Computational Construction of atomistic non-planar char models

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In order to understand the surface chemistry of char, realistic char models are pivotal. In char, the carbon atoms are arranged in an amorphous part and a greater fraction consisting of polyaromatic sheets, resembling stacked graphene-like layers with defects. Atomistic char models commonly focus on flat polyaromatic sheets. In some studies, single sheets are randomly packed, leading to only few stacking motifs, whereas other models employed identical stacked graphene-like layers. However, HRTEM studies of char reveals a large fraction of stacking of curved and non-planar layers.
We propose a novel method for the construction of such char models by starting on the mesoscale with corrugated stacked planes as a template. The atomistic model is generated by the following steps:
First, beads representing the ‚ring centers‘ are distributed on the plane with a soft sphere potential and a force, keeping the beads on the planes. On an ideal flat plane, a hexagonal packing results. Second, the atomisitic carbon positions are determined via a Voronoi tessellation. This results in the formation of graphene sheets on flat planes. On a sphere, fullerenes like C_{60} are formed. It is possible to include defects by gaps in the planes or by using less ring centers than necessary to fill the plane. In a last step, adding hydrogen atoms or functional groups with heteroatoms on dangling bonds can be used to decorate the defects. In a final step, the system is tempered using the reactive force field ReaxFF to remove any unfavorable arrangements.
In this way, fullerenes, carbon nanotubes or stacked graphene layers, can be generated depending on the templating planes initially used. On the long run, this approach will allow the reconstruction of char models using the data from HRTEM images as an experimental boundary condition.