From pseudopotentials for the cemical environment to challanges in the computational treatment of catalysis

Paola Nava*[1], Alexander Punter , and Yannick Carissan

[1]Institut des Sciences Moléculaires de Marseille, UMR7313 (iSm2) – Aix-Marseille Université - AMU – Campus St. Jérôme, 13397 Marseille cedex 20, France

Résumé

We have recently developed pseudopotentials for reproducing properties of chemical fragments based on sp2 and sp3 carbon atoms. The pseudofragments retain only one or two explicit protons and electrons and use non-atom-centered potentials, which are however very closed to the position of the carbon atom, so that the whole pseudofragment can be considered as an atomic building block. The pseudopotentials were extracted at the Hartree-Fock level, but they can be employed in the framework of DFT or multireference calculations. We have used the pseudopotentials to reproduce the molecular absorption spectra for large organic molecules and carbon allotropes, and are found to recreate both absorption and ECD spectra to a good accuracy. Besides that, we are interesting in the computational description of metal complex catalytic activities. The comprehension of the reactivity of first-row transition metal catalyst has become crucial in the last decades, as these elements are more available than second- and third-row transition metals. Their computational treatment is challenging, especially if several spin states are involved in a reaction mechanism. We shall see an example on a cobalt intermediate, where the singlet-triplet energy gap is not trivial to obtain accurately for computational methods.

Mots-Clés: pseudopotential, pseudofragment, first, row transition metal, catalysis

*Intervenant