

Solvent Acidolysis Crystallization of Halide Perovskite Single Crystals

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We introduce a novel solvent acidolysis crystallization (SAC) technique for the synthesis of methylammonium (MA) lead halide perovskites, where the hydrolysis of *N*-methylformamide (NMF) in acidic conditions forms the MA cation in situ.[1] Bulk crystals were grown at room temperature without the need for antisolvents, simplifying the process and reducing the use of toxic and expensive chemicals. The SAC technique also facilitated the creation of MAPbBr₃ single crystals with intense edge emission under UV light.[2] Micro-X-ray diffraction revealed that lattice compression at the edges correlated with longer-lived photogenerated carriers and enhanced detectivities in planar photodetectors. Furthermore, we synthesized mixed dimethylammonium/methylammonium lead tribromide (DMA/MAPbBr₃) crystals, achieving 44% DMA incorporation while maintaining the 3D cubic phase.[3] These mixed crystals exhibited suppressed orthorhombic phase and lower phase-transition temperatures, with higher detectivity at room temperature and significant enhancement below 200 K.

Our research also addressed the formation of metallic lead in perovskite crystals, finding that precursor solution composition and concentration played a crucial role.[4] Black-colored mixed DMA/MAPbBr₃ crystals, despite their unexpected coloring, showed higher crystallinity and fewer defects, underscoring the need for further investigation into precursor solution chemistry.

Additionally, we developed a surface engineering crystallization technique to grow a polycrystalline MAPbBr₃ film on bulk single crystals, resulting in intense green emission under UV light.[5] These surface-engineered crystals exhibited fewer surface defects and significantly improved X-ray detector sensitivity. The polycrystalline nature and surface engineering of MAPbBr₃ single crystals proved essential for superior optoelectronics, achieving high detectivities and photoresponsivities.[6] This highlights the potential of the SAC technique for advancing the synthesis and application of halide perovskites in optoelectronic devices.

References

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Figures

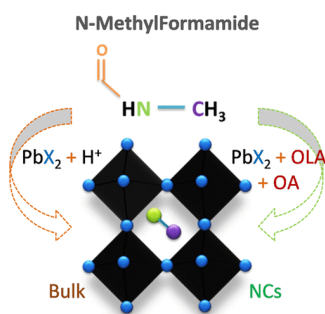


Figure 1: *N*-Methylformamide as a Source of Methylammonium Ions in the Synthesis of Lead Halide Perovskite Nanocrystals and Bulk Crystals, <https://pubs.acs.org/doi/10.1021/acsenergylett.6b00521>