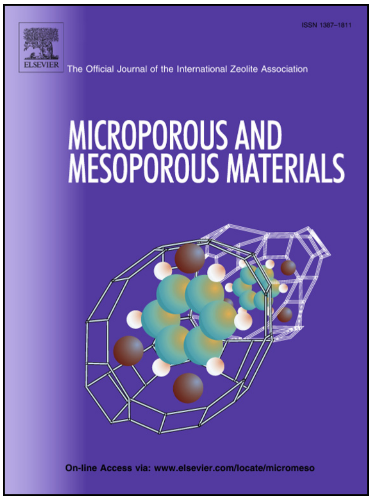

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PII: S1387-1811(14)00302-3
DOI: http://dx.doi.org/10.1016/j.micromeso.2014.05.030
Reference: MICMAT 6576

To appear in: *Microporous and Mesoporous Materials*

Received Date: 17 January 2014
Revised Date: 23 April 2014
Accepted Date: 23 May 2014

Please cite this article as: A. Jackowski, S.I. Zones, K. Chaudhuri, H.S. Lacheen, S.W. Yeh, A hydrothermal synthesis of a 2-dimensional layered silicate followed by a transition to a 3-dimensional aluminosilicate zeolite, *Microporous and Mesoporous Materials* (2014), doi: http://dx.doi.org/10.1016/j.micromeso.2014.05.030

A hydrothermal synthesis of a 2-dimensional layered silicate followed by a transition to a 3-dimensional aluminosilicate zeolite

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Dedicated to the memory of our wonderful zeolite research colleague, Lun-Teh Yuen

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ABSTRACT: We present a study of aluminosilicate zeolite synthesis where the first crystalline product observed is layered. It then transforms with time to the zeolite SSZ-81. The reaction has two crystalline phases that precede the SSZ-81 formation because FAU zeolite is used as the Al source that, eventually, is incorporated into the final organo-aluminosilicate zeolite product. The course of events is followed by microscopy, XRD, changes in solution pH, product organic mass, and TGA. In addition, the reaction using a chromium-exchanged FAU was performed that then allowed for tracking the same changes using EDX analyses in the SEM.

KEYWORDS: zeolite, SSZ-81, crystallization kinetics, EDX

INTRODUCTION

There is a recent surge in interest on layered materials as synthesis precursors to 3D zeolites, with the former then delaminated to create exfoliated structures with catalytic properties. The major material in this vein has been the MCM-22 (P) that precedes the formation of the 3D zeolite with the IZA code **MWW**. The hydrothermal synthesis product had been described initially by Puppe [1], Zones [2], and Rubin [3]. The structure of the Rubin material, MCM-22, was realized by Lawton et al [4] and then initial work in layer separation was begun by Roth et al [5-7] in a series of **MCM** materials. Corma also described the preparation of ITQ-2 via delamination studies [8]. Work has continued for more than a decade (see Roth and Cejka [9]) and continued breakthroughs and modifications have been made (Inagaki [10], Roth and Dorset [11], Tsapatsis [12], Zones and Katz [13-15], Pastore [16], Lercher [17], Schreyeck [18]). Recently, the zeolite precursor PREFER has been delaminated to UCB-2 via a swollen intermediate state [14]. Roth and Cejka also demonstrated a very novel utilization of features within a germanosilicate structure by describing the delamination of an actual 3D zeolite, engineered around the hydrolysis of Ge-containing double four ring groups in the 14-ring zeolite structure UTL [19].

The use of layered precursors could be modified with the use of an aluminum source from a zeolite. **FAU** as an Al source in zeolite synthesis has been shown to impact kinetics, beginning with studies on SSZ-13 formation [20], and then later in the syntheses of novel zeolites like SSZ-26 [21] and SSZ-42 [22]. Recently a similar style of conversion to a Levynite product was described [23]. Other studies have demonstrated the use of chromium-exchanged **FAU** as the Al source altered the reaction kinetics that facilities the formation of more Al-rich SSZ-23, an **STT** structure, instead of the expected SSZ-13 [24].

In this study a hydrothermal synthesis containing Y zeolite as an Al source that persists in the reaction through a period of layered silicate formation was investigated. The progression of phases forming and further converting to an organic cation (SDA) containing 3D zeolite was monitored. The changes in reaction products as a function of time are followed by examining the solid products via elemental composition, TGA of the SDA, XRD, morphological changes by SEM, overall mass increases with product formation, and changes in solution pH that were previously highlighted as an important indicator of crystallization of SDA-containing high silica zeolites by Zones et al [25-27]. Approaches to alter the product structure using chromium exchanged **FAU** were also undertaken that led to a convenient method to trace the aluminum migration during reaction.

EXPERIMENTAL

SYNTHESIS

The SDA, I,5-diquinuclidinyl pentane dication (Figure 1) was prepared as previously described [28]. The diquaternary SDA was used as a dihydroxide and mixed with NaOH, Na-Y zeolite as Al source, and Cabosil MS, a very pure source of silica. Reagents, in proportions given in Table 1, were combined into the Teflon-lined Parr 4823 reactors. Reactions were also performed with a Reheis F-2000, which is an aluminum hydroxide, substituted for Na-Y with the same proportions in Table 1. Individual time points were taken for the use of a single reactor. Initially points were taken for runs 2-14 days and then more detailed characterization was carried out on points where transitions occur. The runs were carried out at 160 ºC, 43 RPM with reactors loaded onto a spit in a Lindberg Blue M convection oven. At the end of the specified run time the reactor was removed and cooled to room temperature with-