Recycling lead and transparent conductors from perovskite solar modules

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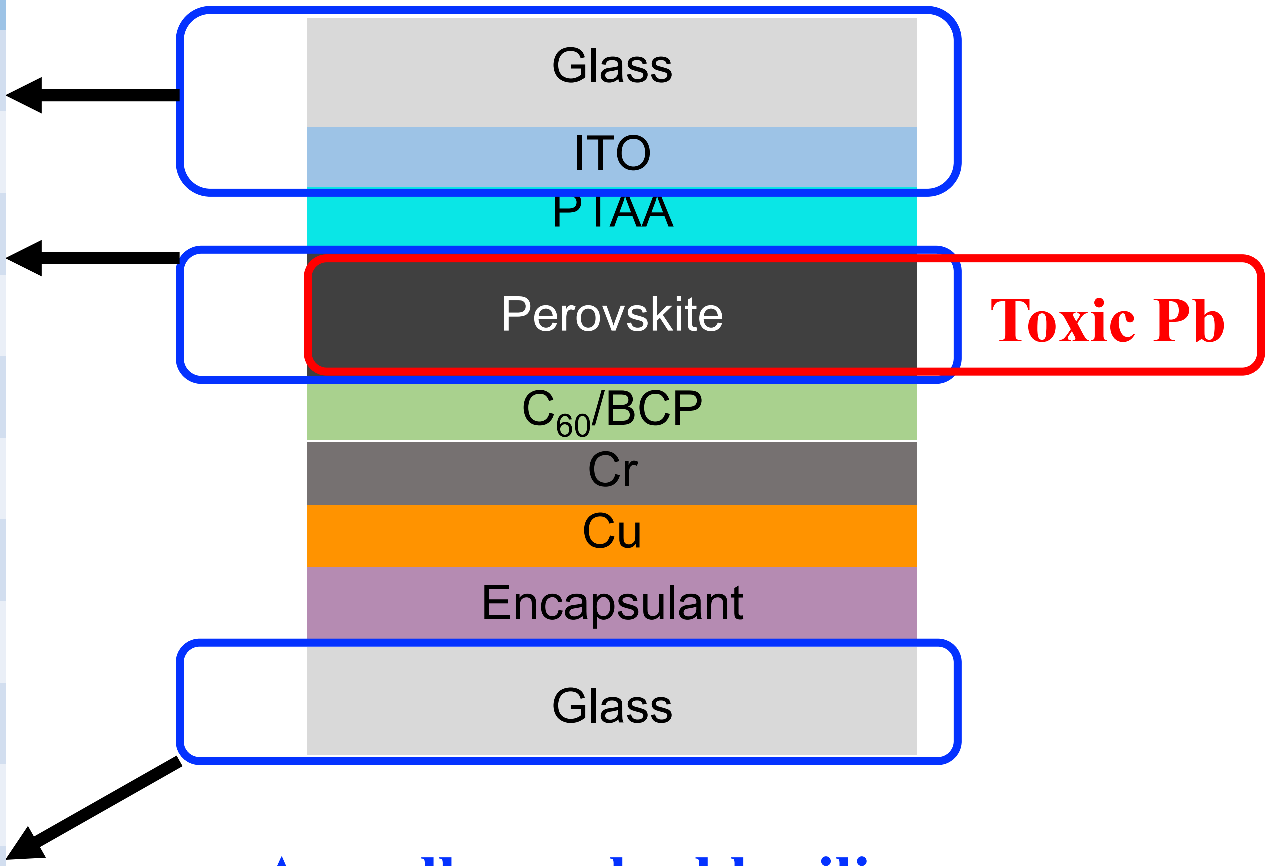
What should or can be recycled?

- Recycle **valuable components** for economically attractive

Module materials	Cost($/m^2)
ITO/glass (0.67 – 3.2 mm)	6.4 (6.4 – 12)
PTAA (10 nm)	5.29
PbI_2 (Perovskite 1 µm)	3.21
FAI (Perovskite 1 µm)	1.55
CsI (Perovskite 1 µm)	0.37
C_{60} (30 nm)	3.00
BCP (6 nm)	0.51
Cr (30 nm)	0.06
Cu (150 nm)	0.34
Encapsulant (150-400 µm)	1.7 (1.54 – 2.0)
Back glass (2 – 2.5 mm)	2.4 (2.4 – 5.04)
Total materials	24.8

- Recycle **toxic components** for environmentally sustainable

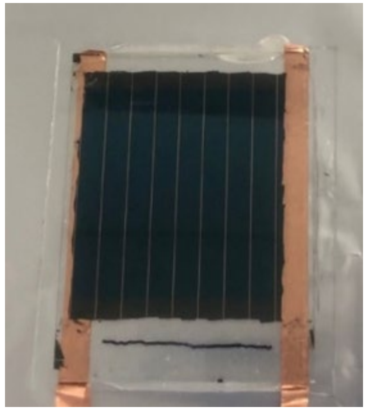


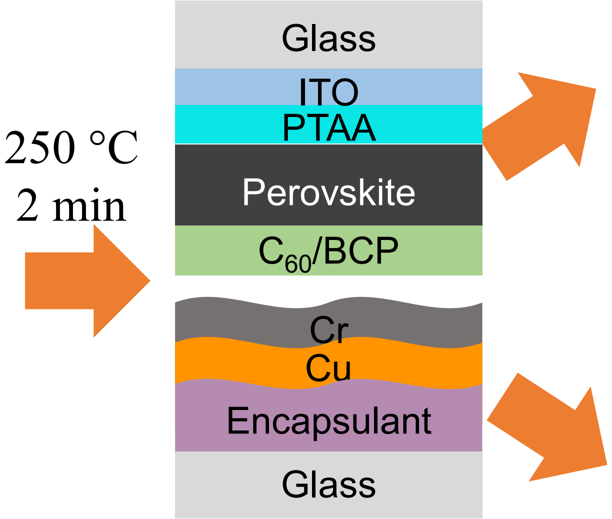
As well as valuable silicon bottom cell in tandem

Thermal delamination of encapsulated perovskite solar module

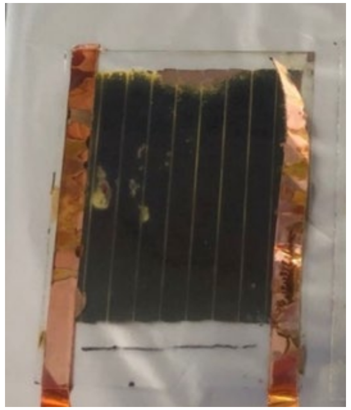
- **Method:** thermal delamination (thermal stress at 250 °C for 2 min)
- **Result: 1)** Recycled components: intact ITO/glass and cover glass
  2) Expose the perovskite layer

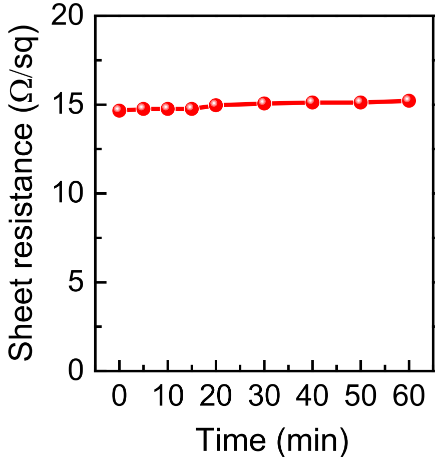
Perovskite module



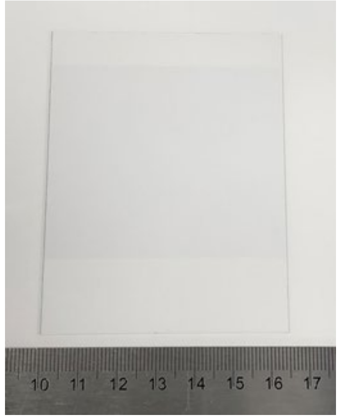


ITO/glass side





Recycled ITO/glass



Cover glass side

Recycled cover glass

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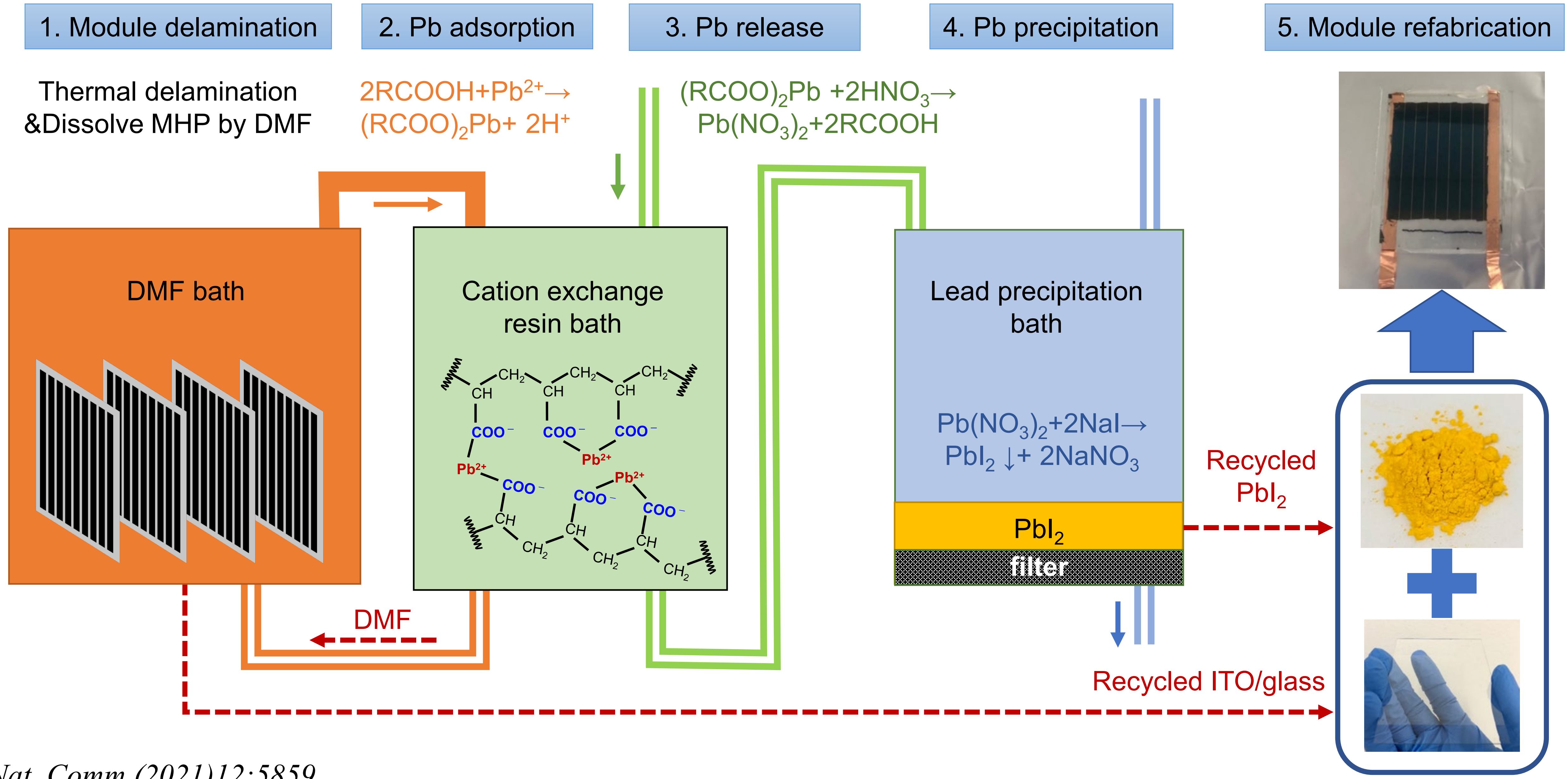
Overview of the Recycling Process



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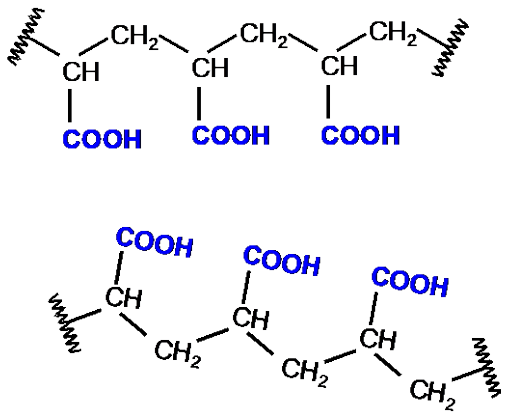
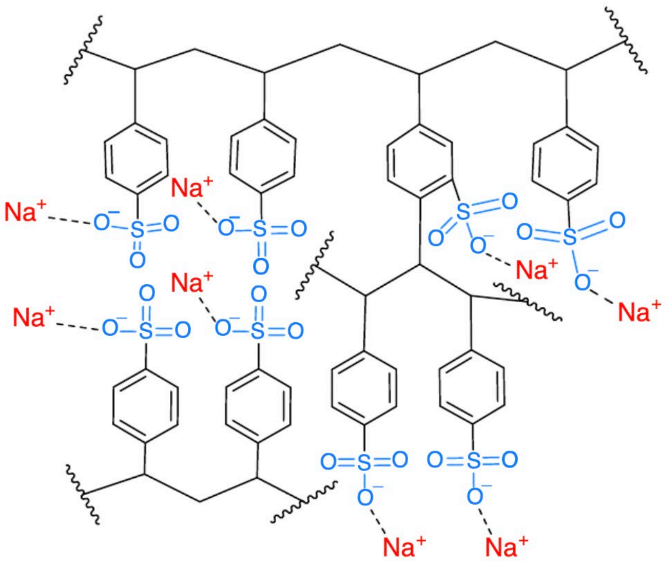
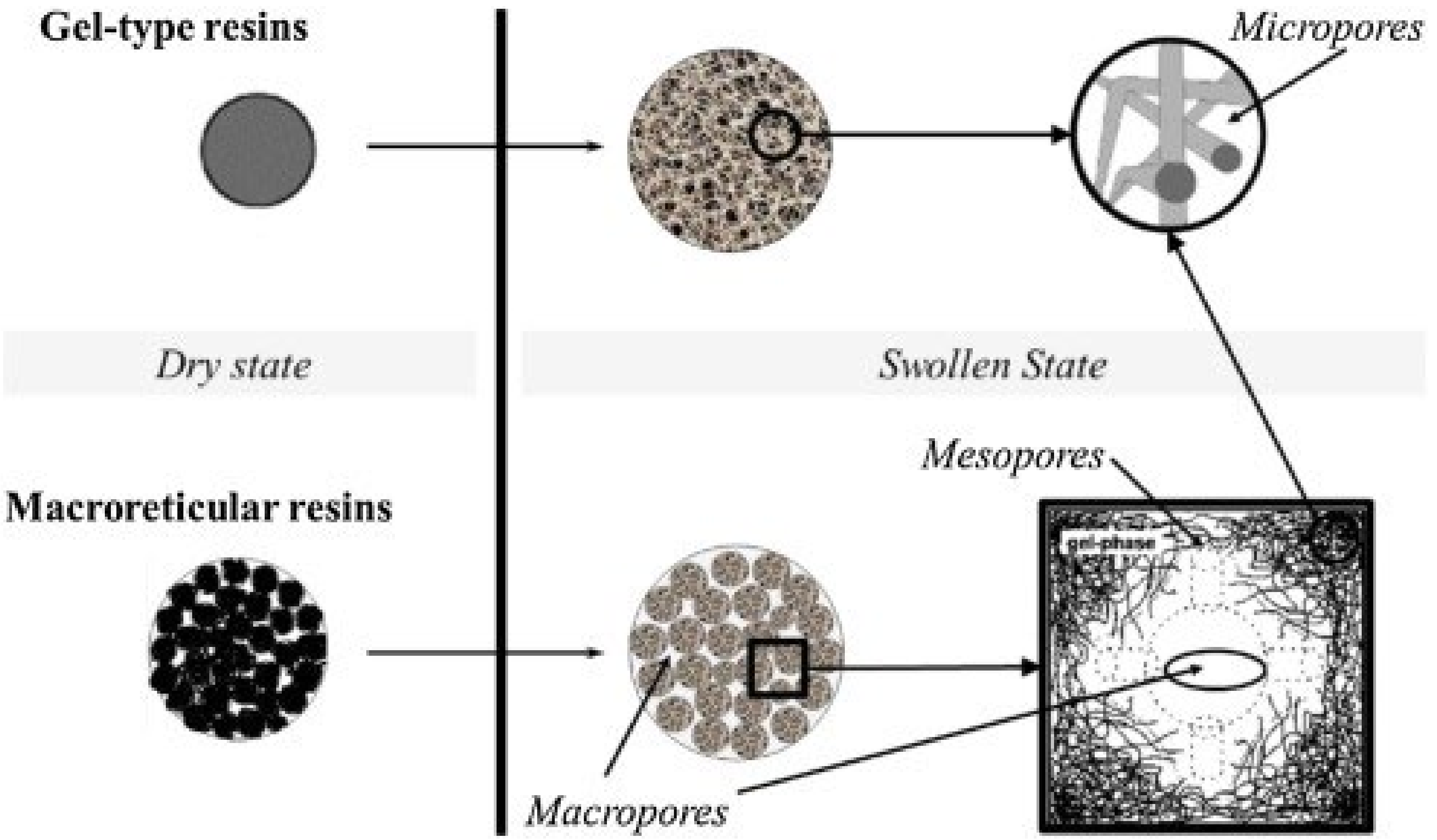
What should be the lead absorbing material?

Weak acid cation exchange resin (WAC)



Strong acid cation exchange resin (SAC)





Lead adsorption process

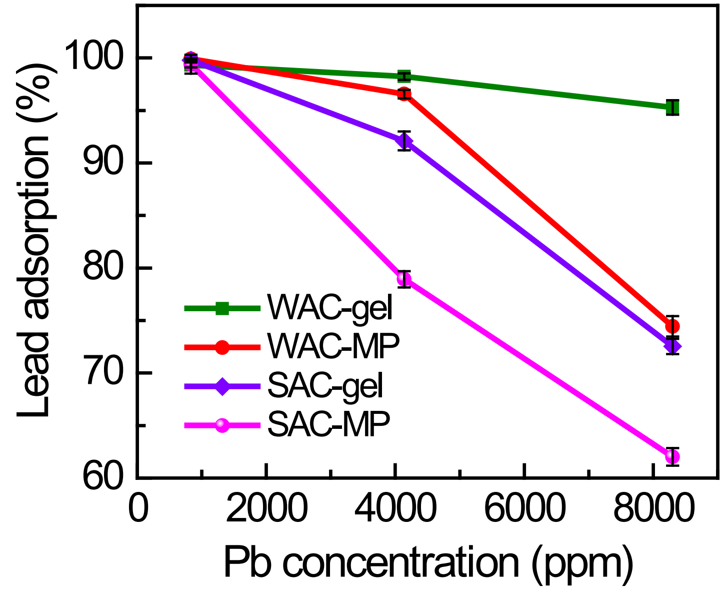
$2R\text{-}H + Pb^{2+} \rightarrow (R)_2\text{-}Pb + 2H^+$

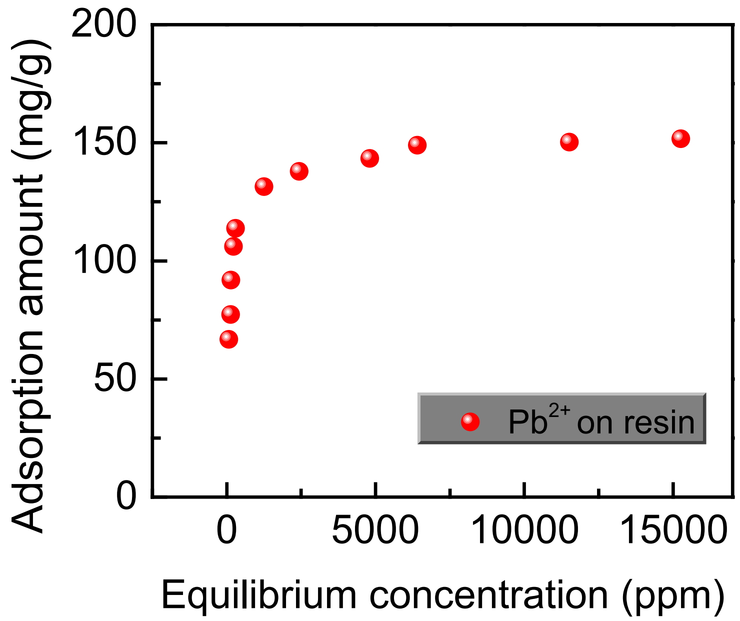
Equilibrium constant: $K_{H^+}^{Pb^{2+}} = \frac{[Pb^{2+}]_r \times [H^+]_s^2}{[Pb^{2+}]_s \times [H^+]_r^2}$

Distribute coefficient: $D = \frac{[Pb^{2+}]_r}{[Pb^{2+}]_s} = K_{H^+}^{Pb^{2+}} \times \frac{[H^+]_r^2}{[H^+]_s^2}$

Resin	Capacity	Affinity
WAC-gel	4 eq/L	$Pb^{2+} < H^+$
WAC-MP	4 eq/L	$Pb^{2+} < H^+$
SAC-gel	1.9 eq/L	$Pb^{2+} > H^+$
SAC-MP	1.7 eq/L	$Pb^{2+} > H^+$

eq/L: equivalents per liter of resin





❖ What is advantage of WAC resin for lead recycling?

1. Large resin total capacity for efficient Pb adsorption in DMF solution

Lead release process

$2R\text{-}H + Pb^{2+} \leftarrow (R)_2\text{-}Pb + 2H^+$

❖ Resin regeneration in aqueous HNO_3 solution

❖ Because $Pb(NO_3)_2$ is soluble in water, chose HNO_3 as regenerant

Resin	Capacity	Affinity
WAC-gel	4 eq/L	$Pb^{2+} < H^+$
WAC-MP	4 eq/L	$Pb^{2+} < H^+$
SAC-gel	1.9 eq/L	$Pb^{2+} > H^+$
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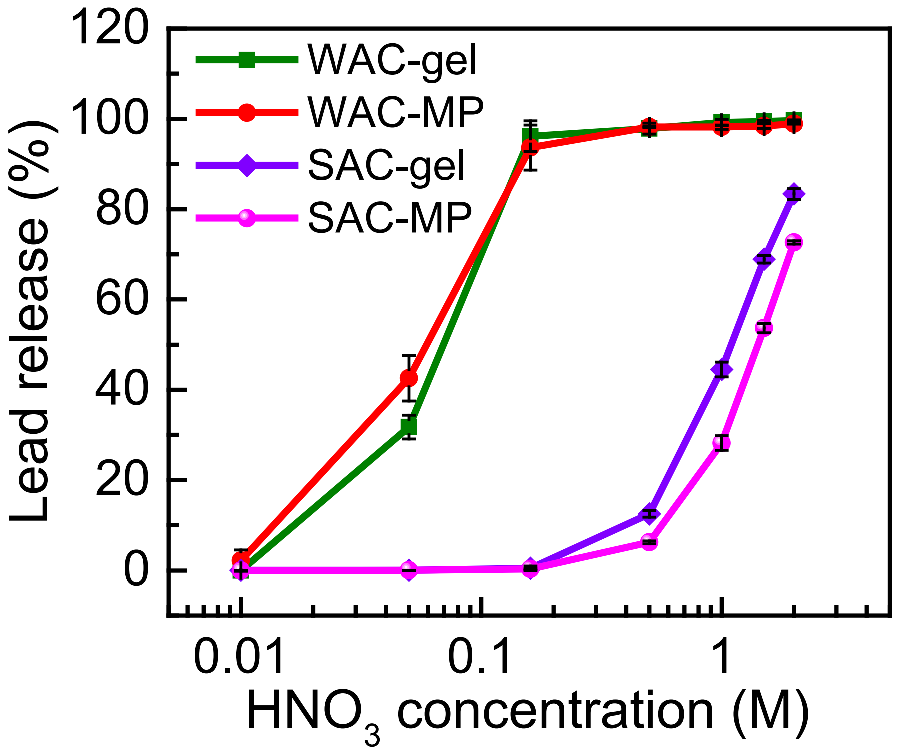


❖ What is advantage of WAC resin for lead recycling?

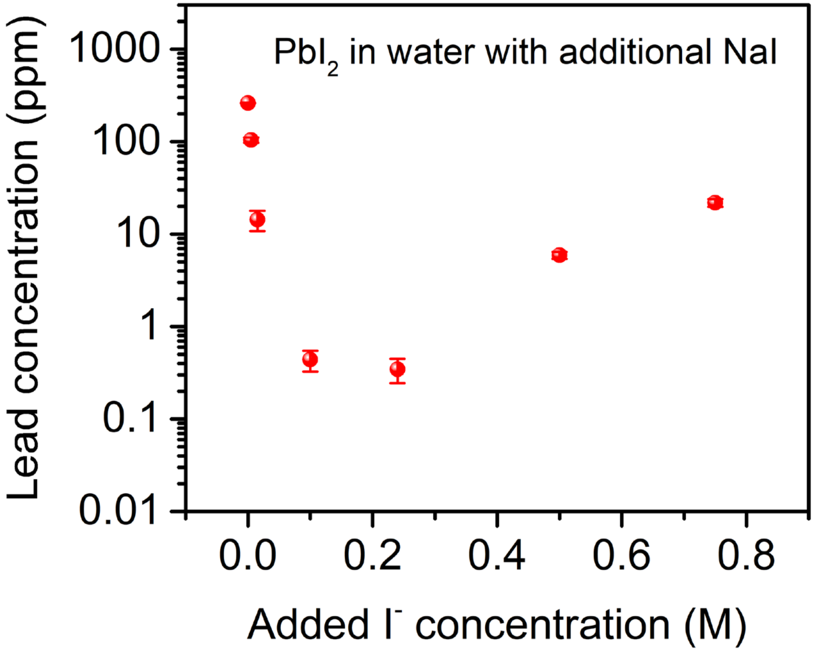
1. Large resin total capacity for efficient Pb adsorption in DMF solution
2. Higher affinity to H^+ than Pb^{2+}, easy release of Pb during regeneration

Lead precipitation process

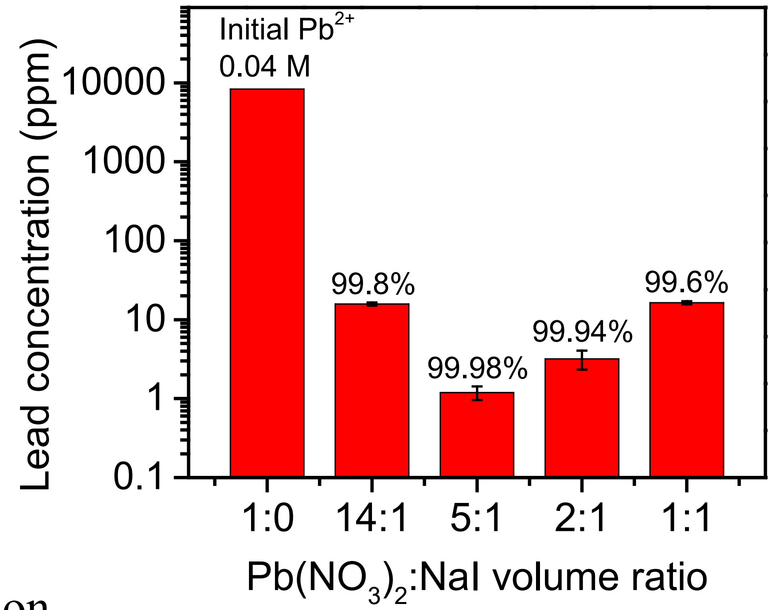
Convert $Pb(NO_3)_2$ in solution to PbI_2 precipitation for reuse, with efficiency of >99.9%

❖ Mechanism: utilize different solubility

- $Pb(NO_3)_2$: 597 g/L
- PbI_2:0.58 g/L (260 ppm)
- PbI_2 with 0.1M NaI: ~1 ppm

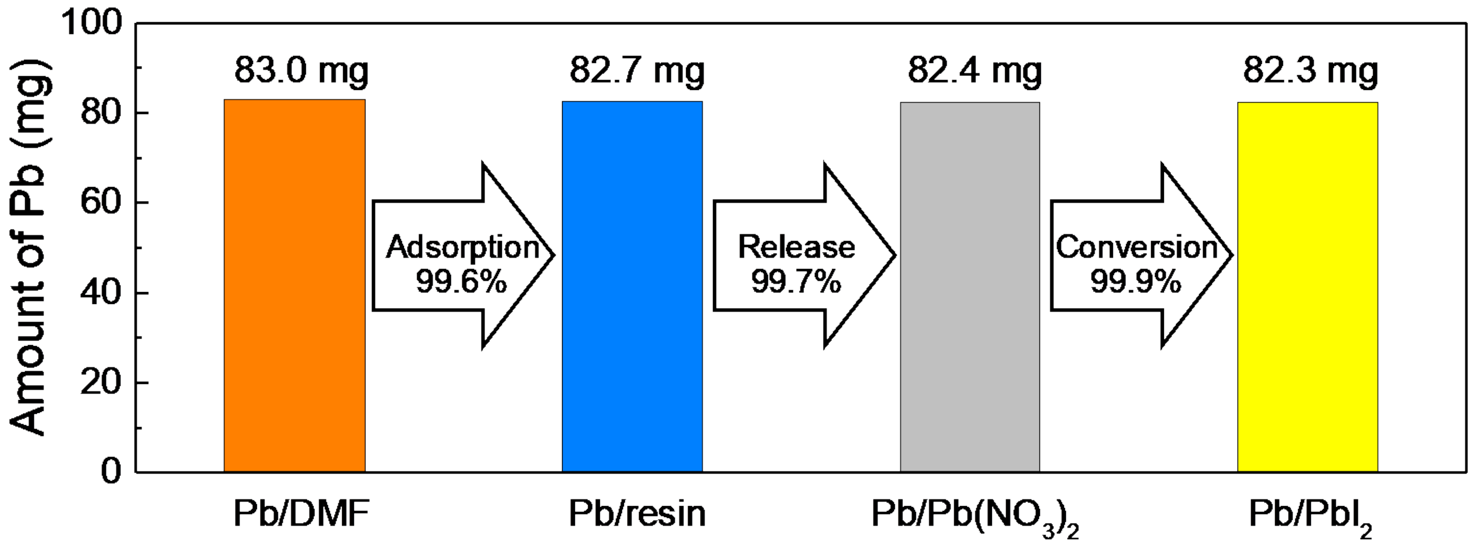


NaI added into $Pb(NO_3)_2$ solution



Lead recycling efficiency

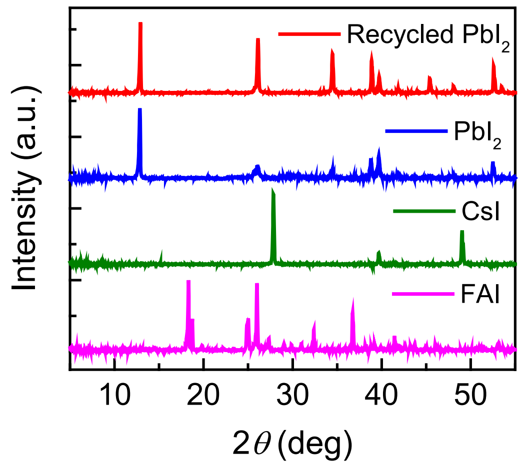
- Proposed adsorbent: weak acid cation exchange resin
  1. 99.6% lead adsorption ratio from DMF solution
  2. 99.7% lead release ratio from resin to clean solution as $Pb(NO_3)_2$
  3. 99.9% of conversion ratio from $Pb(NO_3)_2$ to PbI_2
- Overall Pb recycling efficiency: 99.2%

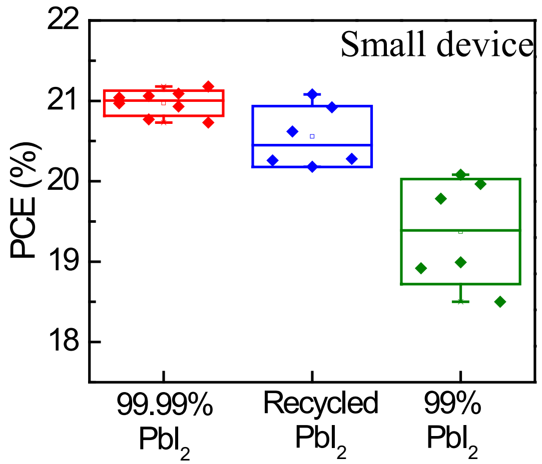


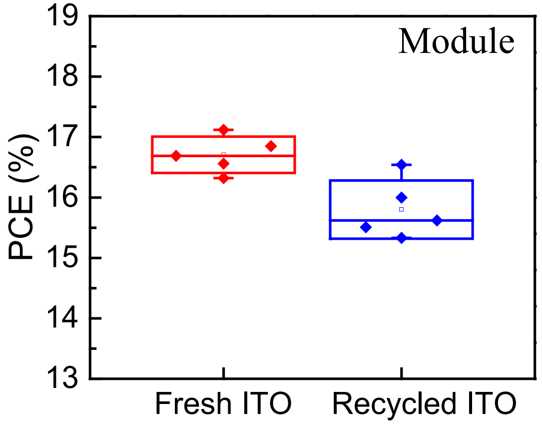
Recycled materials properties and efficiency

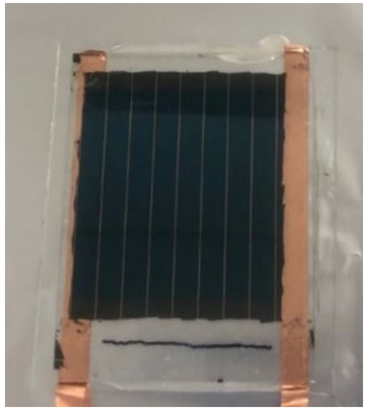
❖ Recycling Pb from $CsFAPbI_3$ perovskite modules generate pure PbI_2

❖ Refabricated devices based on recycled PbI_2 and recycled ITO give comparable PCE as commercial raw materials.









Cost analysis

Recycled value: \$12
~50% of module material value

Recycling consumption: \$1.35
~11% of recycled value

Module materials	**Cost(\$/m^2)**
Total materials cost	24.8
Recycled components	**Cost(\$/m^2)**
ITO/glass (0.67 – 3.2 mm)	6.4 (6.4 – 12)
PbI_2	3.21
Back glass (2 – 2.5 mm)	2.4 (2.4 – 5.04)
Total recycled	12.0
Recycling consumption	Cost(\$/m^2)
DMF (reusable)	2.41/5
Resin (reusable)	1.20/5
DCB	0.09
HNO_3	0.05
NaI	0.49
Total consumption (reuse DMF and resin for 5 times)	1.35

Conclusion

- Weak acid cation exchange resin has excellent Pb adsorption from DMF solvent, as well as near 100% Pb release ratio during regeneration;
- Over 99% lead recycling ratio from decommissioned perovskite solar modules;
- No obvious photovoltaic performance drop for the perovskite solar devices based on recycled PbI_2 or recycled ITO/glass compared to the fresh counterparts;
- Cost analysis shows this recycling technology is economically attractive, in addition to its notable environmentally sustainable impacts.