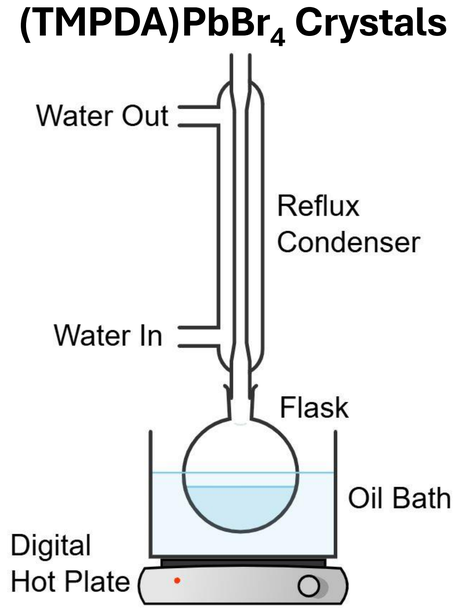
Growth and Pellet Fabrication of 2D Perovskite Scintillators

Amy Dickinson[1], Carol Crean[1], Joydip Ghosh[2], Phasit Petisiwaveth[1,3], Paul Sellin[1]

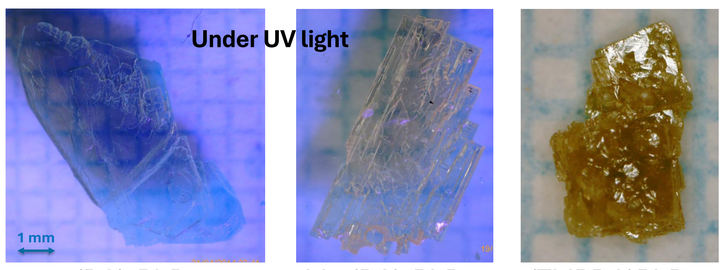
1. School of Mathematics and Physics, University of Surrey, Guildford, United Kingdom
2. Department of Chemistry, University of Oxford, United Kingdom
3. School of Physics, University of Wollongong, Australia

- 2D perovskite materials are promising for radiation detection applications due to their high x-ray and gamma absorption, high light yields and fast decay times. These materials show great versatility as they can be tuned by swapping out the organic spacers and halides to suit the desired application [1].
- Growing high-quality, high light yield crystals is challenging due to random nucleation and uncontrolled growth rates [2]. The fabrication of scintillator pellets has been proven as a useful technique to provide uniformity, and we are now applying this to 2D Perovskites [3].
- This work presents a comprehensive investigation of the optical properties of 2D perovskite scintillators and the effects of adding dopants.
- This work introduces a pellet fabrication method used to produce uniform perovskite pellets.

Synthesis of $(BA)_2PbBr_4$ and $(TMPDA)PbBr_4$ Crystals



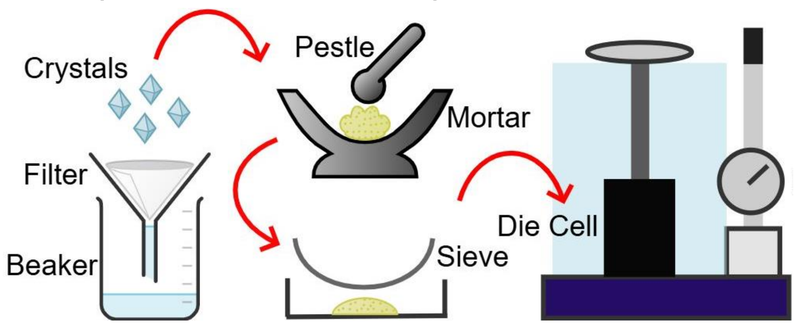
The solution is heated to 100°C and then cooled to room-temperature over 7-14 days.



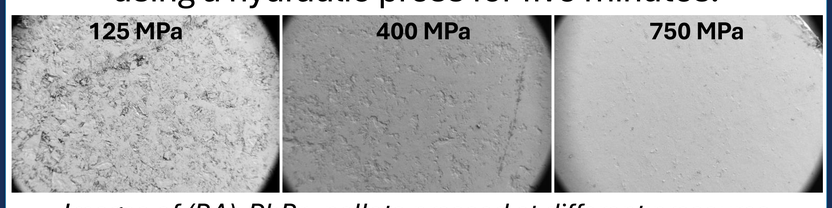
$(BA)_2PbBr_4$ | Mn:$(BA)_2PbBr_4$ | $(TMPDA)PbBr_4$

Pellet Fabrication

The crystals are washed several times using acetonitrile, and dried. They're then roughly ground into a powder using a pestle and mortar. The powder is sieved and poured into a die cell.



The powder is pressed at constant pressure using a hydraulic press for five minutes.

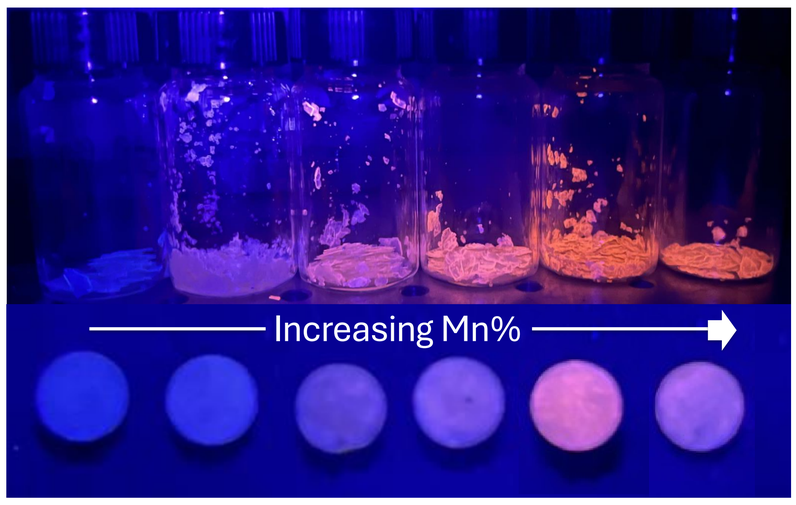


Images of $(BA)_2PbBr_4$ pellets pressed at different pressures under an electron microscope at 50x magnification

At higher pressures, the surface has fewer pits and density increases.

Manganese-doped BA_2PbBr_4

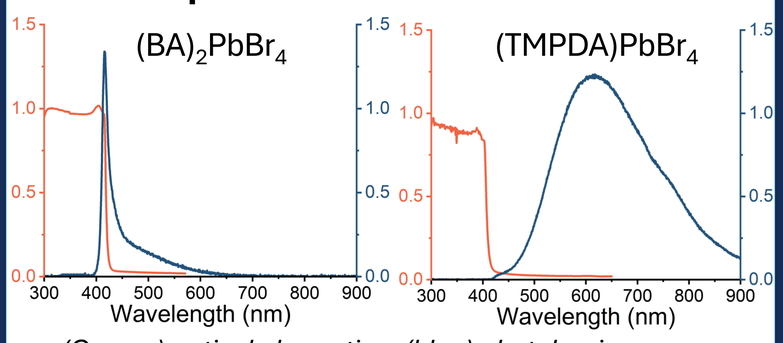
Adding dopants during synthesis provides a way to tune band gaps and increase light yield. The dopant percentage influences these factors. Manganese doping introduces a second 'orange' peak while increasing overall light yield.



(Top) Mn:$(BA)_2PbBr_4$ crystals; (bottom) Mn:$(BA)_2PbBr_4$ pellets

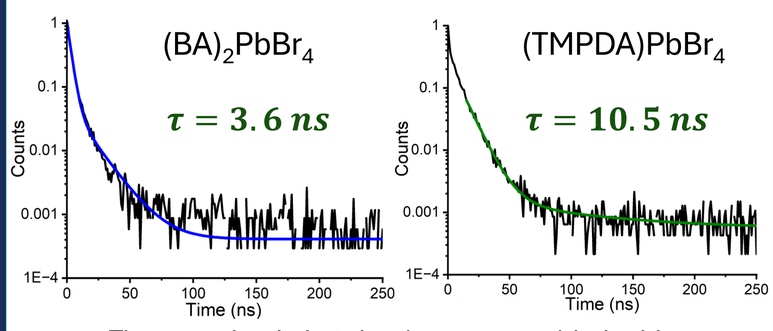
Other dopants such as Li^+ and Rb^+ can be used to enhance light yields without effecting the band gap. The wide variety of available dopants increases the tunability of 2D perovskites.

Optical Characterisations



(Orange) optical absorption; (blue) photoluminescence

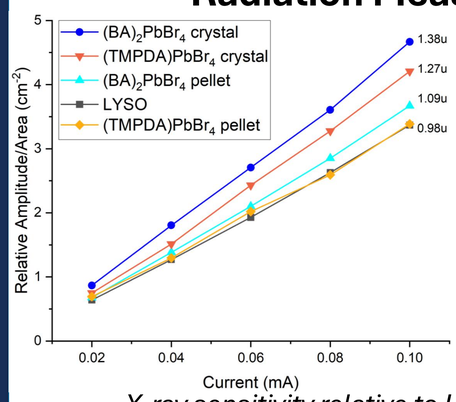
$(BA)_2PbBr_4$ belongs to the Ruddlesden-Popper (RP) sub-group of perovskites while $(TMPDA)PbBr_4$ is a Dion-Jacobson (DJ). RP typically have narrow emission peaks and small Stokes shifts whereas DJ are usually broad emitters with large Stokes shifts.



Time-resolved photoluminescence with double exponential fit

These 2D perovskites all have very short decay times which implies efficient radiative recombination processes within the crystals. $(TMPDA)PbBr_4$ has an unusually short lifetime for a broad emitter which is promising when large Stokes shifts and a fast decay is needed.

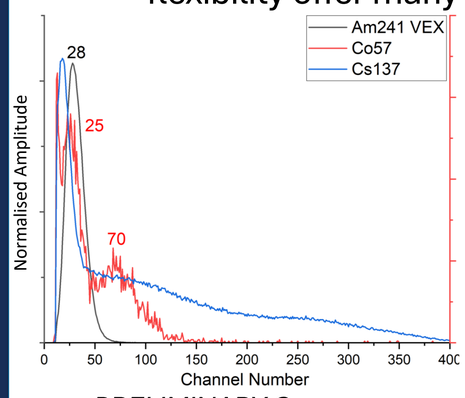
Radiation Measurements



The light yields under x-ray excitation for $(BA)_2PbBr_4$ and $(TMPDA)PbBr_4$ crystals are higher than LYSO which is 33,000 photons/MeV [4].

X-ray sensitivity relative to LYSO at 80kpV, 0.1mA

The pellets show a lower sensitivity at 79% and 77% of the $(BA)_2PbBr_4$ and $(TMPDA)PbBr_4$ crystals respectively. Although the pellets are less sensitive, their uniformity and structural flexibility offer many more benefits.



Under gamma-source excitation, the $(BA)_2PbBr_4$ crystals show promising responses, with resolvable peaks.

PRELIMINARY Gamma-source scintillation using PMT

Future work will develop this technique further.

Conclusion

- 2D perovskite scintillators have versatile applications within radiation physics due to the tuneability of the cations, halides and dopants.
- $(BA)_2PbBr_4$ and $(TMPDA)PbBr_4$ crystals can be synthesised using a slow-cooling method however, controlling growth rates is challenging. Successful trials of Manganese doped $(BA)_2PbBr_4$ has also been shown.
- Pellets have been successfully fabricated using a cold-pressing method and has improved uniformity and reproducibility.
- The light yields under x-ray excitation for $(BA)_2PbBr_4$ and $(TMPDA)PbBr_4$ crystals are higher than LYSO, with the pellets showing slightly lower sensitivity at 79% and 77% respectively.

Future Work

Future work will focus on the optimisation of these 2D perovskite pellets by investigating hot-pressing and sintering techniques. Also, exploring other dopants to further increase light yields will benefit the development of pellets.

References

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[2] Sheikh, T. and Nag, A., 2019. Mn Doping in Centimeter-Sized Layered 2D Butylammonium Lead Bromide (BA_2PbBr_4) Single Crystals and Their Optical Properties.
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