

Unique Catalytic Activity of Mesoporous Catalysts in Friedel-Crafts Benzylation of Benzene

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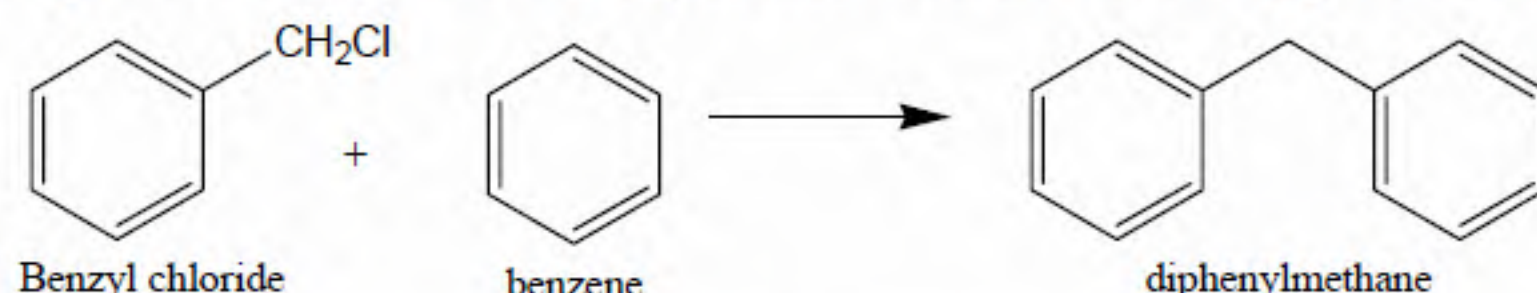
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Abstract

Different samples of metal-incorporated MCM-41 were prepared and used as catalysts in Friedel-Craft's benzylation of benzene. The catalytic performance was evaluated by off-line GC analysis. Fe-MCM-41 exhibited excellent activity, the sample with Si/Fe ratio = 10 showed 92% conversion with 95% selectivity towards diphenylmethane within a few minutes. Generally, the activity per Fe-site was an order of magnitude higher for the samples containing a combination of Fe₂O₃ nano-particles and isolated Fe³⁺ sites. A synergy of two catalytic centers (particles and isolated sites) is proposed to explain the high performance of the highly loaded samples. The catalytic performance of Fe-MCM-41 was superior to other metal-containing MCM-41 (e.g. Ga, Sn, and Ti) catalysts, or other Fe-containing mesoporous materials (e.g. Fe-HMS).

Introduction

The electrophilic substitution reaction of alkyl groups in an aromatic system are known since 1877. Friedel-Crafts type reactions are considered as one of the most important reactions in organic synthesis. Benzylation of benzene using benzyl chloride to afford diphenylmethane is an interesting example as substitute for polychlorobenzene in the application of dielectrics.



Although, homogeneous, Lewis or Brønsted acids were commonly used as catalysts in Friedel-Crafts type reactions [1] they create several problems such as polyalkylations and rearrangements which are difficult to avoid, in addition to corrosion, toxicity, and catalyst recovery. An important step forward to phase put these problems is the application of solid acid catalysts in these reactions.

A number of solid catalysts have been evaluated. Clark et al.[2,3] reported montmorillonite-supported zinc (Clayzinc) and nickel chloride as highly active for catalyzing Friedel-Crafts alkylations. Another approach was clays [4,5]. Choudary found that iron pillared clays (FePILCs) were efficient catalysts, producing quantitative conversions with greatly reduced amounts of catalysts and shorter reaction times [6]. However, FePILCs were quite labile even under ambient conditions[7]. Aging the materials for periods of 3 months at 25°C resulted in a loss of the 011 ordering, redistribution of iron and a concomitant change in the degree of polycation polymerization. At the same time, the poor thermal stability of clay materials gives rise to difficulties in regeneration. Heteropolyacid salts[7-9] and zeolites (e.g. HY [10], and H-β[11]) were also used as catalysts for Friedel-Crafts reactions. It is interesting to mention that, in spite of its very strong acidity, zeolite H-ZSM-5 shows little or no activity in the