Chemical Engineering Journal 293 (2016) 243-251



Contents lists available at ScienceDirect

Chemical Engineering Journal

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

Low temperature catalytic combustion of ethylene over cobalt oxide supported mesoporous carbon spheres



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HIGHLIGHTS

• CoO loaded mesoporous carbon spheres were developed for practical C₂H₄ removal.

• The catalyst could convert ethylene completely at 185 °C.

• The reaction order for C_2H_4 oxidation was determined to be 0.44.

• The activation energy for C₂H₄ oxidation was calculated to be 79.2 kJ/mol.

ARTICLE INFO

Article history: Received 13 September 2015 Received in revised form 25 January 2016 Accepted 19 February 2016 Available online 27 February 2016

Keywords: Ethylene removal Mesoporous carbon spheres Catalytic oxidation Cobalt oxide

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A novel catalyst has been developed based on cobalt oxide supported mesoporous carbon spheres (MCSs) for the catalytic combustion of ethylene (C_2H_4). Owing to the unique and well-developed 3D mesoporous structure, the cobalt oxide could be homogenously dispersed on the carbon framework at nanoscale dimensions. The cobalt oxide loaded on MCSs is detected to be in the form of CoO owing to the reduction effect of the carbon support. The MCSs support could facilitate the oxidation ability of CoO, involving more active oxygen species on the catalyst surface. The MCSs supported CoO catalyst could convert 1000 ppm C₂H₄ completely at 185 °C, exhibiting a highest catalytic performance among the synthesized catalysts. The reaction order and active energy for C₂H₄ oxidation are determined to be 0.44 and 79.2 kJ/mol, respectively. A GHSV study and a stability test further indicate that the MCSs supported CoO catalyst possesses high catalytic activity and stable catalytic performance for low concentration C₂H₄, which could benefit for the industrial application of the catalyst in the future.

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1. Introduction

Ethylene (C_2H_4) is a plant growth regulator released from fruit and vegetable tissues, which could accelerate the growth, aging and even spoiling of plants [1–3]. The adverse impact of C_2H_4 on fresh food preservation has been estimated to cause significant postharvest food losses (as high as 10–80%), which has become a growing area of research and development to alleviate this impact [4]. Currently, several strategies have been proposed for the C_2H_4 removal, including adsorption [1,5], direct oxidation [6,7] and catalytic oxidation [8,9]. Adsorption usually employs adsorbents with well-developed porosity, such as activated carbon [10–12] and zeolites [13], which have been widely used in air purification. However, these adsorbents usually reversibly trap C₂H₄ by either ion-quadrupole interaction or Van der Waals force, which show very low C₂H₄ adsorption selectivity, particularly when dealing with very trace C₂H₄ concentration. Direct oxidation by strong oxidizers such as potassium permanganate [14] or ozone [15] has been evidenced to be efficient to eliminate trace C₂H₄ even at low temperature. However, the main drawbacks of oxidation are the high toxicity and non-renewability, limiting their wide application in food storage areas. Catalytic combustion, using solid catalysts composed of an active phase (noble metal or transitionmetal oxide) and a high-surface-area porous support, has been well studied as an effective and feasible technology for chemically oxidizing C₂H₄ into carbon dioxide and water at low or mediate temperatures [9]. This method presents a number of significant potential advantages including non-toxicity, high catalytic activity and selectivity for C_2H_4 as well as stable catalytic performance.

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Fig. 1. (a) Schematic illustration of the preparation of MCSs and MCS/Co; (b) photo of synthesized MCSs; (c) SEM image of the MCSs; (d) SEM image of cobalt oxide loaded MCSs (MCS/Co-30).

Generally, the active phase for C_2H_4 oxidation is noble metals such as Pd [9], Au [16,17], Pt [18] and Ag [19,20], which show a high oxidative activity for low-concentration C_2H_4 . Unfortunately, their very high costs limit the application in the large volume storage facilities. The transition-metal oxides, such as cobalt oxide (CoO_x, in the form of Co₃O₄ or CoO), copper oxide (CuO), manganese oxide (MnO_x) and titanium dioxide (TiO₂), are economically viable alternatives, which also exhibit high reactivity for C_2H_4 oxidation [21–25]. Cobalt oxide presents a high catalytic activity in various catalysis processes, such as the oxidation of carbon monoxide or organic molecules [26–32]. Even in some cases it is more reactive than supported Pd or Pt catalysts [33], which is the catalyst of choice for this reaction. However, a relatively high temperature is needed to fulfill a high conversion efficiency, which might be attributed to the assorted polycrystals of cobalt oxide, exhibiting different reactivities and usually with lower catalytic



Fig. 2. TEM images of (a) MCS/Co-5, (b) MCS/Co-10, (c) MCS/Co-30, (d) HRTEM image of MCS/Co-10.

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