



A comparative investigation on the low-temperature catalytic oxidation of acetone over porous aluminosilicate-supported cerium oxides



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HIGHLIGHTS

- Mesoporous Ce/Al-MSPs were prepared via aerosol-assisted self-assembly method.
- Both redox property and acidity played crucial roles in the acetone oxidation.
- High stability over 24 h on the acetone removal at 150 °C.
- One-step AASA gave catalyst with superior characteristics, activity and stability.

ARTICLE INFO

Article history:

Received 6 September 2014
Received in revised form 25 November 2014
Accepted 8 December 2014
Available online 13 December 2014

Keywords:

VOCs oxidation
Cerium oxide
Mesoporous silica
ZSM-5
Aerosol flow

ABSTRACT

The performance of cerium oxide supported on mesoporous aluminosilicate particles (Al-MSPs) with a nominal Ce and Al loading of 2.8 and 1.2 wt.%, respectively, was evaluated for the low-temperature oxidation of acetone. The influence of the preparation methods, including one-step aerosol-assisted self-assembly (AASA) method and post-synthetic wet impregnation method, was examined. The results showed that the surface area and pore diameter of Ce/Al-MSPs(50/25) prepared by AASA method were similar to those of Ce–Al-MSPs made via traditional impregnation method, while a better metallic dispersion of Ce and Al oxide species on Ce/Al-MSPs(50/25) was observed as compared to that of Ce–Al-MSPs. The catalytic performances of Ce/Al-MSPs(50/25) and Ce–Al-MSPs were compared with that of cerium-impregnated H-ZSM-5 (Ce-ZSM-5) zeolite catalyst. The activity of freshly made catalysts followed the order: Ce/Al-MSPs(50/25) > Ce-ZSM-5 > Ce–Al-MSPs. The superior catalytic behavior of the Ce/Al-MSPs(50/25) could be due to the combined effect of strong surface reducibility and high acid property. Although Ce-ZSM-5 and Ce–Al-MSPs presented high activity for acetone removal in the beginning, their activities quickly decreased due to the coking effect. In contrast, Ce/Al-MSPs(50/25) showed a good stability during a 24 h test at the reaction temperature of 150 °C. Both surface reducibility and acidity played crucial roles for their high catalytic performance and high stability at low temperatures.

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1. Introduction

Catalytic oxidation is recognized as one of the effective techniques for volatile organic compounds (VOCs) elimination due to its high-efficiency and energy-saving process [1,2]. Noble metal-based catalysts show good activity at low temperatures for the complete oxidation of VOCs; however, their industrial applications might be significantly restricted due to their high prices [3–5]. Recently, supported metal oxides (Cu, Mn, Ce, Fe, Ti, etc.) have received considerable attention for VOCs removal because of their

relatively low cost and high catalytic activity in several oxidation reactions [6–9].

In the preparation of supported catalysts, the choice of a suitable support and/or preparation method are of great importance to obtain catalysts with high activity, since support materials play crucial roles in determining the physicochemical properties of the formed active phase (e.g. oxidation state and metallic dispersion), which are all crucial parameters for their catalytic behavior [10–13]. Porous materials are often employed as catalyst supports by providing high surface areas and large pores to disperse the active particles and improve the efficiency of the catalyst [14–19]. It is well-known that ordered mesoporous silica materials, such as MCM-41 and SBA-15 have high surface area, uniform and tailorable pore distribution (2–50 nm), which are good for the synthesis of well-dispersed metal/metal oxide nanoparticles via their hosting into pore channels [20–22]. Recently, more attention has been

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focused on the application of metal-containing mesoporous silica catalysts for end-of-pipe pollution control [23–27].

Metal-loaded mesoporous silica materials are usually prepared by the wet impregnation and grafting methods, which easily result in the appearance of bulk metal species, blocking the channels of mesoporous silica and affecting its catalytic activity [28–30]. Moreover, most of the synthetic processes were conducted through solution precipitation methods using batch reactors, which require multi-step and time-consuming procedures. An alternative approach to metal-containing mesoporous silicas is the exploitation of vapor-phase routes which is the continuous flow set-up and this allows an easy scale-up of the synthesis [31,32]. Mesoporous metal-containing silica spherical particles (MSPs), which were synthesized over one-step aerosol-assisted self-assembly (AASA) process, are relatively novel materials for environmental protection applications [33,34]. The spherical MSPs presents advantages of higher production rate, higher packing density and lower pressure drop than those mesoporous materials manufactured via batch processes [35]. These advantages make MSPs more practical for gas adsorption and separation in terms of overall engineering considerations.

Our research group has been the first to prepare metal-containing MSPs catalysts for acetone oxidation through detailing the relationship between metallic species and catalytic activities of catalysts [36,37]. Among tested bimetallic catalysts of Ce/Mn, Ce/Al and Ce/Cu-MSPs, the Ce/Al-MSPs appeared to have the best performance on acetone oxidation, and it could have ~70% acetone removal at reaction temperature of 150 °C. The oxidation of acetone over Ce/Al-MSPs catalyst was a two-center mechanism involving the acetone chemisorbed on Al-MSPs and the oxygen atom chemisorbed on Ce. This reaction can be described in the following way:



However, to the authors' knowledge, the systematic studies on influences of preparation methods (wet impregnation and AASA method) on the VOCs catalytic activities have not been reported yet. The catalysts prepared respectively by the traditional wet impregnation method and the novel AASA method are characterized by XRD, BET, ICP-MS, TEM, ^{27}Al NMR, XPS, NH_3 -TPD and H_2 -TPR techniques, and their catalytic activities are compared in terms of acetone oxidation. The performances of these two catalysts are also compared with that of the Ce-impregnated commercial H-ZSM-5 zeolite (Ce-ZSM-5) to understand their industrial applicability.

2. Experimental

2.1. Catalysts preparation

Two metals of Ce and Al were chosen as the metal species for synthesizing the bimetallic Ce/Al-MSPs catalysts. The metal precursors were prepared from cerium nitrate and aluminum nitrate solutions, respectively. Cetyltrimethylammonium bromide (CTAB) was used as the structure directing template and tetraethoxysilane (TEOS) was used as the silicon precursor for obtaining the mesoporous structure of silica supports. The molar gel composition of the synthesized mixture was 1 SiO_2 :0.18 CTAB:10 ethanol:80 H_2O :0.008 HCl:0.02 Ce:1/X Al. The synthesized samples are denoted as Ce/Al-MSPs(50/X), where 50 corresponds to the molar ratio of Si/Ce and X corresponds to the molar ratio of Si/Al in the precursor solution.

All of the precursors were mixed together and stirred for 30 min to obtain a clear solution. The solution was then nebulized by an ultrasonic atomizer (1.8 MHz) and the atomized droplets were passed through two heaters by a dry and clean air stream with flow

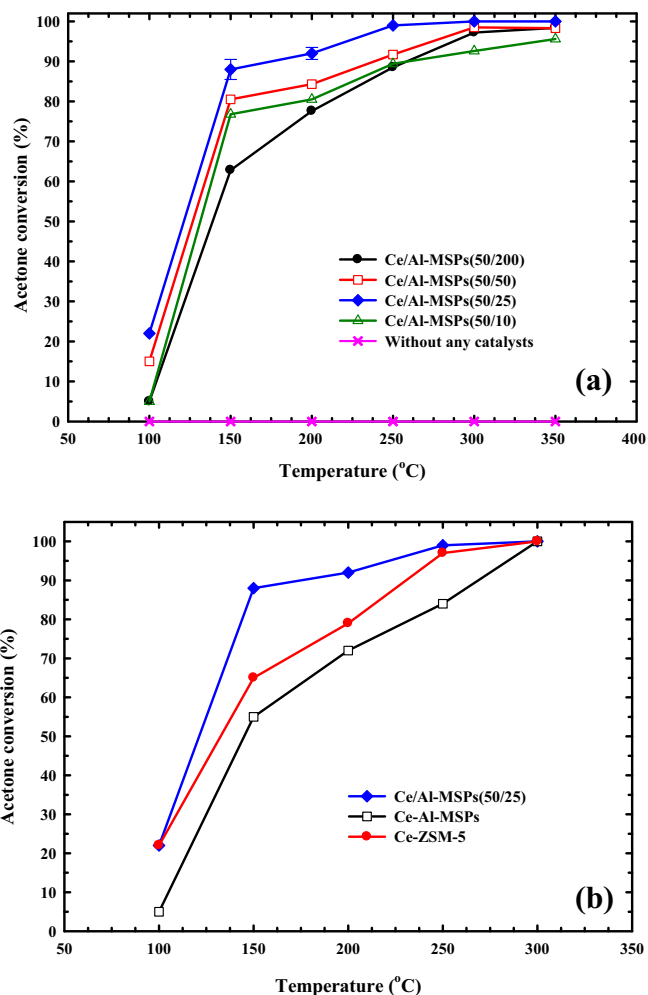


Fig. 1. (a) Acetone conversion over the Ce/Al-MSPs(50/X) catalysts (reaction condition: acetone concentration: 1000 ppmv, O_2 concentration: 20 vol% and GHSV: 15,000 h^{-1}). (b) Comparison of acetone conversion over the Ce/Al-MSPs(50/25), Ce-Al-MSPs and Ce-ZSM-5 catalysts (reaction condition: acetone concentration: 1000 ppmv, O_2 concentration: 20 vol% and GHSV: 15,000 h^{-1}).

rate maintained at 2 LSTP min^{-1} . The first and second heating zones of the aerosol process were controlled at temperatures of 150 and 550 °C, respectively. After the heating process, the as-prepared samples were collected downstream of the reactor by a high-efficiency filter. They were then further calcined at 550 °C for 6 h in air. Detailed procedures on the AASA aerosol method can be referred to our prior studies [36,37].

In order to verify the performance of the Ce/Al-MSPs catalysts, Ce-containing catalysts including mesoporous Ce-Al-MSPs and microporous Ce-ZSM-5 was also prepared by impregnating cerium precursors on mesoporous Al-MSPs support and the commercial ZSM-5 zeolite, respectively. The as-synthesized Al-MSPs and the commercial ZSM-5 (CBV5524G, Zeolyst) was calcined at 550 °C for 6 h, respectively, and then impregnated with cerium nitrate precursor at room temperature under 30 min ultrasonic process. The solution was then dried at 120 °C for 12 h and then calcined at 550 °C for 6 h. The resulting materials are marked as Ce-Al-MSPs and Ce-ZSM-5.

2.2. Catalytic test

The acetone is chosen as the target volatile organic compound, which is commonly used as a solvent in chemical and semi-conductor processing plants. The oxidation of acetone was carried

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