

Information about the Dataset Provided

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Introduction

This document outlines the dataset provided for the publication titled "Insight into the Li/LiPON Interface at Molecular Level: Interfacial Decomposition and Reconfiguration" authored by Kangli Wang, Jürgen Janek, and Doreen Mollenhauer (available at <https://doi.org/10.1021/acs.chemmater.4c00377>). Within the dataset, folders typically contain subfolders, each representing specific surface and interface configurations considered in the study.

Computational details

All total energy calculations were based on density functional theory (DFT) as implemented in the Vienna Ab-initio Simulation Package (VASP).¹⁻⁴ The Perdew-Burke-Ernzerhof (PBE) density functional within the generalized gradient approximation was employed to describe the exchange-correlation interaction.⁵ The van der Waals interactions were incorporated using the approach of Grimme through the D3 dispersion correction with Becke-Jonson damping.^{6,7} The interactions between core and valence electrons were described by projector-augmented wave potentials (PAW P).^{8,9} A plane-wave cutoff energy of 500 eV was employed to optimize the geometric structures, including bulk, surface, and interface. Moreover, convergence during the self-consistent field iteration was considered to be achieved when the energy change was less than 10^{-5} eV, and for ionic relaxations, the convergence criterion was set to atomic forces less than 0.01 eV/Å. For the optimization of the bulk, we set the Monkhorst-Pack k-points mesh to $20 \times 20 \times 20$ for the conventional cells of Li (space group: $Im\bar{3}m$), Li₂O (space group: $Fm\bar{3}m$), α -Li₃N (space group: $P6/mmm$), β -Li₃N (space group: $P6_3/mmc$), and Li₃P (space group:

$P6_3/mmc$), $12 \times 12 \times 12$ for β -Li₃PO₄ (space group: $Pmn2_1$), $12 \times 12 \times 6$ for γ -Li₃PO₄ (space group: $Pnma$), $8 \times 16 \times 16$ for 1D-LiPON and $8 \times 8 \times 8$ for 2.5D-LiPON. For the optimization of the surface structure, the k-points samplings were reset based on the geometry of each structure with one k-point in the surface normal direction and the same number of k-points in-plane as in the bulk calculations. For the optimization of the interface structure, the k-points samplings were adjusted inversely proportional to the size of the supercell to maintain the same k-point grid density as in the bulk calculations.

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