Methane Adsorption on Fullerenes

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Molecules on corrugated surfaces often show competition between the interaction with each other and with the surface sites, leading to a set of phases of various symmetries. Additionally, adsorption of fuels such as methane on substances with large surface areas such as some carbons are very promising with respect to fuel storage [1]. Indeed, many studies on methane storage in various materials [2-6] have appeared in recent years. In this work the physisorption of methane on fullerene complexes is studied by means of molecular dynamics simulations. These simulations give insight into the structure of the methane-fullerene clusters at low temperatures. They provide geometric and energetic information on these clusters at a microscopic level and are thus complementary to mass-spectrometric experiments that measure their overall composition.

A force field for the interaction of methane and fullerenes, as well as the methane-methane interaction is developed. The methane-fullerene interaction is treated with density functional theory via the dispersion-corrected functional ωB97X-D [7,8], methane-methane interaction is computed with CCSD theory. Considering methane in a spherically-symmetric approximation did not yield correct results; therefore atom-atom pair potentials were implemented. In the simulation methane molecules were rigid and the (neutral or positively charged) fullerene mono-, di-, tri- and tetramers fixed in space at optimized geometries. Simulations with clouds of methane molecules surrounding the fullerene complexes at experimental conditions show a clear shell structure, groove and dimple sites. These sites are analyzed both geometrically and energetically. A clear definition for groove and dimple sites is given. The results, for example given in terms of 'magic numbers' as observed in experiments, are in good agreement with experiments [9].

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