

Rapid Perovskite Formation by CH_3NH_2 Gas-Induced Intercalation and Reaction of PbI_2

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Solution processable perovskite solar cells traditionally use $\text{CH}_3\text{NH}_3\text{I}$ solid powder as one of the two precursors that requires solvation into a solution and a spin-coating step; resulted films need post-annealing ($\sim 1\text{h}$) for complete conversion to $\text{CH}_3\text{NH}_3\text{PbI}_3$. Here we describe a method to form perovskite in ambient air by exposing PbI_2 films to the simpler CH_3NH_2 gas precursor (as opposed to $\text{CH}_3\text{NH}_3\text{I}$ solid powders)¹. The reaction completes within a few seconds forming complete-coverage perovskite films with a roughness of 2 nm. The non-stoichiometric reaction produces Pb oxides as by-product, which are reconverted by further HI gas exposures. With combined measurements of thin film crystal structure, chemical state, and absorption properties, we elucidate the chemical reaction mechanisms underlying these gas-induced processes. Fabricated solar cell devices show efficiency of 15.3%, which stays almost the same after 133 days. Such a gas-induced reaction also enabled the successful preparation of high quality perovskite films with a size of 100 cm^2 . We also demonstrate that this method is versatile and enables the formation of a wide range of perovskite materials, e.g. $\text{CH}_3\text{NH}_3\text{PbCl}_3$, $\text{CH}_3\text{NH}_3\text{PbBr}_3$, $\text{CH}_3\text{NH}_3\text{PbI}_3$, $\text{CH}_3\text{NH}_3\text{SnBr}_3$, $\text{CH}_3\text{NH}_3\text{PbBr}_x\text{I}_{3-x}$, $\text{CH}_3\text{NH}_3\text{PbBr}_x\text{Cl}_{3-x}$.

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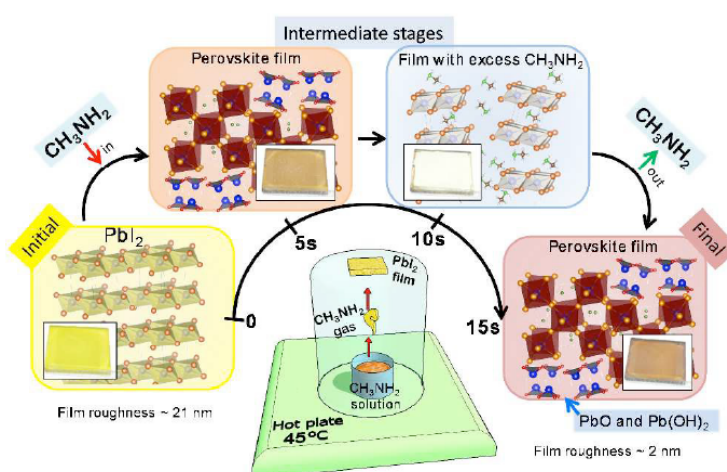


Figure 1. CH_3NH_2 gas induced perovskite formation. Illustration of the setup used to form perovskite from simple precursors of CH_3NH_2 gas and PbI_2 film is depicted in the lower middle part of the figure: a PbI_2 film is exposed to CH_3NH_2 gas from a heated solution. The different stages of the reaction are schematized from left to right with an arrow corresponding to the time scale of the process.