Surface Morphology Review



Figure 4: Surface Morphologies of the As-Deposited (Cu,Mn,Co)<sub>3</sub>O and After Reduction Treatment. [10]



Figure 5: SEM Image of As Deposited [8]



Figure 6: Spinel Coating Thickness [9]

The first study referred to the use of SS430 as the substrate. Figure 4.4 shows the as-deposited  $(Cu,Mn,Co)_3O_4$  and the surface morphology after reduction treatment. The rapid growth of the oxide scale is very thick, which prevents the normal deposition of  $(Cu,Mn,Co)_3O_4$  spinel from adhering to the SUS430 steel substrate; in addition, the coating will peel off after densification. SEM micrographs are used to analyse the porosity of the  $(Cu,Mn,Co)_3O_4$  spinel layer at different stages. The porosity of the as-deposited  $(Cu,Mn,Co)_3O_4$  spinel (17.7% ± 4.22%) increases to 18.8% ± 3.76% after reduction, which corresponds to an increase in vacancy condensation caused by released oxygen. [10]

For the second reference,  $Cu_{1.3}Mn_{1.7}O_4$  spinel coated with Crofer 22APU studied about densification. Figure 4.5. shows the SEM image of the cross-section of the as deposited. The coating is uniform and has good adhesion, with a thickness of about ~26 µm. Although the particles are dense, the as-deposited coating has many small pores, so further densification is required. Reduction and reoxidation are effective strategies for coating densification. It was found that the  $Cu_{1.3}Mn_{1.7}O_4$  spinel coating was reduced to Cu and MnO phases after annealing in a forming gas of 850 °C for 1 hour. [8]

The third research study from use Crofer 22 APU as the substrate and CeO2-doped  $(Co,Mn)_3O_4$  as coating material. Coating thickness before the oxidation process is equal to 4 µm. After oxidized at 8000 °C for 250 h, the average result for coating thickness varies from 5.5 µm to 5.7 µm. The analysis of the SEM image shown in Figure 4.6 shows that a dense and uniform Mn1.5Co1.5O4 spinel coating with a thickness of about  $5.8 \pm 0.3$  µm is formed before oxidation. In addition, it is found that the thickness of the Cr2O3 oxide scale is about  $3.5 \pm 0.3$  µm between the Mn1.5Co1.5O4 spinel coating and Crofer 22 APU substrate. [11]

The research found that the particles size below  $1\mu m$  able to give the improvement towards surface area for absorption process in ceramic body. So that, LSCF-SDCc with 5wt% Ag has met the criteria of particle size for composite cathode in SOFC. Based on previous studies that have been reviewed, the coating is effectively deposited on the surface of the substrate. the thicker the coating thickness it will make the good protective layer for the EPD process. The coating shows a fine coating structure with the substrate, which is exactly what this study expected. [12]

#### 4. Conclusion

This research is concentrated on previous studies to find out the characterization of the interconnects by varying the applied voltages from 30 V to 60 V and the deposition time from 20 to 60 seconds. The most effective spinel coating parameter for Solid Oxide Fuel Cell (SOFC) have been investigated. The objective of the study was to review the characterization of SOFC using spinel coated substrate with various voltage and coating duration parameter. All methods reviewed in this study used The EPD as a method for coating spinel coatings on interconnects with various voltage and duration. According to the previous analysis, the analysis was phase analysis, morphology analysis, elemental analysis, and weight deposition analysis. For phase analysis, the coating layer on substrate coating microstructure indicate the good spinel for interconnect coating. Then, for morphology analysis it determined the surface morphology for each coating that have the different quality of deposition regarding the influenced of applied voltage and duration. Hence, when the parameters increase, the coating layer becomes denser and uniform. Moreover, for elemental analysis, the coating material used have a base of Mn and Co and related to the diffusion of Cr. that smaller concentration than 5 g/l will decrease the deposition rate.

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#### References

- I. Aznam, J. C. W. Mah, A. Muchtar, M. R. Somalu, and M. J. Ghazali, "Electrophoretic deposition of (Cu,Mn,Co) 3 O 4 spinel coating on SUS430 ferritic stainless steel: Process and performance evaluation for solid oxide fuel cell interconnect applications," *J. Eur. Ceram. Soc.*, vol. 41, pp. 1360–1373, 2021.
- [2] M. A. Hassan, O. Bin Mamat, and M. Mehdi, "Review: Influence of alloy addition and spinel coatings on Cr-based metallic interconnects of solid oxide fuel cells," *International Journal of Hydrogen Energy*, vol. 45, no. 46. Elsevier Ltd, pp. 25191– 25209, Sep. 21, 2020.
- [3] M. A. S. Pingwei, Spinel Coatings For Solid Oxide Fuel Cell Interconnects And Crystal Structure Of Cu-Mn-0 Title: Spinel Coatings For Solid Oxide Fuel Cell Interconnects And Crystal Structure Of Cu-Mn-0. 2009.
- [4] P. Guo, Y. Lai, Y. Shao, Y. Zhang, and Y. Wang, "metals Thermal Growth Cu 1.2 Mn 1.8 O 4 Spinel Coatings on Metal Interconnects for Solid Oxide Fuel Cell Applications."
- [5] J. Xiao, W. Zhang, C. Xiong, B. Chi, J. Pu, and L. Jian, "Oxidation behavior of Cudoped MnCo 2 O 4 spinel coating on ferritic stainless steels for solid oxide fuel cell interconnects," 2016.
- [6] F. Smeacetto *et al.*, "Electrophoretic deposition of Mn1.5Co1.5O4 on metallic interconnect and interaction with glass-ceramic sealant for solid oxide fuel cells

application," J. Power Sources, vol. 280, pp. 379-386, Apr. 2015.

- [7] J. Xiao, W. Zhang, C. Xiong, B. Chi, J. Pu, and L. Jian, "Oxidation of MnCu0.5Co1.5O4 spinel coated SUS430 alloy interconnect in anode and cathode atmospheres for intermediate temperature solid oxide fuel cell," *Int. J. Hydrogen Energy*, vol. 40, no. 4, pp. 1868–1876, Jan. 2015.
- [8] T. University, "Electrophoretically Deposited Copper Manganese Spinel Protective Coatings On Metallic Interconnects For Prevention Of Cr-Poisoning In Solid Oxide Fuel Cells," 2011.
- [9] Y. Zhang, A. Javed, M. Zhou, S. Liang, and P. Xiao, "Fabrication of Mn-Co spinel coatings on Crofer 22 APU stainless steel by electrophoretic deposition for interconnect applications in solid oxide fuel cells," *Int. J. Appl. Ceram. Technol.*, vol. 11, no. 2, pp. 332–341, Mar. 2014,.
- [10] I. Aznam, J. C. W. Mah, A. Muchtar, M. R. Somalu, and M. J. Ghazali, "Electrophoretic deposition of (Cu,Mn,Co)3O4 spinel coating on SUS430 ferritic stainless steel: Process and performance evaluation for solid oxide fuel cell interconnect applications," *J. Eur. Ceram. Soc.*, vol. 41, no. 2, pp. 1360–1373, 2021, doi: 10.1016/j.jeurceramsoc.2020.09.074.
- [11] H. Zhang, Z. Zhan, and X. Liu, "Electrophoretic deposition of (Mn,Co)3O4 spinel coating for solid oxide fuel cell interconnects," *J. Power Sources*, vol. 196, no. 19, pp. 8041–8047, Oct. 2011.
- [12] J. W. Fergus, "Effect of cathode and electrolyte transport properties on chromium poisoning in solid oxide fuel cells," *Int. J. Hydrogen Energy*, vol. 32, no. 16, pp. 3664– 3671, Nov. 2007,