Materials Letters 113 (2013) 96–99

Contents lists available at ScienceDirect

Materials Letters

journal homepage: www.elsevier.com/locate/matlet

Synthesis, characterisation, and CO oxidation activity of M/Al_2O_3 meso phase catalysts (M=Ce, V, Cu)

N.K. Renuka *, A.V. Shijina [1], A.K. Praveen [1]

Department of Chemistry, University of Calicut, Kerala 673635, India

ARTICLE INFO

Article history:
Received 20 May 2013
Accepted 18 September 2013
Available online 25 September 2013

Keywords:
Porous materials
Texture
Supported metal oxide
CO oxidation

ABSTRACT

Here we compare the ability of V, Ce, and Cu supported mesoporous alumina catalysts in facilitating the oxidation of carbon monoxide, an identified major air pollutant. The catalyst systems are characterised using physico–chemical methods including SEM, TEM, XRD, BET surface area analysis, porosity studies, TPR, and DR-UV analysis. Cu/Al system has been identified as the most efficient catalyst among the series. The highest activity of copper based system is attributed to the low temperature reducibility of the dispersed copper ions when compared to the other two active metal ion species.

1. Introduction

Today, research on carbon monoxide removal is in full swing universally, as the presence of CO in atmosphere pilots to serious environmental problems. Automotive exhaust gas has been identified as a major source for atmospheric CO. Reduction of CO amount in automotive exhaust gas is one of the greatest challenges due to the strict emission regulations adopted all over the world. Catalytic oxidative removal is an established facile method to minimise the concentration of this hazardous gas in vehicle exhaust gas. Supported noble metals as well as metal oxides have been extensively surveyed to this effect. Among the various metal oxides that serve as active support, alumina with high surface area and fine textural features stands superior in catalytic field. Noble metal supported on alumina and mixed/supported alumina have been investigated by various research groups [1–8]. Other applications of alumina reach out various fields including lithographic pattern designing, bone demineralisation process, chromatography etc. [9,10].

The present study is aimed at comparing the CO oxidation activity of some well acclaimed oxidation catalysts, vis., CuO, V_2O_5 and CeO_2, that are dispersed over meso alumina. The mesoporous phase is marked by higher surface area and desirable pore features than conventional alumina. The only report on the said reaction over meso alumina discusses the activity of dispersed noble metals, Pt, Pd and Ag [11]. In our work, meso alumina support is synthesised by dodecyl amine assisted template route, and is characterised adopting various analytical methods. The exhibited trend in carbon monoxide activity is correlated with the reducibility of the catalyst systems. The study reveals the superiority of dispersed copper oxide in facilitating carbon monoxide oxidation.

2. Materials and methods

Mesoporous alumina support has been prepared according to the reported procedure [12]. Wet impregnation procedure using ammonium metavanadae was used to yield V/Al catalyst system. Ce and Cu loaded aluminas were synthesised using deposition precipitation of oxides from the respective nitrate solutions by ammonia precipitation. The weight percentage of the active metal oxide in the support has been selected as the criterion for naming the catalysts. The low and wide angle powder X-ray diffraction (XRD) patterns of the support were obtained by a Brucker Nanostar instrument and a Rigaku D/MAX- diffractometer respectively. (Fourier Transform Infra red) FTIR spectra were recorded using JASCO FTIR-4100 spectrometer. Transmission electron microscopy (TEM) and Scanning electron microscopy (SEM) images were achieved with a Philips CM 200 transmission electron microscope, and JEOL Model JSM 6390LV instrument respectively. Diffuse Reflectance UV (DR-UV) spectra were taken with $BaSO_4$ as reference using a Jasco V-550 spectrophotometer. The BET surface area and pore size distribution were obtained using a Micromeritics Gemini Surface Area analyser by the nitrogen adsorption method. Micromeritics Pulse Chemisorb-2705 instrument yielded the temperature programmed reduction (TPR) analysis data. In order to trace the catalytic activity, 0.3 g of the catalyst samples were activated at 300 °C for 1 h, and the reaction was carried out

* Corresponding author. Tel.: +91 494 2401414; fax: +91 494 2400269.
E-mail address: nkrenu@gmail.com (N.K. Renuka).
[1] Tel.: +91 494 2401144; fax : +91 494 2400269.

0167-577X/$ - see front matter © 2013 Elsevier B.V. All rights reserved.
http://dx.doi.org/10.1016/j.matlet.2013.09.073

in a quartz reactor. Gas flow was adjusted to a space velocity of 28,800 h^{-1}, which contained 6% V/V of oxygen, 1% V/V of carbon monoxide and the rest nitrogen. CO and CO_2 in the outlet gas are separated using "Pouropack" packed column followed by converting to methane by the "methanator" and finally detected separately by GC-FID.

3. Results and discussion

Identification of texture and mesoporous morphology of alumina are done from the electron microscopy images displayed in Fig. 1, which reveals nanoparticles of size below 50 nm (Fig. 1(a)), and worm hole type mesopores in alumina prepared via surfactant assisted route (Fig. 1(b)). Type IV adsorption isotherm typical for mesoporous materials with H1 type hysteresis loop was displayed by alumina (not included here). Mesopores of size 3–14 nm were also noticed in the support. Absence of bands characteristic of dodecyl amine (2920 and 2850 cm^{-1}) was confirmed from the FTIR spectra, indicating complete template removal from the calcined support material. XRD pattern (Fig. 2(a)) revealed a rather amorphous nature of meso alumina. However, from the relative intensity of the diffraction peaks, the γ-phase was of alumina was identified in the support, that displayed major peaks at 2θ values 36.79°, 45.73° and 66.49°, corresponding to d values 2.53, 1.93 and 1.39 nm respectively (JCPDS reference number 00-010-0425). The XRD peak characteristic of meso order of alumina, appearing at high d spacing is depicted in Fig. 2(b), which indicated worm hole

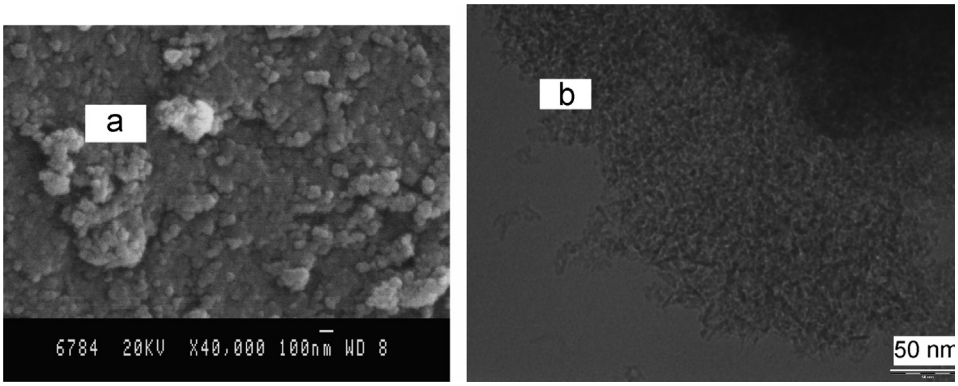

Fig. 1. SEM image (a) and TEM image (b) of γ-Al_2O_3 mesophase.

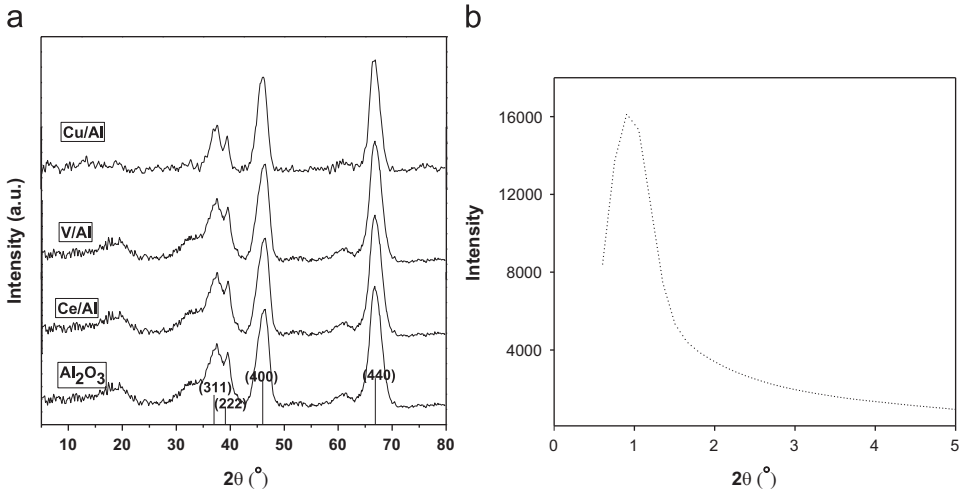

Fig. 2. XRD patterns of catalysts. Wide angle (a) and low angle pattern of support (b).

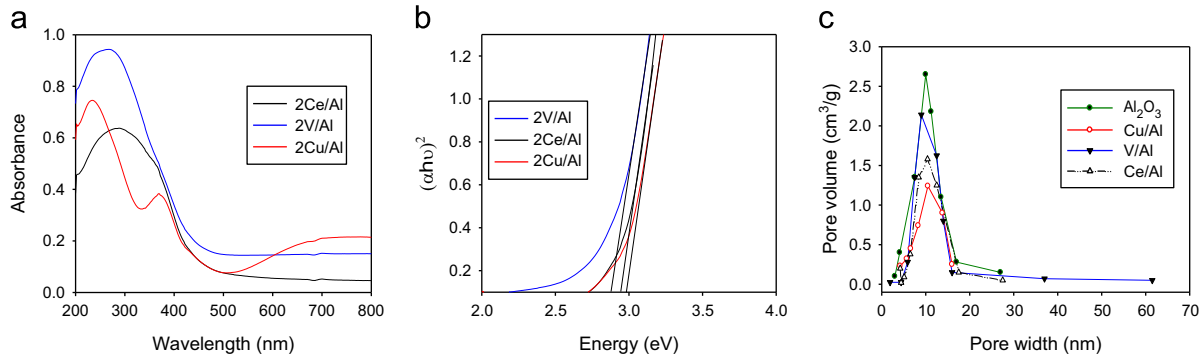

Fig. 3. UV absorbance spectra of the supported catalysts (a); Kubelka–Munk plot (b); pore size distribution of support and 2 wt% metal supported systems (c).