Characterisation and performance of vapour-deposited lead halide perovskite films for radiation detection applications

M. Zahid[1], A.Dickinson[1], J. Jayarathne[1], C. Crean[2], P.J. Sellin[1]

1. School of Mathematics and Physics, University of Surrey, Guildford, United Kingdom
2. School of Chemistry and Chemical Engineering, University of Surrey, Guildford, United Kingdom

UNIVERSITY OF SURREY

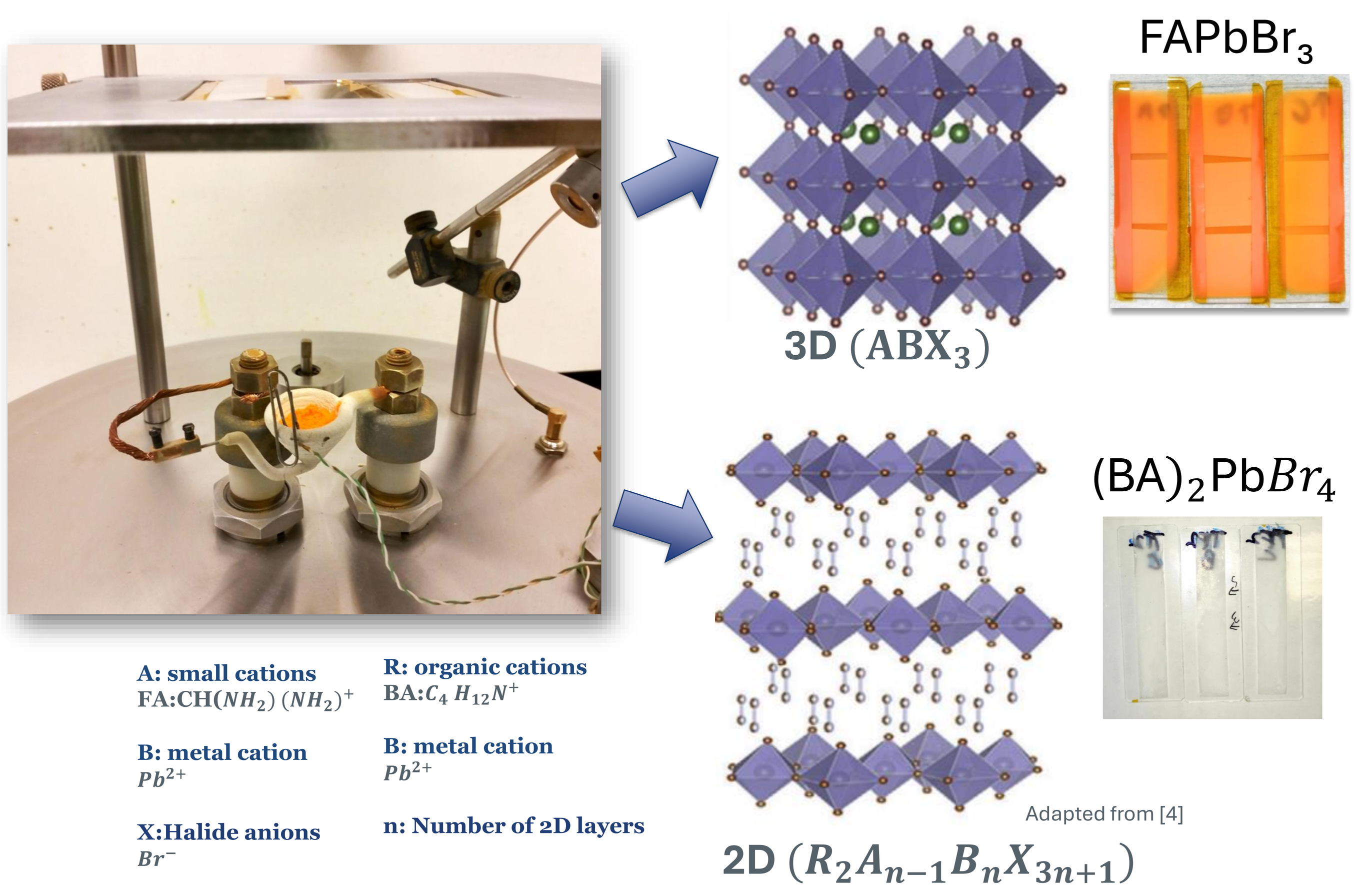
Introduction

- Perovskite thin film (TFs) show potential use as large-area imaging detectors due to its uniform detector film formation, compatibility with flexible substrates, and easy integration with electronic systems [1].
- Lead halide perovskite materials are promising for radiation detectors due to their high Z-number and density, good charge transport properties and potential use as scintillator [2].

Problem Statement & Research Aim

- Solution processing is predominantly fabricated for thin film but suffers from reproducibility and uniformity[3].
- Although single-source vacuum deposition (SSVD) for thin films is well established for photovoltaic applications and offers a simple process and more uniform results, it is largely unexplored as a radiation detector.
- In this work, the fabricated SSVD 3D and 2D lead halide perovskite thin films are characterised and their scintillation performance are reported.

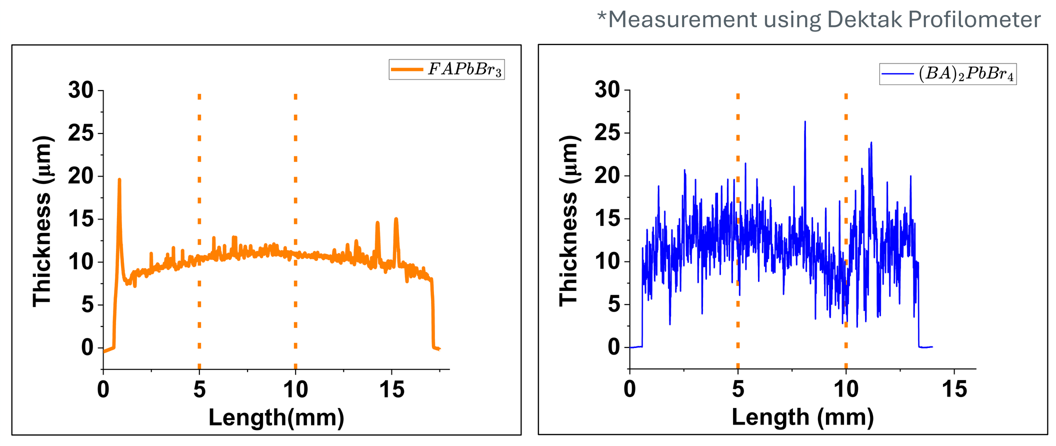
Single-Source Vapour Deposition (SSVD) for 3D & 2D perovskites



- Lead halide perovskite powder FAPbBr$_3$ (FA^+ = CH$(NH_2)_2^+$ and $(BA)_2PbBr_4$ (BA = C_4H_9N) were sublimated as single precursor, on static substrate.
- For FAPbBr$_3$, the source substrate distance was 9 cm and $(BA)_2PbBr_4$ was 5 cm, resulting in a deposition rate of 0.018 μm/min and 0.033 μm/min respectively.
- Compared with 3D perovskite, 2D perovskite has layered structured separated by large volume organic cation (BA) component. This quantum well structure exhibits not only bright light scintillation and superior intrinsic stability [5] .

Thickness & Roughness Analysis

*Measurement using Dektak Profilometer

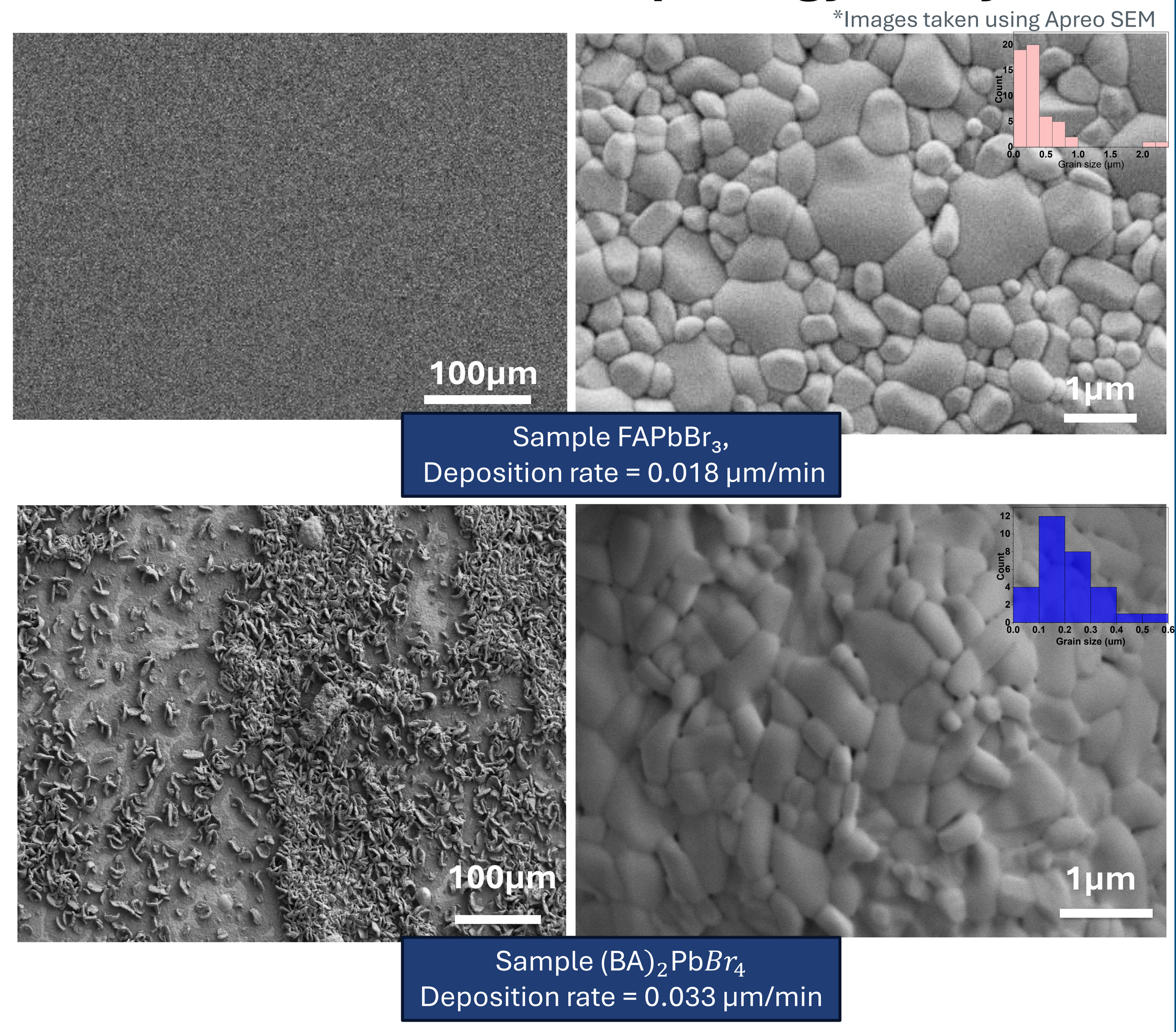


Sample	Mean thickness (μm)	Arithmetic Roughness (R_a) (μm)
FAPbBr$_3$	11.0 ± 4.2	0.3 ± 0.2
$(BA)_2PbBr_4$	11.5 ± 3.6	2.5 ± 1.8

- The maximum thickness achieved was 11.5 μm with sample $(BA)_2PbBr_4$ but with higher roughness.
- Sample FAPbBr$_3$ shows uniform thickness and smoother profile suggesting low deposition rate improve the film uniformity.
- Film thickness and film roughness can be controlled by optimising the deposition rate.

SEM Structural & Morphology Analysis

*Images taken using Apreo SEM

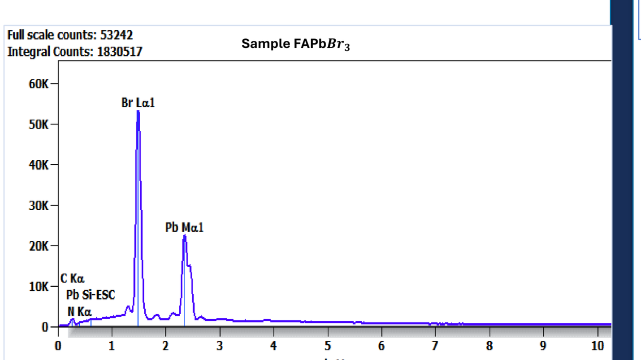


- At 100um scale image, FAPbBr$_3$ film shows smoother morphology than $(BA)_2PbBr_4$ film, consistent with the measured thickness profile.
- The low deposition rate in FAPbBr$_3$ film allows sufficient surface diffusion and adatom rearrangement that contribute for smoother morphology.
- At the 1 μm scale, both samples show irregular grain sizes and pin hole free.
- Absence of pinholes is important for higher light scintillation as it reduces defect densities and suppressed non radiative recombination.

EDS Compositional Analysis

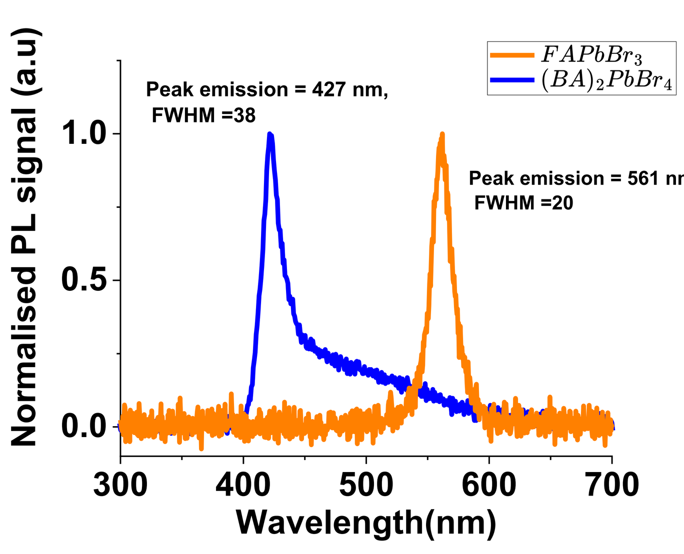
*Analysis using Pathfinder software.

Element Atom (%)	Sample FAPbBr_3	Sample $(BA)_2$PbBr_4
Br-L	53.6±0.4	39.6±1.7
Pb-M	17.8 ±0.3	9.7±0.3
Ratio Pb:Br	1:3	1:4



- Similar with 3D perovskite thin film, 2D perovskite thin film showed a consistent stoichiometric ratio of lead halide compound.

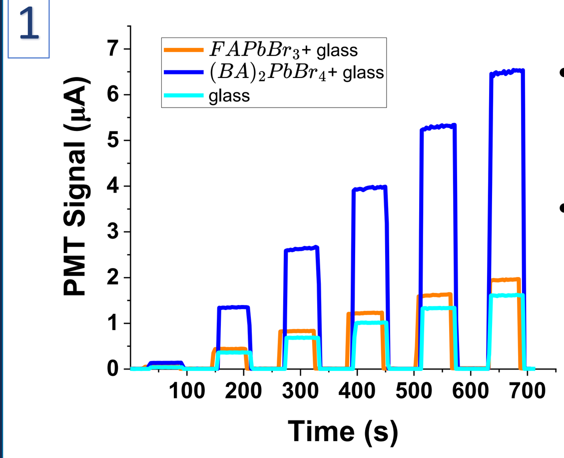
Photoluminescence(PL) Analysis



- The peak emission of FAPbBr$_3$ and $(BA)_2PbBr_4$ were 560 nm and 430 nm [6] respectively and consistent with its crystal properties, suggesting the same composition after the vacuum deposition process.
- The blue shift of peak emission from 3D to 2D perovskite originates from the incorporation of large organic cation into perovskite lattice, suggesting the wavelength of perovskite can be tuned to match the spectral sensitivity of scintillation reader (e.g PMT and SiPM).

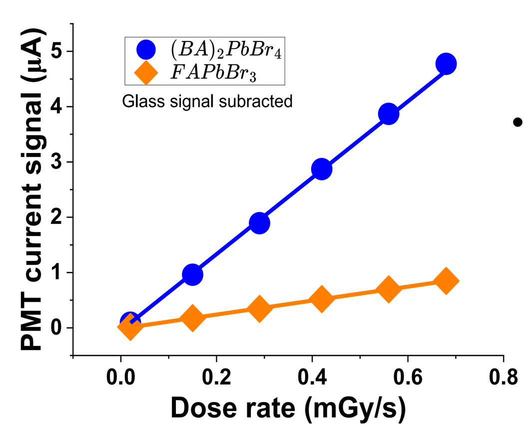
X-ray Scintillator Performance

1



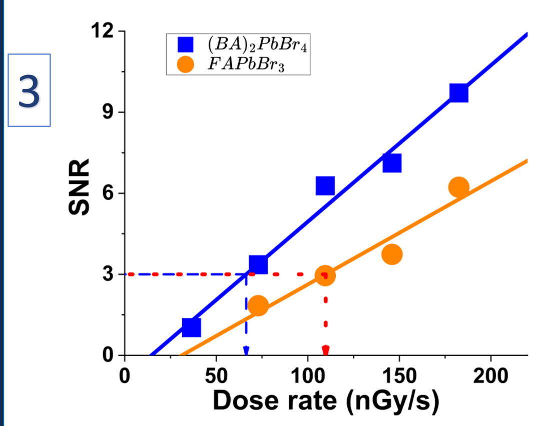
- Both thin films shows rapid ON/OFF photocurrent response.
- The photocurrent increase linearly with X-ray, which is important for reliable measurement.

2



- The X-ray sensitivity of sample $(BA)_2PbBr_4$ was higher than sample FAPbBr$_3$.
- This is due to 2D structural that consist inorganic sheets separated by organic space cations that form natural quantum wells, resulting in higher exciton binding energy and radiative recombination.

3



*All measurements using 50kV Amptek Mini X-ray tube

- A linear dose response between SNR and the X-ray dose rate.
- The estimated limit of detection (LOD) for FAPbBr$_3$ and $(BA)_2PbBr_4$ were 110 nGy/s and 66 nGy/s respectively indicates suitability for low dose X-ray applications.

Conclusions

- 3D and 2D lead halide perovskite thin films were fabricated via SSVD shows potential as radiation detector.
- Both materials show linear dose response, and 2D perovskite thin film shows higher RL sensitivity compared to 3D counterparts.
- Using SSVD, improved film smoothness can be achieved by optimising the deposition rate.
- PL signal can be tuned to match the spectral sensitivity of the scintillation reader devices by selecting an appropriate organic cation.

Future Work

- Optimisation of the vacuum evaporator system to achieve a thicker and more uniform thin film.

Kementerian Kesihatan Malaysia

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