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Insights into the behavioral difference of water in the presence of GM1

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ABSTRACT

Studies on the structure and dynamics of interfacial water, emphasizing on the properties of water near the surface of biomolecules, are well reported, but there is a lack of evidence on the behavior of water near a comparatively rough surface containing molecules with a bulky head group like GM1. In this report we comparatively analyze the structure and dynamics of water as a function of distance from the lipid head group in GM1 containing lipid bilayers, with the lipid bilayers where GM1 is not present. This approach effectively demonstrates the behavioral difference and hence delayed convergence from bound water to bulk water in the presence of GM1 compared to a relatively smooth surface.
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1. Introduction

Water is the most crucial constituent of lipid membranes, whose interaction is essential to maintain the structure and function of cell membrane [1–4]. Depending on the location in the lipid membrane, water molecules reveal very diverse properties [5] and a number of classifications is considered such as bulk, interfacial, bound and penetrating water according to their position in lipid membrane [6–8].

Studies of water at lipid membrane interface have a long history. Many experimental techniques like NMR [9–11] along with time resolved fluorescence spectroscopy [12–15] and neutron scattering [11,16] have been used to study the structure and dynamics of hydration water. Study of water at protein surfaces [17–20], water next to DNA and RNA [21] were also performed. In bio-mimicking systems such as in micelles, reverse micelles, the structure and dynamics of water were also investigated [22–24]. Beside experiments, computer simulation also aids to understand the interfacial properties of water molecules [25,26]. The orientation and polarization of the interfacial water [27–32], water conductance through carbon nanotube [33], the hydrogen bonding structure of water and of the lipid headgroups has also been assessed [32,34,35] using MD simulations. Theory of solvation of small and large apolar species in water was also developed [36]. The detailed structure of interfacial water for charged lipids has been extensively studied using vibrational sum frequency generation spectroscopy [37], and it was revealed very recently using molecular dynamics simulation study that a mosaic of water orientation exists on the surface of neutral zwitterionic phospholipid bilayers [38]. Though there exist a large amount of data on the structure and dynamics of interfacial water in lipid bilayer with different lipids, the works mostly emphasize on the properties of water near the surface of biomolecules [39–43] and there is a lack of evidences about the behavior of water near a comparatively rough surface containing molecule with bulky head group like GM1 (Monosialotetrahexosylganglioside) which is an important glycosphingolipid, present in the outer layer of the cell membrane [44] and it plays significant role to promote the toxicity of amyloid proteins present in raft membranes [45]. In the current work we mainly want to highlight the effect of difference in surface composition of lipid bilayer on the properties of water and in that respect this work is different from the previous ones as here the water properties are illustrated as function of the bulkiness of the surface. In this report we comparatively analyze the structure and dynamics of water as a function of distance from the lipid head group in GM1 containing lipid bilayer with the lipid bilayers where

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GM1 is not present and found that water behaves in a different way at the moderately rough surface and thus the interfacial property persevered for a prolonged region in comparison to a relatively smooth surface. We have also reported if the variation of water property is due to the difference in composition of lipid bilayer and established that this is solely due to the difference in surface. Although there is a lot of works on water structure and dynamics near the surface of biomolecules, our main motive is to illustrate the change in water properties and hence delayed convergence from bound water to bulk water in presence of GM1.

2. Computational methodology

2.1. System setup

We present an overview of our MD simulation studies of the structure and dynamic behavior of interfacial water in different lipid bilayers. Three independent 100 ns long atomistic simulations were performed for three different lipid bilayers the NAMD2.7 [46] program package with the CHARMM27force field [47] including dihedral cross term corrections (CMAP) [48] which was found to be suitable in many lipid–protein molecular dynamics study [49]. TIP3P water model [50] is used. One structure is ensemble of hydrated POPC (1-palmitoyl-2-oleoyl-sn-glycero-3-phosphocho line) in a bilayer. One is composed of Cholesterol (33.33 mol%) along with POPC and another contains GM1 (0.042 mol%), Cholesterol (0.25 mol%) and POPC. Pure POPC lipid bilayer was consisted of 72 lipids with 4674 water molecules. Cholesterol/POPC bilayer contains 30 Cholesterol molecules with 60 POPC and was solvated by 3549 water molecules. The ternary bilayer (GM1/CHOL/POPC) is consisted of 60 POPC, 30 Cholesterol and 4 GM1, 4 sodium ions to maintain electro neutrality and solvated by 4443 water molecules. The initial configuration was constructed using a web-based interface, CHARMM-GUI membrane builder [51].

2.2. Simulation protocol

All MD simulations were carried out under the isobaric–isothermal (NPT) ensemble with imposed 3D periodic boundary conditions. A time step of 2 fs was used to integrate the equation of motion. The temperatures were maintained at 300 K for the simulations using Langevin dynamics, while the pressure was kept constant at 1 bar using a Nose–Hoover–Langevin piston [52]. The smooth particle mesh Ewald method was used to calculate long range electrostatic calculations [53]. Short range interactions were cut-off at 10 Å. All bond lengths involving hydrogen atoms were held fixed using the RATTLE4 [54] and SETTLE [55] algorithm.

2.3. Analysis

The trajectory analysis was performed with CHARMM (Chemistry at Harvard Macromolecular Mechanics) [56] and the snapshots were generated by VMD [57]. The tetrahedral order parameter (q) in the system is defined as [58]:

$$q = 1 - \frac{3}{8}\sum_{j=1}^{3}\sum_{k=j+1}^{4}\left[\cos\theta_{jik} + \frac{1}{3}\right]^2 \quad (1)$$

Normalization with respect to total number of water molecules was done for the calculation of interaction energy and hydrogen bonds.

The rotational autocorrelation function of dipole vector of water is calculated from the given equation [59]:

$$\Gamma_l(t) = \langle P_l(\hat{\mu}(0)\cdot\hat{\mu}(t))\rangle \quad (2)$$

where P_l are the Legendre polynomials of the order l and $\hat{\mu}(t)$ the unit vector along the P-N molecular axis at time t, the brackets "$\langle\rangle$" denote time average.

3. Results and discussions

3.1. Structure of water

Consistent with previous studies [60], the orientation of water molecules, depending upon the lipid head group orientation was illustrated in Fig. S1 in Supporting information. We have found orientation of H-atoms of waters towards the PO_4^- group and reverse orientation for the choline group which is in accordance with the recent molecular dynamics simulation study [38]. Same type of orientation was also recognized near Cholesterol and GM1 head groups which concur well with the previous findings that the orientation of water molecules depends upon their local environment [31,61,62]. The local structure of water molecules was further estimated by calculating the tetrahedral order parameter q of water molecules [63] along their vertical position.

Depending upon the depth from the lipid surface, solvent layers are divided mainly in five regions, each in a shell of area within 4 Å, from the phosphorus atom of phosphate group up to 20 Å along the Z axis. One more layer exists beyond 20 Å. The q parameter of water in the three systems for the different regions is shown in Fig. 1a. The tetrahedral order parameter of water is very high in the region 0–4 Å, and it is lower at the region 4–8 Å, after 8 Å the averaged tetrahedral order parameter for the rest regions are almost identical for the systems except the system containing GM1. But in the presence of GM1, the values are not same beyond 8 Å; the value drops simultaneously up to 16 Å. The contrast trend in presence of GM1 in our simulation predicts that structure of water is little bit different near the head group of GM1 which has the ordering effect on the structure of water till 16 Å. This is because the presence of GM1 makes water to be interfacial up to 16 Å. Compared to TIP3P water model, the computed q values are little bit higher than those reported earlier [64–66] which may be due to the presence of lipid bilayer. This is supported by the plot showing the average water–water hydrogen bonds and water–lipid hydrogen bonds per water molecule at different distance from the P atom of the bilayer along Z axis in the three systems (Fig. 1b). The hydrogen bonding pattern of water agrees well with previous finding [67]. It can be seen in the GM1 containing bilayer that the number of water–water H-bonds per molecule is greater in the interval of z from 40 Å to 50 Å than that of the POPC/CHOL bilayer and almost doubled in comparison to the pure POPC bilayer which confirms more ordered and compact arrangement of water in GM1 containing system.

For a global structural analysis of water, the atom density distribution of water along bilayer normal was calculated for the pure POPC bilayer and GM1 containing bilayer and plotted (Fig. 2a). Moreover the atom density of the phosphate atom of phosphate group of POPC lipid and the same for the oxygen atom of the saccharide head group of GM1 are also plotted. The peak of oxygen atom for GM1 resides above ~8 Å above the phosphate plane. We have observed in GM1 containing membrane, the density of water 10 Å above from the phosphate plane, is almost 50 units greater rather than in the same region in the pure POPC bilayer. Along with our previous finding, our simulation confirms ordered arrangement of water in presence of GM1 which makes water bound for a longer region than in the pure POPC bilayer.

In order to investigate the energetics of the local structure of water, we have plotted the normalized interaction energy of water molecules with the variation of normalized intermolecular hydrogen bonds as a function of the distance along membrane normal

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