

## Radiation Damage in Complex Oxide Materials – Accurate yet Fast Simulations by Fitting Atomistic Potentials

Researchers at the Australian Nuclear Science and Technology Organization and the University of Sydney have used the atomistic simulation tool GULP to study the  $(\text{Sr}_{1-3x/2}\text{La}_x)\text{TiO}_3$  perovskite system. The atomistic method represents significant time and cost savings over previously used density functional theory (DFT) methods, and accurately reproduces bulk and short-range properties including local order/disorder effects.

Oxide materials are suitable host materials for high-level radioactive waste and it is thus important to understand the atomic-level radiation damage effects that are difficult to explain experimentally. Reporting in the journal *Nuclear Instruments and Methods in Physics Research B*<sup>1</sup> the researchers used the GULP<sup>2</sup> program to study  $(\text{Sr}_{1-3x/2}\text{La}_x)\text{TiO}_3$  perovskite materials that show interesting behavior under external radiation owing to their complex structures.

Previous atomistic simulation studies have often miscalculated key properties such as bulk modulus, thermal expansivity, heat capacity, and defect energies. The scientists successfully used the forcefield fitting capabilities of GULP, and developed a forcefield that accurately reproduces bulk and short-range properties including local order/disorder effects.

GULP was first used to investigate different forcefield potential expressions in their ability to fit  $\text{TiO}_2$  crystal structures. This led to a simple Born–Mayer pair potential model with partial charges being adopted as the simplest and most practical. It has only two fitted parameters per atom type plus the charges, which can be determined from DFT methods using Mulliken population analysis.

The parameter fitting proceeded by first using published crystallographic and elastic constant data to develop a model for perovskite  $\text{SrTiO}_3$ . Next Lanthanum was added by fitting to a series of ab initio generated structures for  $(\text{Sr},\text{La})\text{TiO}_3$  perovskites.

The results were tested against ab initio structures, relative energies, and formal charge models (Figs. 1, 2), and indicated that the model is able to describe defective states making it suitable for simulating the atomic level effects of radiation damage.

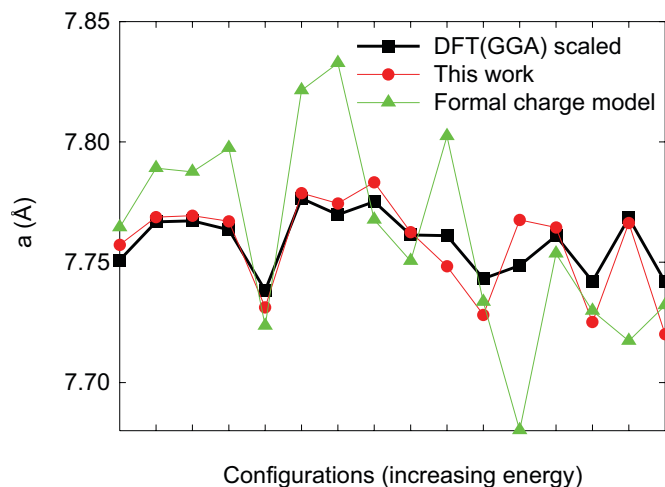


Figure 1. Model testing against ab initio structures, relative energies and formal charge models.

### Module used

GULP

### Industry sectors

Nuclear

Power Generation

### Organizations

Australian Nuclear Science and

Technology Organization

The University of Sydney

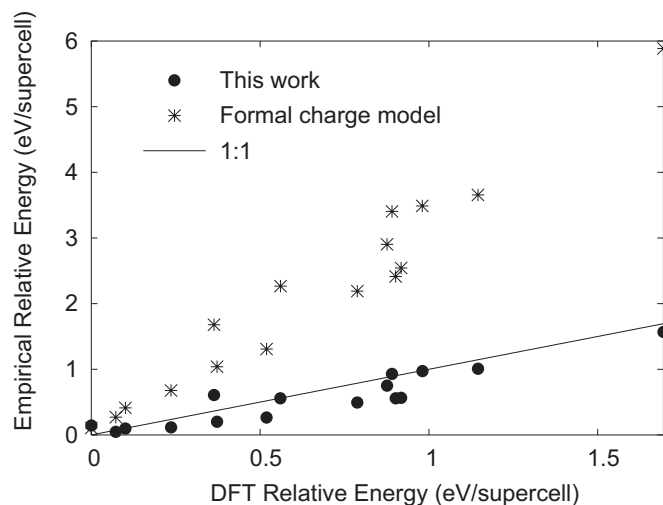


Figure 2. Model testing of empirical versus DFT relative total energy.

Dr Thomas, Australian Nuclear Science and Technology Organization, said, “The computational tool (GULP) enabled the generation of a simple but accurate forcefield; this has been used in further work on defect ordering and radiation damage in these complex perovskites.”

“Experiments have revealed some interesting phenomena - short- and long-range vacancy ordering, as well as unexpected radiation resistance properties - but could not explain them. Computational methods are more suited to exploring the basic physics of such atomic-scale phenomena. In particular, our recent work on the physics of defect ordering in (Sr,La)TiO<sub>3</sub> perovskites has recently been submitted to *Phys. Rev. B* and is currently in review.”

The study demonstrates how the combination of DFT methods, experimental data, and GULP forcefield fitting methods can be used to develop simple, fast yet accurate models of even quite complex materials.

#### References

1. Thomas, B. S., Marks, N. A., Begg, B. D., *Nuclear Instruments and Methods in Physics Research B*, **2005**, 228, 288–292.
2. Gale, J.D., Rohl, A.L., The General Utility Lattice Program (GULP), *Molecular Simulation*, **2003**, 29, 291.