Propane Dynamics in Porous Silica: A Neutron Scattering and MD Simulation Study

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Studying the structure and dynamics of confined hydrocarbons in porous silica substrates is important as it serves as a good proxy for understanding the interaction of gases with porous silica rich rocks in terrestrial environments. Neutron scattering is a technique that is ideally suited to study structure and processes at the molecular length and time scales. In particular, quasi-elastic neutron scattering (QENS) can be used to study the diffusive motion of a hydrocarbon by investigating the broadening of an elastic line in the spectra. Molecular dynamics (MD) simulations can provide information on the same length and time scales as QENS experiments and thus complement the latter. We have used a combination of QENS and MD simulations to study diffusion of propane confined in 20 nm pores of silica aerogel. QENS data reveals that the translational diffusion of confined propane becomes enhanced at high pressure. This observation is corroborated by the intermediate scattering functions calculated from the MD simulation trajectories. Simulation data show that the rotational motion is out of the energy window of the SNS/BASIS instrument. An investigation of the structure of confined propane shows that the origin of anomalous pressure dependence of diffusivity is a result of a large fraction of propane molecules adsorbed at the pore wall that remain immobile at low pressure while this fraction is reduced at high pressure leading to higher diffusivity. Effect of presence of water on the dynamics of propane confined in 1.5 nm pores of MCM-41-S was also studied using a combination of QENS and MD simulations. Water is found to suppress the mobility of confined propane by building molecular bridges across the pore that block the motion of propane.