Theoretical modeling of thermodynamical properties of actinide complexes

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The aim of presented work is to establish a new set of thermodynamic constants for plutonium oxides and hydroxydes in gas phase by accurate ab-initio methods. Theoretical studies are driven by the importance of Pu compounds in nuclear science and industry and by large discrepancies in experimental values of these constants [1]. Chemistry of Pu is very complex [2] and its theoretical description requires the use of methods which account for electron correlation and relativistic effects. This poster will show preliminary values of enthalpies of reactions involving oxides and hydroxides of Pu, obtained with relativistic Hamiltonians: Zeroth Order Regular Approximation (ZORA) and Douglas-Kroll-Hess (DKH); various methods: based on density functional theory (DFT) and wave function theories (WFT) which use the concept of complete active space (CAS); various basis sets: all-electron (AE) and pseudopotentials (PP). In modelling of compounds in condensed phase, additional difficulty arises from the need to describe its environment and the compound-environment interactions. One possibility to deal with this problem is to use the Frozen Density Embedding (FDE) scheme [3–4], in which the compound of interest (e.g. Pu oxide) is described by accurate methods (WFT), while environment is modelled by cheaper techniques (such as DFT). This poster will present basic concepts behind FDE.

References

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