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Preparation and Characterization of Dual Layer Thin Layer Lanthanum Strontium Cobalt Ferrite /Alumina Hollow Fiber Membrane using Dip-coating and Brush-coating Techniques

(Penyediaan dan Pencirian Lapisan Dual Lapisan Nipis Lantanum Strontium Ferit Kobalt/ Membran Gentian Alumina Berongga menggunakan Teknik Penyalutan Celup dan Penyalutan Berus)

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ABSTRACT

This paper reports the preparation of the dual layer ceramic hollow fiber membrane that made of alumina and a mixed ion electron conducting (MIEC) material for simultaneous reaction and separation applications. Alumina hollow fiber membrane was prepared using the phase inversion process followed by a sintering technique at elevated temperature. The alumina hollow fiber membrane was used as membrane support onto which a thin and dense layer of lanthanum strontium cobalt ferrite (LSCF) was deposited. The main objective of this study was to investigate the LSCF coating formulations used in the deposition of LSCF layer onto alumina substrate membrane. The sintering temperature of thin LSCF layer was varied to investigate gas-tightness properties of LSCF membrane. A series of characterizations were conducted for both the support and the LSCF membrane. The result showed that the thin layer membranes with thicknesses ranging from 3 to 20 µm were successfully deposited on the surface of alumina hollow fiber support. The sintering process improved the gas-tightness properties but the sintering temperature above 1150°C caused defects on the surface of LSCF membrane.

Keywords: Asymmetric alumina hollow fiber; dip coating; lanthanum strontium cobalt ferrite; mixed ion electron conducting; phase inversion based spinning

ABSTRAK

Kertas ini melaporkan penyediaan membran dwi lapisan seramik gentian berongga yang diperbuat daripada alumina dan bahan elektron ion campur berjalan (MIEC) untuk aplikasi tindak balas dan perpisahan serentak. Membran alumina gentian berongga telah disediakan dengan menggunakan proses fasa balikan diikuti oleh teknik pensinteran pada suhu tinggi. Membran alumina gentian berongga digunakan sebagai membran sokongan dengan lapisan lantanum strontium kobalt ferit (LSCF) yang nipis dan padat akan dimendapkan di atasnya. Objektif utama penyelidikan ini adalah untuk mengkaji formula salutan LSCF yang digunakan dalam pemendapan lapisan LSCF ke atas membran alumina sokongan. Suhu pensinteran untuk lapisan nipis LSCF telah diubah untuk mengkaji sifat kedap gas LSCF membran. Terdapat beberapa siri pencirian telah dijalankan untuk kedua-dua membran sokongan dan membran LSCF. Hasil kajian menunjukkan lapisan nipis membran dengan ketebalan antara 3 hingga 20 µm boleh dimendapkan di atas permukaan alumina sokongan gentian berongga. Proses pensinteran meningkatkan sifat kedap gas tetapi peningkatan suhu melebihi 1150°C menyebabkan kerosakan pada permukaan membran LSCF.

Kata kunci: Alumina tidak simetri gentian berongga; fasa songsang berasaskan pemintalan; kobalt ferit lantanum strontium; pengaliran elektron campuran ion; penyalutan celup

INTRODUCTION

Ceramic membrane has started to receive substantial attention in various high temperature applications. There are many types of ceramic materials used for ceramic membrane preparations such as aluminium oxide (Al_2O_3), titanium dioxide (TiO_2), silicon dioxide (SiO_2), zirconium dioxide (ZrO_2) and a ceramic material known as the mixed ion electron conducting (MIEC) (Li 2007). Among them, the MIEC material has a unique characteristic to be used as membrane at high temperature applications. This material is an ionic semiconductor that can conduct both ions and electronic charge carriers (electron and/ or holes) without any electric supply (Riess 2003). The fundamental operation of MIEC is based on its oxygen vacancies behavior under oxygen partial pressure gradient. The oxygen ion transport within oxygen vacancy was explained by Wagner theory (Sunarso et al. 2008; Wei et al. 2013). Lanthanum strontium cobalt ferrite (LSCF) is one of the MIEC material that possesses long term stability, excellent oxygen permeability and highly electronic conductivity (Tan et al. 2008, 2005; Wang et al. 2009).

LSCF membranes have to be prepared in a hollow fiber configuration using the combination of the phase inversion and the sintering technique (Tan et al. 2012, 2010). The membranes consist of a thin layer membrane supported on the porous finger-like structure. Both thin layer membrane and the support are made of similar material, which is LSCF. Although this approach is very promising, it may increase the production cost if the ceramic is rather costly. Therefore, the use of porous support for the membrane serves as a good alternative to reduce a fabrication cost (Büchler et al. 2007; Rahman et al. 2012a). In this context, alumina hollow fiber membrane that has an asymmetrical structure composes of finger-like structure and sponge-like region can be great advantages to be used as the support for LSCF membrane (Rahman et al. 2012b). The development of a thin LSCF membrane on the alumina support can simultaneously enhance oxygen permeation flux (Gerdes & Luss 2006). This dual layer hollow fiber membrane can be prepared using an advance technique which is known as the co-extrusion/co-sintering technique (Droushiotis et al. 2010; Othman et al. 2010; Wu et al. 2010). The co-extrusion process involves the pressing of two different ceramic suspensions simultaneously via a triple orifice spinneret into a coagulation bath, to complete phase inversion process (Li et al. 2014; Othman et al. 2010). However, the co-extrusion/co-sintering possess has its own limitation especially when the ceramic materials used in the process have significant thermal expansion coefficient (CTE) differences (Li et al. 2014; Wu et al. 2011).

Therefore, the conventional coating techniques were used to prepare the dual layer membranes. The objective of this study was to investigate the LSCF coating formulations used in the deposition of LSCF layer onto an alumina support. Two different ceramic suspensions were formulated to deposit the precursors of LSCF membrane onto the outer surface of alumina hollow fiber support. So far, the preparations thin layer LSCF deposited onto alumina substrate using different formulations of LSCF suspensions have not been extensively discussed. The morphology of the dual layer membranes were characterized using the SEM-EDX analysis. The gas-tightness test was used to study the effect of sintering temperature on the permeability of LSCF membrane.

Experimental Details

MATERIAL

Alumina powder 1 μm (6-8 m2/g), 0.5 μm (32-40 m2/g) and 0.01 μm (100 m2/g) were purchased from Johnson Mattey, $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-\alpha}$ (LSCF6428) powders with a d_{50} in range 0.2-0.5 μm which purchased from KCeracell Co. Ltd, polyethersulfone (Radal A300, Ameco Performance, USA), N-methyl-2-pyrrolidone (AR grade, Qrec[tm]) and Arlacel P135 (Polyethyleneglycol 30-dipolyhydroxystearate, Uniqema) were used in the fabrication of alumina/LSCF dual layer hollow fiber membrane.

PREPARATION OF ASYMMETRIC ALUMINA HOLLOW FIBER MEMBRANE

1.3 wt. % of Arlacel was mixed in the NMP solutions prior to 53 wt. % of alumina powders addition. Different particle sizes were used at a ratio of 1:2:7 (0.01:0.5:1 μm). The details on the membrane fabrication can be found elsewhere including experimental setup (Paiman et al. 2015; Rahman et al. 2015). Finally, the sintering process was held for 8 h at 1400°C for a rate of 5°C min^{-1} to form high packing density ceramic body.

DEPOSITION OF LSCF THIN LAYER ONTO ALUMINA HOLLOW FIBER

Two different types of LSCF suspensions were prepared. First, a LSCF suspension was prepared by mixing LSCF powder with ethanol with ratio of 1:1, 1:2 and 1:4. Another formulation was prepared using a mixture of ceramic powder (i.e. 25 and 30 wt. %), polymer binder, dispersant and organic solvent. The dual layer alumina/LSCF ceramic hollow fiber membranes were dried in oven at 60°C for 24 h and later were sintered at temperature ranging from 1100 to 1200°C using a high temperature tubular furnace (XL-1700).

CHARACTERIZATION METHODS

The cross sectional images of alumina substrate and dual layer alumina/LSCF hollow fiber membrane were captured using the SEM machine (SEM, TM3000 Hitachi). The elemental surface mapping analysis with the energy dispersive X-ray spectroscopy detector (EDX, TM3000 Hitachi) was carried out on the dual layer membrane. The mechanical properties of dual layer alumina/LSCF hollow fiber membrane before and after coating were carried out using the Instron Model 5544 tensile tester provided with a load cell of 1 kN. The gas permeation test was carried out using a soap bubble meter. Helium (He) was used as sample gas, flowing at a flow rate ranging from 0.2 to 1 bar. The gas tightness test was carried out on the dual layer alumina/LSCF hollow fiber membrane using the gas permeation apparatus which can be found elsewhere (Tan et al. 2005).

Result and Discussion

PREPARATION OF ALUMINA HOLLOW FIBER AS CERAMIC SUBSTRATE

Asymmetric alumina hollow fibers were used as the ceramic substrate to deposit the thin layer of LSCF membrane on its outer surface. Figure 1 shows the SEM images of the ceramic support prepared using 53 wt. % of ceramic loading at ceramic/polymer ratio of 6. The figure shows that the outer and inner diameters of ceramic support were 1.68 and 1.08 mm, respectively. The thickness of the ceramic support was consisted of sponge-like region and finger-like voids. The finger-like voids which originated from the lumen covered 56% of support thickness. It has

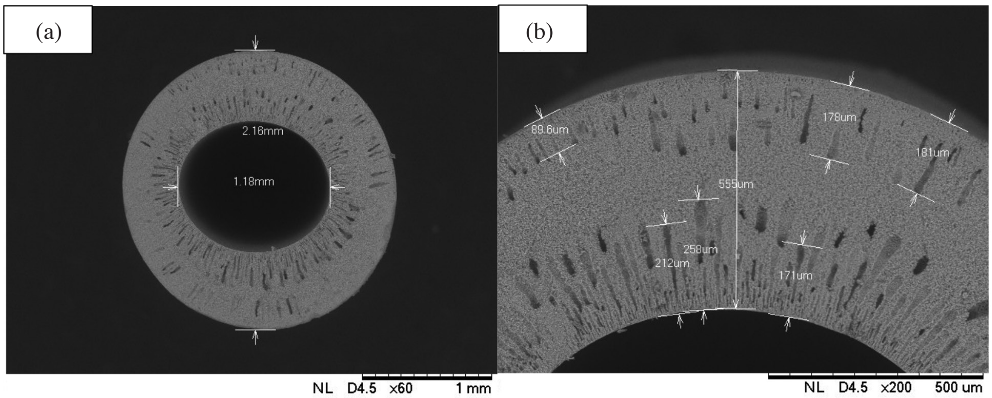

FIGURE 1. SEM images for asymmetric alumina hollow fibers (a) Overall cross section and (b) Wall thickness of hollow fibers

been known that the alumina hollow fiber membrane prepared using the phase inversion technique possessed the asymmetrical structure due to the hydrodynamically unstable viscous fingering phenomenon occurred during the phase inversion process (Kingsbury & Li 2009). This structure is favorable to be used as ceramic support as it provided porous channels to reduce a mass resistance without deteriorating the mechanical strength. This support has a mechanical strength of 24 MPa, which was sufficient to deposit LSCF membrane on its outer layer and to assemble into a membrane module. Figure 2 shows the permeability data of alumina hollow fiber support sintered at 1400°C. The permeation of helium gas increased from 1.08±3.37 × 10^4 mol.m^{-2} Pa^{-1}.s^{-1} to 13.87±3.17 × 10^4 mol.m^{-2} Pa^{-1}.s^{-1} when the pressure was increased from 0.2 to 1.0 bar.

PREPARATION OF LSCF MEMBRANE

In this study, two different types of ceramic suspensions were used to deposit LSCF layer onto the alumina hollow fiber support. In order to deposit LSCF membrane on the alumina support, the LSCF suspensions were prepared by mixing ceramic powder and pure ethanol using different weight ratios i.e. 1:1, 1:2 and 1:4. The SEM-EDX analysis

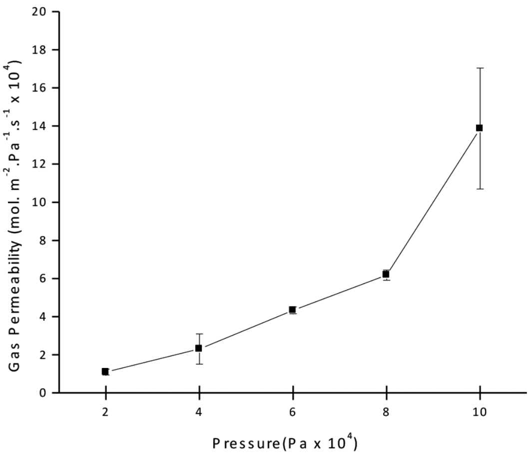

FIGURE 2. Gas permeabilities of alumina substrates

was used to characterize the adhesion of LSCF membranes, sintered at 1200°C on alumina hollow fiber support as shown in Figure 3. Based on the SEM images, LSCF membranes were successfully prepared on the alumina hollow fiber support using mixtures of ceramic powder and ethanol. Using this approach, LSCF membranes with the thicknesses of 17.3, 5.5 and 3.1 μm were obtained from ceramic suspensions with LSCF/ethanol ratios of 1:1, 1:2 and 1:4, respectively. In order to study the migration of LSCF particles into the porous alumina support, the EDX mapping was carried out on each SEM images. The migration of LSCF particles was based on each colors represent by La, Sr, Co and Fe species, respectively. It can be clearly seen that the migration of LSCF particles still occurred on each samples. Nevertheless, the LSCF membrane prepared using the LSCF/ethanol ratio of 1:1 was still adhered on the alumina support. LSCF suspension prepared using ratio of LSCF/ethanol of 1:2 and 1:4 which lower concentration depicted poor stability and could diffuse into alumina substrate easily as in Figure 3. Therefore, the gas-tightness test was carried out only on the LSCF membrane prepared using the ceramic suspension with LSCF/ethanol ratio of 1:1 to study the resistance towards flowing gases. The result showed that, even though the LSCF membrane was seen to have a good adhesion, it had a poor gastight property of 2.1434 × 10^{-5} ± 0.38 mol m^{-2} $Pa^{-1}s^{-1}$ to withstand different inert gas pressures. As reported by Tan et al. (2005), the gastight LSCF layer should have less than 10^{-10} mol.m^{-2} Pa^{-1}.s^{-1} of gas permeation.

In order to improve the coating of LSCF membrane on the ceramic support, the formulation of ceramic suspension was changed. Adopting the formulation of alumina support, the ceramic suspension for preparing the LSCF membrane on the alumina support was consisted of LSCF powder, NMP and polymer binder. The ratio between ceramic particle and polymer binder was set at 10:1 and the rest was consisted of NMP. The ceramic loading of LSCF suspensions were varied ranging from 20 to 40 wt. % to find a suspension with suitable viscosity for the dip coating technique. Based on an initial observation, the LSCF suspension prepared using 20 wt. % was too diluted. It was expected that suspension with low viscosity would lead to the diffusion of LSCF

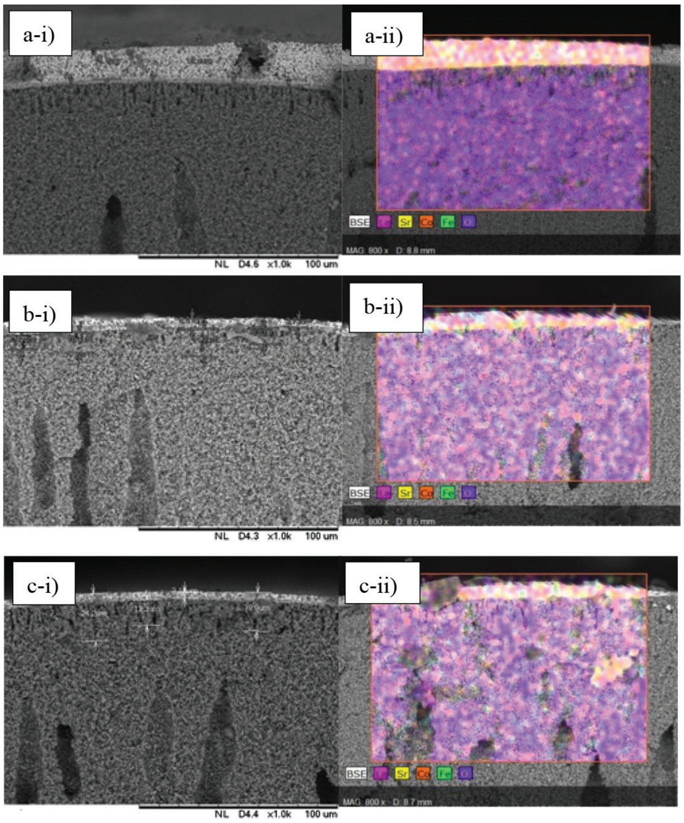

FIGURE 3. Alumina substrate deposited by LSCF using paint brush technique at different ratios (a-i) & (a-ii) SEM/EDX for 1:1 ratio (b-i) & (b-ii) SEM/EDX for 1:2 ratio (c-i) & (c-ii) SEM/EDX for 1:4 ratio

into pore structure of alumina hollow fiber support and consequently producing poor LSCF membrane. When the ceramic loading was increased from 25 to 30 wt. %, the LSCF membranes were successfully produced on the outer surface of alumina hollow fiber support using dip-coating technique. However, when ceramic loading was further increased more than 35 wt. %, a viscous LSCF suspension was produced, which was almost impossible to be used for dip coating technique. In order to study the adhesion of LSCF membrane on alumina hollow fiber support, the SEM-EDX analysis was used to observe the cross-section of the dual layer membrane. Figure 4 shows the SEM images of supported LSCF membranes prepared using 25 and 30 wt. % of LSCF loading. These figures show that both supported membranes have good adhesion onto alumina support. The EDX mapping also suggested that the diffusion of LSCF particle in the alumina hollow fiber support can be reduced. However, the thickness of the LSCF membrane was still affected by the ceramic loading of ceramic suspension. The LSCF membrane prepared using 30 wt. % ceramic loading has a thickness of 15 μm, compared to 7 μm thickness prepared using 25 wt. %.

EFFECT OF SINTERING TEMPERATURE ON THE MORPHOLOGY AND GAS-TIGHTNESS PROPERTIES OF LSCF MEMBRANE

In order to obtain a thin and dense supported LSCF membrane, the effect of sintering temperature was studied as it gives a significant effect on the gas-tightness property of the ceramic membrane (Zeng et al. 2007). The supported LSCF membrane should be as thin as possible suitable for the application that requires higher oxygen permeability. Furthermore, the sintering process is expected to improve the adhesion of the LSCF membrane and to remove the delamination and defects on membrane surface. In this study, the sintering temperature were varied at 1100°C, 1150°C and 1200°C for both supported LSCF membranes prepared using 25 and 30 wt. % ceramic loadings. Figure 5(a)-5(i) and Figure 5(b)-(i) shows the surface of supported LSCF membrane sintered at 1100°C. The SEM images show that the ceramic particles started to bind to adjacent particles, causing a neck growth to occur before the densification phase. However, this temperature was not sufficient to produce a thin layer and gas tight membrane. When the temperature was raised to 1150°C, LSCF particles were sintered as shown in Figure 5(a-ii) and Figure 5(b-ii). However, some cracks can be seen on the surface of LSCF membrane prepared using 30 wt. % ceramic loading. Figure (5a-iii) and Figure (5b-iii) shows the SEM images of LSCF membrane sintered at 1200°C. The figure shows that the LSCF membrane has major cracks on its surface based on direct physical observation. This suggests that the LSCF membrane should not sintered at temperatures above 1150°C.

Based on this study, it can be deduced that a very thin LSCF layer was not suitable to be sintered at a temperature

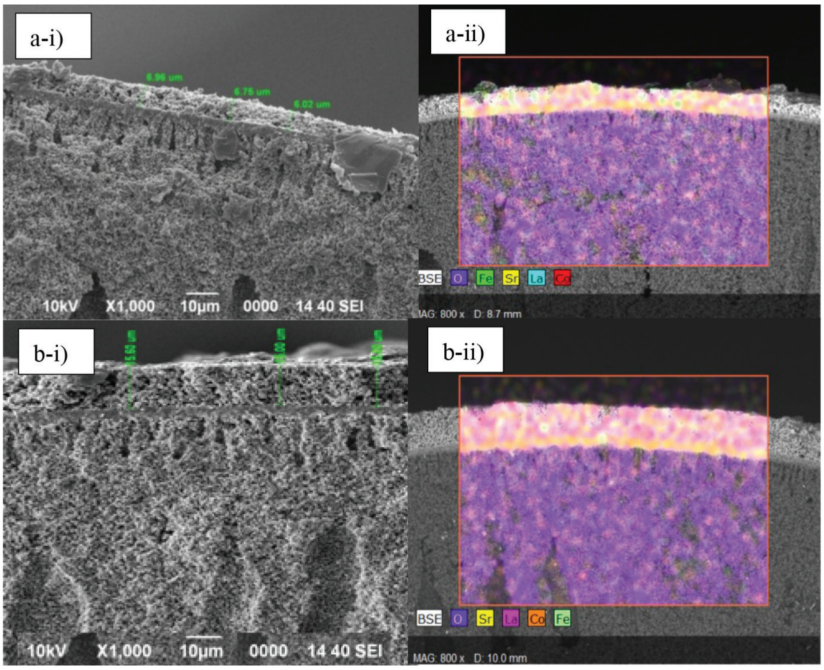

FIGURE 4. SEM-EDX of alumina substrate deposited by LSCF using dip coating technique (a) 25% of LSCF and (b) 30% of LSCF, (i) SEM images and (ii) EDX mapping

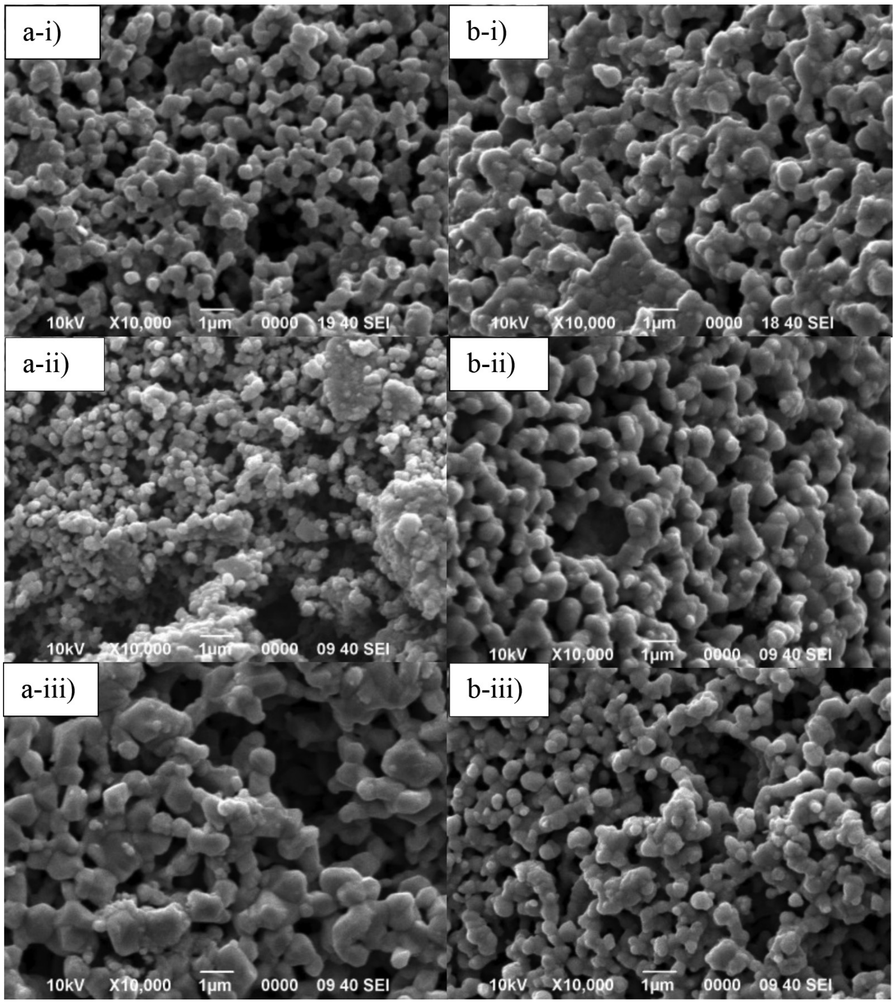

FIGURE 5. SEM images of surface of dual layer alumina/LSCF (a) 25% of LSCF, (b) 30% of LSCF, (i) sintered at 1100°C, (ii) sintered at 1150°C and (iii) sintered at 1200°C

higher than 1150°C. Bigger crack was expected to form on the membrane surface if the sintering process was carried out beyond this temperature. Gas tightness tests were carried out to prove that the dual-layer LSCF/ alumina hollow fiber membrane prepared using the single dip-coating was dense and gas-tight. The supported LSCF membrane with the lowest permeability of gas nitrogen was determined as dense and gastight as shown in Figure 6. At temperature of 1100°C, the nitrogen permeations were 7.711×10^{-6} mol m^{-2} $Pa^{-1}s^{-1}$ and 4.76×10^{-6} mol m^{-2} $Pa^{-1}s^{-1}$ for LSCF membranes prepared by 25 and 30 wt. %, respectively. The LSCF membrane prepared by 25 wt. % depicted lowest permeability (2.401×10^{-6} mol m^{-2} $Pa^{-1}s^{-1}$). This result might be due to increase of packing density of LSCF membrane. Meanwhile, the nitrogen permeation of 11.257×10^{-6} mol m^{-2} $Pa^{-1}s^{-1}$ obtained using the membrane from 30 wt. % of LSCF loading. The membrane sintered at 1200°C exhibited poor gastight properties due to the formation of cracks on LSCF surface. The nitrogen permeation of the membranes were 28.157×10^{-6} mol m^{-2} $Pa^{-1}s^{-1}$ and 12.862×10^{-6} mol m^{-2} $Pa^{-1}s^{-1}$ for 25 and 30 wt. % of LSCF loading, respectively. The graph showed that the LSCF membrane with 25 wt. % loading sintered at 1150°C depicted the lowest permeability compared to other sintering temperature.

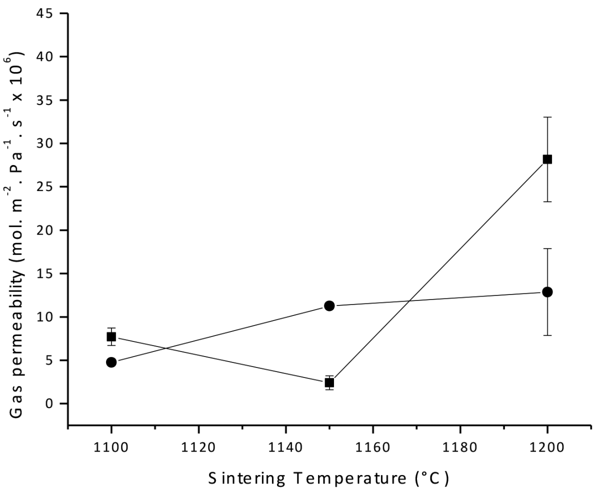

FIGURE 6. Gas tightness test for dual layer alumina/LSCF (■) 25% of LSCF and (●) 30% of LSCF

Conclusion

Asymmetric alumina hollow fiber membrane was successfully prepared using phase inversion followed by sintering technique at high temperature. The outer and inner diameters of alumina support membrane were 1.68 and 1.08 mm, respectively. The alumina support was offered excellent mechanical strength, better permeability and highly porous. Due to incompatibility of thermal expansion coefficient between alumina and LSCF, the co-extrusion was not suggested for preparing the dual layer alumina/ LSCF hollow fiber membrane. The LSCF membranes were successfully prepared using a ceramic suspension that consisted of ceramic particle, polymer binder, solvent and additive. The coating process enabled 3 to 20 μm LSCF membranes to be deposited onto alumina hollow fiber support. The thin LSCF layer should not be sintered at temperature above 1150°C. The LSCF membrane, prepared using 25 wt. % LSCF loading sintered at sintering temperature of 1150°C, has the lowest nitrogen flux, was assumed to have a dense structure. The nitrogen permeation value was 2.401×10^{-6} mol m^{-2} $Pa^{-1}s^{-1}$. It was expected that this value can be further reduced if multiple coating technique was applied on the alumina substrate.

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