SOFC solid electrolytes are known for their ionic conductivity characteristics, which increase with increasing SOFC operating temperature. Using COMSOL Multiphysics numerical simulation, analysis of SOFC power performance with yttria-stabilized zirconia (YSZ) and lithium sodium carbonate - gadolinium-doped ceria ({LiNa}2CO3-GDC) electrolytes was conducted to determine the potential of these electrolytes in their application in SOFC. The ionic conductivity of YSZ was differentiated based on the mole value of the yttria content, namely 8, 8.95, 10 and 11.54 mol. Meanwhile, GDC varied based on the (LiNa)₂CO₃ content such as 7.8, 10, 16.8 and 30 %. With the numerical model, the calculation error is an average of 7.32 % and 6.89 % for the experimental power and voltage values. In SOFC with the YSZ electrolyte, it was found that the power output can increase 26.4-35 times with an increase in operating temperature from 500 °C to 750 °C. SOFC with 8YSZ can produce the highest power compared to other YSZ, which is 123 A/m^2 at a current of 198 A/m² with an operating temperature of 500 °C and 3,440 A/m^2 at a current of 5,549 A/m^2 with an operating temperature of 750 °C. Whereas in SOFC with the GDC electrolyte, it was found that the power output can increase 18.6-22.6 times with an increase in operating temperature from 500 °C to 750 °C. SOFC with 30 % (LiNa)₂CO₃-GDC produced the highest power compared to other GDC, which is 231 A/m² at a current of 444 A/m² with an operating temperature of 500 °C and 5,240 A/m^2 at a current of 10,077 A/m² with an operating temperature of 750 °C. YSZ also showed the potential for an increase in power output as the SOFC temperature increases above 750 °C, while the 30 % variation (LiNa) 2CO3-GDC shows a limited increase in ionic conductivity at 750 °C

Keywords: SOFC, ionic conductivity, electrolyte, power performance, COMSOL Multiphysics, YSZ, GDC

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ANALYSIS OF THE EFFECT OF IONIC CONDUCTIVITY OF ELECTROLYTE MATERIALS ON THE SOLID OXIDE FUEL CELL PERFORMANCE

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1. Introduction

The environmental impact caused by the use of various energy conversion machines is a decrease in air quality, which will adversely affect the ecosystem, so efforts to overcome this are continuously carried out. Efforts are being made to find energy conversion machines that do not have a negative impact on the environment. One of the alternative energy conversion machines that are suitable for solving this problem are fuel cells. They only use hydrogen and oxygen to generate electricity through electrochemical reactions, which are much cleaner than conventional energy conversion machines, such as combustion engines and boilers that produce a lot of CO₂ and CO compounds.

When compared to other energy conversion machines, fuel cells are promising because they have the cleanest technology (zero pollution) and have a rather high efficiency of electricity generation. The exhaust emissions from fuel cells are water and heat, which can also be used as fuel cell by-products. In terms of operation and electricity production capacity, several types of fuel cells are used. One type of fuel cells that can replace power plants in energy conversion engines are solid oxide fuel cells (SOFC). SOFC are very suitable when applied to power plants because of the stability of equipment infrastructure in the long term, fuel flexibility,

and relatively low cost, in addition to the large production of electrical energy [1].

SOFC have many advantages, one of which is the potential as a regeneration system in other energy conversion machines because of their ability to operate at high temperatures [2]. With these advantages, it is estimated that the need for SOFC procurement will increase in the future. Therefore, the improvement of SOFC performance will be continued.

One of the important matters in SOFC systems is the ionic conductivity of the electrolyte material. It increases oxygen concentration in the species diffusivity. Therefore, optimizing the electrolyte design to increase the ionic conductivity is the focus of this study. In this research, YSZ material (8 mol % Y₂O₃-ZrO₂) was selected as electrolyte material. Research on fuel cells is still limited, especially research on the behavior of ionic reactions of hydrogen and oxygen radicals in delivering ions to SOFC electrolytes, which have never been analyzed due to limitations in research instruments. In recent years, studies have been undertaken to address barriers and problems for the practical application and commercialization of SOFC. There are several factors that influence SOFC performance such as geometry, material composition, and fuel stoichiometry factors, but numerical simulation research is quite useful

in solving complex problems. Due to increased computing power and improved simulation models and software, it is possible to calculate transport phenomena in fuel cells, such as the effect of fuel rate, temperature, anode pressure, fuel composition, and cathode pressure on fuel cell performance. So through this research, the effect of SOFC electrolyte materials on ionic conductivity needs to be studied. This ionic conductivity behavior involves changes in heat and fluid in the mechanism, so numerical simulations need to be carried out to predict and analyze their effects and changes.

2. Literature review and problem statement

Many parameters greatly affect SOFC performance, such as the magnitude of the cell potential difference, the thickness of the main components, and the operating temperature to the porosity of the material [3]. These parameters are very influential because they have the effect of circulating the fuel cell electrolyte ions in SOFC to react to produce electricity. The flow of these ions is greatly influenced by the ionic conductivity of the electrolyte material used, so the selection of the appropriate electrolyte material is an important factor [4]. Each electrolyte material has a different composition and performance in the ion flow, which will affect electrical energy production. Electrolytes are the heart of SOFC, which flow oxide ions from cathode to anode where they react with hydrocarbons to form H_2O and CO_2 , thereby allowing electrochemical reactions. Oxide ion conduction occurs through the hopping mechanism of the oxygen void, which will be activated thermally, so that the role of ionic conductivity is very large. In order to achieve high ionic conductivity in electrolytic materials, it is expected that smaller-sized cations are more likely to have sufficient mobility in the lattice.

The main requirements for the electrolyte to work efficiently are reliable mechanical properties and high conductivity of the oxide ion. Therefore, the choice of SOFC electrolyte material is a crucial thing [5]. SOFC electrolyte materials have microstructural parts, such as grain and grain boundaries that can affect their ionic conductivity. In SOFC electrolytes, grain and grain boundaries will be a medium for ion transfer from cathode to anode using the hopping method [6], where chemical reactions and coloumb forces will move these ions [7]. This ionic activity is the main factor in the ionic conductivity of a material. Conductivity is the ability of a material to move ions. The conductivity value of a material varies according to the atomic composition contained in the material and depending on the environmental temperature of the material. Brodnikovska et al [8] examined the relationship of doping to the ionic conductivity of electrolytes. They found that the addition of the right doping agent will increase the number of ions that can move freely, making ions able to transfer quickly, thereby reducing total resistance to the material and increasing the ionic conductivity of the material. Also, the addition of doping agent can change the grain boundary arrangement and create a smoother ion diffusion path, where each doping substance can produce a different grain arrangement and the number of ionic vacancies, where the vacuum of ions (which are generally oxygen ions) will affect the number of hopping processes in solid electrolytes [9].

During the past decades, many electrolytes have been made from ceramic materials. One of them is yttria-stabilized zirconia (YSZ), which is widely used as a solid electrolyte material in SOFC [10]. Other electrolytes are scandia-stabilized zirconia (ScSZ) and gadolinia-doped ceria (GDC). GDC is a solid electrolyte material that has higher conductivity compared to YSZ [11].

The comparison of the ionic conductivity of 8YSZ and 10Sc1CeSZ (scandia-stabilized zirconia) showed the lack of the number of vacant ions (oxygen ions), which will result in the majority of ions being transferred at the grain boundaries, where the conductivity of the grain boundaries is smaller [8]. 8YSZ has a smaller number of ionic vacancies than 10Sc1CeSZ, which could be due to the smaller number of doping substances in 8YSZ. Adding a doping agent affects the grain size and ionic conductivity of electrolyte materials. By adding barium doping to 8YSZ, the grain size of the electrolyte, as well as the ionic conductivity value, was increased [12].

Based on the previous research, the power performance of SOFC is strongly influenced by the ionic conductivity of electrolytes. On the other hand, the ionic conductivity of electrolytes, especially YSZ and GDC, changes depending on the percentage of dopping added into these electrolytes. Therefore, it is required to conduct research on the effect of dopping given to YSZ and GDC on the SOFC performance. The present research investigates numerically the effect of percentage of adding ceria on the GDC electrolytes and different mol values of yttria-stabilized zirconia on the ionic conductivity of electrolytes and power performance of SOFC.

3. The aim and objectives of the study

The aim of the study is to analyze the effect of ionic conductivity of electrolyte materials such as yttria-stabilized zirconia and lithium sodium carbonate – gadolinium-doped ceria on solid oxide fuel cell performance using numerical simulation.

To achieve this aim, the following objectives were set:

- to analyze the power performance of solid oxide fuel cells using the lithium sodium carbonate – gadolinium-doped ceria electrolyte for different ionic conductivity and operating temperature of fuel cells;
- to analyze the power performance of solid oxide fuel cells using yttria-stabilized zirconia for different ionic conductivity and operating temperature of fuel cells;
- to compare power performance between the yttria-stabilized zirconia and lithium sodium carbonate – gadolinium-doped ceria electrolytes for different ionic conductivity and operation temperature of fuel cells.

4. Materials and methods

4. 1. SOFC properties

The effect of ionic conductivity of electrolyte materials on SOFC power performance was investigated by numerical simulation using COMSOL Multiphysics 5.3a software, which solves differential equations based on the finite element method. The model has an integrated Battery and Fuel Cells module. It is an excellent modeling tool for SOFC modeling and simulation. The SOFC model was developed in COMSOL and it allowed the user to control several model parameters to optimize fuel cell efficiency.

The numerical simulation results were validated by comparing the experimental results. Also, the physical properties considered in the governing equation are listed in Table 1. The Battery and Fuel Cells module has many options, which can be used to combine various parameters related to the actual physics of the processes involved in real-world fuel cell problems. In this study, the characteristics of the electrolyte material will be validated with the results of experimental data from literature reviews and will be proven by a numerical simulation model (Table 2).

Table 1
Physical properties of SOFC for validation

SOFC properties	Values	
Electrolyte	8YSZ	
Length of flow channel (m)	1e-3	
Width of gas flow channel (m)	5e-4	
Height of gas flow channel (m)	5e-4	
Rib width (m)	5e-4	
Open-circuit voltage (Volt)	1.12	
Electrolyte thickness (m)	1.5e-4	
Electrode thickness (m)	5e-5	
Pressure (atm)	1	
Temperature (°C)	750	
Porosity	0.4	

Table 2

Curent density (Am ⁻²)	P (Wm ⁻²) [15]	P (Wm ⁻²) [14]
1,000	938.503	791.280
2,000	1858.103	1661.92
2,500	2207.903	2082.52
3,700	2787.503	2626.45
4,100	3198.023	3155.58
5,500	3383.863	3408.56
6,000	3445.663	3508.74
7,100	3449.867	3685.41
8,000	3265.891	3369.84

4. 2. SOFC geometry model

SOFC numerical simulation was represented by a SOFC stack with a model geometry as shown in Fig. 1. The fuel cell model consists of five components, two electrodes (anode and cathode) clamping the electrolyte in the middle and two gas flow channels on the outside of the electrode. In this study, oxygen flowed through the upper gas channel while the lower gas channel was used to flow hydrogen. Oxygen and hydrogen flows moved in opposite directions through the channel.

The variable investigated in this research was the ionic conductivity of SOFC electrolyte materials. The electrolyte material was taken from the deduction of the literature, YSZ [13] and GDC [14]. YSZ (yttria-stabilized zirconia) was one of the solid electrolytes used for SOFC, and the ionic conductivity of YSZ was due to the absence of oxygen, where Y_3^+ atoms replace some Zr_4^+ . The replacement of these atoms increased the oxygen void, then the appearance of different ionic conductivity values for each YSZ was varied. The type of electrolyte was the commonly used electrolyte, that is SOFC. (LiNa)₂CO₃ – GDC (lithium sodium carbon-

ate – gadolinium-doped ceria) is one of the SOFC electrolyte options that have advantages over YSZ, where the application of GDC can be operated at lower temperatures than YSZ, in which the Ce (cerium) atom will be given an additional doping substance in the form of Gd (gadolinium) to form GDC. Then, (LiNa) $_2$ CO $_3$ will be heated with GDC to increase the ionic conductivity of the grain change. Table 3 displays the variables of electrolyte ionic conductivity used in this simulation.

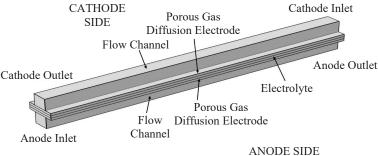


Fig. 1. SOFC geometry model

Table 3 lonic conductivity of electrolytes

, , , , , , ,			
Electrolyte	Ionic conductivity (S/m)	Tempera- ture (°C)	
8.04 mol % Y ₂ O ₃ -ZrO ₂ (YSZ) [13]	0.1	500	
	3.2	750	
8.95 mol % Y ₂ O ₃ -ZrO ₂ (YSZ) [13]	0.08	500	
	2.1	750	
10.01 mol % Y ₂ O ₃ -ZrO ₂ (YSZ) [13]	0.07	500	
	1.9	750	
11.54 mol % Y ₂ O ₃ -ZrO ₂ (YSZ) [13]	0.03	500	
	1.1	750	
0 %wt (LiNa) ₂ CO ₃ – GDC [14]	0.6	500	
	4.5	750	
7.8 %wt (LiNa) ₂ CO ₃ – GDC [14]	1.7	500	
	4.07	750	
16.8 %wt (LiNa) ₂ CO ₃ – GDC [14]	3.8	500	
	21.05	750	
30 %wt (LiNa) ₂ CO ₃ – GDC [14]	4.4	500	
	20.3	750	

The SOFC numerical simulation results will be compared with the validation data specified from previous literature research. The calculation error can be determined to find the relevance of the SOFC model to the actual tools in the experiment and simulation models in previous numerical studies. After that, the data collection according to the independent variables can be conducted. The validated SOFC model includes parameters that are independent variables to determine the effect of ionic conductivity on SOFC power performance.

4. 3. Governing equation

Solid oxide fuel cells use certain materials as components of solid electrolytes or electrodes. In this research model, the mathematical model used is based on (1), (2). These equations will provide a relationship between P (electric power) and V (cell potential difference)

Power density
$$[Wm^{-2}] = V_{cell}[Volt] \times Current density [Am^{-2}],$$
 (1)

$$V_{cell} = V_{nemst} - (V_{ohm} + V_{act} + V_{concen}), \tag{2}$$

where Power density is the amount of power per unit area produced by SOFC (W/m²); V_{cell} is the amount of the total potential difference that operates on the actual SOFC; Current density is the amount of current per unit area (A/m²); V_{nernst} is the amount of the change in the cell potential difference value when the reactants are not at their standard conditions (Volts); V_{ohm} is the ohmic loss (Volt); V_{act} is the loss of activation (Volt); V_{concen} is the loss of concentration (Volt).

Fuel cells produce electrical energy based on the amount of the potential difference between the cathode and anode (1). The potential difference will be directly proportional to the process of electron and ion transfer in the SOFC system. SOFC's electric current exists because of the transfer of ions and electrons in the SOFC circuit. In addition to the potential difference, the amount of electrical load or electric current will also affect the power generated in SOFC. The greater the potential difference and the electric current, the more power SOFC can produce.

 V_{cell} is the amount of the total potential difference that actually operates on SOFC. In actual conditions, the measured potential difference value at the SOFC output is the potential difference that has been deducted from overpotential. Overpotential is a voltage loss that exists due to phenomena in SOFC, such as heat generation due to electrical current or water trapped in SOFC components. This overpotential value is shown in equation (2) with the formula

$$V_{nernst} - (V_{ohm} + V_{act} + V_{concen}).$$

 V_{nernst} is the ideal voltage that has not been deducted by overpotential, whereas V_{ohm} , V_{act} and V_{concen} are 3 types of overpotential that occur in SOFC. OCV or V_{nernst} itself will be calculated according to the formula (3).

$$V_{nemst} = OCV = E^o + \frac{RT}{nF} \ln \frac{\text{product concentration}}{\text{reactant concentration}}.$$
 (3)

The concentration of reactants and products affect the value of the potential difference of cells, *R* is the ideal gas constant, 8.314 J/mol K, T is the temperature in kelvin, n is the number of electrons, where the fuel cell redox process with hydrogen fuel will involve 2 electrons and F is the Faraday's constant, 96485.3329 coulombs per mole of electrons. The reactant concentration and product concentration in equation (3) represent the reduction and oxidation activities in SOFC. The V_{nernst} or OCV formula itself is a derivation from the Gibbs equation. (3) explains how the effect of the ratio of reactants and products can affect the large difference between the cathode and anode. This ratio is also known as an equilibrium constant. The equilibrium constant is a number that represents an equilibrium condition or condition when a chemical reaction stops. While reactions that have not yet reached the equilibrium constant, a chemical reaction will continue to react until they reach equilibrium conditions. This ratio represents a redox process in SOFC, if reactants and products have reached equilibrium ratio, the product will stop reacting even in TPB (Triple Phase Boundary). As a result, the reaction ended. Usually, the equilibrium condition can be avoided by increasing the concentration of the product. Because of this phenomenon, the Gibbs equation derived into equation (2) is used to define V_{cell} in SOFC.

Concentration-dependent Butler-Volmer equation [15]

$$i_{loc}^{an} = i_o^{an} \begin{bmatrix} c_{\text{H}_2}^{TPB} \\ c_{\text{H}_2}^{o} \end{bmatrix} \exp \left(\frac{\alpha^{an} F}{RT} \eta_{act}^{an} \right) - \\ - \frac{c_{\text{H}_2O}}{c_{\text{H}_2O}^{o}} \exp \left(\frac{\beta^{an} F}{RT} \eta_{act}^{an} \right) \end{bmatrix} \left[\text{Am}^{-2} \right], \tag{4}$$

$$i_{loc}^{cat} = i_o^{cat} \begin{bmatrix} c_{\mathrm{H}_2}^{TPB} & \exp\left(\frac{\alpha^{cat}F}{RT}\eta_{act}^{cat}\right) - \\ -\frac{c_{\mathrm{O}_2}^{TPB}}{c_{\mathrm{O}_2}^{o}} & \exp\left(\frac{\beta^{cat}F}{RT}\eta_{act}^{cat}\right) \end{bmatrix} \left[\mathrm{Am}^{-2}\right], \tag{5}$$

where α^{an} ; β^{an} ; α^{cat} ; β^{cat} are the transfer coefficients: 0.5; 1; 3.5 and 0.5 respectively.

In SOFC, the value of electric current arises because of electron transfer. With the explanation in equation (3), electrons will move when a reaction occurs or there is a potential difference in non-equilibrium conditions. (4) represents the Butler-Volmer equation used in this numerical simulation. The Butler-Volmer equation is used to determine the effect of potential differences on the amount of current that will appear in actual conditions.

Formula (6) explains the relationship between currents and ionic conductivity [11]. This formula is used to represent the amount of electric current that can be produced based on its ionic conductivity. Ionic conductivity is the material's ability to move ions. In actual phenomena, ionic conductivity arises based on many aspects, for example, porosity and material composition.

$$I_a = \sigma_a x E_a. \tag{6}$$

In (5), I is the electric current, σ is the ionic conductivity (S/m) and E is the potential component a, while the letter "a" represents the electrode or electrolyte component. With (5), the statement of the relationship of electric power, voltage and electric current in (1) can be related to the value of ionic conductivity. Then the phenomenon that arises due to differences in types of ionic conductivity can be explained by (2) regarding overpotential, (3) regarding the equilibrium conditions and potential difference and (4) regarding the effect of potential differences on the SOFC electric current. As a result, the SOFC simulation by distinguishing the ionic conductivity of its electrolyte can be analyzed.

The formulas of ionic conductivity for YSZ are represented below

$$\sigma_{ion.}T = 4e_o^2 \frac{Do}{k} \exp\left(-\frac{Ea}{kT}\right) \times \left\{ -\frac{1}{2} \left(\frac{n_y}{2} + \frac{1}{K}\right) + \frac{1}{2} \sqrt{\left(\frac{n_y}{2} + \frac{1}{K}\right)^2 + \frac{2n_y}{K}} \right[s \frac{K}{c} m \right\},$$
 (7)

$$K = K_o \exp\left(-\frac{\Delta H_{as}}{kT}\right). \tag{8}$$

Table 4

The YSZ ionic conductivity will be based on the parameters listed above, where the conductivity will depend on temperature [13]. While the effective conductivity of the electrodes and electrolytes is:

$$\sigma_{ion-a}^{eff} = (1 - \varepsilon_a)(1 - \phi_{ano})\sigma_{ion}, \tag{9}$$

$$\sigma_{ion-c}^{eff} = (1 - \varepsilon_c)(1 - \phi_{cat})\sigma_{ion}, \tag{10}$$

$$\sigma_{ano} = \frac{95e6}{T} \exp\left(\frac{-1150}{T}\right) [S/m], \tag{11}$$

$$\sigma_{cat} = \frac{42e6}{T} \exp\left(\frac{-1200}{T}\right) [S/m], \tag{12}$$

$$\sigma_{cat}^{eff} = \sigma_{cat} \left((1 - \varepsilon) \phi_{cat} P_{el} \right)^{3.5}, \tag{13}$$

$$\sigma_{ano}^{eff} = \sigma_{ano} \left((1 - \varepsilon) \phi_{ano} P_{el} \right)^{3.5}. \tag{14}$$

Probabilities for an electron-conducting particle to belong to connecting ends of the composite, subscript i here represents electron- or ion-conducting species. This formula is also used to represent the electrochemically active specific surface area [18].

$$P_{i} = \left[1 - \left(\frac{4.236 - Z_{ii}}{2.472}\right)^{2.5}\right]^{0.4},\tag{15}$$

$$Z_{ii} = \frac{n_i Z_i^2}{Z},\tag{16}$$

$$n_{el} = \frac{\alpha^3 \phi_{el}}{1 - \phi_{el} + \alpha^3 \phi_{el}}; \quad n_{io} = 1 - n_{el},$$
 (17)

$$Z_{el} = 3 + \frac{z - 3}{n_{el} + (1 - n_{el})\alpha^{2}}; \quad Z_{io} = 3 + \frac{(z - 3)\alpha^{2}}{n_{el} + (1 - n_{el})\alpha^{2}}, \quad (18)$$

$$\alpha = \frac{r_{io}}{r_{el}}.\tag{19}$$

Electrochemically active specific surface area formulas [18] are shown below.

$$A_v = \pi \left(\sin^2\theta\right) r_{el}^2 n_t n_{el} n_{io} \frac{Z_{el} Z_{io}}{Z} P_{el} P_{io},$$

$$n_{t} = \frac{1 - \varepsilon}{\frac{4}{3} \pi r_{el}^{3} \left[n_{el} + (1 - n_{el}) \alpha^{3} \right]}.$$
 (20)

α

 n_t

Mi, Mj

vi, vj

between electron/ion particles

Ratio of particle size

Number of particles per unit

volume

Relative molecular mass (g/mol)

Kinetic volume

The diffusion term used is based on COMSOL Multiphysics modeling of Maxwell-Stefan [19].

$$D_{ij} = \frac{\left(3.16 \times 10^{-8} T^{1.75}\right)}{Pope\left(v_i^{1/3} + v_i^{1/3}\right)} \left(\frac{1}{M_i} + \frac{1}{M_j}\right)^{0.5}.$$
 (21)

Parameters	Definition	Value	
$i_{loc}^{an}; i_{loc}^{cat}$	Local current density	Calculated	
$i_o^{an}; i_o^{cat}$	Reference current density	Calculated	
$c_i^{TPB}; c_i^o$	Concentration on TPB, Concentration on inlet	Calculated	
$egin{array}{ccc} lpha^{an}; & eta^{an}; \ lpha^{cat}; & eta^{cat} \end{array}$	Transfer coefficient	0.5; 1; 3.5; 0.5	
$i_{ m H_2}^*$	Experiment current of hydrogen	Calculated	
$p_{\scriptscriptstyle \mathrm{H}_2}^*$	Experiment pressure of hydrogen	Calculated	
$i_{\mathrm{O}_2}^*$	Experiment current of oxygen	Calculated	
A_{des}	Pre-exponential factor	5.59e ¹⁵ [m ² ·mol ⁻¹ ·s ⁻¹]	
E_{des}	Activation energy	8.812e ⁴ [J mol ⁻¹]	
Γ	Surface site density	Calculated	
γ_o	Sticking probability	0.01	
A_a	Pre-exponential factor	2.07e ⁹ [Am ⁻²]	
E_a	Activation energy	8.78e ⁴ [J mol ⁻¹]	
A_{O_2}	Pre-exponential factor	4.9e ¹³ [Pa]	
$E_{{ m O}_2}$	Activation energy	2e ⁵ [J mol ⁻¹]	
A_c	Pre-exponential factor	5.19e ⁸ [Am ⁻²]	
E_c	Activation energy	8.86e ⁴ [J mol ⁻¹]	
p	Partial pressure	Calculated	
T	Temperature	1073 [K]	
k	Boltzman constant	8.62e ⁻⁵ [eV/K]	
e_o^2	(elementary charge) ²	10 ^{-18.8} [eV][S][s]	
K	Mass action constant	calculated	
n _y	Yttria concentration	Based on YSZ type	
K_o	Standard mass action constant	Based on YSZ type	
Ea	YSZ – activation energy	Based on YSZ type	
D_o	YSZ – high temperature limit of diffusion coefficient	Based on YSZ type	
ΔH_{as}	YSZ – standard association enthalpy	Based on YSZ type	
ε	Porosity	0.4 (anode and cathode)	
Φ	Electron-conducting particle fraction	0.6 (anode); 0.5 (cathode)	
Pel	Probabilities for an electron-conducting particle to belong to connecting ends of the composite	Calculated	
Z	Random packing system of spherical particles	6	
Φ_{el}	Electron/ion-conducting fraction	$\Phi_{cat};\Phi_{ano}$	
r_{el} , r_{io}	Radius of electron/ion-con- ducting particle	5e ⁻⁷ [m]	
n_{el} ; n_{io}	Fraction number of electron/ ion-conducting particles	Calculated	
$Z_e; Z_{io}$	Coordination number of electron/ion-conducting particles	Calculated	
$Z_{elel}; Z_{ioio}$	Average coordination number	Calculated	

Nomenclature

Calculated

Calculated

Based on species

6e⁻⁶ H₂; 12.7e⁻⁶ H₂O; 17.9e⁻⁶ N₂; 16.6e⁻⁶ O₂

5. Results of the power performance of SOFC

Fig. 2, 3 show the results of this simulation compared to the research data in the literature as validation.

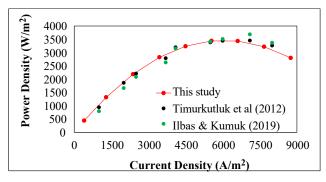


Fig. 2. Results of power simulations on SOFC models

In Fig. 2, the results of the SOFC's power simulation model of this study are compared with validation data. It can be seen that the SOFC model of this study has a power value with an average error of 3.60 % when compared to the power value of the reference experimental results [20]. While, compared to the power value in the reference simulation model [21], this model has an average error of 9.58 %. Table 4 represents the error of this model when compared to the validation reference [20, 21].

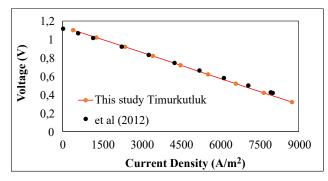


Fig. 3. Results of voltage simulations on SOFC models

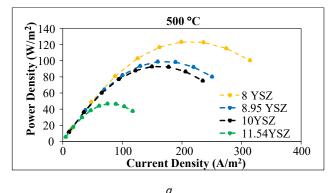
In Fig. 3, the voltage simulation results of this study are compared with the validation data [20]. It can be seen that the SOFC model of this study has a voltage value with an average error of 3.60 % when compared to the voltage value of previous literature.

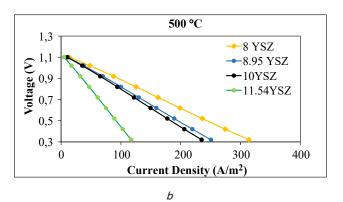
5. 1. Analysis of the YSZ electrolyte

The YSZ's composition can affect the value of its ionic conductivity. Fig. 4, 5 show the results of the electrical power performance of SOFC at 500 °C and 750 °C along with YSZ ionic conductivity values with variations of the yttria's mol value. The different ionic conductivities at 500 °C and 750 °C in these figures are examples of different activation energy values, while at higher temperatures, ion mobility and ionic conductivity increase. According to previous research [16, 17], the increase in ionic conductivity will be higher along with the increase in temperature due to the less activation energy needed. This is due to the very high level of ionic mobility, which can reduce the activation energy for a SOFC system to operate. The difference in ionic conduc-

tivity will be easily seen at low temperatures compared to high temperatures.

The graph in Fig. 4 shows the simulation results with variations of YSZ at a temperature of $500\,^{\circ}$ C. In this study, 8YSZ has the highest power value at $123\,\mathrm{W/m^2}$ at a current density of $198\,\mathrm{A/m^2}$. In this SOFC simulation, the highest current value can be reached by 8YSZ at $397\,\mathrm{A/m^2}$. The maximum power values for each variation of electrolytes are shown in Table 5. 8YSZ has the largest grain size and the smallest porosity compared to $8.95\mathrm{YSZ}$, $10\mathrm{YSZ}$ and $11.54\mathrm{YSZ}$. So, the greater composition of yttria doping will increase the conductivity of YSZ [22].





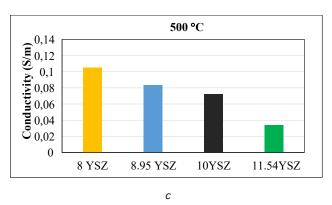


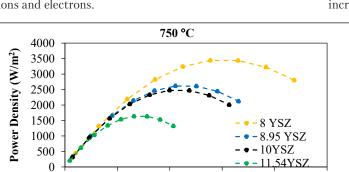
Fig. 4. SOFC power performance with 8YSZ, 8.95YSZ, 10YSZ and 11.54YSZ at 500 °C: a — power density; b—voltage; c—conductivity

The graph in Fig. 5 shows the simulation results with variations of YSZ at 750 °C. At 750 °C, 8YSZ has the highest power value at 3440 W/m^2 at a current density of $5,549 \text{ A/m}^2$. In this SOFC simulation, the highest current value can be reached by 8YSZ at $10,959 \text{ A/m}^2$. The amount of power and current

0

2000

is directly proportional to the highest ionic conductivity value of 8YSZ compared to other versions of YSZ. The maximum value in Fig. 5 is also caused by differences in current at different operational voltages. Where at a temperature of 750 $^{\circ}\text{C}$, the same voltage value at a temperature of 500 $^{\circ}\text{C}$ can cause a higher current because of the greater mobility of ions and electrons.



а

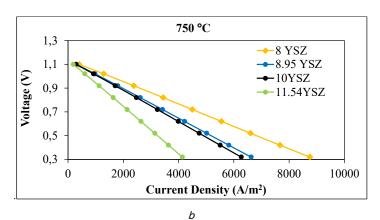
Current Density (A/m²)

4000

6000

8000

10000



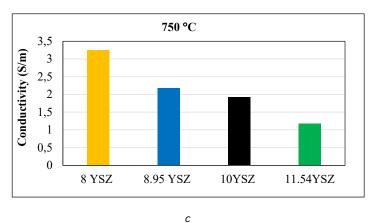


Fig. 5. SOFC power performance with 8YSZ, 8.95YSZ, 10YSZ and 11.54YSZ at 750 °C: a — power density; b — voltage; c — conductivity

Compared to the temperature of 500 °C in Fig. 4, 8YSZ, 8.95YSZ, 10YSZ and 11.54YSZ maximum power increases up to 27.9 times, 26.4 times, 26.6 times and 35 times, respectively. Increases of the maximum power in SOFC are the result of high temperatures that cause a high rate of ion mobility. This high ion mobility increases conductivity.

This phenomenon also shows the influence of yttria content in YSZ on the ionic conductivity of the electrolyte material at high temperatures, such as at 11.54YSZ. The higher the yttria levels, the higher the maximum power increase at high temperatures. This phenomenon is shown when the total power increase of 11.54YSZ is compared to the total power increase at 8YSZ, 8.95YSZ and 10YSZ.

5. 2. Analysis of the GDC electrolyte

The graphs in Fig. 6 are the results of simulation with a variation of $(\text{LiNa})_2\text{CO}_3\text{-GDC}$ at a temperature of 500 °C. In this study, the 30 wt % variation had the highest power value at 231 W/m² at a current of 444 A/m². At the same voltage, the highest current value can be achieved by 30 wt % at 442 A/m². The amount of power and current is directly proportional to the value of 30 wt % (LiNa) $_2\text{CO}_3\text{-GDC}$'s ionic conductivity, which has the highest value among other GDC electrolytes in this simulation. The maximum power value of SOFC with electrolytes 8 wt %, 16.8 wt % and 30 wt % at 500 °C can be seen in Table 5.

According to previous researches, it can be concluded that as carbonate co-doping increases, as at 30 wt % (LiNa)₂CO₃-GDC, it will increase the grain size. Then by reducing the number of material grain boundaries, it will increase the power output. Porosity can also influence the power value of the GDC. 30 wt % carbonate is the optimal value, where conductivity starts decreasing as carbonate co-doping content is increased in the GDC electrolyte [23]. The peak power in this SOFC simulation occurs because of voltage and current multiplication, according to Fig. 6, current increases as voltage decreases.

The graph in Fig. 7 shows the simulation results of (LiNa) $_2$ CO $_3$ -GDC at temperatures of 750 °C. In this study, 16.8 wt % and 30 wt % had the highest power values at 5,262 W/m 2 at 10,120 A/m 2 and 5,240 W/m 2 at 10,077 A/m 2 , respectively. At the same voltage, the highest current can be achieved by 16.8 wt % and 30 wt % at more than 18,000 A/m 2 . The same at 500 °C, the value of ionic conductivity is directly proportional to electric power production. In the simulation both at 500 °C and 750 °C, the maximum power value can be achieved with voltage and current that have the highest multiplication result. In reality, this is due to the actual reactant and product ratio that affects the voltage (potential difference) between electrodes and also affects the electric current production.

In this study, 16.8 wt % and 30 wt % can produce similar power, this proves the limit of the increase in ion mobility in an electrolyte system. Besides the activation energy required by high-temperature SOFC is much smaller than low-temperature SOFC, the ion mobility can be boosted by a greater porosity at 16.8 wt %. It is shown the power produced by 16.8 wt % is greater than the power generated at 30 wt %, which has a lower porosity. Compared to the simulation at 500 °C in Fig. 7,

0 wt %, 7.8 wt %, 16.8 wt %, 30 wt % experienced a maximum increase in power up to 18.6 times, 22.04 times, 22.81 times and 22.6 times, of which 30 wt % can produce the highest power at a temperature of 500 $^{\circ}$ C but at a temperature of 750 $^{\circ}$ C, the power produced by 30 wt % can be equivalent to the power produced by 16.8 wt %.

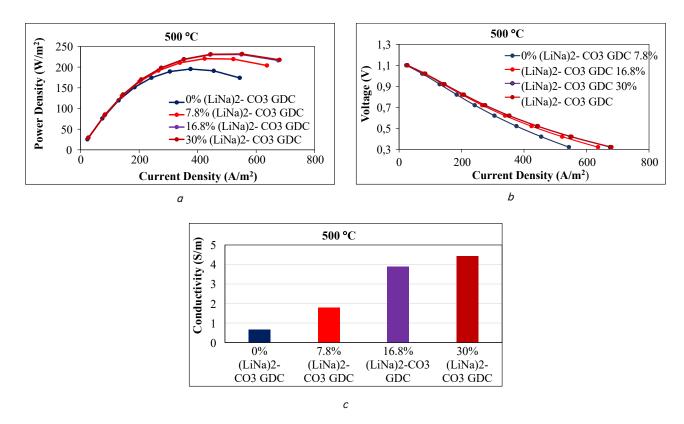


Fig. 6. Performance of fuel cells with the (LiNa) $_2$ CO $_3$ -GDC electrolyte at 500 °C: a – power density; b – voltage; c – conductivity

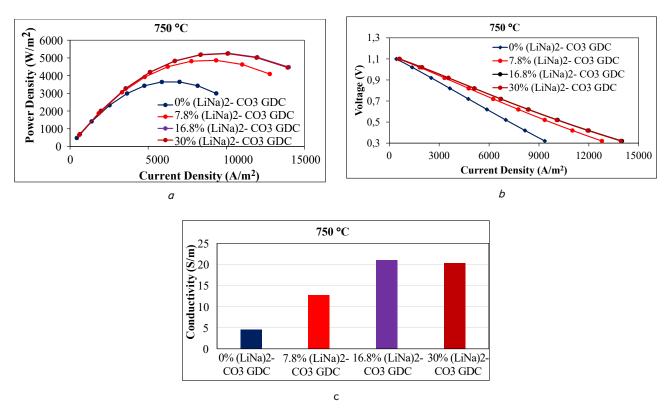


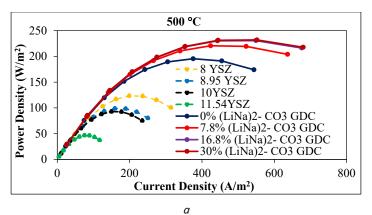
Fig. 7. Performance of fuel cells with the (LiNa)₂CO₃-GDC electrolyte at 750 °C: a – power density; b – voltage; c – conductivity

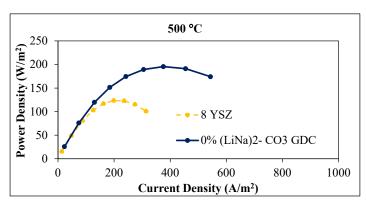
5. 3. Performance comparison between YSZ and GDC electrolytes for SOFC

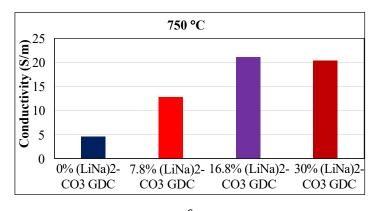
The YSZ and GDC electrolyte simulation shows the results that indicate the potential of the two electrolytes to be

a major component in SOFC. GDC's main component, ceria, seems to have better potential compared to zirconia (YSZ's main component) in operating SOFC at low temperatures. Ceria consists of cerium, an atom with atomic number 58

that has lower ionization energy compared to zirconium (zirconia's main component) with atomic number 40. This shows the difference between GDC and YSZ in the ability to react with oxygen, based on valence electrons. The following data show a comparison between YSZ and GDC in this simulation study. Fig. 8 also shows the highest SOFC power of YSZ electrolytes compared to the lowest SOFC power of GDC electrolytes at 500 °C.







b

Fig. 8. Comparison of simulation results between YSZ and GDC electrolytes at 500° C: a- power density; b- power density for 8 YSZ and 0 % GDC; c- conductivity

In Fig. 8, the gap difference can be seen between SOFC power using YSZ electrolytes and the power value of SOFC results using GDC electrolytes. In this study, the value of YSZ power is always below the GDC one at 500 °C, which is caused by the lower ionic conductivity of all YSZ variations than the GDC electrolyte's ionic conductivity. Fig. 8

also shows 8YSZ and 0 wt % carbonate co-doping GDC ionic conductivity values, where 8YSZ (the variation of yttria-Zr with the largest ionic conductivity among other YSZ variations) has a conductivity value that is 6.2 times lower than that of GDC with 0 wt % co-doping carbonate at 500 °C. Although GDC with 0 wt % carbonate doping is the variation that has the lowest conductivity than other GDC variations.

But in Fig. 9, it can be seen the gap difference between the power of SOFC results using YSZ electrolytes and the value of SOFC results using GDC electrolytes is getting smaller than at temperatures of 500 °C, although the value of the YSZ power is always below the GDC one at 750 °C. At 500 °C, the ionic conductivity ratio of 8YSZ and 0 wt % carbonate-GDC is 5:31, but at 750 °C, the conductivity ratio between 8YSZ and 0 wt % carbonate-GDC reaches 5:7. This shows a difference in the ability to transport ions in both electrolytes.

YSZ, which generally operates at temperatures of around 700 °C to 1,000 °C, shows unfavorable performance at 500 °C, but at 750 °C, YSZ's power performance can increase dramatically. According to Table 5, the maximum power of the YSZ variation ranged from 46 to 123 W/m² at a temperature of 500 °C, while a maximum power can be reached between 1,635 to 3,440 W/m² at a temperature of 750 °C. This increase shows the potential of YSZ to operate at high temperatures with high maximum power.

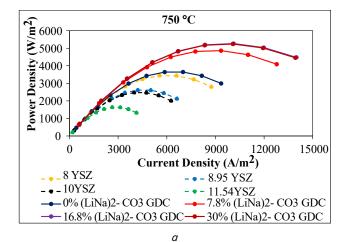
Table 5 Maximum power generated by each variation

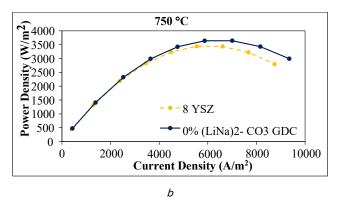
Model	Max Power (Wm ⁻²)	Model	Max Power (Wm ⁻²)
8YSZ-750	3440.80	8YSZ-500	123.33
8.95YSZ-750	2614.36	8.95YSZ-500	98.86
10YSZ-750	2474.24	10YSZ-500	92.81
11YSZ-750	1635.95	11YSZ-500	46.62
0LiNaGDC-750	3643.31	0LiNaGDC-500	195.43
7LiNaGDC-750	4860.82	7LiNaGDC-500	220.54
16LiNaGDC-750	5262.68	16LiNaGDC-500	230.62
30LiNaGDC-750	5240.28	30LiNaGDC-500	231.86

(LiNa)₂CO₃ GDC has good potential at low-temperature SOFC, this is indicated by a maximum power higher than for YSZ at 500 °C. However, at 750 °C, the maximum power produced does not increase significantly as the increase in maximum power at YSZ variations. GDC also shows the inability to operate at certain high temperatures, such as at 30 wt % (LiNa)₂CO₃ GDC, where its maximum power can be rivaled by 16.8 wt % (LiNa)₂CO₃ GDC.

Fig. 10 shows the dependence of ion-conducting particle size on the maximum power performance of SOFC.

The particle size of ionic conductivity has a significant effect on the power performance of SOFC. Increasing the radius particle of ionic conductivity decreased the maximum power performance of SOFC. As can be seen in this figure, if the particle size of ionic conductivity was doubled from 0.5 to 1 micron, the maximum power decreases by almost a half, by about 45 percent.





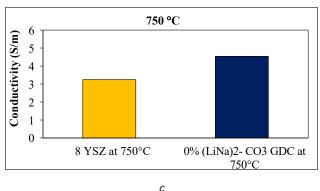


Fig. 9. Comparison of simulation results between YSZ and GDC electrolytes at 750 °C: a — power density; b — power density for 8 YSZ and 0 % GDC; c — conductivity

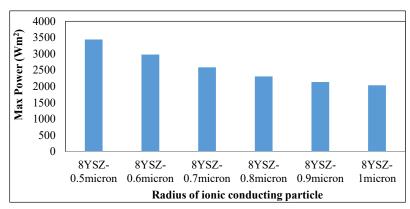


Fig. 10. Relation of ion-conducting particle size and maximum SOFC power

6. Discussion of doping content effect on grain size

The YSZ's composition can affect the value of its ionic conductivity. The more yttria doping put in the zirconia electrolyte, the greater the oxygen vacancy. For the yttria concentration increase above the 8 mol value, the ionic conductivity value of electrolytes tends to decrease, even when oxygen vacancy continues to grow [13]. On the other hand, the greater the yttria's mol value, the smaller the grain size. The larger grain size will increase ionic conductivity. This large grain size is responsible for decreasing grain boundary. Reducing grain boundaries will increase ionic conductivity in SOFC electrolytes. This is the reason why the greater composition of yttria doping will increase the conductivity of the YSZ

electrolyte and reduce the power output of SOFC. This increase in power output can also be caused by thermal expansion, which can reduce the porosity value of YSZ electrolytes and increase ionic conductivity, where the content of yttria doping agents can change the zirconia expansion characteristics. According to the studies [24], the length expansion at ceria ranges from 1.2 % at 800 °C whereas, in the research of zirconia variation expansion, the expansion of zirconia length ranges from 0.4 to 0.8 % at 800 °C. Both of these can also support a reduction in the porosity value, which can increase the value of ionic conductivity by increasing the Triple Phase Boundary. Most TPB is lost when the porosity is too high.

GDC is one of the solid electrolytes suitable for replacing YSZ. The co-doping composition of the GDC such as (LiNa) $_2$ CO $_3$ GDC containing a mixture of ceria, gadolinium and lithium sodium carbonate can affect the ionic conductivity values of the GDC electrolyte. (LiNa) $_2$ CO $_3$ GDC is one of the alkali carbonates that is proven to be able to increase the strength of the material and also can boost ionic conductivity at low temperatures. The GDC grain shape lengthens and enlarges with its (LiNa) $_2$ CO $_3$ GDC content as well as the increasing of ionic conductivity value. In this research, when carbonate co-doping increases, as at 30 wt % (LiNa) $_2$ CO $_3$ GDC-GDC, it will increase the grain size. Then by reducing the number of material grain boundaries, it will increase the power output. Porosity can also affect

the power value on GDC. 30 wt % carbonate is the optimal value, where conductivity starts decreasing as carbonate co-doping is increased in the GDC electrolyte content.

While the conductivity is changed in this model based on experimental results [14] and calculation [13]. The grain radius remained unchanged in this model while the conductivity changed. As shown in Fig. 10, increasing ionic particle radius will decrease the maximum power of SOFC in the same type of electrolyte. It is verified that the maximum power of SOFC decreases as the YSZ dopant increases and the limitation is seen in 30 wt % (LiNa)₂CO₃ GDC. In this research, the prediction of the optimum mol value of YSZ and the optimum dopping added to GDC to obtain the best SOFC performance have been well known. Further research is still needed to prove experimentally the tendency that has been achieved in the present study.

7. Conclusions

1. For different operating temperatures, the power performance of SOFC with the yttria-stabilized zirconia electrolyte has the same tendency. The power output of SOFC decreased with increasing the percentage of the yttria's mol value. The 8YSZ electrolyte produces the highest power at

 $500\,^{\circ}\text{C}$ and $750\,^{\circ}\text{C}$ due to the highest conductivity of 8YSZ compared to other YSZ. At $750\,^{\circ}\text{C}$, the maximum power of SOFC increases by 35 times compared to the operating temperature of $500\,^{\circ}\text{C}$.

2.30% (LiNa)₂CO₃ GDC produces the highest power at 500 °C. Besides, at a temperature of 750 °C, variations of 16.8% and 30% reach the highest power output of SOFC. This indicates that there is a temperature limit on GDC to increase the ionic conductivity of the electrolyte.

3. GDC shows the potential to produce high power at SOFC with low operating temperatures, while YSZ shows the potential to produce high power at SOFC with higher temperatures. The simulation model also shows that the increase in doping will increase the grain size, so the maximum power of SOFC will decrease. For GDC, increased doping increases the ionic conductivity but the loss of high-radius particles will limit the performance.

Acknowledgments

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