On the structure and dynamics of water associated with single-supported zwitterionic and anionic membranes

We have used high-resolution quasielastic neutron scattering (QENS) to investigate the dynamics of water molecules (time scale of motion similar to ~10^{-11} - 10^{-9} s) in proximity to single-supported bilayers of the zwitterionic lipid DMPC (1,2-dimyristoyl-sn-glycero-3-phosphorylcholine) and the anionic lipid DMPG (1,2-dimyristoyl-sn-glycero-3-phosphoglycerol) in the temperature range 160-295 K. For both membranes, the temperature dependence of the intensity of neutrons scattered elastically and incoherently from these samples indicates a series of freezing/melting transitions of the membrane-associated water, which have not been observed in previous studies of multilayer membranes. We interpret these successive phase transitions as evidence of different types of water that are common to the two membranes and which are defined by their local environment: bulk-like water located furthest from the membrane and two types of confined water in closer proximity to the lipids. Specifically, we propose a water type termed "confined 2" located within and just above the lipid head groups of the membrane and confined 1 water that lies between the bulk-like and confined 2 water. Confined 1 water is only present at temperatures below the freezing point of bulk-like water. We then go on to determine the temperature dependence of the translational diffusion coefficient of the water associated with single-supported DMPG membranes containing two different amounts of water as we have previously done for DMPC. To our knowledge, there have been no previous studies comparing the dynamics of water in proximity to zwitterionic and anionic membranes. Our analysis of the water dynamics of the DMPG and DMPC membranes supports the classification of water types that we have inferred from their freezing/melting behavior. However, just as we observe large differences in the freezing/melting behavior between these model membranes for the samewater type, our measurements demonstrate variation between these membranes in the dynamics of their associated water over a wide temperature range. In particular, there are differences in the diffusive motion of water closest to the lipid head groups. Previously, QENS spectra of the DMPC membranes have revealed the motion of water bound to the lipid head groups. For the DMPG membrane, we have found some evidence of such bound water molecules; but the signal is too weak for a quantitative analysis. However, we observe confined 2 water in the DMPG membrane to undergo slow translational diffusion in the head group region, which was unobserved for DMPC. The weak temperature dependence of its translational diffusion coefficient allows extrapolation to physiological temperatures for comparison with molecular dynamics simulations.

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