Abstract

Electron diffraction (ED) is a powerful tool for the structure determination of crystalline materials. It offers an alternative to single crystal X-ray diffraction (SCXRD) that is often limited by the size of synthesized crystals. Electron diffraction allows analysis of materials at the nanoscale, thus it is particularly useful for samples which crystals are too small for other methods. ED utilizes electrons for collection of diffraction patterns that can be performed in a transmission electron microscope (TEM). Collected ED patterns are further analyzed allowing for the determination of unit cell parameters, lattice type, and even the crystal structure itself. Nevertheless, the effective structure determination from standard ED patterns requires in-depth expertise and data collection is time consuming. Current development of ED methods focuses on the facilitation and automatization of data collection and processing. Notably, highly advanced, continuous rotation electron diffraction (cRED) data collection takes only a few minutes enabling structure determination within a single day.

In this work, I present the utilization of cRED method for the structural characterization of zeolites. These materials are often synthesized as polycrystalline samples with crystals of nanometer in size. This makes zeolites unsuitable for structure determination using the SCXRD. Powder X-ray diffraction (PXRD) is a standard technique for the verification of zeolite structures, however, the structure determination from PXRD patterns is complex and time-consuming, e.g., due to the wide, overlapping peaks. Determination of the structure of zeolites is often essential for understanding their functionalities and potential applications. Traditionally, zeolites are synthesized through hydrothermal methods, which are non-controllable and lead to unpredictable structures. The ADOR synthesis approach offers an alternative strategy for creating new zeolite structures from parent germanosilicates. This stepwise process consists of four steps, namely: assembly of the parent material, disassembly of it into layered precursor, organization of the layers, and reassembly of them to create a daughter zeolite. This method can produce

zeolites that are not possible to prepare by standard methods, however often the quality of ADOR-prepared crystals is not suitable for their structure determination by PXRD.

The aim of this study was to utilize the cRED method to establish a unified workflow for the structure determination of novel daughter ADOR zeolites based on the analysis, mostly focused on the structure, of the parent material. To provide the control over the process optimization both parent and daughter zeolites had to be well-known and described zeolites. Thus, the most studied ADORable zeolite, UTL germanosilicate was used as a parent material. It was utilized to synthesize two daughter zeolites with recognized structures, namely IPC-2 and IPC-4. The structure, morphology, and texture of the parent UTL was investigated at first, providing valuable insights that subsequently facilitated the cRED determination of the structures of daughter materials. The successful description of investigated structures and comparison of the outcome with the literature data confirmed the validity of this method. Subsequently, it was applied to the case study of the structure determination of IPC-20, a recently published daughter zeolite prepared from IWV germanosilicate. Successful structure solution of this novel material further proved the comprehensiveness of established approach that significantly simplifies the structure determination of new ADOR zeolites.