

Materials Advances

Accepted Manuscript

This article can be cited before page numbers have been issued, to do this please use: M. Brown, S. Borbon Rojas, L. Fernandez Izquierdo, R. Haroldson and M. A. Quevedo-López, *Mater. Adv.*, 2026, DOI: 10.1039/D6MA00161K.



This is an Accepted Manuscript, which has been through the Royal Society of Chemistry peer review process and has been accepted for publication.

Accepted Manuscripts are published online shortly after acceptance, before technical editing, formatting and proof reading. Using this free service, authors can make their results available to the community, in citable form, before we publish the edited article. We will replace this Accepted Manuscript with the edited and formatted Advance Article as soon as it is available.

You can find more information about Accepted Manuscripts in the [Information for Authors](#).

Please note that technical editing may introduce minor changes to the text and/or graphics, which may alter content. The journal's standard [Terms & Conditions](#) and the [Ethical guidelines](#) still apply. In no event shall the Royal Society of Chemistry be held responsible for any errors or omissions in this Accepted Manuscript or any consequences arising from the use of any information it contains.

7 Optimized Grain Growth in CsPbBr₃ Thick Films Using Mist 8 Assisted Dissolution-Recrystallization for Enhanced 9 Optoelectronic Performance

10 Monet Brown,^a Susana Borbon-Rojas,^a Leunam Fernandez-Izquierdo,^a Ross Haroldson^a and
11 Manuel Quevedo-Lopez^{*a}

12 All-inorganic metal halide perovskite (MHP) thick films offer exceptional promise for photodetector applications (visible, UV,
13 X-ray and gamma-ray) due to the stability of inorganic compositions and enhanced radiation absorption of thick-film
14 architectures. However, their performance is often limited, compared to their single crystal counterparts, due to small grain
15 sizes and high grain boundary densities which introduce trap states that reduce device efficiency. Here, we introduce a mist-
16 assisted dissolution–recrystallization (MADR) process to enhance the microstructure of blade-coated CsPbBr₃ thick films
17 (~200 μm). Applying a CsPbBr₃/DMSO mist at controlled concentrations (0.1–0.4 M) followed by low-temperature annealing
18 (60–110 °C) drives a 3.8-fold increase in average grain size from 0.83 ± 0.28 to 3.19 ± 1.57 μm. Under optimal conditions (0.1
19 M mist, 85 °C anneal), the MADR-treated film exhibits enlarged grains, reduced trap density, and improved crystallinity,
20 resulting in a ~10³ order photocurrent enhancement from 1.02 × 10⁻⁷ to 4.5 × 10⁻⁴ A/cm² at 5 V under 365 nm illumination
21 (125 mW/cm²). This MADR approach on blade coated films provides scalable, high-quality perovskite films with low thermal
22 budget and broad substrate compatibility, offering a versatile route for radiation detection and optoelectronic devices.

23 Introduction

24 The exceptional properties of CsPbBr₃ such as a wide bandgap
25 (□2.3 eV), high atomic numbers (Cs = 55, Pb = 82, and Br = 35)
26 and large mobility-lifetime product of (3.2 × 10⁻³ cm²/V) have
27 driven substantial interest in optoelectronic applications
28 including sensors, lasers, and photovoltaics.^{1–3} The wide band
29 gap reduces dark current, improving the signal-to-noise ratio,
30 while the high atomic numbers increase the photoelectric
31 absorption.^{2,4} These properties have enabled perovskite X-ray
32 detectors to achieve ~200-fold higher sensitivity and ~100-
33 fold lower detection limits than conventional a-Se-based X-ray
34 detectors.^{4,5} However, these results are primarily achieved in
35 single crystals of CsPbBr₃, which are challenging to scale for
36 commercial manufacturing.^{4,6} While scalable methods exist for
37 perovskite thin films (<1 μm), clinically relevant X-ray energies
38 (10 to 100 keV) require 100 to 500 μm thickness for full
39 attenuation.^{4,7–9} Similarly, exceptional performance has been
40 reported for visible-UV photodetectors. Bin Yang et al. reported
41 solution grown polycrystalline CsPbBr₃ thick films with
42 detectivities of 10¹⁴ Jones, exceeding single crystals having a
43 reported detectivity of 10¹¹ Jones under U-V exposure.^{10,11}
44 However, solution submersion methods are incompatible with
45 many device architectures.⁴ Blade coating has emerged as a
46 scalable fabrication method for perovskite thick films; however,
47 achieving high material quality with controlled grain size
48 remains a critical challenge.^{4,12–14}

49 Kim et al. report that the mobility-lifetime product (μτ) of the
50 MAPbI₃ blade-coated thick film (1.0 × 10⁻⁴ cm² V⁻¹) is two orders
51

52 of magnitude lower than that of single-crystal MAPbI₃ (1.2 × 10⁻²
53 cm² V⁻¹), highlighting the need to improve performance in
54 blade-coated films.^{15,16} Cheng et al. addressed this need in
55 MAPbI₃ blade-coated thick films using a pore-filling method that
56 enhanced X-ray sensitivity 5-fold, from 323.86 to 1616.01
57 μC-Gyair⁻¹ cm⁻² under an electric field of 44.44 V-mm⁻¹.⁶ While
58 this approach has advanced hybrid perovskite thick films, all
59 inorganic perovskite films remain essential for applications
60 requiring long term operational stability in ambient
61 environments.^{17,18} For all-inorganic systems such as CsPbBr₃, Shi
62 et al. and Gou et al. employed dissolution-recrystallization to
63 improve material quality and increase sensitivity via a powder
64 “washing” pre-deposition treatment and a surface treatment,
65 respectively.^{17,19} These approaches increased the sensitivity
66 from 798 to 14,430 μC-mGyair⁻¹·cm⁻² at 100 V-mm⁻¹ (Shi et al.)
67 and the methods reported by Gou et al. resulted in a 2-fold
68 sensitivity increase.^{17,19} Building on these strategies, this work
69 introduces a novel mist-assisted dissolution-recrystallization
70 technique to increase bulk grain size and crystallite size in all-
71 inorganic perovskite thick films (~200 μm).

72 Leveraging crystallization kinetics to control grain coarsening
73 and crystallite size in blade-coated CsPbBr₃ thick films, this work
74 systematically decouples the processing conditions that govern
75 dissolution-recrystallization dynamics, establishing a
76 framework for enhancing film quality and device-relevant
77 optoelectronic properties at low processing temperatures.
78 Grain size refers to the dimensions of individual particles
79 observed by microscopy, whereas crystallite size represents the
80 coherent scattering domain size determined from XRD analysis.
81 This method employs a DMSO-based mist solution containing
82 dissolved CsPbBr₃ (0.1 M, 0.2 M, or 0.4 M) applied to blade-
83 coated CsPbBr₃ thick films. The mist penetrates the bulk of the
film and partially dissolves the perovskite, creating a saturated
or supersaturated environment. Upon annealing at controlled
temperatures (60, 85, or 110 °C), the system undergoes Ostwald

^a Department of Material Science & Engineering, University of Texas at Dallas, 2601
N. Floyd Rd. RL10 Richardson, TX. 75080

† The data supporting this article have been included as part of the Supplementary Information. Supplementary information: Figure S1, Tables S1, Figure S2, Figure S3, Table S2 and Figure S4 and further experimental details. See DOI: [URL – format] https://doi.org/DOI]



1 ripening within the metastable zone, where grain coarsening
2 distinct from nucleation and crystal growth, proceeds
3 surface-energy-driven material transport from smaller grains
4 larger grains.^{20,21} This mist-assisted dissolution-recrystallization
5 (MADR) technique achieves substantial grain size increases
6 with optimal conditions (0.1 M, 85 °C) yielding grain sizes of $3.19 \pm 1.57 \mu\text{m}$
7 compared to $0.83 \pm 0.28 \mu\text{m}$ in the as-deposited film.
8 Correspondingly, photocurrent response, a key metric
9 evaluating charge collection efficiency in photodetector
10 applications, increases by over three orders of magnitude, from
11 1.02×10^{-7} to 4.5×10^{-4} A/cm² at 5 V in a Pt/CsPbBr₃/Pt planar
12 device illuminated with a 365 nm LED at 125 mW/cm². These
13 results demonstrate that controlled temperature
14 concentration dynamics enable targeted grain coarsening
15 CsPbBr₃, substantially improving optoelectronic performance.
16 This solution-based approach is generalizable to diverse
17 perovskite systems and can be applied iteratively to multiple
18 performance enhancements, offering a scalable pathway
19 toward high-efficiency thick film perovskite optoelectronic
20 devices and radiation detectors.

21 Results

22 While CsPbBr₃ single-crystals and thin films have been reported
23 to exhibit excellent optoelectronic properties, thick-films (100–
24 500 μm) required for commercially relevant high energy photonics
25 applications often struggle to achieve large grains, high
26 crystallinity, and high carrier mobility.⁴ Overcoming these
27 processing challenges is therefore essential for translating the
28 intrinsic properties of MHPs into practical device performance.
29 **Figure 1(a) and (b)** show cross-section SEM images of the
30 deposited CsPbBr₃ thick film fabricated by blade coating. The
31 deposited film exhibits a highly porous microstructure with
32 small grains, characteristic of blade-coated films.^{4,13} **Figure 1(c)**
33 shows the grain size distribution of the as-deposited CsPbBr₃
34 film, with an average grain size of $0.83 \pm 0.28 \mu\text{m}$. Such a small



35 **Figure 1:** (a) and (b): SEM cross section of as-deposited
36 CsPbBr₃ 200 μm thick film. (c) Grain size distribution of the as-
37 deposited CsPbBr₃ 200 μm thick film. The average grain size of
38 50 grains is $0.83 \pm 0.28 \mu\text{m}$.

39
40
41
42
43
44
45 grain-to-thickness ratio and high porosity commonly leads to
46 reduced device performance.^{4,19} Subsequently, MADR
47 treatment was applied to improve film quality. We

systematically investigated the effects of annealing
temperature and mist concentration on grain morphology and
crystallite size. The DMSO/CsPbBr₃ mist induces partial
dissolution when applied to the porous, fine-grained as-
deposited film and subsequent annealing promotes grain
growth that increases film density, grain size, and crystallite
size.

Impact of Mist Concentration and Annealing Temperature on CsPbBr₃ Film Microstructure.

To promote grain growth, MADR was performed on as-deposited
CsPbBr₃ thick films. Exposure to pure DMSO mist caused
nonuniform grain size distribution (**Figure S1**), whereas
incorporating CsPbBr₃ into the mist enabled more homogeneous
solvent-film interactions and improved film uniformity. A
DMSO/CsPbBr₃ mist solution was applied at concentrations of 0.1
M, 0.2 M, and 0.4 M due to the solubility limit of CsPbBr₃ in DMSO
(0.5 M).²² Cross-sectional SEM images of films treated at each
mist concentration are shown in **Figure 2**. Low mist concentration
(0.1 M) produced larger grains (1.91–3.19 μm), attributed to more
uncoordinated DMSO molecules that promote Ostwald
ripening.^{20,21} Conversely, at higher mist concentrations (0.4 M)
there is limited dissolution, yielding smaller grains (0.97–1.52 μm).
Recrystallization temperatures of 60, 85, and 110 °C were
selected to probe solvent assisted growth kinetics below the 189
°C boiling point of DMSO (**Figure 2**). Annealing at 85 °C produced
the largest grain size increase (1.45 – 3.19 μm). At 60 °C,
insufficient thermal energy limits grain coarsening (0.97–2.09
 μm), whereas at 110 °C, faster DMSO evaporation suppresses
further growth (1.52–1.91 μm). Grain sizes and corresponding
standard deviations are summarized in **Table S1**.

These observations are consistent with established crystallization
kinetics models: within the solubility and supersolubility
boundaries (metastable zone), Ostwald ripening is the dominant
mechanism; whereas above the supersolubility boundary (labile
region), supersaturation promotes spontaneous crystallization
and suppresses ripening.^{20,21,23,24} Under optimal conditions,
average grain size increased from $0.83 \pm 0.28 \mu\text{m}$ in the as-
deposited film to $3.19 \pm 1.57 \mu\text{m}$ with 0.1 M mist and a 85 °C
anneal, indicating that this condition lies near the center of the
metastable zone where ripening is most pronounced.^{23,26} These
parameters collectively define useful boundaries for
distinguishing the metastable and labile regions. XRD analysis,
discussed in the next section, was conducted to validate the
proposed mechanism by quantifying crystallite size evolution.
Larger crystallite sizes at higher mist concentrations corroborate
the SEM results and crystallization kinetics described above.²³



1

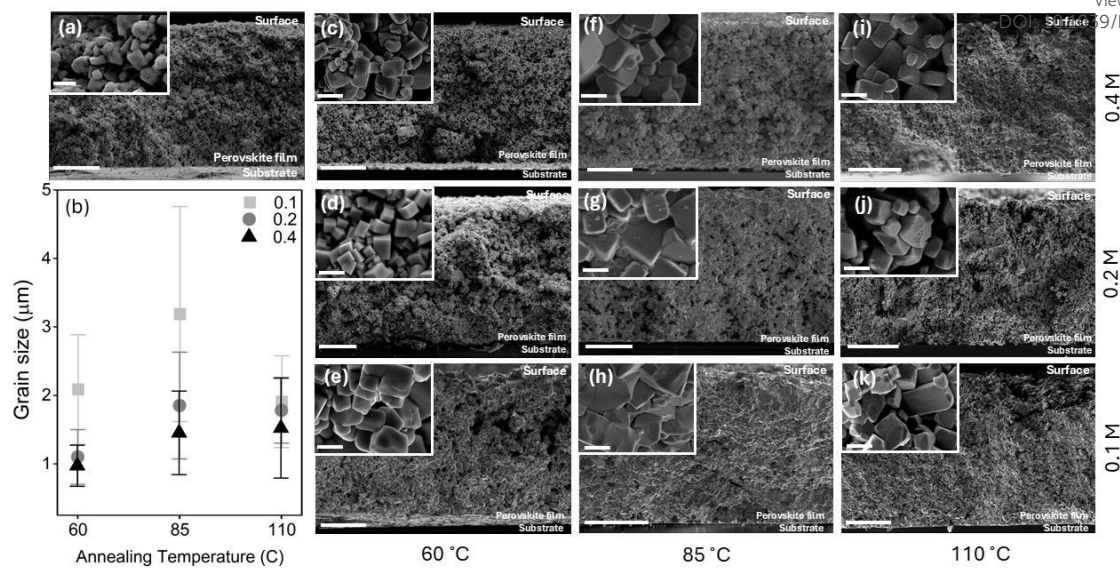
View Article Online
DOI: 10.1039/C6MA00161K

Figure 2: SEM cross sectional images of (a) as-deposited bladed-coated CsPbBr₃ film and (b) grain size distribution of mist-assisted dissolved and recrystallized CsPbBr₃ films. The films were annealed at (c-e) 60 °C, (f-h) 85 °C, and (i-k) 110 °C using a mist solution of DMSO and CsPbBr₃ powder at concentrations of 0.1 M (bottom row), 0.2 M (middle row), and 0.4 M (top row). All scale bars represent 50 μm and 2 μm in insets.

Impact of Mist Concentration and Annealing Temperature on CsPbBr₃ Crystallinity

The effects of mist solution concentration and annealing temperature on film composition were evaluated by X-ray diffraction (Figure 3). At moderate mist concentration (0.2 M), secondary Cs-Pb-Br phases formed at 60 °C, whereas higher mist concentration (0.4 M) induces secondary phase formation at both 60 and 85 °C. At 60 °C, diffraction peaks at 11.6, 12.5, 30.9, and 2θ (Degrees) correspond to the tetragonal CsPb₂Br₅ and rhombohedral Cs₄PbBr₆ (Figure S2). Peaks associated with rhombohedral Cs₄PbBr₆ appeared in 0.4 M films annealed at 60 and 85 °C. No reflections associated with secondary phases were observed at 110 °C. Phase compositions are summarized in Table S1.

The XRD data were further analyzed to estimate crystallite size using the Scherrer equation.²⁷ As shown in Figure 3(d), MADR treatment at 85 °C with 0.1 M mist concentration increased crystallite size to 44.22 ± 10.81 nm from 40.92 ± 2.66 nm in the as-deposited film. At 110 °C with 0.4 M mist concentration, the crystallite size increased to 125.48 ± 9.56 nm, the maximum observed value. Crystal growth occurs via the expansion of a single crystalline domain through coalescence of domains with coherent planes, a process favored by the high supersaturation conditions that bring crystallites into proximity.²⁸ Grain growth results from the aggregation of multiple domains with incoherent planes. Correlation of XRD and SEM results show that variations in mist concentration and annealing temperature modulate the balance between these mechanisms, determining which mechanism dominates under specific MADR conditions.

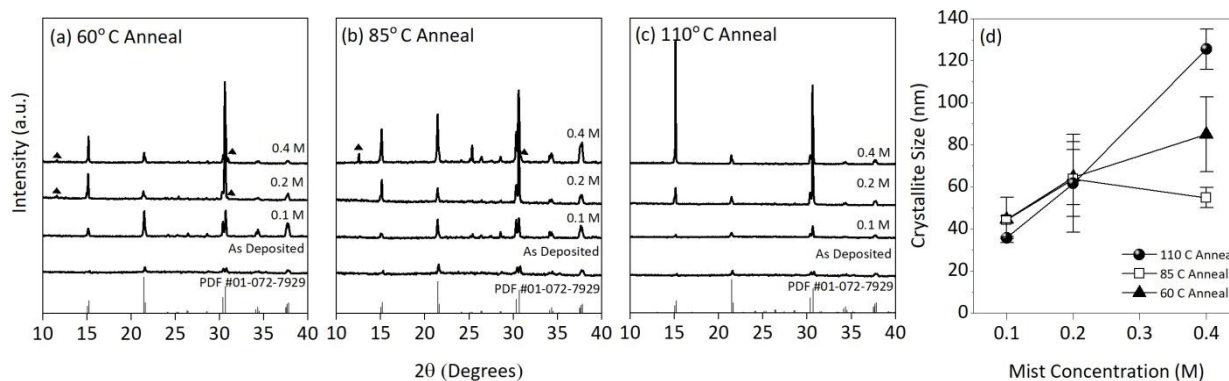


Figure 3: X-ray diffraction patterns for the as-deposited and treated films at each mist solution concentration (0.1, 0.2 and 0.4 M) and annealed at (a) 60 °C, (b) 85 °C, (c) 110 °C are presented. Peaks that do not correspond to Orthorhombic CsPbBr₃ are accompanied by a triangle with tetragonal CsPb₂Br₅ and rhombohedral Cs₄PbBr₆ present at 60 °C and rhombohedral Cs₄PbBr₆ at 85 °C. (d) Crystallite size as a function of concentration and mist solution concentration.

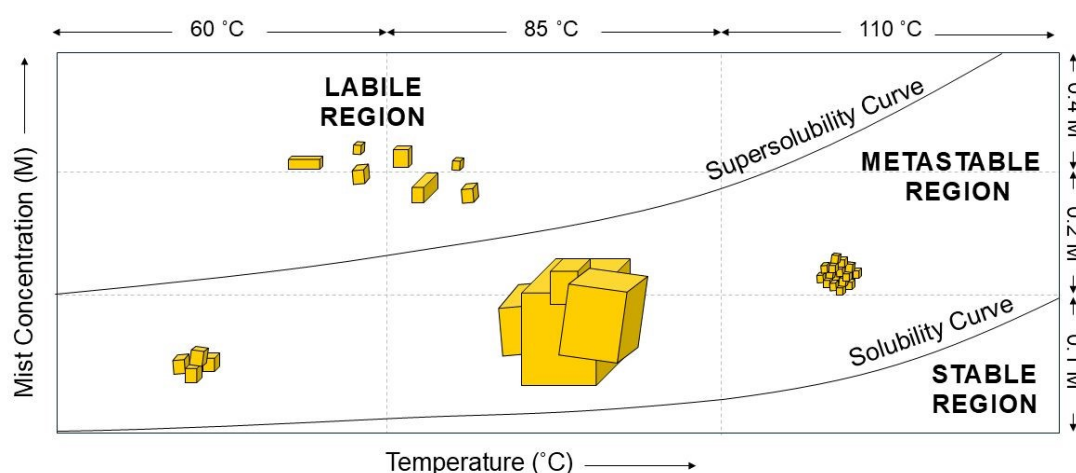


Figure 4: Schematic showing grain growth and crystal growth as a function of mist concentration and annealing temperature in CsPbBr₃ films as an adaptation of classic crystallization models.^{23,24} Optimal grain growth occurs at moderate temperature (85 °C) and low concentration (0.1 M) within the metastable region. Higher concentrations promotes rapid crystallization, yielding smaller grains with larger crystallite sizes.

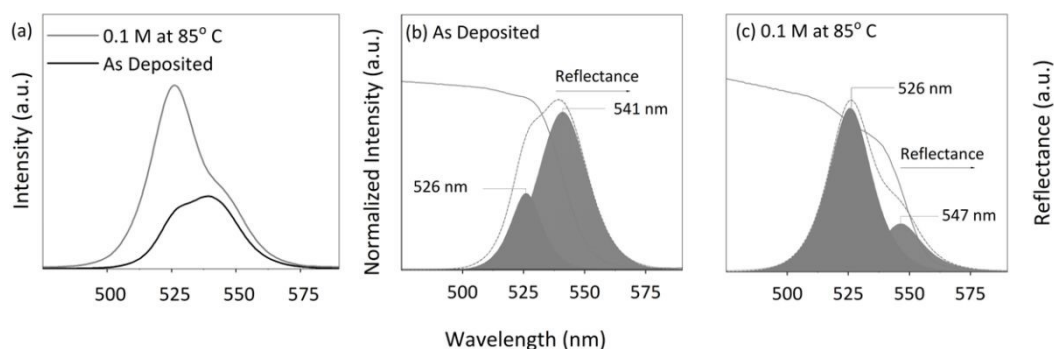


The combined SEM and XRD results are interpreted within a thermodynamic-kinetic model for crystallization. At low concentrations, the system lies within the metastable region where abundant uncoordinated DMSO molecules promote coarsening through Ostwald ripening, resulting in larger grain sizes but minimal change in crystallite size.^{21,23,26} Increasing concentration shifts the system toward the labile region, where reduced DMSO availability limits solvent-induced ripening, and unstable supersaturation promotes spontaneous crystallization, producing smaller grains with larger crystallite size.^{21,23,31} Annealing temperature further modulates these effects as DMSO with a boiling point of 189 °C, exhibits slow evaporation kinetics at 60 °C that progressively increase up to 110 °C. Therefore, SEM grain size peaks at moderate temperature 85 °C where thermal energy is sufficient for grain coarsening, while XRD crystallite size increases with temperature as faster solvent evaporation at 110 °C favors crystalline ordering and lacks the solvent availability for grain growth (Figure S4). The resulting microstructure is therefore governed by the coupled effects of supersaturation and temperature, as illustrated in Figure 4.

Impact of Grain Growth and Crystallite Size on Optoelectronic Properties

The recrystallized film with the largest grains and lowest porosity (0.1 M, 85 °C) was selected, as grain boundaries and pores act as primary barriers to charge transport in polycrystalline films.^{16,29} This film was compared to the as deposited film and evaluated using steady-state photoluminescence (PL). As shown in Figure 5, two emission bands appear: a primary CsPbBr₃ band at 525 nm and a secondary band near 540 nm, attributed to defect-related emission in CsPbBr₃.¹⁰ Band-fitting results are shown in Figures 5(b) and (c). The PL spectra were fitted with a mixed Gaussian-Lorentzian function in OriginPro 2025, yielding reduced chi-squared values $<1 \times 10^{-5}$ (see Table S2). The recrystallized film exhibits reduced relative intensity in the 540 nm defect-related band and enhanced emission at 525 nm, indicating that MADR treatment significantly reduces defect states. Furthermore, the PL emission of the 0.2 M film deposited at 60 °C, which contains secondary phases (CsPb₂Br₅ and Cs₄PbBr₆) as identified by XRD, is blue-shifted relative to the 0.2 M films deposited at 85 °C and 110 °C, which consist of phase-pure CsPbBr₃, consistent with prior reports of blue-shifted emission in Cs-Pb-Br systems containing Cs₄PbBr₆ secondary phases (see Figure S5).³⁰

Figure 5: (a) Photoluminescence Spectroscopy of the as-deposited film and recrystallized film saturated with a 0.1 M solution and annealed at 85 °C. Figures (b -c) show the fitting of the PL spectra.



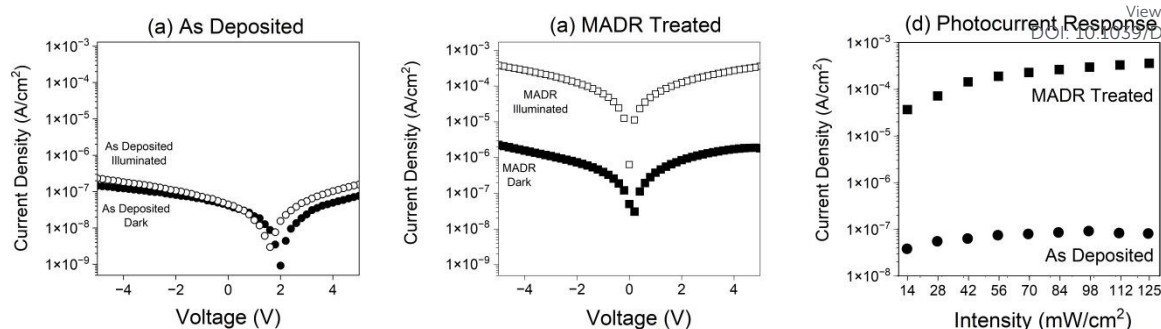
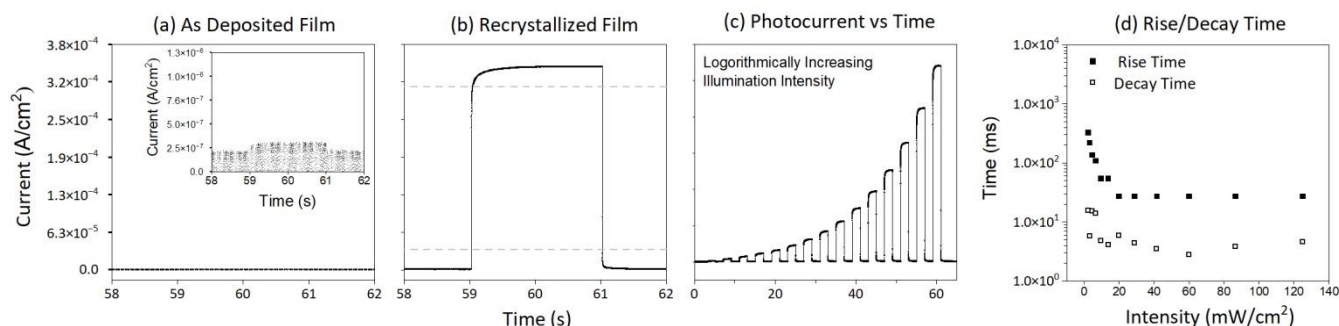


Figure 6: IV characterization of (a) the as-deposited film compared to (b) the film recrystallized with a 0.1 M solution at 85°C under dark conditions and illuminated at 125 mW/cm² and (c) photocurrent response of the as-deposited film and the recrystallized film across illumination intensities at 5 V.

The photocurrent response for the planar Pt/CsPbBr₃/Pt device fabricated from the as-deposited film and selected recrystallized film was evaluated using a 365 nm light source (from dark to 125 mW/cm², (Figure 6a-b)). The recrystallized film shows high photocurrent, consistent with reduced defect density from steady state PL. MADR-induced grain coarsening reduces grain boundary density and passivates trap states, eliminating asymmetry observed in the as-deposited film.^{31,32} Figure 6(c) shows the photocurrent response at 5 V as a function of illumination intensity. At 5 V bias, the as deposited film exhibits a photocurrent density of 1.02×10^{-7} A/cm², whereas the recrystallized film shows a 440 fold enhancement to 4.5×10^{-4} A/cm² (Figure 5c). The sheet conductance of the as-deposited film was determined to be 1.42×10^{-11} S, increasing to 2.72×10^{-10} S following MADR treatment, indirectly indicating enhanced charge transport in the recrystallized film (see Section S2).

Current-time measurements under a 125 mW cm⁻² illumination intensity results in a photocurrent response of 9.3×10^{-8} A/cm² for the as-deposited film, while the recrystallized film reaches 3.85×10^{-4} A/cm². The recrystallized film also exhibits rise and decay times of <27.4 ms and <4.6 ms respectively (Figure 7a-b). As shown in Figure 7(c) and (d), both photocurrent and response speed improve with increasing illumination intensity (from 2.2 to 125 mW/cm²). Higher light intensity generates more electron-hole pairs and progressively fills trap states, reducing the density of available trapping sites that would otherwise delay charge

Figure 7: Time-current response of (a) the as-deposited film and (b) the film recrystallized with a 0.1 M solution at 85°C. (c) The time-current response of the recrystallized film was measured with logarithmically spaced illumination intensity from 0.03 mW/cm² to 125



mW/cm². (d) Rise and decay times of the recrystallized sample across logarithmically spaced illumination intensities from 2.2 mW/cm² to 125 mW/cm². Measurements were made under 5 V bias with a 365 nm light source.

collection.³³

The as-deposited film exhibited a responsivity of 1.07×10^{-5} A/W and a specific detectivity of 3.58×10^6 Jones. Following MADR treatment, the responsivity and detectivity increased to 3.59×10^{-3} A/W and 5.04×10^9 Jones, respectively, demonstrating a significant enhancement in photodetector performance upon recrystallization (see Section S3). To assess ambient stability, the MADR treated film was stored unencapsulated between 20–30°C and 45–65% RH for 6 months, after which it retained a sustained photocurrent response exceeding 10 continuous minutes, demonstrating reasonable long-term operational stability (see Figure S6).

Hot-pressed CsPbBr₃ pellets fabricated from mechanochemically synthesized ball-milled powder exhibit a comparable grain size (~0.35 μm, converted from the reported grain area) and a higher absolute photocurrent response (~10⁻⁷ A) relative to the as-deposited film in this work (0.83 ± 0.28 μm and 9×10^{-11} A).³ MADR treatment induces a pronounced increase in photocurrent from the low response of the as deposited film (9×10^{-11} A, corresponding to 1.02×10^{-7} A/cm²) to a photocurrent density of 4.5×10^{-4} A/cm² under identical measurement conditions. Solution-grown polycrystalline films, such as those reported by Yang et al., show higher photocurrent densities (1.2×10^{-2} A/cm², calculated from the reported data), indicating the upper performance limits achievable with highly crystalline thick films.¹⁰

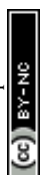


Table 1: CsPbBr₃ thick films are compared by fabrication method and their properties (grain size, thickness, processing temperature, and photocurrent response).

Fabrication Method	Average Grain Size (μm)	Thickness (μm)	Processing Temperature (°C)	Photocurrent (A)	Photocurrent Density (A/cm ²)	Ref.
Mechanochemical and Hot Pressing	0.35	400-700	100	~10 ⁻⁸	-	3
Blade Coated	0.83 ± 0.28	~ 200	85	1.02 × 10 ⁻¹¹	1.02 × 10 ⁻⁷	This work
Dissolution-Recrystallization	3.19 ± 1.57	~ 200	85	3.75 × 10 ⁻⁷	4.5 × 10 ⁻⁴	This work
Solution Grown Microcrystals	-	~ 200	140	6.0 × 10 ⁻³	1.2 × 10 ⁻²	10

MADR-treated blade-coated films nonetheless exhibit competitive photocurrent response, with further improvements anticipated through iterative dissolution-recrystallization cycles. When considered alongside scalability, low thermal budget, and broad substrate compatibility, this approach represents a feasible route to large-scale perovskite thick film fabrication for optoelectronic applications. A comparison of these fabrication techniques are summarized in **Table 1**.

Experimental

The schematic of the MADR method for bulk recrystallization of blade-coated CsPbBr₃ films is shown in **Figure 8**. In this method, the CsBr and PbBr₂ precursors are ball-milled to synthesize the CsPbBr₃ powder. The CsPbBr₃ powder is then used to prepare a slurry to deposit a 200 μm thick film onto a glass substrate using a conventional Dr. Blade method. After film deposition, the CsPbBr₃ films are partially dissolved using a CsPbBr₃/DMSO mist solution at concentrations of 0.1, 0.2 and 0.4 M and then annealed at either 60, 85 or 110 °C for 30 minutes. Platinum contacts were used for the fabrication of the devices used for optoelectronic characterization.

Materials

Cesium bromide (CsBr, 99.9%) was obtained from Alfa-Aesar and Lead (II) bromide (PbBr₂, 99.99%) and dimethyl sulfoxide (DMSO) from Sigma-Aldrich. For the ball milling process, stainless-steel balls (2 mm), milling jars, and a high-energy vertical planetary ball mill were acquired from MSE Supplies LLC.

Blade Coated Film

Glass substrates underwent sequential 10-minute sonication steps in acetone, isopropyl alcohol, deionized water, and methanol. After drying with nitrogen, the substrates were annealed at 115 °C for 2 minutes and cleaned with oxygen-based reactive ion etching (RIE) for 10 minutes to remove organic residues and moisture. CsPbBr₃ films were then deposited on the glass by blade coating. The CsPbBr₃ powder precursors were synthesized via mechanochemical ball milling following a previously reported method.³⁴ The resulting CsPbBr₃ powder was sieved through a 0.2 μm pore-sized mesh to prevent agglomerated particles in the blade coating slurry. To prepare the slurry for blade coating, 1.5 mg of ethyl cellulose was dissolved in a mixture of 0.3 mL of n-n dodecane and 0.7 mL of α-terpineol by sonicating for 20 minutes. The slurry is then prepared using 30 μL of the prepared solution per 100 mg of CsPbBr₃ ball milled

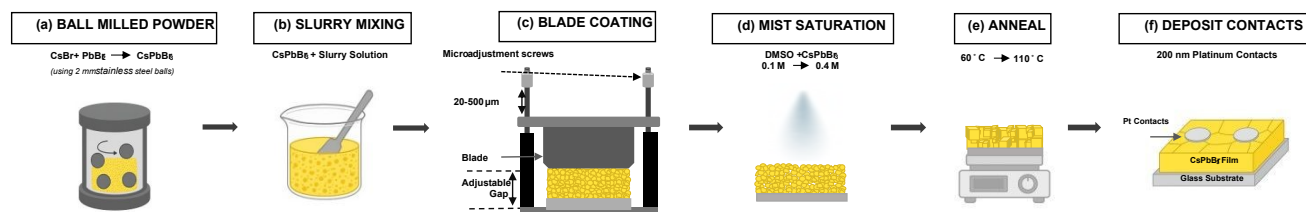


Figure 8. Experimental setup for: (a) ball milling of perovskite powder, (b) slurry preparation, (c) film deposition by blade coating, (d) film treatment with a mist solution, (e) annealing to facilitate recrystallization, and (f) deposition of Platinum contacts (E-beam evaporation).



1 powder. This slurry was then used to blade-coat CsPbBr₃ films onto 53
 2 a glass substrate at 2 mm/s at room temperature (Figure 8c). The 54
 3 blade coated films were annealed at 85 °C for 10 minutes with a 55
 4 heating ramp of 1 °C/min. 56

5 57
 6 58

6 Mist-Assisted Dissolution-Recrystallization (MADR)

7 Next, the film was exposed to a mist solution for dissolution- 59
 8 recrystallization. The mist solution was prepared by dissolving 60
 9 ball-milled CsPbBr₃ powder in Dimethyl sulfoxide (DMSO) at 0.1, 61
 10 0.2, and 0.4 M, followed by stirring for 18 hours at room 62
 11 temperature. Then 5 mL of the mist solution was applied to 200 63
 12 μm thick films through a 0.5 mm nozzle propelled by nitrogen at 64
 13 50 psi with a sample-to-nozzle distance of 15 inches, as shown 65
 14 **Figure 1d**. Then annealed for 30 minutes at either 60, 85, or 160 66
 15 °C using a ramp rate of 1 °C/min (**Figure 8e**). 67

17 Device Fabrication

18 Devices were fabricated by depositing platinum contacts (7 × 10⁻⁴ 67
 19 cm², 2 × 10⁻² cm apart with an active area of 2 × 10⁻⁴ cm²) onto the 68
 20 surface of CsPbBr₃ thick films via e-beam physical vapor 69
 21 deposition to fabricate a planar Pt/CsPbBr₃/Pt architecture. 70

22 Material and Device Characterizations

23 Scanning electron microscopy (SEM) images of the polycrystalline 71
 24 blade coated thick films were obtained using a Zeiss Sigma 72
 25 500VP. Cross-sectional images were acquired by cleaving the 73
 26 glass substrate supported films. The average grain size was 74
 27 measured using ImageJ software.³⁵ X-ray diffraction (XRD) 75
 28 patterns were obtained using a Rigaku SmartLab X-ray diffraction 76
 29 system equipped with a copper CuKα radiation source (λ = 1.5406 77
 30 Å) at a current of 30 mA and voltage of 40 kV and crystallite size 78
 31 was calculated using the Scherer equation.²⁷ UV-Vis 79
 32 measurements were performed using an Agilent UV-Vis-NIR 80
 33 spectrophotometer with a Mercury arc lamp and multiangle 81
 34 reflection (R) capability. Steady state photoluminescence (PL) was 82
 35 measured with a 405 nm light emitting diode (LED), a 0.8 mm 83
 36 aperture, and an output of 100 mW/cm². Photocurrent response 84
 37 was measured using a 365 nm LED modulated from dark to 125 85
 38 mW/cm². Current-Voltage characteristics of the planar 86
 39 Pt/CsPbBr₃/Pt device were measured in a probe station with a 87
 40 Keithley 4200 system. 88
 41 89
 42 90
 43 91
 44 92
 45 93
 46 94
 47 95

42 Conclusion

43 This study demonstrates a mist-assisted dissolution- 84
 44 recrystallization (MADR) process for scalable control 85
 45 microstructure in blade-coated CsPbBr₃ thick films. At low mist 86
 46 concentrations, partial dissolution promotes grain coarsening 87
 47 through Ostwald ripening, yielding larger grains. High mist 88
 48 concentrations reduce dissolution, resulting in smaller grains. 89
 49 Annealing temperature modulates these effects: insufficient 90
 50 heating limits ripening, while excessive heating accelerates 91
 51 solvent evaporation and quenches grain growth. Careful control 92
 52 of mist concentration and annealing temperature enables precise 93
 94
 95

grain and crystallite size tuning, significantly improving photo 96
 response. This approach offers a pathway for engineering high- 97
 quality perovskite thick films for X-ray detection, photodetectors, 98
 and other applications requiring high-performance, large-area 99
 devices. 100

Author contributions

Monet Brown: Conceptualization, Data curation, Formal 101
 analysis, Investigation, Methodology, Visualization, Writing- 102
 original draft, Writing-review and editing 103

Susana Borbon-Rojas: Methodology, Writing -review and 104
 editing 105

Leunam Fernandez-Izquierdo: Validation, Writing -review 106
 and editing 107

Ross Haroldson: Data curation 108

Manuel Quevedo-Lopez: Conceptualization, Funding 109
 acquisition, Project administration, Resources, Supervision, 110
 Validation, Writing-review and editing 111

Conflicts of interest

There are no conflicts to declare. 112

Data availability

The data supporting this article have been included as part of 113
 the Supplementary Information. Supplementary information: 114
 Figure S1, Tables S1, Figure S2, Figure S3, Table S2 and Figure S4 115
 and further experimental details. See DOI: [URL – format 116
<https://doi.org/DOI>] 117

Acknowledgements

The authors would like to express their gratitude to the 118
 University of Texas at Dallas and The Texas Instruments Chair in 119
 Nanoelectronics for partial financial support. 120

References

- 1 Y. Hua, X. Sun, X. Li, F. Cui, Z. Yue, J. Liu, H. Liu, G. Zhang and X. 121
 Tao, *J. Mater. Chem. C*, 2023, **11**, 9153–9160. 122
- 2 N. Grundmanis, A. Sarakovskis, A. Lupilov, V. Gostilo, A. Owens 123
 and K. Pudzs, *Nuclear Instruments and Methods in Physics 124
 Research Section A: Accelerators, Spectrometers, Detectors and 125
 Associated Equipment*, 2025, **1073**, 170305. 126
- 3 M. Sotelo-Lerma, L. Fernandez-Izquierdo, M. A. Ruiz-Molina, I. 127
 Borges-Doren, R. Haroldson and M. Quevedo-Lopez, *Materials*, 128
 DOI:10.3390/ma17205123. 129
- 4 Y. Haruta, M. Kawakami, Y. Nakano, S. Kundu, S. Wada, T. 130
 Ikenoue, M. Miyake, T. Hirato and M. I. Saidaminov, *Chem. 131
 Mater.*, 2022, **34**, 5323–5333. 132



- 1 5 J. Peng, C. Q. Xia, Y. Xu, R. Li, L. Cui, J. K. Clegg, L. M. Herz, M. B43
 2 Johnston and Q. Lin, *Nature Communications*, 2021, **12**, 1531. 44
 3 6 J. Cheng, C. Xue, M. Yang, X. Wang, Z. Xu, N. Li, X. Zhang, X. 45
 4 Feng, X. Liu, Y. Liu, S. F. Liu and Z. Yang, *ACS Appl. Mater. 46*
 5 *Interfaces*, 2024, **16**, 36649–36657. 47
 6 7 T. Zou, B. Xiang, Y. Xu, Y. Wang, C. Liu, J. Chen, K. Wang, Q. Dai, 48
 7 S. Zhang, Y. -Y. Noh, and H. Zhou, *IEEE Journal of the Electron 49*
 8 *Devices Society*, 2021, **9**, 96–101. 50
 9 8 S. V. N. Pammi, V.-D. Tran, R. Maddaka, J.-H. Eom, J. S. Jung, H. 51
 10 M. Jeong, M.-D. Kim, V. Pecunia and S. G. Yoon, *Advanced 52*
 11 *Optical Materials*, 2020, **8**, 2000845. 53
 12 9 S. V. N. Pammi, H.-W. Lee, J.-H. Eom and S.-G. Yoon, *ACS Appl. 54*
 13 *Energy Mater.*, 2018, **1**, 3301–3312. 55
 14 10 B. Yang, F. Zhang, J. Chen, S. Yang, X. Xia, T. Pullerits, W. Deng 56
 15 and K. Han, *Advanced Materials*, 2017, **29**, 1703758. 57
 16 11 Z. Zhang, H. Li, H. Di, D. Liu, W. Jiang, J. Ren, Z. Fan, F. Liao, L. 58
 17 G. Li, Y. Xiong and Y. Zhao, *ACS Appl. Electron. Mater.*, 2023, **5**, 59
 18 388–396. 60
 19 12 K.-W. Huang, M.-H. Li, Y.-T. Chen, Z.-X. Wen, C.-F. Lin and P. 61
 20 Chen, *J. Mater. Chem. C*, 2024, **12**, 1533–1542. 62
 21 13 M. Xia, Z. Song, H. Wu, X. Du, X. He, J. Pang, H. Luo, L. Jin, G. Li, 63
 22 G. Niu and J. Tang, *Advanced Functional Materials*, 2022, **32**, 64
 23 2110729. 65
 24 14 B. Yang, W. Pan, H. Wu, G. Niu, J.-H. Yuan, K.-H. Xue, L. Yin, X. 66
 25 Du, X.-S. Miao, X. Yang, Q. Xie and J. Tang, *Nature 67*
 26 *Communications*, 2019, **10**, 1989. 68
 27 15 H. Wei, Y. Fang, P. Mulligan, W. Chuirazzi, H.-H. Fang, C. Wang, 69
 28 B. R. Ecker, Y. Gao, M. A. Loi, L. Cao and J. Huang, *Nature 70*
 29 *Photonics*, 2016, **10**, 333–339. 71
 30 16 Y. C. Kim, K. H. Kim, D.-Y. Son, D.-N. Jeong, J.-Y. Seo, Y. S. Choi, 72
 31 T. Han, S. Y. Lee and N.-G. Park, *Nature*, 2017, **550**, 87–91. 73
 32 17 Z. Gou, S. Huanglong, W. Ke, H. Sun, H. Tian, X. Gao, X. Zhu, D. 74
 33 Yang and P. Wangyang, *physica status solidi (RRL) – Rapid 75*
 34 *Research Letters*, 2019, **13**, 1900094. 76
 35 18 A. Choubey, N. Perumal, S. Narendhiran, S. P. Muthu and R. 77
 36 Perumalsamy, *International Journal of Energy Research*, 2022, **78**
 37 **46**, 9310–9322. 79
 38 19 T. Shi, W. Liu, J. Zhu, X. Fan, Z. Zhang, X. He, R. He, J. Wang, K. 80
 39 Chen, Y. Ge, X. Sun, Y. Liu, P. K. Chu and X.-F. Yu, *Nano Research*, 81
 40 2023, **16**, 9983–9989. 82
 41 20 X. Cao, L. Zhi, Y. Li, F. Fang, X. Cui, L. Ci, K. Ding and J. Wei, *ACS*
 42 *Appl. Energy Mater.*, 2018, **1**, 868–875.
- 21 T. Vetter, M. Igglund, D. R. Ochsenbein, F. S. Hänseler and M. 43
 Mazzotti, *Crystal Growth & Design*, 2013, **13**, 4890–4905. 44
 22 X. Cao, G. Zhang, L. Hao, X. Ding, T. Dong, X. Li, Q. Zeng, X. He, Y. 45
 Jia and J. Wei, *Journal of Alloys and Compounds*, 2022, **919**, 46
 165722. 47
 23 Y. Zhang, in *Modelling of Chemical Process Systems*, ed. S. A. 48
 Imtiaz, Elsevier, 2023, pp. 181–202. 49
 24 J.-P. Duroudier, in *Crystallization and Crystallizers*, ed. J.-P. 50
 Duroudier, Elsevier, 2016, pp. 39–78. 51
 25 S. Kim and A. S. Myerson, *Ind. Eng. Chem. Res.*, 1996, **35**, 1078– 52
 1084. 53
 26 M. A. Behrens, A. Franzén, S. Carlert, U. Skantze, L. Lindfors and 54
 U. Olsson, *Soft Matter*, 2025, **21**, 2349–2354. 55
 27 P. Scherrer, *Math. Phys.*, 1918, **2**, 98–100. 56
 28 S. Li, Y. Xiao, R. Su, W. Xu, D. Luo, P. Huang, L. Dai, P. Chen, P. 57
 Caprioglio, K. A. Elmostekawy, M. Dubajic, C. Chosy, J. Hu, I. 58
 Habib, A. Dasgupta, D. Guo, Y. Boeije, S. J. Zelewski, Z. Lu, T. 59
 Huang, Q. Li, J. Wang, H. Yan, H.-H. Chen, C. Li, B. A. I. Lewis, D. 60
 Wang, J. Wu, L. Zhao, B. Han, J. Wang, L. M. Herz, J. R. Durrant, 61
 K. S. Novoselov, Z.-H. Lu, Q. Gong, S. D. Stranks, H. J. Snaith and 62
 R. Zhu, *Nature*, 2024, **635**, 874–881. 63
 29 T. Meier, H. Bässler and A. Köhler, *Advanced Optical Materials*, 64
 2021, **9**, 2100115. 65
 30 J. Li, H. Zhang, S. Wang, D. Long, M. Li, D. Wang and T. Zhang, 66
Materials, 2018, **11**, 717. 67
 31 W. Xu, T. Du, M. Sachs, T. J. Macdonald, G. Min, L. Mohan, K. 68
 Stewart, C.-T. Lin, J. Wu, R. Pacalaj, S. A. Haque, M. A. McLachlan 69
 and J. R. Durrant, *Cell Reports Physical Science*, 2022, **3**, 100890. 70
 32 A. B. Andrabi and A. T. Mallajosyula, *Solar Energy*, 2023, **264**, 71
 112020. 72
 33 J. Pan, Z. Chen, T. Zhang, B. Hu, H. Ning, Z. Meng, Z. Su, D. 73
 Nodari, W. Xu, G. Min, M. Chen, X. Liu, N. Gasparini, S. A. Haque, 74
 P. R. F. Barnes, F. Gao and A. A. Bakulin, *Nature 75*
Communications, 2023, **14**, 8000. 76
 34 M. Shekarnoush, L. Fernandez-Izquierdo, F. S. Aguirre-Tostado, 77
 Z. H. Shamsi and M. A. Quevedo-Lopez, *Chem. Mater.*, 78
 DOI:10.1021/acs.chemmater.3c01440. 79
 35 R. Wayne, Image J (version 1.54p) National Institutes of Health 80
 and the Laboratory for Optical and Computational 81
 Instrumentation 2025. 82

The data supporting this article have been included as part of the Supplementary Information. Supplementary information: Figure S1, Tables S1, Section S1, Figure S2, Figure S3, Figure S4, Table S2, Figure S5, Section S2, Section S3 and Figure S6. Further data supporting the findings of this study are available from the corresponding author upon reasonable request.

[View Article Online](#)

[DOI: 10.1039/D6MA00018K](#)

