Large Area Deposition of $CH_3NH_3PbI_3$ Films Using Aerosol-Assisted Chemical Vapour Deposition

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Aim

- To deposit large area dense, uniform perovskite films.

Why

- High quality perovskite will lead to improvement in the efficiency of the solar cells.
- Present methods of deposition are challenging to scale up, so restrict commercialization of cell production.

How

- Use of Atmospheric pressure CVD, as this is a cost effect method with a proven ability to integrate into industrial processes.
- Optimisation of deposition parameters and method.

Single stage process

Initially small area trials (2 cm x 10 cm) were carried out by the single stage process to determine critical parameters.

(a) Substrate position relative to the precursor inlet.

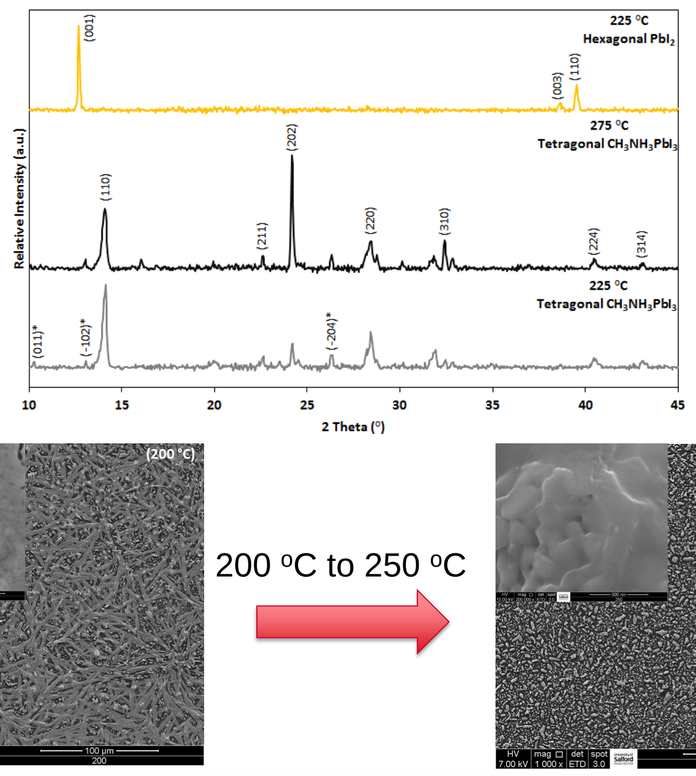
gas inlet

Perovskite Lead Iodide

This suggests that the mass transport of the methyl ammonium iodide (MAI) is the rate limiting step, resulting in a concentration gradient with most of the reaction taking place near the gas inlet and hence depleted further along with little to no MAI available for intercalation.

(b) Substrate temperature

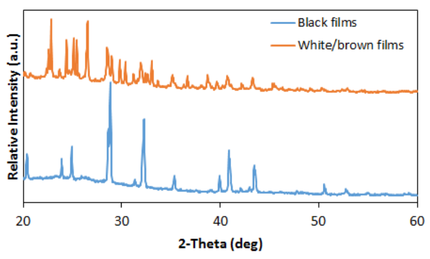
Ts < 150 °C only formation of PbI_2
Ts > 200 °C for perovskite formation. Tetragonal polycrystalline
Ts > 250 °C change in preferred orientation from (110) to (202)

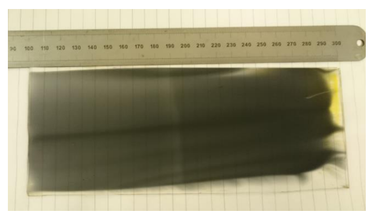


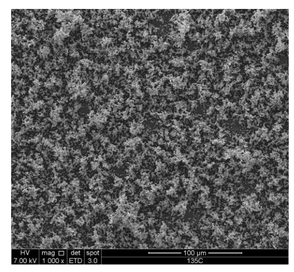
200 °C to 250 °C

Development to larger area

Problems
(a) While cooling films rapidly changed from black to white and then in air to brown. This is due to the intercalation of MAI molecules into PbI_2/$MAPbI_3$ lattices. Reducing the film exposure to MAI and removing while still warm retained the black perovskite.





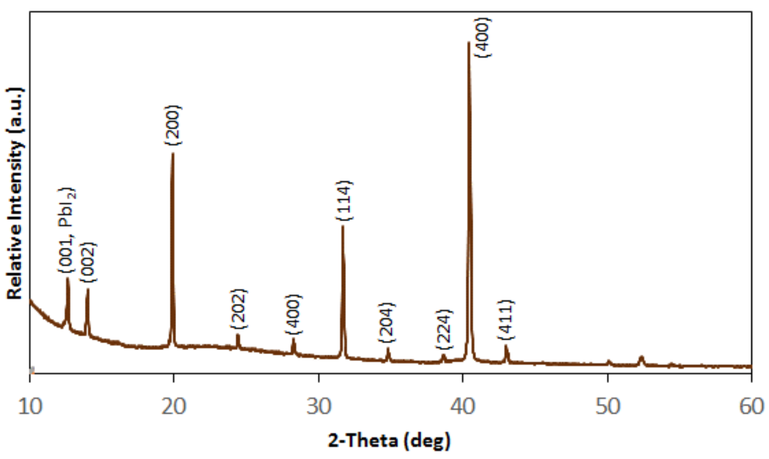


Problems
(b) The more serious issue was the increase in homogeneous reactions, leading to the formation of very soft, powdery films.

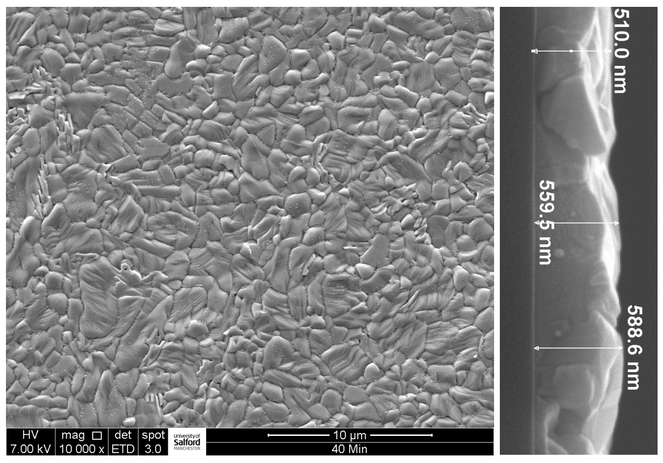
Three stage process

For better control of film development, a three step process was tested with each precursor being added sequentially under different deposition conditions [1].

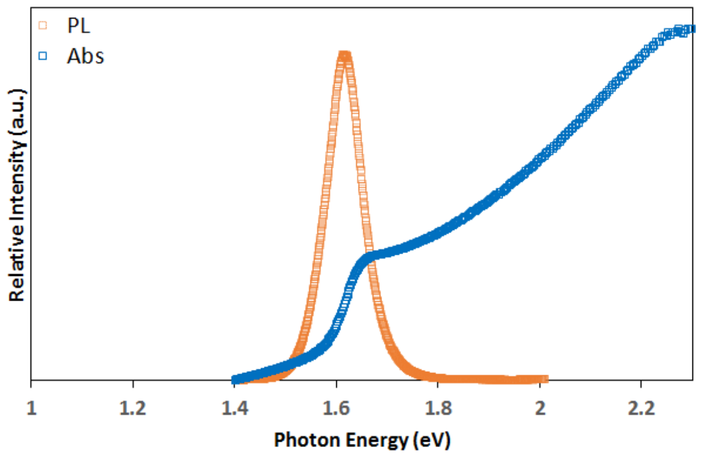
Robust, highly dense and uniform $CH_3NH_3PbI_3$ layers



XRD showing highly polycrystalline, tetragonal perovskite films.



Uniform perovskite films with ~ 0.53 µm in thickness.



Absorption and emission peak at 1.62 eV (765 nm).

[1]S. Chen et al, Cryst.Eng.Comm, 17 (2015) 7486.

Conclusion

Atmospheric pressure CVD has been used to deposit thin films of polycrystalline perovskite on glass. A single step process worked successfully for small area deposition, but it was necessary to move to a three step process for better control of the individual parameters needed for each reactant for large scale deposition of dense, pin-hole free perovskite films.

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