ATOMISTIC SIMULATION OF INTERACTIONS OF CARBON NANOPARTICLES WITH ORGANIC COATINGS

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Polyester paint coatings may discolor when exposed to long term harsh weathering conditions. The principal source of contamination has been suggested as rain or air carried carbonaceous particles that adhere to the paint surface. Computational studies of interfacial adhesion between modified polyester and carbon surfaces can provide valuable insight into the design of contamination resistant paint coatings. To enable a realistic nanoscale representation of a cured paint surface, we have developed a methodology for construction of atomistic models of crosslinked polymer networks[1]. In this study, the fully atomistic model of crosslinked polyester has been modified on the atomic scale, using hydrophobic and hydrophilic functional groups, with varying degree of surface coverage. Graphite, amorphous carbon[3] and fullerene models have been used to simulate the carbonaceous contaminants.

Theoretical adhesion calculations[2,4] have been used to characterize the interfacial interactions between polyester and carbon surfaces under a variety of computational protocols which include accentuation of polyester atomic surface roughness (Figure 1) and relaxation of the interface. Adhesion energetics have been calculated using the COMPASS forcefield[5] to describe interatomic interactions. COMPASS (Condensed – phase Optimized Molecular Potential for Atomistic Simulation Studies) is an ab-initio force field with parameters derived from approximate solutions of the full Hartree Fock equations, and optimized for a wide range of condensed matter properties using experimental data. The parameterization to condensed phase properties used in this forcefield is ideal for application to adhesive energy calculations between dense surfaces and has been demonstrated to produce reliable results for our systems of interest [1,2,4,5].

Results show that chemical modification and atomic scale roughness of a polyester surface can significantly affect adhesion with the carbonaceous solids (Figure 2). Relaxation of the interface leads to reorganisation of the polyester substituents, increasing the interfacial contact area and thereby reducing the effects of atomic surface roughness. This is more pronounced at elevated temperatures where the mobility of the polyester components is far greater.

We have also examined the interaction of neutral and charged fullerenes with model silica and polyester surfaces. Classical Molecular Dynamics simulations at 298 K indicate that Van der Waals forces are sufficiently strong in most cases to cause physisorption of the neutral fullerene particle on to the surfaces (Figure 3). The adhesion of C_{60} to both the silica and polyester surfaces depends greatly on the charge on the particle. Crosslinking and functionalization of the polyester can improve resistance to the neutral fullerene. However, for charged fullerenes, the same functionalization can actually reduce resistance to adhesion.

References:

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Figure 1. Schematic representation of polyester / amorphous carbon interface.

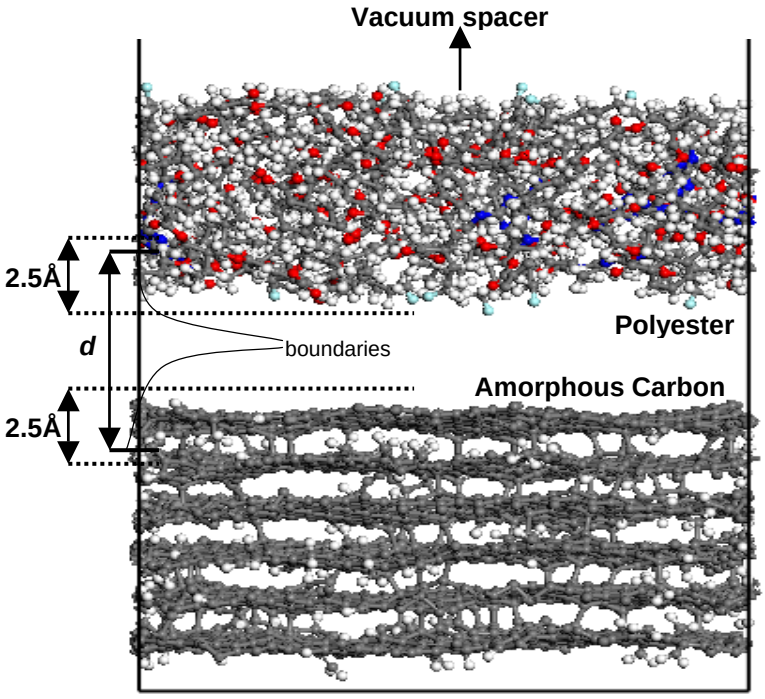

Figure 2. Sample adhesion curves for Graphite – Polyester interfaces showing the effects of nanoscale polyester surface treatments.

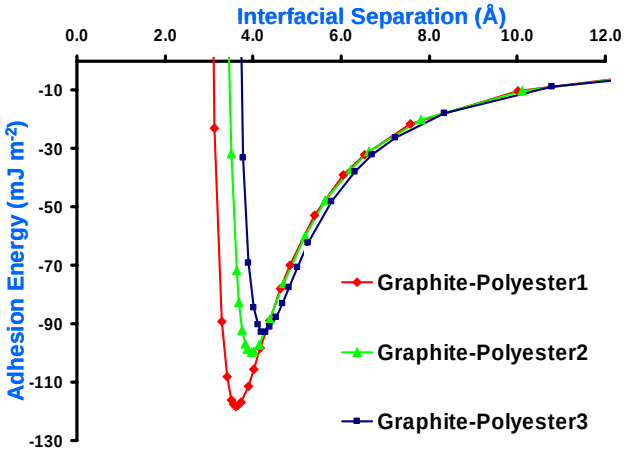

Figure 3. Slice of Van der Waals surface highlighting interaction of fullerene with polyester surface.