Supramolecular organization of water–ethanol solution in ferrierite under pressure

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Turning disorder into organization is a key issue in science. In particular, supramolecular organization induced by external stimuli has opened new paths for the bottom-up fabrication of nanostructures. By using a combined strategy based on the synergy of X-ray powder diffraction experiments and modeling studies [1], we showed that high pressure - in combination with the shape and space constraints of a hydrophobic all-silica zeolite - separate an ethanol–water liquid mixture into ethanol dimer wires and water tetramer squares (Figure 1).

Separation of ethanol from water was accomplished in an all-silica ferrierite (Si-FER), by using as pressure transmitting medium a mixture of (1:3) ethanol and water in the 0.20 to 1.34 GPa pressure range. The system was studied in situ by high-pressure synchrotron X-ray powder diffraction at BM01 beamline at ESRF and refined via first principles modeling.

Upon separation, the confined supramolecular blocks alternate in a binary two-dimensional architecture that remains stable upon complete pressure release. This unique architecture might rationalize the high selectivity of Si-FER in the separation of alcohol–water mixtures, a key issue in biofuel production. Moreover, as ferrierite is an exceptionally selective catalyst for biofuel synthesis in its hydrophilic Brønsted acid form, while only its pore entrances are actually exploited, we suggest that pressure could also be beneficial for the catalytic performance by enforcing the intrusion of reactants.

These results support the combined use of high pressures and porous networks as a viable strategy for driving the organization of molecules or nano-objects towards complex, pre-defined patterns relevant for the realization of novel functional nanocomposites.


Keywords: ferrierite, high pressure synchrotron X-ray powder diffraction, ab initio molecular dynamics simulations