Available online at www.sciencedirect.com
ScienceDirect
Microporous and Mesoporous Materials 101 (2007) 30–42
MICROPOROUS AND MESOPOROUS MATERIALS
www.elsevier.com/locate/micromeso

New zeolite topologies based on intergrowths of FAU/EMT systems

Gema González [a,*], Carmen Soraya González [a], Werner Stracke [b], Rudolf Reichelt [b], Luis García [c]

[a] Lab. Ciencia e Ing. Materiales, Dpt. Ingenieria, Instituto Venezolano de Investigaciones Científicas, Caracas 1020 A, Venezuela
[b] Institute for Medical Physics and Biophysics, University of Muenster, Muenster, Germany
[c] Escuela Ing. Química, Fac. Ingeniería, Universidad Central de Venezuela, Caracas, Venezuela

Received 27 July 2006; received in revised form 23 October 2006; accepted 1 November 2006
Available online 5 January 2007

Abstract

The controlled synthesis of intergrowths of zeolites FAU and EMT was carried out by the mixture of the organic structure-directing agent (SDA) 15-Crown-5 (T_1) and 18-Crown-6 (T_2) in different proportions ($T_1/T_2 = 25\%$, 50% and 75%), using T/Al_2O_3 ratios of 0.3 and 0.7, *n*-heptane and *m*-xylene catalytic reactions were used as model reactions for elucidating the pore and cage size of the solids.

The samples were characterized by the following techniques XRD, FTIR, HRSEM, STM and EDX, N_2 adsorption and TPD-NH_3. The FAU samples were formed by octahedral submicrometric crystallites, EMT samples have hexagonal plate morphology of 2–5 μm. The intergrowth crystals are micrometric hexagonal plates through whose hexagonal faces intergrow the octahedral crystallites of FAU. The intergrowth proportions obtained were evaluated by comparison of the experimental XRD patterns with the simulated patterns using the DiFFaX computing program. The intergrowth proportion obtained depends on the molar ratio of template/Al_2O_3 and the relative proportion of SDA used in the gel. For a SDA proportion $T_1/T_2 = 50\%$ and a $T/Al_2O_3 = 0.70$, an intergrowth proportion 50% of FAU and 50% of EMT was obtained, with a stacking arrangement of clusters. When using the same T_1/T_2 but $T/Al_2O_3 = 0.30$, the intergrowth proportion was 12% of FAU and 88% of EMT.

Keywords: Intergrowth; FAU; EMT; *m*-Xylene; *n*-Heptane; Structure-directing agent

1. Introduction

The development and the use of molecular sieves have caused a great impact in different industrial processes due to its versatility, related directly to their structural acidic and sorption properties. The importance of zeolites as catalysts for refining processes has been mainly associated to the regular distribution of channels and cavities which determine the distribution of product species in terms of geometry and size. These properties can be modified through the variation of the parameters of the synthesis. In this way, it is possible to obtain materials with new framework structures and potentially better catalysts. In the search of new topologies it has been considered, that materials with controlled stacking disorder form intergrowth structures which could be an interesting alternative for catalytic applications. The structural disorder and its correlation with the catalytic activity have not been studied very well, neither the controlled synthesis of structural disorder. This is the fundamental base for the controlled design of microporous materials of disordered structures. In the present work, a detailed study of the synthesis of intergrowths FAU/EMT has been carried out.

The zeolite FAU is one of the most important zeolites for catalytic applications, due to its pore size and acid site accesibility. This zeolite crystal has a face centered cubic structure with a space group *Fd*3m and a unit cell length of $a = 24.74$ Å. It is formed by eight supercages

* Corresponding author. Fax: +58 212 5041418.
E-mail address: gemagonz@ivic.ve (G. González).

1387-1811/$ - see front matter
doi:10.1016/j.micromeso.2006.11.008

(~1.2 nm^3) per unit cell with four 12-membered ring pore openings (0.74 nm) per cage, and can be described by the stacking of sodalite layers in a ABCABC sequence [1]. These layers are related by a center of inversion in the double 6-member rings. Each layer is shifted laterally before connecting to the previous one. The EMT zeolite was synthesized for the first time in 1990, using 18-Crown-6 ether [2]. It has an hexagonal unit cell with a space group $P6_3/mmc$ and lattice parameters $a = b = 17.45$ Å and $c = 28.46$ Å. The stacking of layers results in a ABAB sequence, with a mirror plane relation between neighbouring layers. The arrangement of sodalites cages creates two types: a "hypocage" [3] (0.5 nm^3), with three 12-membered ring pore openings and a larger "hypercage" [4] (1.3 nm^3),

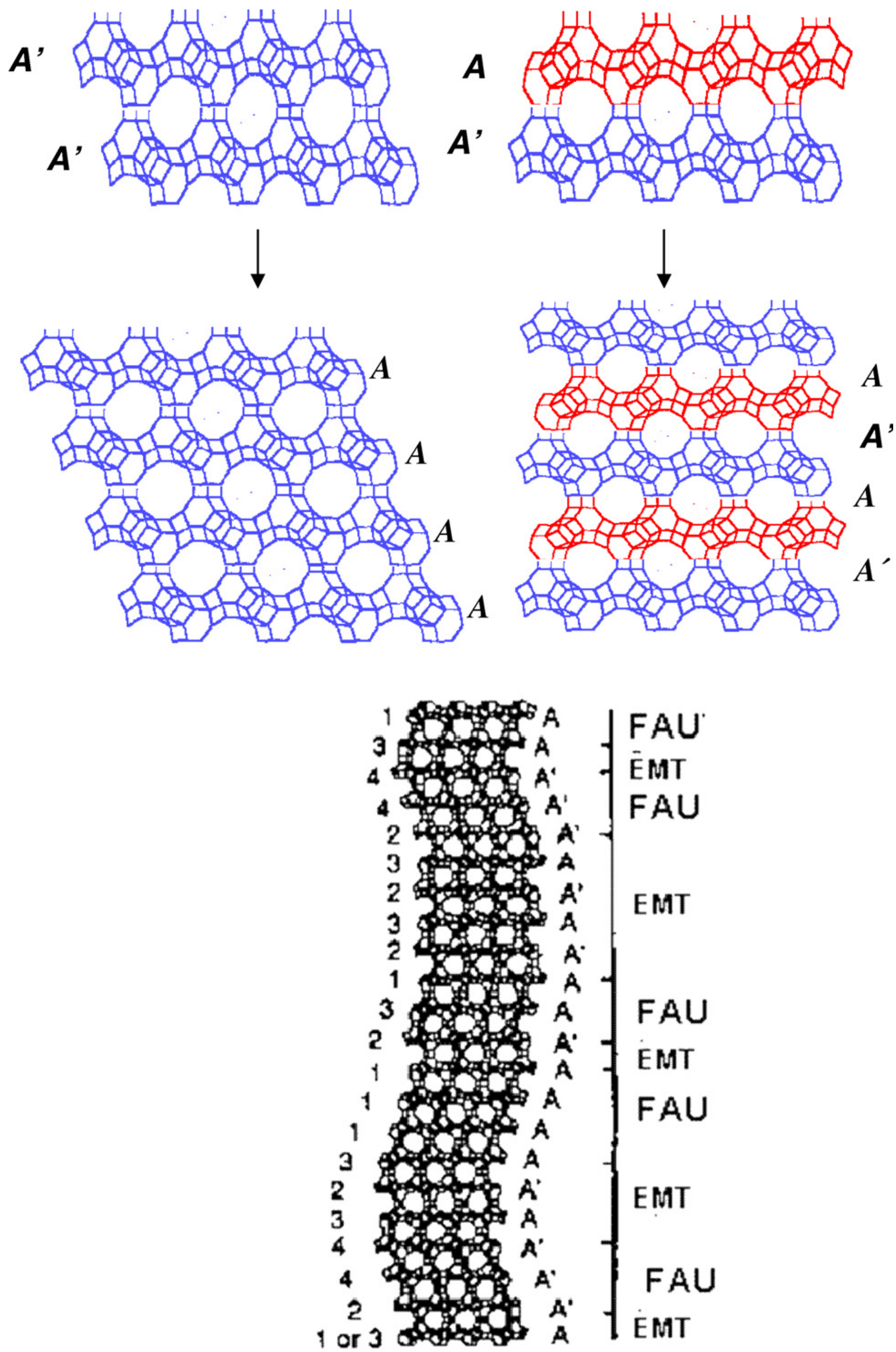

Fig. 1. Layer stacking formation of FAU and EMT structures. Layers AA and A′A′ are related by inversion centers symmetry operations, their stacking results in FAU structure. Layers AA′ and AA′ are related by mirror plane operations, their stacking results in EMT structure. The combination of these results in an intergrowth structure.