# Supporting Information

## Green Solvent Engineering for Sustainable Recovery and Circular Use of Perovskite Solar Cells

Yash Taneja [a, b], Ranbir Singh [a, b, *]

[a]School of Mechanical and Materials Engineering, Indian Institute of Technology Mandi, Kamand, Mandi, Himachal Pradesh, India

[b]Advanced Energy Conversion Laboratory (AECL), Indian Institute of Technology (IIT) Mandi, Mandi, Himachal Pradesh, 175005, India

Corresponding Email ID: *ranbir@iitmandi.ac.in*

### Solar Cell Fabrication using Fresh Materials

Perovskite solar cells with Glass/ITO/$SnO_2$/$MAFAPbI_3$/Spiro-OMeTAD/Au architecture were fabricated within a nitrogen filled glove box. Etched ITO substrates were cleaned using detergent, deionized water, acetone, and isopropyl alcohol (IPA). Substrates were then placed in an oven for 2 hrs at 70 °C before ozone plasma treatment for 20 minutes. A 15% $SnO_2$ colloidal dispersion in water was then spin-coated onto the substrates at 2000 rpm for 20 seconds, followed by heating at 130 °C for 20 minutes. For $MAFAPbI_3$ film deposition, MAI:FAI:$PbI_2$ (1.26M:0.14M:1.4M) were mixed in 1 mL of solvent (4:1 anhydrous DMF:DMSO).[1] The precursor was spin-coated onto the $SnO_2$ layer with an antisolvent method at a spin rate of 4000 rpm. 200 µL of chlorobenzene was immediately dripped on the film, followed by thermal annealing at 120 °C for 5 minutes. To create the hole transport layer, a spiro-OMeTAD solution was prepared by dissolving 60 mg of the material in 1 mL of chlorobenzene. Freshly prepared solutions of 16 µL of Li-TFSI (from a 520 mg/mL acetonitrile stock), 10 µL of TBP, and 6 µL of FK209 (from a 300 mg/mL acetonitrile stock) were added to the spiro-OMeTAD solution. This solution was spin-coated onto the substrate

at 3000 rpm for a duration of 20 seconds. Finally, 80 nm thick Au was thermally evaporated as a top electrode under vacuum (< $4\times10^{-6}$ Torr) using a mask.

**Solar Cell Fabrication using Recycled Materials**

A similar process was used for device fabrication using the recycled materials. The recycled Glass/ITO substrates, which were already etched, went under sequential sonication in distilled water, IPA, acetone, and IPA. After cleaning, the substrates were treated in a UV/ozone chamber for 30 minutes to ensure complete surface activation. Following ozonisation, a fresh ETL layer of similar $SnO_2$ was deposited using the spin coating method at 2000 rpm. The films were kept on the hot plate at 130 °C for 20 minutes. Following this, the recycled perovskite powder, which was dissolved in (4:1) DMF: DMSO solution, was spin-coated at 1000 rpm for 10 seconds, followed by a second spin-coating step at 4000 rpm for 30 seconds. After 10 seconds of spinning at 4000 rpm, 300 μL of chlorobenzene antisolvent was dripped onto the perovskite-coated substrates, and the film was annealed at 120 °C for 5 minutes. The recovered spiro-OMeTAD powder was first dried and then dissolved in 200 μL of chlorobenzene. The solution was then spin-coated at 1500 rpm on the perovskite film. Finally, the Au was thermally deposited as a top electrode under vacuum (< $4\times10^{-6}$ Torr) using a metal mask.

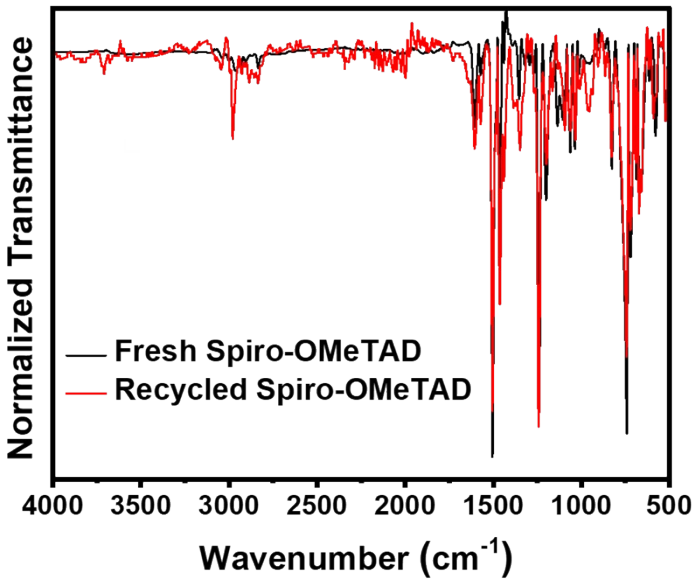


**Figure S1.** FTIR spectra for fresh and recycled spiro-OMeTAD.

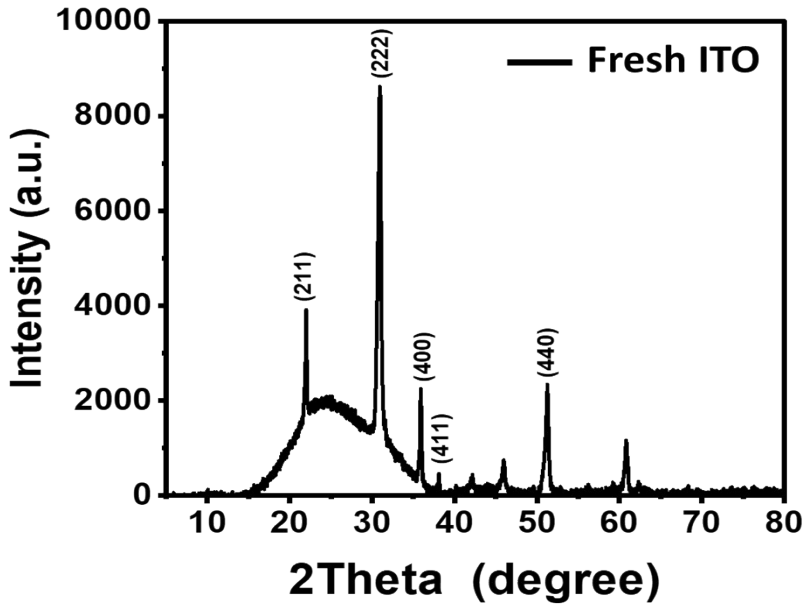


**Figure S2.** XRD plot for fresh glass/ITO substrate.

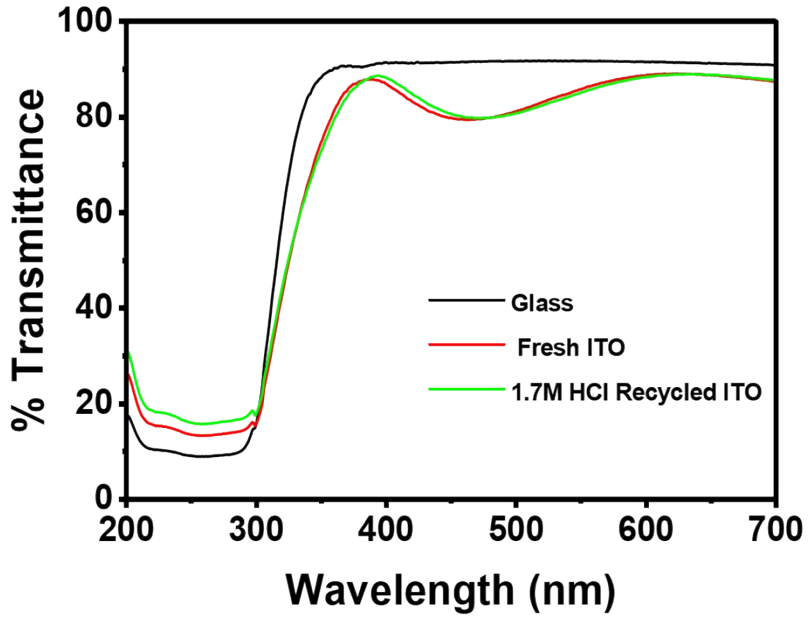


**Figure S3.** Transmittance spectra of glass, fresh ITO, and recycled ITO substrates.

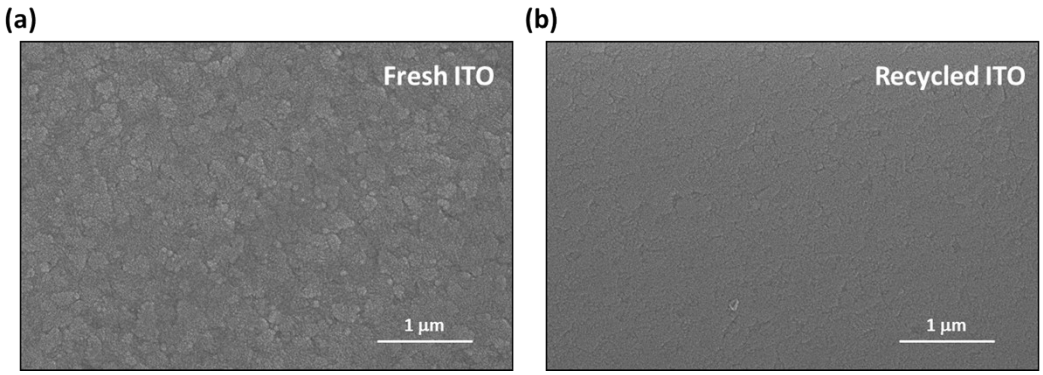


**Figure S4.** FE-SEM images of (a) fresh and (b) recycled ITO.

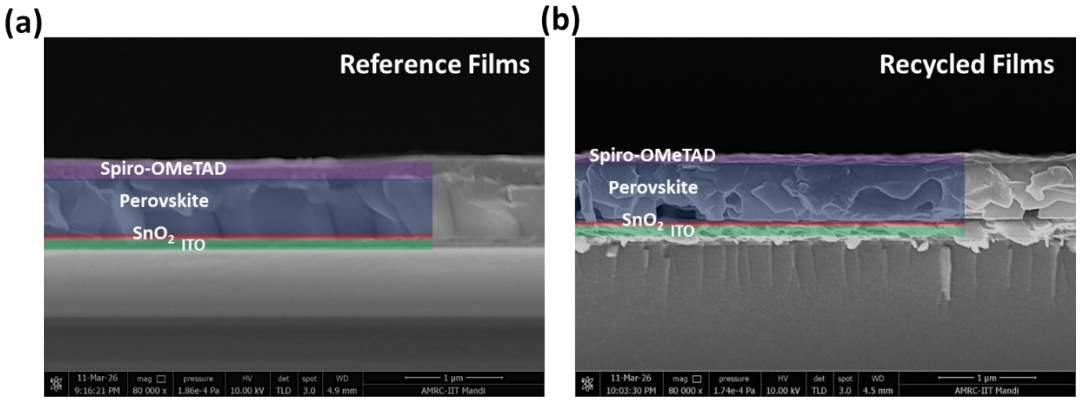


**Figure S5.** Cross-sectional SEM images of the device architecture. All the films (a) fabricated with fresh materials and (b) fabricated with recycled materials showing the multilayer structure consisting of ITO/$SnO_2$/perovskite/Spiro-OMeTAD, exhibiting a similar layered configuration.

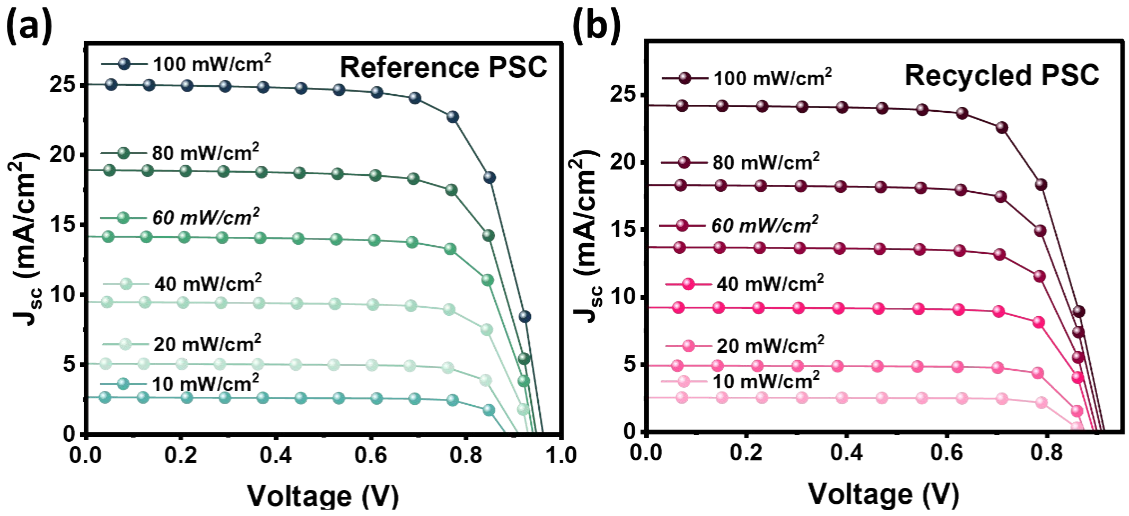


**Figure S6.** *J-V* curves for (a) reference and (b) recycled PSC at various light intensities (10, 20, 40, 60, 80, and 100 mW/cm$^2$).

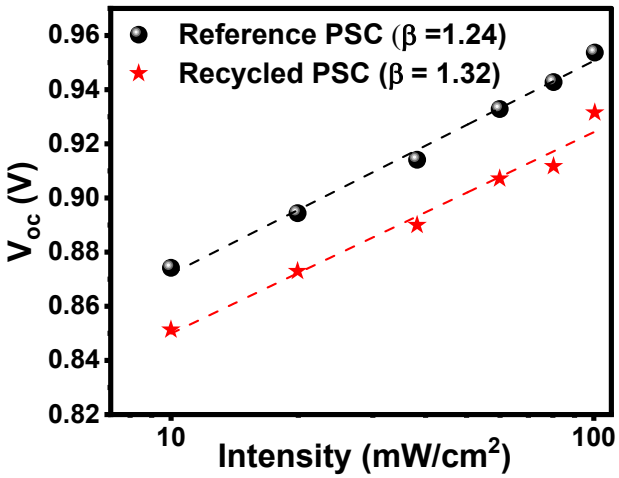


**Figure S7.** Light-intensity dependence of the $V_{oc}$ for the reference and recycled PSCs. The semi-logarithmic plot displays the experimental data points fitted with a linear regression.

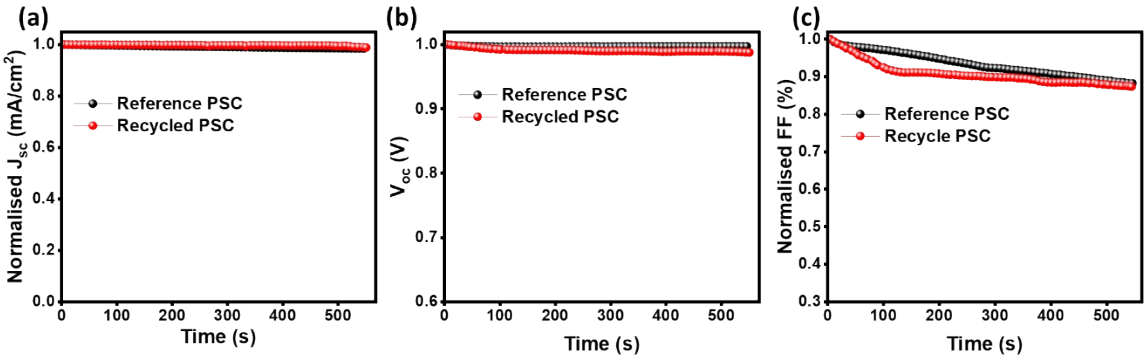


**Figure S8.** Light soaking stability data for 500 seconds of continuous illumination using AM 1.5G light source (a) *J-V,* (b) $V_{oc}$, and (c) FF of fresh and recycled PSCs.

**Table S1.** showing the EIS parameters like series resistance ($R_s$), recombination resistance ($R_{se}$), and charge transfer resistance ($R_{ct}$) for reference and recycled PSCs.

| **Device** | **$R_s$ (Ohm)** | **$R_{re}$ (Ohm)** | **$R_{ct}$ (Ohm)** |
|---|---|---|---|
| Reference PSC | 8.306 | $3.44 \times 10^4$ | 4890 |
| Recycled PSC | 21.46 | $3.44 \times 10^4$ | 5624 |

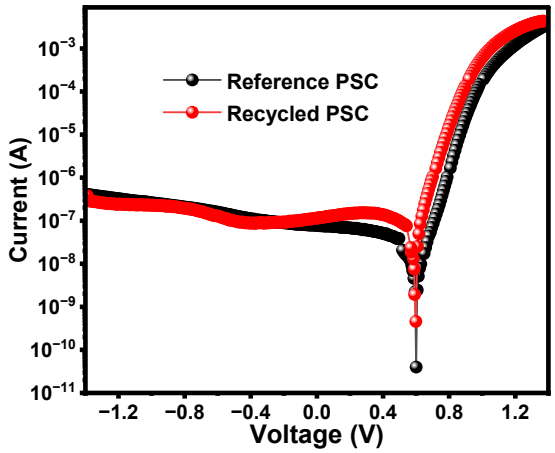


**Figure S9.** Schematic showing dark-current ($I_D$) vs voltage (V) curve for reference and recycled PSC devices.

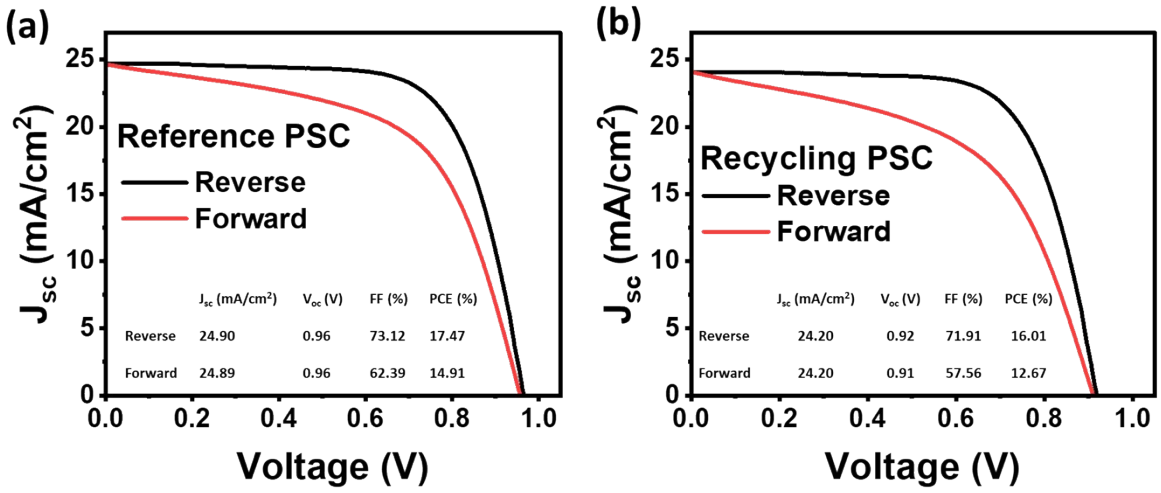


**Figure S10.** Schematic showing the reverse and forward J-V scans for (a) reference and (b) recycled PSC device.

**Table S2.** showing the thickness and approximate (approx.) mass of each layer used in PSCs fabrication. For quantitative analysis, 10 substrates of 1.5 $cm^2$ area are used to calculate the mass of each layer in a PSC device according to the previous studies.[3]

| Layer | Thickness | Approx. Measured Mass (in mg) |
|---|---|---|
| ITO | ~ 120 (nm) | ~ 1.8 |
| $SnO_2$ (Unrecovered) | ~ 24 (nm) | ~ 0.4 |
| Perovskite | ~ 560 (nm) | ~ 5.4 |
| Spiro-OMeTAD | ~ 200 (nm) | ~ 2.3 |
| Gold (Au) | ~ 80 nm | ~ 1.4 |
| Total Thin Film Mass | | ~ 11.3 |

Total mass of the thin-films: ~ 11.3 mg

Mass of unrecovered $SnO_2$: ~ 0.4 mg

Percentage of $SnO_2$: (0.4 mg / 11.3 mg) × 100 = 3.53%

Total recovery: 100%  3.53% = 96.47%

In case the mass of the 10 glass substrates with 1.2 mm thickness (~5041.1 mg) is included, the recovery percentage increases to over 99.5%.

**Table S3.** shows a color-coded assessment of various solvents based on green chemistry parameters, highlighting their relative safety in terms of human health, environmental impact, and aquatic toxicity.[2] (Higher values (green) signify safer, more environmentally benign properties, whereas yellow represents intermediate scores (4, 5, and 6), indicating moderate safety. Conversely, lower scores (red) indicate significant health, safety, or environmental hazards.)

| Solvents | Recycling | Bio-treatment | VOC# Emission | Aquatic impact | Air Impact | Health hazard | Exposure potential | Explosion Danger |
|---|---|---|---|---|---|---|---|---|
| **Water** | - | 4 | 6 | 10 | 10 | 10 | 9 | 9 |
| **DMSO** | 4 | 6 | 9 | 8 | 6 | 7 | 9 | 9 |
| **DMF** | 4 | 3 | 8 | 9 | 4 | 1 | 6 | 9 |
| **Ethyl acetate** | 6 | 6 | 4 | 9 | 5 | 10 | 7 | 8 |
| **γ-butyrolactone** | - | - | 5 | - | - | - | - | 9 |
| **2-methoxyethanol** | 5 | 4 | 7 | 10 | 5 | 1 | 2 | 8 |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| **Chlorobenzene** | 9 | 7 | 7 | 10 | 1 | 4 | 4 | 8 |

[#] VOC signifies volatile organic compounds.


## References


1. Singh, R., et al., *Enhancing the power conversion efficiency of perovskite solar cells via the controlled growth of perovskite nanowires.* Nano Energy, 2018. **51**: p. 192-198.
2. Alder, C.M., et al., *Updating and further expanding GSK's solvent sustainability guide.* Green Chemistry, 2016. **18**(13): p. 3879-3890.
3. Wu, Z., et al., *Closing the loop: recycling of MAPbI 3 perovskite solar cells.* Energy & Environmental Science, 2024. **17**(12): p. 4248-4262.