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Computer Simulation and Phase Diagram Prediction of Li2S1-xSex Systems

Lithium-sulfur (Li-S) batteries are an attractive Li-ion alternative that provides large capacity (1672 mAhg−1) and energy density (2500 Wh kg−1) while also being low cost, earth-abundant,and lightweight [1–3]. Li/S suffers from the low conductivity of S and the solubility of intermediary polysulfide species during cycling. It has been reported that Se and mixed SexSy represent an attractive new class of cathode materials with promising electrochemical
performance in reactions with both Li and Na ions [4]. Notably, unlike existing Li/S batteries that only operate at high temperature, these new Se and Li/SexSy electrodes are capable of room temperature cycling. In order to study large systems and impact of temperature effectively, empirical interatomic potentials of Li2S and Li2Se were derived and validated against available experimental and calculated structure, elastic properties and phonon spectra. In addition, complex high temperature transformations and melting of Li2S and Li2Se were reproduced, as deduced from molecular dynamics simulations. Furthermore, cluster expansion and Monte-Carlo simulations were employed to determine phase changes and high temperature properties of mixed Li2S-Se. The former generated 42 new stable multi-component Li2S-Se structures and ranked metastable structures by enthalpy of formation. Monte Carlo simulations produced thermodynamic properties of Li2S-Se system for the entire range of Se concentrations obtained from cluster expansion and it demonstrated that Li2S-Se is a phase separating system at 0K but changes to mixed system at approximately 350K which was confirmed by constructed by phase diagram of Li2S-Se system. It was finally demonstrated that molecular dynamics and Monte Carlo simulations techniques yield consistent results on phase separation and high temperature behavior of Li2S-Se at 50% of sulphur and selenium. This work involved a series of computational codes such as DL_POLY, LAMMPS, VASP, Monte Carlo, Cluster Expansion, Material Studio and GULP. High performance computing played a significant role in terms of time taken to complete the calculations. UNCLE code which is within MedeA environment which handles complex systems there was an initial benchmark between local cluster and CHPC Lengau which we found out that the latter produced fast and accurate results.

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