Application of the DoE (design of experiments) for a simplified and rapid optimization of the one step spin-coating deposition of mixed Sn-Pb perovskite

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For the past years tin-lead perovskites have attracted much attention, as their band-gap can be tuned over a much wider range than full lead counterparts.[1] However, mixed tin-lead perovskites still suffer from two major drawbacks: poor stability towards air and a very fast crystallization, because of the lower solubility and greater Lewis acidity of tin halides than lead analogs. Fast crystallization results in poor film quality (*e.g.* pinholes, dendrites), which causes inefficient charge transport/collection, severe charge recombination and device shorting, resulting in inferior device performances.[2] Therefore, mixed tin-lead perovskites need precise control of the nucleation. For this study, we choose to focus on the spin-coating deposition technique, which consists in drop casting the perovskite ink onto a spinning substrate. This technique is cheap and easy to set up, yet, many parameters have to be taken into account, such as the solution drop casting method, the number of plateaus, their speed/acceleration/duration, or the nitrogen flow flux over the substrate. This variety of parameters makes this process time-consuming and tedious to optimize. This work aims to apply a statistical method to effectively optimize the spin-coating process of a mixed tin-lead perovskite, starting from the parameters already in use in the lab for the deposition of a full-lead analog.

The design of experiment (DoE) method allows a simultaneous and very efficient screening of multiple parameters (*e.g.* speed, duration), and provides a rapid quantification in a given response (*e.g.* layer thickness, absorption). Two consecutive DoEs are described in this work. For each set of parameters, UV-vis spectroscopy, SEM and confocal imaging, and XRD characterization are used to analyze the perovskite films. The performances of resulting devices are assessed by I(V) and EQE characterization. Firstly, in the case of a 1-step spin-coating deposition (as depicted in Fig.1a), the durations and speeds of the two plateaus are investigated. This DoE highlights the great influence of the duration of the second plateau over the film morphology, and behind it the washing-solvent introduction, in accordance with the literature.[3] Thus second DoE focuses on the washing-solvent drop casting (*e.g.* deposition time, flow speed, nature). For each measurement a statistical software allowed to determine most influent parameters and to demonstrate the validity of this DoE approach. From these studies we successfully optimized the spin-coater parameters with a very limited number of experiments and showed the power and relevance of DoEs. We highlighted the main parameters affecting the nucleation kinetics and therefore were able to tune them accordingly to produce a dense, pinhole-free perovskite layer. This time-saving method can not only be applied for any changes in the perovskite ink but more broadly can be used with other deposition techniques.

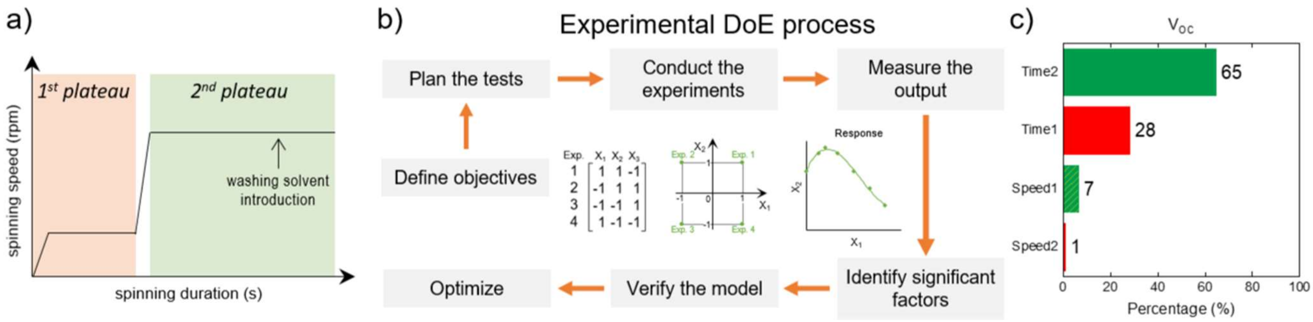

Figure 1: a) Scheme of the spinning speed evolution versus time for a 2-plateaus 1-step spin-coating deposition b) Conceptual diagram of a DoE c) Example of the Pareto effect for the V_{OC}, where the colors are indicators of negative/positive impact of the parameters and the length, its the weight.

References:

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