Abstract

The PhD thesis concerns the synthesis of novel zeolite materials, investigation of their properties and their possible use in catalytic application. The work was focused on the two-dimensional zeolites. The thesis was worked out at the Department of Synthesis and Catalysis at J. Heyrovský Institute of Physical Chemistry, AS CR.

Germanosilicate UTL (Si/Ge molar ratio 4.0-6.5) was found to undergo unique structural changes in the neutral or acid environment leading to transformation of its three-dimensional framework into two-dimensional layered material denoted IPC-1P. The UTL degradation, so called top-down synthesis, was enabled due to a presence of double-four-units (D4Rs), which can be seen as supporting units/pillars between the rigid layers. The preferential location of Ge in D4Rs makes the units an ideal target for their selective degradation. The interlayer space in lamellar IPC-1P was modified by swelling with long-organic chain surfactant (material IPC-1SW). To keep the interlayer space permanently expanded (up to 3.3 nm) the silica amorphous pillars were subsequently introduced (material IPC-1PI). The integrity of the layers and their preserved UTL character was confirmed in all members of IPC-1 family by HRTEM and electron diffraction measuring.

The layers of IPC-1P were condensed back and formed new zeolite structures. Depending on the chosen linkage, two novel zeolites were prepared, IPC-2 and IPC-4. The original D4R units in UTL were replaced by new single-four-ring (S4R) units in IPC-2 or by single Si-O-Si bridges in IPC-4. Three zeolites are closely related as they have the same UTL-like layers and differ only in the layer linkers. The size of their channel systems decreases with the size of the linkage between the layers in order UTL (14-12-ring) > IPC-2 (12-10-ring) > IPC-4 (10-8-ring). Zeolites IPC-2 and IPC-4 were verified by IZA Structure Commission as novel zeolites with three letter codes OKO (for COK-14, which is isostructure to IPC-2) and PCR (Prague Chemistry fouR), respectively.

The novel materials, IPC-1PI, IPC-2 and IPC-4, were prepared also with aluminium and they were tested for their catalytic activity in alkylation of toluene with isopropyl alcohol and the pillared Al-IPC-1PI in the reaction of styrene with phenol/*tert*-butylphenol.

The new approach for the synthesis of zeolites was designated the ADoR strategy: first the synthesis of the parent zeolite - Assembly, then its hydrolysis into layered material – Disassembly, followed by organisation of the layers and finally calcination into novel material – Reassembly. The mechanism of the ADoR was studied in detail under varying hydrolysis conditions.

The ADoR concept was successfully applied on other germanosilicate zeolite IWW. Its hydrolysis led to a lamellar material denoted IPC-5P. The layered IPC-5P was converted back to the three-dimensional structure by incorporation of silylating agent. Apart from the zeolite UTL and its IPC-1P, IPC-5P strongly tends to a formation of its original IWW framework. The 3D-2D-3D transformation of zeolite framework is a very unique case, which until now has not been described.