

Spin-Coating-Speed-Dependent Structural and Optical Properties of Open-Air-Processed MAPbI₃ Thin Films: Experimentally Informed SCAPS-1D Device Simulation

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Abstract

Spin coating remains one of the most accessible routes for depositing metal-halide perovskite absorbers, yet the strong coupling between rotational speed, solvent removal, crystallization and film properties complicates process optimization under ambient conditions. This study investigates the influence of spin-coating speed (2000–5000 rpm) on the structural and optical properties of open-air-processed methylammonium lead iodide (MAPbI₃) thin films and uses the experimentally determined optical bandgaps to inform one-dimensional device simulation. Films were deposited from a 1.25 M MAPbI₃ precursor using ethyl acetate antisolvent treatment and annealed at 110 °C for 10 min under controlled cleanroom conditions (24.4 °C and 41.5% relative humidity). X-ray diffraction confirmed tetragonal MAPbI₃ with preferential (110) orientation and no detectable PbI₂ reflection. The response to spin speed was non-monotonic: the film deposited at 3000 rpm exhibited the narrowest (110) peak (0.352°), largest crystallite size (22.73 nm), lowest microstrain (1.257×10^{-2}) and lowest dislocation density (1.936×10^{-3} linesnm⁻²), indicating the best crystalline quality. UV–visible analysis yielded optical bandgaps of 1.59–1.60 eV. When these

bandgaps were incorporated into an FTO/TiO₂/MAPbI₃/Spiro-OMeTAD/carbon SCAPS-1D model, the 1.59 eV absorber produced a prototype efficiency of 16.67%, compared with 16.47% for the 1.60 eV absorber. Subsequent optimization of layer thicknesses, carrier densities, bulk and interface defect densities, and resistive parameters increased the predicted power-conversion efficiency to 25.38%, with an open-circuit voltage of 1.25 V, short-circuit current density of 24.24 mA/cm² and fill factor of 83.61%. The results identify 3000 rpm as the most favourable condition for crystalline film quality and demonstrate how experimentally constrained optical data can improve the physical relevance of device-level optimization.

Keywords:

MAPbI₃ thin films; spin-coating speed; ambient-air processing; crystallization; optical bandgap; SCAPS-1D; perovskite solar cells

Introduction

Metal-halide perovskite solar cells have developed rapidly from the first photovoltaic use of organolead halide perovskites to a versatile thin-film technology with exceptional optoelectronic performance [1–3]. Their appeal arises from strong visible-light absorption, a direct and compositionally tunable bandgap, long carrier-diffusion lengths, low exciton-binding energy and compatibility with low-temperature solution processing [4–6]. These attributes offer a pathway to lightweight and potentially low-cost photovoltaics. Nevertheless, the translation of high material quality into reproducible device performance remains limited by the sensitivity of the perovskite layer to precursor chemistry, solvent coordination, crystallization kinetics, environmental conditions and interfaces [3,6,7].

Methylammonium lead iodide (CH₃NH₃PbI₃, MAPbI₃) remains an important model absorber for studying process–structure–property relationships in lead-halide perovskites. It normally exhibits a direct optical bandgap near 1.55–1.60 eV, a sharp absorption edge and strong absorption throughout much of the visible spectrum [5,8]. Although more compositionally complex perovskites have achieved greater operational stability, MAPbI₃ offers comparatively simple precursor stoichiometry and well-established structural and optical signatures. It is therefore

particularly useful for isolating how a deposition variable modifies crystallinity, defect formation, optical response and the photovoltaic quantities predicted from those properties.

Spin coating is widely employed for laboratory-scale perovskite deposition because it enables rapid control of solution spreading, wet-film thinning and solvent evaporation. During rotation, centrifugal flow, viscous resistance and solvent loss collectively determine the final thickness and uniformity of the coating; film thickness commonly decreases with increasing angular velocity according to a process-dependent power-law relationship [9]. In perovskite precursor films, however, rotational speed does more than change thickness. It also changes the residence time of coordinating solvents, precursor supersaturation, nucleation density and the interval available for crystal growth. A speed that is too low may leave a thick wet layer that dries non-uniformly, whereas excessive speed can remove solution and solvent so rapidly that crystal coalescence is restricted. An intermediate speed can consequently provide the best compromise between coverage, nucleation and grain development.

Solvent and antisolvent engineering further govern this kinetic competition. Coordinating solvents such as dimethyl sulfoxide can form intermediate adducts with lead-halide precursors, slowing uncontrolled crystallization, while timely antisolvent application rapidly extracts the host solvent and drives the wet film towards supersaturation [10,11]. The identity, volume, timing and delivery position of the antisolvent must therefore be controlled alongside spin speed. Recent ambient-processing studies have confirmed that antisolvent selection can substantially change perovskite phase formation, surface coverage and optical response [12], and that single-step MAPbI₃ deposition in air requires coordinated optimization of precursor temperature, solvent composition, spin parameters and annealing conditions [13].

Ambient fabrication adds another important source of variability. Water vapour can alter precursor coordination, intermediate-phase formation, nucleation and annealing behaviour, with the outcome depending on humidity level and the specific processing route [7,14]. Reproducible open-air studies should therefore report temperature and relative humidity rather than treating the surrounding atmosphere as an unspecified background condition. Establishing a process window under measured ambient conditions is particularly important for laboratories and future manufacturing environments where continuous inert-atmosphere control may be technically or economically restrictive.

The structural consequences of deposition can be evaluated from X-ray diffraction through phase identity, preferred orientation, peak width and derived microstructural parameters. For tetragonal MAPbI₃, the intense reflections near 14° and 28° are commonly associated with the (110) and (220) planes, whereas a reflection near 12.6° can indicate residual PbI₂. Peak broadening may be used to estimate coherent crystallite size, although strain and instrumental broadening should be considered when interpreting Scherrer-derived values. Optical spectroscopy provides complementary information because deposition-induced differences in thickness, coverage, scattering and structural disorder modify absorbance and the apparent absorption edge. Accurate bandgap comparison consequently requires consistent baseline correction and use of the measured thickness when calculating the absorption coefficient [5,8].

Experimental characterization alone does not directly reveal how a small change in bandgap or defect-related material quality propagates through a complete photovoltaic stack. SCAPS-1D addresses this limitation by solving Poisson's equation together with carrier-continuity and drift-diffusion equations for multilayer thin-film devices [15,16]. It permits independent examination of absorber thickness, band alignment, doping, carrier mobility, bulk and interface defect densities, contact properties and parasitic resistances. Studies have combined experimental MAPbI₃ data with SCAPS-1D modelling to connect measured absorber properties with prospective device performance [17]. However, simulated efficiencies should be interpreted as model-dependent projections rather than experimental device records, particularly when parameters are optimized independently of demonstrated processing constraints.

Many reports optimize either MAPbI₃ film preparation or numerical device parameters, but fewer establish a direct experimental-to-simulation workflow in which a deliberately varied deposition parameter is linked to measured structural and optical responses and then introduced into device modelling. Moreover, apparently simple trends with spin speed may conceal a non-monotonic crystallization response. This makes it necessary to distinguish the experimentally observed effect of deposition speed from subsequent numerical sensitivity analyses in which other layer and device parameters are varied.

Accordingly, this study investigates MAPbI₃ films deposited in open air at 2000, 3000, 4000 and 5000 rpm while holding precursor concentration and volume, acceleration, coating duration, ethyl acetate antisolvent treatment, annealing conditions and the measured cleanroom environment

constant. The films are compared using X-ray diffraction and UV–visible spectroscopy to establish their phase, crystallite-related parameters and optical bandgaps. The experimentally obtained bandgaps are subsequently incorporated into a planar FTO/TiO₂/MAPbI₃/Spiro-OMeTAD/carbon SCAPS-1D model. Parametric analyses of layer thickness, carrier density, bulk and interface defects, series and shunt resistance, and temperature are then used to identify the dominant performance limitations and a physically interpretable route towards an optimized device.

2.0 Materials synthesis and Method

2.1 ITO Substrate Preparation and Cleaning

Patterned indium tin oxide (ITO)-coated glass substrates for photovoltaic and organic light-emitting diode applications were obtained from Ossila Ltd. (product code S2006C1; batch number: 250428-01F). Each substrate measured 25 mm × 25 mm, nominal sheet resistance of 20 Ω/sq and had a glass thickness of 1.1 mm. Prior to the deposition of the perovskite layer, the substrates were sequentially cleaned using a Digital Pro+ digital ultrasonic cleaner equipped with independently adjustable bath-temperature and sonication-time controls. The substrates were initially ultrasonicated for 15 min in a 1:4 (v/v) Hellmanex™ III/deionized-water solution prepared using Hellmanex™ III critical-cleaning concentrate (Ossila Ltd.; product code C141-25mL). The deionized water used throughout the cleaning process had an electrical conductivity of 0.999 $\mu S\ cm^{-1}$, an electrical resistivity of 0.52 $M\Omega\ cm$, and a total dissolved solids concentration of 0.499 ppm, as indicated by the integrated conductivity/resistivity meter of the BIOBASE water-purification system. Following this treatment, the substrates were thoroughly rinsed with fresh deionized water to remove residual alkaline cleaning agents. The substrates were subsequently ultrasonicated sequentially in deionized water, acetone (BDH; GPR grade; minimum assay 99.5 %) and isopropyl alcohol, also referred to as propan-2-ol (BDH; GPR grade; minimum assay 99.5 %), for 10 min in each cleaning medium. All ultrasonication steps were performed at 50 °C, with thorough rinsing using fresh portions of the corresponding cleaning liquid between successive stages. This procedure facilitated the removal of dust particles, grease, organic residues and other surface contaminants from the ITO surface. Following solvent cleaning, the substrates were dried under a stream of high-purity nitrogen gas (N₂, 99.99 % purity) to remove residual solvent and loosely bound surface particles. They were subsequently treated in an Ossila UV–

ozone cleaner for 10 *min* as the final surface-cleaning step. The UV–ozone treatment removed residual organic contaminants and increased the surface energy and wettability of the ITO, thereby promoting improved interfacial contact and more uniform deposition of the subsequent functional layers. The cleaned substrates were used immediately for thin-film deposition to minimize surface recontamination.

2.2 Preparation of MAPbI₃ Perovskite Precursor Solution

A 1.25 *M* MAPbI₃ precursor solution was prepared by dissolving 0.864 *g* (1.874 *mmol*) of lead(II) iodide (PbI₂; Sigma-Aldrich; 99 % purity, corresponding to a maximum nominal impurity content of 1 %; product no. 211168) and 0.298 *g* (1.875 *mmol*) of methylammonium iodide (MAI: recrystallised, >99.5 % purity; Ossila Ltd.; product code M271-5g) in 1.2 *mL* of N,N-dimethylformamide (DMF; Molychem India LLP; LR grade; minimum assay 99 %, maximum nominal impurity content 1 %; product code 14185) and 0.3 *mL* of dimethyl sulfoxide (DMSO; Molychem India LLP; LR grade; minimum assay 99 %, maximum nominal impurity content 1 %; product code 14187), producing a total solvent volume of 1.5 *mL* and an approximately equimolar PbI₂:MAI ratio of 1:1. The resulting DMF:DMSO volume ratio was 4:1. The solution was magnetically stirred at 60 °C for 10 *min* to facilitate complete dissolution and homogenization. Before spin-coating deposition, the precursor solution was passed through a 0.45 µm polyvinylidene fluoride (PVDF) syringe filter to remove undissolved particles and aggregates that could cause pinholes, nonuniform nucleation and local variations in film thickness. The precursor preparation was conducted in a cleanroom under an intelligent air-purification fume hood. The recorded environmental and processing conditions were a relative humidity of 41.5 %, an ambient temperature of 24.4 °C, a fume-hood face velocity of 0.29 *m s*⁻¹ and a total volatile organic compound concentration of 1 *ppm*. The precursor mixture was magnetically stirred at 60 °C for 10 *min* to facilitate complete dissolution of the precursor salts and produce a homogeneous MAPbI₃ precursor solution suitable for subsequent spin-coating deposition. All recorded chemicals were used as received without further purification.

2.3 Spin coating and thermal annealing of MAPbI₃ films

The MAPbI₃ precursor solution was deposited onto the cleaned ITO substrates using a VTC-100PA vacuum spin coater under the same cleanroom conditions employed for precursor

preparation: 41.5 % relative humidity, 24.4 °C ambient temperature, 0.29 m s⁻¹ fume-hood face velocity and 1 ppm TVOC. For each sample, 100 µL of the precursor solution was applied to the centre of the stationary substrate using the static-dispensing method before applying the spin-coating conditions presented in Table 1. This method was selected to promote complete initial surface coverage, minimize precursor loss and improve the reproducibility of radial spreading and film formation. Following precursor application, the spin-coating programme was initiated immediately to limit uncontrolled solvent evaporation and premature crystallization. Ethyl acetate (EA, 300 µL) was employed as the antisolvent to induce rapid solvent extraction, precursor supersaturation and perovskite nucleation. The antisolvent was dispensed at the centre of the rotating substrate at $t = 15$ s after initiation of the 30 s spin-coating cycle, corresponding to the beginning of the final 15 s. It was delivered as a single continuous dispense rather than as multiple discrete drops, while the micropipette tip was maintained approximately 1 cm above the substrate. Continuous central dispensing promoted rapid and relatively uniform antisolvent distribution across the rotating wet film, minimized localized solvent-extraction gradients and avoided repeated impact disturbances that could cause splashing, film displacement, nonuniform nucleation or thickness variations. Spin speed was the deliberately varied deposition parameter; precursor formulation and volume, acceleration/deceleration, total duration, antisolvent identity, volume, timing and delivery geometry, annealing conditions and environment were held constant.

Table 1. Spin-coating conditions used for MAPbI₃ film deposition.

Sample ID	Spin speed (rpm)	Acceleration/deceleration (rpm s ⁻¹)	Duration (s)	Antisolvent treatment (µL)
S1	5000	2000	30	300
S2	4000	2000	30	300
S3	3000	2000	30	300
S4	2000	2000	30	300

Immediately after spin coating, the wet films were transferred onto a preheated hotplate and annealed at 110 °C for 10 min. Thermal annealing promoted the evaporation of residual DMF, DMSO and antisolvent, the conversion of the intermediate precursor phase into crystalline

MAPbI₃, and the development of the characteristic dark perovskite film. Following annealing, the samples were allowed to cool naturally to room temperature before storage or further characterization.

The experimental design enabled the effects of spin speed, coating sequence, antisolvent type and antisolvent application time on the resulting MAPbI₃ film thickness to be evaluated.

2.4 Optical and Structural Characterization of MAPbI₃ Thin Films

The optical absorption spectra of the MAPbI₃ thin films were measured using a LAMBDA 365+ UV–Vis spectrophotometer controlled with UV WinLab software. The spectra were recorded in absorbance mode over the wavelength range of 200–1100 nm, with the instrument scanning from 1100 to 200 nm in order to evaluate the optical absorption behaviour and estimate the optical bandgap of the films. A spectral bandwidth of 1.00 nm, scan speed of 480 nm min⁻¹ and data interval of 1 nm were employed. Each sample was measured using a single scan cycle with a cycle time of 1 s. Both the UV and visible lamps were activated, with automatic lamp changeover at 400 nm. A cleaned, uncoated ITO substrate was used as the reference blank to correct for the optical contribution of the underlying ITO-coated glass. The crystalline structure and phase composition were examined by X-ray diffraction using a benchtop X-ray diffractometer (Aeris, Malvern Panalytical, the Netherlands; Type No. 9430 070 99991).

2.5 Numerical simulation of perovskite solar cell

Numerical simulation of the perovskite solar cell was performed to investigate how the experimentally determined optical bandgap of the MAPbI₃ absorber influences the photovoltaic performance of the proposed device. The simulated cell employed the conventional n–i–p configuration:

FTO/TiO₂/MAPbI₃/Spiro-OMeTAD/Carbon

Numerical device modelling provides insight into the physical processes governing charge generation, transport, recombination and collection within the solar cell. It also enables systematic evaluation of material and device parameters that may be difficult to vary independently during experimental fabrication.

The simulations were carried out using the one-dimensional Solar Cell Capacitance Simulator, SCAPS-1D. The software was developed at Ghent University, Belgium, by Burgelman and co-workers for the numerical analysis of thin-film semiconductor devices. SCAPS-1D calculates the steady-state electrical and optoelectronic behaviour of a multilayer device by simultaneously solving Poisson's equation, the electron and hole continuity equations, and the drift–diffusion current-density equations under specified boundary and operating conditions.

The simulation procedure accounts for the thickness, bandgap, electron affinity, dielectric permittivity, effective density of states, charge-carrier mobility, doping concentration and defect density of each semiconductor layer. It can also incorporate interface defect states, bulk recombination, contact work functions, series resistance and shunt resistance. From these inputs, SCAPS-1D predicts the current density–voltage characteristics, external quantum efficiency, energy-band profile, carrier concentrations, generation and recombination rates, capacitance–voltage response and capacitance–frequency behaviour of the device [19][20][21].

The electrostatic potential within the device is governed by Poisson's equation (Eq. 1).

$$\frac{d\psi(x)}{dx^2} = -\frac{q}{\varepsilon} [p - n + N_D^+ - N_A^- + p_t - n_t], \quad 1$$

where, ψ represent the electric field, ε denote dielectric constant of the semiconductor equal to $\varepsilon = \varepsilon_r \varepsilon_0$, where ε_r is the relative permittivity and ε_0 is vacuum permittivity, q symbolize electron charge, N_A^- and N_D^+ are acceptor and donor carrier density, respectively, E is the electrostatic potential, p and n are the hole and electron concentration, n_t and p_t are the trapped electron and hole concentration, respectively.

The steady-state electron and hole continuity equations are expressed as in Eqs. 2 and 3:

$$\frac{1}{d} \frac{dJ_n}{dx} = G_n - R_n, \quad 2$$

$$\frac{1}{d} \frac{dJ_p}{dx} = G_p - R_p, \quad 3$$

where J_n and J_p are the electron and hole current densities, G_n and G_p are the electron and hole generation rates, and R_n and R_p represent the corresponding recombination rates.

The electron and hole current densities are described by the drift–diffusion relation (Eqs.4 and 5).

$$J_n = qn\mu_n E + qD_n \frac{dn}{dx}, \quad 4$$

$$J_p = qp\mu_p E - qD_p \frac{dp}{dx}, \quad 5$$

$$D_{n,p} = \left(\frac{k_B T}{q} \mu_{n,p} \right), \quad 6$$

where E is the electric field, μ_n and μ_p are the electron and hole mobilities, and D_n and D_p are the electron and hole diffusion coefficients. The diffusion coefficients are related to the carrier mobilities through the Einstein relation (Eq. 6), where k_B is the Boltzmann constant and T is the absolute temperature.

2.5.1 Device structure and operating mechanism

The simulated perovskite solar cell shown in Fig.1 employs the planar n–i–p configuration FTO/TiO₂/MAPbI₃/Spiro-OMeTAD/Carbon. FTO acts as the transparent front electrode, while TiO₂ selectively extracts electrons and blocks holes. MAPbI₃ serves as the light-absorbing layer in which electron–hole pairs are generated. Spiro-OMeTAD extracts holes and suppresses electron transfer towards the rear contact, whereas carbon collects the holes and functions as the back electrode. Under illumination, electrons move from MAPbI₃ through TiO₂ to FTO, while holes move from MAPbI₃ through Spiro-OMeTAD to carbon [22].

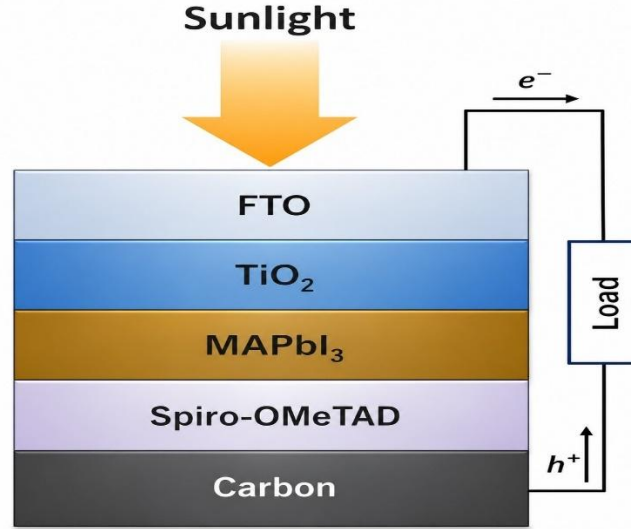


Fig. 1. Schematic diagram of the simulated PSC.

2.5.2 Basic input parameters and simulation conditions

The SCAPS-1D model was constructed using the thickness, bandgap, electron affinity, relative dielectric permittivity, effective density of states, carrier mobility, doping concentration and bulk defect density of the constituent device layers, as summarized in Table 2. The experimentally measured MAPbI₃ bandgap values of 1.59 – 1.60 eV were incorporated into the model to evaluate the influence of the deposition conditions on photovoltaic performance. Defect states were introduced at the TiO₂/MAPbI₃ and MAPbI₃/Spiro-OMeTAD interfaces to account for non-ideal interfacial recombination; the corresponding interface parameters are presented in Table 3. The work functions of the carbon and ITO contacts were set to 5.0 eV and 4.4 eV, respectively [19][21]. Simulations were performed under AM1.5G illumination at an incident power density of 100 mW cm⁻², an operating temperature of 300 K, a frequency of 1 × 10⁶ Hz, and a voltage range of 0 – 1.50 V [19][21].

Table 2. SCAPS-1D parameters for simulating the structure of the PSC [21][23][24]

Parameters	Layer			
		TiO ₂	Spiro-OMeTAD	MAPbI ₃
		[23]	[21]	[24]
Thickness (μm)	0.1	0.06	0.05	0.5
E _g (eV)	3.5	3.2	2.9	1.59, 1.60

ϵ_r	9.0	10.0	2.2	6.5
χ (eV)	4.3	4.2	3.0	3.9
N_C (cm ⁻³)	2.2×10^{18}	2.2×10^{18}	2.2×10^{18}	2.2×10^{18}
N_V (cm ⁻³)	1.8×10^{19}	1.8×10^{19}	1.8×10^{19}	1.8×10^{19}
N_A (cm ⁻³)	0	0	1×10^{18}	1×10^{16}
N_D (cm ⁻³)	1×10^{18}	1×10^{18}	0	0
μ_n (cm ² V ⁻¹ s ⁻¹)	20	20	1×10^{-4}	2
μ_p (cm ² V ⁻¹ s ⁻¹)	10	10	2×10^{-4}	2
N_t (cm ⁻³)	1×10^{15}	1×10^{15}	1×10^{15}	1×10^{14}
Defect type	Neutral	Single donor	Single acceptor	Single acceptor

Table. 3. Interface layer parameters [21][24]

Parameters	TiO ₂ /MAPbI ₃ interface	MAPbI ₃ /Spiro- OMeTAD
Defect type	Neutral	Neutral
Cross section for electrons (cm ²)	1×10^{19}	1×10^{19}
Cross section for holes (cm ²)	1×10^{19}	1×10^{19}
Energetic distribution	Single	Single
Energy level with respect to E _v (eV)	0.65	0.65
Characteristic energy (eV)	0.10	0.10
Total density (cm ⁻³)	1×10^{12}	1×10^{12}

3.0 Results and discussion

3.1 Crystal structure

Fig. 2 presents the X-ray diffraction patterns of MAPbI₃ thin films deposited under varying spin coating speed. All the samples exhibit the characteristic diffraction reflections of crystalline MAPbI₃, confirming successful formation of the perovskite phase under the investigated open-air deposition conditions. The most intense peak occurs at $2\theta \approx 13.94\text{--}14.02^\circ$ and is assigned to the (110) plane of tetragonal MAPbI₃. The persistence of this dominant reflection in all the films indicates preferential crystal growth along the (110) orientation, irrespective of the applied spin speed.

Additional MAPbI₃ reflections are observed at approximately 19.81°, 23.34°, 24.47°, 28.25°, 31.51°, 34.86°, 40.52°, 43.1° and 50.25°, corresponding approximately to the (112), (211), (202), (220), (310), (312), (224), (314) and (404) crystallographic planes, respectively. The strong (110) and (220) reflections confirm the formation of the tetragonal MAPbI₃ structure. The peak near 30.52–30.73°, particularly visible in the thinner films, is assigned mainly to the (222) reflection of the underlying ITO substrate rather than to MAPbI₃. Its visibility is likely enhanced at higher spin speeds because faster rotation produces a thinner perovskite layer, allowing the substrate contribution to become more pronounced.

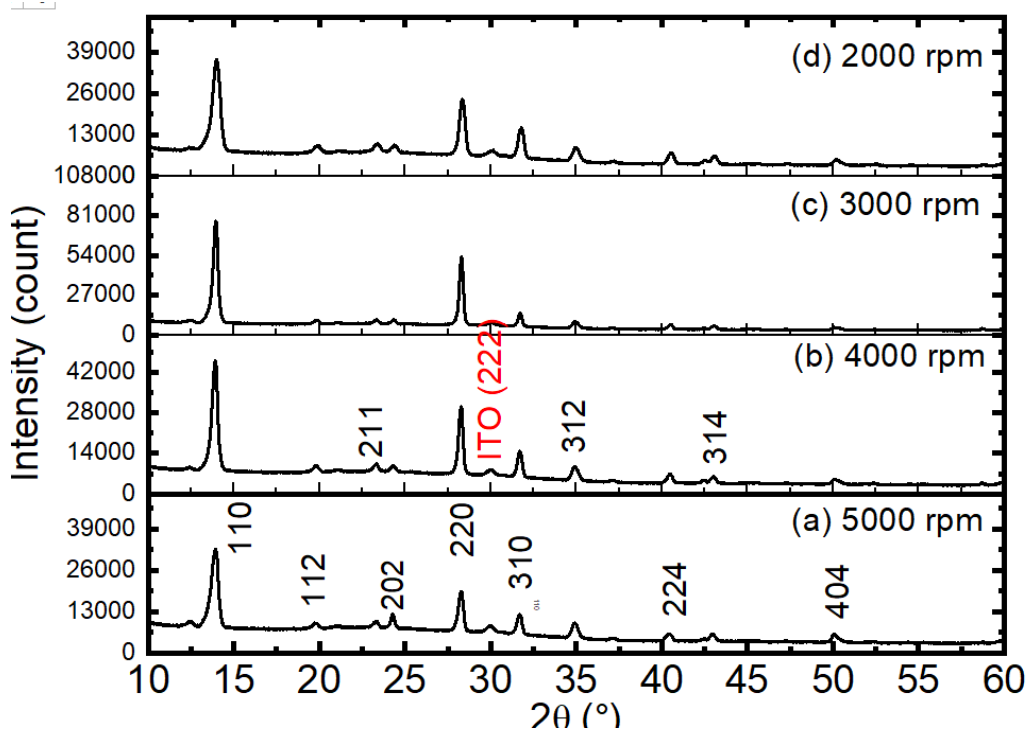


Fig. 2. XRD patterns of the samples deposited under varying speeds (a) 5000, (b) 4000, (c) 3000, (d) 2000 rpm.

No distinct diffraction peak is evident near $2\theta \approx 12.63^\circ$, which is the principal (001) reflection of crystalline PbI₂. This suggests that the precursor materials were largely converted into MAPbI₃, with no appreciable residual PbI₂ detectable by XRD.

The structural parameters calculated from the dominant MAPbI₃ (110) reflection further demonstrate the influence of spin speed on crystal quality. The peak position changes only slightly

from 13.94 to 14.02 °, indicating that deposition speed does not significantly alter the crystal phase. The small displacement of the peak may instead arise from minor changes in residual strain, lattice distortion or film–substrate interaction.

The 3000 rpm film exhibits the smallest FWHM of 0.35211°, and consequently possesses the largest crystallite size of 22.732 nm. It also has the lowest microstrain of 1.257×10^{-2} and the lowest dislocation density of 1.936×10^{-3} lines/nm². These results indicate that deposition at 3000 rpm produces the highest degree of crystalline ordering, with fewer lattice imperfections and reduced internal strain.

At 4000 rpm, the FWHM increases to 0.42891°, giving a crystallite size of 18.66 nm, a microstrain of 1.532×10^{-2} , and a dislocation density of 2.872×10^{-3} lines/nm². Although the crystal quality remains reasonably high, the smaller crystallite size and increased strain relative to the 3000 rpm film suggest that faster solvent removal begins to restrict crystal growth.

The 5000 rpm film has a broader FWHM of 0.58067°, corresponding to a crystallite size of 13.78 nm. Its microstrain and dislocation density increase to 2.072×10^{-2} and 5.264×10^{-3} lines/nm², respectively. At this high spin speed, rapid thinning and accelerated solvent evaporation may reduce the time available for precursor diffusion and crystal coalescence, thereby producing smaller coherent crystallites and a higher concentration of structural imperfections.

Similarly, the 2000 rpm sample exhibits the broadest peak, with an FWHM of 0.60842°, and the smallest crystallite size of 13.16 nm. It also presents the highest microstrain of 2.160×10^{-2} and dislocation density of 5.778×10^{-3} lines/nm². At the relatively low spin speed, the thicker wet precursor layer may dry less uniformly, promoting non-uniform nucleation, residual stress and structural disorder.

This non-monotonic behaviour indicates that an intermediate spin speed provides a better balance between film thinning, solvent evaporation, nucleation and grain growth. Among the investigated conditions, 3000 rpm yields the best structural quality, as demonstrated by its largest crystallite size, lowest microstrain and lowest dislocation density. These features are generally favourable for photovoltaic applications because reduced structural disorder can suppress defect-assisted recombination and facilitate charge transport through the MAPbI₃ absorber.

The crystallite size was determined using Debye–Scherrer equation (Eq. 7) [25]

$$D = \frac{K\lambda}{\beta \cos \theta} \quad 7$$

where $K = 0.9$, $\lambda = 0.15406$ nm for Cu K α radiation, β = FWHM in radians, and $\theta = 2\theta/2$, the crystallite sizes for the MAPbI₃ (110) peak are shown in Table 4. The Microstrain was evaluated by Eq. 8, while the dislocation density was calculated using Eq. 9 [25]

$$\varepsilon = \frac{\beta}{4 \tan \theta} \quad 8$$

$$\delta = \frac{1}{D^2} \quad 9$$

Table 4. Structural parameters for the samples

Deposition speed (rpm)	Peak	2theta (°)	FWHM (°)	FWHM (rad)	Crystallite size (nm)	Microstrain	Dislocation density (lines/nm ²)
5000		13.941	0.58067	0.01014	13.78	2.072×10^{-2}	5.264×10^{-3}
4000		13.930	0.42891	0.00749	18.66	1.532×10^{-2}	2.872×10^{-3}
3000	110	13.941	0.35211	0.00615	22.73	1.257×10^{-2}	1.936×10^{-3}
2000		14.017	0.60842	0.01062	13.16	2.160×10^{-2}	5.778×10^{-3}

3.2 Optical Properties

3.2.1 Optical absorbance, transmittance, absorption coefficient and refractive index

The absorption coefficient (α) (Fig.3a) was estimated using Eq. (10), while the extinction coefficient (k) (Fig. 4a) was determined using Eq. (11). These parameters were subsequently used to calculate the refractive index (n) according to Eq. (12) and the real part of the dielectric constant (ε_r) using Eq. (13). The estimated bandgap values and the optical parameters selected for comparison are summarized in Table 5 [26][27].

$$k = \frac{\alpha\lambda}{4\pi} \quad 10$$

$$\alpha = \frac{2.303A}{t} \quad 11$$

$$n = \frac{1 + \sqrt{R}}{1 - \sqrt{R}} \quad 12$$

$$\varepsilon_r = n^2 - k^2 \quad 13$$

where A represent absorbance, R is reflectance, and t is film thickness.

Table 5. Estimated bandgap and selected optical parameters of MAPbI₃ thin films deposited under different spin-coating conditions.

Deposition speed (rpm)	E _g (eV)	Refractive index (n) at 886 nm	Dielectric constant (ε _r) at 886 nm
5000	1.59	2.32	4.86
4000	1.10	2.32	4.86
3000	1.10	2.16	4.36
2000	1.10	2.16	4.36

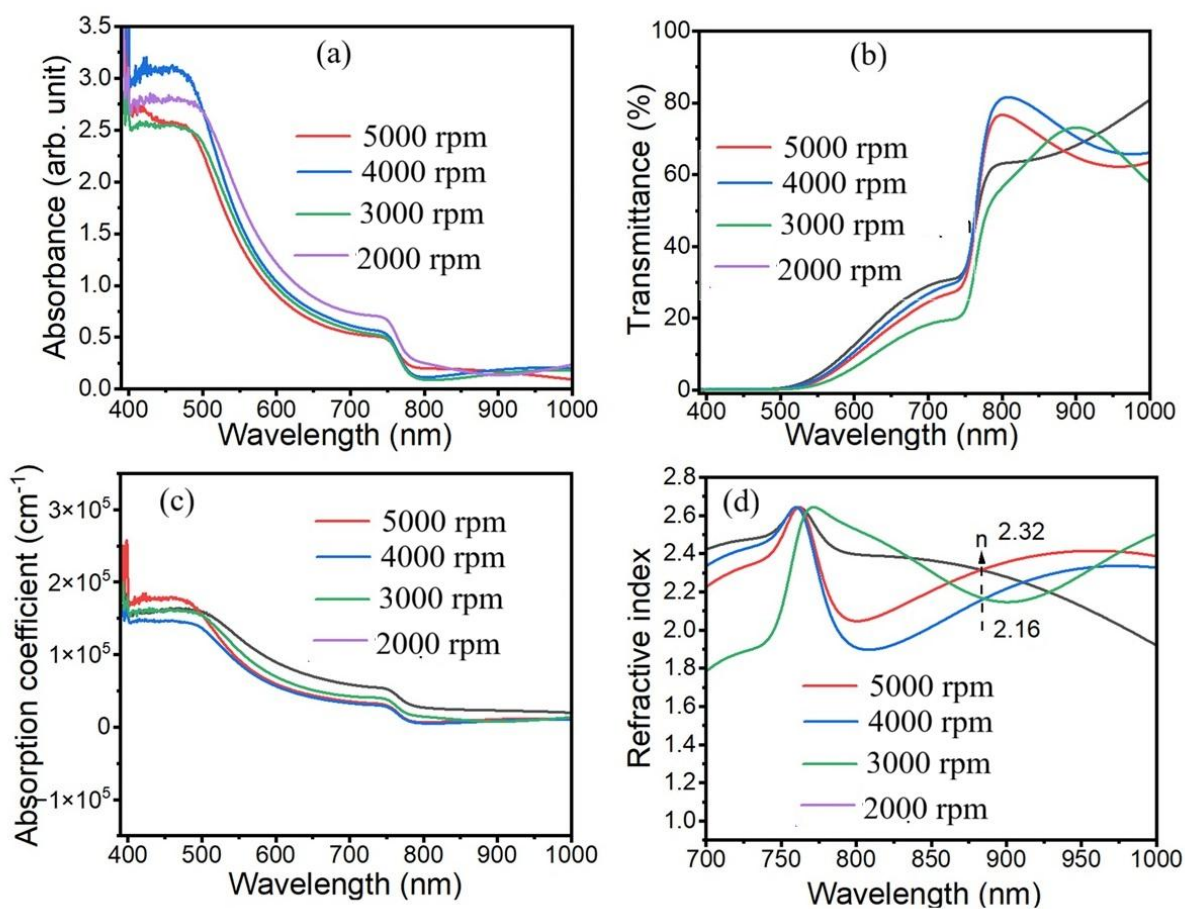


Fig. 3 (a) Optical absorption, (b) Optical transmittance, (c) Absorption coefficient, (d) Wavelength-dependent refractive index.

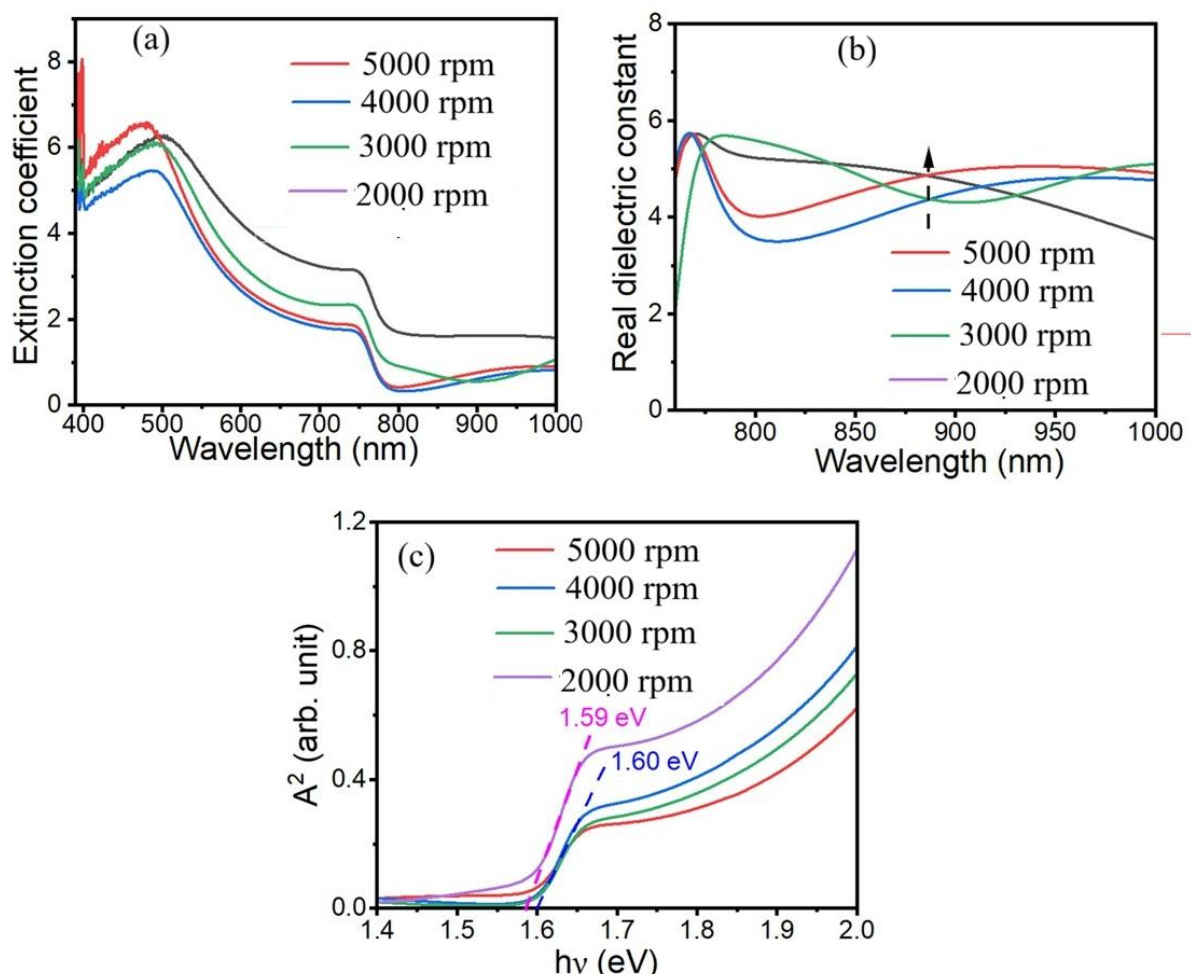


Fig. 4. (a) Extinction coefficient vs. wavelength, (b) Real dielectric constant vs. wavelength, (c) Graph of A^2 vs. $h\nu$ for optical bandgap determination.

4. Assessment of photovoltaic performance by SCAPS-1D simulation

4.1 Effect of experimentally determined bandgap on prototype-cell performance

This section discusses the effect of bandgap of the prepared MAPbI₃ films on photovoltaic performance of simulated FTO/TiO₂/MAPbI₃/Spiro-OMeTAD/C PSC architecture. The literature reports that the bandgap of MAPbI₃ is one of the most important parameters controlling the photovoltaic performance of PSCs because it determines the spectral absorption range, open-circuit voltage (V_{OC}), short circuit current density (J_{SC}), and the power conversion efficiency (PCE)

[28][29][30]. Shockley and Queisser [31] established that the photovoltaic efficiency limit of a single-junction solar cell is fundamentally determined by the absorber bandgap through the balancing between current generation and voltage loss. De Wolf et al. [32] further demonstrated that the sharp absorption edge and bandgap quality of perovskites are closely related to photovoltaic performance.

In this section we simulated the prototype devices by isolating the effect of the experimentally determined MAPbI₃ bandgap while keeping absorber thickness and all other device parameters constant. The intention is finding the optimal parameters for the optimized device with boosted theoretical performance to provide a pathway for practical device design. The bulk-layer and interface-defect parameters used as inputs for the SCAPS-1D simulations are detailed in Tables 1 and 2, respectively

Fig.5 present the simulated performance of the FTO/TiO₂/MAPbI₃/Spiro-OMeTAD/carbon prototype cells using the experimentally determined MAPbI₃ bandgaps. Only the absorber bandgap was varied, while the thickness and all other electrical, optical and defect-related parameters were kept constant. Therefore, the results isolate the effect of bandgap and do not represent the full influence of spin-coating speed.

The prototype based on the 5000 *rpm* film with ($E_g=1.59$ eV) produced the best performance, with $V_{OC}=1.10$ V, $J_{SC}=21.14$ mA/cm², FF=71.85%, and PCE=16.67%. The devices based on the 1.60 eV absorbers deposited at 4000, 3000 and 2000 rpm produced identical values of $V_{OC}=1.10$ V, $J_{SC}=20.92$) mA/cm², FF=71.73%, and PCE=16.47%, because they were simulated with the same bandgap and otherwise identical input parameters.

Table 6. Effect of bandgap of the prepared MAPbI₃ films on photovoltaic performance of the prototype PSCs

Deposition speed (rpm)	E_g (eV)	Photovoltaic performance			
		V_{OC} (V)	J_{SC} (mA/cm ²)	FF (%)	PCE (%)
5000	1.59	1.10	21.14	71.85	16.67
4000	1.60	1.10	20.92	71.73	16.47

3000	1.60	1.10	20.92	71.73	16.47
2000	1.60	1.10	20.92	71.73	16.47

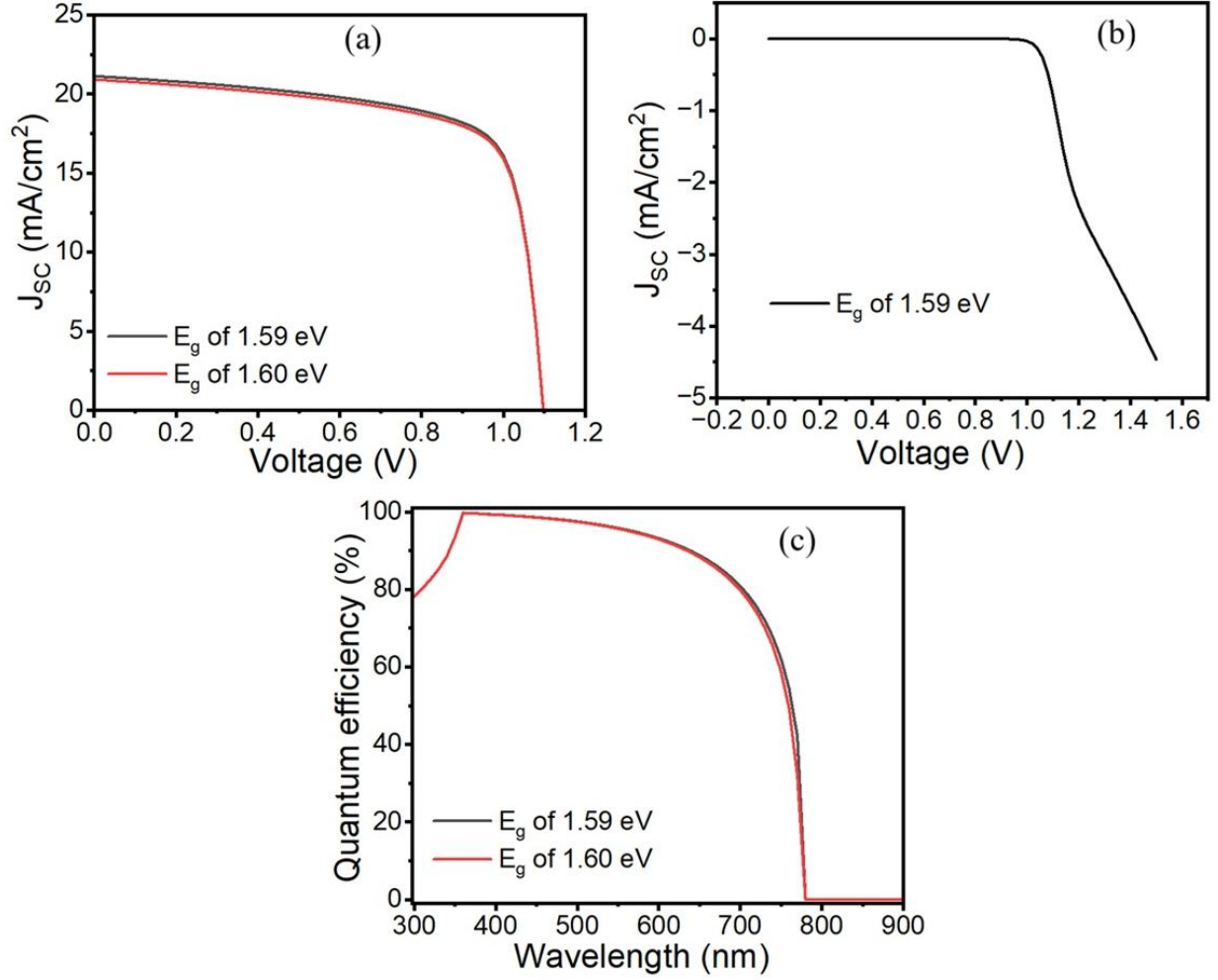


Fig. 5. (a) J-V plots under light for the PSCs obtained from the bandgaps of the synthesized MAPbI₃ absorber layer spin coated at varying speeds, (b) J-V plot under dark for the samples with the optimal bandgap of 1.59 eV. (c) Associated QE plots for device under illumination.

The PCE takes the combined effect of V_{oc} , J_{sc} , and FF according to Eq. 14.

$$PCE(\%) = \left(\frac{V_{oc} \times J_{sc} \times FF}{P_{in}} \right) \times 100$$

The slightly higher J_{SC} of the 1.59 eV device results from its marginally broader absorption range. Its lower photon-energy threshold allows absorption of additional photons near the red edge of the solar spectrum, increasing electron–hole-pair generation and raising J_{SC} by approximately 1.05%. This behaviour is visible in Fig. 5a, where the illuminated J–V curves largely overlap, although the 1.59 eV cell shows a slightly higher current-density plateau. Because the transport, defect and resistance parameters were unchanged, the photocurrent increase is mainly optical rather than a consequence of improved transport or reduced recombination.

The simulated V_{oc} remains unchanged because the 0.01 eV bandgap difference is too small to produce a visible variation at the reported precision. Moreover, V_{oc} is also controlled by quasi-Fermi-level splitting, recombination, carrier concentrations, contacts and band alignment. The FF changes only slightly from 71.73% to 71.85%, indicating almost identical charge extraction, electric-field distribution and resistive losses in both devices.

The combined increase in J_{SC} and FF raises the PCE from 16.47% to 16.67%. Thus, the 1.59 eV absorber provides a slightly better balance between photon harvesting and voltage generation. Fig. 5c supports this interpretation, as the 1.59 eV cell has a slightly red-shifted absorption cut-off of about 780 nm compared with approximately 775 nm for the 1.60 eV cell, resulting in a marginally higher integrated photocurrent.

Figure 5b shows that the dark current of the 1.59 eV device remains low at small forward bias because the junction barrier and selective contacts restrict carrier injection. At higher forward voltage, the current rises rapidly as the barrier is reduced and diode injection and recombination become dominant. However, spin-coating speed can also modify film thickness, morphology, crystallinity, defect density and interface quality, which must be included separately to reproduce its full effect on device performance.

4.1.1 Energy-band characteristics

Fig. 6 presents the energy-band profiles of the FTO/TiO₂/MAPbI₃/Spiro-OMeTAD device for MAPbI₃ bandgaps of 1.60 and 1.59 eV under illumination, together with the dark-state profile of the 1.59 eV device. The plots show the conduction-band edge (E_C), valence-band edge (E_V), electron quasi-Fermi level (F_n), and hole quasi-Fermi level (F_p) across the device thickness. The conduction-band offset at the TiO₂/MAPbI₃ interface is -0.30 eV.

Thus, the interface exhibits a CBO of -0.30 eV, corresponding to a small cliff-like conduction-band alignment. This alignment promotes electron transfer from MAPbI₃ into TiO₂ because the TiO₂ conduction band lies approximately 0.30 eV below that of the absorber. However, an excessively large negative CBO could increase interfacial electron-hole recombination; therefore, control of the TiO₂/MAPbI₃ interface remains important.

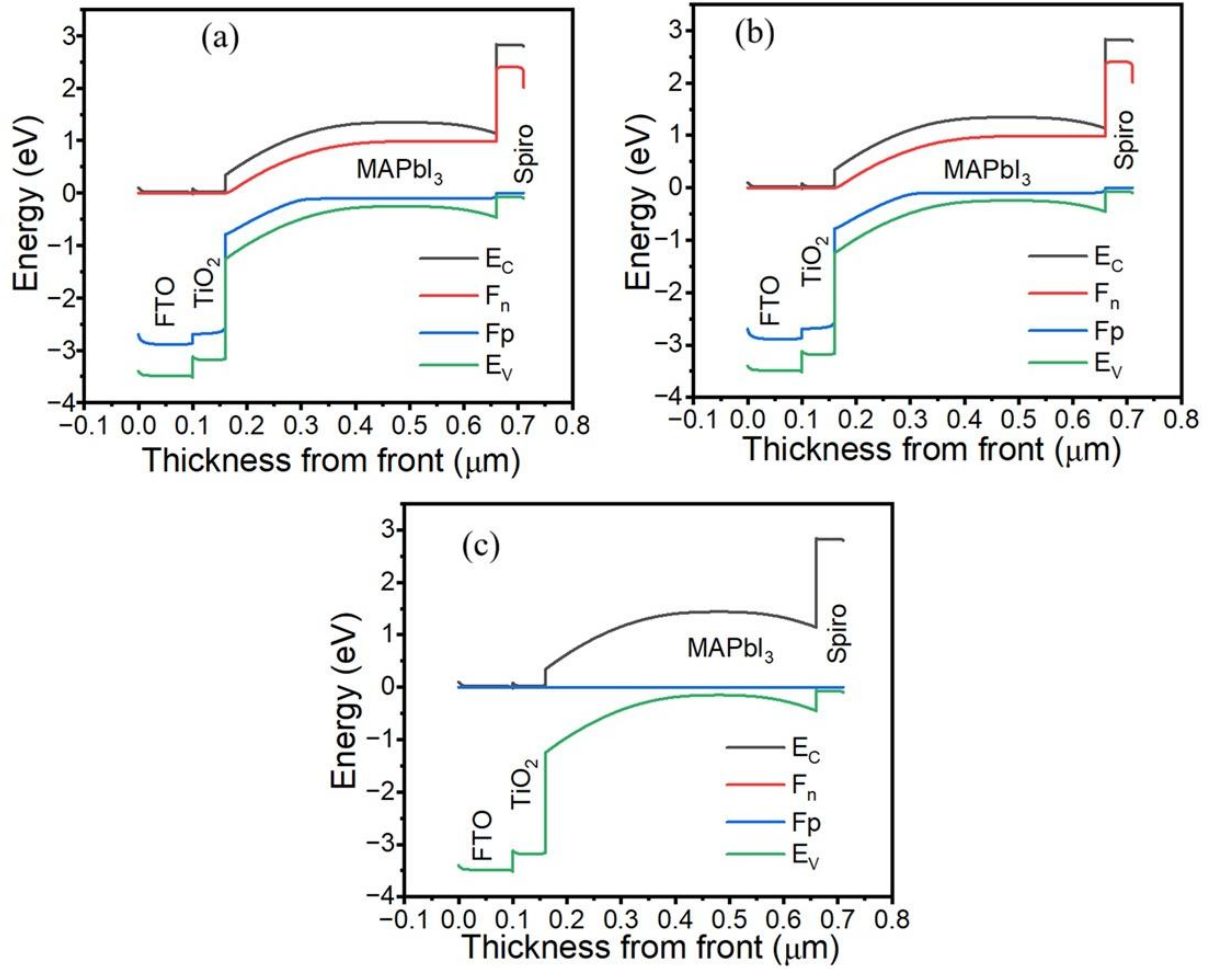


Fig. 6. Energy band plots of the films with bandgap of 1.60 eV under illumination, (b) Energy band plots of the film with bandgap of 1.59 eV under illumination, (c) Energy band plots of the film with bandgap of 1.59 eV under dark.

The valence-band positions of MAPbI₃ are -5.49 and -5.50 eV for bandgaps of 1.59 and 1.60 eV, respectively. Consequently, the MAPbI₃/Spiro-OMeTAD interface has a valence-band offset (VBO) magnitude of 0.41 eV for the 1.59 eV absorber and 0.40 eV for the 1.60 eV absorber. In another convention, these correspond to -0.41 and -0.40 eV, respectively. The offset provides an energetic driving force for hole transfer into Spiro-OMeTAD, while its high conduction-band position blocks electrons from reaching the rear contact.

Under illumination, the bending of E_C and E_V across MAPbI₃ indicates an internal electric field that assists photogenerated-carrier separation. Electrons are directed towards TiO₂/FTO, while holes are transported towards Spiro-OMeTAD/carbon. The separation between F_n and F_p represents the light-induced quasi-Fermi-level splitting and is directly related to the generated photovoltage. The nearly identical splitting and band bending for the two absorber bandgaps agree with their common V_{OC} of 1.10 V.

Under dark conditions, F_n and F_p converge towards a common equilibrium Fermi level because excess photogenerated carriers are absent. Nevertheless, the band bending remains, demonstrating that the built-in field originates from equilibrium charge redistribution across the contacts and heterointerfaces. Overall, Fig. 6 confirms selective carrier extraction, while the nearly unchanged CBO and VBO explain why the small bandgap variation primarily affects optical absorption and J_{SC} , rather than substantially changing the interfacial transport energetics.

4.2 Effect of layer thickness on photovoltaic performance

Fig. 7 shows that the photovoltaic response depends strongly on the function of each layer in the FTO/TiO₂/MAPbI₃/Spiro-OMeTAD/carbon device. Varying the TiO₂ thickness from 0.04 to 0.12 μm produces almost no change in V_{OC} or J_{SC} , while FF and PCE increase only slightly from 71.84 to 71.89% and from 16.66 to 16.68% , respectively (Fig. 7a). This weak dependence indicates that TiO₂ is already sufficiently thick for selective electron extraction and hole blocking throughout the investigated range. Because TiO₂ contributes little to useful photogeneration, further thickness increase provides no significant photocurrent benefit and only marginally modifies charge transport.

In contrast, MAPbI₃ thickness has the strongest influence on device performance. Increasing the absorber thickness from 0.5 to 1.0 μm raises J_{sc} from 21.14 to 22.86 mA/cm^2 , FF from 71.85 to

77.51%, and PCE from 16.67 to 19.78%, while V_{oc} increases slightly from 1.10 to 1.12 V (Fig. 7b). A thicker absorber increases the optical path length and the probability that incident photons are absorbed, particularly near the band-edge region where the absorption coefficient is lower. The resulting increase in electron–hole-pair generation explains the improved J_{sc} and the enhanced long-wavelength quantum efficiency in Fig. 8b. The modest increase in V_{oc} is consistent with greater photogenerated carrier density and stronger quasi-Fermi-level splitting. The improved FF suggests that, within this simulated thickness range, the additional carrier generation outweighs transport and recombination losses. However, experimentally excessive absorber thickness may eventually increase bulk recombination because carriers must travel longer distances before collection.

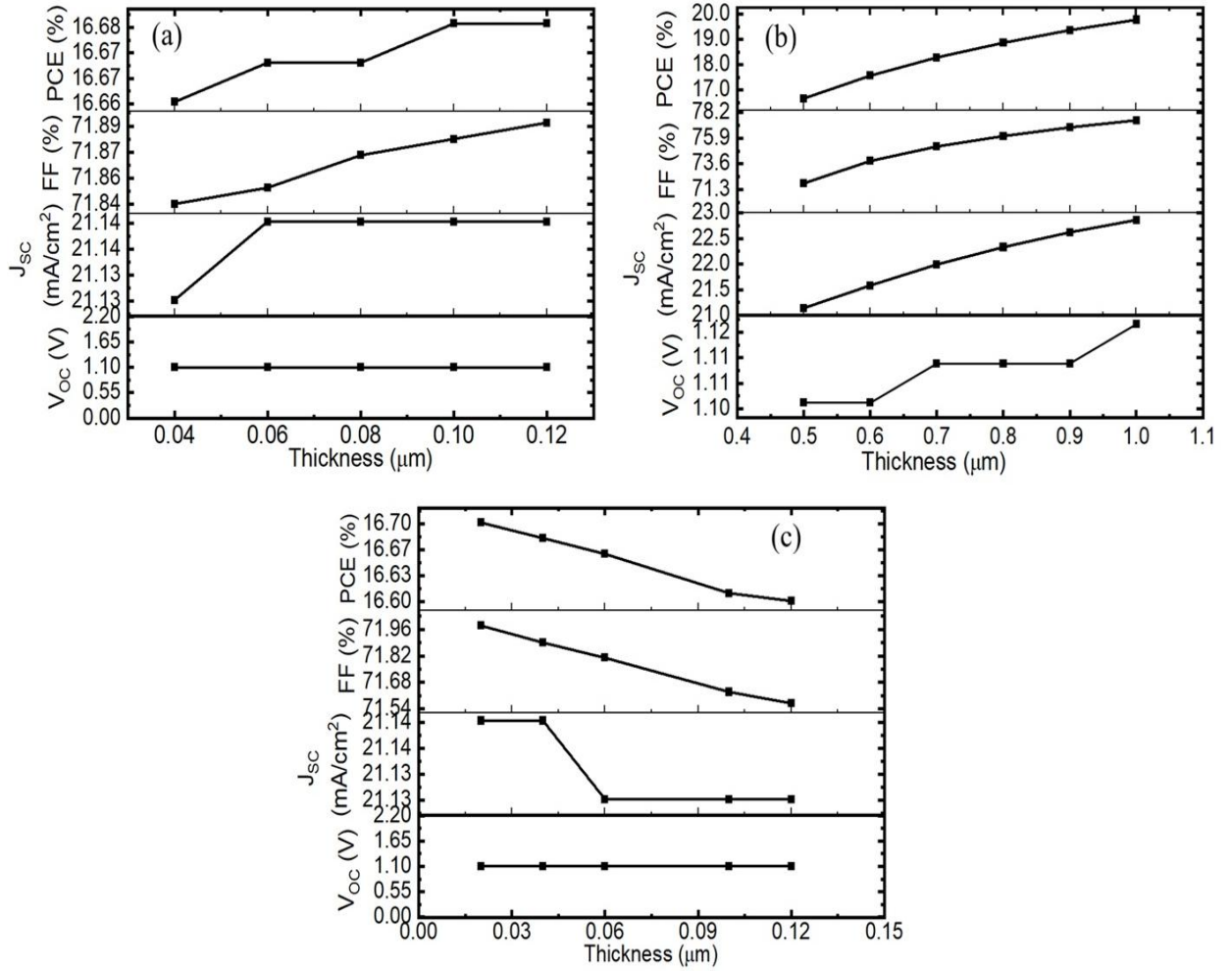


Fig. 7. (a) Dependence of V_{OC} , J_{SC} , FF and PCE on thickness of TiO_2 , (b) Dependence of V_{OC} , J_{SC} , FF and PCE on thickness of $MAPbI_3$, (c) Dependence of V_{OC} , J_{SC} , FF and PCE on thickness of Spiro-OMeTAD.

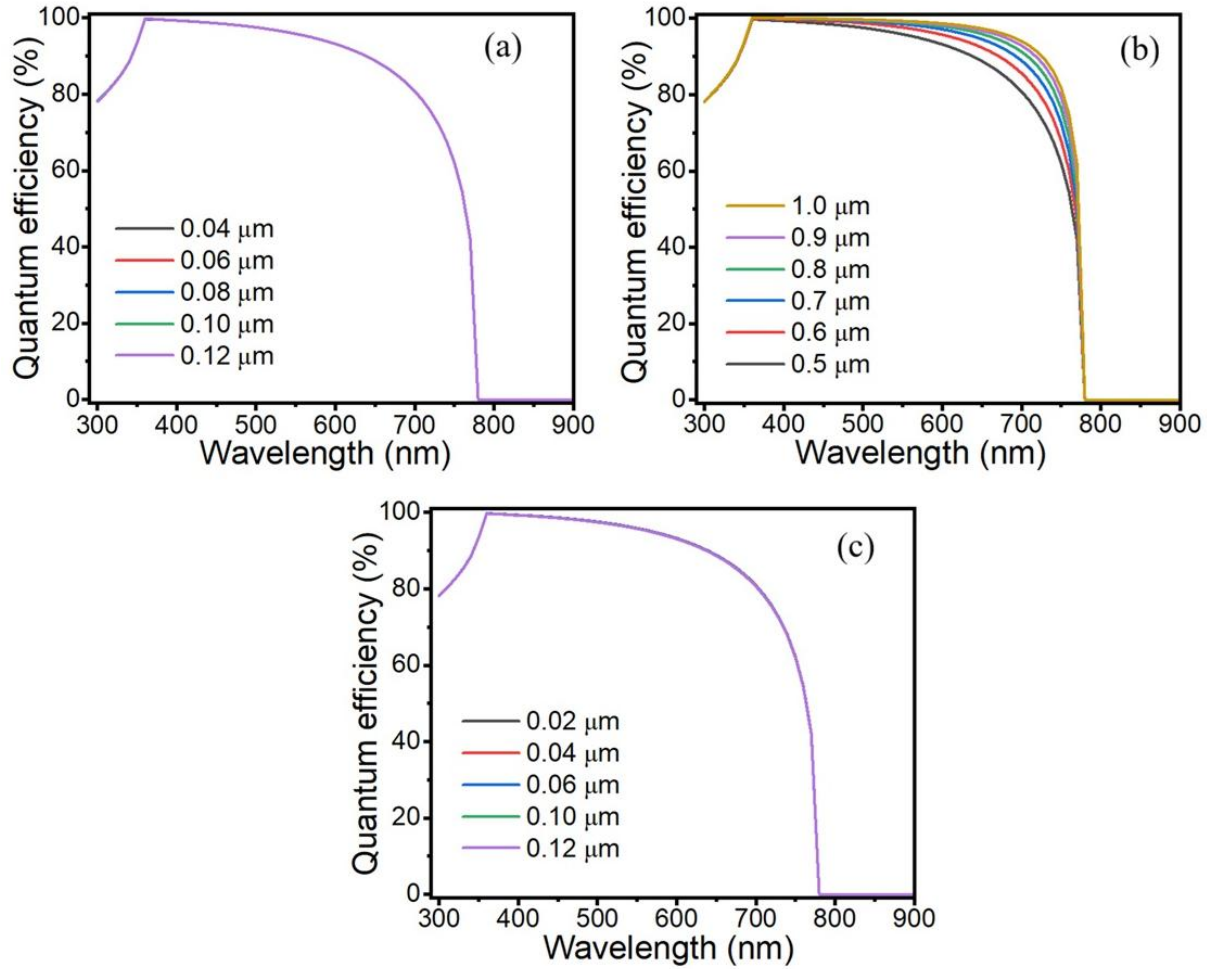


Fig. 8. QE plots for the effect of thickness of (a) TiO_2 , (b) $MAPbI_3$, (c) Spiro-OMeTAD.

The Spiro-OMeTAD thickness produces the opposite trend. As its thickness increases from 0.02 to 0.12 μm , V_{oc} and J_{sc} remain nearly constant, whereas FF decreases from 71.98 to 71.57% and PCE falls from 16.70 to 16.60% (Fig. 7c). Since Spiro-OMeTAD does not significantly contribute to photogeneration, increasing its thickness mainly lengthens the hole-transport pathway and raises the effective transport resistance. This increases the probability of carrier loss and slightly reduces charge-extraction efficiency. The nearly overlapping QE curves in Fig. 8c confirm that the effect

is predominantly electrical rather than optical. Accordingly, the selected values of 0.12 μm TiO_2 , 1.0 μm MAPbI_3 and 0.02 μm Spiro-OMeTAD provide the best simulated performance.

4.3 Effect of Carrier density

Fig. 9 show that increasing the carrier density of both TiO_2 and Spiro-OMeTAD improves the photovoltaic performance of the FTO/ TiO_2 /MAPbI₃/Spiro-OMeTAD/carbon cell. The improvement is mainly associated with enhanced electrical conductivity, stronger selective-contact behaviour and more efficient extraction of photogenerated carriers. For TiO_2 , increasing the donor density from 10^{14} to 10^{18} cm^{-3} raises Voc from 1.07 to 1.10 V, Jsc from 20.52 to 21.14 mA/cm^2 , FF from 64.45 to 71.85%, and PCE from 14.18 to 16.67%. At low donor density, the relatively poor conductivity of TiO_2 increases electron-transport resistance and promotes carrier accumulation near the TiO_2 /MAPbI₃ interface, which increases the probability of interfacial recombination. Higher donor density increases the electron concentration, lowers the resistive loss of the ETL and improves electron extraction towards FTO. It also shifts the TiO_2 Fermi level closer to the conduction band, strengthening the junction field and improving carrier separation. The pronounced rise in FF therefore reflects reduced transport resistance and recombination loss, while the modest increases in Voc and Jsc indicate improved quasi-Fermi-level splitting and charge collection.

A similar trend is observed for Spiro-OMeTAD. Increasing its acceptor density from 10^{15} to 10^{19} cm^{-3} increases Voc from 1.06 to 1.14 V, Jsc from 20.93 to 21.30 mA/cm^2 , FF from 66.79 to 73.03%, and PCE from 14.82 to 17.71%. Higher acceptor density increases the hole concentration and conductivity of Spiro-OMeTAD, thereby reducing the resistance encountered by holes travelling towards the carbon electrode. It also reduces hole accumulation at the MAPbI₃/Spiro-OMeTAD interface and suppresses interfacial recombination. The resulting improvement in quasi-Fermi-level splitting produces the marked increase in Voc, while more efficient hole extraction improves FF and PCE.

The results further show that PCE is more sensitive to the carrier density of Spiro-OMeTAD than to that of TiO_2 over the investigated ranges. This indicates that hole extraction and transport through the HTL constitute a comparatively stronger limitation in the prototype cell. Overall, carrier densities of 10^{18} cm^{-3} for TiO_2 and 10^{15} cm^{-3} for Spiro-OMeTAD provide the best simulated

performance by reducing transport resistance, limiting interfacial carrier accumulation and promoting efficient, selective charge collection.

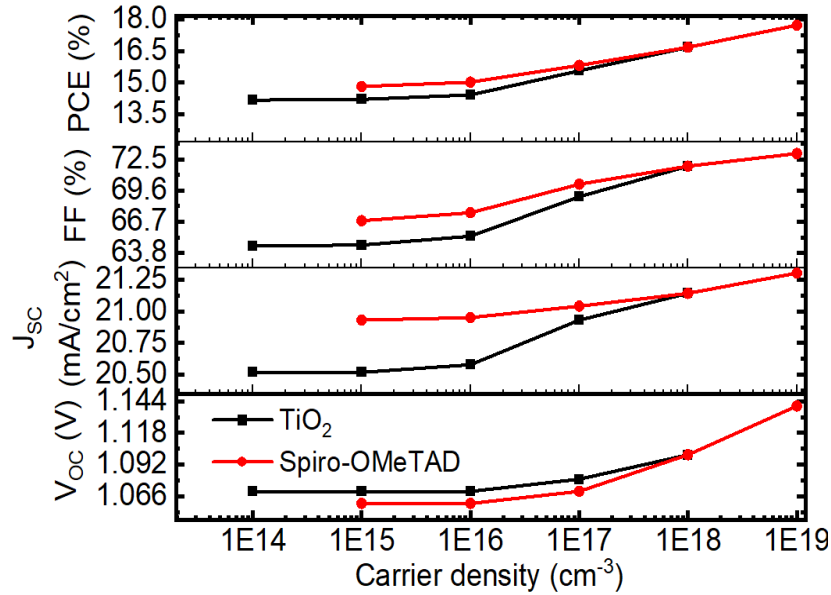


Fig. 9. Dependence of V_{OC} , J_{SC} , FF and PCE on carrier density of TiO_2 and Spiro-OMeTAD.

4.4 Effect of absorber bulk-defect density

Fig. 10 and the corresponding table show that increasing the MAPbI₃ bulk-defect density strongly degrades the photovoltaic performance of the prototype cell. As the defect density rises from 10^{14} to 10^{18} cm⁻³, J_{SC} decreases from 21.14 to 7.90 mA/cm², FF falls from 71.85 to 60.81%, and PCE declines sharply from 16.67 to 4.55%.

Bulk defects introduce localized energy states within the MAPbI₃ bandgap that act as electron and hole capture centres. Increasing their concentration enhances Shockley-Read-Hall recombination, shortens carrier lifetime and reduces the diffusion length. Consequently, a larger fraction of the photogenerated carriers recombines inside the absorber before reaching the TiO_2 and Spiro-OMeTAD interfaces. This explains the strong reduction in J_{SC} , because fewer carriers contribute to the external photocurrent.

The increased recombination also limits the separation of the electron and hole quasi-Fermi levels, thereby reducing the attainable photovoltage. Simultaneously, trap-mediated carrier loss weakens charge collection near the maximum-power region and increases the non-ideal diode response, producing the observed deterioration in FF. Since PCE depends on the combined values of V_{oc} , J_{sc} , and FF, the simultaneous recombination-induced losses cause the pronounced efficiency reduction shown in Fig. 10.

The device retains comparatively good performance at defect densities of 10^{14} and 10^{15} cm^{-3} , but substantial degradation begins at 10^{16} cm^{-3} , where PCE decreases to 14.13%. At 10^{18} cm^{-3} , severe trap-assisted recombination dominates carrier transport, reducing the PCE to 4.55%. Therefore, maintaining a low MAPbI₃ defect density is essential for preserving carrier lifetime, quasi-Fermi-level splitting and efficient charge collection.

The reported $V_{oc}=6.48 \text{ V}$ at 10^{17} cm^{-3} is physically inconsistent with the 1.59 eV absorber bandgap and with the stated PCE of 10.00%; this table entry should be checked against the original SCAPS output before publication.

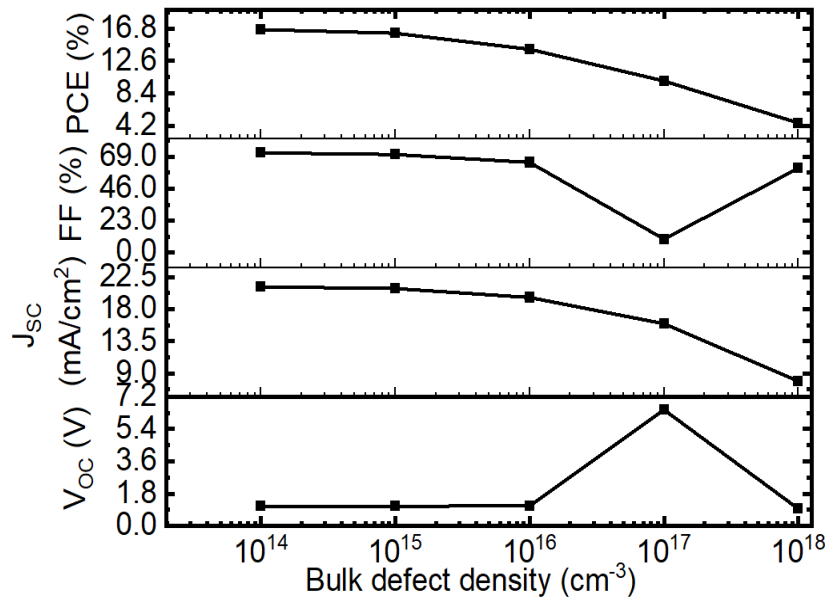


Fig. 10. Dependence of V_{oc} , J_{sc} , FF and PCE on bulk defect density of MAPbI₃.

4.5 Impact interface defect density

Fig. 11 and the corresponding tables show that increasing the interface-defect density degrades device performance by introducing interfacial trap states that capture photogenerated carriers and promote Shockley-Read-Hall recombination. The extent of the degradation, however, differs considerably between the $\text{TiO}_2/\text{MAPbI}_3$ and $\text{MAPbI}_3/\text{Spiro-OMeTAD}$ interfaces.

At the $\text{TiO}_2/\text{MAPbI}_3$ interface, increasing the defect density from 10^{10} to 10^{14} cm^{-2} reduces V_{OC} from 1.11 to 1.01 V and PCE from 16.77 to 15.72%, while J_{sc} remains nearly constant at approximately 21.15 mA/cm^2 . The nearly unchanged photocurrent indicates that electron collection through TiO_2 remains effective, even at relatively high interface-defect density. However, the additional traps increase electron-hole recombination near the ETL/absorber junction, reducing quasi-Fermi-level splitting and therefore V_{OC} . The increase in FF from 71.43 to 73.64% does not indicate improved interface quality; rather, it partly compensates for the voltage loss and may arise from changes in the simulated J–V curvature. The overall reduction in PCE confirms that the net effect of increasing interface defects is detrimental.

The $\text{MAPbI}_3/\text{Spiro-OMeTAD}$ interface is considerably more sensitive. Increasing its defect density from 10^{10} to 10^{14} cm^{-2} decreases V_{OC} from 1.19 to 1.08 V, J_{sc} from 22.37 to 19.83 mA/cm^2 , FF from 81.31 to 70.25%, and PCE from 21.71 to 15.03%. Trap states at this interface capture electrons and holes before selective hole extraction, enhancing non-radiative recombination and carrier accumulation. This reduces carrier lifetime, weakens quasi-Fermi-level splitting and increases extraction losses, accounting for the simultaneous decline in voltage, photocurrent and FF.

The stronger response of the rear interface indicates that hole extraction through the $\text{MAPbI}_3/\text{Spiro-OMeTAD}$ junction is a major performance-limiting process in the prototype device. Consequently, reducing the defect density at this interface through improved film coverage, chemical passivation and better interfacial contact should provide a larger efficiency gain than equivalent improvement at the $\text{TiO}_2/\text{MAPbI}_3$ interface. Overall, an interface-defect density near 10^{10} cm^{-2} is most favourable because it suppresses trap-assisted recombination and preserves efficient carrier separation and collection.

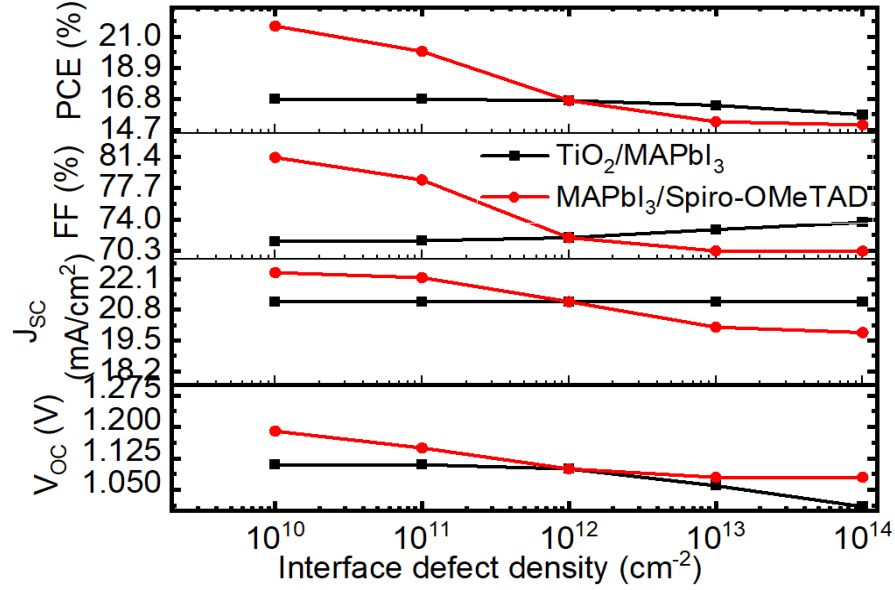


Fig. 11. Dependence of V_{OC} , J_{SC} , FF and PCE on defect density of $TiO_2/MAPbI_3$, and $MAPbI_3/Spiro-OMeTAD$ interface.

4.6 Effect of series and shunt resistance

Fig. 12 shows that the photovoltaic response is highly sensitive to both series resistance and shunt resistance. Increasing from 0 to $50 \Omega cm^2$ reduces J_{SC} from 21.14 to 18.13 mA/cm^2 , FF from 71.85 to 28.08%, and PCE from 16.67 to 5.59%, while V_{OC} remains nearly constant at 1.10 V.

Series resistance represents ohmic losses arising from the transport layers, electrodes, bulk materials and interfacial contacts. As series resistance increases, part of the photogenerated voltage is lost internally as an ($V = I.R_s$) drop, particularly near the maximum-power and short-circuit regions. This limits carrier extraction and distorts the illuminated J–V curve, producing the pronounced reduction in FF. The decrease in J_{SC} at high series resistance indicates that severe transport resistance also restricts the collection of photogenerated carriers. In contrast, V_{OC} is largely unaffected because the current is zero under open-circuit conditions and the associated resistive voltage drop is therefore negligible.

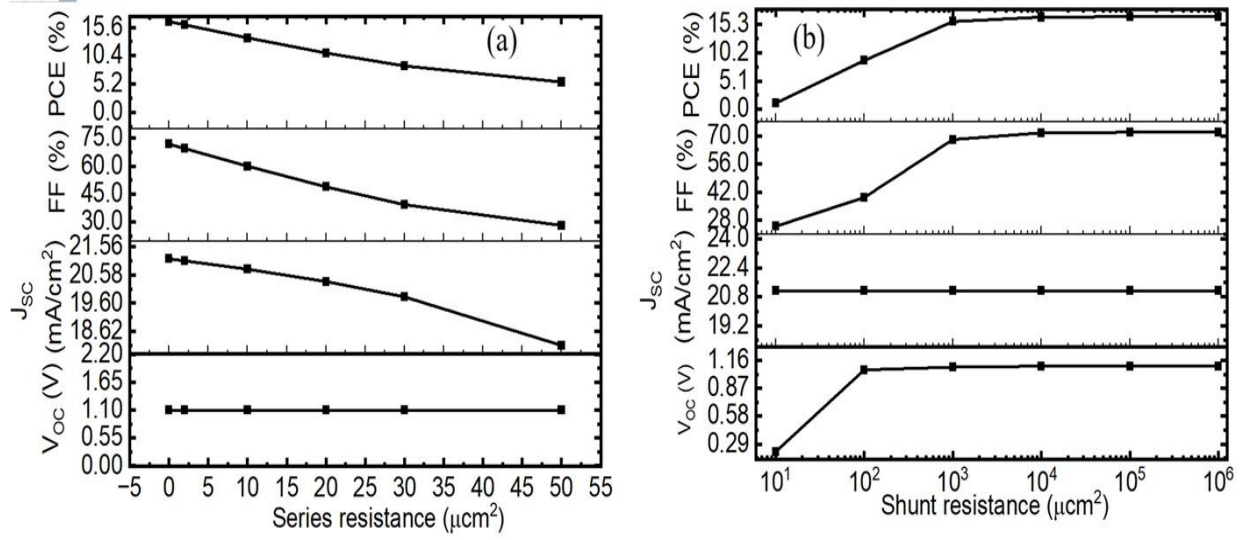


Fig. 12. Dependence of V_{OC} , J_{SC} , FF and PCE on (a) series and (b) shunt resistances of the stack device.

Shunt resistance exhibits the opposite effect. Increasing shunt resistance from 10^1 to $10^6 \Omega\text{cm}^2$ raises V_{oc} from 0.21 to 1.10 V, FF from 24.98 to 71.85%, and PCE from 1.10 to 16.67%, whereas J_{sc} remains constant at 21.14 mA/cm². A low shunt resistance provides an alternative leakage pathway across the junction, allowing photogenerated current to bypass the external load. This leakage weakens the photovoltage, increases the slope of the J–V curve near short circuit and severely lowers FF and PCE. As shunt resistance increases, leakage becomes progressively suppressed, enabling more of the generated current to flow through the external circuit.

The improvement becomes small above approximately 10^4 – $10^5 \Omega\text{cm}^2$, indicating that shunt leakage is then negligible and other recombination or transport processes control performance. Overall, efficient operation requires a low series resistance to minimize ohmic voltage losses and a high shunt resistance to suppress parasitic leakage. The best simulated performance is therefore obtained at series resistance of $0 \Omega\text{cm}^2$ and shunt resistance of $10^6 \Omega\text{cm}^2$.

4.7 Photovoltaic performance of optimized device

Simulations of the prototype device was used to identify optimal/ best layer thicknesses of 0.12 μm for TiO_2 , 1.0 μm for MAPbI_3 , and 0.02 μm for the HTL. Carrier density of the HTL was slight increased to 10^{19} cm^{-3} , while the bulk defect density of the ETL and HTL were reduced to 10^{14} cm^{-3} . The interface defect density was reduced to 10^{10} cm^{-2} . It should be noted that optimized values of the bulk [33][34][35] and interface [36][37][38] defect density are supported by experimental works. All other parameters remained unchanged as used in the prototype device.

Figure 13 demonstrates the improved photovoltaic response obtained after jointly optimizing the layer thicknesses, carrier densities, and bulk and interface defect densities of the FTO/ TiO_2 / MAPbI_3 /Spiro-OMeTAD/carbon device. At 300 K, the optimized cell delivers V_{OC} of 1.25 V, J_{SC} of 24.24 mA/cm^2 , FF of 83.61%, and PCE of 25.38%, representing a substantial improvement over the prototype efficiency of 16.67%. The PCE of the optimized cell is consistent with the Shockley-Queisser limit for absorber with bandgaps of 1.50 and 1.60 eV [39].

The more rectangular illuminated J-V curve in Fig. 13a indicates reduced transport and recombination losses. The higher J_{SC} originates mainly from the increased MAPbI_3 thickness, which improves photon absorption and photocarrier generation, while the higher V_{oc} reflects reduced non-radiative recombination and stronger electron-hole quasi-Fermi-level splitting. The improved FF confirms more efficient carrier extraction, lower effective resistive losses and reduced carrier accumulation at the transport-layer interfaces.

The quantum-efficiency spectrum in Fig. 13b remains close to unity over most of the visible region before falling sharply near the MAPbI_3 absorption edge. This broad and high response demonstrates efficient photon-to-carrier conversion and collection across the absorber's active spectral range. The decline at longer wavelengths occurs because photons with energies below the MAPbI_3 bandgap cannot generate electron-hole pairs.

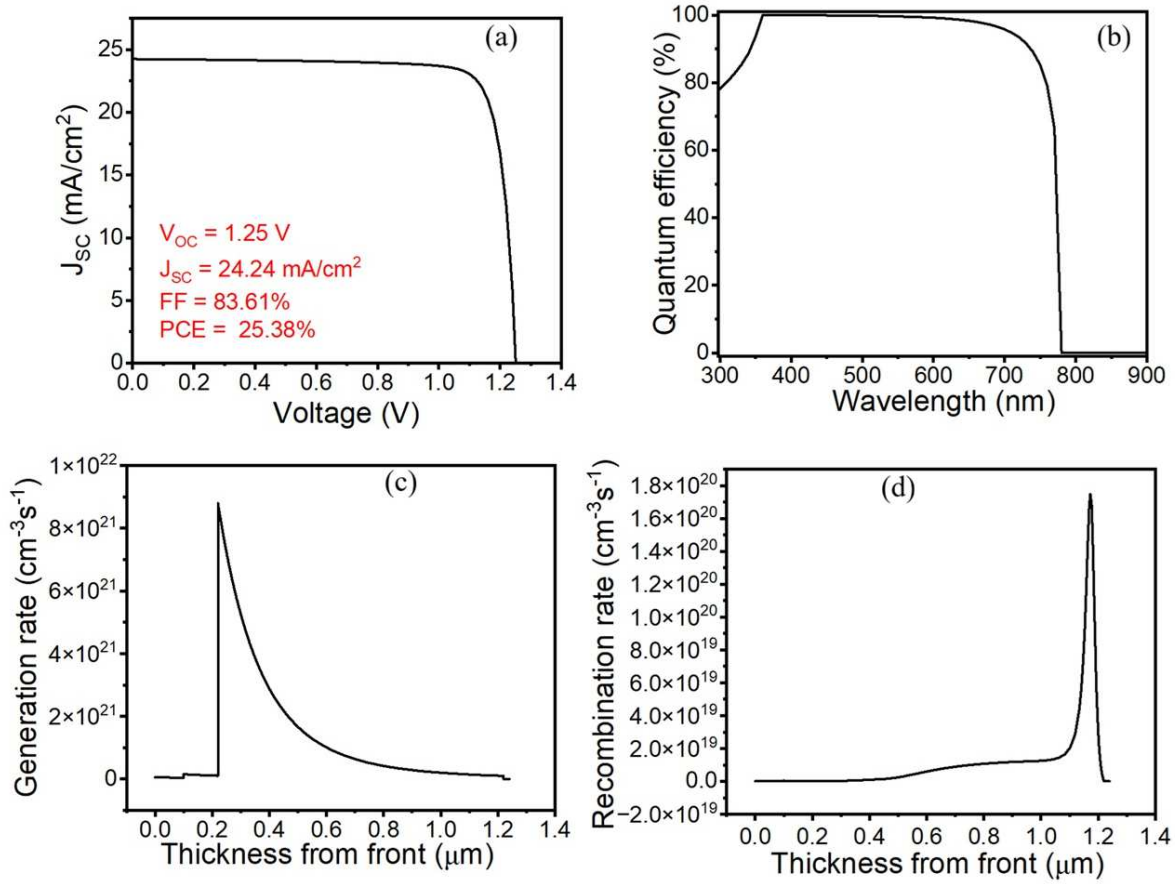


Fig. 13. (a) J-V curve for the optimized device, (b) QE plot, (c) Carrier generation rate, (d) Carrier recombination rate.

Figure 13c shows that carrier generation occurs predominantly within the MAPbI₃ absorber and is strongest near the illuminated TiO₂/MAPbI₃ side. This profile arises because the incident photon flux is greatest near the front of the device and decreases with penetration depth as light is progressively absorbed. The enlarged absorber thickness increases the integrated generation volume, thereby contributing to the enhanced photocurrent.

The recombination profile in Fig. 13d is substantially lower than the generation rate over most of the absorber, confirming that a large fraction of the generated carriers survives long enough to reach the selective contacts. The remaining recombination is concentrated near the heterointerfaces, where electrons and holes may coexist and interact with interfacial trap states. Reducing the bulk and interface defect densities to 10^{14} cm⁻³ and 10^{10} cm⁻², respectively, suppresses trap-assisted Shockley-Read-Hall recombination, increases carrier lifetime and

preserves the quasi-Fermi-level splitting. The predicted PCE of 25.38% is therefore governed not by a single parameter, but by the coordinated improvement of photogeneration, carrier lifetime, transport and collection throughout the device.

4.8 Effect of temperature on optimized device

Fig. 14 show that increasing the operating temperature from 290 to 340 K causes a gradual reduction in the optimized-device efficiency from 25.44 to 24.78%. The dominant loss originates from V_{OC} , which decreases from 1.26 to 1.21 V, while J_{SC} increases only slightly from 24.24 to 24.28 mA/cm² and FF rises from 83.24 to 84.50%.

The reduction in V_{OC} is mainly associated with enhanced thermally activated recombination and increased diode saturation current. At higher temperature, the intrinsic carrier concentration increases and the effective bandgap may narrow slightly, thereby raising the reverse saturation current and reducing the electron–hole quasi-Fermi-level splitting under illumination. This causes the systematic leftward shift of the J–V curves in Fig. 14b and lowers the maximum attainable photovoltage.

The small increase in J_{sc} results from the marginal extension of the absorption edge and enhanced thermal excitation of carriers. However, this gain is very small because MAPbI₃ already absorbs strongly over most of the visible spectrum. The nearly overlapping QE curves in Fig. 14c confirm that temperature has little effect on the spectral collection efficiency within the investigated range.

The moderate increase in FF indicates that the simulated device experiences slightly improved charge transport or reduced effective resistive limitation at elevated temperature. Nevertheless, this improvement is insufficient to compensate for the stronger voltage loss. Consequently, PCE decreases monotonically as temperature rises.

Overall, the optimized device shows relatively good thermal tolerance, with only a 0.66 percentage-point efficiency loss between 290 and 340 K. Even so, the negative temperature coefficient of V_{OC} remains the principal factor controlling the decline in photovoltaic performance, indicating that suppression of thermally activated recombination is essential for maintaining high efficiency under elevated operating temperatures.

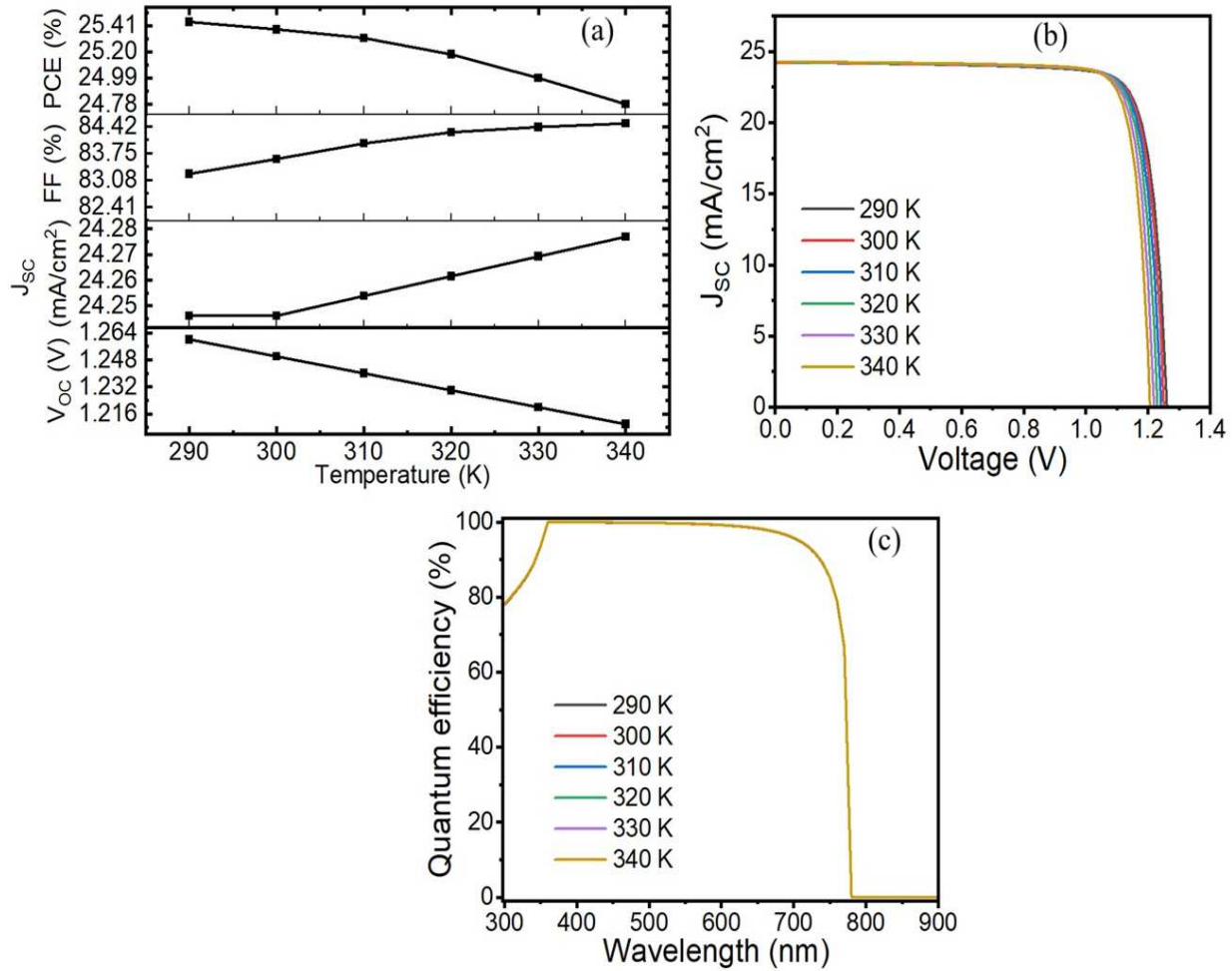


Fig. 14. (a) Dependence of V_{OC} , J_{SC} , FF and PCE on temperature of the device, (b) J-V curve for the effect of temperature, (c) QE plots.

4.9 Comparison with recent experimental and numerical studies

The present results are broadly consistent with recent studies showing that the optimum spin-coating condition for MAPbI₃ is not universal, but depends on the combined rates of wet-film thinning, solvent extraction, intermediate-phase conversion and thermal crystallization. Kılıç et al. [13] recently obtained their optimum ambient-air MAPbI₃ film at 4000 rpm for 40 s followed by annealing at 105 °C for 30 min. Their film showed dominant (110) orientation and no detectable PbI₂ phase. In the present study, the best structural quality occurred at 3000 rpm after a shorter

30s coating cycle and annealing at 110 °C for 10 min. The agreement in phase identity and preferred orientation, but difference in optimum speed, demonstrates that spin speed should be interpreted together with coating time, precursor/solvent system, antisolvent delivery and annealing schedule rather than as an independent universal control variable.

The crystallite size of 22.73 nm obtained at 3000 rpm is lower than the approximately 33.38 nm reported for a fresh, ethyl-acetate-treated ambient MAPbI₃ film by Nur-E-Alam et al. [12]. Their toluene-treated film reached approximately 57.22 nm, further illustrating the strong dependence of coherent-domain size on antisolvent chemistry and processing history. Nevertheless, the present non-monotonic trend is physically meaningful: both 2000 and 5000 rpm produced smaller crystallites and larger microstrain than 3000 rpm. This indicates that either insufficient wet-film thinning at low speed or excessively rapid solvent removal at high speed can hinder ordered crystal growth. The absence of a detectable PbI₂ peak in the present films also agrees with the phase-pure ambient MAPbI₃ reported by Kılıç et al. [13].

The measured optical bandgaps of 1.59–1.60 eV fall within the commonly reported range for polycrystalline MAPbI₃, although they are slightly higher than the 1.57 eV obtained for an ethyl-acetate-processed film by Nur-E-Alam et al. [12] and the 1.53 eV reported by Rabhi et al. [17]. Differences of this magnitude can arise from precursor composition, thickness, strain, phase purity, optical baseline treatment and the method used to construct the Tauc plot. The narrow 0.01 eV spread across the present samples indicates that varying spin speed changed crystallite-related structural quality much more strongly than the intrinsic absorption threshold. Consequently, the small simulated performance difference between the 1.59 and 1.60 eV prototype absorbers should not be interpreted as the complete photovoltaic effect of spin speed, because experimentally induced changes in thickness, coverage, grain boundaries and defect density were held constant in that particular simulation.

The optimized SCAPS-1D efficiency of 25.38% is also credible relative to recent numerical MAPbI₃ studies, but it remains model-dependent. Ali et al. [18] reported 23.32% after optimizing MAPbI₃ and transport-layer thicknesses, incorporating Ti₃C₂ MXene and examining temperature effects. Rabhi et al. [17] predicted 28.39% for an experimentally informed but different AZO/SnO₂/MAPbI₃/D-PBTTT-14/RGO architecture. The present value lies between these reports. Its difference from 23.32% or 28.39% should not be treated as a direct ranking because the studies

used different electrodes, transport materials, band offsets, optical losses, mobilities and defect assumptions. More importantly, the reduction of MAPbI₃ bulk-defect density and interface-defect density in the present optimization explains much of the increase from the 16.67% prototype to 25.38%, confirming that recombination control is at least as important as the small measured bandgap variation.

Overall, comparison with recent literature supports three principal outcomes of this work. First, ambient-air processing can yield phase-pure, preferentially (110)-oriented MAPbI₃ when the deposition and annealing window is carefully controlled. Second, 3000 rpm represents a process-specific optimum for the present formulation and ethyl-acetate protocol rather than a universal spin-speed optimum. Third, the experimentally measured bandgap provides a defensible constraint for SCAPS-1D, while realistic prediction of the full spin-speed effect requires the measured thickness and experimentally supported bulk and interface defect densities to be varied together. These distinctions strengthen the significance of the study by connecting a reproducible ambient deposition window to both measured film quality and bounded device-level performance projections.

Table 7. Comparison of the present results with recent experimental and SCAPS-1D studies of MAPbI₃.

Study	Processing/model distinction	Main reported outcome	Relation to this work
This study	Ambient MAPbI ₃ ; EA antisolvent; 2000–5000 rpm; 110 °C, 10 min	3000 rpm optimum; D = 22.73 nm; E _g = 1.59–1.60 eV; optimized SCAPS PCE = 25.38%	Reference case
Nur-E-Alam <i>et al.</i> (2024) [12]	Ambient MAPbI ₃ ; four antisolvents; ≈300 nm films	EA: D = 33.38 nm and E _g = 1.57 eV; EA gave the lowest degradation among the tested antisolvents	Supports EA use; shows antisolvent-dependent crystallite size and bandgap

Study	Processing/model distinction	Main reported outcome	Relation to this work
Rabhi <i>et al.</i> (2024) [17]	Experimental MAPbI ₃ plus SCAPS; alternative AZO/SnO ₂ /MAPbI ₃ /D-PBTTT-14/RGO stack	Tetragonal MAPbI ₃ ; E _g = 1.53 eV; optimized PCE = 28.39%	Higher predicted PCE reflects different transport layers, contacts and assumptions
Ali <i>et al.</i> (2025) [18]	SCAPS MAPbI ₃ optimization with Ti ₃ C ₂ MXene and layer-thickness/temperature studies	Optimized PCE = 23.32%	Present 25.38% lies within the range of recent optimized simulations
Kılıç <i>et al.</i> (2026) [13]	Ambient one-step MAPbI ₃ ; 4000 rpm, 40 s; 105 °C, 30 min	Dominant (110) orientation; no detectable PbI ₂	Confirms ambient feasibility; different optimum demonstrates process dependence

5. Conclusion

This study established a process-specific relationship between spin-coating speed and the structural and optical properties of open-air-processed MAPbI₃ thin films. X-ray diffraction confirmed tetragonal MAPbI₃ with preferential (110) orientation and no detectable PbI₂ residue across the investigated conditions. The response to spin speed was non-monotonic: deposition at 3000 rpm produced the largest crystallite size (22.73 nm), lowest microstrain (1.257×10^{-2}) and lowest dislocation density (1.936×10^{-3} lines nm⁻²), whereas both lower and higher speeds yielded greater structural disorder. Thus, 3000 rpm provided the most favourable balance between wet-film thinning, solvent removal, nucleation and crystal growth for the present precursor formulation, ethyl-acetate treatment and annealing protocol. The optical bandgaps remained within a narrow range of 1.59–1.60 eV, indicating that spin speed affected crystalline quality more strongly than the fundamental absorption threshold.

When the measured bandgaps were introduced into the FTO/TiO₂/MAPbI₃/Spiro-OMeTAD/carbon SCAPS-1D model, the 1.59 eV prototype produced V_{OC} , J_{SC} , FF, and PCE of 1.10 V, 21.14 mA cm⁻², 71.85%, and 16.67%, respectively, only slightly exceeding the 16.47% obtained with the 1.60 eV bandgap absorbers. This small difference confirms that bandgap variation alone does not represent the full photovoltaic effect of spin speed. The parametric analysis instead identified absorber thickness, transport-layer carrier density, bulk and interface defect densities, and parasitic resistances as the dominant performance controls. Their coordinated optimization produced an optimized device with V_{OC} of 1.25 V, J_{SC} of 24.24 mAcm⁻², FF of 83.61% and PCE of 25.38%. Because these values are numerical projections based on optimized inputs, they should not be interpreted as experimentally demonstrated device performance.

Competing Interests

The authors have no relevant financial or non-financial interests to disclose

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Data Availability

The data for this work will be made available on request.

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