Disorders in Ceria: Insights from Embedded-cluster and Supercell Calculations

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This presentation focuses on the current understanding of defect chemistry in ceria from theoretical modelling using embedded-cluster and supercell approaches. We combined Mott-Littleton defect calculations, hybrid quantum mechanics/molecular mechanics (QM/MM) embedded-cluster approaches,[1] plane-wave DFT calculations, and Monte Carlo simulations in developing a thorough understanding of the disorder in ceria. We developed a robust shell-model potential for ceria, considering accurate ionic polarisabilities, defect structures, and formation energies calculated by QM/MM.[2] These developments set a firm basis for our ongoing and future research. Our shell-model potential supports accuracy defect modelling at the dilute limit using QM/MM models with hybrid functionals, or large-scale atomistic simulations of highly disordered reduced ceria (CeO_{2-x}) using Monte Carlo techniques. Our potential and QM/MM models have also supported the understanding of experimentally observed variations in band edge positions and work functions in ceria and other metal oxides.[3] By accounting for the advances in the theoretical modelling of disorders in ceria, we aim to elucidate the origins of its widespread applications in catalytic science.

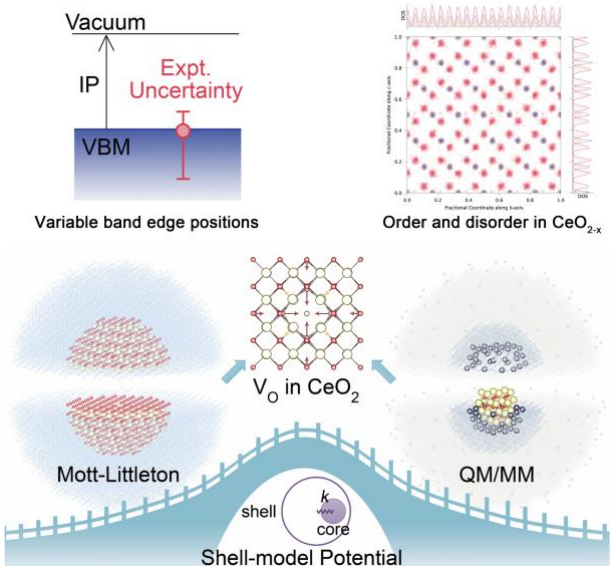

Figure 1: A new shell-model potential supports various embedded-cluster and supercell calculations, improving the understanding of defect chemistry and band structure of ceria.

Acknowledgements

The authors acknowledge the use of the THOMAS, YOUNG, and ARCHER2 UK National Supercomputing Service (http://www.archer2.ac.uk) via membership of UK's HEC Materials Chemistry Consortium, which is funded by EPSRC (Grant No. EP/X035859, EP/P020194, EP/T022213, and EP/R029431). We also acknowledge funding provided by EPSRC under Grant No. EP/W014580, EP/W014378, EP/W026775, EP/R001847, EP/K038419, and EP/I030662.

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