

Short communication

# Preparation of Pd/(Ce<sub>1-x</sub>Y<sub>x</sub>)O<sub>2</sub>/γ-Al<sub>2</sub>O<sub>3</sub>/cordierite catalysts and its catalytic combustion activity for methane

Hengcheng Liao\*, Miaomiao Liu, Peiyuang Zuo

School of Materials Science and Engineering, Southeast University, Jiangsu Key Laboratory for Advanced Metallic Materials, Nanjing 211189, China



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## ABSTRACT

A series of (Ce<sub>1-x</sub>Y<sub>x</sub>)O<sub>2</sub> ( $x = 0, 0.15, 0.35, 0.5$ ) coatings on γ-Al<sub>2</sub>O<sub>3</sub> pre-coated cordierite honeycomb were prepared by sol-gel method, and then palladium was loaded by aqueous solution impregnation deposition with Pd(NO<sub>3</sub>)<sub>2</sub> as precursor. The structure and morphology of samples were evaluated and the catalytic combustion activity for methane was also discussed. (Ce<sub>1-x</sub>Y<sub>x</sub>)O<sub>2</sub> synthesized by sol-gel has a single-phase cubic fluorite structure. Increasing the Y/Ce ratio can significantly improve the inner surface morphology of the honeycomb channels and also the coating mechanical stability, and leads to a considerable improvement in the catalytic activity of the prepared catalysts for methane.

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## 1. Introduction

With increasingly stringent requirements for protecting our environment, catalytic combustion of nature gas (methane) has attracted more and more attention [1]. Monolithic catalysts are widely used for catalytic combustion of methane and other organics. As a supporter, cordierite ceramic honeycomb (abbreviated to COR) has very small specific surface area (usually <1 m<sup>2</sup>/g) [2], thus γ-Al<sub>2</sub>O<sub>3</sub> coating is usually used as a transition layer to create a higher surface area [3], however, at high temperature, it is prone to transform into the θ-phase, finally into the α-form, which has a low specific surface area.

In order to prevent the transformation of γ-Al<sub>2</sub>O<sub>3</sub> and also to enhance catalytic activities, some rare-earth element oxides (such as CeO<sub>2</sub>, La<sub>2</sub>O<sub>3</sub>, and Y<sub>2</sub>O<sub>3</sub>) are commonly used as additives [4–6]. CeO<sub>2</sub> has been widely studied in recent years for catalytic application. CeO<sub>2</sub> has many advantages, such as: strong ability to store/release oxygen [7,8], high thermal and structure stabilities [9], and good ability to disperse the noble metal active component [10,11]. Doping divalent or trivalent ions in CeO<sub>2</sub> could improve its oxygen storage capacity [12,13]. Addition of Y to (Ce,Zr)O<sub>2</sub> solid solution could improve oxygen vacancies and promote the reduction of Ce<sup>4+</sup> [14], and also improved the reduction–reoxidation properties of the active PdO species [15].

In this paper, (Ce<sub>1-x</sub>Y<sub>x</sub>)O<sub>2</sub> solid solution coatings were prepared by sol-gel method. The effect of Y/Ce ratio on the phase constitution, surface morphology, and mechanical stabilities was investigated and the catalytic performance of the prepared Pd/(Ce<sub>1-x</sub>Y<sub>x</sub>)O<sub>2</sub>/γ-Al<sub>2</sub>O<sub>3</sub>/COR catalysts for methane was also discussed.

## 2. Experimental

2.1. Preparation of (Ce<sub>1-x</sub>Y<sub>x</sub>)O<sub>2</sub>/γ-Al<sub>2</sub>O<sub>3</sub>/COR

A commercial honeycomb ceramic was cut into cuboid samples (6 mm × 6 mm × 30 mm) which were pretreated by conventional process. Then they were immersed in the γ-Al<sub>2</sub>O<sub>3</sub> transition sol for 5 mins and the excess γ-Al<sub>2</sub>O<sub>3</sub> sol inside the honeycomb channels was blown off, and then they were dried at 120 °C for 2 h and roasted at 550 °C for 2 h in a muffle furnace with flowing air. The amount of the loaded γ-Al<sub>2</sub>O<sub>3</sub> coating can reach 12 wt.% – 15 wt.% after double repeating. The prepared samples above were labeled as γ-Al<sub>2</sub>O<sub>3</sub>/COR.

Ce and Y nitrates were used as Ce- and Y- precursors, and the mole ratios of Ce and Y was 1:0, 0.85:0.15, 0.65:0.35, and 0.5:0.5. The mixed aqueous solution of Ce- and Y- nitrates and citric acid was prepared with distilled water. Citric acid was added as a complexing agent in 1:2 M ratio with respect to the sum of the metal salts. The mixed solution were heated at 70 °C in a water bath and constantly stirred until a stable sol was obtained. The γ-Al<sub>2</sub>O<sub>3</sub>/CORs were impregnated in this sol for 5 min. The excess sol inside the channels need blow off.

\* Corresponding author at: School of Materials Science and Engineering, Jiangning Campus of Southeast University, Nanjing 211189, China.  
E-mail address: [hengchengliao@seu.edu.cn](mailto:hengchengliao@seu.edu.cn) (H. Liao).

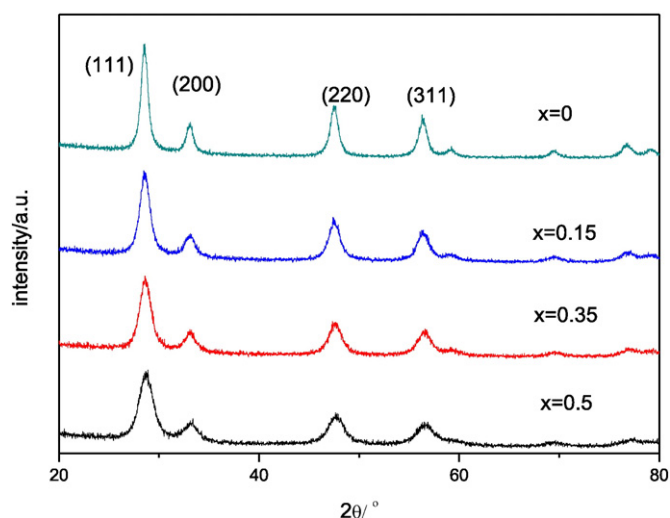


Fig. 1. XRD patterns of  $(\text{Ce}_{1-x}\text{Y}_x)\text{O}_2$  ( $x = 0, 0.15, 0.35, 0.5$ ) powders after calcination.

Table 1

Lattice constant and crystallite size of  $(\text{Ce}_{1-x}\text{Y}_x)\text{O}_2$ .

$x$	Lattice constant $a/\text{nm}$	Crystallite size $D/\text{nm}$
0	0.5413	11.8
0.15	0.5402	9.5
0.35	0.5395	8.8
0.5	0.5378	8.4

Subsequently, they were dried at 120 °C for 2 h in air and roasted at 550 °C for 2 h. This processed sample is labeled as  $(\text{Ce}_{1-x}\text{Y}_x)\text{O}_2/\gamma\text{-Al}_2\text{O}_3/\text{COR}$ .

## 2.2. Preparation of catalysts

Catalysts were prepared by a classical aqueous solution impregnation method, in which  $\text{Pd}(\text{NO}_3)_2$  is as precursors. The  $(\text{Ce}_{1-x}\text{Y}_x)\text{O}_2/\gamma\text{-Al}_2\text{O}_3/\text{COR}$ s were completely impregnated in the Pd-solution for 30 min, and the excess aqueous solution inside the channels was blown off. Then they were dried at 120 °C for 2 h in air and roasted at 450 °C for 2 h. The final sample is labeled as  $\text{Pd}/(\text{Ce}_{1-x}\text{Y}_x)\text{O}_2/\gamma\text{-Al}_2\text{O}_3/\text{COR}$ .

## 2.3. Characterization

The XRD patterns of  $(\text{Ce}_{1-x}\text{Y}_x)\text{O}_2$  dried gel powder were collected on a Bruker apparatus (D8-Discover), using  $\text{Cu K}\alpha$  radiation ( $\lambda = 1.5418 \text{ \AA}$ ). The X-ray tube was operated at 40 kV and 30 mA and the scanning rate was 0.02°/step in the range of  $20^\circ \leq 2\theta \leq 90^\circ$ . The specific surface area was measured by  $\text{N}_2$  adsorption at 77 K on ASAP 2000 M with the Brunauer–Emmet–Teller (BET) method. The surface morphology of the  $(\text{Ce}_{1-x}\text{Y}_x)\text{O}_2$  coating was examined by XL30 environmental SEM.

Ultrasonic oscillating test was performed, to evaluate the cohesive strength of coating on the cordierite substrate, in a KQ-250B ultrasonic bath with a power of 220 W for 30 min. Before test, the dry sample was weighted as  $m_{\text{before}}$ , and after test, the sample was dried again, and then weighted as  $m_{\text{after}}$ . The mass losses of the coating was calculated using  $\Delta W\% = (m_{\text{before}} - m_{\text{after}}) / m_{\text{before}} \times 100\%$ .

Inductively coupled plasma-optical emission spectroscopy (ICP-OES) was performed to measure the loading amount of palladium, cerium and yttrium in the prepared monolithic catalysts.

## 2.4. Catalytic activity evaluation

The catalytic combustion of methane on the  $\text{Pd}/(\text{Ce}_{1-x}\text{Y}_x)\text{O}_2/\gamma\text{-Al}_2\text{O}_3/\text{COR}$  catalysts was carried out in a conventional fixed-bed flow quartz micro-reactor (length = 400 mm, i.d. = 10 mm). A gas mixture

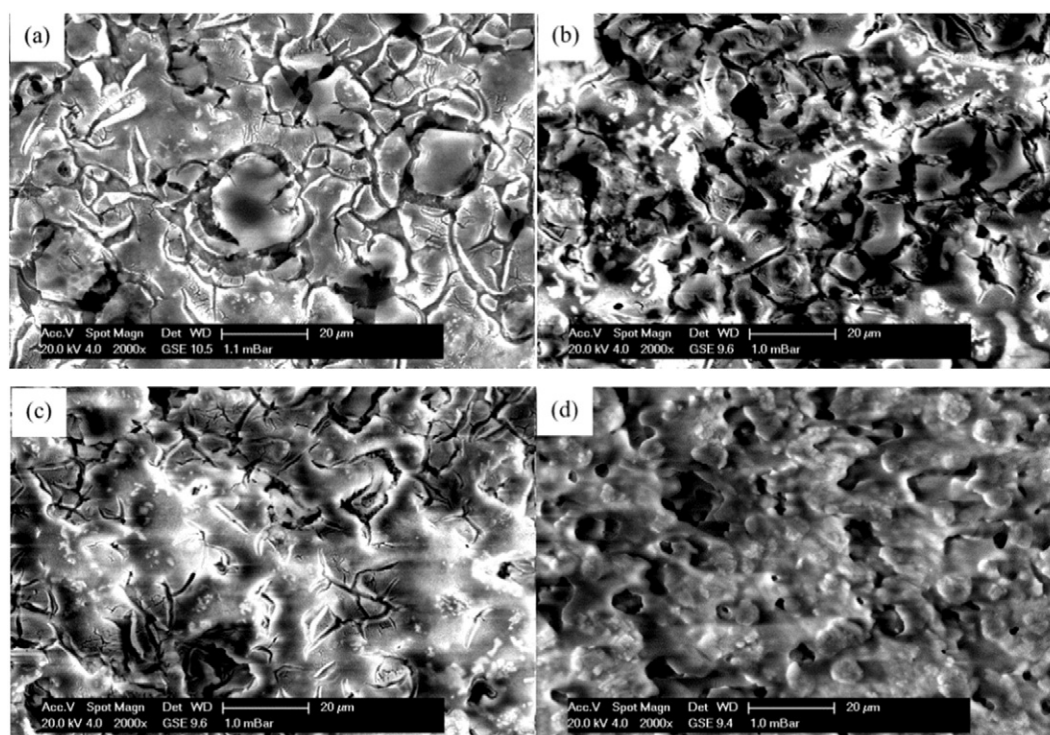


Fig. 2. The inner surface morphology of  $(\text{Ce}_{1-x}\text{Y}_x)\text{O}_2/\gamma\text{-Al}_2\text{O}_3/\text{COR}$ s (a)  $x = 0$ ; (b)  $x = 0.15$ ; (c)  $x = 0.35$ ; (d)  $x = 0.5$ .

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