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Molecular Simulation



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EDITORIAL

Molecular simulation of framework materials

We are pleased to introduce this special issue of the journal Molecular Simulation on the topic of molecular simulation of framework materials. This broad category of materials includes well-established inorganic materials, such as zeolites, the very topical metal organic frameworks (MOFs), as well as covalent organic frameworks and other molecular framework materials. These materials are all built following the same pattern, with an underlying net formed by nodes linked by atomic or molecular bridging units, although the nature of the interactions involved can vary widely from strong covalent bonds, coordinative bonds, $\pi - \pi$ stacking, to hydrogen bonds, etc. Framework materials have received great attention and have been the focus of an ever-growing research effort in the past decade as their properties can be tuned for specific applications, such as gas storage and separation, catalysis, and drug delivery, among others.

In the past few years, the state of the art in molecular simulation of framework materials has moved on from 'simply' simulating individual adsorption isotherms to large-scale screening studies, modelling framework effects such as guest response, thermal expansion, mechanical behaviour or amorphisation to name just a few. Computational characterisation of materials and their properties now cover a large range of time and length scales, from high-accuracy quantum chemistry calculations of structures, electronic properties and excited states, to force fieldbased molecular dynamics studies of diffusion and Monte Carlo simulations of adsorption processes of complex mixtures. Moreover, molecular simulation is nowadays often a key component in large-scale collaborative studies of novel framework materials. Here, in conjunction with the use of various in situ and in operando experimental techniques, it allows to obtain a very detailed picture on the molecular scale and provides physical insight into the behaviour of very complex systems. This trend, which is happening broadly throughout the chemical sciences, has also gained traction in popular depictions of science, where experimental and computational aspects are now seen to go hand in hand (as shown for example in Figure 1).

The aim of this special issue is to provide a resource for the molecular simulation community interested in these materials, covering the development of new simulation methodologies and the application of established methods to novel materials and phenomena. It includes the following contributions:

- Louis Vanduyfhuys et al. offer a novel model for the understanding and prediction of breathing in flexible MOFs, using a semi-analytical mean-field approach.
- Hideki Tanaka et al. propose a potential theory for the gate adsorption in soft porous crystals, based on the Polanyi potential theory of adsorption, using grand canonical Monte Carlo simulations and free energy analysis methods.
- Benjamin Sikora et al. offer a new Monte Carlo simulation method, called Continuous Fractional Component, for the description of adsorption in MOFs in the high-density regime, near saturation uptake.
- Sanliang Ling et al. present a study of the adsorption of small organic molecules in the flexible MOF MIL-53-Fe, highlighting in particular the



Figure 1. (Colour online) Complementarity of computational and experimental chemistry, as depicted by LEGO academic pictures. Reproduced with permission from LegoAcademics. [1]

conformational disorder of the adsorbate and potential of the material for selective adsorption.

- Daiane Damasceno Borges et al. explore the adsorption of small gas molecules on the iron tetracarboxylate MOF MIL-102, by a combination of Density Functional Theory calculations and force field-based Grand Canonical Monte Carlo simulations.
- Wilfried Louisfrema et al. combine neutron diffraction and molecular simulation to elucidate the dehydration-induced cation redistribution in faujasite zeolite Na₅₈Y, introducing a new method for the assignment of cationic sites from the local environment.
- Nicholas Burtch et al. use force field-based molecular dynamics and Monte Carlo simulations to study the behaviour of water near metal sites in a series of functionalised dynamic Zn-DMOF materials.
- Jeffery A. Greathouse et al. explore the adsorption of small gas molecules in pores of small diameter, namely idealised carbon nanotubes. In particular, they study effects of pore entry and diffusion through molecular dynamics simulations.
- Cigdem Altintas et al. demonstrate the potential of molecular simulation for the selection of porous coordination network-based mixed matrix membranes for CO₂/N₂ separations, calculating gas permeability and selectivity of 200 new mixed matrix membranes.
- Stephen A. Wells et al. review the GASP software package for geometric simulations of flexibility in polyhedral and molecular framework structures,

providing insight into the links between local bonding and steric geometry and global flexibility of materials.

- Caroline Mellot-Dranieks reviews advances in crystal structure and electronic structure predictions for MOFs. Focusing in particular on photocatalytic and optoelectronic applications, this paper highlights how the insights gained can lead to guidelines for the discovery of MOFs with targeted properties.
- Jordi Toda et al. demonstrate how important the choice of the force field is for correctly predicting xylene diffusion in 10-ring zeolites where the 10-ring pore opening acts as bottlenecks.

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