Superlattice Formation and Mechanical Response of One-Component Polymer-Grafted Nanoparticle Assemblies: A Coarse-Grained Simulation Study

Arata Kuroishi [1], Yusei Kobayashi [1,2], Masashi Yamakawa [1,2]

[1] Faculty of Mechanical Engineering, Kyoto Institute of Technology, Goshokaido-cho, Matsugasaki, Sakyo-ku, Kyoto 606-8585, Japan, [2] High-Performance Simulation Research Center, Kyoto Institute of Technology, Goshokaido-cho, Matsugasaki, Sakyo-ku, Kyoto 606-8585, Japan

heishixintai@gmail.com

Numerous studies have shown that the mechanical properties of systems composed of polymer-grafted nanoparticles (PGNPs) are strongly dependent on the self-assembled structures formed by these particles. In recent years, increasing attention has been directed toward the self-assembly of polymer-grafted nanocrystals, as polymer ligands exhibit significant conformational flexibility and versatile functional characteristics, enabling the formation of highly ordered and diverse superlattice structures. Previous research has identified grafting density, polymer chain length, and chain stiffness as key parameters governing self-assembled morphologies[1][2]. Although nanoparticle shape is also expected to play a crucial role in determining both superlattice structures and mechanical behavior, its specific influence has not yet been systematically elucidated.

To address this issue, we perform coarse-grained molecular dynamics simulations to investigate the effects of grafting density, polymer chain length, and nanoparticle shape on the formation of ordered superlattice structures (simple cubic phase) in one-component PGNP systems. Our results demonstrate that, even under identical grafting density and polymer length, distinct superlattice structures can be stabilized solely by variations in particle shape, emphasizing the critical role of particle anisotropy in determining equilibrium morphologies.

Furthermore, to obtain a comprehensive understanding of the relationship between structure and mechanical response, uniaxial tensile simulations are performed not only on well-ordered superlattices but also on various structures obtained under different conditions. The results reveal that particle rearrangements occur during deformation, indicating that the internal structure dynamically evolves under applied strain.

Keywords:

Polymer nanocomposite, Superlattice structures, Anisotropic interactions, Mechanical response, Molecular dynamics

References:

[1] C Yu. H Guo. *J. Chem. Theory Comput.* 2024, 20, 1625−1635.

[2] G Hou. X Xia. J Liu. W Wang. M Dong. L Zhang. *J. Phys. Chem. B* , 2019, 123, 2157−2168.