Supporting Information

Synergistic Effect of Anti-Solvent Bath Method and Optimized Annealing Conditions to Spawn High Quality Triple Cation Thin Films

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1	Cs(0.05)FA(0.79)MA(0.16)PbI(2.7)Br(0.3)					
Material	Cs	FAI	MAI	Pbl	PbBr	
Weight (mg)	19	300.2	60.8	826.2	91.8	
2	Cs(0.05)F/	Cs(0.05)FA(0.67)MA(0.28)PbI(2.7)Br(0.3)				
Material	Cs	FAI	MAI	Pbl	PbBr	
Weight (mg)	19	254.6	106.4	826.2	91.8	
3	Cs(0.05)F/	Cs(0.05)FA(0.55)MA(0.40)PbI(2.7)Br(0.3)				
Material	Cs	FAI	MAI	Pbl	PbBr	
Weight (mg)	19	209	152	826.2	91.8	
4	Cs(0.05)F/	Cs(0.05)FA(0.40)MA(0.55)PbI(2.7)Br(0.3)				
Material	Cs	FAI	MAI	Pbl	PbBr	
Weight (mg)	19	152	209	826.2	91.8	
5	Cs(0.05)F/	Cs(0.05)FA(0.28)MA(0.67)PbI(2.7)Br(0.3)				
Material	Cs	FAI	MAI	Pbl	PbBr	
Weight (mg)	19	106.4	254.6	826.2	91.8	
6	Cs(0.05)FA(0.16)MA(0.79)PbI(2.7)Br(0.3)					
Material	Cs	FAI	MAI	Pbl	PbBr	
Weight (mg)	19	60.8	300.2	826.2	91.8	

Table S1: Table of triple cation perovskite solution values and weights

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Figure S1: Full XRD spectra for FAvsMA triple cation perovskite. The symbols '#' and '*' represent δ -phase perovskite and cubic PbI₂, respectively. Peaks were indexed via literature [1].

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Table S2: Summary of FWHM and crystallite size for the FA;MA triple cation perovskite thin films - based off the XRD results in Figure 1 and Equation 1

Sample	Before Heat FWHM (degree)	After Heat FWHM (degree)	
0.79 FA : 0.16 MA	1.03	1.1704	
0.67 FA : 0.28 MA	0.1993	0.205	
0.55 FA : 0.40 MA	0.1384	0.2613	
0.40 FA : 0.55 MA	0.1645	0.1417	
0.28 FA : 0.67 MA	0.1690	0.1159	
0.16 FA : 0.79 MA	0.1672	0.1399	



Figure S2: SEM images all at 30,000x magnification of FAvsMA triple cation perovskite thin films before and after thermal degradation on hotplate at 85° C for 2 hours. The lines across the images with circles are from the Matlab program used to calculate grain size. Program is open source available on MathWorks written by Matthias Funk from the Karlsruhe Institute of Technology. The numbering scheme coordinates with the numbers found in Table S1, for the solution amounts of FAvsMA - with 1 being FA rich and 6 being MA rich.

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Figure S3: Calculated mean for grain size of films before and after heating. Mean calculated from open source program available on MathWorks written by Matthias Funk from the Karlsruhe Institute of Technology.



Figure S4: UV-vis measurements of FAvsMA films both before and after thermal degradation on hotplate. The 'AH' in the plot legend stands for 'after heat', denoting that measurement as the one after hotplate degradation. Slight increase in absorption was observed for the MA rich perovskite film (0.16 FA:0.79 MA).

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Figure S5: Thickness of $Cs_{0.05}FA_{0.16}MA_{0.79}PbI_{2.7}Br_{0.3}$ thin films grown on FTO/TiO₂ substrates. Same films were used in the UV-vis measurements portrayed in Figure 2 of the main text. Thickness was measured via profilometer, details of measurement device can be found in the Experimental section of the main text.

Table S3: FWHM and mean grain size (calculated from the histograms and Matlab line cut program) for the MA rich solution (0.16 FA : 0.79 MA). The FWHM comes from the XRD characterization, while the mean grain size comes from the analysis of the SEM images.

Sample	FWHM (degree)	Mean Grain Size (nm)
80° C - 30 min	0.114	312.5
80° C - 60 min	0.1199	329.63
80° C - 90 min	0.1387	316.67
80° C - 120 min	0.1139	877.78
150° C - 30 min	0.1432	253.39
150° C - 60 min	0.1474	n/a
150° C - 90 min	0.1405	n/a
150° C - 120 min	0.1392	n/a

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Figure S6: XRD results for the MA rich (0.16 FA:0.79 MA) films annealed at 80° C for 30, 60, 90, and 120 minutes. Once complete, films were measured, then placed back on hot plate for 2 hours at 85° C as an attempt to induce degradation. Once time had elapsed, films were removed and measured once again. The original XRD peak is shown in the corresponding solid color. The same film, once thermal degradation had been applied, is overlaid in dashed red.

References:

1. Sun, Y., et al., *Triple-cation mixed-halide perovskites: towards efficient, annealing-free and air-stable solar cells enabled by Pb (SCN) 2 additive.* Scientific reports, 2017. **7**(1): p. 1-7.

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