

Supporting information

Closing the Loop: Circular Recycling of MAPbI₃ Perovskite Solar Cells

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1 Experimental Procedures

1.1 Materials

Indium tin oxide (ITO)/glass substrates were purchased from Advanced Election Technology Co., Ltd. SnO₂ 15% in H₂O colloidal dispersion was purchased from Alfa Aesar. Methylammonium iodide (MAI, 99.5%) and Spiro-OMeTAD (2-N,2-N',2-N',2-N',7-N,7-N',7-N',7-N'-octakis(4-methoxyphenyl)-9,9'-spirobi[fluorene]-2,2',7,7'-tetramine, 99.5%) were purchased from Xi'an Yuri Solar Co., Ltd (former name: Xi'an Polymer Light Technology Corp., Ltd). Lead iodide (PbI₂, 99.99%) were purchased from TCL Development Co., Ltd. Ethanol (>98%, technical, denatured) was purchased from Carl Roth. Chlorobenzene (anhydrous, 99.8%), isopropanol (ACS reagent), ethyl acetate (anhydrous, 99.8%), N-Methyl-2-Pyrrolidone (NMP, anhydrous, 99.5%), 4-tert-butylpyridine (tBP), N,N-dimethylformamide (DMF, anhydrous, 98%), γ -Butyrolactone (GBL, 99%), Dichloromethane (DCM), lithium bis(trifluoromethylsulphonyl)imide (Li-TFSI, 99.95%), tris(2-(1Hpyrazol-1-yl)-4-tert-butylpyridine)cobalt(III) bis(tri-fluoromethylsulphonyl)imide (FK209 Co (III) TFSI salt) and acetonitrile (anhydrous, 99.8%) were purchased from Sigma Aldrich. Silica gel (60A, 40-63u) was purchased from Fluorochem. Deionized water was obtained with an in-house filter. Acetone (\geq 99%, technical) and isopropanol (\geq 98%, technical) for cleaning was purchased from VMR.

1.2 Solution preparation

SnO₂ solution was prepared by dispersing 1 mL SnO₂ colloidal dispersion into 5 mL mixture of deionized water and isopropanol ($V_{\text{water}}:V_{\text{isopropanol}} = 1:1$) to achieve a diluted 2.5% concentration. 1.5 M fresh MAPbI₃ solution was prepared by mixing 1.5 mmol MAI and 1.5 mmol PbI₂ in 1 mL mixture of DMF and NMP ($V_{\text{DMF}}:V_{\text{NMP}} = 4:1$). The solution is then stirred for 1 hour at 70 °C. 1.5 M recycled MAPbI₃ solution was prepared by dissolving 1.5 mmol recycled MAPbI₃ in 1 mL of mixed DMF and NMP ($V_{\text{DMF}}:V_{\text{NMP}} = 4:1$), which was also stirred for 1 hour at 70 °C. To prepare Spiro-OMeTAD solution, 72.3 mg Spiro-OMeTAD was dissolved into 1 mL chlorobenzene, followed by adding 17.5 μ L Li-TFSI solution (520 mg/mL in acetonitrile), 28.8 μ L tBP and 28.8 μ L FK209 Co (III) TFSI solution (300 mg/mL in acetonitrile) in sequence.

1.3 Device fabrication

ITO/glass substrates were sonicated in acetone and then isopropanol for 20 min each, followed by drying with a N₂ stream and then 5 min O₂ plasma treatment. SnO₂ solution was filtered with a nylon filter (pore size: 0.20 μ m) and sonicated for a few seconds. 80 μ L SnO₂ solution was spin-coated at 4000 rpm for 20 s onto an ITO/glass substrate or a recovered ITO/SnO₂ sample from old devices in air

and then annealed at 150 °C for 20 min. After electron transport layer deposition (SnO₂ deposition), the substrates were transferred to a N₂-filled glovebox for the following layers. Perovskite solutions were filtered through 0.20 μm polytetrafluoroethylene (PTFE) filters before use. Subsequently, 50 μL perovskite solution was spun onto ITO/SnO₂ substrates at 1000 rpm for 10 s, and then 4000 rpm for 40 s. During the second step, 300 μL chlorobenzene was dropped continuously onto the film at 30 s, followed by annealing at 110 °C for 10 min. Then ITO/SnO₂/perovskite substrate was coated with a layer of Spiro-OMeTAD (50 μL) at 5000 rpm for 40 s with no post-annealing. This layer was coated dynamically – Spiro-OMeTAD was cast on the sample when the spin coater was already on. Subsequently, the samples coated with Spiro-OMeTAD were stored overnight in low humidity conditions for the oxidation of Spiro-OMeTAD to take place. Lastly, a layer of Au electrode was deposited on the sample via thermal evaporation. The effective device area is 0.063 cm². For devices made with recycled components, all the parameters are the same as for devices made with virgin components.

1.4 Recycling

Spiro-OMeTAD

Big pseudo-modules (125 mm*85 mm) were prepared for the recycling of Spiro-OMeTAD. The structure of pseudo-module is ITO/SnO₂/MAPbI₃/ Spiro-OMeTAD. Au deposition was excluded for pseudo-modules. SnO₂, MAPbI₃ and Spiro-OMeTAD were deposited via drop-casting or doctor blade coating. Spiro-OMeTAD was dissolved with chlorobenzene. Then flash column chromatography was carried out to purify the harvested Spiro-OMeTAD solution (the eluent (DCM: EA=19:1) was selected via thin layer chromatography). We employed 20 ml syringes as chromatography columns, using approximately 6 g of silica gel, to separate around 100-200 mg of Spiro-OMeTAD (estimated quantity). We then concentrated Spiro-OMeTAD with a rotary evaporator, followed by drying in vacuum at room temperature. Additional FK209 Co (III) TFSI, Li-TFSI and tBP were added to prepare Spiro-OMeTAD solution for cell fabrication.

MAPbI₃

MAPbI₃ was collected from pseudo-modules as well. To dissolve MAPbI₃, we used pipettes to apply a minimal amount of GBL onto the modules, which were positioned almost vertically and placed in a petri dish. Ethyl acetate, as the anti-solvent, was added into the obtained solution till the precipitates no longer showed significant increase (the amount of ethyl acetate was roughly 3 times as high as that of GBL). It should be noted that the whole process was done in a glovebox to minimize contact with air and thus iodine loss. Also, it would be better to use oxygen-free ethyl acetate. Afterward, recycled MAPbI₃ was collected via centrifugation at 5000 rpm for 3 min, followed by drying in vacuum for at least 24 hours at room temperature.

ITO/SnO₂ and Au

ITO/SnO₂ and Au were harvested from old functional devices (25mm*25mm) stored in a glovebox. Spiro-OMeTAD was dissolved by swirling the device in chlorobenzene or ethyl acetate for about 5 min. Then the sample was swirled in GBL for 2 min to remove MAPbI₃. Au was fallen into the solution and naturally separated from the substrate. The gold flakes were collected via filtration. The obtained ITO/SnO₂ substrate was washed with ethanol and dried with a gentle N₂ stream.

1.5 Characterization

The current-voltage characteristics of the solar cells were measured using a Keithley 7001 Switch System and a Keysight B2901A Precision Source/Measure Unit. The Keithley 7001 facilitated the selection and switching between different cells on a sample automatically. Meanwhile, the Keysight B2901A was employed to apply a controlled voltage to the cells and measure the resultant currents. The measurements were carried out under illumination of 100 mW cm^{-2} AM1.5 G (Wavelabs Sinus-70 light engine). Light intensity was calibrated with a crystalline Si cell. The current voltage characteristics were scanned from 1.2 to -0.6 V (reverse scan), with a scan step of 40 mV.

UV-Vis spectra were acquired with a PerkinElmer lambda 950, using a 150 mm integrating sphere. The absorbance spectra of Spiro-OMeTAD were recorded in the wavelength range of 300–600 nm, while the transmittance spectra of MAPbI₃ were measured from 400 to 850 nm. Fresh and recycled Spiro-OMeTAD HTM solutions, doped with additives, were statically spin-coated on ITO glasses at 4000 rpm for 30s, followed by annealing at 75 °C for 5 min. Both fresh and recycled MAPbI₃ films were spin-coated on common glass substrates using the same parameters as employed for MAPbI₃ layers in solar cell devices (detailed in the Device fabrication section).

X-ray diffraction (XRD) measurements were carried out with classical ex situ Bragg-Brentano geometry using a PANalytical X'pert powder diffractometer with filtered Cu-K α radiation ($\lambda = 1.54178 \text{ \AA}$) and an X'Celerator solid-state stripe detector operated at 40 kV and 30 mA. The XRD patterns were collected from 10 to 50°, with a step size of 0.0394°. The miller indices of MAPbI₃ were assigned using the software Mercury based on a crystallographic information file (CIF) downloaded from the Crystallography Open Database (COD) (<http://www.crystallography.net/cod>). The COD ID is 4124388. Both fresh and recycled MAPbI₃ films were prepared following the same procedures employed for MAPbI₃ films intended for UV-Vis measurements.

Time-integrated photoluminescence and time-resolved photoluminescence spectra were recorded with PicoQuant Fluotime 300 at room temperature and in air. The time-integrated photoluminescence spectra were recorded from 450 to 900 nm. A diode laser (LDH-P-C-405B) was employed to excite the samples at 402.2 nm, using a repetition rate of 0.2 MHz and a FWHM of 50 ps. The laser beam area is $7540 \text{ }\mu\text{m}^2$. The exciton carrier density for both the fresh and recycled MAPbI₃ film is $4.040\text{E}+22 \text{ e/cm}^3$. To mitigate the effects of photo-brightening and ensure reliable results, the measurement protocol included performing three replicates to achieve overlapping spectra. If overlap was not observed, the method necessitated shifting to an unexposed spot and reducing the laser intensity to secure three matching spectra. Figure S14 shows the TR-PL spectra for fresh and recycled MAPbI₃ measured three times consecutively at the same spot. The spectrum presented in the main article is one of the overlapped spectra. Casting procedures for both fresh and recycled MAPbI₃ films mirrored those utilized for MAPbI₃ designated for UV-Vis measurements. The MAPbI₃ films were coated on common glasses.

Scanning electron microscope images were taken at 15 kV acceleration voltage using a JEOL JSM-7610F Schottky Field Emission Scanning Electron Microscope, with a SEI detector and a working distance around 6 mm. The ITO/SnO₂ glass samples were mounted on a specimen stage with carbon tapes and silver pastes.

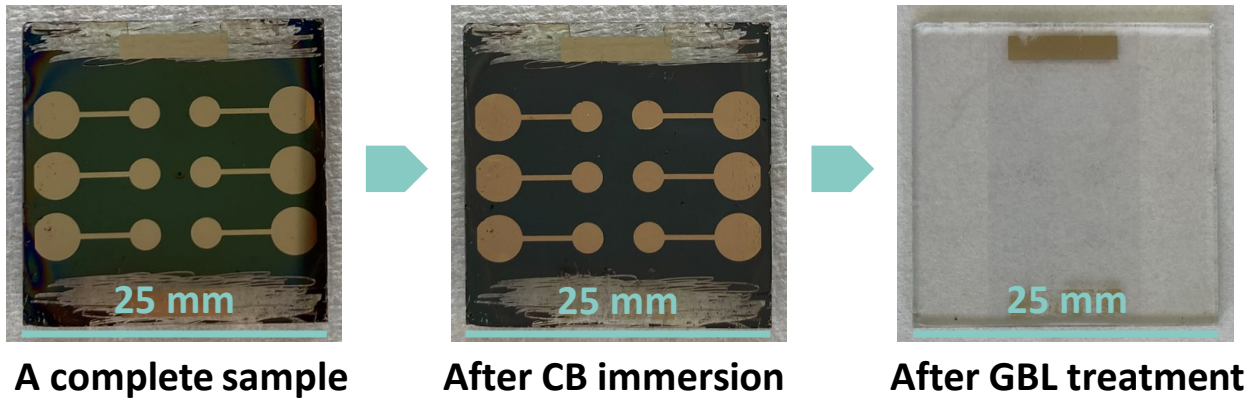
Optical images of solar cells were captured using a Zeiss Axio Imager.M1 microscope at a 2.5x magnification.

1.6 Notes

Language models, ChatGPT and Claude, were used when drafting this manuscript to improve readability and language.

Table S1: Summary of previous reports. CB: Chlorobenzene; EA: ethyl acetate; in the ‘Recycled Layer and Ratio’ column, the numerators in fractions represent the count of recycled layers, and the denominators indicate the total number of layers in the solar device; %: the metrics (PCE, J_{sc} , V_{oc} and FF) of a device with recycled materials divided by the same metrics of a device with fresh materials (reference); savings: reduced cost of a recycled device/the cost of the reference; recycled mass (%): recycled mass/material on stack.

Recycling method	Recycled Layer and Ratio	PCE (%)	J_{sc} (%)	V_{oc} (%)	FF (%)	Savings (%)	Recycled mass (%)	Ref
DMF/GBL/DMSO » rinsing	FTO glass, mpTiO ₂ ; 2/5	99.56	100.35	101.98	97.14	N/A	N/A	[1]
CB » EtOH » DMF » EtOH washing	FTO glass, TiO ₂ ; 2/5	99.38	89.14	99.07	108.96	N/A	N/A	[2]
DMF » CB » washing	FTO glass; 1/4	91.46	96.16	96.94	97.06	N/A	N/A	[3]
Tape » DMSO/DMF » washing	SnO ₂ , ITO glass; 2/6	100	100.40	97.37	102.75	N/A	N/A	[4]
Tape » CB » heat » in situ MAI solution coating	FTO, TiO ₂ , PbI ₂ ; 2.5/5	103.41	101.31	98.99	103.12	N/A	N/A	[5]
Heat » DMF » ion exchange resin for Pb » Pb ²⁺ + I ⁻ » PbI ₂	ITO glass, back glass; 2/8	~95	N/A	N/A	N/A	N/A	N/A (material cost of a virgin module and recycling cost are provided)	[6]
	Pb; 0.5/8	97.14	N/A	N/A	N/A			
DMF » NH ₃ ·H ₂ O » HI » PbI ₂	FTO glass, c-TiO ₂ , m-TiO ₂ ; 3/5	98.5	104.19	0.98	96.84	N/A	N/A	[7]
	Pb; 0.5/5	93.34	98.32	94.57	99.95			
Tape » CB » H ₂ O » cooling crystallization for PbI ₂	FTO glass; 1/5	103.4	102.08	101.94	98.63	N/A	N/A	[8]
	PbI ₂ ; 0.5/5	92.47	96.39	99.03	94.59			
Butylamine » heat » toluene » EtOH » PbI ₂ crystallization to MAPbI ₃ crystals	ITO glass, NiO _x , PbI ₂ ; 2.5/5	100.62	100.33	100	99.10	N/A	N/A	[9]
Methylamine + THF » cleaning for ITO/SnO ₂ and Au » Au melting » ACN for MAPbI _{3-x} Br _x » EA extraction for Spiro-OMeTAD	ITO glass, SnO ₂ , MAPbI _{3-x} Br _x , Spiro-OMeTAD and Au; 5/5	99.04	99.56	99.74	99.63	N/A	N/A	[10]
CB » GBL » EtOH washing » chromatography for Spiro-OMeTAD » anti-solvent crystallization for MAPbI ₃ » fresh SnO ₂ for ITO/SnO ₂	ITO glass, SnO ₂ , MAPbI ₃ , Spiro-OMeTAD ; 4/5	97.16	97.21	99.08	101.33	63.7 at lab scale; 92.4 (Au), 61.4 (Ag) at industrial scale	99.97	This work



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Figure S1: Photographs of a sample at different stages of recycling. CB: chlorobenzene. The Au stripes at the sides of a recovered or a recycled ITO/SnO₂ were scraped off before thermal evaporation as for a sample with a fresh substrate.

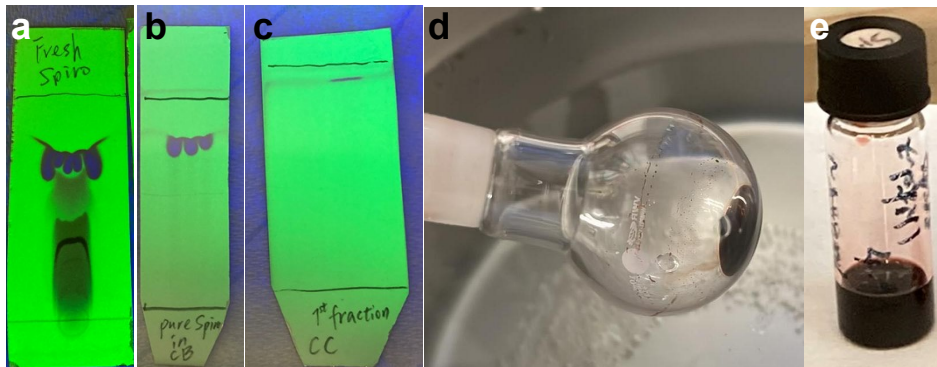


Figure S2: Thin-layer chromatography plates for a) fresh Spiro-OMeTAD doped with additives, b) pure Spiro-OMeTAD and c) column purified recycled Spiro-OMeTAD; d) recycled Spiro-OMeTAD after rotary evaporation; e) fresh Spiro-OMeTAD solution.

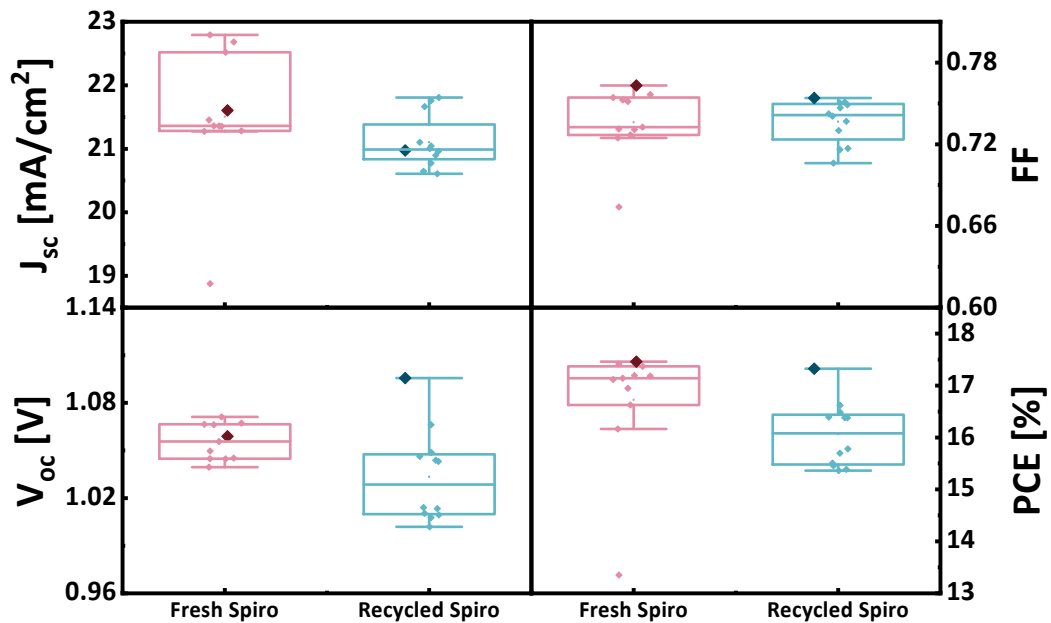


Figure S3 : J_{sc} , V_{oc} , FF and PCE of 12 solar cells using fresh Spiro-OMeTAD versus using recycled Spiro-OMeTAD. The magnified data points in a darker hue are the champion devices shown in the main article.

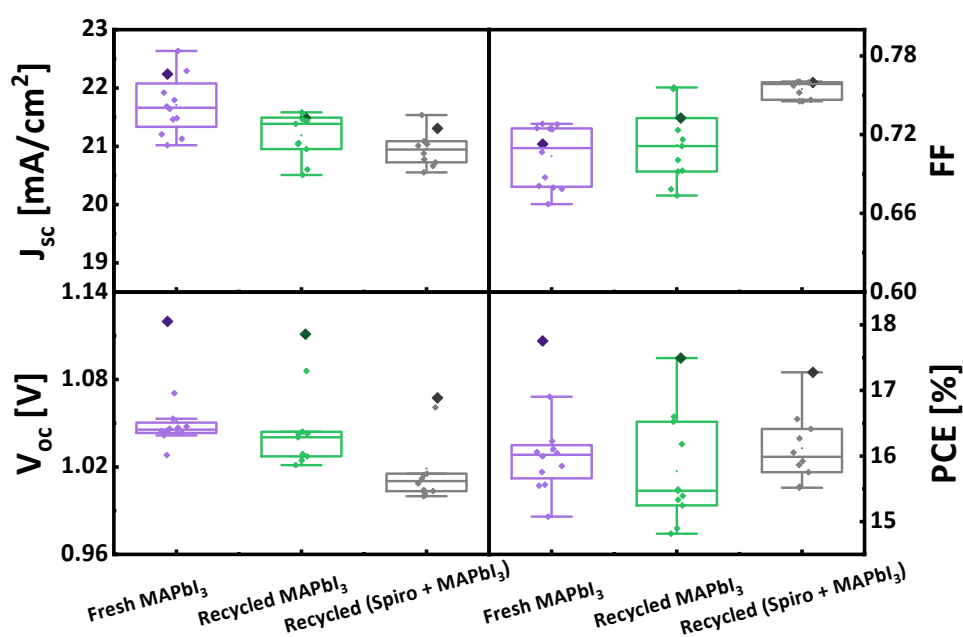


Figure S4 : J_{sc} , V_{oc} , FF and PCE of 12 solar cells made with fresh MAPbI_3 versus recycled MAPbI_3 versus recycled MAPbI_3 and Spiro-OMeTAD. The magnified data points in a darker hue are the champion devices shown in the main article.

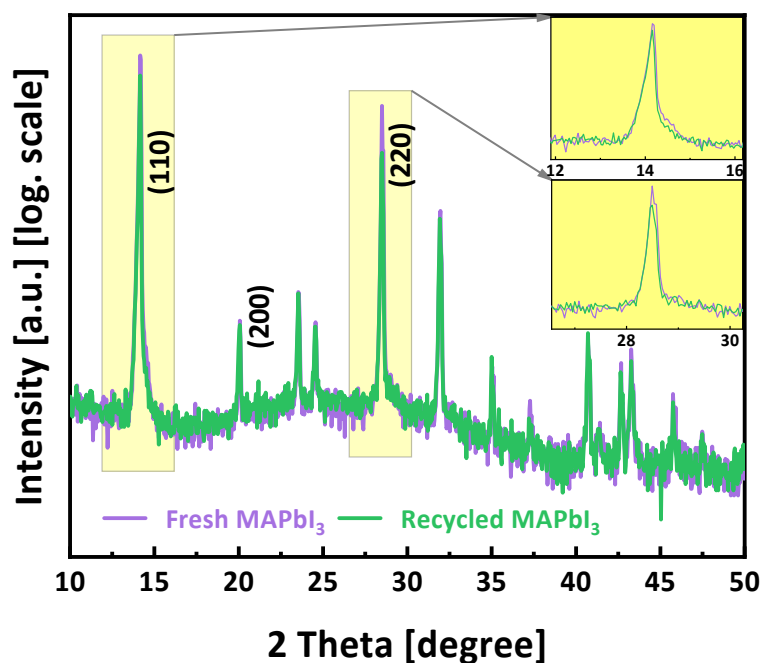


Figure S5: Superimposed XRD patterns of fresh and recycled MAPbI_3 films on common glass, with insets highlighting zoomed-in peak comparisons.

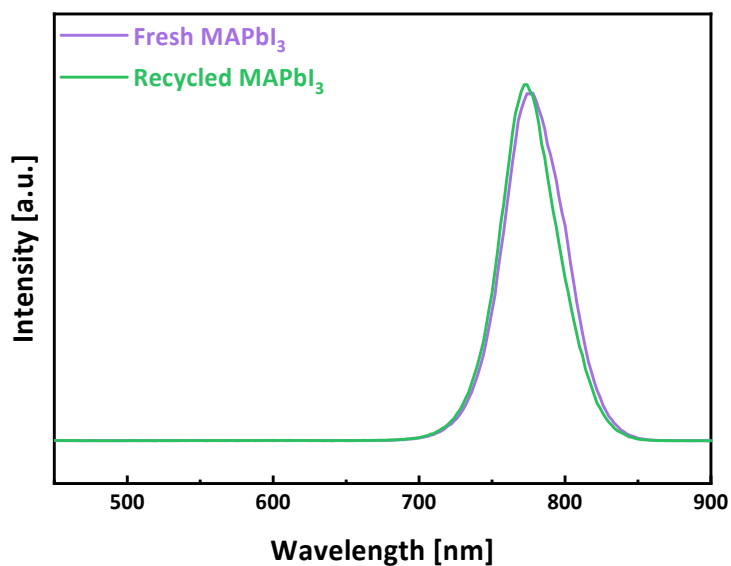


Figure S6: TI-PL spectra of fresh and recycled MAPbI_3 films on common glass.

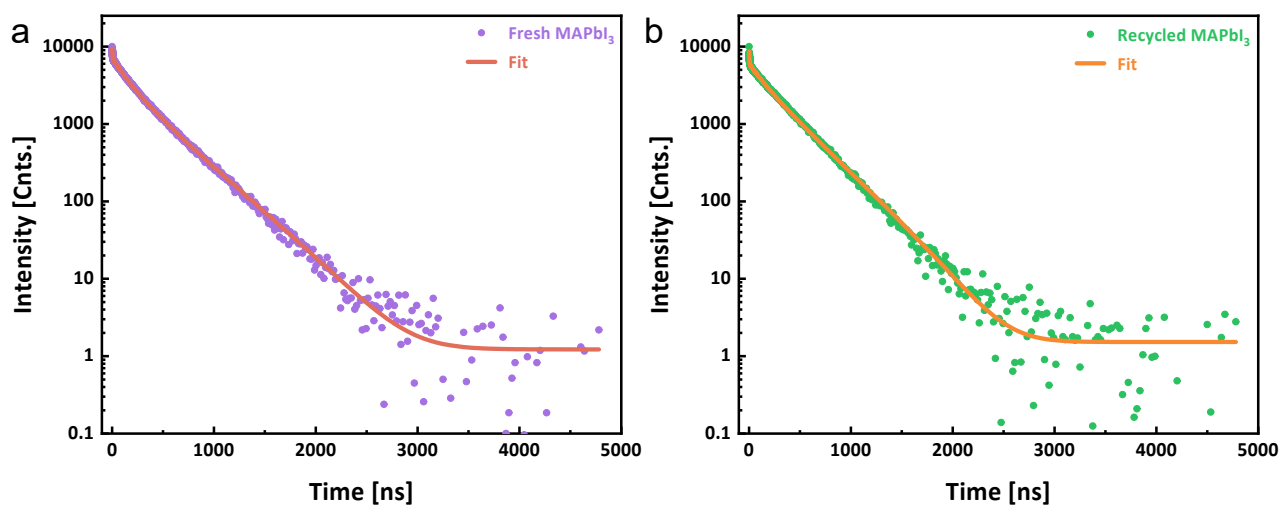


Figure S7 : Fitted TR-PL spectra for fresh and recycled MAPbI_3 films.

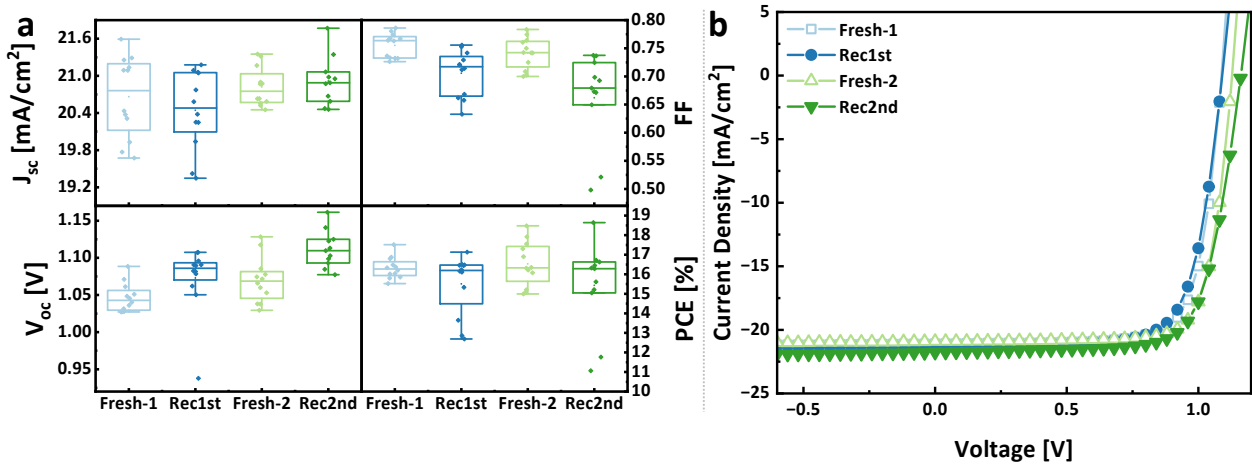


Figure S8: Two-cycle recycling of ITO/SnO₂: a) J_{sc} , V_{oc} , FF and PCE of 12 solar cells made with ITO/SnO₂ recycled once (Rec1st) versus ITO/SnO₂ recycled twice (Rec2nd) versus fresh materials (Fresh-1 is for comparison with Rec1st — both made in the same batch, Fresh-2 is for comparison with Rec2nd); b) JV curves of the champion devices.

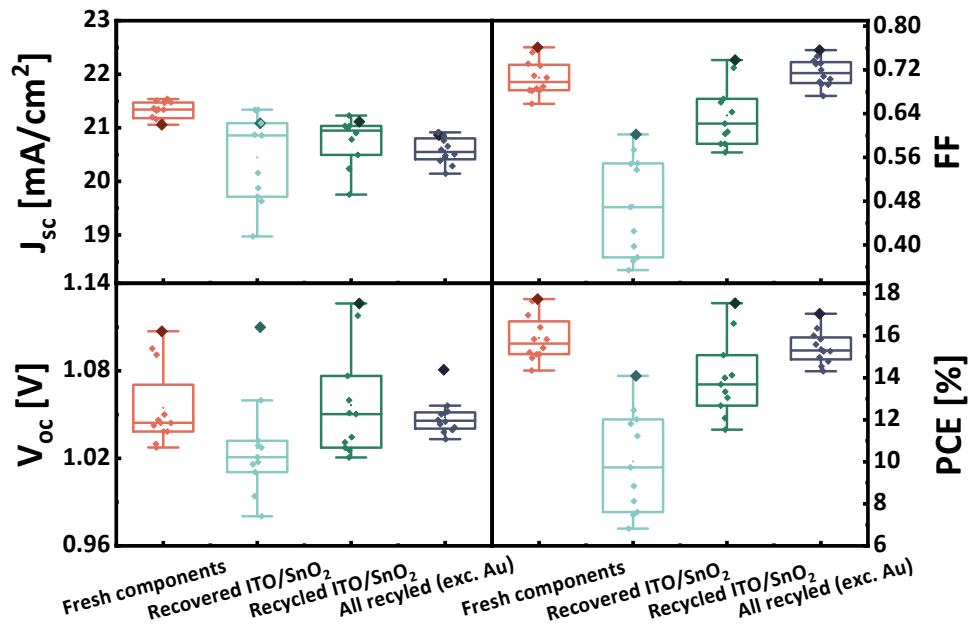


Figure S9 : J_{sc} , V_{oc} , FF and PCE of 12 solar cells made with fresh components exclusively versus recovered ITO/SnO₂ versus recycled ITO/SnO₂ (recovered ITO/SnO₂ deposited with fresh SnO₂) versus recycled MAPbI₃, Spiro-OMeTAD and ITO/SnO₂. The magnified data points in a darker hue are the champion devices shown in the main article.

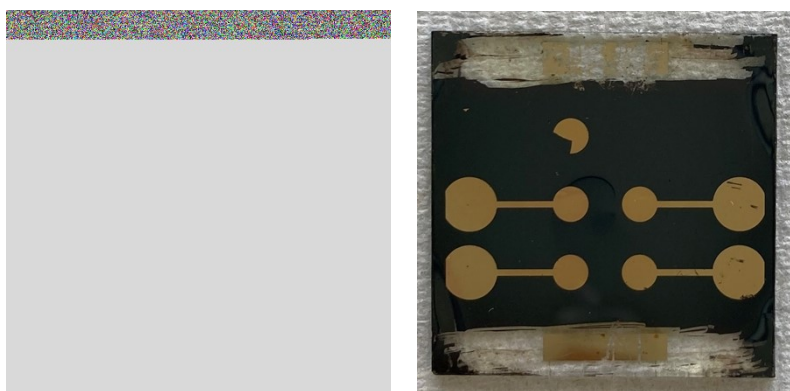


Figure S10: Two perovskite solar samples after chlorobenzene treatment.

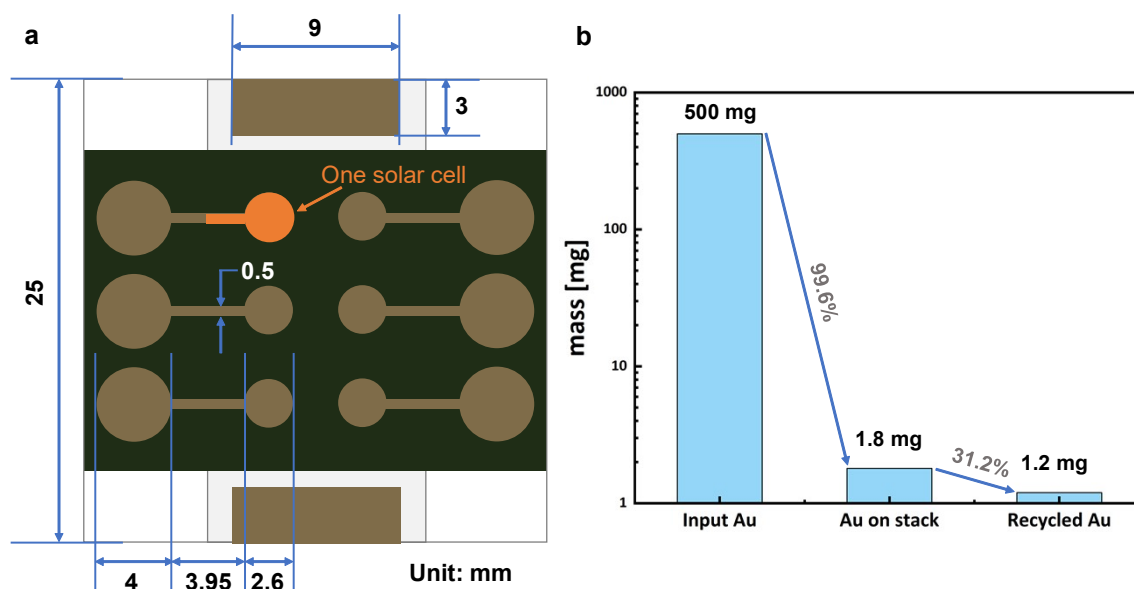


Figure S11: a) Designed dimensions of a perovskite solar sample consisting of six individual cells (the area highlighted in orange denotes a single solar cell); b) Mass flow of gold per batch (18 samples), from input to recycling.

Table S2: The lab-scale cost comparison between a 1 m² hybrid module and a fresh one.

Layer	COST (\$)	
	Fresh [C_{VL}]	Hybrid [C_{HL}]
ITO glass	244.30	0.37
SnO ₂	0.17	0.17
MAPbI ₃	7.62	6.5
Spiro-OMeTAD	79.01	46.8
Au	104.02	104.02
Combined	435.12	157.86

The cost of hybrid MAPbI₃ layer and hybrid Spiro-OMeTAD layer can be deduced by:

$$C_{HL} = C_{VL} - (C_{Vc} - C_{Hc}) \quad (1)$$

where C_{HL} is the cost of a hybrid layer, C_{VL} is the cost of a virgin layer, C_{Vc} is the cost of virgin component, and C_{Hc} is the total cost of one component for a hybrid module. C_{Vc} and C_{VL} can be found in Table 2.

Table S3: Material expenses, generated waste and waste management costs in our labs in Erlangen, 2021 – 2022.

Total estimated researchers focusing on experiments		50			
Lab	i-MEET		HTM PV		
Year	2021	2022	2021	2022	
Expenses on TCO glasses	3,745.2 €	4,258 €	1,260 €	7,214.0 €	
Total expenses for materials	60,248.7 €	51,891.5 €	53,268.0 €	55,459.5 €	
Amount of generated waste	900 L	1290 L	371 kg	456 kg	
Expenses for waste management	N/A	N/A	1,757.2 €	2001,4 €	

Table S4: Material price estimates for purchase quantities required when scaling up solar panel production ten thousand times. The purchase volume here is the closest to the required volume available, while maintaining the material grades identical to those used in our experiments. A 10% price learning was adopted based on literature [11], and accounts for the material price estimates. As for Au and Ag, the cost reduction for every doubling is not practical due to their unique value. Spiro-OMeTAD is a highly expensive HTM in the current market, and thus a more sustainable and cheaper synthesis is assumed to be utilized instead [12].

Material	Usage for 1 m ²	unit	purchase volume	price (\$)	price per unit (\$/unit)	required volume for 10000 m ²	# Doublings	calculated price per unit (\$/unit) [P_V]	source
ITO glass	1	m ²	\	\	\	10000	\	7 (front glass) + 1.30 (ITO)	[13]
Deionized water	33	mL	\	\	\	330000	\	0.000012	[14]
Acetone	33	mL	200000	1436.4	0.0072	330000	0.73	0.0067	VMR
Isopropanol	33	mL	10000	85.17	0.0085	330000	5.05	0.0051	VMR
SnO ₂ (15 %)	0.58	g	2000	259.2	0.13	5800	1.54	0.11	VMR
Deionized water	1.25	mL	\	\	\	12500	\	0.000012	[14]
Isopropanol	1.25	mL	20000	574.56	0.029	12500	\	0.029	Sigma
MAI	0.14	g	10	75	7.5	1400	7.13	3.54	Xi'an Yuri
PbI ₂	1.38	g	25	58	2.32	13800	9.11	0.89	Xi'an Yuri
DMF	2.4	mL	45000	2106	0.047	24000	\	0.047	Sigma
NMP	0.6	mL	18000	509.76	0.028	6000	\	0.028	Sigma
Spiro-OMeTAD (alt. synt.)	0.774	g	1	31.76	31.76	7740	12.92	8.14	[12]
LITFSI	0.097	g	250	1123.2	4.49	970	1.96	3.66	Sigma
FK 209 Co (III) TFSI	0.092	g	500	5318.1	10.64	920	0.88	9.69	Greatcellsolar
Acetonitrile	0.495	mL	2000	264.6	0.13	4950	1.31	0.12	Sigma
Chlorobenzene	10.7	mL	1000	246.24	0.25	107000	6.75	0.12	Sigma
tBP	0.284	g	500	816.48	1.63	2840	2.51	1.25	Sigma
Au	1.65	g	1	63.04	\	\	\	63.04	Market price
Ag	2.5 [13]	g	1	0.73	\	\	\	0.73	Market price
Ethanol	33	mL	200000	970.92	0.0049	330000	0.73	0.0045	Carl Roth
SnO ₂ (15 %)	3.32	g	2000	259.2	0.13	33206	4.06	0.0855	VMR
Deionized water	7.21	mL	\	\	\	72100	\	0.000012	[14]
Isopropanol	7.21	mL	20000	574.56	0.029	72100	1.85	0.024	Sigma
GBL	3.05	g	20000	662.04	0.033	30500	0.61	0.031	Sigma
Ethyl acetate	8.2	mL	2000	240.84	0.12	82000	5.36	0.069	Sigma
Chlorobenzene	1.9	mL	1000	246.24	0.25	19000	4.25	0.16	Sigma
Silica gel	14	g	20000	698	0.035	140000	2.81	0.026	Goldleaf Scientific
DCM	133	mL	205000	2308.8	0.011	1330000	2.7	0.0085	Chem Impex
Ethyl acetate	7	mL	20000	629.64	0.031	70000	1.81	0.0261	Sigma

Table S5: Material costs to prepare a 1 m² solar panel based on the price estimates in Table S3.

Layer	Material	Usage for 1 m ²	unit	calculated price per unit (\$/unit) [P_{Vj}]	Subtotal cost (\$) [C_{Vc}]	Total cost per layer (\$) [C_{VL}]
ITO glass	ITO glass	1	m ²	7 (front glass) + 1.30 (ITO)	8.3	8.69
	Deionized water	33	mL	0.000012	0.0004	
	Acetone	33	mL	0.0067	0.22	
	Isopropanol	33	mL	0.0051	0.17	
SnO ₂	SnO ₂ (15 %)	0.58	g	0.11	0.064	0.10
	Deionized water	1.25	mL	0.000012	0.00002	
	Isopropanol	1.25	mL	0.029	0.036	
MAPbI ₃	MAI	0.14	g	3.54	0.50	1.85
	PbI ₂	1.38	g	0.89	1.23	
	DMF	2.4	mL	0.047	0.11	
	NMP	0.6	mL	0.028	0.017	
Spiro-OMeTAD	Spiro-OMeTAD (alt. synt.)	0.774	g	8.14	6.30	9.26
	LiTFSI	0.097	g	3.66	0.35	
	FK 209 Co (III) TFSI	0.092	g	9.69	0.89	
	Acetonitrile	0.495	mL	0.12	0.057	
	Chlorobenzene	10.7	mL	0.12	1.29	
	tBP	0.284	g	1.25	0.36	
Au	Au	1.65	g	63.04	104	104
Ag	Ag	2.5 [14]	g	0.73	1.83	1.83

Table S6: Recycling costs for a 1 m² solar panel based on price estimates in Table S3. At an industrial scale, the ample availability of recovered metal electrodes eliminates the restriction of reuse from the limited lab-scale recovery. After the recovery process, the electrodes should be subjected to a sequential cleaning procedure involving chlorobenzene, GBL, and ethanol to remove any residual HTM and perovskite. The quantities of chlorobenzene and GBL used are assumed to be consistent with those required for the respective layer recycling processes. Ethanol is employed as the final cleaning agent, and a volume of 10 mL should be sufficient for cleaning 1-2 grams of electrodes. For recycling at this scale, the likelihood of the utilized solvents being recycled and reused is high, thus we include a scenario where the solvents for recycling are recycled and reused with a reuse rate of 90%, and replaced with fresh solvents every 5 m². The recycled solvents include ethanol, GBL, ethyl acetate, chlorobenzene, and dichloromethane.

Target material	Chemical	Unit	Calculated price per unit (\$/unit) [P_V]	Usage [U]	Subtotal cost (\$)	Total cost (\$) [C_R]	Usage [U_{SR}]	Subtotal cost (\$) if sol rec [C_{SR}]	Total cost (\$) if sol rec [C_R]
SnO₂ + ITO glass	Ethanol	mL	0.0045	33	0.15	0.25	9.24	0.042	0.14
	SnO ₂ (15%)	g	0.11	0.58	0.064		0.58	0.064	
	Deionized water	mL	0.000012	1.25	0.00002		1.25	0.00002	
	Isopropanol	mL	0.029	1.25	0.036		1.25	0.036	
MAPbI₃	GBL	g	0.031	3.05	0.095	0.66	0.854	0.027	0.18
	Ethyl acetate	mL	0.069	8.2	0.56		2.296	0.16	
Spiro-OMeTAD	Chlorobenzene	mL	0.16	1.9	0.30	1.97	0.532	0.084	0.82
	Silica gel	g	0.026	14	0.36		14	0.36	
	Dichloromethane	mL	0.0085	133	1.13		37.24	0.32	
	Ethyl acetate	mL	0.026	7	0.18		1.96	0.051	
Au/Ag	Chlorobenzene	mL	0.16	1.9	0.30	0.44	0.532	0.084	0.12
	GBL	g	0.031	3.05	0.095		0.854	0.027	
	Ethanol	mL	0.0045	10	0.045		2.8	0.013	

sol rec: solvents are recycled.

When a solvent is recycled, the cost of this solvent required for 1 m² (i.e. Subtotal cost (\$) if sol rec in the Table) can be deduced by:

$$C_{SR} = U_{SR} * P_V = [(U + U * 0.1 * 4)/5] * P_V \quad (2)$$

where C_{SR} is the cost when the solvent is recycled, U_{SR} is the usage of a chemical with solvents recycling, U is the usage of a chemical without solvents recycling, P_V is the calculated price per unit. U_{SR} , U and P_V can be found in Table S5.

Table S7: The industrial-scale cost comparison between a 1 m² hybrid module and a fresh one.

Layer	COST		
	Fresh [C_{VL}]	Hybrid [C_{HL}]	Hybrid-sol rec [C_{HL}]
ITO glass	8.69	0.15	0.042
SnO ₂	0.1	0.1	0.1
MAPbI ₃	1.85	2.25	1.77
Spiro-OMeTAD	9.26	7.49	6.33
Au	104.02	1.49	1.17
Combined (Au as electrode)	123.92	11.48	9.412
Ag	1.83	0.46	0.14
Combined (Ag as electrode)	21.73	10.45	8.382

The costs of hybrid layers at an industrial scale were calculated in the same way as for the lab scale, with the exception of metal electrodes. We assume a gold recycling rate of 99% for both thermal evaporation losses and solar panels. We applied the same rules for Ag. Thus, the cost of a hybrid electrode is deduced by:

$$C_{He} = C_R + [M_o + M_o * 0.01 * (10000 - 1)] * P_V / 10000 \quad (3)$$

Where C_R is available in Table S5, while M_o and P_V for Au and Ag can be found in Table S3.

Table S8: Material on a 1 m² module (it is assumed that there is a 10% material loss except for ITO glass and Au in device fabrication, as mentioned in the main article, and 60% material utilization ratio for Au [15]), and the corresponding recycled mass.

Material	MASS (g)	
	Material on stack	recycled mass (lab scale)
ITO glass	5000	5000
SnO ₂	0.078	0.078
MAPbI ₃	1.368	1.19
Spiro-OMeTAD	0.697	0.46
Au	0.99	0
LiTFSI	0.09	0
FK 209 Co (III) TFSI	0.081	0
Total	5003.3	5001.7

Table S9: Electricity consumption for recycling.

Recycling target	steps with electricity consumption	Power (kW)	Time (h)	Electricity (kWh)	overall electricity consumption (kWh)
Spiro-OMeTAD	flash column chromatography	0.06	100/60	0.1	4.81
	Rotary Evaporation	1.4	140/60	3.27	
	vacuum drying	0.06	12	0.72	
MAPbI ₃	centrifugation	0.07	3/60	0.0035	
	vacuum drying	0.06	12	0.72	
ITO/SnO ₂	\	\	\	\	

Table S10: Characterization factors for raw materials required for a 1 m² module (Part I). The primary data were adapted from literature [16]. Due to the absence of sufficient data, FK209 Co(III) TFSI was excluded from our analysis; however, given its low usage of only 0.092 g, its exclusion is expected to have a negligible impact.

Item	Unit	Usage	Cumulative energy demand (MJ-Eq)	Agricultural land occupation (m ² a)	Climate change (kg CO ₂ -Eq)	Fossil depletion (kg oil-Eq)	Freshwater ecotoxicity (kg 1,4-DCB-Eq)	Freshwater eutrophication (kg P-Eq)	Human toxicity (kg 1,4-DCB-Eq)	Ionising radiation (kg U235-Eq)	Marine ecotoxicity (kg 1,4-DCB-Eq)	Marine eutrophication (kg N-Eq)	Metal depletion (kg Fe-Eq)
ITO glass	m ²	1.00E+00	2.92E+02	5.40E-02	1.59E+01	6.87E+00	8.25E-02	1.06E-03	1.66E+02	1.67E-01	6.65E+01	1.51E-02	6.41E-01
Deionized water	kg	3.46E-02	8.31E-04	2.78E-06	5.55E-05	1.60E-05	8.32E-07	2.95E-08	1.10E-03	6.02E-06	9.48E-04	5.95E-08	3.97E-06
Acetone	kg	2.59E-02	1.74E+00	9.88E-06	5.00E-02	4.00E-02	4.22E-04	4.03E-06	2.75E-02	8.00E-06	5.42E-02	4.68E-05	3.22E-05
Isopropanol	kg	2.68E-02	1.62E+00	9.83E-04	4.26E-02	3.70E-02	2.48E-04	7.03E-06	4.09E-01	9.87E-04	3.05E-01	2.99E-05	1.73E-03
Tin oxide	kg	8.70E-05	2.36E-02	5.25E-05	1.44E-03	4.36E-04	1.87E-05	7.21E-07	2.60E-02	2.35E-04	2.17E-02	4.27E-06	1.07E-01
MAI	kg	1.40E-04	4.80E-01	2.26E-03	2.07E-02	9.55E-03	7.80E-04	1.84E-05	9.15E-01	2.21E-03	6.96E-01	2.94E-05	1.65E-03
Lead iodide	kg	1.38E-03	7.49E-02	1.44E-03	5.38E-03	1.43E-03	9.83E-05	2.73E-06	2.15E-01	2.64E-04	1.08E-01	7.12E-06	8.72E-04
DMF	kg	2.47E-03	1.92E-01	2.87E-04	6.39E-03	4.30E-03	8.99E-05	2.43E-06	1.46E-01	6.32E-04	9.51E-02	3.08E-05	4.11E-04
NMP	kg	5.67E-04	6.17E-02	2.05E-04	3.18E-03	1.67E-03	6.01E-06	1.33E-07	1.20E-02	1.42E-04	4.26E-03	7.34E-07	1.38E-04
Spiro-OMeTAD	kg	7.74E-04	1.52E+00	2.30E-02	8.51E-02	2.75E-02	5.74E-03	9.90E-05	5.07E+00	8.65E-03	4.29E+00	9.52E-05	3.91E-03
LiTFSI	kg	9.70E-05	6.76E-02	3.57E-04	3.66E-03	1.45E-03	2.11E-04	4.01E-06	1.80E-01	1.96E-04	1.48E-01	4.39E-06	1.31E-04
Acetonitrile	kg	3.89E-04	4.30E-02	2.39E-05	1.43E-03	9.87E-04	6.83E-06	1.67E-07	1.12E-02	3.44E-05	8.73E-03	6.54E-06	4.19E-05
Chlorobenzene	kg	1.18E-02	9.29E-01	1.36E-03	3.26E-02	2.05E-02	1.19E-03	1.57E-05	5.96E-01	1.84E-03	4.86E-01	3.23E-05	1.57E-03
tBP	kg	2.84E-04	5.16E-02	1.75E-04	2.89E-03	1.06E-03	4.10E-05	1.35E-06	6.11E-02	2.55E-04	4.83E-02	3.55E-06	1.92E-04
Silver	kg	2.50E-03	8.82E+00	2.79E-02	5.47E-01	1.91E-01	5.32E-01	1.43E-02	1.13E+03	3.96E-02	5.92E+02	4.24E-03	3.46E+00

Table S11: Characterization factors for raw materials required for a 1 m² module (Part II). The primary data were adapted from literature [16]. Due to the absence of sufficient data, FK209 Co(III) TFSI was excluded from our analysis; however, given its low usage of only 0.092 g, its exclusion is expected to have a negligible impact.

Item	Unit	Usage	Natural land transformation (m ²)	Ozone depletion (kg CFC-11-Eq)	Particulate matter formation (kg PM10-Eq)	Photochemical oxidant formation (kg NMVOC)	Terrestrial acidification (kg SO ₂ -Eq)	Terrestrial ecotoxicity (kg 1,4-DCB-Eq)	Urban land occupation (m ² a)	Water depletion (m ³)	Ecosystem quality (points)	Human health (points)	Resources (points)
ITO glass	m ²	1.00E+00	2.61E-03	1.38E-06	2.00E-02	4.89E-02	7.22E-02	7.88E-03	1.58E-02	1.02E-02	4.89E-01	1.72E+00	8.54E-01
Deionized water	kg	3.46E-02	7.66E-09	2.53E-11	1.98E-07	1.70E-07	3.06E-07	4.94E-08	5.29E-07	4.54E-05	2.60E-06	9.94E-06	2.10E-06
Acetone	kg	2.59E-02	-4.86E-09	2.47E-11	7.24E-05	2.35E-04	2.63E-04	4.64E-06	4.27E-06	3.58E-06	1.38E-03	2.15E-03	4.79E-03
Isopropanol	kg	2.68E-02	3.94E-06	1.73E-09	6.45E-05	2.10E-04	1.81E-04	1.40E-05	1.39E-04	1.13E-04	1.47E-03	4.43E-03	4.51E-03
Tin oxide	kg	8.70E-05	6.21E-06	1.17E-10	2.22E-05	1.39E-05	3.37E-05	7.82E-07	9.48E-05	5.11E-06	5.30E-04	2.81E-04	5.04E-03
MAI	kg	1.40E-04	4.37E-06	7.48E-09	6.76E-05	9.10E-05	1.71E-04	3.25E-05	3.12E-04	4.06E-04	1.30E-03	7.14E-03	1.22E-03
Lead iodide	kg	1.38E-03	9.15E-07	6.16E-10	1.52E-05	2.15E-05	4.31E-05	7.42E-06	5.55E-05	1.62E-05	2.64E-04	1.68E-03	2.12E-04
DMF	kg	2.47E-03	2.70E-06	1.28E-09	1.78E-05	2.16E-05	4.54E-05	4.54E-05	5.45E-05	1.91E-05	2.84E-04	1.26E-03	5.34E-04
NMP	kg	5.67E-04	-1.75E-07	2.93E-10	5.72E-06	9.95E-06	1.44E-05	2.52E-06	2.22E-05	1.55E-04	1.28E-04	2.07E-04	5.31E-04
Spiro-OMeTAD	kg	7.74E-04	7.42E-06	8.00E-09	4.80E-04	3.39E-04	4.63E-04	9.61E-05	1.26E-03	2.79E-04	4.94E-03	3.87E-02	3.47E-03
LiTFSI	kg	9.70E-05	6.14E-07	5.12E-10	8.76E-06	1.28E-05	2.76E-05	3.41E-06	3.46E-05	8.80E-06	1.71E-04	1.38E-03	1.80E-04
Acetonitrile	kg	3.89E-04	1.62E-07	7.75E-11	2.43E-06	3.67E-06	1.03E-05	9.58E-07	3.82E-06	3.68E-06	4.77E-05	1.32E-04	1.20E-04
Chlorobenzene	kg	1.18E-02	3.00E-06	8.42E-09	9.78E-05	2.60E-04	1.75E-04	2.25E-05	3.20E-04	1.00E-04	1.46E-03	5.45E-03	2.53E-03
tBP	kg	2.84E-04	4.24E-07	7.97E-10	8.16E-06	9.01E-06	1.60E-05	2.94E-06	2.58E-05	9.26E-06	1.30E-04	5.36E-04	1.36E-04
Silver	kg	2.50E-03	2.75E-04	7.40E-08	3.15E-03	1.05E-02	1.01E-02	6.52E-04	4.74E-02	1.07E-02	1.92E-01	7.75E+00	1.83E-01

Percentage of Each Material in Cumulative Energy Demand (MJ-Eq)

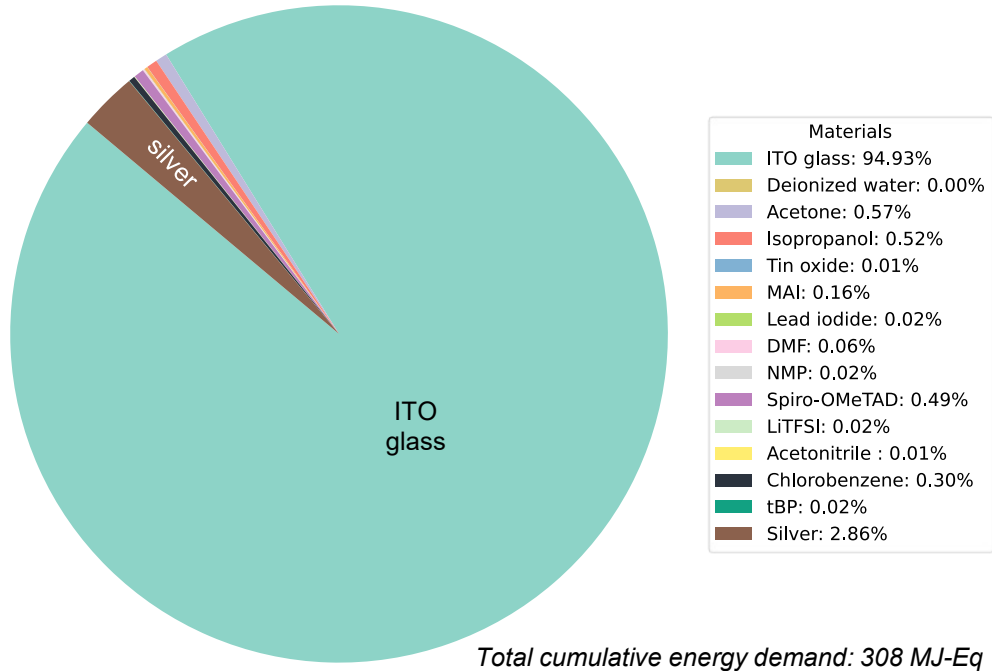


Figure S12: Cumulative energy demand composition of virgin materials required for a 1 m² module.

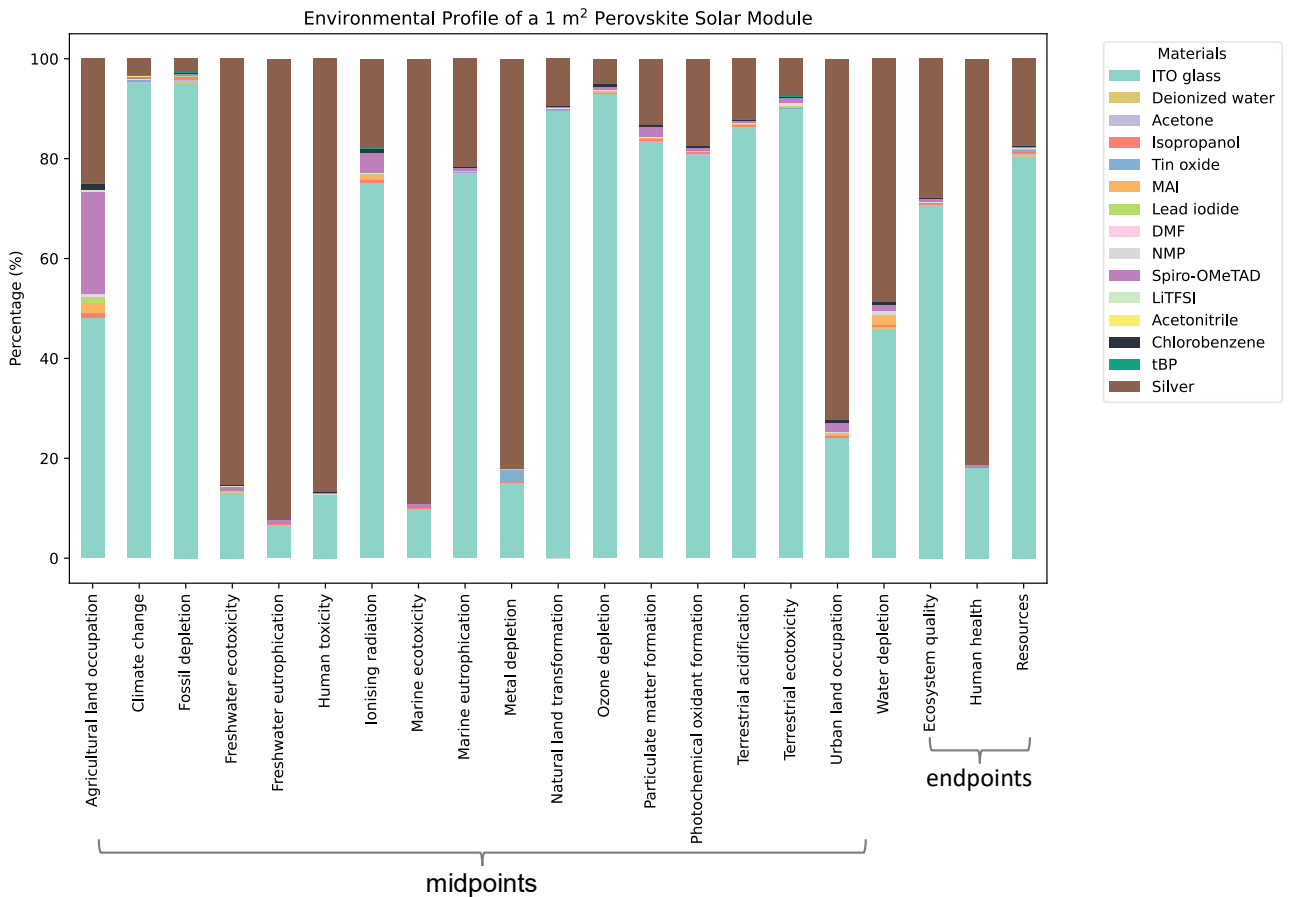


Figure S13: Environmental profile of a 1 m² module.

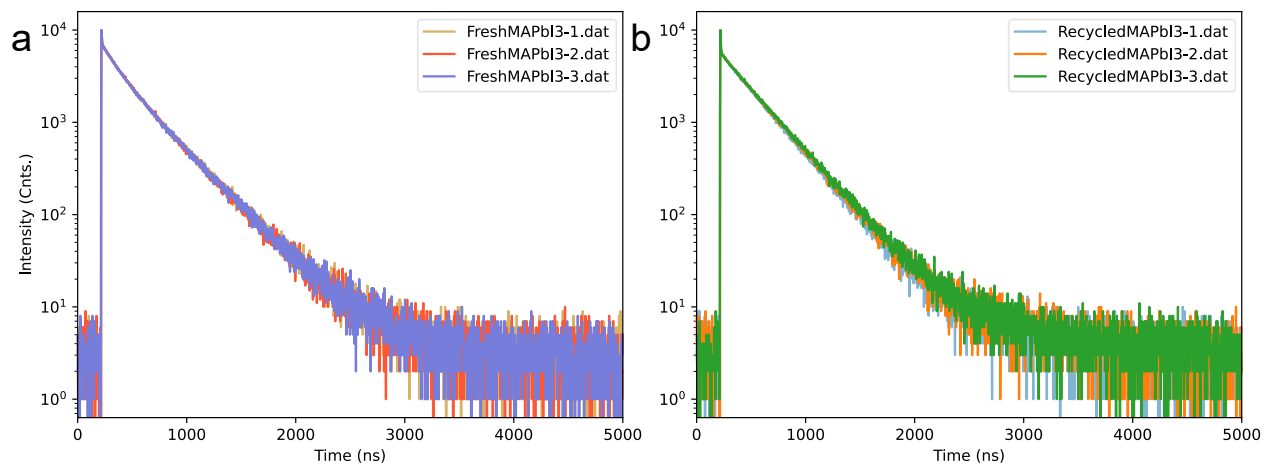


Figure S14: TR-PL spectra for a) fresh MAPbI₃ and b) recycled MAPbI₃, each measured three times consecutively at the same spot.

There are two primary uncertainty sources for PCE:

- 1) Discrepancy between the designed electrode area and the actual area. Table S12 displays the designed electrode area and the areas measured from optical microscope images (Figure S15). Analysis of their differences indicates an average deviation of $1.14\% \pm 0.55\%$, leading to electrode areas ranging from 98.31% to 101.69% of the designed area. Since PCE inversely correlates with electrode area according to Equation (4), the area discrepancies imply that the real PCE could diverge from the measured values, potentially being up to 1.66% lower or 1.72% higher.

$$PCE(\%) = \frac{P_{out}}{P_{in}} * 100 = \frac{J_{max} * V_{max}}{P_{in}} * 100 = \frac{I_{max} * V_{max}}{Area * P_{in}} * 100 \quad (4)$$

- 2) JV measurement setup uncertainty. To determine the uncertainty of the setup, we conducted ten JV characteristic measurements of an OPV cell. We chose OPV cells for their superior stability compared to perovskite solar cells. The PCE results revealed a coefficient of variation of 2.72%.

The overall uncertainty, calculated using the root of the sum of squares (RSS) method, is 3.22%. This calculation excludes the smaller 1.66% deviation from electrode area discrepancies to avoid double counting the same source of uncertainty. Consequently, the actual PCE is estimated to be within 3.22% less or more than the measured PCE.

Table S12: Measured electrode area of four cells versus the designed area.

	designed	Cell 1	Cell 2	Cell 3	Cell 4
Total area (μm^2)	6296792	6196054	6321202	6392110	6363482

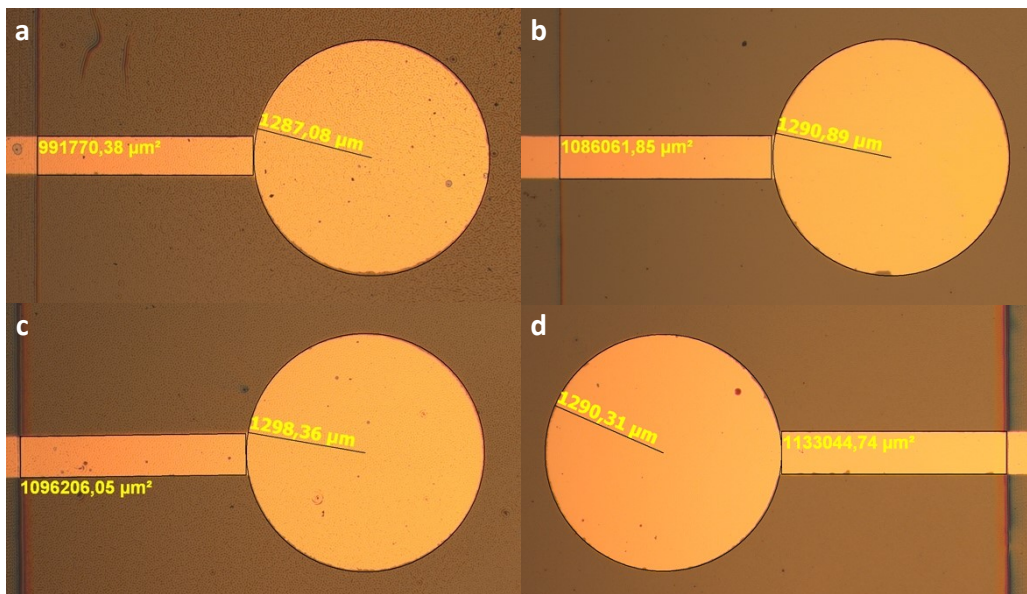


Figure S15: Optical microscope images of 4 solar cells.

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