

Water Dynamics from the Surface to the Interior of a Supramolecular Nanostructure

J.H. Ortony, B. Qiao, C.J. Newcomb, T.J. Keller, L.C. Palmer, E. Deiss-Yehiely, M. Olvera de la Cruz, S. Han, and S.I. Stupp

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Water within and surrounding the structure of a biological system adopts context-specific dynamics that mediate virtually all of the events involved in the inner workings of a cell. These events range from protein folding and molecular recognition to the formation of hierarchical structures. Water dynamics are mediated by the chemistry and geometry of interfaces where water and biomolecules meet. Here we investigate experimentally and computationally the translational dynamics of vicinal water molecules within the volume of a supramolecular peptide nanofiber measuring 6.7nm in diameter. Using Overhauser dynamic nuclear polarization relaxometry, we show that drastic differences exist in water motion within a distance of about one nanometer from the surface, with rapid diffusion in the hydrophobic interior and immobilized water on the nanofiber surface. These results demonstrate that water associated with materials designed at the nanoscale is not simply a solvent, but rather an integral part of their structure and potential functions.

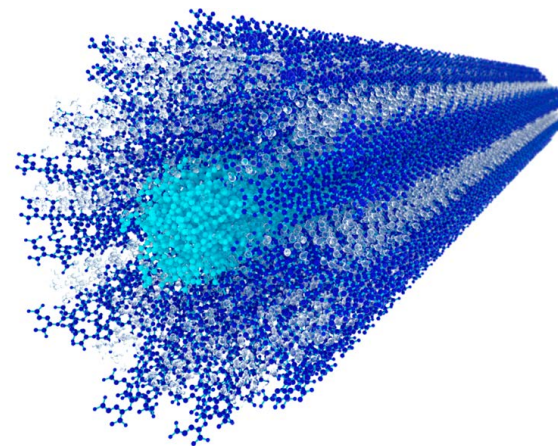


Figure. (a) Schematic illustration of a high-aspect-ratio peptide amphiphile nanofiber, which contains significant internal water, particularly between the β -sheet structures that propagate down the long axis of the nanofiber. Note that the water depicted between β -sheets does not imply structural water and that water at the surface of the nanofiber has been excluded for visual clarity. The peptides are shown in blue, the hydrophobic tails in cyan, and the water molecules in gray.