



Catalytic ozonation of *p*-chlorobenzoic acid in aqueous solution using Fe-MCM-41 as catalyst



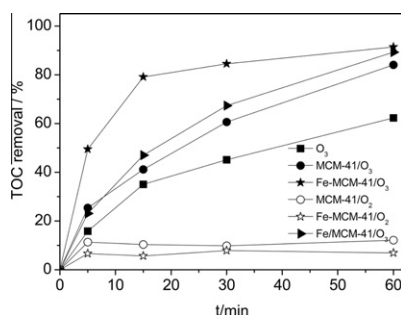
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HIGHLIGHTS

- ▶ Fe-MCM-41 was first used as a heterogeneous catalyst for ozonation process.
- ▶ Fe leaching of Fe-MCM-41 was greatly reduced compared with that of Fe/MCM-41.
- ▶ Brönsted acid sites on the Fe-MCM-41 catalyst were thought to be the active sites.
- ▶ Generation mechanism of hydroxyl radicals was proposed.

GRAPHICAL ABSTRACT



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ABSTRACT

MCM-41 and iron-substituted MCM-41 (Fe-MCM-41) were synthesized successfully and characterized using X-ray diffraction (XRD), nitrogen adsorption–desorption and transmission electron microscopy (TEM). The mesopores of the sample remained well ordering and of hexagonal structure. Fe-MCM-41 samples possessed high BET surface areas, high pore volumes and narrowed pore size distribution. Their catalytic activities were compared in terms of *p*-chlorobenzoic acid (*p*-CBA) and TOC removal. Ozonation catalyzed by Fe-MCM-41 exhibited extraordinarily high catalytic performance over other studied processes. 100% conversion of *p*-CBA and 91.3% TOC were achieved in 10 min and 60 min, respectively. Fe-MCM-41 as a novel heterogeneous catalyst in ozonation process was addressed. The presence of *tert*-butanol (TBA) inhibiting the degradation of *p*-CBA in the Fe-MCM-41/O₃ process verified that the hydroxyl radicals derived from ozone decomposition were responsible for the improvement of catalytic ozonation. The mechanism explaining the formation of hydroxyl radicals on Fe-MCM-41 surface was proposed. Negative centers or Brönsted acid sites developed by the substitution of trivalent iron into the walls of MCM-41 and the hydroxyl groups formed as a result of coordinative unsaturation were found to be the active sites enhancing catalytic performance. The superior stability of iron ions in Fe-MCM-41 catalyst during the ozonation process was also examined.

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1. Introduction

Ozone (oxidation potential 2.07 eV) has been used widely and effectively in water and wastewater treatment to eliminate most inorganic and organic substances since the late 19th century [1].

However, high energy demand and partial oxidation of some refractory organic compounds confine the practical application of ozonation process. The molecular ozone reacts especially reactively with specific functional groups, such as amino groups, double bonds and nucleophilic positions [2,3] but lead to the formation of aldehydes and carboxylic acids, both of which are inert with ozone, thus total mineralization is not always achievable. In such case, radical processes have to be implemented in order to improve the ozonation efficiency.

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