

493i Self Assembly of 1,4-Benzenedithiolate-Tetrahydrofuran Mixtures on Gold Surface: a Monte Carlo Simulation Study

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We report a Monte Carlo simulation study of self assembly of 1,4-benzenedithiolate-tetrahydrofuran mixtures on gold (111) surface. Both 1,4-benzenedithiolate (BDT) and tetrahydrofuran (THF) are modeled as rigid molecules. The potential model for BDT molecules is based on the universal force field plus partial charges derived from our recent ab initio calculations. Tetrahydrofuran is modeled by an empirically established united-atom Lennard-Jones potential plus electrostatic interaction from partial charges. The nonbonded interactions between gold atoms and molecules are modeled by Lennard-Jones pair potentials from UFF. The chemical bonding between the sulfur atom in BDT and Au atoms is modeled by a Morse potential. A pseudo two-dimensional Ewald summation technique is used to compute the electrostatic energies. We use grand canonical Monte Carlo method to obtain the equilibrium adsorption coverage of BDT-THF mixtures on Au (111) surface. Canonical ensemble (NVT) simulation is then used to explore further the structural information of the equilibrated coverage. Our results indicate that BDT molecules prefer to anchor to the Au surface with one of the sulfur atoms bonded with Au atoms. The solvent THF molecules form clusters either above BDT monolayer, or occupy the vacancy on the Au surface which is not covered by BDT molecules. BDT molecules adsorb on Au surface perpendicularly with a tilt angle of approximately 20 degrees to the surface normal. In addition, the perpendicularly oriented BDT monolayer forms an ordered structure on Au surface. In comparison, the THF molecules always exhibit amorphous structure on Au surface. The tilting and ordering of BDT molecules on Au do not depend on the BDT coverage. We also compared several different parameterizations of Morse potential, from the published literatures and from our recent quantum mechanical ab initio calculations. We found that these different Morse potentials give qualitatively similar BDT adsorption configurations on Au surface.