

Synthesis of HZSM-5 zeolite with *b*-axis oriented crystals using isomaltulose as a novel modifier of crystallization

Alexander A. Karavaev, Igor V. Mishin, Kirill A. Cherednichenko, Alexander A. Panin and Leonid M. Kustov

Synthesis of HZSM-5 zeolite with *b*-axis oriented crystals

To synthesize HZSM-5 zeolites directly in the H-form with *b*-axis oriented crystals, a mixture containing tetraethyl ortosilicate $\text{Si}(\text{O-Et})_4$, (99%, SIGMA ALDRICH), distilled water, tetra-*n*-propylammonium hydroxide TPA-OH (1 M, MACKLIN)), aluminum isopropoxide $\text{Al}(\text{O-i-Pr})_3$ (>98%, TOKYO CHEMICAL INDUSTRY), isomaltulose (STOING) was used. The molar ratio of reagents $\text{Si}(\text{O-Et})_4$: H_2O : TPA-OH : $\text{Al}(\text{O-i-Pr})_3$: isomaltulose was 1:13.8:0.15:0.008:0.03 for HZSM-5-I-0.03 and 1:13.8:0.15:0.008:0.05 for HZSM-5-I-0.05. Calculated $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio was 280.

Tetra-*n*-propylammonium hydroxide and tetraethyl ortosilicate were dissolved in distilled water and intensively stirred at 65 °C for 6 hours. Then aluminum isopropoxide and isomaltulose were added to the resulting gel and stirred until the mixture was completely homogenized, but without subsequent aging. The resulting suspension was placed in a Teflon autoclave and subjected to microwave treatment in an M6 system (Preekem Instruments, China, frequency 2,45 GGz). Crystallization was carried out for 2 hours at 200 °C. The obtained particles of zeolite, designated as H-ZSM-5-I-0.03 and H-ZSM-5-I-0.05, were isolated by centrifugation in an ultracentrifuge at 1800 rpm, washed with distilled water and dried at 170 °C for 2 hours, then they were calcined at 550 °C for 6 hours to remove the organic template.

Material characterization Powder X-ray diffraction (XRD) data were collected using a Thermo Fisher Scientific ARL X' TRA (Cu – K α radiation, $\lambda = 1.54187 \text{ \AA}$) diffractometer in the range of 2θ angles from 5 to 50°.

The shape and size of the particles, as well as the structure of the synthesized zeolite, were determined with a scanning (F50 Pro, operating voltage 10 kV) electron microscopy, and transmission (JEOL JEM-2100, operating voltage 200 kV) electron microscopy.

The textural properties of the zeolite were determined by a low-temperature N₂ physisorption using a Belsorp Mini-X (MicrotracBEL, Japan) instrument. The specific surface area was calculated by the BET model. The total pore volume was calculated at $p/p_0 = 0.99$ from the adsorption branch of the isotherm. The micropore volume was calculated by the MP method. Mesopore size distribution, volume of mesopores were determined according to the Barrett–Joyner–Halenda (BJH) method from the desorption curve; micropore size distribution was obtained according to the MP method from the adsorption curve; determination of the external surface area was carried out according to the t -plot method using the adsorption curve.