DEVELOPING THE NEXT GENERATION OF PEROVSKITE SOLAR CELLS

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Organic-inorganic halide perovskites are at the brink of commercialization as the next generation of light-absorbing materials for solar energy harvesting devices. Perovskites have large absorption coefficients, long charge-carrier lifetimes and diffusion lengths, and a tunable absorption spectrum. Furthermore, these materials can be low-temperature solution-processed, which transfers to low-cost manufacturing and cost-competitive products. The remarkable material properties of perovskites enable a broad product-market fit, encompassing traditional and new applications for solar technology. Perovskites can be deposited on flexible substrates for flexible solar cells, applied in thermochromic windows for power generation and building cooling, or tuned for tandem solar cell application to include in high performance solar panels. However, perovskites are intrinsically unstable, which has so far prevented their commercialization. Despite large research efforts, including over two thousand publications per year, perovskite solar cells degrade in under one year of operation. In a saturated research field, new ideas are needed to inspire alternative approaches to solve the perovskite stability problem. In this dissertation, we detail research efforts surrounding the concept of a self-healing perovskite solar cell.

A self-healing perovskite solar cell can be classified with two distinctions: mechanically healing and molecularly healing. First, mechanically self-healing involves the materials ability to recover its intrinsic properties after mechanical damage such as tares, lacerations, or cracking. This type of healing was unique to the organic polymer community and ultra-rare in semiconducting materials. By combining a self-healing polymer with perovskite material, we developed a self-healing semiconducting perovskite composite material which can heal using synergistic grain growth and solid-state diffusion processes at slightly elevated temperatures. The material is demonstrated in flexible solar cells with improved bending durability and a power conversion efficiency reaching 10%. The addition of fluidic polymer enables macroscopic perovskite material movement, which is otherwise brittle and rigid. The results inspire the use of polymer scaffolds for mechanically self-healing solar cells.

The second type of healing, molecular healing, involves healing defects within the rigid crystal domains which results from ion migration. The same phenomenon which leads to the device degradation, also assists the recovery of the device performance after resting the device in the dark. During device operation, perovskite ions diffuse in the perovskite lattice and accumulate at the device interfaces where they undergo chemical reactions or leave the perovskite layer, ultimately consuming the perovskite precursors. The photovoltaic performance can be recovered if irreversible degradation is limited. Ideally, degradation and recovery can match day and night cycling to dramatically extend the lifetime of perovskite solar cells. In this dissertation, we introduce the application of chalcogenide chemistry in the fabrication of perovskite solar cells to control the thin film crystallization process, ultimately to reduce defects in the perovskite bulk and introduce surface functionality which extends the device stability. This new strategy will help enable the realization of a molecularly self-healing perovskite solar cell by reducing irreversible degradation. Lastly, we present a few other new ideas to inspire future research in perovskite solar cells and assist in the commercialization of the next generation of photovoltaics.