FABRICATION OF CERAMIC SEPARATOR WITH CHANNEL BY EXTRUSION

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ABSTRACT

For the purposes of reducing manufacturing costs, we have been trying to fabricate SOFC components by wet processing. But the channels of the separator are usually made by machine tools with high costs. We have now produced this component by extrusion. The content of glue and plasticizer in the clay-like compound for extrusion was important. After sintering, the surface of the component showed a deficiency of chromium.

INTRODUCTION

For solid oxide fuel cells (SOFCs) applications, doped lanthanum chromite (LaCrO3) is the popular material for the separator, because of similar thermal expansion as yttria stabilized zirconia (YSZ) and good chemical stability[1]. In the planar type, the separator requires gas channels for both fuel and oxidant gas. The most popular method for making this utilizes machine tools after sintering. But, the sintered ceramics is hard to manufacture because of brittle fracture. Even if the channels are processed by auto-machine, the manufacturing cost is high. On the other hands, the green body sheet, made by a tape casting[2], is processed as channel structure. This method is effective for the avoidance of brittle fracture owing to flexibility of the green body. Nevertheless, this method still makes the manufacture complex.

Extrusion is usually easy to make the planar type material. For example, this method is one of the most suitable processing method for an uniform shape such as a square pillar and a column. The green body is flexible because the clay-like material has a plastic deformation. If the shape of the dies which are attached to the end of extrusion machine, have channel structure such as a comb, they will be able to provide channels on the sheet. So, for the purpose of reducing of manufacturing costs, we selected the extruding method. We have already done for YSZ electrolyte [3]. In this paper, we discuss the conditions for making the...
ceramic-green sheet with channels by extrusion.

EXPERIMENTAL

Compound preparation

A summary of the preparation of LSCO compound for extrusion is shown in Table 1 and Figure 1. The shape of molds is shown in Figure 2. By optimization of the compound for LSCO sheets, we could minimize dry shrinkage and deformation after processing, and make workability easier during extrusion processing. The compound was prepared from strontium-doped lanthanum chromite (LSCO, Nikkatou, Japan) powder, methyl-cellulose (MC) for binder, glycerol (Gly) for plasticizer, ammonium polyacrylate as dispersant and distilled water as solvent[3].

These components were placed in a kneading-dough and mixed. The volume ratio of binder and plasticizer was 0.15 to 0.25. After kneading, the compound was stored at 4°C for 3 days. The viscosity of slip (water and ceramic powder) was adjusted by addition of dispersant. The value of viscosity was measured by a viscometer (Tokyo Sokki).

The green extruded LSCO sheet had a width of 80 mm and a thickness of 2 mm. It had channels after extruding with comb-like dies.

Sintering LSCO sheet and its properties

The green LSCO sheet was cut, dried at room temperature and plastic agents were burned out in air, and then they were fired at 1450 to 1600°C for 4 hours. The fired LSCO shrunk between 8 and 12%, at 1450 to 1600°C, respectively. We determined their relative densities by the Archimedes method using kerosene (Figure 4). A scanning electron microscopy (SEM) photograph of the cross section was taken for observation of the LSCO surface (Figure 5). An electron probe micro analyzer (EPMA) determined the concentration of chromium.

RESULTS AND DISCUSSIONS

The LSCO green body is shown in Figure 3. The height of channel, which was designed at 1.0 mm in dies, was 0.8 mm. The pressure of the extruding machine was 10 kgf/cm². The relative densities, after firing at 1500°C, are shown in Figure 6. Under 15 vol% of Methylcellulose (MC) content, the clay-like compound could not hold its own shape. On the other hand, at over 35 vol% of MC content, the compound was too hard to be extruded. Under 10 vol% of glycerol (Gly) content, the green body after extrusion had many
cracks. On the contrary, at over 35 vol% of Gly content, the roughness of the surface increased. We decided MC 20 vol%, Gly 15 vol% (20-15) as the optimum condition because this gave significantly high relative density. Relationship between the sintering temperature and relative density is shown in Figure 7. The relative density at 1450°C was slightly higher than that at 1500°C. Generally, the relative density is higher at higher sintering temperature. But, in this study, the relative density at 1500°C had the minimum value.

According to Tai [3], the evaluation of chromate gas by the following reaction makes porosity of samples higher:

\[
\text{Cr}_2\text{O}_3(s) + \frac{3}{2}\text{O}_2 \rightarrow 2\text{CrO}_3 (g)
\]

To confirm this mechanism, the concentrations of chromium and lanthanum were determined at cross-sections by EPMA mapping (Figure 8). The concentration of lanthanum was distributed uniformly. On the other hand, that of chromium was distributed to the inside. The surface of the sample showed a deficiency of chromium, and its depth was about 10 micrometers.

The Figure 9 shows the relationship between the sintering temperature and the grain growth at 1450°C and 1600°C. It seems that the grain size increased from 0.5 micron to 2 micron with increasing temperature. But, the pore size at 1600°C was larger than that at 1450°C. This made the relative density at higher temperature lower.

**CONCLUSIONS**

The LSCO ceramic separators which were made by extrusion process, were found suitable for the planar type SOFC. By using dies with rectangular shapes, channel structure could be made. In this process, it is important for the extrusion that the contents of compound are optimized.

**REFERENCES**

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Table 1. Summary of Studies on the preparation of Compounds

| Component | Operations |
|-----------|------------|
| 30mol% Strontium -doped Lanthanum Chromite | Powder |
| Dispersant | Deflocculation of powder |
| Methylcellulose (MC) | Binder |
| Glycerol (Gly) | Plasticizer |
| Distilled Water | Solvent |

Figure 1. Flow chart of the Extruding process for the LSCO sheet
Figure 2. Schematic of die for extrusion

Figure 3. Green body of LSCO plate

Figure 4. Relative densities of LSCO (fired at 1500°C)
Figure 5. The cross sectional photograph of a fired LSCO plate by SEM

Sintering temperature at 1600°C

- Filled to make green bodies with channels
- Opened to make green bodies with channels

Optimum condition

Figure 6. The optimum conditions for extrusion
Figure 7. The dependence of density on sintering temperatures

Figure 9. SEM micrographs of fired LSCO plates
Figure 8. EPMA mapping of lanthanum and chromium