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Computational modeling of thermal interfaces in graphene based nanostructures

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In my Ph.D. work, the thermal transport across graphene platelets as thermally conductive material was investigated by computational tools, with the aim to contribute to the development of polymeric composites materials with enhanced thermal properties.

Most of the efforts focused on the improvement on the thermal transport in graphene-based nanostructures by the reduction of the thermal boundary resistance within the flakes, for the ideal purpose to create a thermally conductive network of particles. The idea developed in this work relates to the adoption of chemically bound molecules between the graphene platelets, called molecular junctions, able to act as thermal bridges between the graphene platelets. Grafting molecules across the edges of graphene flakes ideally preserve the efficiency in phonon transfer inside the graphene pristine structure and improved heat transfer between the platelets.

The functionalization with species exploiting non-bonding interaction was studied via Molecular Dynamics (MD) simulations as a possible path to reduce thermal resistance among suspended nanoplatelets. Encouraging results from pending alkyl chains induced further investigations on more chemically viable functionalization, such as phenols. However, the use of phenols and other non-covalently bound junctions as thermal bridges provided overall a limited improvement of the thermal boundary conductance (TBC).

Molecular junctions exploiting covalent bond represented a significantly more efficient media to enhance TBC between suspended particles of graphene compared to non-covalent ones. Shorter species were found more efficient to drive heat than longer ones, due to diffusive mismatch of phonons between linker and graphene while a plateauing trend was found for longer chains. Overall, the TBC improvement was found to be the lowest for junctions made of aliphatic moieties, characterized by the long and flexible alkyl chain and the highest for the short and rigid polyaromatic bridging molecules. Moreover, the elastic modulus of the molecular junctions was estimated and found correlated with the TBC. The analysis of vibrational modes between molecular junctions and the adopted graphene flakes found limited overlap for aliphatic/aromatic junctions where aromatic and polyaromatic structures displayed closer vibrational spectra.

The results obtained from molecular junctions bridging suspended graphene platelets were a useful achievement to implement molecular junctions in a context closer to an actual composite. To fulfill this purpose, a polydimethylsiloxane (PDMS) polymer matrix was simulated surrounding the platelets and the molecular junctions. In this latter study, the use of molecular junctions was demonstrated to be an efficient approach to increase the thermal conductance of the interface. In fact, for the chemical species investigated, limited to a partially aliphatic, an aromatic, and a polyaromatic with strong aromatic coupling, the TBC increased up to about 5-times the initial value.

Such improvement in thermal transfer inside the junction was attributed, on the one hand, to the length of the molecular junction that forced the flakes to keep the distance between the platelets and, on the other hand, to the chemistry of the junction, as previously observed in suspended flakes. The strongly coupled junction, based on anthracene, was found able to behave similarly to a uniform material and was thus considered a theoretical upper value for molecularly joined graphene nanocomposites.

Beside the effect of TBC between platelets (filler-filler), inside a composite material, the large area of interaction between polymer and filler is known to have an important role in the heat flux within the

composite. Therefore, to investigate the thermal transport inside a composite material, an upper-scale implementation is needed.

Following this premise, MD and Finite Element Method (FEM) were then coupled in a multi-scale approach to investigate thermal transport in the continuum for novel composite materials. In MD simulations, carried out by the thermal equilibration method, the interfacial thermal conductance (ITC) between the surface of PDMS polymer and two different filler candidates, graphene and borophene were determined. The so calculated values were then fitted into FEM simulations. Overall, the graphene-based composites reported higher thermal conductivities than the counterpart made of borophene flakes did. Moreover, in the models with higher aspect ratio, thus flakes with higher lateral size, were the ones in which the composite conductivity scales more with graphene compared to borophene. Despite this finding, the much lower thermal conductivity (TC) of borophene, two orders of magnitude lower than graphene, was compensated by the slightly higher thermal conductance at the interface with polymer. This phenomenon suggested that an optimal route to improve TC of composites should include proper interface design to improve the boundary conductance, for example by chemical functionalization even at the cost of a moderate reduction of the TC. Nevertheless, the strategies to reach percolation by design of molecularly joined three-dimensional networks still represent one of the most powerful mechanism to improve the TC of composite. Within this approach, the use of molecular linkers contributes to the creation of a percolative network and thus improving composite TC by reducing thermal resistance in filler-filler contact.