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Selective synthesis of benzophenone over two-dimensional mesostructured CrSBA-15

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ABSTRACT

Well hexagonally ordered CrSBA-15 catalysts synthesized by different hydrothermal methods were used, for the first time, for the selective synthesis of benzophenone (BP═O) by liquid-phase oxidation of diphenylmethane (DPM) under various reaction conditions. To investigate the leaching of chromium species on the surface of silica networks, the results of original and reusable Cr-containing mesoporous silica catalysts were correlated and compared, and the catalytic activity of washed CrSBA-15(8) was also demonstrated. Based on the all catalytic results for this reaction, CrSBA-15(8) was found to be a highly active and an environmentally friendly solid catalyst and has superior catalytic activity than other Cr-containing mesoporous catalysts.

1. Introduction

Benzylic oxidation is an important transformation in synthetic organic chemistry [1–3]. Oxidation of diphenylmethane (DPM) to benzophenone (BP═O) is an important example for the benzylic oxidation. BP═O is primarily used as an important intermediate chemical in the production of perfumes and pharmaceuticals, and used as a fragrance enhancer and photo-initiator, and it is conventionally synthesized by the oxidation of DPM in the presence of chromic acid [4] and nitric acid [5], and using the Friedel–Craft's acylation of benzene with benzoyl chloride in the presence of $AlCl_3$ [6]. However, these processes are still expensive, polluting and risky. The large amounts of catalyst need for these catalytic reactions. After completion of the catalytic process, the separation of catalyst from the reaction mixture is very difficult. A lot of tarry wastes can be formed in the catalytic system. The chromium reagents are environmentally undesirable. To overcome these problems, a solid acid catalyst is mainly used for the decrease of the production cost, owing in the bulk chemical industries. For example, several microporous catalysts are used for the synthesis of BP═O. Particularly, the microstructured CrAPO-5 and Cr-PILC catalysts are used for the production of BP═O by the oxidation of DPM with *t*-butylhydrogenperoxide (TBHP) in the liquid phase reaction conditions. However, they produce a less conversion of DPM at high reaction time because of small pore diameter which restricts the diffusion of reactants/intermediate and products [7,8]. Mesoporous CrMCM-41 catalysts are also used in the oxidation of DPM using various oxidizing agents under different reaction medium [9–12], and they produce a good DPM conversion as well as BP═O selectivity. Several researchers tried to incorporate a very high heteroatom species on the surface of SBA-15 under strong acidic hydrothermal method, however, most of them failed to introduce more heteroatomic species into SBA-15 with catalytic active sites of tetrahedral coordination because the formation of metal-oxo species in the mesoporous material is much less. To overcome this problem, pH-adjusting direct hydrothermal (pH-aDH) method has been introduced and used for the synthesis of MSBA-15 catalysts (M = Al, Cr, Mn, Sn, Ga, Ce, Ti, Nb and V) with high metal-content, and the synthesized MSBA-15 catalysts have been used for certain catalytic reactions [13–25]. Selvaraj and Kawi [16] reported the highly ordered CrSBA-15 catalyst synthesized with enhanced hydrothermal stability. The CrSBA-15 catalyst has a huge number of tetrahedral chromium in the framework of silica walls because it has thicker pore walls and higher hydrothermal stability than CrMCM-41 [16]. The well hexagonally ordered two dimensional mesoporous CrSBA-15 catalysts are successfully used in the production of anthraquinone and verbenone with the ranging of selectivity from 88 to 100% [16,26,27]. However, to the best

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