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First-Principles DFT Study on the Effect of Lithiation on the Spinel LixMn2O4 Structure: Calibration of CASTEP and ONETEP Simulation Codes.

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Li-Mn-O layered-spinel composites are among the promising and economically viable, high energy density cathode materials for improving the performance of lithium ion batteries. A number of studies have focused on the specific capacity of these composite materials. However, the complex internal structural changes affecting their performance during the discharge process (lithiation) are not yet fully understood. As such, we perform the spin polarised density functional theory DFT calculations using the CASTEP (traditional DFT) and the ONETEP (linear-scaling DFT) codes to elucidate the effect of lithiation on the electronic structure of spinel LixMn2O4 ($0 \leq x \leq 2$). The electronic structure calculations were performed under the generalized gradient approximation (GGA). Electronic structure analysis depicted semiconducting properties for delithiated-Mn2O4 with a band gap of ~0.65 eV whilst, LiMn2O4 and lithiated-LiMn2O4 were found to be conductors. Furthermore, it was found that less amount of energy is required for electrons to occupy the eg orbitals of LiMn2O4 than of the eg orbitals of the delithiated-Mn2O4. This indicates that lithiation favours Mn3+ which is in line with what was observed experimentally. The LiMn2O4 Density of States (DoS) calculated with ONETEP clearly distinguish the dx2-y2 and dz2 orbitals. The dx2-y2 orbital is filled and the dz2 orbital is empty, which is consistent with the dual-existence of Mn4+ and Mn3+. We also performed a scaling test with ONETEP on supercells of LiMn2O4 spinel structure and the best performance was achieved by ensuring that the product of MPI processes and OMPI_THREADS are equivalent to the requested number of cores in the Lengau cluster. Our current findings forms a basis for moving from traditional DFT to linear-scaling DFT which will enable the study of the electronic properties of Li-Mn-O layered-spinel nanoarchitectures.

Apply to be considered for a student ; award (Yes / No)?

Yes

Level for award;(Hons, MSc, PhD, N/A)?

MSc

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