Dynamics of Bridged Water Molecules in the Vicinity of a Collagen Microfibril

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It is well known that the MRI spin relaxation rates measured in ordered collagenous tissues such as cartilage and tendon are anisotropic across the depth of the tissues. This anisotropy termed as the "magic angle effect" is a consequence of incomplete averaging of dipolar spin interactions due to binding of water molecules to the collagen fibers. Two main hypotheses are proposed regarding the microscopic origin of the magic angle effect: The ice-like water bridges and spatial confinement of water molecules in cavities in the collagen network. This study investigated the nature of the bonds made by water molecules and collagen using a microfibril incorporating molecular dynamics (MD) simulations. All-atom MD simulations of a microfibril consisting of seven tropocollagen fragments in a water box were carried out. The radial distribution function of water molecules was calculated to determine the thickness of the different hydration shells. The continuous residence time of water molecules in each hydration shell was calculated and the molecules that are residing a longer time in the first and second hydration shell were considered for further analysis. The translational motion of long residing molecules was studied to understand their bonding nature and twenty-four long residing molecules (resident time which are longer than 2 ns) were identified. Several bridge sites in collagen that these water molecules make bonds with were also identified. The average duration where the water molecules were translationally restricted was found to be 54.02 ps which is considerably longer than previously reported water bridge residence times which were less than 10 ps. However, during this time, water molecules were not rotationally restricted. Depending on the nature of the hydrogen bonds formed, three types of rotational motion could be identified. Few water bridges that formed between the long residing water molecules and the collagen were ice-like water bridges. However, most bonds between collagen and water molecules were flipping between atoms of the bonded molecules being partially ice-like or trapped.

Keywords: *Collagen, Hydration Shell, Molecular Dynamics Simulation, Magic Angle Effect, Spin Relaxation*

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