Ceria Chemistry at the Nano-scale

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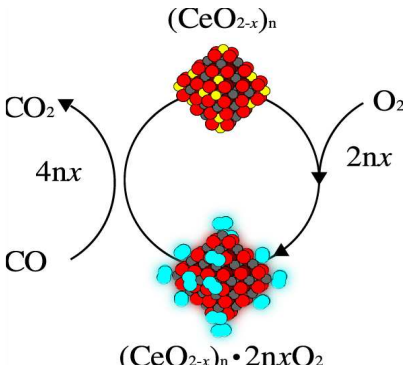
The chemistry of CeO_2 (ceria) is rich and intriguing, and has important technical applications, for example in solid oxide fuel cells and for the purification of exhaust gases in vehicle emissions control – all of this largely a consequence of ceria's exceptional reduction-oxidation properties enabled by the duality of cerium, which easily toggles between Ce^{4+} and Ce^{3+} thanks to its 4*f* electron.

In this talk we will discuss ceria clusters and nanoparticles in reducing, oxidative and humid environments and the powerful analyses and predictions that can be achieved though computational materials modelling. Realistic materials modelling calculations need to be able to mimic these intricate details and at the same time treat large (and dynamic) systems. This is a challenge.

We have used a range of theoretical methods to study ceria, including DFT, DFTB (DFT-based tight-binding), as well as force-field calculations with a newly parametrized reactive force-field – all within a multi-scale simulation environment.[1-4] Based on the quantum-chemical calculations, we find that small ceria nanoparticles of certain shapes (such as perfect octahedra) under low-temperature conditions can be stabilized through the adsorption of oxygen molecules in the form of superoxo species (in agreement with experimental studies; blue in *the figure*), and water in the form of hydroxo species. Moreover, based on force-field simulations we can predict the relative stabilities of very large ceria nanoparticles of different shapes. These and other examples will be discussed.



References

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