

NANO-CONFINEMENT OF WATER IN SILICALITE AND AIPO₄-5 ZEOLITES: INVESTIGATION OF THE STRUCTURE AND THE DYNAMICS BY MOLECULAR MODELLING

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While silicas are among the chemically simplest of minerals, their structure and dynamics are fascinatingly rich and varied, thanks in part to the subtle interplay of the rigidity of the SiO₄ units and to the flexibility introduced by their association in networks of corner- or edge-sharing tetrahedra. The balance of forces in porous polymorphs like zeolites is even more delicate, so that their phase transition may depend on the presence of defects [1] or occluded molecules, as when adsorption of *p*-xylene in H-ZSM-5 determines a transition from monoclinic to orthorhombic [2]. The framework flexibility is thus an important parameter in problems of molecular adsorption and diffusion in zeolites.

The affinity of zeolites for water is also very varied, depending on the amount and the type of the defects. A perfect silicalite (without structural defects and aluminium) is very hydrophobic while synthesized silicalites are from slightly hydrophobic to very hydrophilic [3]. On the contrary, perfect AIPO₄-5 is very hydrophilic, as is faujasite NaY, containing aluminium and Na⁺ counter-ions.

Molecular dynamics is a useful tool to investigate these materials, either empty or mixed with organic adsorbates, at the nanometric scale. Nevertheless, most simulations:

- assume rigid frameworks ignoring then the flexibility issue,
- poorly represent the inorganic-organic interactions because of the incompatibility of the large charges in oxide models ($q_{\text{Si}} > 2e$) [4] compared to those in typical molecular force fields ($q < 0.5e$).

We provide a new force field for silicas, able to describe their structure and their dynamics and compatible with the molecular interactions with organic systems. We will show that the new force field is valid either for dense silicas like α -quartz or α -cristobalite, or for microporous silica like silicalite. The new force field describes well the affinity of silicalite for water, which will be compared to the adsorption of water in AIPO₄-5.

This study is a necessary preliminary step before encapsulating organic molecules of interest in both hydrophilic and hydrophobic microporous silicas and looking at their adsorption sites and their diffusion.

References:

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