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PREPARATION OF HIERARCHICAL ZSM-5 FROM INDONESIAN KAOLIN BY ADDING SILICA

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Abstract. Hierarchical ZSM-5 was prepared using Bangka kaolin-Indonesia in the absence of structure directing agent. The effect of hydrothermal time and temperature was investigated. The prepared samples were characterized by XRD, XRF, FT-IR, and N_2 adsorption/desorption. The results show that hierarchical ZSM-5 can be obtained at hydrothermal temperature of 393–448 K. Moreover, this method also produces mordenite and quartz as by-products.

Keywords: hierarchical ZSM-5, Bangka kaolin-Indonesia, hydrothermal time and temperature.

1. Introduction

Development of porous material with a higher surface is widely investigated such as adsorption, chromatography, catalysis, sensor, and gas storage [1-4]. Hierarchical is one of the pore modification technologies. Hierarchical ZSM-5 is synthesized and used in the selective condensation of benzaldehyde with 2-hydroxy-acetophenone, esterification of benzaldehyde with hexanoic acid, as well as branched polyethylene [5]. Moreover, hierarchical ZSM-5 shows better catalytic activity to convert methanol than using conventional ZSM-5 as a catalyst [6].

A bulk of works has been reported on preparation of hierarchical ZSM-5. Some researchers reported preparation of hierarchical ZSM-5 by post synthesis as demetalisation and desilication without organic template done [7-9], while the other researchers published the synthesis of hierarchical ZSM-5 by direct template (supramolecular and solids template) [10-13]. The synthesis also requires silica and alumina source, in order to decrease the cost of ZSM-5, many researcher substituted the commercial chemical products by the natural ones. Rice husk ash, fly ash, and kaolin are some materials that have been used for synthesis of zeolite as a source of silica and/or alumina [14-16].

In Indonesia kaolin is a clay mineral reserved in large abundance which reaches 1,068,377,264 tons. Bangka Belitung is the largest mine of kaolin in Indonesia [17]. Generally, kaolin contains 10–95 % of minerals and 85–95 % of kaolinite ($2H_2O{\cdot}Al_2O_3{\cdot}2SiO_2$) [18]. Kaolin may also contain quartz, mica, feldspar, illite, montmorillonite, and a bit of bauxite, zircon, rutile, kyanite, siliminate, graphite, attapulgite and halloysite [19]. The kaolin is used for the synthesis of ZSM-5, for example for the synthesis of ZSM-5 micropores. However, kaolin structure is constructed of a silicon tetrahedral layer and a layer of silicon octahedral units (1:1 clay mineral), causing difficulties to synthesize zeolite directly from kaolin [20]. Therefore kaolin must be enabled to change its structure by metakaolinization. Metakaolin was observed through calcination of kaolin at the temperature of 873–1173 K [21]. In this paper we present a synthesis method to prepare hierarchical ZSM-5 from metakaolin without the addition of structure directing agent. The effect of hydrothermal time and temperature in the synthesis process was examined. The products were characterized by XRD, XRF, FT-IR, and N_2 adsorption/desorption.

2. Experimental

2.1. Material Preparation

ZSM-5 was prepared by using Bangka Belitung kaolin as a raw material, tetraethylorthosilicates (TEOS) as an additional silicon source, sodium hydroxide (NaOH) as a mineralizing agent and provides alkalinity, cethyltrimethylammonium romide (CTAB) as a mesophase template, and silicalite-1 as a seed. All