ASIA-PACIFIC
SOLAR RESEARCH CONFERENCE

The impact of dynamic two-step solution process on film formation of $Cs_{0.15}(MA_{0.7}FA_{0.3})_{0.85}PbI_3$ perovskite and solar cell performance

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Perovskite solar cells have attracted great attention in recent years and much of the research has been focused on the optimization of solution-based fabrication methods for device performance improvement. Sequential process has been used for many state of the art perovskite solar cells. This paper provides further understanding of the mechanism for perovskite film formation fabricated by sequential solution-based methods by comparing two types of sequential methods for $Cs_{0.15}(MA_{0.7}FA_{0.3})_{0.85}PbI_3$ perovskite which involves an initial step of depositing PbI_2-CsI-DMSO-DMF by spin coating followed by the spin coating of MAI-FAI-IPA. In the "static process", there is a stoppage between the two spin coating steps. In the "dynamic process", the MAI-FAI-IPA precursor in the 2nd step is dispensed while the substrate is still in motion after the 1st PbI_2-CsI-DMSO-DMF deposition step. Both static and dyanmic seqential processes are illustrated in Figure 1.

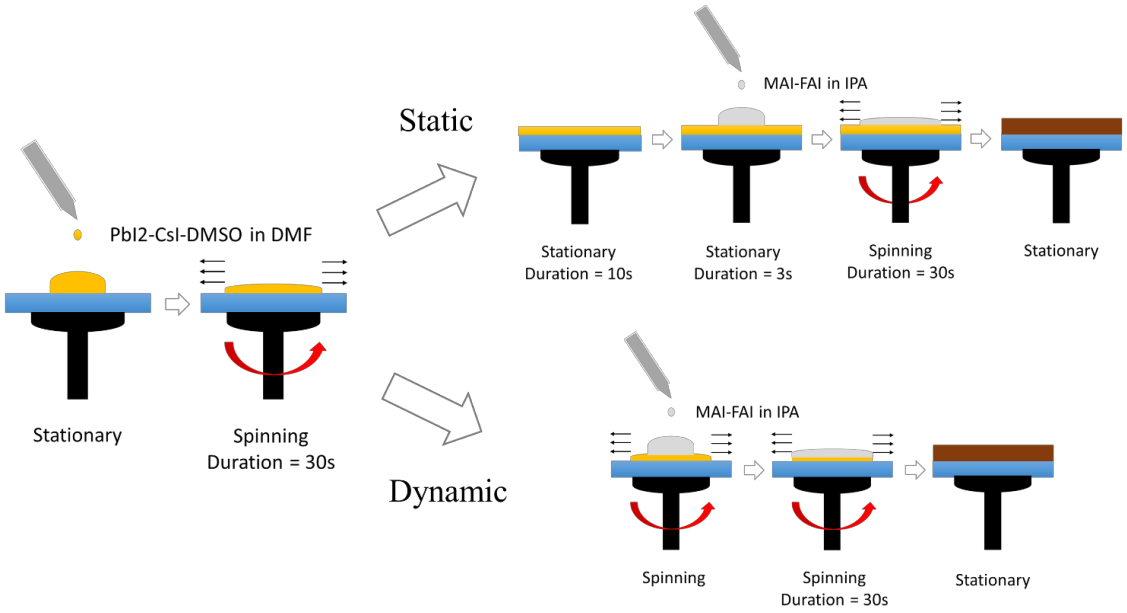

Figure 1. Schematic illustration for the sequential fabrication of CsFAMAPbI3 perovskite film by static and dynamic processes

This is the first time such dynamic process is used for the fabrication of $Cs_{0.15}(MA_{0.7}FA_{0.3})_{0.85}PbI_3$ perovskite film and corresponding solar cells. X-ray diffraction measurements (XRD) and Fourier transform infrared (FTIR) measurements are carried out on as-deposited dynamic processed and static processed films for comparison. The two as-deposited films have different FTIR and XRD peaks, indicating different rate of perovskite formation. The mechanism for film formation is improved in the dynamic process due to the "retainment" of the DMSO complex necessary for the intermediate phase which i) promotes intercalation between the PbI_2-CsI and the MAI-FAI and ii) slows down the crystallization of $Cs_{0.15}(MA_{0.7}FA_{0.3})_{0.85}PbI_3$ for full perovskite conversion and film uniformity. It is also found that as-deposited perovskite film by dynamic process is more ordered with higher degree of preferred orientation.

All of these improvements can be seen in film thickness (thicker) and more uniform morphology (confirmed by scanning electron microscopy and optical measurements); better crystallinity and higher degree of preferred crystal orientation (confirmed by XRD); and higher carrier lifetimes (confirmed by steady state and time-resolved photoluminescence measurements). A champion dynamic processed perovskite device reaches an efficiency of 18.4%, while the average efficiency of these devices reaches 17.0% which is a 3% improvement over the average efficiency of static processed devices (see Table 1).

Table 1. J-V characteristics of devices fabricated by static and dynamic processes

	PCE (%)	Jsc (mA/cm2)	Voc (mV)	FF (%)
Static Process	14.1±1.4	18.6±1.7	1057±31	71.4±5.0
Dynamic Process	17.0±0.6	20.9±0.6	1061±13	76.6±1.7

The new insights provided by this work are important for future optimization of sequential fabrication method for perovskite solar cells as the sequential process has the advantage of flexibility allowing i) solvent incompatibility of different precursors to be resolved; ii) separate optimizations for each step; and iii) different deposition methods to be used for each deposition step.