

## NOTE

### Study of Redox Properties of $Ce_xPr_{1-x}O_{2.8}$ Mixed Oxides

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$Ce_xPr_{1-x}O_{2.8}$  mixed oxide was prepared by citrate sol-gel method and calcined at 900 °C for 4 h. The formation of the solid solutions can improve the redox property of the  $Ce_xPr_{1-x}O_{2.8}$  mixed oxides.  $Ce_{0.5}Pr_{0.5}O_{2.8}$  sample shows a key high capability for redox properties.

**Keywords:**  $Ce_xPr_{1-x}O_{2.8}$  mixed oxide,  $H_2$ -TPR, XRD.

Cerium dioxide ( $CeO_2$ ) provides a major function in the three-way catalysts (TWC), because it has multiple effects on the catalyst state and performance<sup>1</sup>. However,  $CeO_2$  has a major drawback, *i.e.*, significant deactivation of the redox couple, which occurs due to the sintering of  $CeO_2$  particles and the reactions between  $CeO_2$  and  $\gamma-Al_2O_3$  support or active precious metals when it is performed at high temperatures under the driving conditions<sup>2,3</sup>. Consequently the OSC decreases and the activity of the catalysts declines. In order to eliminate these drawbacks, a great number of  $CeO_2$ -based mixed oxide systems<sup>4-8</sup> such as Ce-Zr-O, Ce-Hf-O, Ce-La-O, Ce-Rh-O, Ce-Ti-O, Ce-Y-O, Ce-Pr-O, *etc.* have been conducted. Many studies have shown that the redox properties can be considerably enhanced if additional elements are introduced into the  $CeO_2$  lattice and solid solutions are formed. The redox behavior of  $Ce_xPr_{1-x}O_{2.8}$  mixed oxides is the important feature, because most catalysts used for the oxidation reaction are under a redox cycle. So the present work devoted to preparing a series of  $Ce_xPr_{1-x}O_{2.8}$  mixed oxides by a sol-gel method and to investigating their reduction properties.

**Preparation of  $Ce_xPr_{1-x}O_{2.8}$  mixed oxide:** A series of oxides with the general formula  $Ce_xPr_{1-x}O_{2.8}$  ( $x = 0, 0.3, 0.5, 0.7, 0.8, 0.9, 1$ ) were prepared by citrate a sol-gel method<sup>8</sup>. An appreciate amount of solid power of citric acid was added to each pre-mixed nitrate solution of cerium and praseodymium. The mixture was then vaporized under stirring, until it was slowly becoming transparent gel. The gel that was transparent and viscous was dried at 100 °C in an oven and obtained a solid. The solids were finally calcined at 900 °C for 4 h to obtain yellow powders.

**Temperature programmed reduction (TPR):** The redox properties of  $Ce_xPr_{1-x}O_{2.8}$  mixed oxides were measured by means of temperature-programmed reduction technique. Each  $Ce_xPr_{1-x}O_{2.8}$  mixed oxides (50 mg) were placed in a quartz reactor, which was connected to a conventional TPR apparatus. The sample was reduced in a flow of  $H_2-N_2$  (5: 95) mixed gas at a heating rate of 20 °C/min up to 900 °C.

**X-ray diffraction:** X-ray diffraction patterns were collected on a PHILIPS PW3040/60 powder diffractometer using  $CuK\alpha$  radiation. The working voltage of the instrument was 40 kV and the current was 40 mA. The intensity data were collected at 25 °C in a  $2\theta$  range from 20 to 100 ° with a scan rate of 0.1 %.

Fig. 1 shows the temperature programmed reduction (TPR) profiles of the  $Ce_xPr_{1-x}O_{2.8}$  mixed oxides calcined at 900 °C. There is one large reduction peak at about 630 °C for pure  $Pr_6O_{11}$ . XRD experiment of the fresh  $Pr_6O_{11}$  and the  $Pr_6O_{11}$  samples after reduction were showed in Fig. 2. From Fig. 2, it can be seen that the principal component of the samples after reduction is  $Pr_2O_3$ . So it can be see that  $Pr_6O_{11}$  is transformed to  $Pr_2O_3$  completely after the temperature progress reduction experiment. Then the reduction process is  $PrO_{1.83} \rightarrow PrO_{1.5}$ . For pure  $CeO_2$ , there is no obvious reduced peak at lower temperature, but there is a weak desorption peak above 860 °C which is ascribed to the reduction of the bulk<sup>9</sup>.

For  $Ce_xPr_{1-x}O_{2.8}$  mixed oxides, the reduction peak decreases in the experimental region. When  $x < 0.5$  the reduction temperature shifts to lower region, as the increases of the  $x$  value. When  $x > 0.5$  the reduction temperature have a little change as the increases of the  $x$  value. There is a lowest reduction

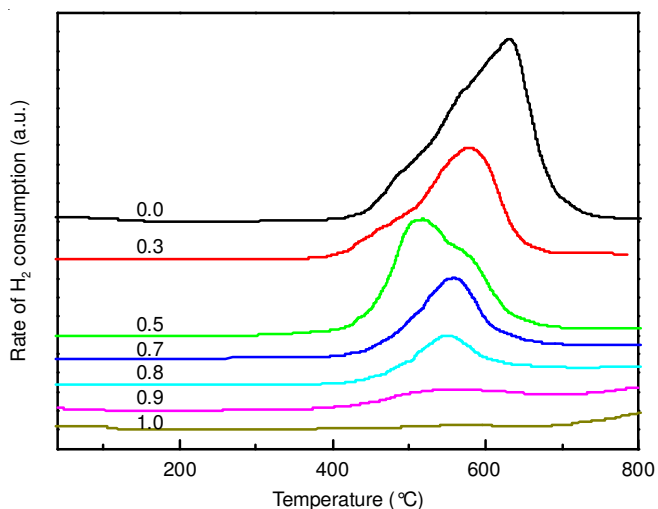


Fig. 1. Temperature programmed reduction profiles of  $Ce_xPr_{1-x}O_2$  mixed oxides calcined at 900 °C

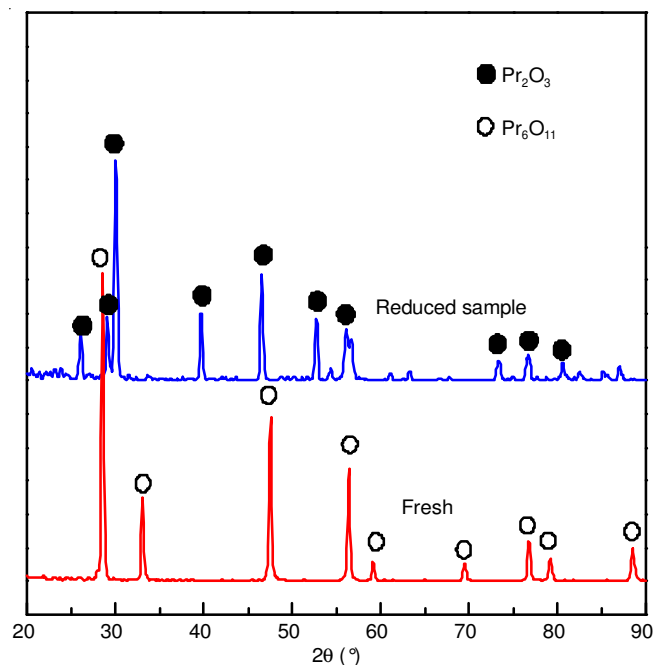


Fig. 2. XRD patterns of the fresh  $Pr_6O_{11}$  and reduced  $Pr_6O_{11}$

temperature at about 510 °C when  $x = 0.5$ . So it can be seen that  $Ce_{0.5}Pr_{0.5}O_{2-\delta}$  sample shows a high capability for redox properties. The formation of the solid solutions can improve the redox property of the  $Ce_xPr_{1-x}O_{2-\delta}$  mixed oxides.

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