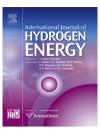


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## Highly dispersed Ru on K-doped meso–macroporous SiO<sub>2</sub> for the preferential oxidation of CO in H<sub>2</sub>-rich gases

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#### ABSTRACT

In this work, highly dispersed Ru nanoparticles which had a uniform small nanoparticle size were supported on K-promoted meso-macroporous  $SiO_2$  by using the simple impregnation method. The effect of the size of Ru nanoparticle on the catalytic performance for the preferential oxidation of CO (CO-PROX) in H<sub>2</sub>-rich gases was investigated. Meanwhile, the related mechanism on size effect was discussed. The catalysts were characterized by using techniques of transmission electron microscopy, temperature-programmed reduction and CO-chemisorption. The results indicate that the K-promoted Ru/SiO<sub>2</sub> catalyst with the size of metal Ru particles at about 7 nm showed obviously higher turnover frequency (TOF) than that of K-Ru/SiO<sub>2</sub> with smaller size of Ru particles of around 2 nm. As for oxidizing CO to CO<sub>2</sub> on specific weight of ruthenium, the catalyst with the smaller size of metal Ru exhibited better performance owing to its much higher specific surface area of metal Ru. The catalyst with the smaller size of Ru nanoparticles showed much better methanation formation resistance for CO and CO<sub>2</sub>. The K-promoted and highly dispersed Ru on SiO<sub>2</sub> exhibited excellent activity and selectivity for the CO-PROX reaction.

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#### Introduction

The fuel of hydrogen for fuel cells is generally produced by using steam reforming of hydrocarbons and water gas shift reactions, which contains around 1 vol.% of CO [1]. However, trace amount of CO leads to the poisoning of the platinum anode of fuel cells [2]. Thus, the carbon monoxide is required to be removed to less than 10 ppm or 100 ppm for the COresistant anodes. The preferential oxidation (PROX) of CO has been supposed to be the most promising method [3]. The reported catalysts for CO-PROX mainly include noble metal catalysts, such as Ru [4], Rh [5], and Pt [6], base metal oxide catalysts, such as CuO–CeO<sub>2</sub> [7] and CoOx-CeO<sub>2</sub> [8], and gold catalysts [9]. Among them, the nano Ru catalyst is one of the most potential candidates [10]. Ru catalyst supported on mesoporous silica displays a high activity and selectivity for CO-PROX. For example, Wang et al. [11] studied the catalysts of Ru supported on different mesoporous SiO<sub>2</sub>, including MCM-41, MCM-48, SBA-15 and KIT-6 for the CO-PROX, and pointed out that the catalytic performance of the catalysts

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highly related to the dispersion of ruthenium, the porosity of the support, the reducibility of ruthenium species and the interaction between Ru and the support. Among catalysts studied, Ru/MCM-41 presented the highest catalytic activity. Previous studies in our lab have found that K-promoted Ru/ meso-macroporous  $SiO_2$  exhibited excellent activity for CO-PROX [12].

The effect of metal nanoparticle size on catalytic activity has aroused considerable interests in terms of academic and practical viewpoints. Chen et al. [13] reported that the

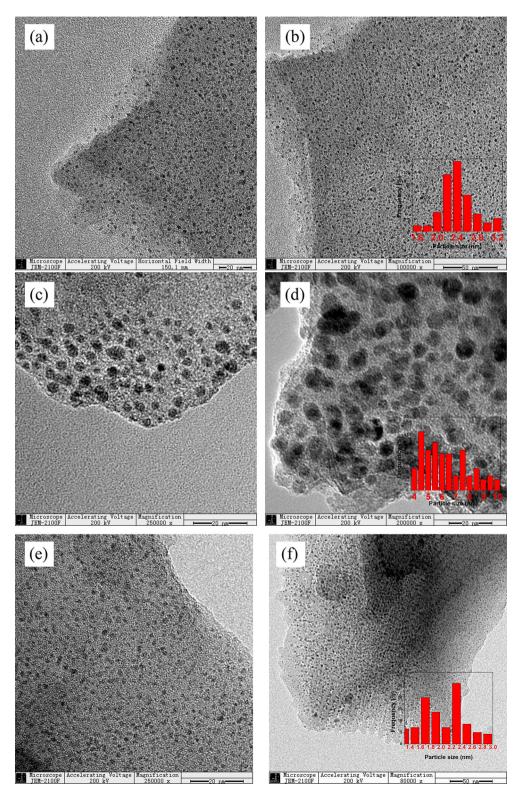


Fig. 1 – TEM images of (a, b) Ru/SiO<sub>2</sub>(SI); (c, d) K-Ru/SiO<sub>2</sub>(COI); (e, f) K-Ru/SiO<sub>2</sub>(SEI); (g, h) K-Ru/SiO<sub>2</sub>(SC), and a corresponding Ru particle size distributions; Scale bar: 20 nm for (a, c, d, e, g), 50 nm for (b, f, h).

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