A Theoretical Study of the Electronic and Magnetic Properties of Pristine and Defected Bulk and Monolayer MoO_3 based on a First Principle DFT calculations

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ABSTRACT

The need for new magnetic, ion conducting and optical materials in the current era of quantum computing, sensing and artificial intelligence, has emerged as an area of renewed significance. However, designing materials for such applications where even slight changes in atomic positions could affected functionality, required accurate atomistic and energetic understanding of materials. Hence, a first-principle Density Functional Theoretical (DFT) study of the electronic and magnetic properties of defected monolayer and bulk MoO_3 was conducted based on methods implemented in Burai version 1.3 and Quantum Espresso version 7.1. Based on these calculations, an indirect band gap of ca. 1.85 eV for bulk MoO_3 obtained from density of states (DOS) and projected density of state (PDOS) reveal the semiconducting nature of this bulk material. Point defect formation energies are calculated using the supercell methodology with a supercell size of 4 x 2 x 4. Substitutional defects including native defects and also the incorporation of vanadium and sulfur were considered for both Mo-rich and O-rich conditions. The effects of such changes in the lattice composition on the magnetic properties of both the bulk and monolayer MoO_3 are also assessed as well as the incorporation of certain defects which could lead to "switchable" magnetic behaviour, controlled by defect concentration with possible quantum computing applications: qubits.

Keywords: Monolayer, crystal-defect, molybdenum-oxide, lattice, DFT

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INTRODUCTION

Molybdenum oxide (MoO_3) has been the center of significant research efforts both in the experimental and theoretical realms due its interesting structural, chemical, optical and related properties which could allow application in various areas.

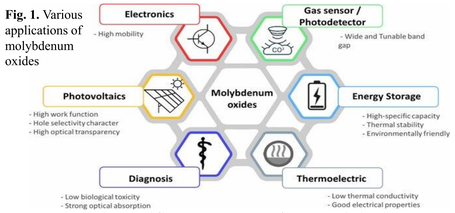

Fig. 1. Various applications of molybdenum oxides

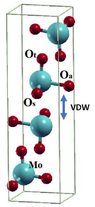

Fig. 2. Unit cell of bulk MoO3 showing Mo, different crystallographic positions of O and direction of VDW forces

However, though significant amount of research has been conducted on this material, the effects of crystal defects on the relative stability of MoO_3 as well as magnetic properties remains an open question. Hence, in this study, a first principle DFT investigation into the effects of crystal defects on the electronic and magnetic properties of bulk and monolayer MoO_3 is conducted.

METHODOLOGY

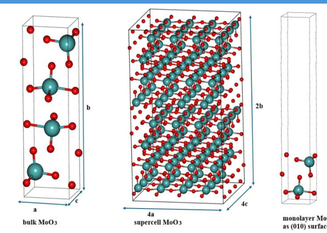

Fig. 3. Cell models for MoO3

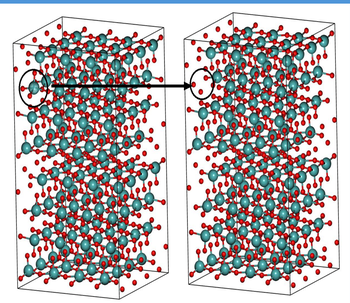

Fig. 4. Schematic diagram of modeling vacancy of Mo

Computer tools used:

B1

QUANTUMESPRESSO

Defect formation energy calculation formula:

$$E_{form} = E_{def} - E_{bulk} + \sum_i n_i \mu_i + qE_F + E_{corr} \quad (1)$$

- E_{def}: Defect formation energy
- E_{bulk}: Total energy of supercell with defect
- $\sum_i n_i \mu_i$: Total energy of bulk supercell
- qE_F: Chemical potentials from phase stability
- E_{corr}: Product of electric charge and Fermi level of pristine supercell
- (1): Correction term

RESULTS

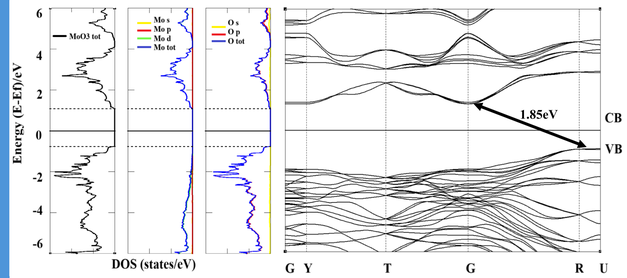

Fig. 5. From left to right: density of states and band gap of bulk MoO3

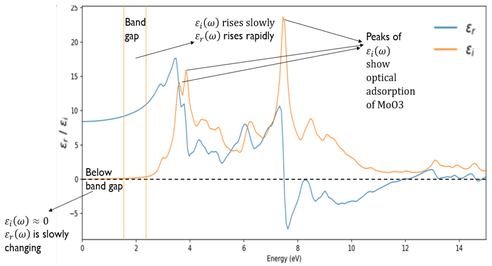

Fig. 6. Real and imaginary part of a dielectric function as a function of energy

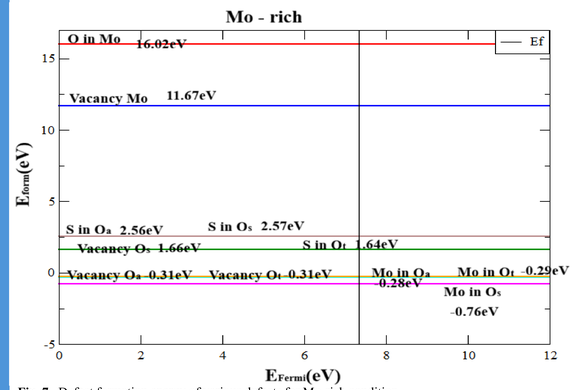

Fig. 7. Defect formation energy of various defects for Mo-rich condition

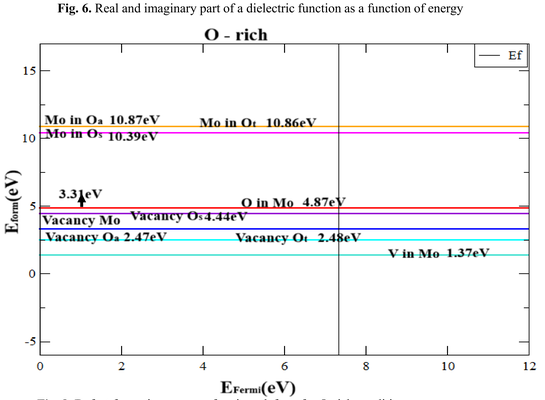

Fig. 8. Defect formation energy of various defects for O-rich condition

CONCLUSIONS AND FUTURE WORK

- Indirect band gap of 1.85eV was obtained from the DFT calculations indicating that in native state MoO_3 is a broad band gap semiconductor.
- For Mo-rich environment the lowest formation energy of neutral defect belongs to substitutional defect such as Mo in position of O and vacancy of terminal and asymmetric oxygen.
- For O-rich environment the lowest formation energy of neutral defect belongs to substitutional defect such as V in position of Mo and vacancy of terminal and asymmetric oxygen.
- Future work predicts completion of defect formation energy calculation for native defects of bulk and MoO_3 monolayer.