

289z Dft Investigation of Platinum Nanoparticles on Graphite

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Our goal is to understand, at a fundamental level, the interactions in a platinum/graphite catalyst used as an anode of a PEM fuel cell and its influence on CO and H₂ oxidation reactions. The metal-metal, metal-support and gas-metal-support interactions can be accurately calculated by using an appropriate density functional, such as the B3LYP, and a sufficient basis set. Several phenomena are investigated as a function of the nanoparticle size, beginning with a single platinum atom on graphite. Due to the graphitic nature of our carbon models, our results should be comparable to an experimental sample of highly-oriented pyrolytic graphite (HOPG). In the DFT studies, preferred adsorbate sites for the Pt clusters on the carbon substrate are investigated, using both periodic and cluster models for the graphite. The graphitic surface models are also geometrically perturbed by introducing structural defects, such as Stone-Wales defects and by introducing carbon vacancies. These structural defects are found to dramatically alter the Pt-C interaction on the surface, as well as the preferred adsorption site. Alternative routes are also presented for stabilizing the Pt-C interactions, such as doping the carbon with other elements, which are shown to alter the electronic structure of the surface. Finally, the adsorption of CO on these carbon-supported Pt nanoparticles is investigated. We find measurable differences between CO on Pt versus CO on the carbon-supported Pt, in terms of both the binding energy and the geometry of the Pt-CO interaction. Ideally, these DFT calculations will be fed into larger-scale calculations, such as Monte Carlo or molecular dynamics, in order to develop a more complete description of fuel cell catalysis.