Transport mechanisms in heterogeneous pore networks: from molecular to meso-scale observations

Alberto Striolo*

University College London, Department of Chemical Engineering

London, United Kingdom

* a.striolo@ucl.ac.uk

Fluids under confinement exhibit interesting peculiarities and deviations from bulk behaviour. Understanding transport mechanisms and predicting transport properties is important for further developing applications such as catalysis and gas separation, and it is becoming essential for developing unconventional hydrocarbon reservoirs. Much can be learned from molecular simulations. We conducted equilibrium, and sometimes non-equilibrium molecular dynamics simulations for fluids, pure and mixed, confined within extremely narrow pores. We considered aqueous systems containing ethanol, H_2S, and methane, as well as organic systems containing alkanes of various molecular weights and CO_2. We will attempt to quantify and generalize the results, which suggest that depending on fluid composition, pore properties, and pore-fluid preferential interactions, a number of different mechanisms dictate the transport properties of the confined fluids. The results sometimes suggest radically different behaviour compared to bulk fluids, which is often the case when we analyse the solubility of gases (methane and H_2S) in confined water. While detailed experimental verification of the observations can be problematic, quasi-elastic neutron scattering (QENS) has proven useful. We will present recent attempts at reconciling observations form atomistic molecular dynamics simulations and QENS, which suggest that synergistic combinations of experiments and simulations yield a powerful tool to discover transport mechanisms in confinement. However, to further practical applications one needs to up-scale these insights. We will present a kinetic Monte Carlo approach, which yields insights useful in particular for quantifying the rate-limiting mechanisms that currently hinder the production of shale gas.