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To date, it was understood that dealumination of zeolite Y was mainly limited to the decomposition of Al^{3+} into extra-framework Al^{4+} species. The present paper shows that the tetra-silicate framework of zeolite Y can be partially decayed to amino-functionalized EFB, a highly stable species (Figure 2 and Figure 2 Figure 2 Figure 2 Figure 2 Figure 2). The extraframework Al^{4+} species introduced in those levels around zeolite Y could be removed by keeping the same zeolite in highly basic medium (e.g., NaOH) for a prolonged time. The NH_4 and OH bonds in the hydroxyl nests of the aluminosilicate host, instead of the framework aluminium, are the source of EFB. The Al/Si molar ratio in the dealuminated zeolite Y was determined as 1.67 by XRD and 13.6 by TGA, while the value of the fresh sample was 1.33. Extraframework Al^{3+} species were found in the form of EFAl, even under milder conditions (solvent-free drying at $80^\circ C$). Instead of the most common approach to dealumination of zeolites, i.e., the formation of secondary pores and/or zeolite amorphization, we showed here the modified

synthesis of zeolite Y samples with presence of considerable amount of cationic species in the form of $\text{Al}(\text{OH})_4/\text{NH}_4/\text{NH}_3/\text{EFAl}$. The latter ones can effectively dealuminate the primary porous network of the initial precursor particles, thereby providing new tools for zeolite dealumination and offer a variety of opportunities for the formation of novel high-silica zeolite Y materials and their utilization in various fields. The achieved materials are suitable for catalysis and other applications. The proposed routes of synthesis provide a molecular approach which allows the synthesis of a variety of zeolite Y precursors with controlled synthesis conditions. Besides, these protocols can be easily implemented in any laboratory performing synthesis of zeolite Y. Thus, this work provides new technological opportunities for the synthesis and subsequent dealumination of zeolite Y and other related zeolite families.

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Rieger et al. [41] have studied the dealumination of the faujasite framework in situ by ion exchange. They were able to exchange 4Al-sites and protons for 1M NH₄Cl solution. The external and interstitial surfaces of the zeolite decreased significantly by up to 45.6 and 32.3%. In contrast to silica, alumina may only be partly replaced by non-framework elements without leaving gaps in the inner part of the crystals [108]. This results again in a tight framework. Noda et al. [105] observed in silicalite-1 that a loss of cations inside the zeolite structure occurs by cation exchange reaction of AlO(OH) units with alkali cations in alkali-assisted dealumination. This cation loss is accompanied by a deformation of tetrahedrally coordinated aluminium in the nanocrystal framework. Some of these small aluminium tetrahedra may be associated with structurally incorporated alkali cations instead of inserted extraframework aluminium cations. Cation exchange reactions may also be

associated with the formation of amorphous aluminium-rich phase inside the framework. Such a phase which is chemically inactive but contains a high concentration of alkali cations is called amorphous hydroxide [109]. It is not fully transformed into a crystalline phase after the removal of extra-framework aluminium cations. This process can be reversible and/or stoichiometric. However, an amorphous hydroxide phase is not stable and after a certain time the Al-OH units will be redissolved into aluminum-containing framework structures again. Especially at high temperatures the transformation of silica is faster than of aluminium. Therefore, in zeolite structures hydrothermal treatment and/or ion exchange leads to a restructuring of the entire structure with the process of changing the composition of the microstructure from one form to another. In other words, the morphology of the sample changes with increasing of time and temperature [110]. The same effect can be detected in the synthesis of zeolite materials with high Si/Al ratio starting from aluminosilicates. The latter

have to be reacted with aluminate solutions at high temperatures in excess of 600°C for the complete removal of the silica matrix which is associated with aluminate-rich zeolitic phases.

In the process of dealumination, the ratio of silica to alumina, which is the main precursor for Na and K ions, does not remain constant but increases with rising temperature [112].

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