

# Fabrication of an Electrochemical Cell by a Rare Earth Stannate Pyrochlore Catalyst and Characterization of NO<sub>x</sub> Gas Decomposition

Sarohan Park and Jooho Moon

Department of Ceramic Engineering, Yonsei University, Seoul 120-749, Korea

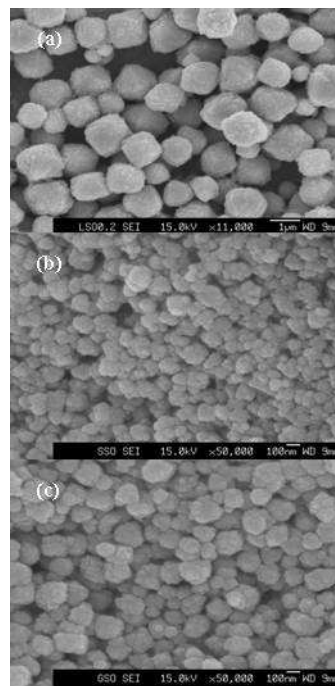
Nowadays air pollution is a critical problem to human. Especially, the pollution caused by automobiles is very harmful to us because their exhaust gases are emitted close to the residential area. Three-way catalytic converter works well for purifying exhaust gases such as hydrocarbons (HC), nitrate oxide (NO<sub>x</sub>) and carbon monoxide (CO). However this system is ineffective for both lean-burn and diesel engines since NO<sub>x</sub> decomposition is difficult in the presence of excess oxygen. To solve this problem, an electrochemical cell in which the electrocatalyst deposited on the oxygen conducting electrolyte serves as a cathode similar to solid oxide fuel cell (SOFC) has been applied to decompose NO<sub>x</sub> under oxidizing atmosphere.

Previous studies have demonstrated that NO<sub>x</sub> can be decomposed into N<sub>2</sub> while the oxygen on the cathode is forced to transport through solid electrolyte to the anode. In such electrochemical cells, the noble metals such as Pd and Pt are generally used for electrode materials. But they are too expensive and suffer from high temperatures instability. In this work, we have fabricated the electrochemical cell using composite electrode materials based on rare earth stannate pyrochlores, Ln<sub>2</sub>Sn<sub>2</sub>O<sub>7</sub> (Ln = La, Sm and Gd). Rare earth stannate pyrochlores exhibit good catalytic activity and high thermal stability, which make them promising when used as a catalytic promoter in the electrode for electrochemical decomposition of NO<sub>x</sub>.

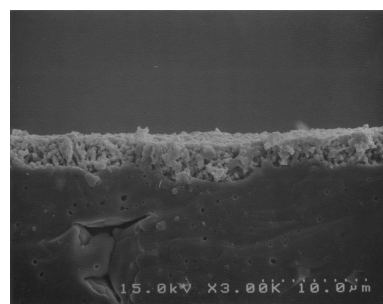
The rare earth stannate pyrochlore were synthesized by hydrothermal method at 200°C for 36 h. The mean particle sizes were in the range of 0.59 – 0.25 μm, while the surface areas were approximately 14 m<sup>2</sup>/g regardless of the type of rare earth elements. The results of XRD and ICP analysis indicated that the product powders were the well-crystallized single phase pyrochlore with a relatively accurate stoichiometry. SEM observation revealed that the synthesized rare earth stannates have a relative narrow size distribution and uniform shape with a rough surface structure as shown in Fig. 1. Composite electrode was produced by a screen printing on yttria stabilized zirconia (YSZ) electrolyte disk using the paste of a mixture of the synthesized rare earth stannate pyrochlores and YSZ powders and were sintered at 1100 ~ 1400°C for 4h in air. Figure 2 shows a cross-sectional view of the microstructure of the composite electrode deposited on YSZ disk after sintering at 1400°C for 4 h. Pt mesh for current collector was contacted to the electrode surface and Pt wire was connected to Pt mesh by Pt paste. The electrochemical cell was mounted on alumina tube equipped in a furnace. The reactant gas of NO 0.1% and O<sub>2</sub> 2% was introduced to the electrochemical cell at 700°C while applying a direct current to the cell by galvanostat. NO decomposition over the electrochemical cells was monitored by on-line gas chromatography and NO<sub>x</sub> analyzer.

Influences of the microstructure and composition of the composite electrode on the decomposition of NO were investigated. Spatial distribution and connectivity of the catalytic active phase and electrochemical active phase play an important role in determining decomposition of NO by the electrochemical cell. Rare earth stannate pyrochlores provide a catalytic active site for NO on which NO is adsorbed and dissociated. YSZ serves as a oxygen

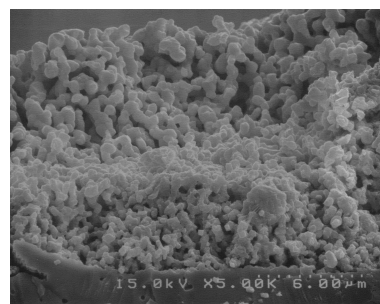
conducting pathway through the electrode layer prior to reaching at the electrolyte. The composite electrodes sintered at temperatures ranging from 1100°C to 1400°C exhibit different decomposition rates since the connectivity of both phases are influenced by sintering temperature. Spatial distribution of the pyrochlore catalyst and YSZ also contributes to NO decomposition. Better catalytic activity was observed in the functionally graded electrode in which the pyrochlore catalyst is dominantly present in the top layer of the composite electrode than the one in which both phases are uniformly distributed throughout the layer. NO decomposition rate over the engineered electrode sintered at 1100°C was 60-70% at 0.35 A/cm<sup>2</sup>



**Figure 1.** SEM micrographs of (a) La<sub>2</sub>Sn<sub>2</sub>O<sub>7</sub>, (b) Sm<sub>2</sub>Sn<sub>2</sub>O<sub>7</sub> and (c) Gd<sub>2</sub>Sn<sub>2</sub>O<sub>7</sub> particles synthesized by a hydrothermal method at 200°C.



**Figure 2.** SEM micrographs of cross-section of the catalyst electrode on the YSZ disk.



**Figure 3.** SEM micrographs of cross-section of the catalyst electrode of different layer.