Liquid-Assisted Mechanochemical Formation of $CsCuCl_3$: Insights from Machine Learning Potentials and DFT

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$CsCuCl_3$ perovskite, a thermally stable and redox-active halide, is a promising candidate for energy conversion and storage applications [1] due to its ionically conductive channels, Cu^{2+}/Cu^{+} redox activity, and tunable semiconducting properties. $CsCuCl_3$ perovskite is efficiently synthesized from $CuCl_2$ and CsCl via liquid assisted grinding (LAG), using dimethyl sulfoxide (DMSO) as a minimal additive, highlighting a sustainable and scalable route to halide perovskites.

In this work we utilize atomistic simulations combining DFT and universal machine learning potential to reveal the role of solvent-mediated Cu^{2+} coordination in perovskite formation. Surface-driven reaction pathways were modeled using a pretrained OC20-compatible universal machine learning interatomic potential (UMA) within the ASE framework [3], enabling efficient sampling of adsorption at reactive interfaces. Interaction of DMSO with 10-1 surface of CuCl2 is investigated on using 3x3 slab model (Figure 1)

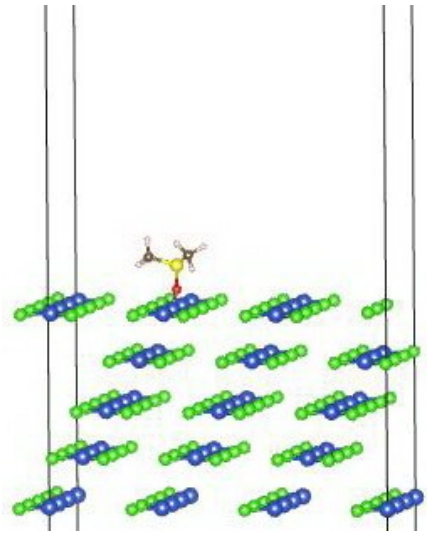


Figure 1. Interaction of DMSO with 10-1 surface of CuCl2.

Complementary density functional theory calculations at B3LYP/ 6-311++G(2d,2p) level of theory using Gaussian 09 program [4] package provide electronic structure insight into Cu–DMSO coordination (Figure 2) and charge redistribution, confirming ligand-induced weakening of Cu–Cl interactions. Mechanochemical experiments validate accelerated formation of $CsCuCl_3$ in the presence of DMSO, supporting a solvent-assisted ion rearrangement mechanism. This work demonstrates how surface-focused machine learning potentials, combined with electronic structure calculations and experiment, can provide mechanistic insight into complex solid-state reactions under mechanochemical conditions.

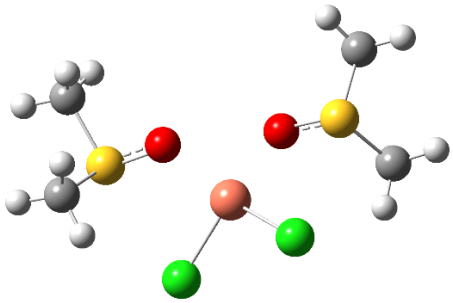


Figure 1. Structure of $CuCl_2$ coordinated with two DMSO molecules.

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