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Full Length Article

Monolithic Mn/Ce-based catalyst of fibrous ceramic membrane for complete oxidation of benzene

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ABSTRACT

Herein, a series of monolithic Mn/Ce-based catalyst of ceramic membranes (CMs) were prepared through impregnation method for volatile organic compounds (VOCs) removal. The porous fibrous CMs with a sinter-locked network structure were fabricated by molding method in a feasible sintering process. Then, the catalytic CMs were systematically analyzed by using specific analytical techniques. The results showed that the fibrous CMs possessed a unique interconnected and uniform pore structure, and MnO_x-CeO_2 active phases were homogenously dispersed into the porous CMs support. The catalytic activity of samples was measured by using benzene as target VOCs. The results revealed that all catalytic CMs were active for oxidation of benzene. Significantly, the catalytic performance was promoted by introducing Ce species into MnO_x. Among all, MnO_x-CeO_2-3:1 catalyst exhibited the lowest 90% benzene conversion temperature (T_{90}) at 244 °C and high stability with continuous benzene stream in the presence of 90 vol.% (20 °C) water vapor under a gaseous hourly space velocity (GHSV) of 5000 h^{-1}, owing to the lower-temperature reducibility and the abundant active oxygen ($O_{Ads.}$) with synergetic effect of MnO_x and CeO_2. The results indicated a promising way to design a high efficiency dual functional CMs for the industrial application of removal VOCs while controlling particulate matters (PMs) from hot gases.

1. Introduction

Monolithic catalysts have good performances on mass and heat transfer, high thermal stability, low pressure drop associated with the high flow rate and good mechanical strength over conventional pellets or powders catalyst [1–3]. Commonly, the monolith support can be divided into metallic or ceramic structure according to the fabrication materials. Metallic monoliths offer advantages such as better heat transfer, good ductility and mechanical stability [4], but limited by its higher cost, lower chemical stability and difficulty in immobilizing catalysts in industrial application [1,5]. Ceramic monoliths have the advantages of a higher porosity, good thermal and chemical stability, strong coating adherence and easier dispersion of used catalysts [6–9], which make it suitable for an excellent monolithic catalyst support to satisfy industrial requirements.

In addition, various industrial processes produce kinds of VOCs containing fine PMs which directly or indirectly threaten the environment and human health [10–15]. Therefore, it must be very desirable to remove VOCs meanwhile dedust does. In fact, porous CMs have been widely used for cleaning dust due to their specific physical and chemical properties which can tolerate the severe conditions with high temperature, high flows, high corrosive atmospheres, thermal shocks and vibrations [13]. In this work, we functionalized the porous CMs with catalytic active components as a monolithic catalyst and studied the effect of monolithic catalyst in removal of VOCs.

Currently, catalytic oxidation is a high-efficiency method in the removal of VOCs by complete oxidizing them into CO_2 and H_2O over specific catalysts at a relatively lower temperature. Mn and Ce-based catalysts exhibited superior catalytic activity for complete oxidation of VOCs [16–19]. Mn oxides have good performance due to their multiple metal oxidation states, high poison resistance, good reducibility and low cost etc. [20,21]. Ce oxides, as a rare earth oxide, has been investigated as an oxygen storage material and perceived an effective component in the VOCs oxidation [22]. MnO_x-CeO_2 mixed oxides showed much higher catalytic activity than those of pure MnO_x and CeO_2 in VOCs oxidation, resulting from the synergistic effect existed in

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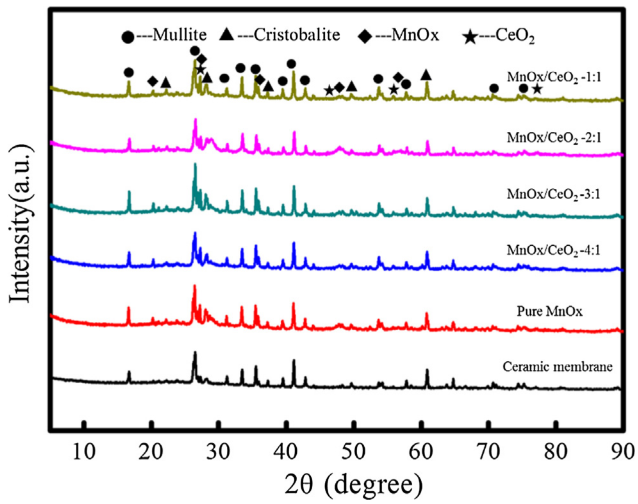

Fig. 1. XRD patterns of CMs and catalytic CMs at different ratio of MnO_x/CeO_2.

the mixed oxides [17,23,24]. However, the catalytic oxidation of industrial VOCs over the conventional powder or pellet catalysts are often limited in practical application. Hence, it is significant to load traditional powder catalysts on to a monolithic support for using in the industrial VOCs removal.

Herein, we successfully prepared a porous CMs by using mullite fiber as raw material and earth-abundant raw chemicals kaolin and feldspar powder as high temperature binder in a relatively low thermal process. Then, the monolithic Mn/Ce-based catalytic CMs were prepared by a simple impregnation method. The Mn/Ce-based catalytic CMs improved gas and heat transfer by acting as a microreactor which is related to the contact of reactant gases with the catalytically active sites on the catalyst surfaces, which is a promising high efficiency dual functional material for treatment the industrial PMs and VOCs pollutions.

2. Experimental

2.1. Preparation of the catalytic ceramic membrane

Porous fibrous ceramic membranes (CMs) were fabricated by molding method in a feasible sintering process. Specifically, a suspension was prepared by dissolving Carboxymethylcellulose (5 g) into deionized water (100 mL) and stirring vigorously for 30 min. Short-cut polycrystalline mullite fibers (d = 10–20 μm, l = 100–200 μm, 30 g) were added to the stirring suspension. Then, Economic accessible ultra-

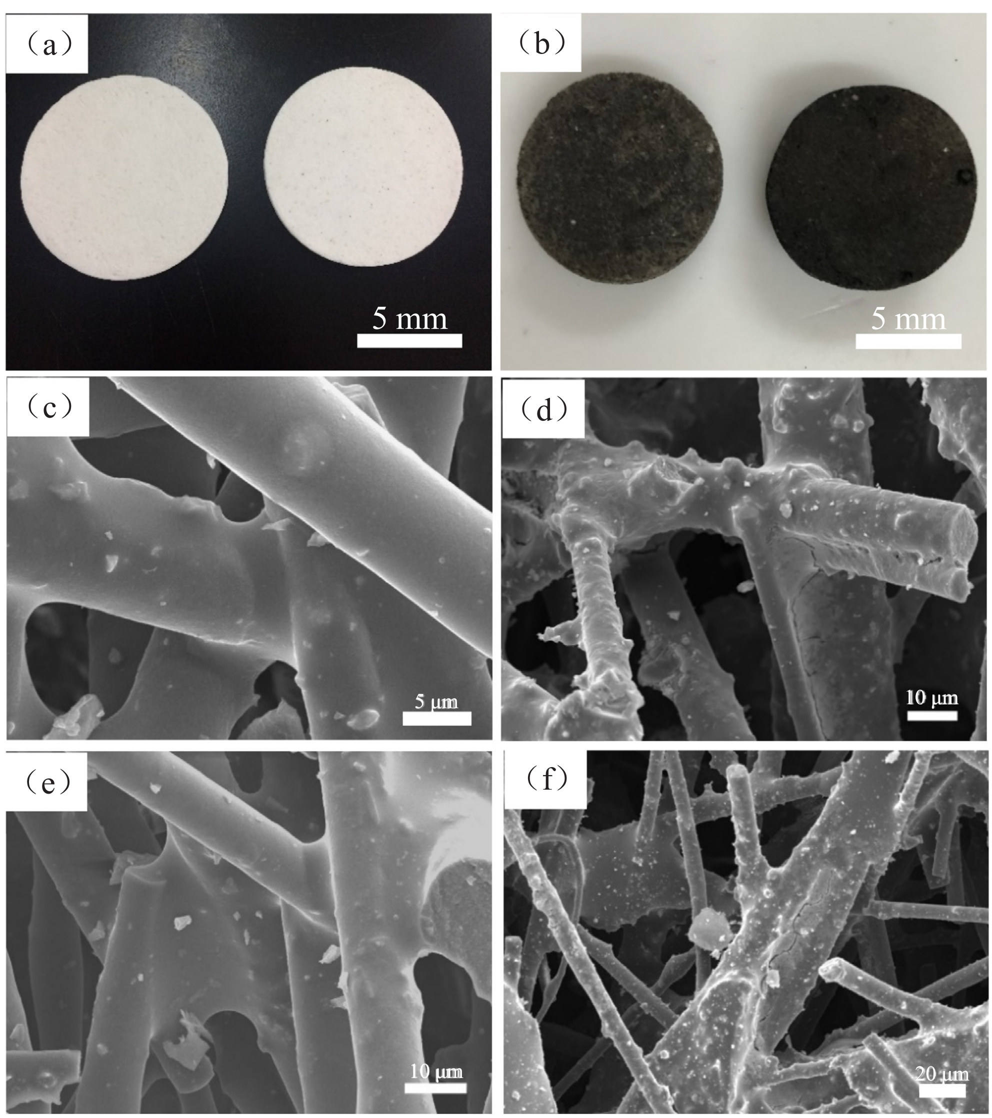

Fig. 2. The overall view of the CMs: (a) before and (b) after impregnation; SEM images of the surface of CMs: (c), (e) before and (d), (f) after adding active phases.