



# Characteristics of Ag Element on LSCF-SDCC-Ag Composite Cathode for LT-SOFC Potential Applications

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DOI: <https://doi.org/10.30880/jamea.2020.01.01.003>

Received 25th October 2020; Accepted 10th February 2020; Available online 29th February 2020

**Abstract:** A composite cathode of Lanthanum Strontium Cobalt Ferrite-Samarium Doped Ceria Carbonate-Argentum (LSCF-SDCC-Ag) for low-temperature SOFC (LT-SOFC) potential applications was investigated in this study. This study focuses on the influence of Ag as an additive element in the improvement of microstructural and the improvement of LSCF-SDCC-Ag composite cathode powder properties. Composite cathode powders LSCF-SDCC-Ag with 1-5 wt.% Ag content were prepared using high energy ball milling (HEBM) technique and underwent sintering process at 500 °C. The prepared samples were characterized using X-ray diffraction (XRD), Fourier Transform Infrared (FTIR) and Energy Dispersive Spectroscopy (EDS). The XRD analysis of LSCF-SDCC-Ag composite cathode powder exhibited good chemical compatibility, as there were no secondary constituent detected via XRD sensitivity. The carbonate phase in LSCF-SDCC-Ag was accomplished to preserve their phase as proven via FTIR analysis. Meanwhile, the quantitative elements for the LSCF-SDCC-Ag composite cathode powder were identified using EDS and from HEBM technique, the cathode successfully achieved the nano-composite state powder characteristic. Hence, from XRD, FTIR and EDS analyses, it can be concluded that the LSCF, SDCC and Ag are compatible with each other to form the composite cathode of LSCF-SDCC-Ag for LT-SOFC applications.

**Keywords:** HEBM, SDCC composite cathode, SOFC, LSCF

## 1. Introduction

The world energy demand is growing at a rate of 1.8 % per year, which is mostly due to the increment of industrialization from the developing countries. The high demands are also associated with the conservation of fossil fuel which leads to the emissions of greenhouse gases and other pollutants. Fuel cells are one of the devices that directly convert fuel to electricity. Solid oxide fuel cells (SOFC) has successfully generated power with sixty percent of efficiencies of output power are common. Previous researches have been conducted to lower the SOFC's operating temperature to improve their life expectancy and reliability [1]. These researches have been focusing on the development of intermediate-temperature (IT) to low-temperature (LT) SOFC ranging from 400 to 600 °C. Consequently, the reduction of operating temperature resulted in significant effects on the inefficiency of the cathode elements. The increase of cathode polarization resistance substantially reduces the electrochemical performance [2]. Previously, research studies reported that the design of a high-performance electrode with a low polarization resistance such as composite cathode has the potential to solve the said problem [1,3].

Recently, the addition of electrolyte material to cathode material resulted in the increased of the triple-phase boundary (TPB) (air/electrode/electrolyte) area, where the electrochemical reactions occurred. A mixed ionic and electronic conductors (MIECs), such as  $\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_{3-\delta}$  (LSCF), is one of the potential candidates that has been utilized in this approach since LSCF has been recognized as one of the promising cathode material for SOFC [4-5]. MIEC

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could enhance the area of TPB and increase the performance of the SOFC. Moreover, an innovative solid carbonate-oxide composite known as samarium doped ceria carbonate (SDCC), which performed well at low temperatures of 400-600 °C, was discovered [6]. In previous research, incorporation of LSCF with SDC carbonates electrolytes enables the production of the superior performance of potential cathode from intermediate- to low-temperature solid oxide fuel cell [7-8]. The use of binary carbonate ((Li/Na)<sub>2</sub>CO<sub>3</sub>) together with SDC to improve the ionic conductivity embarked a new path of research in which the LSCF-SDCC composite cathode is deemed able to perform well at low temperatures of 400-600 °C [9-10]. The combination of LSCF with SDCC has been proven able to yield outstanding properties as composite cathode materials for IT-LTSOFC [11]. The incorporation of LSCF with SDCC is expected to achieve outstanding properties as composite cathode materials for IT-LTSOFC.

In order to develop a new composite cathode material, dissimilar research reported that silver (Ag) is a great potential to be used as an additive element to be mixed with LSCF-SDCC [12]. The mixing is believed able to enhance the conductivity to overcome the polarization resistance of the composite cathode during the low-temperature operation of SOFC [13]. The silver metal was chosen for the LSCF-SDCC composite cathode in LTSOFC with operating temperature below 800 °C. This property is suitable for its good catalytic activity, high electrical conductivity, and relatively low cost. Furthermore, the use of this metal also improves cathode microstructure since the melting point of Ag is 961 °C, which will aid the sintering behavior at cathode [14-15]. This study focuses on the investigation of the LSCF-SDCC-Ag composite cathode to suit the low-temperature application (400-600 °C) of solid oxide fuel cell applications. This research is also designed to investigate the effect of Ag as an additive element in LSCF-SDCC composite cathode materials, as well as to identify its chemical, structural and elemental distribution properties.

## 2. Materials and Method

The raw materials used were the commercial LSCF powder (LSCF6428; Kceracell, Korea) and self-fabricated SDC (Kceracell, Korea) with carbonate powder. The composite electrolytes of SDCC, SDC, Li<sub>2</sub>CO<sub>3</sub>, and Na<sub>2</sub>CO<sub>3</sub> (Sigma Aldrich, USA) powders were ball-milled for 24 hours and dried for 16 hours in the oven, heat-treated at 680 °C. The commercial LSCF powder was then mixed with SDCC in the ratio of 50:50 weight percentage (wt.%) using wet milling techniques, and the mixture was ball-milled with ethanol as the solvent media. The resultant powder obtained was then grounded using agate mortar and calcined in a furnace with a temperature of 750 °C. Ag powder was added via dry mixing with implementation of high energy ball milling (HEBM) method with the addition of 1, 2, 3, 4, and 5 wt.%. The resultant composite cathode powder was then pressed to produce pellets and underwent sintering process at 500 °C. XRD (D8 Advance, Bruker, Germany) was conducted between a range of 2θ from 10° to 80°, to analyze the pellet phase purity and compatibility of the LSCF-SDCC-Ag composite before and after sintering. Cu-Kα was employed as the radiation source, and *Eva Diffrac Plus* (Version 4) software was used to analyze the XRD raw data. Meanwhile, to observe the chemical structural, and the existence of carbonate, Fourier Transform Infrared (FTIR) analysis was carried out based on the Attenuated Total Reflection (ATR) method. FTIR testing was performed in wavenumber ranging from 4000 to 500 cm<sup>-1</sup> (Spectrum 100, Perkin Elmer, USA). Finally, the elemental distribution in LSCF-SDCC-Ag powders were analyzed by employing EDS testing (SEM-EDS, JSM 6380LA-JEOL, Japan).

## 3. Results and Discussion

Fig. 1 illustrates the XRD spectra of LSCF-SDCC with and without Ag addition (1-5 wt.%) which have been sintered at 500 °C. As previously reported [14], the ceria cubic fluorite and perovskite-type structures of SDC (JCPDS No. 75-0157) and LSCF6428 (JCPDS No. 89-5720) were identified respectively from the XRD analysis. Based on the observation attained in this research, the depicted spectra for the incorporation of Ag from 1 to 5 wt.%, did not distress or alter the chemical compatibility of the LSCF-SDCC-Ag composite cathode. Remarkably, there is no additional peak or phase emerged after the composite cathode endures the sintering process at 500 °C. Multiple peaks which correspond to Ag metal retain noticeably for all different contents of Ag on LSCF-SDCC-Ag composite cathode. Composite cathode with the addition with 5 wt.% of Ag recorded the highest peak of crystallinity compared to the peak of the composite cathode with the addition of only 1 wt.% Ag. From this observation, it can be perceived that the chemical and structural compatibility of LSCF-SDCC-Ag composite cathode improves with the increasing of Ag content [16]. However, LSCF-SDCC-Ag composite cathode recorded lower crystallinity compared to the spectra collected from LSCF-SDCC without the addition of Ag content. There was no impurities peak formed in this composite cathode. This phenomenal ascribed that LSCF and SDCC have successfully retained their structures with the incorporation of Ag [14]. Furthermore, the addition of Ag strongly appointed LSCF-SDCC-Ag composite cathode in good chemical and structural compatibility to be used as a composite cathode for SOFC [11].

Figures 2(a) and 2(b) display the FTIR spectrum of LSCF-SDCC and LSCF-SDCC-Ag composite cathode powder respectively. The spectrum in Fig.2(a) exhibits the expected carbonate of CO<sub>3</sub><sup>2-</sup> bands from LSCF-SDCC composite cathode at wavenumbers of 858 and 1411 cm<sup>-1</sup>. The existence of carbonate phases (Li/Na)<sub>2</sub>CO<sub>3</sub> on LSCF-SDCC-Ag

composite cathode with different amount of Ag addition was retained as depicted in Fig. (b). The presence of carbonate phases on LSCF-SDCC-Ag is very crucial to preserve carbonate elements in the composite powder, hence underwent calcinations at 750 °C. From the observation, it is notable that the carbonate from SDCC functioned in an excellent manner as co-ionic conductive, to enhance the electrochemical performance of LSCF-SDCC-Ag composite cathode [17,18].

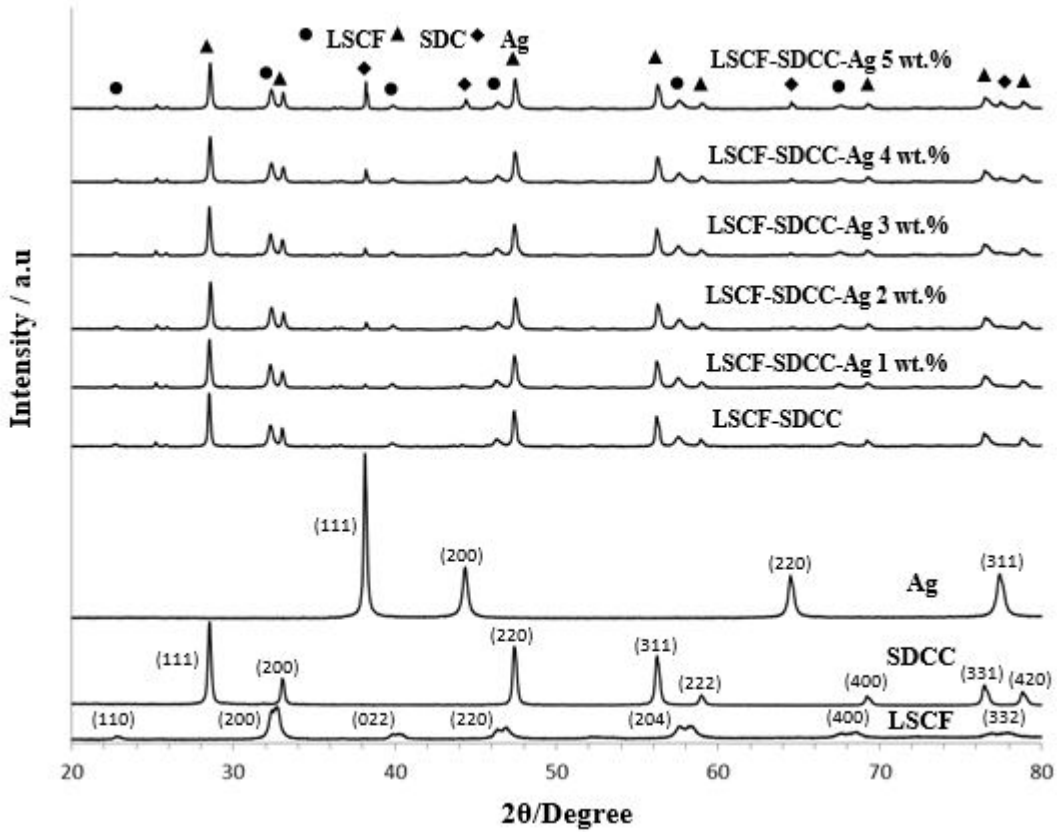


Fig. 1 - XRD spectra of pure LSCF powder, SDCC electrolyte powder, pure Ag powder and LSCF-SDCC-Ag (1-5 wt.%) with sintering temperature of 500 °C

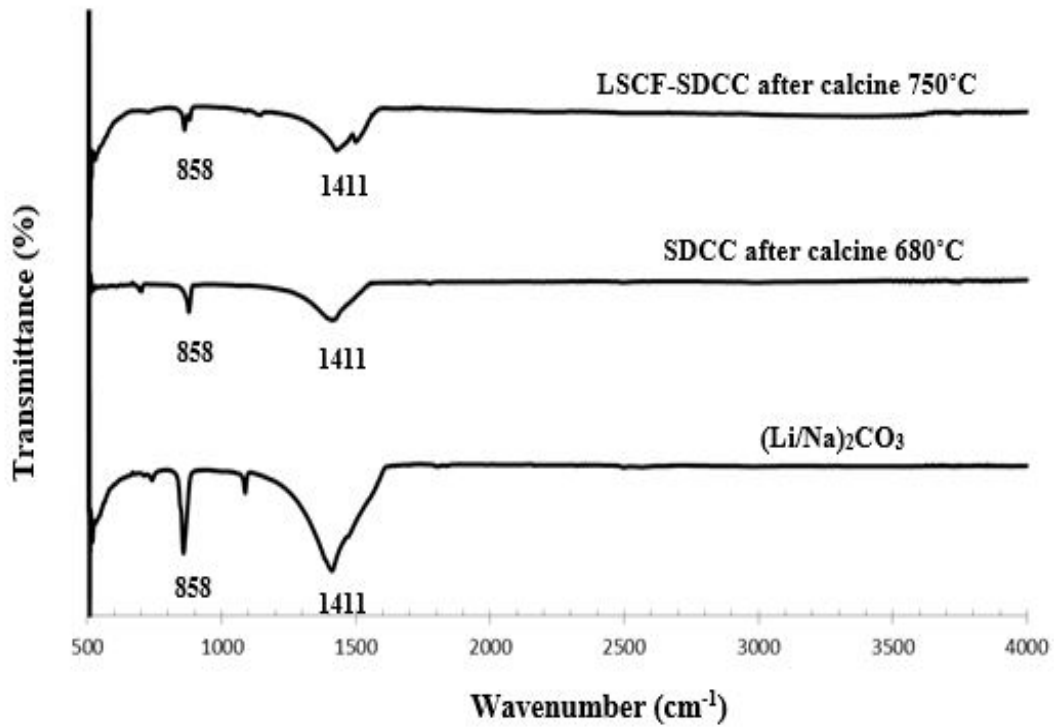


Fig. 2 (a) - FTIR spectra for each carbonate (Li/Na)<sub>2</sub>CO<sub>3</sub>, SDC carbonate (SDCC) and LSCF-SDCC-Ag composite cathode with 1 wt.% of Ag content

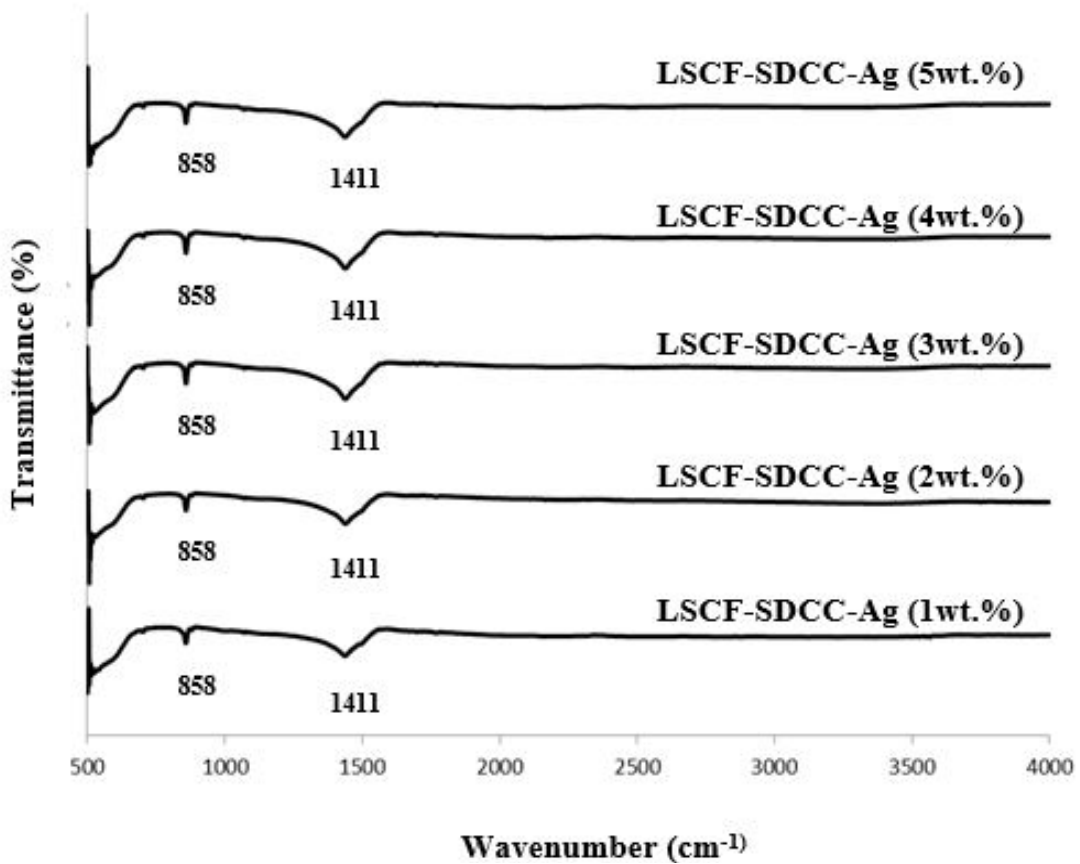
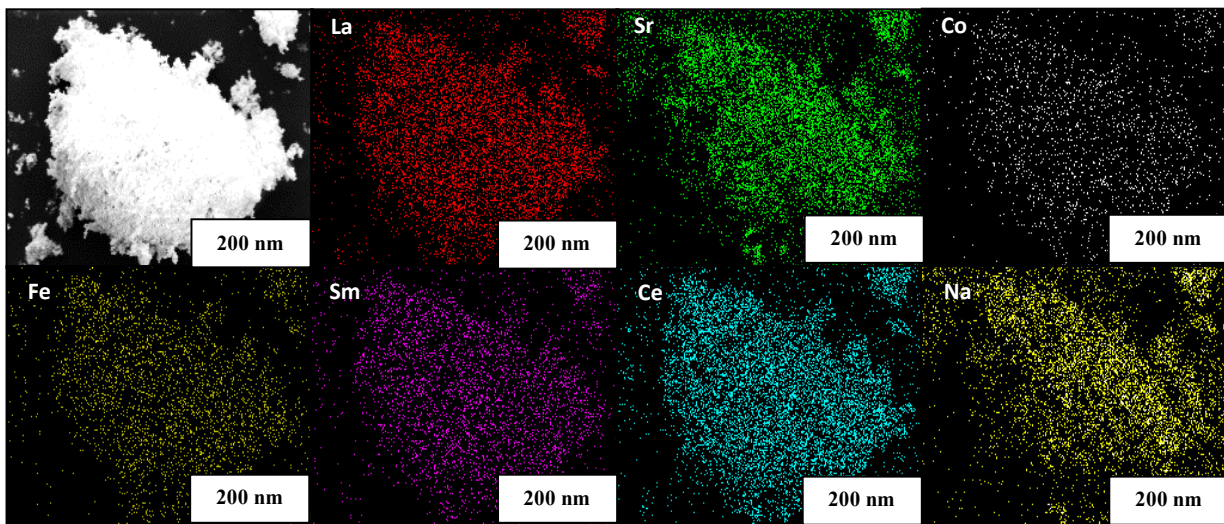
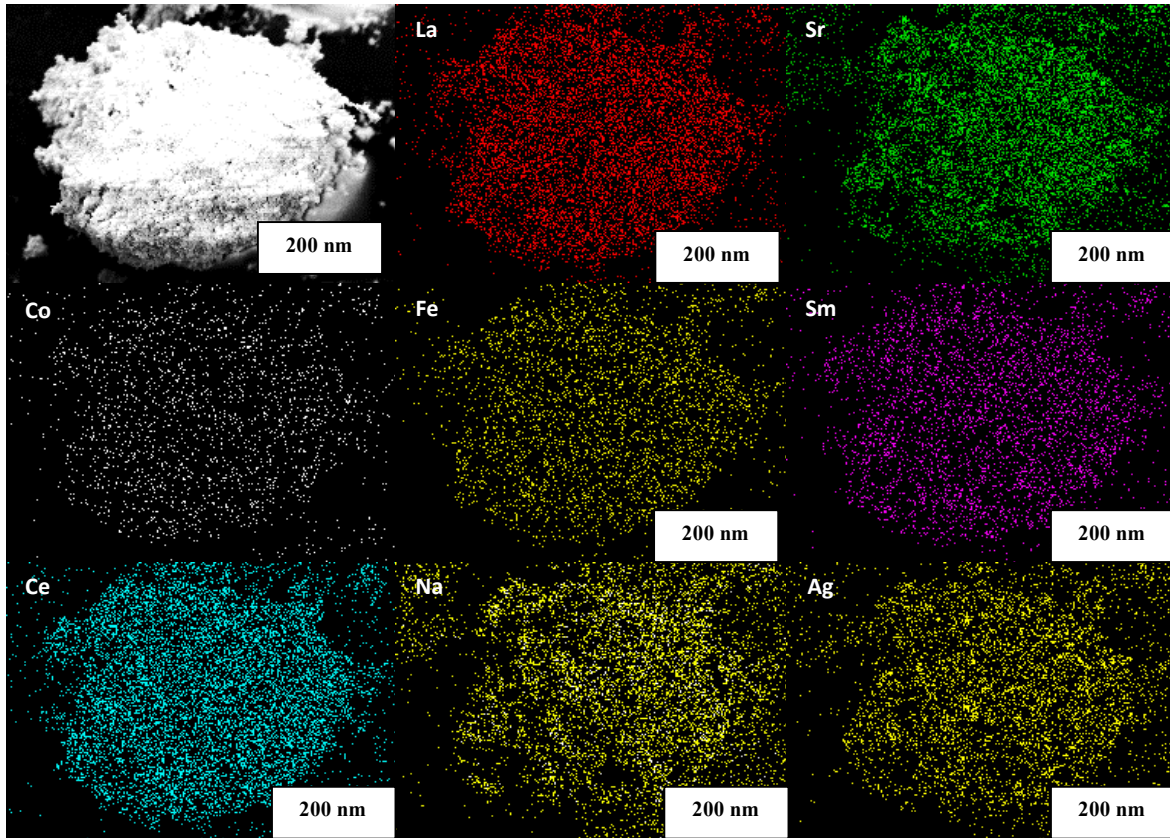


Fig. 2 (b) - FTIR spectra for each of LSCF-SDCC-Ag composite cathode with 1-5 wt.% of Ag content

Element distribution has been recognized as an essential key with a substantial contribution towards the electrochemical performance of composite cathode. Fig.3 and Fig.4 show the image from EDS mapping analysis on LSCF-SDCC and LSCF-SDCC-Ag (3 wt.%), respectively, which significantly evaluate the quantitative of elemental distribution. All elements which have been attributed to LSCF-SDCC and LSCF-SDCC-Ag such as Lanthanum (La), Strontium (Sr), Cobalt (Co), Ferrite (Fe), Samarium (Sm), Cerium (Ce), Argentum (Ag) and Sodium (Na) has been detected via EDS micrograph analysis. The introduction of Ag content into LSCF-SDCC has an unfavorable impact on the composite cathode powder distributions characteristics, due to an absence of any foreign or impurities element from the available image. From the observation, ascribed Ag as an additive element has been promoting on the better powder distribution with all the elements remain detectable and clearly perceived after the addition of 3 wt.% Ag content. Unfortunately, lithium (Li) elements accredited from the composite cathode of LSCF-SDCC and LSCF-SDCC-Ag were not discovered by the EDS analysis, due to low atomic mass property of lithium element [5]. The result discussed earlier has demonstrated that all the critical elements in LSCF-SDCC-Ag composite cathode powder have been homogeneously mixed and well-distributed powder has successfully achieved via HEBM method. The LSCF-SDCC-Ag composite cathode distribution is observed in a proper manner without the appearance of any unwanted or impurities elements [18]. The homogeneity of elements is essential as each element contributes to the electrochemical reactions occurred in the cathode. HEBM method in this research has successfully produced the homogeneously dispersed powders of LSCF-SDCC-Ag composite cathode [5, 11].



**Fig. 3 - Mapping and spectrum EDS illustrates elements distribution in LSCF-SDCC composite cathode**



**Fig. 4 - Mapping and spectrum EDS illustrates elements distribution in LSCF-SDCC-Ag (3 wt.%) composite cathode**

#### 4. Conclusion

The objective of this study is to prepare and characterize new composite cathode powder of LSCF-SDCC-Ag with the addition of additive metal element, Ag in the range between 1 and 5 wt.%. All the composite cathode powder samples have been prepared via high energy ball milling (HEBM) method. From the XRD results, all pure peaks of unsintered and sintered LSCF-SDCC-Ag composite cathode retained their structures, while LSCF-SDCC spectra for the composite cathode recorded no change in phase with the addition of Ag content. Furthermore, the result elucidates from the FTIR testing exhibit that all LSCF-SDFCC-Ag composites cathode with different addition of Ag content revealed the existence of carbonate elements. Meanwhile, the analysis of EDS mapping remarked that the elements were homogeneously mixed and distributed uniformly. As a conclusion, LSCF-SDCC-Ag is presented with excellent microstructural composite cathode powder characteristic to be used in potential LSOFC application.

#### 5. Acknowledgments

This paper was partly sponsored by the Centre for Graduate Studies UTHM, and the authors acknowledge University Tun Hussein Onn Malaysia (UTHM) for their support in providing Multidisciplinary research grant (MDR - 1103) for the project and Postgraduate Students Scholarship (GIPS - Vot 1155).

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