Pre-print submitted version

Advances in Perovskite Optoelectronics: Bridging the Gap Between Laboratory and Fabrication

Subodh G. Mhaisalkar, Nripan Mathews, Henk J. Bolink, and Damodaran Bahulayan*

S. G. Mhaisalkar, N. Mathews, D. Bahulayan
Energy Research Institute @ NTU (ERI@N)
Nanyang Technological University
Research Techno Plaza, X-Frontier Block, Level 5, 50 Nanyang Drive
Singapore 637553, Singapore
E-mail: subodh@ntu.edu.sg

S. G. Mhaisalkar, N. Mathews
School of Materials Science and Engineering
Nanyang Technological University
50 Nanyang Avenue, Singapore 639798, Singapore

H. J. Bolink
Instituto de Ciencia Molecular (ICMol)
Universidad de Valencia
C/Catedrático J. Beltrán 2, 46980 Paterna, Spain

In 2019, hybrid halide perovskites celebrated their 10th anniversary as a “wonder material” for optoelectronic applications. Although the parent perovskite structures were elucidated in the late 19th century, the seminal work by Miyasaka et al. exploiting organic-inorganic hybrid halide perovskites sensitizers for visible-light conversion in solar cells marked the revisit of these materials and has proven to be a game-changer in this field. Extensive investigations were undertaken to develop new materials (all inorganic and organic-inorganic hybrids, in the form of films or alternate morphologies) and deposition techniques, explore interfaces and in-depth characterization, while engineering devices and testing methods for optimum results. Within a short time span, the power conversion efficiency (PCE) of single-junction and tandem perovskite solar cells (PSCs) have exceeded 25% and 29% respectively; thus challenging the dominance

of silicon solar cells. Building-integrated photovoltaics (BIPV) is another hot topic in PSCs, where perovskite solar cells are designed to be semi-transparent for deployment in residential or office building facades. Along with the success in photovoltaics, halide perovskites have also made their impact in light emission, lasing, imaging, spintronics, memristors, and photocatalysis. However, key challenges still lie ahead, particularly on the commercialization of perovskite devices. Poor material and device stability under operational conditions and the lack of reproducibility and scalability have remained problematic; whereas the search for suitable lead-free perovskites continues.

The ICMAT 2019 symposium on “Advances in Perovskite Materials: Photovoltaics, Optoelectronics and Frontier Phenomena” was organized in collaboration with “13 Aseanian Conference on Nano-Hybrid Solar Cells” gathered 230 delegates from 23 countries. The highlights of the five-day event include 220 oral and poster presentations with commemoration and awards to pioneers and young researchers. The symposium dedicated efforts not only to the discussion of progress but also the challenges ahead for perovskite optoelectronics. These twin special issues in Advanced Energy Materials and Energy Technology document some of the recent efforts for bridging the gap between the laboratory and the fabrication of perovskite optoelectronic devices.

The special issue in Advanced Energy Materials presents a collection of state-of-the-art reviews and progress reports on new perovskite materials (including Pb-free alternatives), additives, interfaces, compositional tuning, device physics, and emerging new applications.

Stability of perovskites is a critical topic of discussion in this special issue; with Grätzel, Zhu and You et al. covering topics ranging from fabrication protocols, defect passivation methods and mechanisms, and low-dimensional perovskites. Park discusses engineering

at both HTL and ETL interfaces, grain boundary engineering, additive engineering, and solution chemistry-based large-area coating methodologies (e.g. D-bar coating, bi-facial stamping, etc.) as effective routes to obtain high-efficiency cells and modules. The instability of perovskite materials and devices are also caused by inherent thermodynamic limitations, solvent sensitivity, and imperfect synthetic protocols. In this respect, Bolink et al. present mechanochemical synthesis as a green alternative to produce high purity halide perovskites as dry powders, which can be colloidally dispersed and integrated into optoelectronic devices whereas Baran et al. reviewed the emerging large grain PSCs and single crystal PSCs, highlighting crystal growth and device architectures. Choy, Shapter, and Wilson et al. reviewed the recent progress in the synthesis, composition tuning and application of various charge injection/transport layers and dopant free organic hole transporting materials whereas Qi et al.'s review focused on the importance of considering surface/interface science while designing PSCs. Hysteresis and nonradiative recombination phenomena are also in the line-up with Han et al. focused on the use of additives for hysteresis free PSCs while Stranks and co-workers outlined the studies seeking to minimize nonradiative recombination through light illumination processes.

State of the art reviews on Lead-free perovskites are another attraction in this collection with Miyasaka, Wang, Abate, and Xiao et al. discuss strategies to develop robust organic-free, Pbfree, and dopant-free perovskite solar cells, using bismuth, tin, and other narrow bandgap materials. Theoretical studies on materials and device physics are highlighted by De Angelis, Van de Walle and Banerji et al. covering topics including polaron physics, carrier recombination mechanisms, free carriers, excitons, and sub-bandgap states. The state of the art tandem solar cells and other emerging new applications were covered by Catchpole et al. whereas Bakr et al. compiled newer areas such as perovskite-based photocatalysts, photoelectrodes, and solarto-chemical fuel conversion devices.

The special issue of Energy Technology focuses on long term stability and scaleup with critical reviews covering various deposition methods such as sequential deposition (Ho-Baillie et al.), inkjet technology (Unger et al), surface passivation and composition engineering (Wang and Liao et al.) and the developments in cesium based all inorganic perovskites (Qi et al). The reports from the groups of Vaynzof, Marí, and Hongxia Wang are also covering related areas such as performance and stability with respect to precursor stoichiometry, cation replacement, and dimensionality. The use of spin-coated perovskite singlecrystals (Loi et al.), fluorinated additives to control the crystallinity (Ahmad et al.), tuning of coordinated solvent number (CSN) to obtain uniform pinhole-free films and multiple-source co-evaporation method to incorporate perovskite precursors with varying solubility (Bolink et al.) are also in the line-up. The contributions from the group of Palazon, Bolink, Yao, and El-Mellouhi present some interesting developments in lead-free perovskites whereas the report from the group of Mhaisalkar and Mathews discuss tandem solar cells highlighting the importance of optimal measurement schemes to assess 4 terminal tandem PCE.

The readers can also find some interesting articles on interface materials. For example, the groups of Sarkar, Boix, Makarov, and Xiao reported various interface materials such as amorphous Nb2O5, vacuum-deposited molybdenum oxide (MoOX), p-doped silicon nanoparticles, and thermally deposited dielectric/metal/dielectric (DMD) stack of MoO3/Ag/WO3 as charge injection and transport layers in PSCs. Reports on perovskite nanocrystals (PNCs) are also available as the contributions from the groups of Lee, Yella and Kabra present some interesting new methods for the synthesis of PNCs with better film homogeneity, particle stability, and bandgap tunability.

As highlighted by the various authors of these special issues, halide perovskite materials and devices demonstrate exceptional potential for cost-effective optoelectronics.

Although the PCE of PSCs has surpassed many of its competitors, there remain many challenges to solve before the successful commercialization of these materials and devices. Furthermore, investigations should explore the use of halide perovskites for different applications such as imaging, computing, and communications. We hope that together, these twin special issues will inform the readers of the state-of-the-art and Highlight the pressing challenges ahead for this exciting area of research.

Finally, the organizers of the symposium wish to thank all the contributors to these special issues and the Editors and Editorial staff members at Advanced Energy Materials and Energy Technology for their dedicated efforts to release these special issues on time.