Perovskites for printed flexible electronics

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Abstract. In recent years, halide perovskites have attracted tremendous attention as active materials in various electronic devices including solar cells, light-emitting diodes, photodetectors, etc. Their excellent optical and electrical properties as well as solution processability make perovskite material an ideal candidate for flexible electronic applications that are manufactured by printing in industrial scale. In this article, the material characteristic and synthetic procedure of perovskite crystals are introduced. Deposition techniques and potential challenges of commercializing perovskite-based flexible electronics are briefly discussed.

1. Introduction

Ten years ago, the first organic-inorganic lead halide perovskite based solar cell was reported by T. Miyasaka and coworkers, where an unimpressive power conversion efficiency (PSE) of 3.8% was obtained. [1] The original concept arose from seeking for new promising materials as visible light sensitizers, which are used in dye-sensitized solar cells (DSSC, or Grätzel cell). Although the cell efficiency was not spectacular, the synthesized perovskite materials exhibited good sensitizing ability, well-matched energy levels and superior crystalline structure. Since then, considerable attention of researchers worldwide has been attracted, which leads to, doubtless, a tremendously increased number of publications on this topic (Figure 1).

![Figure 1. Publications on perovskite solar cell from January 2009 to August 2019 (Source: Web of Science, Keywords: Perovskite Solar Cell).](image-url)
The name of perovskite originated from a Russian mineralogist L. A. Perovski, who initially discovered calcium titanium oxide (CaTiO₃) mineral. Hence, the term perovskite usually refers to a generic structure of any compounds that complies with the formula ABX₃. For lead halide perovskite, one common example of the monovalent cation “A” is methylammonium (MA), which is larger than the divalent cation "B" Pb²⁺. “X” anions could be I⁻, Br⁻, Cl⁻ or a mixture of different halides. Crystal structure of perovskite compounds is shown below in Figure 2. [2]

![Crystal structure of metal halide perovskite materials](image)

Figure 2. Crystal structure of metal halide perovskite materials [2].

By employing perovskites with such perovskite two-dimensional crystal structure as active materials in electronic applications, promising device performance can be expected owing to the unique electrical and optical properties of perovskites, including long exciton diffusion length, high charge carriers mobilities, tunable optical bandgap and absorption spectra, high light extinction coefficient, etc. [3-6] In addition to the inherent materials properties, the ease of solution processing enables potentially cost-effective manufacturing of perovskite based electronic devices, which plays a dominant role for industrial scale production and application.

Based on the exceptional characteristics of perovskites, various electronic applications have been explored including optoelectronic devices such as photovoltaics [7-18], light emitting diodes (LED) [19-27], photodetectors [28-33], optical lasers [34-38] as well as field-effect transistors (FETs) [4, 39-43] and resistive-switching memories [44-46].

In this article, synthetic approaches of perovskite materials are described in terms of coating procedure and processing control. Different electronic applications based on perovskites are briefly discussed, mainly focusing on the feasibility of being printable and flexible during industrial scale manufacturing. Further, from technology point of view, the remaining challenges and concerns that impede eruptive application of perovskite-based electronics are pointed.

2. Perovskite layer processing
Unlike silicon based solar cells that typically require lengthy crystal growth, complicated processes with multiple steps and expensive production system for extreme temperature, pressure and materials purity regulation, the fabrication of perovskite thin film absorber is much more facile. Although perovskite films can be manufactured via vacuum-based process as well such as vapor-assisted deposition and thermal co-evaporation, the solution-based wet chemistry processing of thin films has become the most popular and dominant methods which allow for high quality crystals, complete film coverage without pinholes, and consequently superior device performance. [47-48] Generally, solution deposition techniques can be categorized into one-step and two-step methods, as graphically demonstrated in Figure 3.
Figure 3. One-step and two-step coating methods of CH$_3$NH$_3$PbI$_3$ perovskite thin film fabrication [47].

One-step spin coating process is currently the most convenient method for perovskite layer fabrication in laboratory scale. Taking the widely applied CH$_3$NH$_3$PbI$_3$ perovskites as example, one-step coating procedure involves the preliminary dissolution of methylammonium CH$_3$NH$_3$I and inorganic lead halide PbI$_2$ in organic solvents (e.g. DMSO, DMF, DMA or solvent mixture), forming one precursor solution. Subsequently, the prepared solution with mixed reactants in a predefined stoichiometry is spin coated onto substrates. Finally, pure, high quality and homogeneous perovskite films can be achieved after thermal annealing at a range of 100-150 $^\circ$C. [18]

As depicted in Figure 3, two-step coating of perovskite layers separates the incorporation of PbI$_2$ and CH$_3$NH$_3$I in a sequential process [48]. Firstly, PbI$_2$ is dissolved and spin coated onto desired substrates (normally mesoporous metal oxide films). Secondly, CH$_3$NH$_3$I dissolved in a different organic solvent, known as antisolvent, is prepared and exposed to the freshly formed PbI$_2$ precursor. It was found that the conversion to CH$_3$NH$_3$PbI$_3$ hybrid perovskites starts as soon as the two components come into contact. Although one more step is involved into the film fabrication process, two-step coating method demonstrates additional benefits, including more intimate control of film morphology and better process tunability. It was reported that uncontrolled precipitation of perovskites may occur if processed by one-step method, which will, doubtless, lead to inferior device performance. Moreover, the efficiency difference between devices fabricated via both methods was attributed to significant drop of photovoltage and fill factor in one-step deposition, which allows for a further speculation that the film morphology of deposited perovskites is strongly correlated. [47-48]

Both one-step and two-step coating methods may exhibit the potential for cost-efficient and large-scale manufacturing, which means successful process transfer from laboratory scale spin coating to industrial scale production is expected. However, limitation of such pure solution-processing deposition still remains for scaling up. For instance, the production of CH$_3$NH$_3$I requires huge amount of solvent for synthesis and purification. Another fabrication method using methylamine vapor to initiate the recrystallization of perovskites seems to be more promising for industrial scale production. The advantages of this method include low temperature treatment (room temperature), healing effect of methylamine gas on perovskite film, good uniformity of perovskite films, etc. However, the large-scale inhomogeneity is still present for gas treatment deposition method. [49-51]

Various printing techniques have been investigated by researchers from both academia and industry. In Figure 4, three different deposition methods are illustrated including inkjet coating, slot-die coating, roll-to-roll coating and spray coating. [2]
3. Perovskite-based flexible electronics

In addition to the technical development of coating techniques using either solution or gas treatment, how to achieve complete printability on flexible devices remains to be a critical aspect as well. Owing to the solution processability of two-dimensional perovskites, high quality thin films can be potentially coated on flexible substrates, leading to bendable, foldable and wearable electronic devices and thereby apparently widening applications in the future. It has been reported by researchers that perovskite-based flexible solar cells, LED as well as photodetectors can be easily prepared by depositing perovskite layers on indium tin oxide (ITO) / polyethylene terephthalate (PET) substrates [7,11-12,15,21,27,55-58]. Deposition techniques vary a lot for fabrication of flexible devices, it can be imaged that solvent-free melt processing and methylamine vapor exposure methods could be more technically feasible for thin-film coating on flexible substrates. [50,56]

Although promising power conversion efficiency (PCE) and external quantum efficiency for solution-coated flexible solar cells and LED was delivered (device examples are shown in Figure 5), the long-term performance of such electronics remains to be still challenging. However, it was found that the aging of perovskites may not be necessarily related to the deterioration of devices, which means both aged and freshly fabricated films can lead to similarly stable electronic devices. [50] Excellent device stability has already been shown in some applications, such as photovoltaic cells, LEDs and FETs. [2, 20, 23, 42] Furthermore, from the application point of view, it is still questionable how is the necessity of large-scale opto-electronic devices for being flexible. Flexibility of substrates offers additional deposition possibility for the device fabrication like roll-to-roll coating. [53] However, it can be clearly expected that the control of coating process will become more complex in
the industrial line, requiring high performing manufacturing equipment with enough accuracy in terms of heating power, transporting speed, volume precision, etc. Therefore, the replacement of rigid substrates with flexible counterparts is typically accompanied with more challenging process control during device production, high deviation of device properties and consequently lowering of production yield. For example, if roll-to-roll deposition is applied, it would be wise to design a fully integrated equipment with coating functionality of all necessary layers. Thus, extremely high accuracy of equipment geometry and capacity, etc. would be necessary to guarantee a smooth operation, which is also not that easily achievable. Instead, to possibly avoid the complexity, separated small equipment delivering one film by single coating techniques can be expected for the fabrication of rigid substrates based electronic devices. Here, it should be emphasized that a balance between electronic device performance and flexibility exists. To optimally satisfy the requirements of product application and provide possibly superior properties, flexible electronics based on perovskites should be employed in the scenario that rigidity of devices is truly unacceptable.

![Figure 5. Perovskite-based flexible solar cells (left) [15] and flexible perovskite LED (right) [21].](image)

4. Potential issues to be addressed

Nowadays, significant progress has been achieved towards large-scale manufacturing of perovskite photovoltaic modules based on solution processing. Xiexin group announced that perovskite modules with a dimension of 45 cm x 65 cm rigid substrate can be fabricated in their 10 MW pilot line and PCE of 15.3% was achieved for such modules, indicating that commercial and industrial scale of perovskites production is firstly proven. [59] To further expand the success, potential issues in both academia and industry regarding cell performance, lifetime concern, environmental impact, feasibility and applicability of large-scale deposition onto flexible substrates and suitable candidates of buffer layer and electrode materials as well as their deposition techniques will still have to be addressed properly [2,60-64].

![Figure 6. Vision for the future of large-area commercial perovskite solar cells [2].](image)
As depicted by Cannikin law in Figure 6, the shortest boards of the bucket limit its maximum capacity. It is generally believed that the hole transport materials (HTM) and electron transport materials (ETM) are still under rapid development, aiming at substituting traditional materials that are suitable for perovskites’ industrial-scale production. For example, the electron and hole transport materials should be developed with the primary focus on high charge carrier mobility, well-aligned energy levels, processability and photochemical stability. Besides, several R&D topics are relatively mature in terms of development mainly owing to the precious experience from semiconductor industry, such as encapsulation and packaging, electrode materials and appropriate deposition techniques. One can believe that such impeding and promoting factors are not unique for perovskite solar cells. Hence, all these challenges and benefits should be present for the commercialization of other perovskite-based flexible electronic devices as well.

Concerning about ultimate efficiency of single module, one crucial research topic lies on the possibly maintaining the high PCE while scaling-up in module size. Here, defect-free and large-size hybrid organic/inorganic perovskite thin films should be prepared primarily by intimate control of coating procedure and continuous optimization of process parameters. Also, the existence of lead has, doubtless, negative impact on the environment, manufacturing and application. Lead-free perovskite devices are desired obviously, which however still suffers from lowering of efficiencies and stabilities, e.g. Sn, Ge, Cu, Bi, Sb ions-based perovskite solar cells [49,60,65].

5. Concluding remarks

Overall, to best explore the potential of perovskites material-based electronics, solution processability and its realization in industrial manufacturing plays the most important role. Therefore, the processing technologies need to be focused to deliver optimized and facile fabrication in large dimension since no process is born to be fully applicable for mass production purpose. Technology transfer between laboratory and factory would require the collaboration of research scientists and engineers with a wide variety of technical background to address the remaining technical challenges. In the near future, one can expect that printed and flexible perovskite electronics will be appearing in various applications attributing to the excellent product performance, cost-effective price and high global production capacities.

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