

Preparation and CO₂ Permeation Properties of Carbonate-Ceramic Dual-Phase Membranes

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Abstract : In recent years, the carbon dioxide (CO₂) separation technology is required in terms of the reduction of emission of global warming gases and the efficient use of fossil fuels. Since the emission amount of CO₂ gas occupies the large part of greenhouse effect gases, it is considered that CO₂ have the most influence on global warming. Therefore, we need to establish the CO₂ separation technologies with high efficiency at low cost. In this study, we focused on the membrane separation compared with conventional separation technique such as distillation or cryogenic separation. In this study, we prepared carbonate-ceramic dual-phase membranes to separate CO₂ at high temperature. As porous ceramic substrate, the (Pr_{0.9}La_{0.1})₂(Ni_{0.74}Cu_{0.21}Ga_{0.05})O_{4+σ}, La_{0.6}Sr_{0.4}Ti_{0.3}Fe_{0.7}O₃ and Ca_{0.8}Sr_{0.2}Ti_{0.7}Fe_{0.3}O_{3-α} (PLNCG, LSTF and CSTF) were examined. PLNCG, LSTF and CSTF have the perovskite structure. The perovskite structure has high stability and shows ion-conducting doped by another metal ion. PLNCG, LSTF and CSTF have perovskite structure and has high stability and high oxygen ion diffusivity. PLNCG, LSTF and CSTF powders were prepared by a solid-phase process using the appropriate carbonates or oxides. To prepare porous substrates, these powders mixed with carbon black (20 wt%) and a few drops of polyvinyl alcohol (5 wt%) aqueous solution. The powder mixture were packed into stainless steel mold (φφ13 mm) and uniaxially pressed into disk shape under a pressure of 20 MPa for 1 minute. PLNCG, LSTF and CSTF disks were calcined in air for 6 h at 1473, 1573 and 1473 K, respectively. The carbonate mixture (Li₂CO₃/Na₂CO₃/K₂CO₃: 42.5/32.5/25 in mole percent ratio) was placed inside a crucible and heated to 793 K. Porous substrates were infiltrated with the molten carbonate mixture at 793 K. Crystalline structures of the fresh membranes and after the infiltration with the molten carbonate mixtures were determined by X-ray diffraction (XRD) measurement. We confirmed the crystal structure of PLNCG and CSTF slightly changed after infiltration with the molten carbonate mixture. CO₂ permeation experiments with PLNCG-carbonate, LSTF-carbonate and CSTF-carbonate membranes were carried out at 773-1173 K. The gas mixture of CO₂ (20 mol%) and He was introduced at the flow rate of 50 ml/min to one side of membrane. The permeated CO₂ was swept by N₂ (50 ml/min). We confirmed the effect of ceramic materials and temperature on the CO₂ permeation at high temperature.

Keywords : membrane, perovskite structure, dual-phase, carbonate

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