

Computational Studies of Systems Related to the Uranyl Ion, UO_2^{2+}

Colin J. Marsden

IRSAMC, Toulouse, France

I shall discuss first the choice of the "best" computational method for studying actinide compounds; is a version of DFT satisfactory, and if so, which one?

We are interested in the nature of the binding between an actinide and potential ligands. We have studied computationally the interaction between the uranyl ion and over thirty different ligands, both neutral and charged, both mono- and poly-dentate. A statistical analysis of the QSAR type has been performed, to assess the extent to which one can predict ligand binding energies from properties of the isolated ligand. The degree of success obtained is gratifying, but unfortunately the "physical reality" behind the numbers which one obtains is questionable. I shall describe and discuss a rigorous separation of the contribution to ligand binding energies from electrostatic effects and from charge transfer. Extensions of this work to other actinyl ions are in progress.

We are exploring, computationally, the nature of various small molecules related to the uranyl ion. Results will be presented for PuN_2 and PuC_2 . These species will be compared with their better-known analogues PuO_2^{2+} and UO_2^{2+} .