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Methanol intrusion in MFI- Zeolites at High Pressure

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MFI-zeolites are currently used as catalysts in some olefins-production processes, representing an appealing alternative to the high-energy demanding Steam Cracking process, which accounts for 95% of the total worldwide olefins production (Sadrameli, 2016; Arvidsson et al., 2016). More recently, MFI-zeolites have been used in the promising methanol-to-olefins (MTO) synthesis process, which, being able to obtain olefins directly from methanol in place of oil, bears a potential breakthrough industrial impact. At ambient conditions, only the surfaces of the zeolite crystallites are believed to be active in the methanol-to olefins process. However, pressure may favour the intrusion and diffusion of methanol molecules through the zeolitic channels, as observed in other zeolites (Gatta et al., 2018). This phenomenon may bear a significant impact in the industrial applications of MFI zeolites as catalysts, as a "cold" intrusion of methanol into the zeolite cavities might pave the way to increase the efficiency of the MTO conversion process. In this regard, we have synthesized and investigated, by in situ synchrotron powder-XRD, the high-pressure behaviour of six MFI-zeolites with different chemical compositions (framework-Si partially replaced by Al or B and counterbalanced by Na or H as extra-framework cations), by using methanol and silicone oil (as a reference) as P-transmitting fluids, respectively. All the synthesized zeolites are monoclinic (space group P21/n11) at ambient pressure, although a monoclinic-to-orthorhombic phase transition (MOPT) is observed to occur at ~ $P > 0.5$ GPa. Based on the experimental X-ray diffraction patterns and on the high-P evolution of the unit-cell parameters, we ascertain that: i) all the MFI zeolites compressed in silicone oil (acting as non-penetrating fluid) have, overall, the same bulk compressibility, ii) pressure induces the intrusion of methanol through the structural voids and, among the different zeolites, the magnitude of the adsorption phenomenon is different, iii) the MOPT is influenced by both the crystal chemistry and the sorbate (methanol) loading. The experimental findings of this study represent the first step to select the optimal chemical composition of a potential MFI-catalyst for the MTO conversion process operating at high-pressure conditions.

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