Preparation and Analysis of Yttria-Stabilized Zirconia Thin Film by CVD-EVD Method

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1. Introduction

Yttria stabilized zirconia (YSZ) is used for solid oxide electrolyte fuel cells because of its high ionic conductivity. It is desirable for the thickness of the electrolyte film to be approximately 10 to 50 μm in order to reduce the internal resistance of the cell. It's also very important for the yttria concentration to be approximately 8 to 10 mol%. The chemical vapor deposition - electrochemical vapor deposition (CVD-EVD) method is the best technique for depositing YSZ films which are free of porosity and of uniform thickness on porous substrates, because it uses an electrochemical process.1-3)

In this study, the authors investigated the distribution of yttria through YSZ films deposited by the CVD-EVD process. To measure the yttria concentration, secondary ion micro-analysis (SIMS), X-ray micro-analysis (XMA) and inductively coupled plasma atomic emission spectroscopy (ICP-AES) methods were used. The results showed that the experimental results were consistent with each observation.

2. Experimental Procedures

Films were deposited using the CVD-EVD apparatus shown in Fig. 1. The ZrCl₄ and YCl₃ vapor chambers and the reaction chamber had their temperature controlled independently. Vaporized gas was transported to the reaction chamber by Ar carrier gas.

Substrates used were calcia-stabilized zirconia tubes coated with La(Sr)MnOₓ of approximately 15mm in outer diameter and 300mm in length.

SEM observation was carried out using Hitachi S-4000, while the observations by XMA and SIMS were conducted using Shimazu EPMA-8705 and ATOMICA 6500 respectively. YSZ pellets of known density were used for reference. ICP-AES measurements were carried out to confirm the absolute concentration of yttria. The deposited films were detached from the substrates with HCl and dissolved into HCl+HClO₄ before ICP-AES measurement.

3. Results and Discussion

YSZ films were deposited using the CVD-EVD process at the reaction temperature of 1223 K. When the CVD process was completed after several minutes, YSZ films were grown by the EVD process for 8 hours. A SEM photograph of the cross section of the sample is shown in...
Fig. 2. It can be seen that the film is dense and uniform in thickness. Fig. 3 shows XRD spectrum; the crystal structure was found to be cubic.

Fig. 4 shows the yttria distribution through the YSZ film, which was obtained by means of SIMS and XMA. The yttria concentrations obtained by these two methods provide nearly identical results, indicating that the measurements were valid. In Fig. 4, it is shown that the Y2O3 concentration was uniform throughout the film at approximately 10 mol%.

These films were detached from the substrates with HCl, dissolved with HCl-HClO4, and ICP-AES measurements were performed. The resulting concentration is shown in Table 1, in which the values obtained by integrating the experimental results in Fig. 4 are also shown. The results obtained by SIMS and XMA were confirmed by ICP-AES result.

SIMS is considered to be a better analytical method than XMA, because XMA is influenced by such sample conditions as surface roughness more than SIMS.

4. Conclusion

YSZ films were grown on porous La(Sr)MnOx substrates using the CVD-EVD process. The film thickness was about 10 μm. The crystal structure was cubic.

The yttria concentration distribution through the film was investigated using SIMS and XMA. It was found that the concentration was nearly uniform and approximately 10 mol%.

The experimental results indicated that SIMS observation was a good analytical method for YSZ films.

References