Noninvasive Relaxometry Evidence of Linear Pore Size Dependence of Water Diffusion in Nano-confinement

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Abstract: We propose an original experimental method based on NMR at variable magnetic fields experiments (NMRD) and a theoretical analysis of the data that allows probing the spatial dependence of the diffusion coefficient of liquids specifically at proximity to pore surfaces. One of the key results found from these experiments is the linear relationship between average parallel diffusion coefficients and pore radii[1]. Another result is the robustness of the frequency scaling[1, 2, 3, 4] of the master curve approach able to take into account the complexity of the water dynamics at pore surface for samples of different geometries. This approach has proven useful for evaluating the efficiency of the coupling between liquid layers within nano-pore by extracting gradients of diffusion coefficients. The application of this method to water confined in synthesized calibrated nano-pores like MCM-41 for cylindrical geometry[5] and SEOS for spherical geometry[6] has been successful to deal with several dynamical processes on pore surface for different materials. This shows the ability of the proposed method to discriminate between the influence of the geometrical confinement on intra-pore dynamics and the chemistry of the interface induced by different synthesis of the materials. For instance, the frequency selectivity of NMRD profiles has been able to separate the different couplings coming from the spatial heterogeneities on the pore surfaces[1, 2]. This frequency selectivity of NMRD has also allowed discriminating several steps of a complex dynamics process composed by both loops in water and surface diffusion during adsorption events[7, 8, 9]. Based on our experimental and theoretical results, we believe that the proposed noninvasive method allows exploring the interplay between molecular and continuous description of fluid dynamics relevant in physical and biological confinements.

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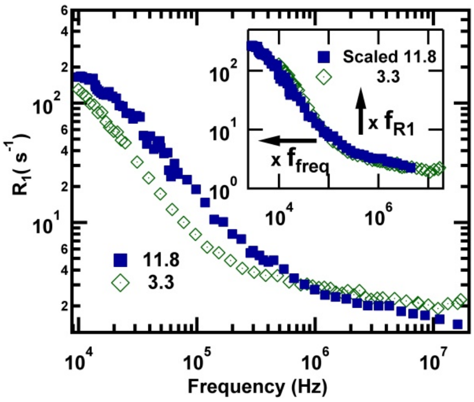

Figure 1 : 1H NMRD profiles of MCM-41 saturated samples with 3.3 nm (diamond) and 11.8 nm (square) pore diameters (main figure). In inset, the master curve obtained by scaling both relaxation rate and frequency by the factors f_{R1} = 1.6 and f_{freq} = 3.3/11.8, respectively is superposed to the MCM-41 of 11.8 nm pore diameter.