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Life cycle assessment of lead recycling processes in perovskite solar cells†

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Life cycle assessment (LCA) was employed to evaluate the environmental impacts of various lead (Pb) recycling processes in perovskite solar cells (PSCs). The analysis identifies solvent recovery and reuse as critical factors in reducing environmental harm, highlighting the need for optimized recycling methods to enhance the sustainability of PSCs.

PSCs have emerged as highly promising candidates in the renewable energy field due to their rapid technological advancements and numerous advantages, accelerating their commercialization. However, as these technologies approach maturity, it is increasingly important to consider the management of their entire life cycle, particularly in addressing the environmental impacts associated with their production, usage, and disposal. PSCs face several challenges, particularly environmental concerns, due to the presence of toxic and rare substances such as Pb and other novel metals.[1,2] Especially, Pb is commonly used in perovskite materials because it contributes to high device efficiency. However, it poses significant environmental and health risks if released during production, use, or disposal.[2] Researchers have explored alternatives like Sn, but these have shown lower performance and stability.[3] LCA is a systematic and effective methodology for evaluating the environmental impacts associated with a product's entire life cycle, from raw material extraction to end-of-life (EoL) disposal.

Recent efforts have focused on assessing the environmental impacts of PSC technologies using LCA, particularly as the importance of managing PSCs after their EoL has gained attention.[4–6] LCA is a valuable tool for evaluating perovskite solar cells (PSCs) by analyzing environmental impacts across their entire lifecycle, from raw materials to disposal. It is particularly effective in assessing the environmental impact of each individual process step. This provides detailed insights into areas where improvements are most needed. LCA helps identify key contributors to environmental impacts, offering guidance for selecting more sustainable materials and processes. In addition to carbon footprint, LCA considers broader environmental concerns, including human health, ozone depletion, and eutrophication. This quantitative approach enables clear comparisons between materials and technologies, aiding in the optimization of processes for sustainable engineering. LCAs have been conducted to suggest effective options for the disposal and recycling of discarded PSCs. Alberola-Borras *et al.* explored the environmental benefits of reusing the TiO_2 scaffold instead of focusing solely on Pb recycling, demonstrating an improvement in the environmental profile.[4] Another LCA study has also highlighted the varying environmental impacts of different recycling techniques for PSC substrates, emphasizing the importance of selecting appropriate methods to minimize environmental harm.[5] Rodriguez-Garcia *et al.* evaluated the environmental impacts of 13 recycling techniques for substrates using LCA, revealing that the use of *N*,*N*-dimethyl-formamide (DMF) is a major contributor to environmental impact and suggesting that solvent recovery could potentially be more environmentally favorable compared to producing fresh coated glass.[6] However, there is still a notable lack of comprehensive LCAs that systematically focus on the recycling of toxic Pb for PSCs. Pb is essential for high-efficiency PSCs and must be managed within their lifecycle to minimize environmental pollution and human exposure.

In this study, we conduct an LCA to evaluate and compare the environmental impacts of several Pb recycling processes. Typically, Pb recycling involves three key stages: dissolution, recovery, and crystallization, as depicted in Fig. 1. The dissolution process involves using appropriate solvents to dissolve each layer of the waste module. During the dissolution process, the specific layers of PSCs can be dissolved using various solvents, which are categorized into multi-step and single-step methods. The

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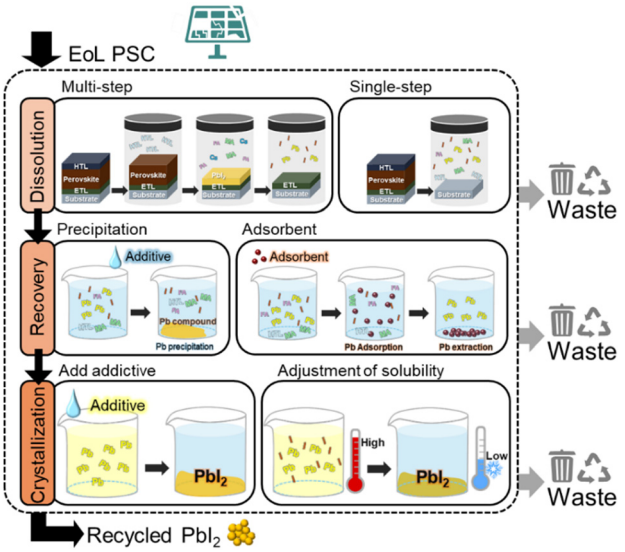

Fig. 1 Schematic illustration of typical Pb recycling process from PSCs including dissolution, recovery, and crystallization steps.

multi-step dissolution method involves selectively dissolving each layer of the PSCs (*e.g.*, hole transport layer (HTL), perovskite layer, electron transport layer (ETL)) separately, allowing for more targeted materials treatment. In contrast, the single-step method involves dissolving layers, including perovskite layer, in a single solution, offering a simpler process. The recovery process focuses on extracting lead materials from a solvent mixture containing various components. Once dissolved, Pb is recovered from the solution using two distinct methods: precipitation, where an additive is introduced to induce Pb compound formation, and an adsorbent-based method, where adsorbents selectively bind to Pb ions for extraction. The final step is crystallization, in which solid PbI_2 forms from its dissolved Pb. This is achieved through either the use of halide salts (*e.g.*, KI, HI, NaI) as additives or by adjusting the solubility through temperature control. Although the primary goal of Pb recycling is to reduce environmental impacts, the processes can also lead to other environmental challenges. These include waste generation and the additional consumption of energy and raw materials, which raise concerns about the practical viability of these methods.

We reviewed and selected five previously reported studies on Pb recycling in PSCs that represent distinct Pb recycling processes and summarized them in Table 1. The life cycle inventory (LCI) of the overall materials and energy process is detailed in Tables S1–S5 (ESI†). The LCI inputs and outputs were derived from laboratory-scale literature data and assumptions, with each process designed to handle the recycling of 1 kg of PbI_2. Two of the most common impact categories were selected: global warming potential (GWP) and human toxicity potential (HTP). GWP is considered the most critical indicator in the energy field, as it measures the potential contribution of the recycling processes to climate change.[7] In contrast, human carcinogenic toxicity potential (HTPc) specifically addresses the environmental and health concerns associated with the presence of Pb in PSCs, making it a crucial factor in evaluating the safety and sustainability of Pb recycling methods.[7] We conducted LCA modeling using SimaPro 9.5.0.2 software in conjunction with the Ecoinvent 3.9.1 database. The environmental impacts were evaluated using the ReCiPe 2016 Midpoint (H) method, which include 18 impact categories.

Fig. 2 compares the LCA results in terms of GWP and HTPc across five different PbI_2 recycling methods, denoted as R1–R5 (Table 1). Results for other environmental categories are provided in Tables S6–S10 (ESI†). The GWP profiles in Fig. 2a reveal significant variation among the five recycling methods. Methods R1, R2, and R4, which incorporate DMF as a solvent, exhibit higher GWP values. Notably, R2 presents the most substantial GWP impact, primarily due to the use of multiple solvents, including DMF, DI water, and ethyl acetate (EA). The multi-step dissolution process in R2 results in cumulative contributions from each solvent, significantly elevating the overall GWP. Similarly, the GWP of R1 and R4 is significantly influenced by the use of organic solvents such as DMF. However, their lower total solvent usage, as detailed in Tables S1–S5 (ESI†), leads to a lower GWP compared to R2.

In contrast, R3, which utilizes hot DI water, shows the lowest GWP, highlighting the environmental advantage of water-based recycling methods. Even though R3 utilized hot water as the dissolution solvent, this did not significantly increase the overall GWP impact due to its relatively low temperature of 50 °C. Furthermore, it is evident that the impact of the adsorbent is minimal compared to the overall environmental impact. The cationic resin in the R4 method accounts for 1.05% of the total environmental profile, while whitlockite in the R5 method accounts for 0.0023%. Similarly, the HTPc profiles in Fig. 2c indicate the highest value for R2, primarily due to the use of multiple solvents, with DMF being a significant contributor. In contrast, R3 and R5 show significantly lower HTPc values, suggesting that water-based processes not only reduce global warming impacts but also mitigate potential human carcinogenic toxicity.

The contribution analysis further emphasizes the impact of solvent selection on both GWP and HTPc (Fig. 2b and d). In R2, DMF and EA contribute 39.59% and 34.15% to the GWP and 49.34% and 44.88% to the HTPc, respectively, underscoring their significant environmental burden.

Conversely, in R4, where isopropanol (IPA) is used in similar amounts to DMF (Table S4, ESI†), it contributes 19.49% to the GWP and 20.78% to the HTPc, which is relatively low compared to that of DMF, indicating that greener solvents can reduce the impact depending on the process specifics. This is further emphasized by the fact that the water used in all recycling processes contributed only 0.02% to 2.20%. Detailed LCA results for each solvent used in Pb recycling are provided in Table S11 (ESI†). The LCA results for human non-carcinogenic toxicity potential (HTPnc) are also presented in Fig. S1 (ESI†) and exhibit a trend consistent with those observed for GWP and HTPc.

The evaluated environmental impacts of all PbI_2 recycling methods (R1–R5) are significantly higher compared to the

Table 1 Pb recycling methods of Perovskite solar cell reported in previous researches

		Absorbents	Solvent	Recycle methods	Pb adsorption rate (%)	Pb recovery rate (%)	Ref.
R1	Zhang *et al.*	—	DMF	Dissolution by DMF and precipitation by NH_4OH and crystallization HI	—	~95.7	8
R2	Binek *et al.*	—	EA, water, DMF	Dissolution by EA, water and DMF and crystallization by cooling	—	~94	9
R3	Schmidt *et al.*	—	Water	Dissolution by hot-water and crystallization by cooling	—	~100	10
R4	Chen *et al.*	Ion-exchange resin	DMF	Dissolution by DMF and adsorption by ion-exchange resin and crystallization by NaI	~99.6	~99	11
R5	Hong *et al.*	Whitlockite	Water	Dissolution by water and adsorption by whitlockite and crystallization by KI	~100	—	12

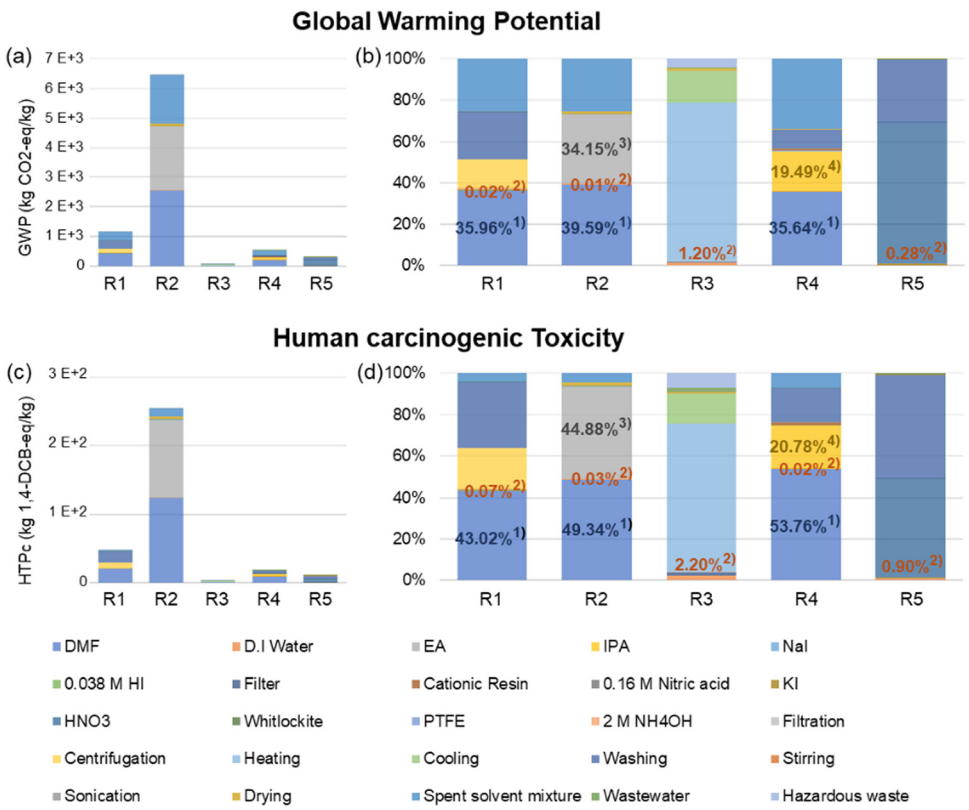

Fig. 2 LCA results and contribution analysis for PbI_2 recycling across five methods (R1–R5): (a) GWP profiles, (b) contribution analysis for GWP, (c) HTPc profiles, (d) contribution analysis for HTPc. (1) DMF, (2) D. I water, (3) EA, (4) isopropanol (IPA).

production of fresh PbI_2, which has a GWP of 8.35 kg CO_2-eq per 1 kg produced.[13] Although PbI_2 occupies a small volume within PSCs, the recycling process requires solvents based on the total volume of the PSCs. This leads to a significant use of organic solvents, which greatly contributes to GWP. However, it's important to consider that, in the PbI_2 recycling process for PSCs, the insoluble substrate and electrodes are typically recycled simultaneously. This simultaneous recycling complicates a direct comparison with the production process of fresh PbI_2, as it can provide additional environmental benefits by reducing the need for new materials. Nevertheless, the key to reducing the environmental impact of the recycling process lies in the efficient use of dissolution solvents.[14] The solvents used in PbI_2 recycling are typically discarded after Pb ions are removed during the recovery and crystallization process. However, they can be reused as dissolution solvents again at the EoL stage.

Fig. 3 shows that how much the environmental impact was reduced with the reuse of solvents. Detailed information on the number of solvent reuses and reduction late in environmental impact is provided in Tables S12–S14 (ESI†). After only 10 reuses, the GWP impact of the R2 method, which initially exhibited the highest value, is reduced by 89.55%, resulting in a GWP significantly lower than that of R1 (inset of Fig. 3a).

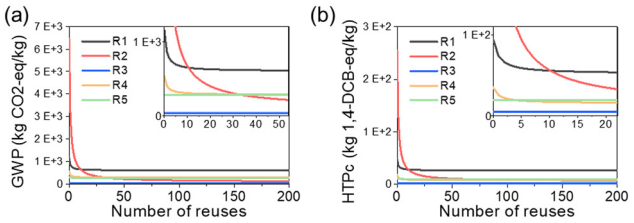

Fig. 3 LCA results of PbI_2 recycling methods (R1–R5) *versus* the number of dissolution solvent reuses (a) GWP, (b) HTPc.

Furthermore, after 34 reuses, it becomes lower than R4 and R5, indicating that R2 is more sustainable Pb recycling process than R1, R4, and R5 when solvents are reused. Conversely, for the R3 and R5, the reuse of water used during Pb recycling process has negligible effects compared to the other methods.

In the HTPc impact category, similar to the trend in the GWP impact results, R2 initially has the highest HTPc impact but significantly decreased with an increasing number of reuse (Fig. 3b and Fig. S2, ESI†). The number of reuses needed to reach the levels of other recycling methods is greater than that for GWP. This may be caused by the different contribution proportions of used solvents compared to GWP. Similar to GWP, the reuse effect is not significant for R3 and R5, where water is used as the dissolution solvent. The HTPnc impact category is also provided in Fig. S3 (ESI†).

Although increasing the number of solvent reuses can effectively reduce environmental impact, the limits of solvent reuse should also be considered. These limits can include factors such as solubility, solvent loss during the process, and concentrated residuals.[15] Prolonged processing increases environmental impacts due to higher energy consumption from extended equipment use. In addition, solvent evaporation during prolonged processing can further exacerbate these impacts. Furthermore, the trade-off between recycled PbI_2 quality and PSC efficiency after repeated recycling must be considered.[11] Another approach to reducing environmental impacts is to optimize the usage of dissolution solvents by utilizing their maximum solubility. Table S15 (ESI†) presents the solvents and their PbI_2 solubility used in the R1–R5 methods. In the R1, R2, and R4 methods, the solubility of DMF is not fully utilized. By optimizing this, it may be possible to develop a recycling process with a lower environmental impact.

In conclusion, we assessed five distinct PbI_2 recycling processes for PSCs from previous studies, with a focus on GWP and HTP through LCA. Our analysis indicates that the utilization of organic solvents such as DMF and EA substantially increases environmental impacts, whereas using water results in significantly lower impacts. Energy consumption and the use of additional adsorbents contributed minimally to the overall environmental footprint. Notably, in the R2 method initially exhibited the highest environmental impact. However, after reusing the solvent 10 times, the GWP impact was reduced by 89.55%, demonstrating the effectiveness of iterative solvent reuse. Thus, to advance a more sustainable PbI_2 recycling process for PSCs, it is imperative to minimize the use of toxic solvents. Additionally, implementing robust strategies for solvent reuse, thereby achieving significant reductions in environmental impacts.

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Data availability

The data supporting this article have been included as part of the ESI.†

Conflicts of interest

There are no conflicts to declare.

Notes and references

1 H.-S. Kim, Y.-J. An, J. I. Kwak, H. J. Kim, H. S. Jung and N.-G. Park, *ACS Energy Lett.*, 2022, **7**, 1154–1177.
2 H. J. Kim, G. S. Han and H. S. Jung, *eScience*, 2024, 100243.
3 L. Serrano-Lujan, N. Espinosa, T. T. Larsen-Olsen, J. Abad, A. Urbina and F. C. Krebs, *Adv. Energy Mater.*, 2015, **5**, 1501119.
4 J.-A. Alberola-Borràs, R. Vidal, E. J. Juárez-Pérez, E. Mas-Marzá, A. Guerrero and I. Mora-Seró, *Sol. Energy Mater. Sol. Cells*, 2018, **179**, 169–177.
5 X. Tian, S. D. Stranks and F. You, *Nat. Sustainability*, 2021, **4**, 821–829.
6 G. Rodriguez-Garcia, E. Aydin, S. De Wolf, B. Carlson, J. Kellar and I. Celik, *ACS Sustainable Chem. Eng.*, 2021, **9**, 15239–15248.
7 V. Muteri, M. Cellura, D. Curto, V. Franzitta, S. Longo, M. Mistretta and M. L. Parisi, *Energies*, 2020, **13**, 252.
8 S. Zhang, L. Shen, M. Huang, Y. Yu, L. Lei, J. Shao, Q. Zhao, Z. Wu, J. Wang and S. Yang, *ACS Sustainable Chem. Eng.*, 2018, **6**, 7558–7564.
9 A. Binek, M. L. Petrus, N. Huber, H. Bristow, Y. Hu, T. Bein and P. Docampo, *ACS Appl. Mater. Interfaces*, 2016, **8**, 12881–12886.
10 F. Schmidt, M. Amrein, S. Hedwig, M. Kober-Czerny, A. Paracchino, V. Holappa, R. Suhonen, A. Schäffer, E. C. Constable and H. J. Snaith, *J. Hazard. Mater.*, 2023, **447**, 130829.
11 B. Chen, C. Fei, S. Chen, H. Gu, X. Xiao and J. Huang, *Nat. Commun.*, 2021, **12**, 5859.
12 J. S. Hong, H. J. Kim, C. H. Sohn, O. Y. Gong, J. H. Choi, K. H. Cho, G. S. Han, K. T. Nam and H. S. Jung, *Energy Environ. Mater.*, 2022, **6**(3), e12374.
13 S. A. Khalifa, S. Spatari, A. T. Fafarman and J. B. Baxter, *ACS Sustainable Chem. Eng.*, 2020, **8**, 16537–16548.
14 G. Rodriguez-Garcia, E. Aydin, S. De Wolf, B. Carlson, J. Kellar and I. Celik, *ACS Sustainable Chem. Eng.*, 2021, **9**, 15239–15248.
15 H. J. Kim, O. Y. Gong, Y. J. Kim, G. W. Yoon, G. S. Han, H. Shin and H. S. Jung, *ACS Energy Lett.*, 2023, **8**, 4330–4337.