Exploring Surface Moisture Stability of Cs-Doped $FA_xMA_{1-x}SnI_3$ Perovskites via Machine Learning Interatomic Potentials and First-Principles Calculations

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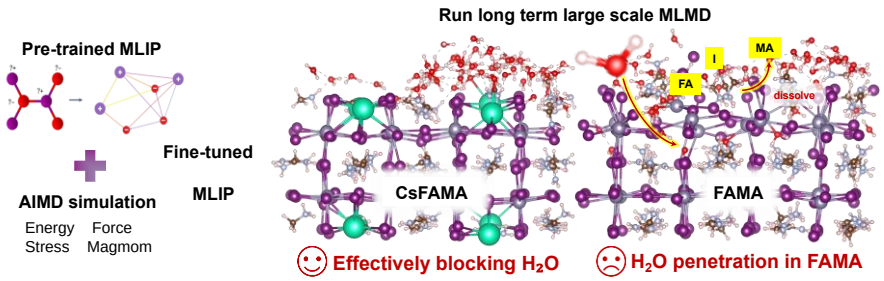
Perovskite solar cells (PSCs) are the most promising devices for converting solar energy into electricity due to their high efficiency. Although Pb-based PSCs have achieved an impressive efficiency of 25.2%, the environmental toxicity of Pb-based perovskites has driven the search for alternative materials, with Sn emerging as a potential substitute for Pb. Sn-based perovskites still face significant challenges, including short lifespans and rapid degradation. To overcome these limitations and enhance the interfacial stability of the $FA_xMA_{1-x}SnI_3$, recent researches focus on strategies such as mixing cations. By replacing the A-site cation with Cs doping has been shown to enhance the stability of Sn-based perovskites.[1] To investigate the mechanism behind this improved stability, we employed theoretical calculations to analyze the formation energy and Gibbs free energy of $FA_{0.75}MA_{0.25}SnI_3$ (FAMA) and $Cs_{0.125}FA_{0.75}MA_{0.125}SnI_3$ (CsFAMA), we found that CsFAMA exhibits lower formation energy and Gibbs free energy. These results suggest that Cs doping enhances the structural stability of perovskites. To further examine moisture stability, by constructing interfaces of FAMA and CsFAMA with water molecules respectively, and performed Ab initio molecular dynamics (AIMD) simulations to uncover the degradation mechanisms. Although AIMD provides precise descriptions of electron interactions and atomic forces, it is computationally expensive. To balance accuracy and efficiency, a pre-trained machine learning interatomic potential, CHGNet, and fine-tuned with AIMD datasets as the training data. With the fine-tuned model, we conducted machine learning molecular dynamics (MLMD) simulations, which can perform long time and large scale simulation. The results reveal that water molecules penetrate deeply into the FAMA lattice, causing the dissolution of FA/MA cations and the leakage of I anions, leading to degradation. In contrast, CsFAMA shows little water molecule penetration, demonstrating that Cs-doping enhances the stability of Sn-based perovskites, mitigates degradation, and improves the performance of perovskite solar cells.



References

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