Abstract

Graphene and related materials (GRMs) have shown great promise for applications in flexible electronic devices and thermal management due to their excellent mechanical properties, thermal conductivity, and electrical conductivity. However, GRMs are prone to aggregation during macroscopic assembly, and the regulation of their interfacial structure and function still faces many challenges. GRM may be assembled in a two-dimensional layered structure obtained from a large number of nanosheets through processes such as filtration, layer-by-layer stacking, or vacuum-assisted filtration. When these nanosheets are densely packed and overlapped in a paper-like arrangement, a thin and flexible film structure is formed, which is therefore referred to as "nanopaper." However, in practical applications, the flexibility and structural stability of graphene nanopaper remain limited. Introducing polymers into GRM nanopaper systems can not only help improve their dispersion and self-assembly but also potentially induce unique polymer crystallization behavior through interfacial interactions, thereby significantly regulating the microstructure and macroscopic properties of the material. However, a systematic understanding of the polymer/GRM interface-induced crystallization behavior, especially the formation mechanism and structural characteristics of multiple crystalline populations, is still lacking.

This dissertation focuses on the core scientific question of polymer/GRM interfacial interactions in polymer physics, conducting research from two levels: the regulation of polymer crystallization behavior and GRM dispersion and assembly. The aim is to systematically reveal the key role of the polymer-GRM filler interface in the formation of multiple crystalline populations, the construction of interface-stabilized crystals, and the regulation of the structure and properties of GRM nanopaper.

On the one hand, this thesis focuses on crystalline polymers loaded into GRM nanopaper, systematically investigating the structural origin, thermal stability characteristics, and formation mechanism of multiple crystalline populations phenomena. By controlling polymer content, molecular weight, GRM type, nanopaper preparation method, polymer structure, the formation conditions of different crystalline structures and their intrinsic relationship with interfacial interactions are analyzed in depth and the influence of polymer crystallization behavior on the thermal conductivity

of GRM nanopaper is preliminarily assessed. On the other hand, this thesis systematically studies the role of polymer in the GRM dispersion process, comparing the effects of different solvent systems, small organic molecules, crystalline polymers and amorphous polymers on GRM dispersion and macroscopic properties of nanopaper, exploring effective strategies for controlling the structure and properties of GRM nanopaper based on interface engineering.

In summary, this dissertation systematically addresses the role of polymer on the formation of multiple crystalline populations, the construction of thermally stable interfacial crystals, and the regulation of GRM nanopaper structure and properties. This provides an important experimental guidance for the design of high-performance GRM composite materials based on polymer crystallization engineering and interfacial regulation, and has significant scientific implications and application prospects for the development of flexible electronic devices and thermal management materials.