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NMR AND DIFFRACTION STUDY OF ACETONE INTERCALATED IN THE LAYER SILICATE RUB-18

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Interaction of organic molecules with inorganic surfaces is of interest because of a variety of technical applications and because of the environmental impact (OGAWA & KURODA, 1995). In order to study these processes intercalates are used as model system. In particular the analysis of the hydrogen bond network established between guest-molecules and the host-compound are of interest. As simplest molecule for the study of the interaction, acetone has been used (BOROVKOV et al., 1982).

The synthesis of Acetone-Rub-18 was carried out starting from H-Rub-18, a derivative of the Na-form (BOROWSKI et al., 2002), by treatment with acetone or D6-acetone at room temperature. The level of intercalation is about 95%. The sample was stored at room temperature in D_2O atmosphere to avoid D-H exchange. DTA, TGA investigations (measured with a BÄHR STH 503, T=293-463 K, heat rate 3 K/min) show two weak signals at 318 and 382 K which are explained as processes involving the intercalated acetone molecule. In the ^1H MAS NMR spectra (spectrometer: Bruker ASX-400, spinning rate - 12 kHz, dwell time - 4 µs, D1 10 s) there are two signals at 2.0 ppm (methyl group) and 6.0 ppm (silanol group). The high temperature (temperature range 298-430 K) ^1H MAS NMR spectra show the splitting of the two main signals into 5 and 6 signals with greater amplitude than at room temperature, respectively. The chemical shift values of the two signals in a ^{29}Si MAS NMR spectroscopy experiment (spinning rate-3.5 kHz, dwell time- 25 µs, D1- 60 s) are typical for Q^3 ($Si(-OSi)_3(-OH)$) and Q^4 ($Si(-OSi)_4$)-connected silicate tetrahedra, respectively. Powder neutron diffraction data were used to investigate the crystal structure of the composite. The data were collected on the high-resolution powder diffractometer HRPT (PSI Villigen, Swiss) at λ=1.886 Å from 5-163° 2Θ with step width 0.1° The symmetry of the structure of D6-Acetone-Rub-18 was determined as $P4_12_12$. Unit cell parameters were refined with least square methods: a_0=7.479(7), c_0=37.334(9) Å. The location of intercalated guest molecules was determined using Fourier syntheses maps. The geometric parameters of the silicate layers shifted only slightly upon the intercalation. The acetone molecules are located in the "semi cages" between silicate layers and stabilize the structure (Fig. 1).

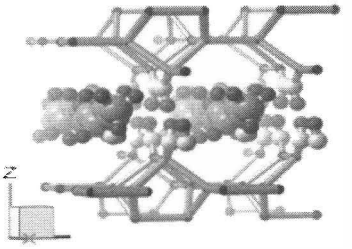

Fig. 1. Fragment of the crystal structure of H-Rub-18 intercalated with D6-Acetone

References

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