Interfacial Engineering of FAPbI3 Perovskite Solar Cells via Tetramethylammonium Iodide (TMAI) Surface Passivation for Enhanced Phase Stability and Efficiency

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The commercialization of **Formamidinium lead iodide (FAPbI3)** perovskite solar cells (PSCs) is primarily hindered by the spontaneous phase transition from the photoactive alpha -phase to the non-perovskite delta -phase under ambient conditions [1]. This study investigates an effective interfacial modification strategy using Tetramethylammonium Iodide (TMAI) as a surface passivation agent to stabilize the alpha - FAPbI3 lattice and **suppress interfacial charge recombination.**

In a typical fabrication, the FAPbI3 perovskite films were deposited via a one-step spin-coating process. The TMAI surface treatment was performed by dynamic spin-coating of a TMAI solution onto the perovskite surface, followed by a brief thermal annealing process to ensure proper interfacial interaction. The introduction of TMAI onto the perovskite surface significantly modifies the surface energy and chemical environment of the film. X-ray diffraction (XRD) analysis confirms that the TMAI treatment effectively inhibits the formation of the delta -phase. Specifically, the iodide (I-) anions provided by TMAI **passivate the iodine vacancies** , while the bulky tetramethylammonium (TMA+) cations reside on the surface, providing a robust **hydrophobic barrier and stabilizing the perovskite lattice** against moisture-induced degradation.

Contact angle measurements reveal an increased surface hydrophobicity following TMAI treatment, suggesting a robust barrier against moisture ingress. The energy level alignment was further elucidated by Ultraviolet Photoelectron Spectroscopy (UPS), which revealed a more favorable band alignment at the perovskite/HTL interface, facilitating efficient hole extraction. Furthermore, steady-state photoluminescence (PL) and J-V characterization indicate a significant reduction in voltage loss and improved carrier extraction efficiency. Optimized devices achieved a power conversion efficiency (PCE) exceeding **21.17%** with remarkably improved long-term stability. This work provides deep insights into the molecular-level interfacial interactions between quaternary ammonium halides and perovskite surfaces, offering a scalable route toward stable and high-performance optoelectronic devices.

Keywords:

Perovskite solar cells, FAPbI3, Interfacial modification, TMAI, Phase stability, Surface passivation

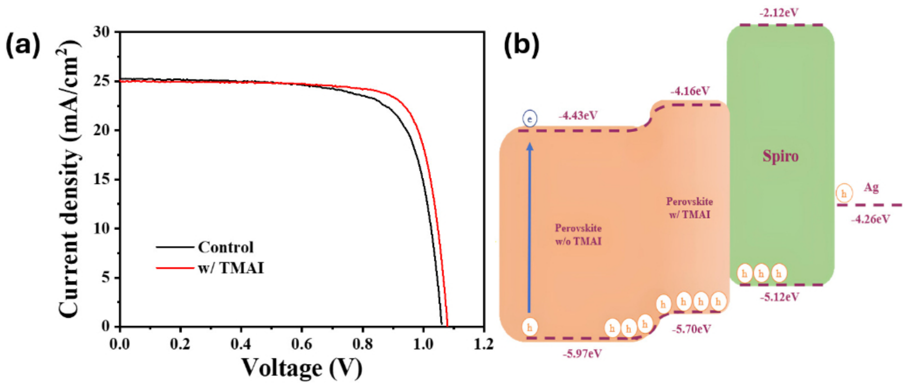

Fig 1. (a) J-V curves of FAPbI3 devices with/without TMAI, achieving 21.17% PCE. (b) Energy level diagram derived from UPS, demonstrating improved band alignment for enhanced interfacial charge extraction.

References:

[1] N. J. Jeon. J. H. Noh. W. S. Yang. Nature. 2015.

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