MJFAS MALAYSIAN JOURNAL OF FUNDAMENTAL AND APPLIED SCIENCES
PRINT ISSN: 2289-5981 | ONLINE ISSN: 2289-599X

RESEARCH ARTICLE

Direct synthesis of ZSM-5 from kaolin and the influence of organic template

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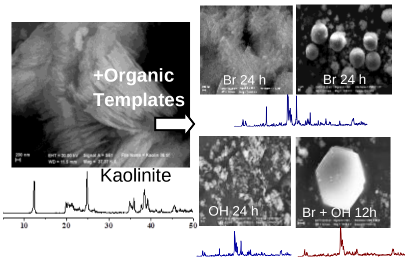
Article history
Received 21 March 2017
Accepted 3 May 2017

Graphical abstract



Abstract

We investigated the transformation of kaolin to ZSM-5 using hydrothermal synthesis method and the influence of organic template as a structure-directing agent. The formation of ZSM-5 from kaolin occurred via dissolution of kaolin to form amorphous silica. Zeolite-like analcime produced when the hydrothermal treatment was extended to 18 h. We found synergistic effect of using a mixture of TPAOH and TPABr organic template to enhance kaolin dissolution, initiate ZSM-5 formation and inhibit the transformation of ZSM-5 to analcime. The ZSM-5 framework formed after crystallization for 6 h by using the mixture of TPAOH and TPABr, while when using TPAOH or TPABr only, ZSM-5 formed at longer crystallization time, i.e. 12 h.

Keywords: Crystallization time, kaolin, phase transformation, TPABr, TPAOH, ZSM-5

INTRODUCTION

ZSM-5 has been widely used as catalyst in petrochemical process because of the high surface area that allows the adsorption of reactant molecules (Kim and Chung, 2003). The ZSM-5 is a robust material with high thermal and acid stability; its unique pore channel system makes a good catalyst to achieve high conversion and selectivity of desired product (Wang *et al.*, 2007). The synthesis of ZSM-5 from natural mineral sources and waste materials has been reported mainly to reduce the cost of production and to utilize the waste minerals. Serpentine (Kim and Chung, 2003), rice husk ash (Prasetyoko *et al.*, 2012), fly ash (Tanaka and Fujii, 2009), expanded perlite (Wang *et al.*, 2007) and kaolinite (Feng *et al.*, 2009; Pan *et al.*, 2014) were previously reported as precursor in ZSM-5 zeolite synthesis. These minerals are generally rich with SiO_2 and Al_2O_3 which make them perfect candidates for zeolite synthesis. The use of metakaolin, a reactive phase of calcined kaolin, as raw materials for the synthesis of ZSM-5 has been reported which requires the heat treatment of kaolin up to 600 – 1100 °C (Feng *et al.*, 2009; Alkan *et al.*, 2005). According to Johnson and Arshad (2014), this activation process promotes kaolin to metastable phase and becomes highly reactive for zeolites formation.

Hydrothermal synthesis of zeolite is influenced by temperature and crystallization time, which these parameters can affect the process of zeolite formation, the Si and Al content in the framework and also the stability of the zeolite (Shiralkar and Clearfield, 1989). In order to orientate the crystal growth, the synthesis was usually performed in the presence of organic template as a structure-directing agent, typically tetrapropylammonium cations. This cation is trapped in the zeolite pores during crystal growth and removed by calcination in air at high temperature (Pan *et al.*, 2014). The role of this organic structure-directing agent mimics the kinetics of complex nucleation and crystallization processes (Byrappa and Yoshimura, 2013). Alkalinity also has a vital role in the crystal growth of zeolite as the OH anions acts as mineralizing agent. Increasing the pH of the mixture improves the dissolution of Si and Al oxides or hydroxides precursor to become a homogeneous solution (Karimi *et al.*, 2012). The increasing of OH^- concentration accelerates the crystals growth and shortens the period of induction time for the nucleation to occur.

The potential of kaolin as mineral source (Si and Al) for ZSM-5 synthesis was investigated in this study. Kaolin was used without prior pretreatment in the presence of TPAOH and/or TPABr as a structure-directing agent. Metakaolinite was generally used for mineral source of ZSM-5 synthesis. Another publication has examine that kaolin can be directly used as mesoporous aluminosilicates (Qoniah *et al.*, 2015). Hydrothermal synthesis was carried out for 6, 12, 18, and 24 h in order to investigate the mechanism of kaolin transformation to ZSM-5. Detail analysis of the solid products using XRD, FTIR and SEM analysis gives information on the purity, crystallinity, morphology and thermal stability of resulting ZSM-5.

EXPERIMENTAL

Materials