2D NMR diffusion-relaxation (DT_2) studies of water in hydrophobic carbon nanotubes

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Introduction

In this work, we investigate, through experimental two-dimensional Nuclear Magnetic Resonance (NMR) diffusion–relaxation ($D\text{-}T_{2eff}$) spectroscopy combined with Molecular Dynamics (MD) simulations, water behavior under extreme nanoscale confinement. To analyze different diffusive mechanisms, we examined the size dependence of water dynamics inside Carbon Nanotubes (CNTs) of different diameters (1.1 nm to 6.0 nm) and temperature range $265 \leq T \leq 305$ K.

Behavior of Water inside CNTs

Carbon nanotubes are able to encapsulate many kinds of materials within their quasi-one-dimensional cavities. Many theoretical studies have shown that materials confined within such small cavities, exhibit novel features which do not appear in the bulk material. In this study we examined water adsorbed inside CNTs in spite of the hydrophobic nature of their wall.

Most of the theoretical studies have shown that water inside hydrophobic nano-channels diffuses faster than bulk water and this enhancement depends on the size of the hydrophobic nanochannels.

Herein, we provide experimental evidence of this dependence by the use of two-dimensional nuclear magnetic resonance diffusion–relaxation ($D\text{–}T_{2eff}$) spectroscopy in the stray field of a superconducting magnet, combined with molecular dynamics simulations. We were able to analyze the size dependence of water dynamics inside Carbon Nanotubes (CNTs) of different diameters (1:1–6:0 nm), in the temperature range of 265–305 K.

The nanotube water is shown to resolve in two or more tubular components acquiring different self-diffusion coefficients strongly depending on the CNT diameter. Notably, a favorable CNT diameter range (3.0–4.5 nm) is observed, in which water molecule dynamics at the center of the CNTs exhibits anomalously enhanced water diffusion, non-Arrhenius temperature dependence, and extraordinary fragility. Such result is of significant importance in the efforts to understand the behavior of water inside hydrophobic nanochannels.

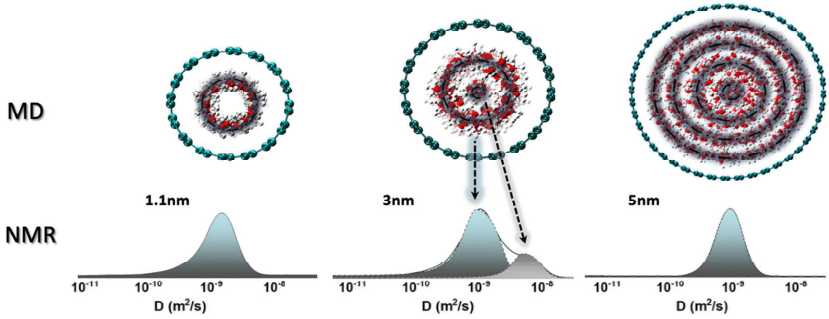

Figure 1. Water diffusion inside CNTs with different sizes (1.1nm, 3nm, 5nm) analyzed by Molecular Dynamic Simulations (MD) and Nuclear Magnetic Resonance techniques (NMR)

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