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青山学院大学審査学位論文の要約

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Elucidation of Pinhole-Induced Degradation
Mechanisms and Strategic Defect Management for
Enhanced Reliability in Perovskite Solar Cells

ペロブスカイト太陽電池の高信頼化に向けたピンホール起因劣化メカニズムの解明と戦略的欠陥管理に関する研究

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Abstract

This research investigates internal morphological defects known as pinholes to improve the long-term durability of perovskite solar cells (PSCs). PSCs are a highly promising photovoltaic technology because they offer a path to high power conversion efficiency (PCE) at a low manufacturing cost compared to conventional silicon solar cells. Their unique physical properties, such as being lightweight and flexible, allow for a wide range of applications where traditional rigid panels are difficult to install, such as curved surfaces or building facades. Despite these advantages, the most significant challenge preventing widespread commercial use is their limited long-term durability. While much research has focused on protecting the perovskite material from external environmental factors like moisture and oxygen, this study addresses the critical problem of internal defects created during the fabrication process.

The efficiency of PSCs has increased at an unprecedented rate, approaching the performance of established crystalline silicon technologies. Because PSCs have the potential to rival or even surpass silicon in terms of efficiency, the technology to strategically manage hidden pinholes to prevent failure and maintain performance over the long term is the final key to successful commercialization. The central focus of this research is the elucidation of a specific degradation mechanism induced by pinholes within the perovskite (PVK) absorber layer. A pinhole is a microscopic void or thin spot that can occur during solution-based processing. The conventional understanding has been that deep pinholes are a problem for initial manufacturing yield because they can create direct electrical short-circuits, or shunts, causing immediate device failure. However, this thesis addresses a more complex and hidden failure mode involving "latent defects". The study hypothesizes that many pinholes are initially benign and do not affect the device's initial performance, but they can later transform into catastrophic, performance-limiting shunts when the device is subjected to operational stress, such as illumination and the resulting flow of photocurrent.

To prove this hypothesis, a systematic investigation was conducted using a controlled model system. PSCs with a standard n-i-p device structure were

fabricated, and pinholes of a defined size and location were intentionally created using a precision laser. This approach enabled a direct comparison between pinhole-containing devices and pinhole-free reference cells. Immediately after fabrication, the devices with intentionally formed pinholes showed high PCE that was nearly identical to the reference cells, confirming that the pinholes were initially harmless and the device was in a "benign state". However, after a stress protocol consisting of dark storage and light soaking, the pinhole-containing devices suffered a catastrophic drop in performance, while the reference cells remained stable.

The physical and electrical changes at the pinhole sites were investigated using several analytical techniques. Lock-in thermography (LIT) was used to non-destructively identify the exact locations of current leakage by detecting the thermal signatures of local shunts. The LIT analysis confirmed that the failure was caused by the formation of intense shunts located exactly at the pinhole sites. Microstructural analysis using scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDS) revealed severe physical damage at these transformed sites. The results showed the delamination and removal of the PVK and gold layers at the shunt locations. Furthermore, 3D depth profiling using laser microscopy confirmed that pinholes that transformed into shunts had physically breached through the entire device stack to the bottom electrode.

To understand the physical trigger for this damage, a two-dimensional device simulation framework was established to model the internal electrical conditions. The simulation results revealed that the unique geometry of a pinhole inherently creates a point of extreme electrical stress. This effect, known as current density concentration, results in peak current densities at the bottom corners of the pinhole that can be more than 30 times greater than in defect-free regions. This localized current generates intense Joule heat, providing the thermal energy to physically destroy the material layers and create a conductive shunt path. The magnitude of this concentration is highly sensitive to the thickness of the PVK layer remaining at the bottom of the pinhole.

Based on the understanding of this mechanism, a practical technological solution was developed. An advanced laser-based repair method, termed "direct pinhole ablation," was created to precisely remove all device layers within the defect area down to the glass substrate. This method was shown

to be highly effective, restoring the performance of severely degraded devices to a level comparable to undamaged reference cells. A key advantage of this technique is that it minimizes the loss of the power-generating active area by targeting only the specific pinhole site. Analysis confirmed that the method is robust and consistently removes the defect regardless of its initial condition.

To translate this repair technology into an industrial tool, the research established a comprehensive and proactive defect management strategy. The strategy is built upon a quantitative trade-off analysis that weighs the benefit of eliminating a potential shunt path against the drawback of the permanent loss of a small fraction of the active area. By using the device simulation model, a decision-making framework was developed to inform whether a repair is beneficial based on the measured physical characteristics of individual pinholes.

The practical implications of this management strategy are significant. First, it can improve manufacturing yield by providing a pathway to recover devices that would otherwise be discarded as failures. Second, it enhances long-term reliability by enabling the proactive repair of latent defects before they can cause failure in the field. Manufacturers can adjust the repair criteria based on strategic priorities, balancing the goal of maintaining high initial efficiency against the proactive avoidance of catastrophic degradation. In conclusion, this work identifies a critical degradation mechanism in PSCs and offers a systematic, data-driven pathway to produce more durable and reliable devices, which is essential for the successful commercialization of this technology as a next-generation renewable energy source.