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Dip-coating of SiO_2 onto ZnO-SiC Composite Membrane

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Abstract: SiC composite membrane was fabricated by mixing with SiC and ZnO powder. This mixture was pressed and sintered at 1,300 °C under air condition. This sintered ZnO-SiC membrane was dip-coated by silica sol and followed by heat-treatment. This membrane was characterized by XRD (X-ray diffraction), FE-SEM (field emission scanning electron microscopy) and BET (Brunauer-Emmett-Teller) instruments. Hydrogen permeation test was conducted at 0.1 MPa pressure and also variation of temperatures. The obtained value of heat-treated membrane after dip-coating at 298 K was obtained as 1.61×10^{-6} mol/(m^2·s·Pa).

Key words: ZnO-SiC membrane, dip-coated, hydrogen permeation test.

1. Introduction

SiC based separation membrane is needed to select proper method to produce support which has a strong chemical and mechanical template on which the selective material is well deposited [1-3]. The choice of membrane materials have important role to be used in densification for network formation as the high temperatures and pressures utilized. The fine micro-structures and densities of the coarse SiC ceramics could be controlled by alternating processes and also using the variety of metal oxides (ZnO, SiO_2, Al_2O_3, etc.) by doping.

SiC as stacking support having high mechanical strength makes the membrane more intensification by doping with ZnO into porous SiC network. ZnO as oxide additive has excellent electro-chemical properties including wide band gap with merits of environmental friendliness and mechanical stability [4, 5]. Additionally, ZnO as active ingredient can be easily synthesized in nano-structured forms with several excellent properties. Because this oxide has regular distribution of size and shape, it could be used to adjust open pores and control porosity of whole SiC system. By adoption of further treatment like a simple dip-coating method onto ZnO-SiC composite membrane, we assumed that, the hydrophilic property of the membrane surface as well as among the particles can be increased. The open pores of the particles and irregular size/shape formed by coarse composite membrane can fill up uniformly and continuously through the sol-gel process by silica sol. This method could control the degree of porosity and pore size distribution of the final product [6, 7].

The present work is to investigate proper fabrication method applying first doping with ZnO onto SiC network and followed by dip-coating process with SiO_2. The surface morphology and micro-structure even for thermo-chemical stability of the product can be affected greatly during the choosing of the proper preparation process. ZnO was used as active ingredient as modification material to improve micro-morphology and electro-chemical properties of the SiC using ceramic filter material or solid support for gas separation. Additional dip-coating approach can be used to fill up the pore and reinforce binding properties between hydrophobic ZnO and SiC interface connected by chemical binding of SiO_2. These membranes were characterized by XRD (X-ray diffraction), FE-SEM (field emission scanning electron microscopy) and BET (Brunauer-Emmett-Teller, BELSORP-max, mini II) analysis. Simple permeation

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tests were also done to develop the possibility of the gas separation membrane.

2. Experimental Procedure

Disc shaped ceramic supports were prepared by mixing 4:1 ratio of β-SiC powder (200-400 mesh) (Aldrich, No.409-21-2) and ZnO powder (99%-100.5%, No.1314-13-2, Aldrich) as doping material including 9 wt.% of phenolic resin as method published in Ref. [8]. Mixed powders were pressed in a mold with a diameter of 14.5 mm and a thickness of 2 mm by uni-axial pressing at 200 bars. The molded membrane was sintered at 1,300 °C for 3 h under air atmosphere. The silica sol was synthesized with TEOS (tetraethyl ortho-silicate) by general sol-gel methods [9]. The dip-coating process was performed at a constant dipping speed of 0.1 mm²/s and dried at room temperature. The membrane was heat-treated at 600 °C after drying since it can remove the all the organic remains. X-ray diffraction analysis (Bruker D8 focus, CuKα, 40 kV, 40 mA) was measured for the phase composition and the crystal structure study. BET surface analyzer was determined the pore size distribution by analyzing the adsorption-desorption of N_2 gas. Field emission-scanning electron microscopy (JEOL-JMS 7500 F) has been used for morphology of the surface and cross section of the membrane. The hydrogen permeability was evaluated from room temperature to 480 K using permeation analyzer at 0.1 MPa.

3. Results and Discussion

XRD patterns in Fig. 1 indicate (a) doped and (b) dip-coated ZnO-SiC composite membranes. All peaks are designated and shown as fine crystalline structures. The phases of the overall SiC are mois-sanite 84R (JCPDS file No.01-073-2086, a, b = 3.07 nm, c = 210.7 nm). The two types of new SiO_2 peaks arising from SiC surface were generated even in just doped one heat-treated under air. One was assigned to cristobalite which is tetragonal (JCPDS file No.00-039-1425, a, b = 4.97 nm, c = 6.92 nm) and the other one was coesite monoclinic structure (JCPDS file No.00-079-0445, a = 7.14 nm, b = 12.37 nm, c = 7.17 nm. The intensity of SiO_2 peaks in the sample of the dip-coated one were increased after dip-coating process while SiC and ZnO peaks were remained as reduced intensity, as shown in Fig. 1

The surface and cross sectional morphologies by FE-SEM are shown in Fig. 2. The just doped has more porous with large cavity than the dip-coated in Fig. 2. The surface and cross section of the dip-coated membrane was surrounded by the spherical and regular particle shape and denser structures than doped one.

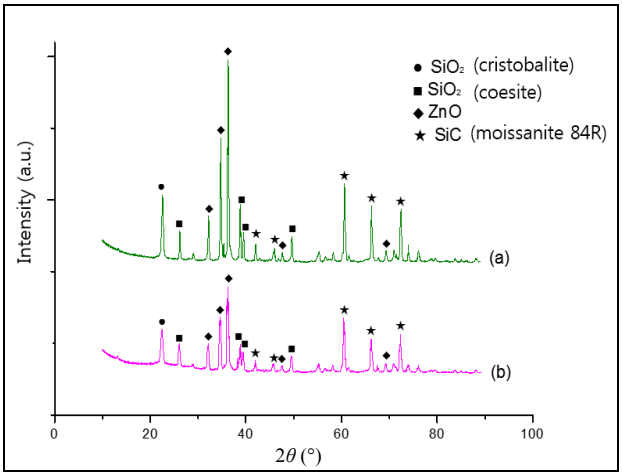


Fig. 1 XRD patterns of (a) doped and (b) dip-coated membrane.

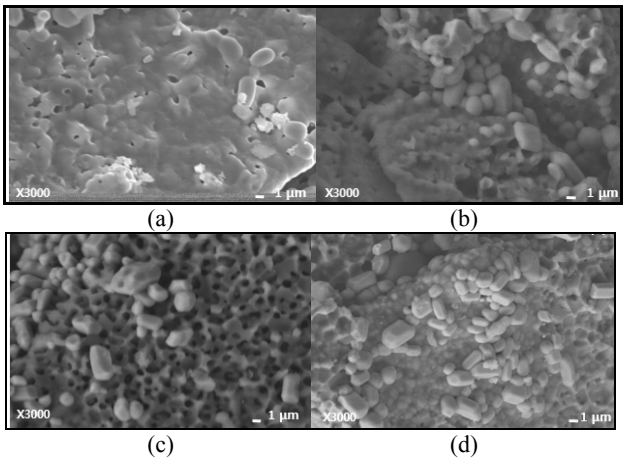
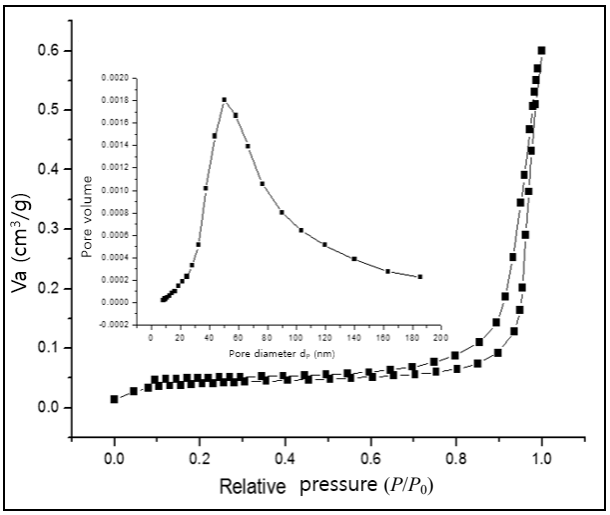
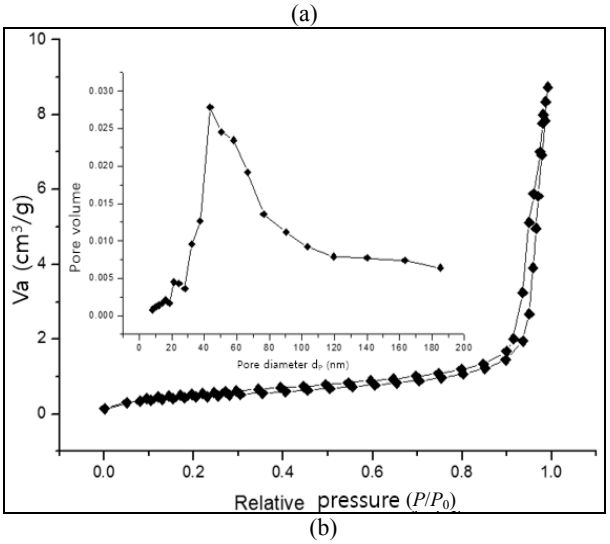


Fig. 2 The surface photographs of (a) doped, (b) dip-coated and cross sectional images of (c) doped, (d) dip-coated membrane with 3,000 magnifications.



(a)



(b)

Fig. 3 N_2 adsorption-desorption isotherms and pore size distributions of (a) doped and (b) dip-coated membrane.

Both doped and dip-coated membranes of N_2 hysteresis loop were not much different and had similar meso-porous shape in Fig. 3. Those are the typical Ⅳ type of the N_2 hysteresis loop as defined by IUPAC (International Union of Pure and Applied Chemistry) with Barrett-Joyner-Halenda method. This type of hysteresis is associated with cylinder pores or voids. Also, the pore size distribution was slightly narrowed down with similar shape in case of dip-coated one.

Even though the pore size distribution was similar shape, the average pore diameter of the doped and dip-coated one was reduced from 31.38 nm to 23.28 nm, respectively.

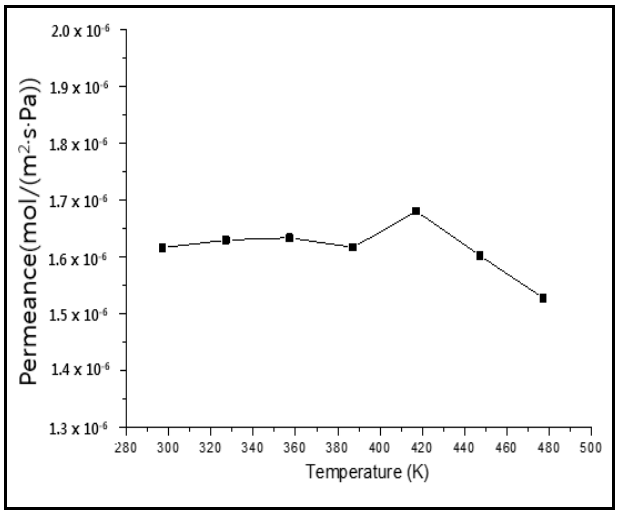


Fig. 4 Hydrogen permeation fluxes of the dip-coated membrane.

Fig. 4 shows hydrogen permeation fluxes only for the dip-coated membrane. These values were slightly decreased as rising temperatures from room temperature to 480 K by following the Knudsen diffusion mechanism, where permeation tendency is in inverse proportion to increasing temperatures [9]. The permeation flux was 1.61×10^{-6} mol/m^2·s·Pa at room temperature. The value of the hydrogen flux could not be obtained from the just doped one since hydrogen was passed through all the temperatures due to large pore size and coarse structure as shown in Fig. 2.

4. Conclusions

SiO_2 dip-coated onto ZnO-SiC composite membrane with meso-porous structure was prepared by doping and further dip-coating process with use of simple and pressure less method. The dip-coating process could control porosity by formation of nano-sized SiO_2 particles and also reinforce binding properties between hydrophobic surface of ZnO and the presented SiO_2 of the SiC surface. Since property and morphology changes can affect the affinity of the permeating species against the pore walls, finally gas permeability of the membrane was affected. More studies with changing factors and also including electro-chemical properties are needed to apply for gas purification and separation membrane.

Acknowledgments

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