Computational aspects of building blocks, nucleation, and synthesis of porous materials

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Details Participants Abstracts

New developments in the parameterization of force fields based on ab initio training data

Abstract

The synthesis of novel types of biporous zeolites has made significant advances during the last years. Our interest lies in the molecular modeling of zeolite precursors of the MFI type that serve as building blocks for the synthesis of a new class of micro- and mesoporous zeolite materials. [1] These building blocks with intrinsic MFI properties have been identified experimentally [2, 3, 4, 5]: the MFI-precursor (1nm x 1nm x 1.3nm), the MFI-nanoslab (4nm x 4nm x 1.3nm), the MFI-tablet (8nm x 8nm x 1.3nm), ... A good forcefield model that is suitable for gaining atomistic insights in the hierarchical growth process must correctly describe two important observations:

* The small building blocks and the edges of the larger structures are flexible.

* During the synthesis, the non-bonding interactions between building blocks, templates, solvent molecules and directing agents are the determinants for the orientation of the distinct blocks before they are connected by condensation reactions.

From these requirements it is clear that predictive simulations on these building blocks depend on an accurate force-field model for zeolites and organic species that incorporates many types of interactions: (i) valence interactions, (ii) hydrogen bonds, (iii) Pauli repulsion, (iv) London dispersion and (v) electrostatic terms, where the latter are capable of describing the response of a building block to a perturbation in the external electrostatic field. In the literature such a force-field is not available, but one can find several parameterizations that fulfill a subset of the listed requirements.[6, 7, 8]

We present several new algorithms for the parameterization of force-field models based on ab initio training data, that are especially being developed for the derivation of a broadly applicable zeolite-guest force-field, and where we have payed attention to the difficulties that are inherent to the conventional parameterization techniques: parameter correlations, model selection, transferability, ... The first algorithm is the Gradient Curves Method that is well suited for the derivation of a valence force field from ab initio data.[9] We will also highlight our current work that focuses on the parameterization of the non-bonding interactions. The latter is mostly based on methods that are already available in the literature.[10, 11, 12, 13]

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