Design of low-silica zeolites based on natural halloysite nanotubes

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In this work, application of a natural halloysite nanoclay as a source of silicon and aluminum atoms for the synthesis of various low-silica zeolites has been proposed.

Halloysite with the chemical formula of $Al_2Si_4O_5(OH)_4*nH_2O$ is formed by rolled aluminosilicate sheets, giving nanotubes with an inner diameter of 10-25 nm. An outer surface of halloysite nanotubes (HNT) comprises of siloxane groups (-Si-O), while inner – of aluminol groups (-Al-OH). Such structure makes it possible to modify HNT by various methods. In particular, treatment of HNT with acid/alkaline agents allows to remove selectively silicon or aluminum atoms, thus changing Si/Al ratio.

Within this study, HNTs were subjected to preliminary thermal and chemical (acid) treatment with further recrystallization into zeolites under hydrothermal conditions. Since acid treatment is not the only way to regulate Si/Al, an experiment was carried out in which halloysite was recrystallized in the presence of tetraethoxysilane (TEOS) as an additional source of silicon. The influence of organic templates addition was investigated.

According to X-ray diffraction data, zeolites obtained from thermally activated HNT possessed SOD structure, while those synthesized from acid etched halloysite had ANA structure, both demonstrated spherical morphology of the crystals (fig. 1a). The sample obtained with TEOS also possessed ANA structure, however, it had unique pompon-like morphology (fig. 1b).

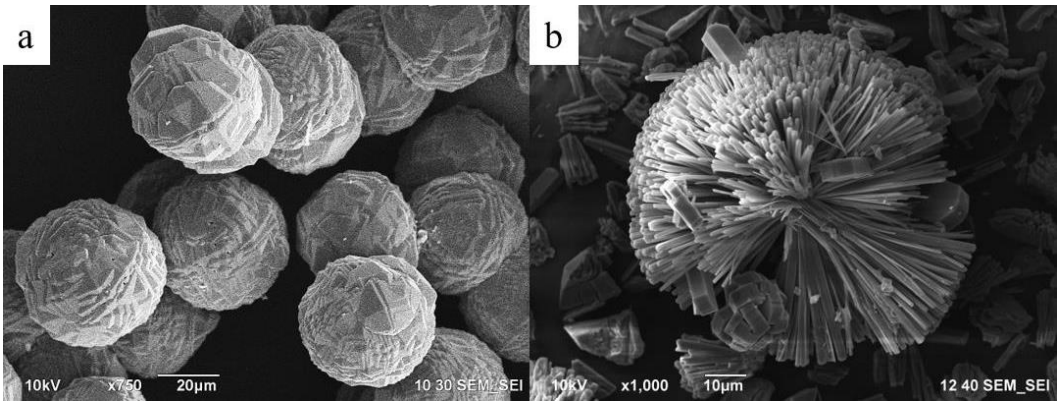

Figure 1. SEM images of the obtained halloysite-based materials

Based on the results obtained, it can be assumed that both HNT and TEOS act as co-templates responsible for the mesopores and crystals of unusual configuration formation.

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