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Low temperature combustion of ethylene in a carbon dioxide stream over a cordierite monolith-supported Cu–Mn Hopcalite catalyst

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ABSTRACT

Complete combustion of trace amount of ethylene in food-grade carbon dioxide is examined over a copper-manganese Hopcalite monolithic catalyst which is prepared with co-precipitation of Hopcalite powder, followed by washcoating on cordierite monolith via a dip-coating method. It is shown that the combustion activity of the Hopcalite monolithic catalyst is closely dependent on the washcoat loading and calcination temperature, and that the calcination temperature has a great effect on the adhesion strength of the Hopcalite washcoat. It is proposed that the catalyst combustion activity originates from a synergic effect of the CuO and amorphous Cu–Mn oxide phases. Solid reaction between the Hopcalite washcoat and cordierite monolith not only improves the adhesion strength of the Hopcalite washcoat, but also prevents the formation and growing of the $Cu_{1.5}Mn_{1.5}O_4$ crystalline particles and thus develops the combustion activity. Furthermore, it leads to doping of Mg in the Hopcalite washcoat, which improves the catalytic stability of the Hopcalite monolithic catalyst.

1. Introduction

Emission of carbon dioxide has become an excessive burden to our environment in recent years. Ethylene epoxidation process (EO/EG) produces almost pure CO_2, and the key for the utilization of this CO_2 stream is its further purification to remove ethylene, aldehyde and alcohol at ppm level [1]. A number of technologies including adsorption, absorption, and catalytic combustion have been employed for the removal of trace amount of ethylene [1]. As is well known, catalytic combustion is one of the most attractive approaches of controlling the emission of organic compounds. For this process, a low reaction temperature is preferred because the combustion cannot provide enough heat to keep the temperature due to the low concentration of the organic components in the carbon dioxide stream. Hopcalite catalyst, copper–manganese mixed oxide with low cost and high activity at low temperatures, is superior for this process. Since its discovery in 1920 [2], Hopcalite catalyst has been investigated extensively [3,4] and has served as efficient catalysts in many industrially important oxidation processes, such as the oxidation of CO, methanol, ethylene, toluene, nitric oxide and combustion reactions [3–6]. It is also very important to the respiratory protection in military, mining and space exploration [7,8].

In view of practical applications, combustion catalysts should be washcoated on structured supports, e.g., ceramic and metallic monoliths, to treat large gas flows with low pressure drop. The development of monolithic catalysts and reactors has been one of the major achievements in the field of heterogeneous catalysis and catalytic reaction engineering [9–18]. For more than 30 years, monolithic catalyst manufacturers have been successful in stationary and automotive exhaust gas treatments, where the gas phase detoxification must be fast with contact time less than a second, since large volumes of gas have to be treated [19–29]. In recent years, monolithic catalysts and reactors are expected to have increasing applications in chemical and biochemical processes, such as in mass production of chemicals, in the treatment of fuel and flue gases, and in other multiphase processes [9,30,31].

In this article, the complete oxidation of ethylene in food-grade CO_2 was carried out over a Cu–Mn Hopcalite monolithic catalyst. The effects of the washcoat loading and calcination temperature were examined. Various techniques such as TPR, XRD and SEM were used for catalyst characterization.

2. Experimental

2.1. Catalyst preparation

Hopcalite catalyst powder was prepared via a co-precipitation technique. Under vigorous stirring, a mixed aqueous solution of 0.333 mol/L copper nitrate and 0.167 mol/L manganese nitrate was

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