1st 8th INSCIC
Yala Rajabhat University
21-22 Feb 2023

PROCEEDING
รายงานสืบเนื่องจาก

การประชุมวิชาการระดับชาติ ด้านวิทยาศาสตร์และเทคโนโลยีเครือข่ายภาคใต้ ครั้งที่ 8 และ การประชุมวิชาการระดับนานาชาติ ด้านวิทยาศาสตร์และเทคโนโลยีเครือข่ายภาคใต้ ครั้งที่ 1

The 8th National Conference on Science and Technology 2023 (NSCIC2023) and
The 1st International Conference on Science and Technology 2023 (INSCIC2023)

วันที่ 21-22 กุมภาพันธ์ 2566
คณะวิทยาศาสตร์เทคโนโลยีและการเกษตร
มหาวิทยาลัยราชภัฏยะลา

รายงานสืบเนื่องจากงานประชุมวิชาการระดับชาติด้านวิทยาศาสตร์และเทคโนโลยีเครือข่ายภาคใต้ ครั้งที่ 8 และ
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ปีที่พิมพ์ 2566

เลขมาตรฐานสากลประจำหนังสืออิเล็กทรอนิกส์ 978-616-8297-28-5
ข้อมูลทางบรรณานุกรมของหอสมุดแห่งชาติ
National Library of Thailand Cataloging in Publication data

ISBN (e-book) 978-616-8297-28-5

จัดพิมพ์แบบ อิเล็กทรอนิกส์

Cathode Electric Signal from a Low-Temperature Ceramic Molten-carbonate Fuel Cell Using a Carbonate Composite Electrolyte

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Abstract

Low-temperature Carbonate Composite Electrolyte of Molten-carbonate Fuel Cell (MCFC) is adapted from Solid Oxide Fuel cells (SOFC) technology, which employs a Barium Oxide (BaO), Yttrium (Y_2O_3), Cobalt Oxide (CO_2O_3), ferric oxide (Fe_2O_3) (BYCF) cathode deposited on the Samarium nitrate doped Cerium (SDC10) and Sodium Carbonate (Na_2CO_3), Lithium Carbonate (Li_2CO_3), Potassium Carbonate (K_2CO_3) electrolyte layer and operated at 600 °C. The structure is formed in pellets of diameter 15 mm. In order to examine the physical and electrical properties of the BYCF cathode test doped electrolytes resulting from the combination of SDC10 and NLK, these electrolytes are referred to as "NLK electrolytes" with varying ratios. It was discovered that the optimal ratio of BYCF-doped SNLK was 70 wt% BYCF/SNLK and 30% cathode, which provided a high electrical conductivity of 0.77 S (s/cm) and a low activation energy of 0.280 KJ/mol, respectively. The minimum amount of energy that must be added to a particle of a substance is proportional to its theoretical porosity of 17.002%. For a chemical reaction to occur, activation energy is required because it causes the following changes in the particles of the reactant: Energy causes the particles of a substance to move faster, increasing the likelihood that they will collide. When properly positioned, collisions provide sufficient energy for a chemical reaction to occur. The findings indicate the feasibility of the molten carbonate fuel cell (MCFC) utilization. However, additional tests are required to prove the possible practicality of the concept and ensure the integrity of the information.

Keywords: Cathode, Low-temperature, activation energy, Electric signal

Introduction

The current demand for electricity is increasing, while the quantity of natural gas is gradually declining. However, the power generation capacity is still inadequate. Insufficient to rely on energy imports from other countries, and fossil fuels are scarce. Therefore, renewable energy, such as solar energy, wind power, hydropower, biomass power, and biogas, has been developed to replace fossil fuels by emphasizing clean energy and environmental friendliness, ensuring effective renewable energy and sustainability.

Fuel cells that are capable of directly converting chemical energy into electrical energy are the source of fuel cell technology in the clean energy category. There are numerous types of fuel cells at present, but only two are widely utilized: polymeric electrolyte membrane fuel cells (PEMFCs) and High temperature fuel cells, such as molten carbonate fuel cells (MCFC) and (SOFC). The polymer electrolyte membrane fuel cell can be used in automobiles and molten carbonate fuel cells can be used in industry (Khan et al., 2017), whereas the cells molten carbonate fuel cells and solid oxide fuel cells can operate at temperatures between 800 and 1000 °C. The disadvantages of high-temperature molten carbonate fuel

cells are actually favorable for industrial facilities. This study examined cathode materials suitable for use between 600 and 800 °C to replace LSM, a perovskite of La(1-x)SrxMnO(3-δ). The most common cathode used at 900 °C, which cannot receive electrical current, specifically in the presence of oxygen ions (O_2-) at 900 °C, where the cathode electrodes should have good electrical conductivity, significantly reduces the efficiency of the cell (Li et al., 2015). The molten carbonate fuel cell can be divided into three components: anode material, cathode material, and electrolyte material. The material for the anode is nickel. It has been used as an anode for a long time because it is a mixture of metals and ceramic (cermet), such as Ni/Yttria-Stabilized Zirconia or metal oxides such as Cerium. An oxide with good electrical conductivity and better compatibility with Yttria-stabilized zirconia than pure metals. In addition, it has been developed Corrosion-resistant anode material of sulfur in the source Natural hydrogen e.g. perovskite $Sr_{(1-x)}La_xTiO_3$ where x = 0.3-0.4 (Sutida et al.,2017). As for the cathode material, it must be a material that conducts electricity well and has a thermal expansion similar to that of the substance. The electrolyte has good conductivity for oxygen ions and is porous enough to allow the flow of oxygen gas. Examples of commonly used substances are lanthanum oxide compounds. Lanthanum, gadolinium, strontium, and yttrium. Popular compound used extensively in research is the perovskite $La_{(1-x)}SrxMn_{(3-\delta)}$ or LSM (Sutida et al., 2017). A good electrolyte layer must be able to carry oxygen ions from the cathode to the pole. The anode is made through the oxygen vacancy hopping mechanism, which uses heat to conduct the reaction and must have mechanical strength and stability for chemical reactions at room temperature up to 1000 °C. According to a research review, materials for electrolytes can be divided into compounds like SDC10-30, which have good electrical and ion conductivity and allow electrons to pass through a high-density electrolyte (Khan et al., 2017). The compound to be used as the material of the electrolyte is NLK, which consists of sodium carbonate (Na_2CO_3), lithium carbonate (Li_2CO_3), and potassium carbonate (K_2CO_3) and has advantages and is able to bring ions together. High density increases electrical conductivity. The low-density fuel gases or oxidants diffuse to the other electrode during operation (Fan et al., 2012). Therefore, the compound between SDC10 and the carbonate compound is chosen because SDC10 has the ability to conduct ions from one electrode to another, does not allow electrons to flow through the electrolyte, and is resistant to changes in size and shape (high density). while SDC10 may be less conductive when the temperature is less than 600 °C. Therefore, carbonates such as Na_2CO_3, Li2CO3, and K_2CO_3 are mixed to increase the electrical conductivity of the compounds SDC10 and carbonate, which are referred to in this study as SNLK compounds. In this study, the BYCF cathode and its compounds were investigated. SDC10 is a mixed electrolyte, whereas NLK is referred to as SNLK due to its thermal expansion difference. Individual cells decompose and cease to function as a result of the heat. To study the influence of the amount of SNLK added to BYCF cathode compounds, it is necessary to dilute SNLK with BYCF, which is referred to as SNLK/BYCF. It is a molten carbonate fuel cell capable of producing electricity between 600 and 800 °C.

Objectives

1. To investigate how the ratio of BYCF/SNLK in the cathode affects its physical and quantitative properties.
2. To examine the proportion of BYCF/SNLK suitable for use with the class electrolyte in cell fuel.

Method

Material preparation

The chemicals used in the experiment were Barium Oxide (BaO) 97% SIGMA-ALDRICH, Yttrium Oxide (Y_2O_3) 99.99% SIGMA-ALDRICH, Cobalt Oxide (CO_2O_3) 99.99% SIGMA-ALDRICH, ferric oxide (Fe_2O_3) 99.99% SIGMA-ALDRICH, Sodium Carbonate (Na_2CO_3) 99.99% SIGMA-ALDRICH, Lithium Carbonate (Li_2CO_3) 99.95% SIGMA-ALDRICH, Potassium Carbonate (K_2CO_3) 99.99% SIGMA-ALDRICH, Ce $(NO_3)_3$, $6H_2O$ 99.5% Acrosorganics, SM (NO3) 3, $6H_2O$ 99.9% Acrosorganics, Ammonia carbonate $(NH_4)_2CO_3$ 99.9% QreC and distilled water. As indicated in Figure 1, the research process consists of three primary components.

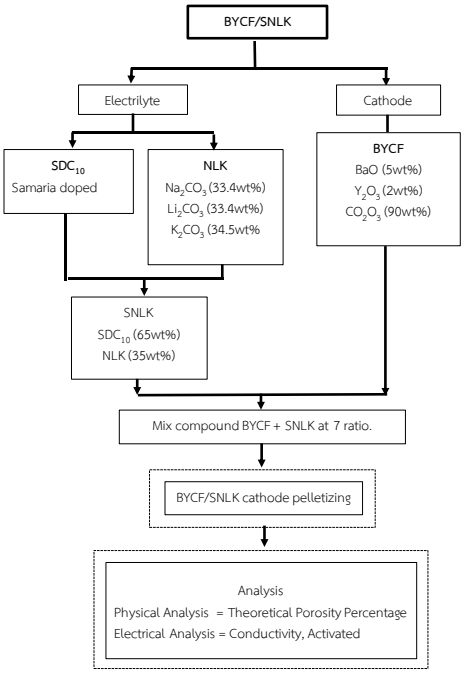

Figure 1. The Research Process.

Preparation of BYCF cathodes for molten carbonate fuel cells operating at various temperatures.

The BYCF (Ba0.054Y0.029Co1.8Fe0.062O2.89) composition was prepared using metal oxide powders of reagent grade from Sigma Aldrich. Through conventional ceramic powder processing, a composition of 5% BaO, 3% Fe_2O_3, 2% Y_2O_3, and 90% Co_2O_3 was obtained (Lund et al., 2017). The BFY was then mixed according to the calculated ratio in a flask with 150 ml of distilled water, 2 drops of PVA glue, and wet mixing for 24 h. After wet mixing was done, it was dried at 100 °C for 24 h then grated into a sample bottle and ground to a dry mix (dry mixing), then filtered and stored for mixing with CoO to produce BYCF cathode. Weighed the CoO and BFY according to the calculated amount (Appendix: Calculation of Preparation Steps). The substance (B: F: Y 5:3:2) was mixed with CoO, and added to 150 ml of distilled water, then added 2 drops of clear liquid glue from the water and wet mashed for 24 h. The substance was dried at 100 °C for 24 h. When the substance was dried, it was scraped into the sample bottle. It was ground and dried again for 24 h. Then the calcite was incubated at 1100 °C for 5 h in a ceramic crucible. Subsequently, the substance was sieved with a 425-mesh sieve, and grind the sifted substance with a grinder to obtain the BYCF substance.

Preparation of SDC10 electrolyte for molten carbonate fuel cells operating at various temperatures.

The preparation process of SDC10($Ce_{0.9}Sm_{0.1}O_{2-\delta}$) using the carbonate method co-precipitation as follows (Manjanna et al., 2013). Weigh Ce $(NO_3)_3$, 6H2O, and Sm (NO3)3, $6H_2O$ by dissolving them in 150 ml of water. Weighed $(NH_4)_2CO_3$ and mixed with 150 ml of distilled water. Stir the solution well to get a new $(NH_4)_2CO_3$ solution. Bring solution A to add by dropping into the $(NH_4)_2CO_3$ solution by constantly stirring the solution. Then form a white powder, measured at pH 9, and 60 °C for 1 h to obtain a white powder or solution B. Bring solution B to the filter and rinse with water several times. Until the pH reached 7, after which ethanol was washed. Wet sludge was then obtained and baked at 80 °C for 12 h. Burned at 600 °C for 4 h and set aside to cool, then scraped the substance off, grind, sieve with a 425-mesh sieve and finely milled to SDC-10 powder.

Preparation of the SDC10+NLK or SNLK electrolyte

The preparation of the SDC10+NLK or SNLK electrolyte was conducted using the steps described by Khan et al. (2018). Weighed Na_2CO_3 (33.4 wt%), Li_2CO_3 (32.1 wt%), and K_2CO_3 (34.5 wt%) from (Appendix C: Calculations of NLK Preparation Procedure), mixed with 150 ml of water, then wet ground with ball milling for 24 h, then the substance was baked at 150 °C for 12 h. SDC10 (65 wt%) was mixed with NLK (35 wt%) and mixed with 150 ml acetone, then wet milled by ball milling for 6 h. The substance was then dried at 150 °C for 12 h then scraped into sample bottles and ground again, prior to being dried at 1,100 °C for 5 h. The mixture of SDC10 + NLK was ground with a grit mill, then sieved with a sieve with a size of 425 mesh to obtain SNLK.

Preparation of SNLK/BYCF cathodes for molten carbonate fuel cells operating at low temperatures.

The preparation was performed as follows: Divide the BYCF/SNLK substance according to the appropriate amount. According to the stoichiometry principle, there was 7 parts, as shown in Table 1. Each ratio was brought into a bottle then added distilled water and 2 drops of PVA glue to improve adhesion when forming, then it was wet ground for 24 h. The substance was dried at 150 °C for 12 then scraped into a sample bottle and then dried for another 12 h. In order to determine the sinter/calcite temperature, calcite was calcined in a high-temperature furnace for 5 hours at 1100 °C and for 1 hour at 690 °C. When the calcite is complete, it is ground with a grit mill, sifted through a 425-mesh sieve, and then extruded into pellets.

Table 1. BYCF/SNLK divided into 7 parts.

Sample	BYCF/SNLK (w/w%)	BYCF (g)	SNLK (g)	SNLK+BYCF(g) Before dry	SNLK/BYCF(g) After dry
1.	100:0	1.2	0	1.2	1
2.	90:10	3.78	0.42	4.2	3.6
3.	80:20	3.36	0.84	4.2	3.5
4.	70:30	2.94	1.26	4.2	3.4
5.	60:40	2.52	1.68	4.2	3.4
6.	50:50	2.1	2.1	4.2	3.4
7.	0:100	0	1.2	1.2	1

Pelletizing process BYCF/SNLK (Lund et al., 2017) BYCF/SNLK was formed into a pellet mold by using a 15-mm compression socket. Each ratio was taken to form pellets with a compressive strength of 3,500 psi (pounds per square inch). Measured the diameter and thickness then weighed them before firing. The pellet-forming substance was burned at a temperature of 690 °C for 1 h with a heating rate of 5 °C/min and cooling

at 5 °C. After burning the BYCF/SNLK pellets, the diameter and thickness were measured and the pellets were weighed to determine their conductivity using the conductivity Electricity material properties study, which consists of electrical conductivity density and activation energy. Electrical conductivity is required because electric current results from the motion of electric charges. In which electric charges can move in many types of media, the properties of the medium that allow electric charges to move through are called conductive. Electrical conductivity at can measure the ability of a material to conduct the work of electric current; the higher the conductivity, because electric current is caused by the movement of electric charges (Xia et al., 2002).

The Activation Energy (Ea) or Activation Energy is the minimal amount of energy a particle of a substance must attain. Activation energy is required to initiate a chemical reaction because it changes the particles involved in the reaction. The energy accelerates the movement of the particles. The more work you do, the greater the likelihood that the particles will collide in the right places, providing them with sufficient energy to trigger a chemical reaction as in Equation 1. (Yonghyeon et. al., 2021)

(1) $$\ln K = \ln A - \left(\frac{E_a}{RT}\right)$$

Where A is the frequency factor for the reaction, R is the universal gas constant (8.3144598 J mol^{-1} K^{-1}), T is the absolute temperature (K), K = the reaction rate coefficient (min^{-1}) and E_a is the activation energy ($kJmol^{-1}$)

Results

Conductivity Electricity

The SI unit for measuring electrical conductivity is the Siemens per centimeter (S/cm). Figure 2 compares the electrical conductivity properties of chemical compounds at temperatures ranging from 50 °C to 800 °C. BYCF/SNLK with a ratio of 90%BYCF to 10%SNLK, 70%BYCF to 30%SNLK, 60%BYCF to 40%SNLK, and 50%BYCF to 50%SNLK, respectively.

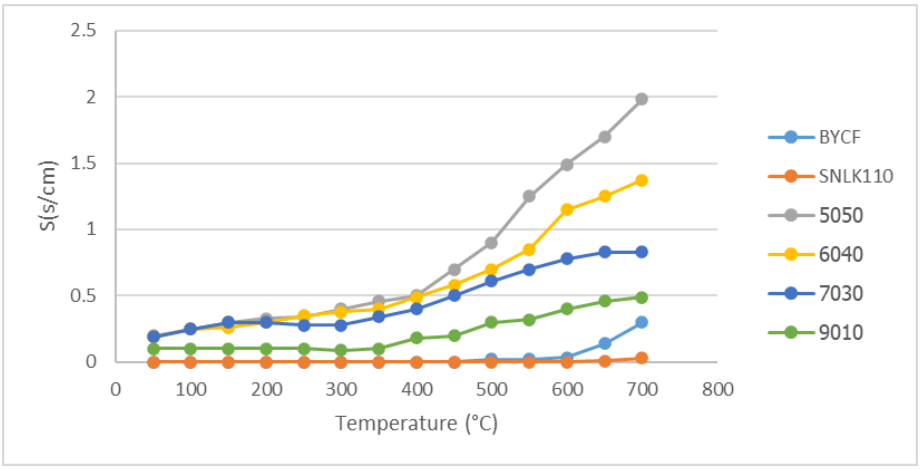

Figure 2. Electrical conductivity comparison of BYCF and SNLK at 50 and 700 °C.

Compounds of the cathode with the ratios 90wt%BYCF/10wt%SNLK, 70wt%BYCF/30wt%SNLK, 60wt%BYCF/40wt%SNLK, and 50wt%BYF/50wt%SNLK. Those containing carbonate compounds have a

higher electrical conductivity between 350 and 650 ° C (Figure 3).

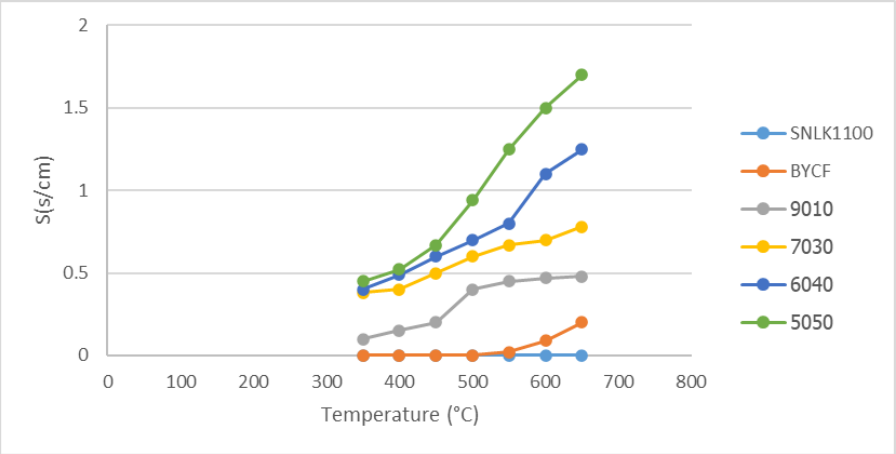

Figure 3 Electrical conductivity comparison of BYCF and SNLK at 350 and 650 °C.

The electrolyte compounds consisted of 100 wt% SNLK600, 100 wt% SNLK1100, and SDC10. These compounds exhibit increased electrical conductivity at temperatures between 350 and 650 °C. The SNLK600 exhibits the best conductivity curve. SNLK600 sintering makes the molten compounds more compatible with one another. NLK can be coated with SDC10, which has good compatibility and higher conductivity between 350 and 650 °C as shown in Figure 4.

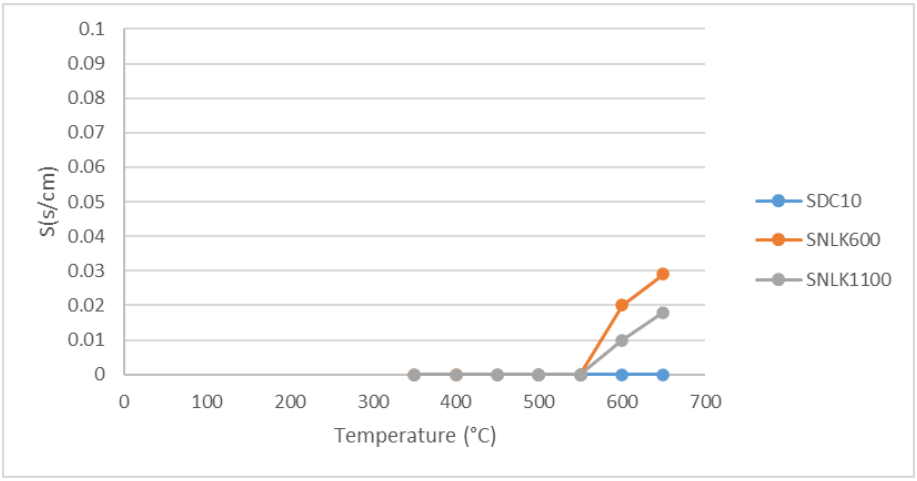

Figure 4 Comparative graph of the electrical conductivity properties of SDC10, SNLK600, and SNLK1100 electrolyte compounds at 350 and 650 °C

The best electrolyte compound, SNLK 600, had a conductivity of 0.029 S(s/cm) at temperatures between 350 and 650 °C, whereas the conductivity of (Zhao et al., 2013) is 1.83 S(s/cm) at temperatures above 800 °C. Consequently, we can conclude that the measured conductivity is less than the comparable value. However, the carbonate-doped electrolyte exhibited electrical conductivity at temperatures lower than the comparator's temperature range. The conductivity of the carbonate-doped electrolyte between 350 and 6500 °C is SNLK600, or 0.029 S (s/cm).

Porosity

Table 2 Percentages of BYCF/SNLK chemical compounds' theoretical strength under sintering at 1,100°C.

Compositions	(%Theoretical Porosity)
100%BYCF	22.456
(90%BYCF+10%SNLK)	16.667
(70%BYCF+30%SNLK)	17.002
(60%BYCF+40%SNLK)	16.743
(50%BYCF+50%SNLK)	16.790
(0%BYCF+100%SNLK.)	16.072

The percentages of BYCF cathode oxide compounds are as follows: 90wt%BYCF+10wt%SNLK, 70wt%BYCF+30wt%SNLK, 60wt%BYCF+40wt%SNLK, 50wt%BYCF+50wt%SNLK, 100wt%BYCF, and 100%SNLK1100 were used to determine the mixing = of BYCF/SNLK oxide compounds with porosity percentages theoretically suitable for fuel cell cathode application. It was discovered that the electrical conductivity of the substances in the ratio of 70%BYCF+30%SNLK is sufficient for low-temperature operation of molten carbonate fuel cells due to a porosity of 17,002%. Due to the fact that a large theoretical porosity permits electrons to flow efficiently through the cathode, if it is too small, oxygen gas will not flow efficiently through the cathode, as determined by comparing the work reference values of other researchers. The microstructure with fine porosity in the cathode layer has approximately 25% porosity (Chourashiya et al., 2011), and the porosity is approximately 23.057% based on the study of the 30wt%GY/BYCF ratio (Chourashiya et al., 2011). Theoretical porosity experiments of compounds in the proportion 70wt%BYCF+30wt%SNLK that are suitable for the manufacture of molten carbonate fuel cells.

Activation Energy

The activation energies of BYCF/SNLK cathode oxide compounds between 350 and 650 °C. At a ratio of 90wt%BYCF+10wt%SNLK, 70wt%BYCF+30wt%SNLK, 60wt%BYCF+40wt%SNLK, 50wt%BYCF+50wt%SNLK, 100wt%BYCF and 100wt%SNLK1100, 100wt%BYCF and 100wt%SNLK1100 are present. The 70%BYCF+30%SNLK ratio, which has an activation energy value of 0.280kJ/mol, has a low activated energy value. The best value obtained was 0.65kJ/mol (Ding et al., 2008) and 1.01 kJ/mol (Zhan et al., 2001) for the activation energy. Thus, the optimal ratio is 70%BYCF+30%SNLK (Table 3).

Table 3. Activation Energy at 350-650 °C

Composition	Activation Energy (KJ/mol)
100%BYCF	2.197
(90%BYCF+10%SNLK)	0.442
(70%BYCF+30%SNLK)	0.280
(60%BYCF+40%SNLK)	0.368
(50%BYCF+50%SNLK)	0.465
(0%BYCF+100%SNLK)	4.713

This indicated that the addition of SNLK carbonate resulted in a greater activation energy. This decrease is a result of SDC10's reduced conductivity at temperatures below 600 °C. Consequently, carbonates such as Na2CO3, Li2CO3, and K2CO3 are combined to decrease the activation energy of SDC10 compounds and

carbonates in reactions with low activation energy. The reaction is more simple and quicker than the reaction with a high activation energy and low intracellular loss.

Discussion

This study provided various ratios (90wt%BYCF/10wt%SNLK, 70wt%BYCF/30wt%SNLK, 60wt%BYCF/40wt%SNLK, 50wt%BYCF/50wt%SNLK, 100wt%SNLK600, 100wt%SNLK110 and 100wt%BYCF) of cathode electrodes for determining the optimal BYCF/SNLK ratio and evaluating the electrical and physical properties to determine the overall conductivity associated with cell performance. The composite BYCF/SNLK ratio with the highest value was the ratio of the activation energy of 70wt%BYCF/SNLK30wt%, which was 0.280KJ/mol, and this compound had the highest conductivity (0.77S(s)/cm). The findings also indicate that the theoretical percentage of porosity is appropriate for the cathode of a molten carbonate fuel cell. According to the electrical conductivity of the 70wt%BYCF/SNLK30wt% ratio, the activation energy is 0.280KJ/mol and the porosity percentage is 17.002%, implying that the value of the porosity percentage is 17.002%. Theoretically, there is a peak capacity suitable for molten carbonate fuel cell (MCFC) applications that is sufficient for fuel cell applications. Despite the promising performance of MCFC demonstrated in this research, additional long-term studies of the cell are still required to establish the concept's potential viability and ensure the integrity of the results.

Acknowledgment

The research was financially supported by the Innovation Fund at Prince Songkla University (RDO601415S). The authors wish to thank Prince of Songkla University in Thailand, the Wind Energy and Energy Storage Systems Centre (WEESYC), the Centre of Excellence for Materials Engineering (CEME), and the Faculty of Environmental Management.

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