Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Competition of opsonins and dysopsonins on the Nanoparticle surface

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Materials and Methods

Molecular Dynamics

Limited time and length scales accessible in the All Atom Molecular Dynamics (AAMD) and also the wide temporal and spatial scopes of Protein Corona (PC) formation motivated the authors to employ Coarse Grained Molecular Dynamics (CGMD) in the current study. During PC formation besides, the hydrophobic interactions are a major contributing factor[1]. Since the Martini forcefield is calibrated mainly based on the oil/water partitioning energy[2], it is a decent choice for considering the hydrophobic interactions during the course of PC formation. Thus, the Martini coarse grained force field is utilized in the current research to study the PC formation on the Graphene Oxide (GO) surface.

The coarse grained models for Human Serum Albumin (HSA) and Immunoglobulin G (IgG) proteins are made from the all atom pdb codes[3] of 1bm0[4] and 1hzh[5] utilizing the martinize code[2]. Whereas in the case of Graphene Oxide (GO), mapping each benzene ring to three SC4 martini beads, first the coarse grained model of pure graphene is built as mentioned in[6]. Using a homemade C++ code, 40% of SC4 beads are chosen randomly to be changed into either epoxy

(SN0) or hydroxyl beads (SP2)[7]. Afterwards, the simulation box is solvated with Martini water beads.

Martini water molecules usually have such a high freezing temperature that they might freeze in the room temperature[8]. To cope with the problem, it is suggested that 10% of water molecules are changed to antifreeze water beads having slightly larger VdW diameter[9]. In the simulations done in our study, using 10%, 20% and even 40% of the larger water molecules did not solve the issue. It seems that higher crystallinity of GO system with respect to common biological systems has severed the problem here. Even replacing 5% of graphene beads to similar beads with a slightly larger VdW diameters, as suggested in the literature[10], does not work in the current circumstances. As the final solution, the polarizable Martini water model is deployed [11]. In this water model each four water molecules are mapped into two massless beads with opposite electrical charges and a neutral VdW bead[12]. Even though the use of the polarizable water molecules increases the computational costs of the simulations, it deciphers the problem of water molecules freezing on the GO system. Thus, in all the simulation done in the current research polarizable water molecules are utilized.

Before the main simulations, IGG, HSA and GO coarse grained molecules are minimized and equilibrated in separate simulation boxes containing normal Martini water beads. Then, proteins alongside with GO and polarizable Martini waters are placed inside the same simulation box and the energy minimization process is initiated. During this stage, the constraints on the distance between massless water beads are converted into linear springs and there is no constraint on the GO beads. Afterwards, the water bead constraints are redeemed and several beads on the edge of the GO system are constrained. Two 1000 step short equilibration stages having time steps of 10

and 15 fs precedes the main simulations with 20 fs time step. To account for the speedup in the Martini simulation[13], the real time reported in the current study is four times the simulation time.

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