Size and support effects interplay on large Pt nanoparticles supported on pristine graphene

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The search for efficient catalysts for anodic and cathodic reactions on fuel cells has been receiving substantial attention both theoretically and experimentally. Fuel cells can be an important part of our pursuit to sustainable growth because they generate electricity with high efficiency and low emission of pollutants [1]. Platinum based catalysts are widely studied as options for fuel cells, with Pt nanoparticles and alloys appearing as a viable option due to the large surface area per material volume, which reduces the loading of catalytic material and the overall fuel cell cost. Moreover, nanoparticles catalysts efficiency can be controlled by changing the nanoparticle size, shape, and the support used where the nanoparticles are dispersed, allowing rational catalyst design for different reactions [2,3].

To study the metal-support interactions and the interplay between support and Pt nanoparticle size effects, we present large-scale DFT simulations for metallic systems using the ONETEP linear-scaling DFT program [4,5]. We simulate large platinum clusters with up to 309 atoms interacting with a single layer graphene support with up to 880 atoms. We compute adsorption, cohesive and formation energies of platinum monolayers and nanoparticles interacting with the support to compare the stability of such clusters when supported, and the changes in catalytic descriptors induced by size and support effects.

Our results show that three-dimensional Pt clusters are more stable than the two-dimensional when in contact with the graphene support and that the system size increases the differences in the stability of such clusters [6]. We also observed geometric and electronic changes in the Pt nanoparticles and in the support induced by the interaction, with interatomic expansions, charge redistributions, and changes in the d-band centre [6]. Moreover, we studied how the interaction of the Pt nanoparticles with probe adsorbates, such as O, CO, and CH_3CH_2OH changes with the nanoparticle size and with the support interaction, showing that the interplay between support and nanoparticle size effects should not be neglected when developing new catalysts.

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