**Effect of Surface States on Oxygen Permeability of La-Sr-Co-Fe Perovskite-Type Oxide Dense Disk**

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## 1. INTRODUCTION

Mixed ionic-electronic conductive (MIEC) Co-based perovskites have been applied to oxygen separation membrane since pioneering work by Teraoka *et. al* in 1985.[1] Since oxygen permeation through MIEC membrane is controlled by surface reactions and/or diffusion of oxide ion, it is important to reveal which is the rate-determining step. In this study, oxygen permeability of $La_{0.1}Sr_{0.9}Co_{0.9}Fe_{0.1}O_{3-\delta}$ (LSCF1991) dense disks with different surface area and roughness were measured, and the contribution of surface reactions in oxygen permeation phenomena was discussed.

## 2. EXPERIMENTAL

After evaporating to dryness of a mixed aqueous solution containing appropriate amounts of metal salts (acetates and nitrates), the residue was precalcined at 350 °C in order to complete the decomposition of metal salts, followed by the calcination at 850 °C for 5h in air. Powder sample thus obtained was compressed into a disk and sintered for 5 h at 1250 °C in air. Both surfaces of sintered disks were polished to nearly 1mm thick by emery paper (from No.80 to No.3000). Laser microscope (LM) was used to observe the disk surface 3-dimensionally and to calculate surface area (Sa) as well as roughness factor. The discoid membrane was welded to a quartz tube with a silver ring. The cell construction of air/membrane/He was used for oxygen permeability measurement. The permeated oxygen in the He side was detected by using TCD gas chromatograph.

## 3. RESULT AND DISCUSSION

LSCF1991 sample disk calcined at 1250 °C was single-phase perovskite-type oxide. Surface states (surface area and roughness factor) of disks were strongly depended on emery papers, that is, polishing by emery paper with lower number (coarser particles) resulted in the rougher surface and higher Sa as shown in Fig. 1. Figure 2 shows temperature dependence of oxygen permeation rate through LSCF1991 membranes polished by different emery papers. Oxygen permeation rate increased with increasing Sa (decreasing the number of emery paper) especially at higher temperature range. In Fig. 3, the rate of oxygen permeation through LSCF1991 was plotted against the normalized surface area (measured Sa/Sa of scanned area by LM). The oxygen permeation rate through the 1 mm-thick membrane ($J(O_2)$) was almost independent of the surface area below 700 °C, indicating that $J(O_2)$ is controlled exclusively by the oxide ion diffusion in the bulk of membrane. Above 750 °C where $J(O_2)$ increased with increasing the surface area, the rates of both diffusion and surface reaction control the oxygen permeation. It has also turned out that the effect of surface area (roughness) on the oxygen permeation flux depends on the thickness of membrane.

**References**

1. Y. Teraoka, H.-M. Zhang, S. Furukawa and N. Yamazoe, *Chem. Lett.*, **1985**, 1743 (1985).

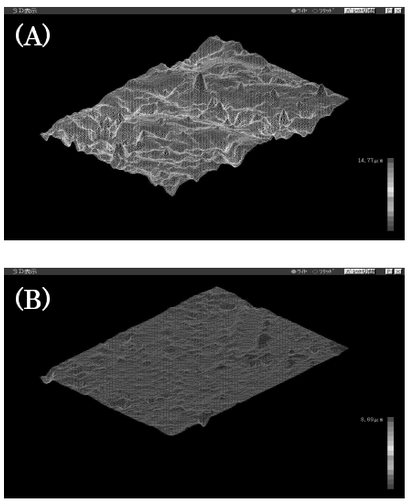


Fig. 1 surface states of the membranes polished by emery papers of (A) No.80 and (B) No.3000.

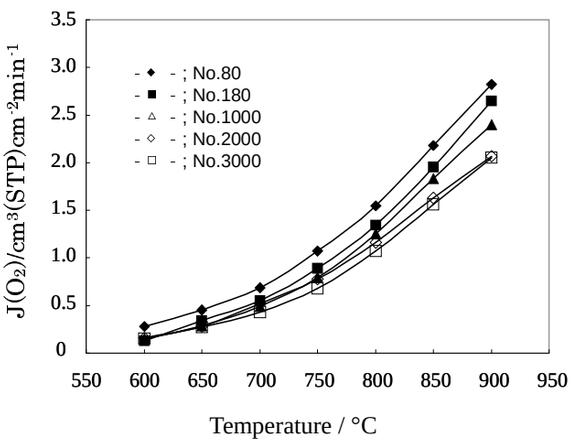


Fig. 2 Oxygen permeation through LSCF1991 membranes polished by different emery papers.

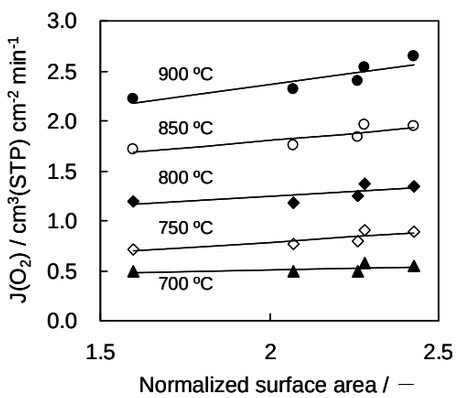


Fig. 3 Rate of oxygen permeation through LSCF1991 as a function of normalized surface area.