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Improving the permeability characteristic of nickle oxide/yttria stabilized zirconia anode through thermal decomposition of organic porous support as sustainable material for green hydrogen ecosystem

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ABSTRACT

Modification of the anode for fuel cell is essential to achieve the effective conversion rate at desired level. It is influenced by the gas distribution during the process. The compact profile of the anode is a direct impact for using sintering as reliable production method, making further material modification is demanded to solve the issue. At this work, practical solution is conducted to maintain the effective gas diffusion for anode, which is achieved through surface decoration enhancement. The study uses organic porous support (PS) as one sustainable and applicable method. The organic PS is made from flour, which evaporates during the sintering process. The diffraction profile for the produced anode indicates no substantial changes in the structural and physical characteristics. The morphology observation implies various models on the pore formation, including an elongated gap, achieved by a higher PS ratio (15 wt%). It promotes the highest permeability up to 0.425 m^2, with maximum partial pressure difference only 4.53 kPa. It shows the achievement of surface modification is reliable to provide substantial improvement on the gas distribution throughout the conversion process. Thus, the contribution of this work is possible for applied as reliable method to improve the pores formation.

Keywords: flour, morphology, pressure, pores, sintering.

INTRODUCTION

Environmental and sustainability are leading issues in current development, particularly in the energy transition. It makes the optimization and integration for both factors taken as primary indicators to offer a reliable solution. The advanced integration of the waste-to-energy concept [1], material recycling for energy storage [2], and sustainable biofuel processing [3] are conducted as a serious approach to provide a practical solution for sustainable development in energy systems. Practical solution for energy fulfillment to reach utility system is supported by implementing storage device, primarily for stationary energy system [4]. Researchers develop various improvement to achieve one reliable solution on the operational level by adding different approach on the given application [5]. Among various energy, hydrogen is considered as the most effective energy carrier that can be stored and converted in different system. It is also possible to utilize hydrogen both for stationary and mobile system. The net cycle for hydrogen energy is considerably positive with vast availability, making this element as the ideal candidate for future energy system. It is also accompanied by the growth in green hydrogen energy, particularly for developing and optimizing fuel cell systems [6].

Hydrogen can be applied in many energy sectors, while current development is specifically focused for electrical production using fuel cell [7]. Fuel cell is highly effective for electric production, especially for mobile system, since it operates quietly and produces water as the emission of process. There are several types of fuel cell, offering various benefit depends on specific requirement. For high temperature operation, fuel cell uses solid oxide (SOFC), which offers several benefits, making it an ideal candidate for the green hydrogen system [8]. The base anodes for the SOFC generally employ NiO/YSZ (nickle oxide/yttria stabilized zirconia), which withstand the given working temperature [9]. Despite its high benefit, operating the SOFC demands further study to achieve the desired function, making the system more reliable for actual consideration.

One major challenge for operation SOFC is the impact of thermal expansion of the component due to high temperature environment. It causes imbalance condition between cathode and electrolyte, causing severe issue on the process operation. Material and system modification offers many advantageous outputs to achieve the designed goal to encounter the disadvantageous for many practical energy material [10–12]. It also applied as solution to overcome the effect of high working temperature condition for SOFC by modifying the grain of the cathode/electrolyte is performed. F. Lu et al. used the concept by implementing thermal expansion concept for Ba-based cathode [13]. It reached lower thermal expansion by 41% and simultaneously provide better power at 650 °C. Employing thermal expansion offset is proven to maximizing the power density. Li et al. achieved the power density for La-based cathode at 1151 mW·cm^{-2} with identic working temperature [14]. At this condition, solution to overcome the thermal expansion is generally encountered for high temperature SOFC.

Another essential problem for the basic condition of SOFC is the gas diffusion which correlates with the porosity of the anode. A particular porous profile is required to ensure the diffusion of the gas during the operation. Modification of the production process was performed. For example, isostatic pressing [15], advanced inert chamber [16], and ultra-thin [17] makes a desirable performance (above 500 mW·cm^{-2}) and density. However, the practical approach relies on the sintering process, which is more beneficial in energy consumption and production costs [18]. It makes another modification is required to solve the given issue by maintaining the effective practical production method through sintering process.

The intense sintering process causes a dense profile, reducing the pore formation within the produced anode [19]. Several approaches were reported to address the problem. Polymeric film enhanced the porous formation of microtubular SOFC, showing that the porous structure improved up to four times [20]. N. Khatun et al. employed radio frequency sputtering for surface modification of Ni-based anode, indicating a higher power value up to 490.18 mW·cm^{-2}, corresponding to a better porous profile [21]. The mesoporous anode was developed in this work [22] for SOFC, improving the power by about 135%, which is related to a better microstructure profile. The reported work indicates a significant improvement in the performance of SOFC by performing surface modification.

The improvement in the performance is directly linked to the ability of gas diffusion for the anode. P. Lu et al. analyzed the multi-structural for the SOFC under the Z-shaped model, demonstrating a slight improvement in the power density (around 3%) by modifying the gas flow direction [23]. The operation of multi-flow arrangement corresponds to partial temperature variation. The issue was addressed by X. Liu et al. by analyzing the relation between temperature distribution and gas flow for segmented SOFC [24]. The finding implied that flow modification affects the temperature distribution, around 12% for counter flow direction. It confirms the urgency of providing steady gas diffusion for the effective operation of SOFC.

The effective gas diffusion is affected by the morphology of the anode. One crucial factor for the morphology is surface decoration enhancement, especially regarding the usability of sustainable material and applicable production methods for possible large-scale production [25]. It motivates for explore a novel method for surface decoration of the anode for a higher permeability at the desired level. This work aims to provide a detailed result for the possibility of using organic porous support (PS) to improve the performance of SOFC anode. The organic PS comes from a biomaterial that can be easily produced and is a considerable, cost-effective material. The evaluation is performed for the detailed material characterization and permeability profile, intending to provide a solid background for developing sustainable SOFC anode.

MATERIALS AND METHOD

The detailed process for producing sample pelleted SOFC is presented in Figure 1. There are two sample categories: anode supported (AS) with base material NiO-YSZ (NY) and modified AS (MAS). MAS was produced by adding porous support (PS) from the base material with the following weight ratio: 5 wt%, 10 wt% and 15 wt%. Basically, the producing method for both categories were similar. Thus, detailed description is focused for producing MAS. The initial step was conducted by mixing NY powder and organic PS at beaker glass based on the designed ratio. Then, alcohol (96%) was added into the mixture. It acted as solvent for the NY and PS powder. The mixture was handled specifically using ultrasonic cleaner which combined by mechanical stirrer for 120 minutes. The purpose for this combined step was to ensure the distribution of NY/PS powder and enhance the homogeneity of the final mixture. At the end of initial step, the MAS was produced as slurry mixture.

The second process was handling the slurry which was obtained from the initial step. The process was taken as the initial decomposition by heating the slurry inside electric oven at 150 °C for 180 minutes. It caused the decomposition of alcohol from the mixture. After that, the slurry turned into powdered MAS and ready for processed through compaction. Compaction was done to obtain solid mixture before handled to sintering. It was done by pressing the powdered MAS at forces 7×10^3, resulting in a compact form solid pellet. Finally, the compacted pellet was handled to sintering at 1200 °C for 360 minutes at specific heating rate of 3 °C/min. The sintering was essential to remove the organic PS from the mixture, causing some surface decoration and form porous structure of the produced pellet. In addition, the specific heating rate was designed to reduce the potential defect which may occurred throughout the sintering.

Surface modification of AS with organic PS was intended to improve the operational characteristic of fuel cell which highly related with the gas flow of within the system. It is highly correlated with the permeability aspect of the component, specifically for the pellet AS. Thus, assessment on the permeability aspect of the MAS was conducted to observe the effect of adding organic PS to the base NY. The schematic process of the evaluation was plotted in Figure 2a. At this process, the evaluated MAS was placed inside the gas chamber. High pressure gas was charged to the chamber. The pressurized gas flowed through the MAS, resulting in pressure different between the inlet and outlet zone. The value was measured directly using differential pressure gauge. The flowed gas from MAS was monitored using gas flowmeter. The dimension of gas chamber is presented in Figure 2b. The all aspects of the measured gas, including the technical dimension of gas chamber, was processed to determine the permeability value and pressure profile throughout the process. The permeability and pressure value are taken to determine the quality of the produced MAS and the effect of adding different ratio of organic PS. The photograph of the assessment apparatus is shown in Figure 2c. Detailed analysis was taken to support the assessment of the MAS.

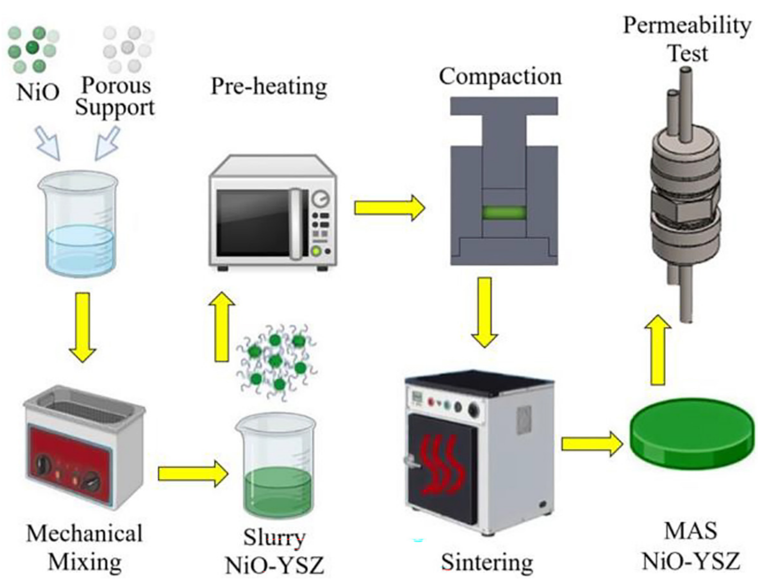

Figure 1. Illustration for obtaining MAS NiO-YSZ with organic PS

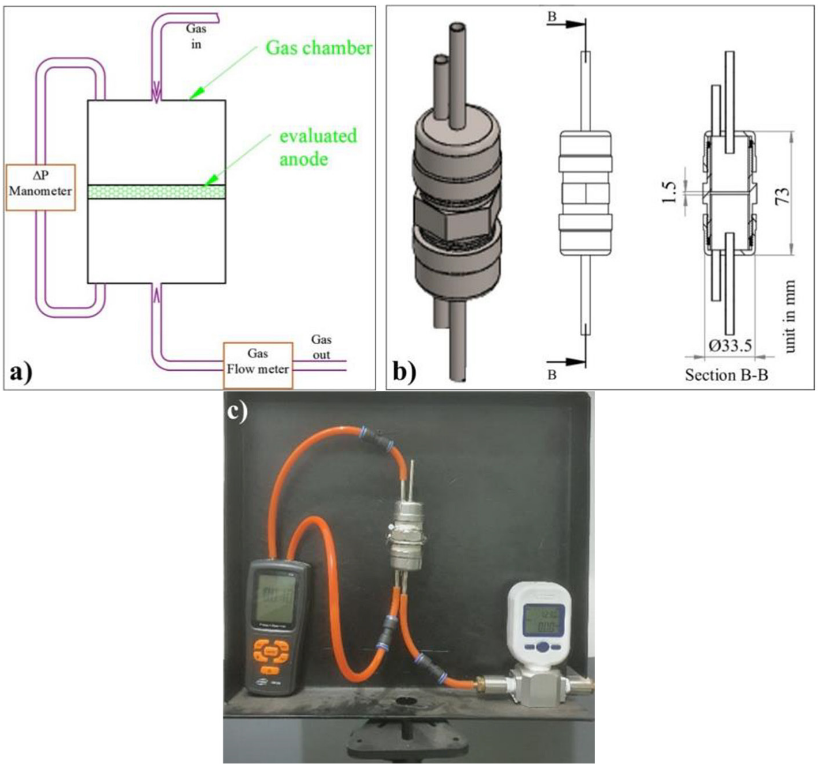

Figure 2. a) Schematic for permeability evaluation, b) 3D model and the dimension of gas chamber, and c) photograph of the equipment

This processed was done by characterizing the MAS and raw material using X-ray diffractometer (PANalytical). The organic PS was assessed for its decomposition profile. It was aimed to understand at which temperature the PS decompose and to help analyzing the probability of porosity within the MAS. In addition, surface decoration for the MAS was also observed by the help of scanning electron (SEM) microscope. Thus, an essential evaluation for the permeability and material characterization were taken to achieve the targeted results and assessment for the role of PS within the MAS.

RESULTS AND DISCUSSION

Figure 3a plots the thermal-decomposition characteristic of the PS. As observed, the PS has sufficient hygroscopic behavior which absorbs moisture from the ambient. It makes the initial decomposition started between 102.5–131.1 °C, representing the evaporation of water molecules within the PS. The liberates moisture accounts for 21.6% of the mass PS. The second thermal-decomposition continue at 253–477 °C, causing the total mass reduces to 15%. The region indicates the liberation of organic compound from the PS

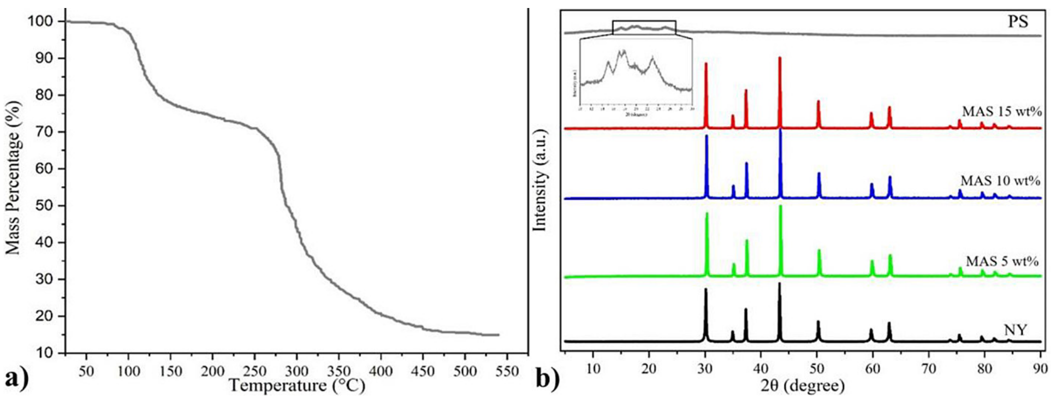

Figure 3. a) TGA porous support (PS), and b) XRD pattern for the base material and MAS

[26]. It confirms the processing of MAS removes most of the PS within the produced pellet. It is also observed according to the diffraction profile as presented in Figure 3b. The amorphous profile from PS is not observed for the produced MAS. In contrast, the MAS has identic diffraction angle with NY representing the crystal face centered cubic (FCC). The crystalline profile for NY has a specific diffraction angle at 2θ: 30.1°, 37.3°, 43.3°, 50.2°, 59.7° and 62.9°, which is similar from this reference [27]. It indicates the successful decomposition of organic PS for the produced MAS, resulting in identic structure for the produced pellet with the base NY. It confirms the addition of organic PS maintain the identic chemical and structural of NY. It is advantageous based on the operation of NY-based anode for SOFC since the essential characteristic of the base NY is achieved while the produced pellet achieves desired surface modification using organic PS.

The sintering makes solid profile for the anode, causing high barrier for gas flow on the operation of FC system. The presence of additional organic PS makes pore structure within the anode, making additional passage for possible gas flow on the operation. It appears notably for the produced MAS according to the microscope image as presented in Figure 4. There is clear evidence to confirm the XRD profile (Figure 3b) since the surface profile of the MAS indicate no substantial trail of the PS. The removed PS due to high temperature sintering making the pore formation for the MAS as the substance evaporate throughout the process. It is indicated by the yellow arrow in Figure 4, as the pore profile is observed for the all MAS. It is the major benefit for employing low cost and sustainable organic matter for making pore structure of pelleted AS-SOFC where the process achieved without altering the chemical structure of the MAS.

Different PS ratio makes the pore profile vary significantly, indicating different mechanism for the surface decoration of the MAS. The low PS ratio causes fractional distribution, making uneven formation of the structure, especially for large particle as indicated by red arrow in Figure 4a. It probably affected as the low ratio of PS evaporates at different rates, altering the position of the powder along with the sintering. Different surface for the MAS 10wt% (Figure 4b) creates larger pore (white box), which is accounted for the solidification of the PS at the mixing process and evaporates simultaneously, resulting in agglomerated of NY powder. The designed surface decoration is likely occurred fractionally for the MAS 5 wt% and 10 wt%, while larger ratio with 15 wt% PS resulting in a better profile. It is highlighted by the presence of elongated pore structure (red box Figure 4c), which is desirable for the gas diffusion. This contribution is linked to the higher PS content that may creating sufficient boundary between the NY powder and evaporates effectively to create

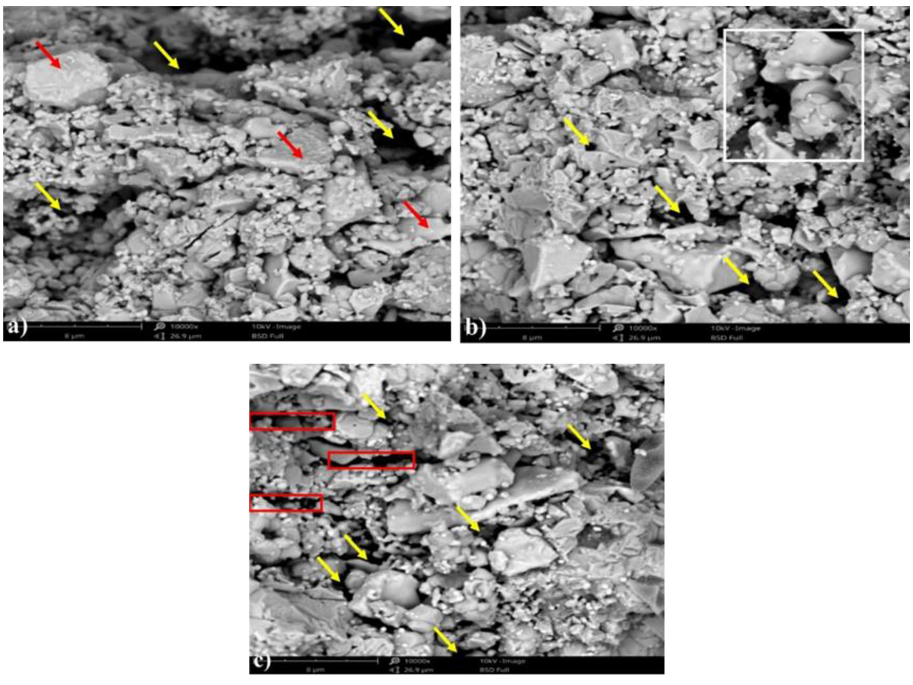

Figure 4. SEM images for: a) MAS 5wt%, b) MAS 10wt%, and c) MAS 15wt%

elongated profile. Despite that, the key achievement for pore structure of the produced MAS is technically achieved. The observed surface of the produced MAS demonstrate the proposed organic PS is technically feasible to achieve pore profile for NY-based anode SOFC, resulting in modified surface with porous characteristic that is favorable for the operation of fuel cell.

The AS requires satisfactory porosity to secure the effective working gas flow throughout the process. The addition of organic PS is intended to improve the pore structure, resulting in a higher porosity value of the AS. This value from several observation is plotted in Figure 5. The minimum and maximum value is added to highlight the variation of the actual condition from the gas characteristic of the SOFC. There is a certain aspect that PS contributes positively to create additional pore structure, resulting in a higher porosity ratio. It emphasized that pore profile as the PS evaporates is successfully achieved to make surface decoration for AS. Moreover, the higher porosity value is achieved than the ratio of PS content from the sample, implying the distribution and evaporation alters the powder structure which contribute to higher porosity. The improvement ratio for MAS 5wt% and 10wt% are similar, around 1.9%, demonstrating the mechanism of pore for the two sample are identic which also confirmed according to their SEM images (Figure 4a and Figure 4b). In contrast, the PS 15 wt% produces a higher improvement, approximately 2.7%, indicating the impact of sufficient effect to create surface decoration for the MAS. The higher improvement supports the SEM images (Figure 4c) which reveals the only sample that has elongated profile, contributing to a better gas flow and indicating a higher porosity. The variation on the porosity value demonstrates positive contribution of the presence organic PS at the initial production of NY-based anode SOFC. The changes in porosity value indicates the role of surface modification by adding organic material that able to fully evaporation throughout the sintering process. It makes the pore profile of the pellet is improved, potentially improves the ability of the operation for NY-based anode SOFC.

The impact of porosity variation relates with the pressure condition from the assessment process. The pressure value is plotted in Figure 6 to highlight the urgency to provide sufficient pore structure for the SOFC. The condition of the pressure for MAS 5wt% is extremely high than MAS 10wt% and 15 wt%. The maximum value is 7.46 kPa, demonstrating the insufficient gas diffusion which makes the pressure at the inlet port becomes higher than the outlet. It is disadvantageous for SOFC since it hinders steady operation to ensure the kinetic rate of the reaction at desired value. Modification with organic PS makes the pore structure for the MAS. Consequently, it has lower pressure differential, with the highest value only 4.97 kPa for MAS 10 wt% and 4.53 kPa for MAS 15wt%. The value confirmed notably that increment for the porosity value for the both type MAS (Figure 5), with the maximum value is achieved by MAS 15wt%. It corresponds to the achievement of the lowest average pressure

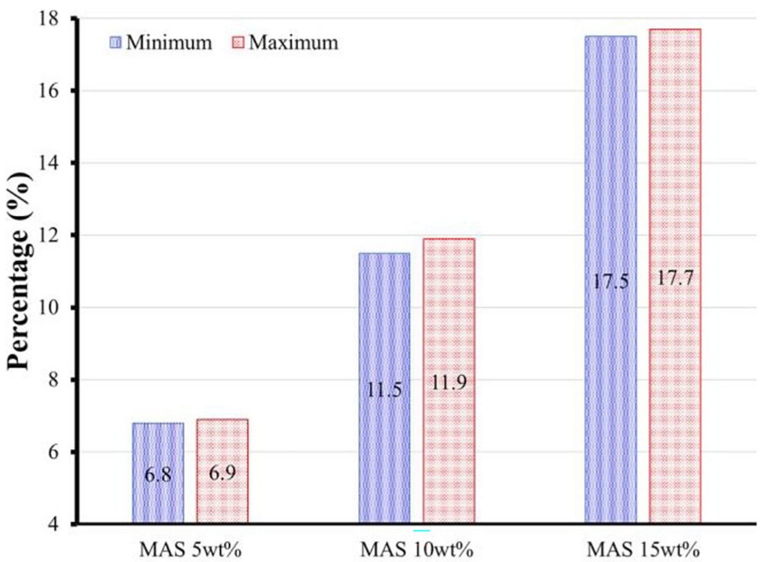

Figure 5. Estimated minimum and maximum porosity ratio of the produced MAS

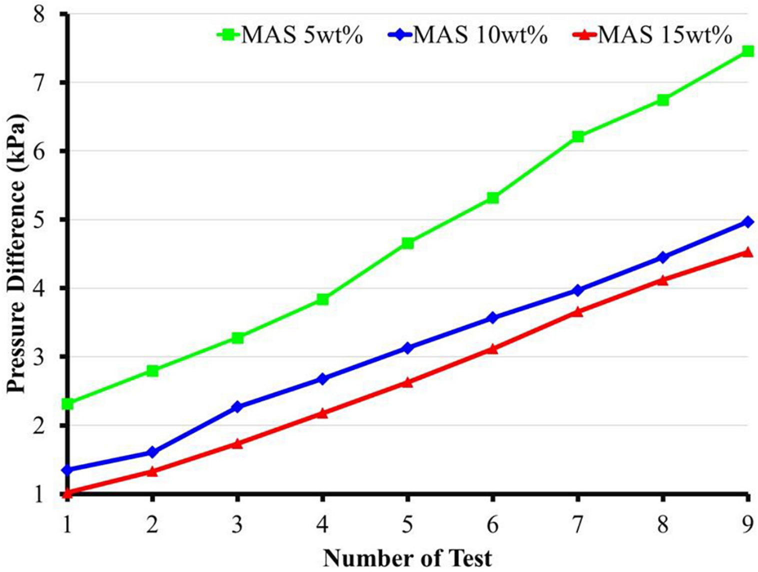

Figure 6. Characteristic of inlet/outlet pressure comparison for the tested MAS

difference for MAS 15wt%, which drops around 13.1 % compared to MAS 10wt%. The given parameter is important to confirm the achievement of suitable pore structure for both MAS, especially for ratio 15 wt% of organic PS. Moreover, the increment of pressure difference along with number of tests demonstrate the actual operation of the high variation on the mechanism of gas diffusion through the pellet. The higher porosity produces a lower pressure difference since it able to maintain the effective gas diffusion. It also indicates the dynamic condition of the operational for fuel cell which requires specific consideration to achieve desired pore structure for the NY-based anode SOFC. Suitable pore structure maintains the effective operation, reducing the pressure difference of the system which makes the operation of fuel cell is expected more steady and potentially increases the average power density.

The initial evaluation shows positive achievement and contribution for employing organic PS to produce pore structure within the MAS. Another assessment was directed to indicating the permeability value according to the parameter and testing results using gas chamber (Figure 2). The permeability for each MAS is shown in Figure 7. Surface decoration makes significant improvement for achieving pore structure despite the high pressure-temperature through sintering process. The obtained porous profile is highly correlated with the

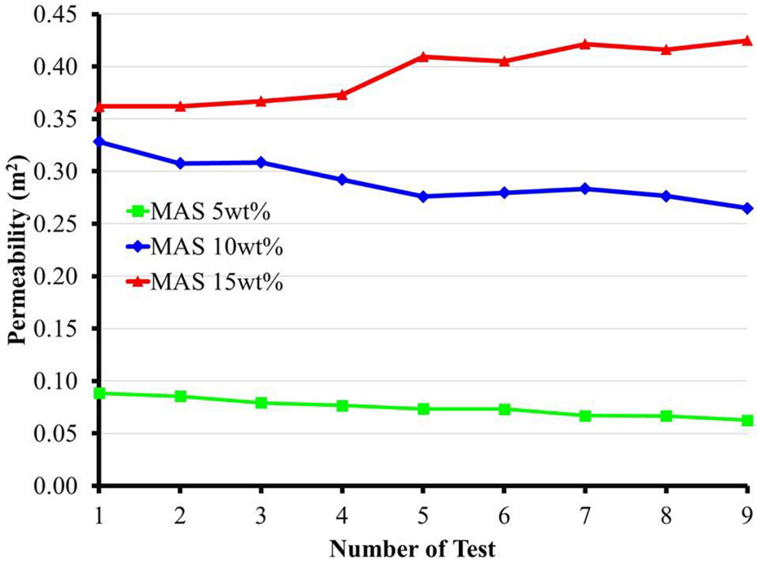

Figure 7. Estimated permeability profile for the tested anode

PS ratio from the process. The low content PS ratio resulting in lower permeability, as seen by MAS 5 wt% with the maximum value 0.088 m^2. Moreover, the deviation throughout the process is remarkably high, making high turbulence condition which result in higher pressure difference (Figure 6).

Improvement the amounts of organic PS into 10 wt% causes the permeability to changes significantly. It reveals the nature condition of the production condition that there is no linear trend between the ratio of PS and permeability value. Thus, the present work provides reliable indicator to set a baseline for using organic PS to perform surface modification for producing MAS-SOFC. It can be seen that the presented permeability for MAS 10 wt% elevates to 0.328 m^2, which technically more than twice time compared to the maximum permeability that achieved by MAS 5wt%. The presence of pore structure for MAS are identic, while increasing the ratio to 10wt% produces different distribution that provides additional configuration such as boundary formation as observed in Figure 4b. The feature allows the permeability value for the MAS 10wt% increased at higher ratio than MAS 5 wt%. It related with the condition to provide steady pressure profile, decreasing the pressure difference for the operation which allows the system to achieve steady operation and favorable conversion rate. The suitable achievement on the porosity level above 15% for MAS 15wt% (Figure 5) becomes one clear indication to obtain the requirement of suitable permeability value for the operation of AS- SOFC. The minimum target which set at 15% is expected to optimize the gas diffusion mechanism [28]. Improvement on the gas diffusion for the operation of SOFC eventually brings positive feedback to maximizing the reaction kinetic and ensure stable operation. Therefore, the achievement is considered suitable to increase the quality of NY-based anode SOFC.

The utilization of pore maker using organic PS from this work becomes one major leap to achieve the desired target for suitable porosity level, providing reliable permeability value up to 0.42 m^2 and simultaneously reduces the pressure difference. The ideal permeability and porosity allow the system to achieve a lower turbulence due to pressure difference, resulting in a favorable kinetic and operational stability. The obtained result is also emphasized the major achievement to obtain elongated profile for the pore structure of MAS 15 wt%, making this feature as another option for further controlled surface decoration to obtain more reliable production conduction of AS using organic PS as sustainable and low cost approach to support green hydrogen ecosystem.

CONCLUSIONS

Surface modification for NY-based anode SOFC is achieved through utilization of organic porous support (PS). The effect of PS ratio (5 wt%, 10 wt% and 15 wt%) essentially contributes to the variation of porosity value for the processed value, ranging from 6.8% to 17.7%. The value indicates the actual positive achievement by adding organic PS to provide pore structure of the produced pellet. Moreover, the XRD profile shows similar pattern between the base NY and MAS, indicating the mixing process maintains the main properties of the NY-based anode SOFC. It is the main feature for using organic PS which evaporates through the sintering process, followed by high potential for actual application considering the low cost material and widely available. The ideal pore structure for MAS 15 wt% reduces the pressure difference about 39.3%, demonstrating the critical aspect on the achievement of porosity value for the given sample. Inline with that, the permeability value for the MAS 15 wt% improves about three times than MAS 5 wt%. It shows different pore formation could be occurred using different ratio of organic PS. Moreover, the suitable achievement for MAS 15 wt% indicates the formation of pore structure is feasible to enhance the gas diffusion mechanism and ensure the effective operation of fuel cell.

The initial evaluation shows the suitability for using organic PS as a sustainable material to produce applicable surface decoration for SOFC anode. Further evaluation is advisable, including for producing a solid-state SOFC arrangement using the proposed method, including the evaluation for analyzing the power density and operation of the anode. The continuous effort to find reliable and applicable production methods is expected to boost the development of a green hydrogen ecosystem and support the energy transition in this era.

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