Supporting Information (SI)

Deep Learning Potential of Mean Force between Polymer Grafted Nanoparticles

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High-Dimensional Potential Energy Surface. The effective interaction between two polymer grafted nanoparticles is translationally and rotationally invariant. It depends on the separation between the centroids of the two nanoparticles, the separation distance between the points of grafting, alignment of a point of grafting with respect to the line connecting the two centroids, alignment of a centroid with respect to the line connecting two points of grafting. Therefore, a higher dimensional representation is required to capture all possible variations of the effective potential energy surface. We employ the Behler-Parrinello-type symmetry functions, G^1 and G^2 for this purpose. Figure S1 shows how G^1 and G^2 vary for different combinations of the exponents that are chosen for this study. The symmetry functions and their slops decay to zero at the cut-off distance. In case of radial symmetry functions, the radial resolution is provided by η. The G^1 is physically related to effective coordination numbers and can be considered as a description of coordination at various distances from an central particle. In case of angular symmetry functions, the parameter λ can have values $+1\ and\ -1$ for shifting the maxima of the cosine function to $\theta_{ijk} = 0$ and $\theta_{ijk} = \pi$, respectively. Here, the angular resolution is provided by ζ. In this study, 25 distinct combination of $\eta, \zeta\ and\ \lambda$ are used to represent a wide distribution of angles formed by interacting sites.

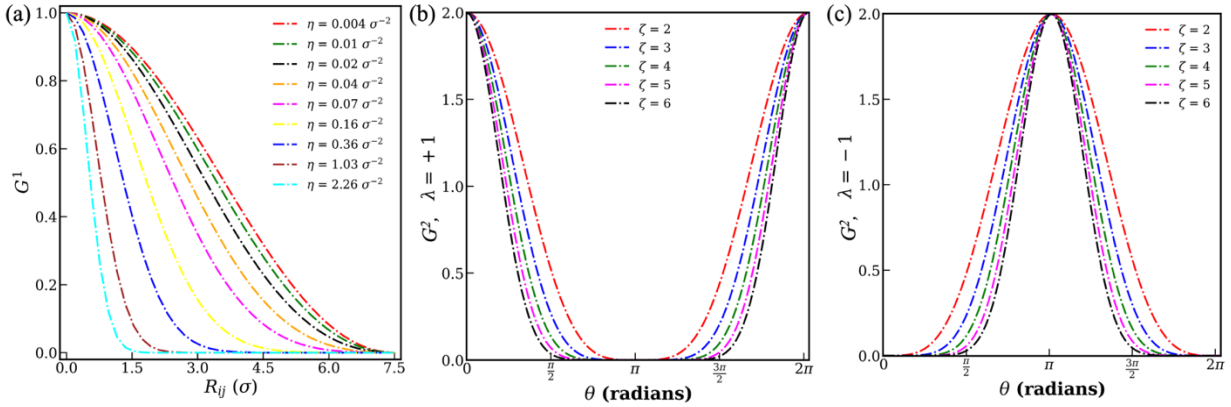

Figure S1: *Radial symmetry functions (G^1) varying from $\eta = 0.004\,\sigma^{-2}$ to $\eta = 2.26\,\sigma^{-2}$ are shown in (a) for eight representative cases. Angular symmetry functions (G^2) for $\lambda = 1$ (b), and $\lambda = -1$ (c) are shown for 10 representative cases. A cutoff distance of $R_c = 7.5\,\sigma$ has been used for G^1 and G^2.*

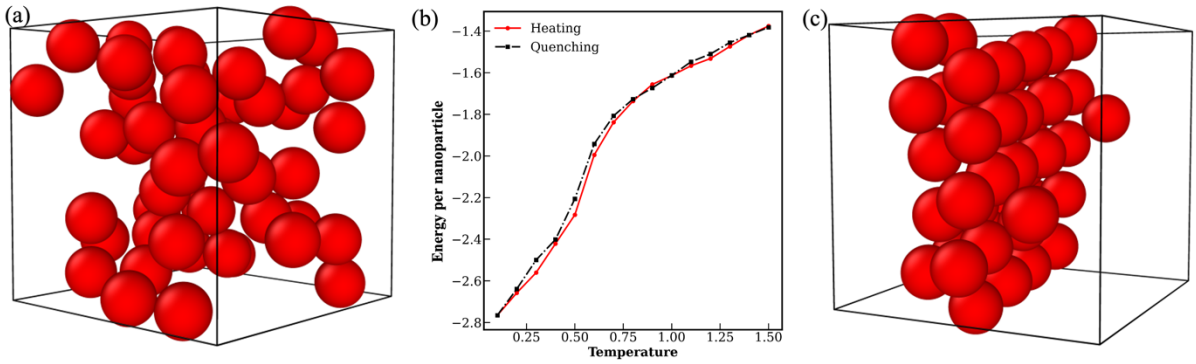

Figure S2: Equilibrium structures of polymer grafted nanoparticles predicted by DL-PMF. The MD snapshot of an equilibrium structure at $T^* = 1.5$ is shown in (a). The energy of the system during a cooling-heating cycle is plotted as a function of temperature in (b). The MD snapshot of the bilayer that has formed at $T^* = 0.24$ is shown in (c).

MD simulations based on deep learning potential. The trained DNN models are used to study self-assembly of nanoparticles. A representative case study is shown in Figure S2 for nanoparticles volume fraction of 0.24. We observe a gradual aggregation of nanoparticles as the system cooled to a lower temperature without much of a hysteresis in energy during a cooling-heating cycle.

The details of the data used for establishing the DL framework. Multiple MD simulations are conducted, each for a given number of NPs and a temperature. The training data set consists of snapshots from all the MD simulations. We have collected ~8000 frames from these MD simulations. The details are summarized in the below Table. This data are divided into 3 sets – training, validation and test sets. The number of data in training, test and validation sets are 5600, 800, and 1200, respectively for developing the deep learning potential of mean force.

Number of Nanoparticles	Temperature	Number of Configurations
50	$T^* = 1.5$	500
50	$T^* = 1.0$	500
50	$T^* = 0.5$	500
50	$T^* = 0.2$	500
30	$T^* = 1.5$	500
30	$T^* = 1.0$	500
30	$T^* = 0.5$	500
30	$T^* = 0.2$	500
15	$T^* = 1.5$	500
15	$T^* = 1.0$	500
15	$T^* = 0.5$	500
15	$T^* = 0.2$	500
2	$T^* = 1.5$	500
2	$T^* = 1.0$	500
2	$T^* = 0.5$	500
2	$T^* = 0.2$	500