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Deconstruction-assisted perovskite formation for sequential solution processing of $Cs_{0.15}(MA_{0.7}FA_{0.3})_{0.85}PbI_3$ solar cells and the effect of chloride additive.

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Organic-inorganic hybrid lead halide perovskite has shown to be one of the best light-harvesting materials for solar cell in the last decade. However, there still is needed a deeper understanding of phase and film formation for better control of device fabrication. In this work, we visualise the formation mechanism of $Cs_{0.15}(MA_{0.7}FA_{0.3})_{0.85}PbI_3$ perovskite by the sequential spin-coating method and how changes in the dispensing timing and substrate motion affect the formation process and properties of the final film quality. In particular, this is the first time that we are able to visualise (Figure 1) and identify the different stages of the film formation (Figure 2) : i) “initial formation”; ii) “perovskite deconstruction” and iii) “perovskite re-crystallisation”. This particularly applies to films that are sequentially spin-coated and involve the use of dimethyl sulfoxide (DMSO) as the “deconstruction” is caused by the formation of intermediate-DMSO-complex. These findings are validated by FTIR and XRD measurements. Comparison among different sequential spin-coating processes also suggests that motion causes an earlier onset of deconstruction, which will lead to a slower re-crystallisation resulting in better quality perovskite film with less non-perovskite phase. This can be achieved by motion dispensing and dynamic processing (where there is no stoppage between the two sequential steps [1]). Reasons for the earlier onset of deconstruction are the higher kinetic energy supplied by the dynamic process. The resultant $Cs_{0.15}(MA_{0.7}FA_{0.3})_{0.85}PbI_3$ film has lower amount of non-perovskite phase and better crystallinity for solar cells with fewer defects and better device performances.

In addition, the effect of chloride (Cl-) incorporation in sequential solution process is also investigated on grain size, crystallinity, photoluminescence response and carrier lifetime. The difference in the effect of Cl- incorporation when done in the 1st step or the 2nd step in a sequential solution process is also investigated. It is found that Cl is effective in producing better morphology and reducing defects. However, it is most effective when MACl incorporated in the first step. Reasons will be presented in the conference. The champion perovskite device has an efficiency of 19.6%, which is a 2.2% absolute improvement over the champion device without Cl additives.

This work has provided more insights into the complex stages involved in perovskite conversion specific to sequential processing and film formation mechanism as a result of additive engineering. The knowledge will aid future process optimisation for better device fabrication.

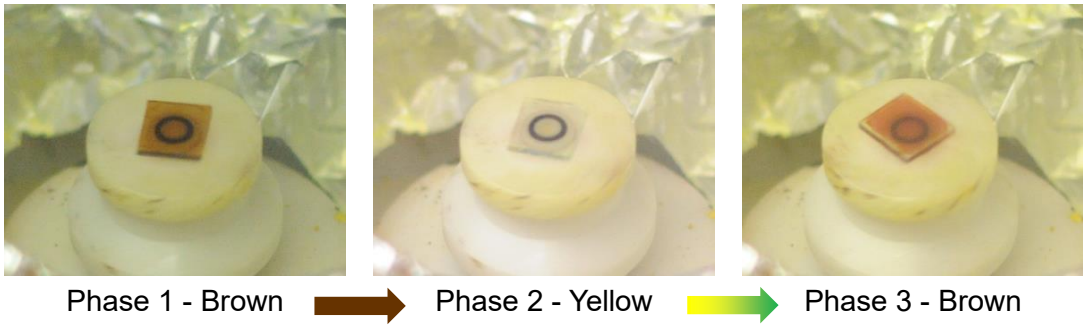

Figure 1 Images of step 2 of a sequential process captured by high speed camera showing colour change representing: Phase 1 - Brown, Phase 2 – Yellow, and Phase 3 - Brown.

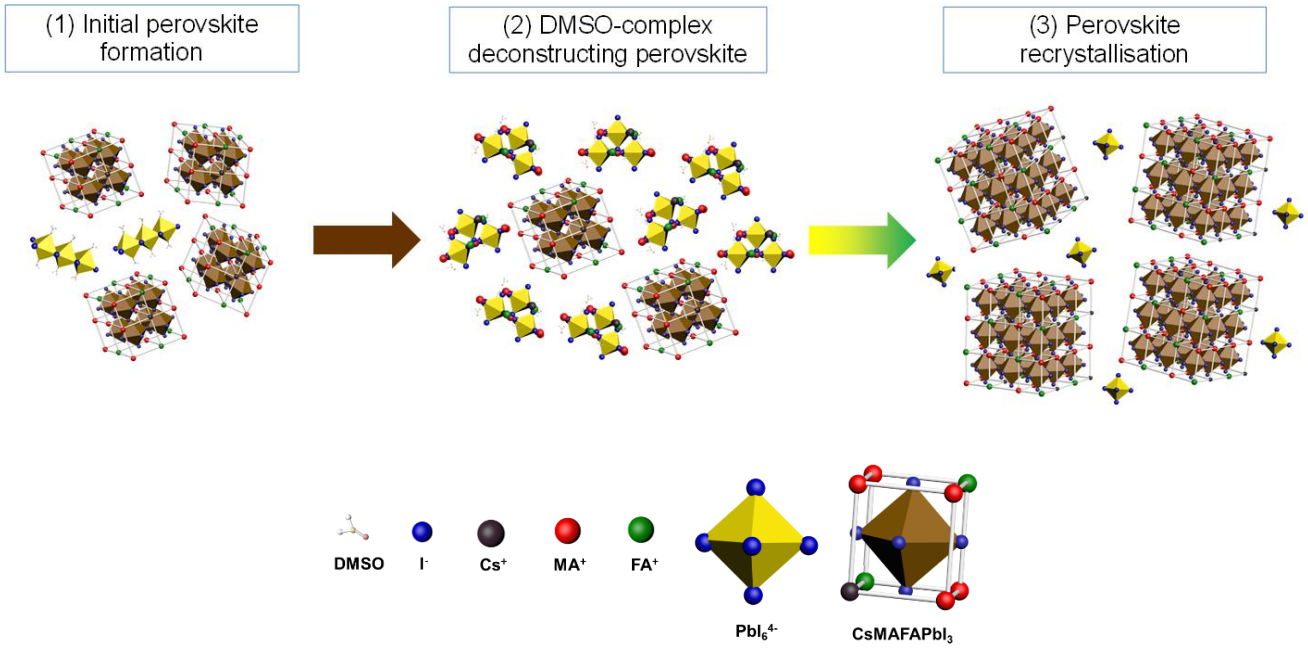

Figure 2 Schematic showing phase transition during the second step of sequential processing: (a) Phase 1 - initial perovskite formation; (b) Phase 2 - perovskite deconstruction due to the formation of $(MA/FA/Cs)_4PbI_6$-2DMSO complex); (c) Phase 3 - perovskite re-crystallisation.

References

[1] J. Bing, J. Kim, M. Zhang, J. Zheng, D. S. Lee, Y. Cho, X. Deng, C. F. J. Lau, Y. Li, M. A. Green, S. Huang, A. W. Y. Ho‐Baillie, "The Impact of a 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", Small, 2019,15,1804858.