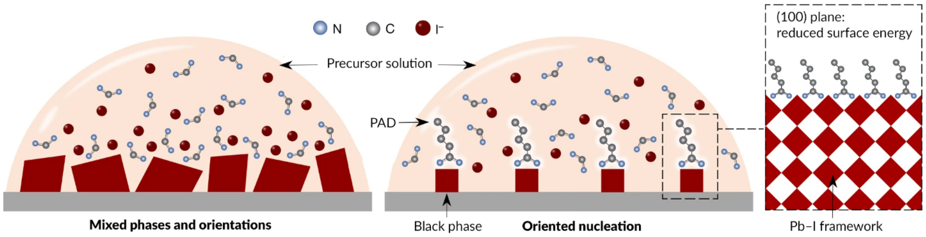
ADVANCED LIGHT SOURCE

ENERGY SCIENCES

Coaxing Molecules to Stand Tall for Better Solar Cells



The introduction of an additive, pentanamidine hydrochloride (PAD), to the precursor solution of a perovskite material, formamidinium lead iodide ($FAPbI_3$), induced oriented nucleation and the preferential formation of the perovskite's photoactive black phase.

Scientific Achievement

Multimodal probes at the Advanced Light Source (ALS) and the Molecular Foundry revealed a way to prevent the formation of undesirable phases in a perovskite-type compound that shows promise for the efficient harvesting of light for solar cells.

Significance and Impact

The work led to new fabrication protocols that resulted in devices with improved power-conversion efficiencies and operational stability.

A new class of PV materials

The harnessing of solar power is a critical pillar in the next generation of electrical infrastructure, and it will require large-scale manufacture and deployment of photovoltaic (PV) panels. A relatively new class of materials, organic–inorganic halide perovskites, offers an alternative to silicon for PV power generation.

Halide perovskites are prime candidates for research because they offer a range of benefits, including being up to 1000 times thinner than silicon, absorbing incoming solar irradiation more efficiently, and having a better matched bandgap for the solar spectrum. Perovskite thin-film fabrication also offers lower-cost processes, but more research, such as this study, needs to be done to capitalize on that potential.

Polytypes–black and yellow phases

Formamidinium lead iodide ($FAPbI_3$) is one of the most promising halide perovskite candidates for PV applications. One challenge, however, is the existence of polytypes of $FAPbI_3$ (different configurations of the same atoms), called black and yellow phases. At room temperature, the yellow phase is the most stable, but it is relatively transparent to the solar spectrum. In contrast, the black phase absorbs most of the solar spectrum, but it is energetically unfavorable and therefore not normally stable at room temperature. As a consequence, the photoactive black phase is hampered by conversion to the non-photoactive yellow phase. To counteract this, researchers have developed a new method of perovskite crystallization that reliably suppresses yellow-phase formation by encouraging the oriented nucleation of $FAPbI_3$ crystals.

Multimodal crossover capabilities

The crystallization of $FAPbI_3$ is a rapid process, making it difficult to monitor in real time. To overcome this challenge, the researchers made use of significant crossover between the expertise and capabilities available at Berkeley Lab's Molecular Foundry and the ALS.

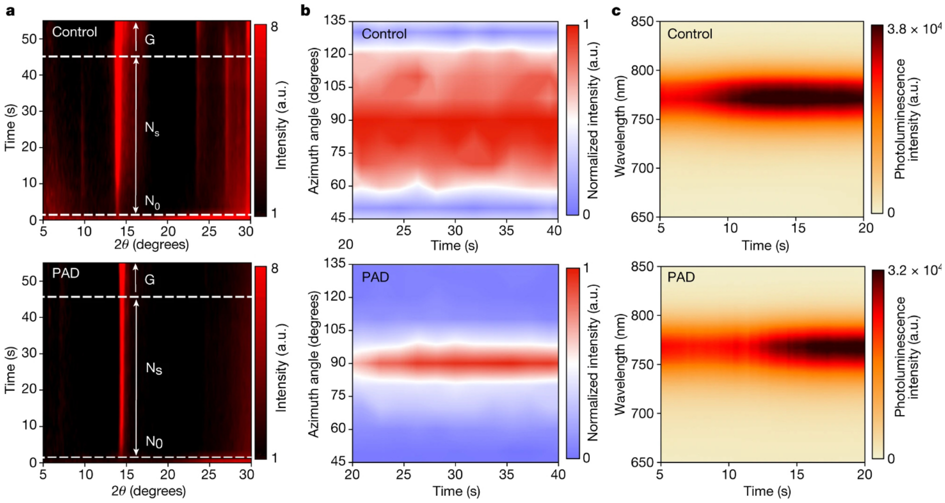
At ALS Beamline 12.3.2, x-ray microdiffraction revealed key features of the crystal structure in real time, as it developed from precursor solutions. This required building a spin coater that worked within the beamline's hutch. While the film was fabricated, diffraction and photoluminescence data could be collected simultaneously. Sample preparation and complementary measurements were performed at the Foundry. With these capabilities, the researchers were able to monitor crystal phase changes and study the influence of additives on the crystallization pathway.

PAD additive success

The results showed that when pentanamidine hydrochloride (PAD) was added to the starting materials, the yellow phase and other intermediate phases were suppressed. According to the researchers' analysis, PAD leads to a decrease in surface energy for the (100) plane, driving the preferential formation of (100)-oriented black-phase $FAPbI_3$.

PAD also allowed for more synthesis control by slowing down the crystallization process, and the well-oriented growth produced crystals of higher quality. Devices made using the PAD-altered process demonstrated consistently higher power-conversion efficiency and considerably improved operational stability. The additive strategy was applied to different fabrication processes and was found to be universally applicable as a strategy to achieve stable black-phase perovskite.

In the future, the researchers hope to expand these advanced characterization capabilities to industry-relevant and scalable processes, as well as broadening the scope to other material classes beyond perovskites, such as organic semiconductors and other functional materials.



(a) In situ grazing-incidence x-ray mirodiffraction measurements of perovskite films fabricated without PAD (top) and with PAD (bottom). N_0 denotes the start of the nucleation, N_s denotes the nucleation stage, and G denotes the growth stage. During the nucleation stage, the control film showed diffraction peaks corresponding to multiple intermediate phases, whereas the film using PAD featured a clean (100) diffraction pattern of black-phase perovskite. (b) Evolution of the azimuth angle. The control azimuth angle showed a broad distribution, indicating a fairly random crystallographic orientation. With PAD, a sharp peak centred at 90° verified a nucleation stage with a preferred out-of-plane orientation along the (100) facet. (c) Evolution of photoluminescence spectra. In situ photoluminescence measurements provided evidence of the sluggish nucleation kinetics with PAD.

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