

Short communication

Combustion synthesis of copper catalysts for selective CO oxidation

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Received 5 December 2007; received in revised form 22 December 2007; accepted 31 December 2007

Available online 4 January 2008

Abstract

Copper catalysts supported on ceria, zirconia and niobia were prepared by combustion method with urea, containing a CuO loading of 6 wt.%, and tested on selective oxidation of CO. The characterization of the samples by X-ray diffraction (XRD) presented the formation of solid solution on CuO–CeO₂ catalyst and a change in crystalline structure of the support with copper insertion on ZrO₂ and Nb₂O₅ catalysts. The analysis of temperature-programmed reduction (TPR) revealed different interaction degrees of copper with the supports, with reduction peaks between 222 and 390 °C. The temperature-programmed desorption of CO (TPD-CO) profiles showed formation of CO₂ and H₂ only for the ceria and zirconia catalysts. In relation to the catalytic tests, the CuO–CeO₂ catalyst presented the best performance, with CO conversion of 95% at 150 °C up to 45 h on stream, and CO₂ selectivity of 55%.

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Keywords: Copper; Ceria; Zirconia; Niobia; Selective oxidation; Carbon monoxide

1. Introduction

Selective CO removal from streams containing an excess of H₂ is the current challenge in heterogeneous catalysis research especially for preparation of H₂ suitable for proton exchange membrane fuel cell (PEMFC). The PEM fuel cells have attracted considerable attention for its application in transportation due to its low operation temperature, high efficiency and compactness [1,2]. Hydrogen is usually produced through steam reforming or partial oxidation of hydrocarbons or alcohols, followed by water–gas shift (WGS) reaction [3]. The effluent gases from such a process contain about 40–75% H₂, 15–25% CO₂, 0.5–2% CO, a few% H₂O and N₂ [4,5]. However, for use in PEMFC, CO levels have to be reduced to below 100 ppm and, preferably, 10 ppm, because of the poisoning of Pt–Ru electrocatalysts used as anode materials [6].

Several options for CO removal include palladium diffusion membranes, methanation and selective oxidation (SELOX) of CO [2,7]. Among the aforementioned methods, SELOX of CO is regarded as the most cost-effective, simplest and straightforward method. Therefore, the development of selective CO oxidation catalysts has stimulated considerable interest worldwide. The catalyst for the SELOX reaction must meet the requirements of high CO oxidation activity combined with high selectivity, to avoid the undesired H₂ oxidation side reaction. Another important requirement is that these catalysts should have good tolerance for CO₂ and H₂O in the feedstock.

The catalysts for SELOX of CO reported in the literature can be classified basically in three types: (a) noble metal catalysts (Pt, Pd, Rh supported on Al₂O₃, SiO₂ and zeolites) [8–11]. These catalysts exhibit good activity and stability in temperature range 150–250 °C, but the selectivity is not good enough to meet the practical requirements; (b) gold-based catalysts, which were much more active than Pt-group metal catalysts at low temperatures (<100 °C) but not so resistant towards deactivation by CO₂ and H₂O in the feed [12–14]; (c) several kinds of metal oxide catalysts, such as the oxides of Cu, Ni, Co, Mn alone or in combination [15,16]. Among these oxides, the CuO–CeO₂ mixed oxides have been shown as the most promising candidate

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