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Revive or reuse: Environmental and energy insights into mesoporous perovskite solar cells recycling

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ABSTRACT

With growing interest in perovskite solar cells, identifying the optimal recycling strategy becomes important. Strategies focusing on revival or layer-reuse were introduced. Yet their energy footprints and environmental impacts should be understood. Thus, revival – the highest level of recycling – and layer-reuse of carbon-based mesoporous perovskite solar cells were modelled alongside the revival of NiO- and Au-NiO-based cells. Subsequent analysis focused on energy return on energy investment combined with life cycle assessment investigating systems from cradle to end of second-life (second-life after recycling). The results showed that revival had higher return of energy (43) while having lower impacts (as low as 8 g CO_2-eq./kWh) compared to reuse of ZrO_2 electrode (as low as 8.8 g CO_2-eq./kWh). However, the degradation and lifetime of second-life devices might render layer-reuse more advantageous, highlighting the small nuances between different recycling levels. Additionally, the environmental impacts of nickel-based cells were significantly higher compared to carbon-based cells. Based on these findings future perovskite solar cell research could target effective revival or reuse.

1. Introduction

The resources available on earth are being dissipated [1]. There is growing demand for metals, especially for the energy transition and renewable electricity generation [2–4]. Solar panels are no exception, as they often require valuable metals such as silver or copper [5], which altogether rose the debate about sustainability of green technologies [6] and increased the interest into circular economy [7–10]. One way forward would be to limit the use of scarce and valuable materials, which perovskite solar cells (PSCs) promise [11]. However, the perovskite crystals often contain lead, which mandates proper end-of-life handling to avoid lead leakage [12]. Furthermore, perovskite cells are prone to degrade much faster than Si-based solar cells, raising the need to explore techniques to prolong their lifetime to succeed in their commercialization [13,14]. Thus, besides stabilization and encapsulation, they are good candidates to integrate design for the circular economy in early stages of their commercialization [11].

Within the circular economy, the concept of design for recycling is a way to keep materials in use for as long as possible [15]. Recycling should be targeted once direct reuse is not viable. However, not all recycling has the same value [16], thus, to better understand the levels of recycling, a recycling hierarchy for energy systems was introduced [17]: It stated that the higher the value of the product of recycling, the lower the resource consumption to perform that recycling. In the case of solar panels, the highest value would be revival of the degraded panel – synonymous with refurbishment or repair – and it would require the least energy. Lowest in the hierarchy lies the recovery of elements – like gold, silver or copper. Recovery of elements requires most resources because of the energy intensive liberation, separation of elements and subsequent purification.

In the context of PSCs, “revival” refers to replacing the degraded perovskite crystals with fresh perovskite Fig. 1b. In such a case, the entire structure that hosts the perovskite, as well as the energy invested in producing that structure, is retained. Thus, the product of recycling carries highest value. Meanwhile, component reuse involves retaining parts of the mesoporous scaffold, such as an electrode deposited on glass, while replacing other elements, including the carbon back-electrode and the degraded perovskite layer see Fig. 1a. As can be seen, the remanufacturing of cells utilizing reused parts already requires replacing more parts of the cell and likely more energy. While the simple hierarchy gives some indication, the reality of the recycling process is more complicated. For instance, the performance and lifetime of the recycled devices needs

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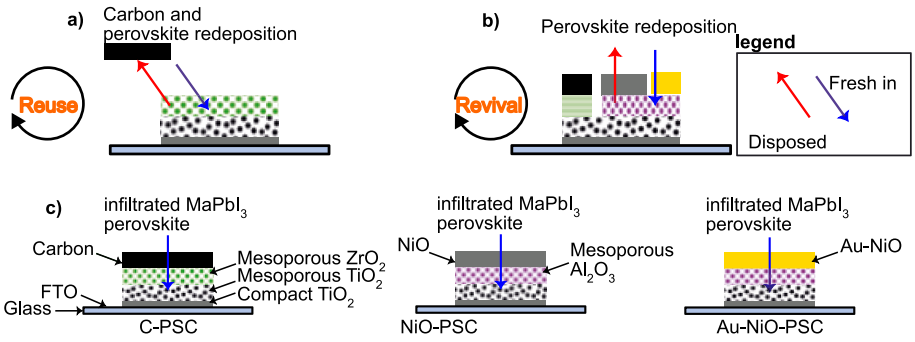

Fig. 1. Schematics depicting a) C-PSC - reuse of ZrO_2 electrode - perovskite and carbon electrode are removed and redeposited and the rest is reused. b) Revival of C-PSC, indicating that only the perovskite is removed and redeposited, while the scaffold remains unchanged. c) Composition of all the studied structures - C-PSC, NiO-PSC, Au-NiO-PSC (FTO - fluorine doped tin oxide. MAPbI3 - methylammonium lead iodide).

to be accounted for.

For experimental attempts to revive PSCs, a noticeable decrease of efficiency was reported [18–20]. However, reusing parts of the cell could potentially result in better retention of the initial efficiency [21] and overall higher cumulative electricity production. To link the consumed resources (i.e., energy expended during fabrication of an energy source) with the potential delivered energy; energy return of investment (EROI) [22] was conceptualized. This technique has already been used to analyse the manufacturing of silicon and thin-film photovoltaics [23]. It enabled connecting the expended energy during manufacturing with the electricity later delivered by the solar panels. Moreover, EROI can and should account for the efficiency of the panels and their degradation over time. Celik et al. [23] showed that some thin-film cells outperform Si-based panels, while perovskite fell short in their comparison. Yet energy is not the only resource consumed; for example, materials and land for the solar panels are as well.

To understand the potential environmental load of materials used during PSC manufacturing, life cycle assessment (LCA) has been often utilized. For instance, LCA was used to study the influence of scaled-up manufacturing [24–27] or to analyse the influence of recycling [28–30]. Moreover, early design has been assessed using LCA [31,32]; however, studies assessing cradle to cradle are scarce [21,33]. The scarcity of holistic studies and the lack of comparison across different PSC recycling levels represent a significant gap in current scientific knowledge.

Importantly, there is a gap in understanding energy demands and environmental impacts of higher levels of recycling of PSCs. Especially what is missing is a harmonized comparison of revival against reuse of layers, see Fig. 1. Thus, the following research focuses on filling this gap and sets out to test whether the simplified hierarchy of recycling upholds under detailed scrutiny. EROI analysis, together with analysis of life cycle impacts, is conducted across two potential lifetimes of mesoporous PSCs. The mesoporous scaffold architecture of PSCs, with infiltrated perovskite, enables revival of the perovskite layer [18–20]. Therefore, they are subject of present analysis. Moreover, the scaffolds could be reused (carbon electrode is replaced in addition to replacing the perovskite) besides revival [21], which enables comparison of the two highest levels of recycling [17]. There are more efficient PSC documented [34], yet mainly mesoporous PSC offer the possibility to compare the revival with partial reuse of the cell- "reuse of the ZrO_2 electrode".

Finally, the study was narrowed down to TiO_2/ZrO_2/Carbon- [21,35] (C-PSC), TiO_2/Al_2O_3/NiO- [18] (NiO-PSC) and TiO_2/Al_2O_3/Au-NiO-scaffolds [19] (Au-NiO-PSC), as for these architectures robust experimental data was available. For schematics of the studied PSC structures see Fig. 1. Building on present modelling work, the research community could target designs which enable revival or reuse of layer and evaluate, for instance, the technoeconomic implications as well as social impacts of higher levels of recycling. Furthermore, the detailed methods and results of EROI and LCA analysis are presented and then discussed.

2. Methodology

In present modelling work, HSC chemistry [36] was utilized to model the manufacturing and recycling of three different types of mesoporous PSCs: carbon-based [21,37], AuNiOx [19] and NiOx [18](Fig. 1 shows the structure of the solar device, while Fig. 2 shows the unit processes in the systems). Later on, the process data was used as part of the life cycle inventory to be utilized for LCA in OpenLCA 2.4.1 [38] complemented by Ecoinvent database [39]. Using extensive thermodynamic database in HSC chemistry, the enthalpy balance of each step was calculated to estimate the energy needs. Such detailed description is vital for detailed energy flow analysis. Not all data on perovskites and some of the solvents was available in the software or Ecoinvent database, thus the missing data was modelled according to previous literature and implemented in the software (see supplementary information).

2.1. System description

For the present analysis four different system models were constructed:

1. Revival of C-PSC (following Akulenko et al., [20]). Revival was modelled so that only the degraded perovskite was replaced with a fresh perovskite.
2. Reuse of ZrO_2 electrode – (following Bogachuk et al., [21]), Reuse of ZrO_2 electrode was modelled so that the scaffold wass reused, however the back-electrode was replaced by a fresh one. The perovskite was replaced as well.
3. Revival of NiO-PSC (following Ku et al., [18])
4. Revival of Au-NiO-PSC (following Li et al., [19])

Revival here refers to the action of removing degraded perovskite and replacing it with fresh material to enable the second life of the devices, as demonstrated for the carbon structure in Fig. 2 (for other systems, see Supplementary Figs. S1 and S2). Reuse of the ZrO_2 electrode means replacing the perovskite layer and rear carbon electrode with pristine carbon layer and perovskite, thus including additional steps beyond revival. Each system consists of manufacturing, the use phase, recycling—either revival or reuse—and the second use phase, see Fig. 2 where each process step for C-PSC (for all structures refer to Figs. S1 and S2) is presented. First use phase was fixed to last 5 years as a baseline [29], while 10 and 25 years were also modelled to reflect possible improvements in stability. The second use phase had variable duration, always starting from 0 years and ending at a maximum equal to the first use phase (i.e., 5, 10, or 25 years accordingly). These models are built on the available literature and are described in further detail in the supplementary document.

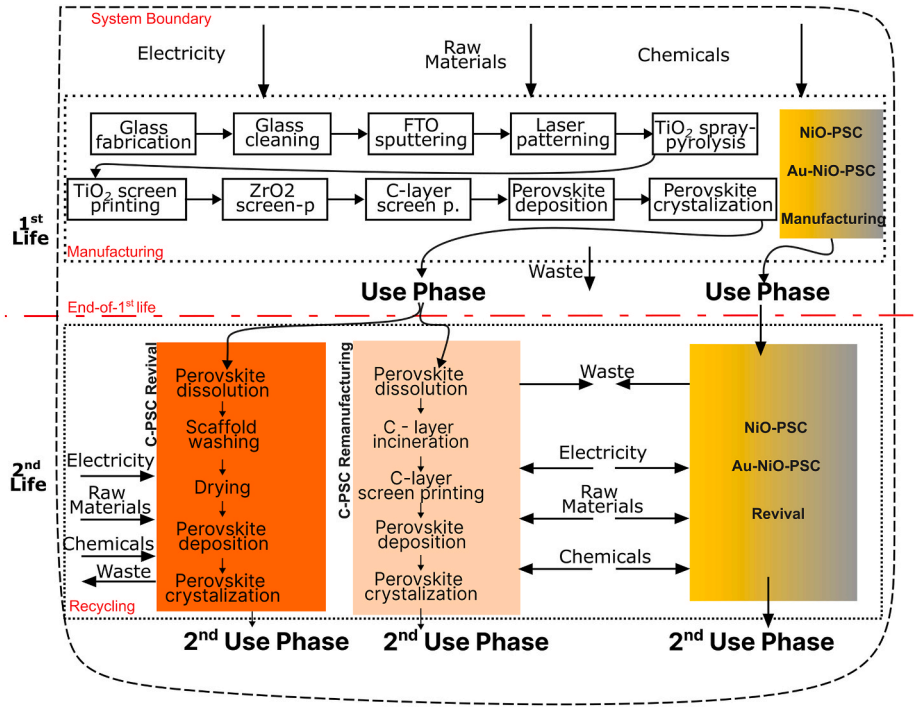

Fig. 2. Overview of the systems under study, simplified to depict details only for the C-PSC revival (following [20]) and ZrO_2 electrode reuse, following [21]). NiO- and Au-NiO-PSC only shown as "black boxes", while their brake-down of process steps are in Supplementary Figs. S1 and S2. The upper part of the figure shows manufacturing processes, the red dashed line in the middle shows the end-of-1st life after which recycling (revival or reuse) follows – bottom part of the figure. All the systems end after 2nd use phase. No transportation involved.

2.2. Energy return on energy invested

EROI was calculated to visualize the impacts of reuse or revival. By calculating EROI, the influence of especially second-life efficiency, lifetime and energy expenditure can be evaluated comparatively. EROI was calculated as follows:

$$\mathrm{EROI} = \frac{\mathrm{E_{Out_{first}}} + \mathrm{E_{Out_{second}}}}{\mathrm{E_{manufacturing}} + \mathrm{E_{EOL}}} \quad (1)$$

Where the numerator is the electricity produced over the assumed two lifetimes (E_{Out_first}; E_{Out_second}) and the denominator sums the electricity consumed during manufacturing and end-of-life (EOL) treatment ($E_{manufacturing}$; E_{EOL}). The electricity production was calculated as follows:

$$\mathrm{E_{Out_{first}}} = \mathrm{I{\cdot}A{\cdot}UF{\cdot}t{\cdot}}\eta_{\mathrm{first}} \quad (2)$$

Where I is the average daily insolation (assumed as 1700 kWh/m^2 per year [23]) t is the number of years of operation during the first cycle(5, 10 or 25 years), A is the total active area of the modelled cells, η_{first} is the maximum efficiency of the cell during the first cycle. UF is the utilization factor (UF = 0.8). The electricity production was linearly decreased during the first lifetime to 80% of initially efficiency at the end of the first lifetime period. Similarly, electricity production can be calculated for the second cycle:

$$\mathrm{E_{Out_{second}}} = \mathrm{I{\cdot}A{\cdot}UF{\cdot}t{\cdot}}\eta \quad (3)$$

Where the efficiency η and lifetime t were varied from zero to the initial maximum (i.e. 5, 10 or 25 years depending on the first life) to visualize impacts of efficiency losses and lowered life expectancy for revived or refurbished solar cells. Furthermore, scenarios were generated to explore the effects of extending lifetime and increasing efficiency—up to 25 years and 25%, respectively—for both potential lifetimes (before and after recycling). The electricity was accounted for as is, omitting the adjustment to primary energy. This way EROI showed how much electricity was returned to the society using the solar cells compared to electricity diverted from other potential uses [40].

2.3. LCA methodology

2.3.1. Goal and scope definition

Following ISO 14040, the goal of the present LCA was to establish which mesoporous structure and approach to recycling yield lower impacts per generated electricity over two lifetimes. The impacts of reviving carbon-, Au-NiO-, and NiO-based mesoporous structures were compared against each other. Furthermore, the comparison included the reuse of ZrO_2-electrode from the C-PSC. These two approaches to recycling were benchmarked against the same-structured non-recycled cells (single-use cells). The impacts were evaluated using a functional unit of: "1 kWh of electricity produced by the mesoporous PSC over two lifetimes" (calculated using equations (2) and (3)). Production and recycling were assumed to be carried out in the same location in Europe, with processes assumed to take place in 2025. We omitted forecasting the recycling into the future across the expected lifetimes in order not to over-complicate the present study. The study aims to inform the general solar cell research audience.

The present assessment focused on the cradle-to-end-of-2nd-life for each of the devices and recycling strategies, following the attributional LCA. The final treatment after the end-of-2nd-life was omitted, as the final disposal of the studied cells is not yet established. The studied systems and their respective system boundaries are presented in Fig. 2. Possible impacts of waste streams were attributed to the cells, so no benefits of potential waste recycling were assumed. (For the single-use cells used as a benchmark, the cut-off is after the end-of-1st-life; no recycling was assumed, as highlighted by the red line in Fig. 2). The use of recycled materials during production was avoided in the model. Transportation was left out of the model to focus solely on the manufacturing and recycling processes, which might be obscured by inclusion of transportation. Similarly, the impacts of the use phase (electricity generation) are usually negligible [41], so they were not

accounted for.

2.3.2. Life cycle inventory and data collection

Sources of data for the life cycle inventory are detailed in Supplementary Tables S1 and S2. Generally, each process starts with the sourcing of raw materials, followed by the manufacturing phase, the use phase, and then revival or reuse, as shown in Fig. 2. Furthermore, the inventory data for the materials entering the described process was obtained from Ecoinvent database using Cut-off system model [39].

2.3.3. Life cycle impact analysis

The life cycle impact assessment followed the ReCiPe 2016 methodology with the hierarchist perspective [42]. The assessment focused on the midpoint categories embedded in the method. Emphasis was placed on global warming potential because solar cells are termed as low-carbon technology. Moreover, toxicity-related impact categories received special attention, as they are of interest due to the presence of lead in the perovskite.

The systems were modelled using openLCA software. The impacts initially calculated per m^2 of cells were modified to follow the functional unit based on equation (4), where E_{Out} – energy delivered, were calculated following equations (2) and (3). The impacts were then presented as a function of second-life efficiency and second lifetime, similarly to Celik et al. [43] to study the effects of degradation and efficiency loss during the second lifetime.

$$\text{Impact per kWh} = \frac{\text{Impact per m}^2}{\left(E_{Out_{first}} + E_{Out_{second}}\right)} \quad (4)$$

A sensitivity analysis of energy consumption during sputtering was conducted to visualize its influence as there was initially large variations in the background literature. Furthermore, the influence of prolonging the lifetime from the initial 5 years to 10 or 25 years was tested, as well as potential improvements in cell efficiency up to 25%.

3. Results

3.1. Energy return on energy invested

EROI highlights the scale of energy return that each energy conversion device, in this case the solar cell, can provide during its use. In this work, the direct manufacturing and remanufacturing (revival or reuse of layers) electricity was compared to the potentially delivered electricity through the first and second life of PSCs. This comparison was used to analyse the benefits of different levels of recycling and between different PSC structures.

The visualisation in Fig. 3 presents contour plots in which the colour gradient indicates increasing EROI as a function of second-life lifetime and second-life efficiency (note that the first lifetime was fixed at 5 years, and the initial efficiency was assumed to decrease linearly over that period to 80% of its initial value, yielding a minimum EROI of 17.5

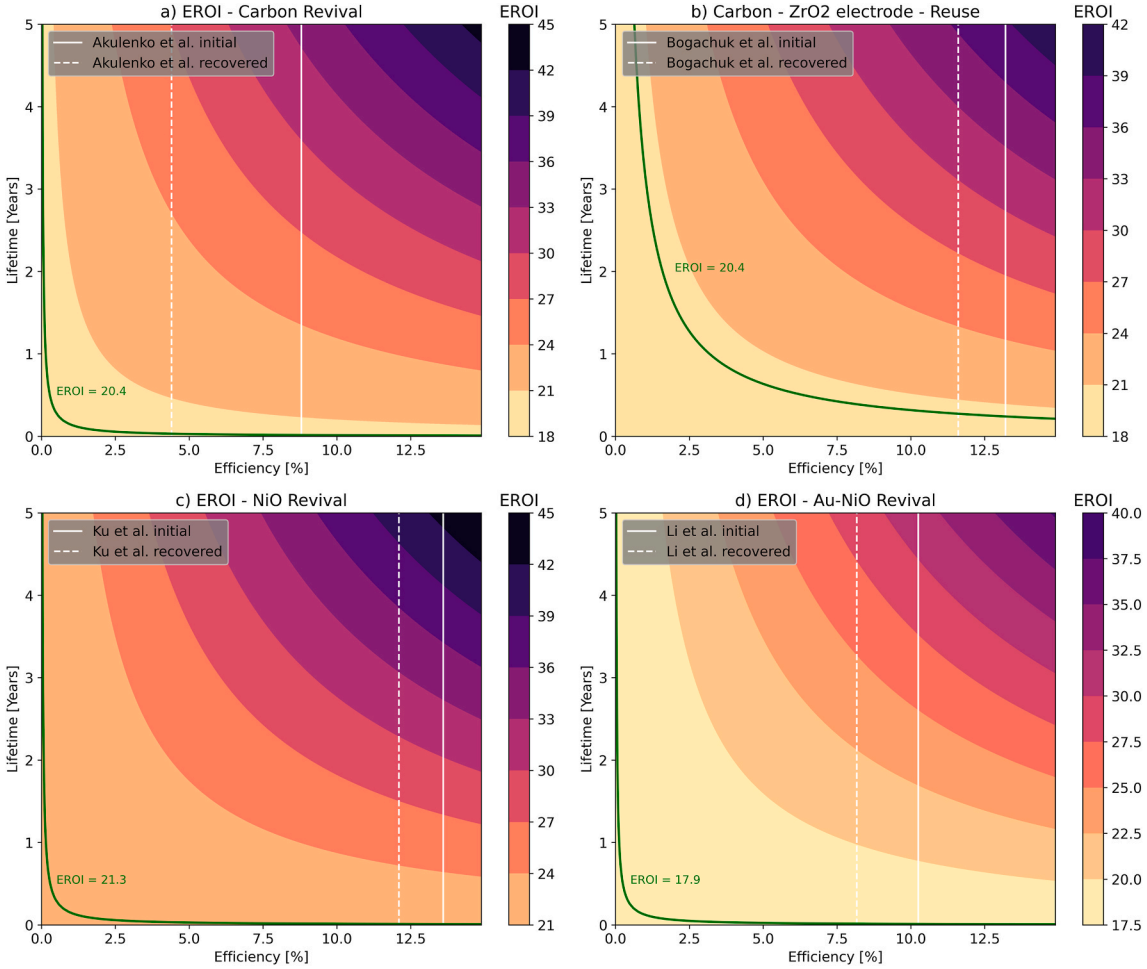

Fig. 3. EROI values for the 4 simulated process. a: C-PSC revival; b: C-PSC layer reuse; c: NiO-PSC structure revival; d: Au-NiO-PSC based structure revival (The green line in each of the plots highlights the maximum EROI value of a pristine device. The dashed white lines highlight efficiency after refurbishment – reuse of the TiO_2-ZrO_2 electrode - or revival, while the white lines highlight initial reported efficiency).

up to 21). The purpose of the colour gradient is to visualize scenarios with varying second-life efficiency and lifetime. The right-hand side of the plots in Fig. 3 represents the most ideal case, in which the highest efficiency reported for C-PSC (14.89%, applied to all modelled systems) is maintained during the second life.

As seen from Fig. 3 all recycling approaches resulted in an overwhelming net benefit (EROI values larger than 1, assuming standard-performing devices during the first life). The benefit increased as both lifetime and efficiency increase. Over the maximum estimated combined lifetime (5 + 5 years), the most beneficial was the revival of C-PSC (EROI of 43), while the Au-NiO-PSC had the lowest return (EROI of max. 37). This lowest EROI was attributed to the energy-intensive manufacturing, especially the back-electrode deposition done via thermal evaporation (8.7 kWh per m^2, adjusted from Ref. [24]). In between, yet very close to each other, were the revived NiO-PSC (41.24) and the reuse of ZrO_2-electrode (41.15).

Furthermore, EROI allows for determining a tolerable decrease in efficiency and lifetime of the second life devices compared to their pristine counterparts. EROI of using two pristine devices is illustrated with the green solid contour lines in Fig. 3a–d. The area above the contour line represents conditions where recycled devices provided a higher return of energy. Thus, focusing on all the revival systems - Fig. 3a, b, c – reviving the solar cells had a profound benefit over pristine devices. This benefit arose from lower electricity consumption during the washing-out and redepositing of the $MAPbI_3$ perovskite compared to the manufacturing electricity consumption. In contrast, the benefit was less profound if the carbon layer was removed and then redeposited. Consequently, Fig. 3b (right edge of the plot) shows that to surpass the pristine devices, the cell with the reused ZrO_2-electrode needed to operate for at least ¼ of a year at the highest efficiency, which is already in the realm of current perovskite stability [44]. Thus, strictly from the energy return point of view, there is significant room for a decrease in the second-life efficiency and lifetime.

When shifting the focus on the levels of recycling [17], it is worth comparing the reuse of the ZrO_2-electrode with full revival of the carbon devices. Based purely on the maximum EROI, the revival achieved higher return of energy, see right-hand upper corner of Fig. 3a. However, comparing the reported experimentally achieved efficiencies (solid and dashed white lines in Fig. 3), it is clear that currently the reuse can provide higher benefit. Based on the experimental studies the revived cell struggle with efficiency decrease, thus if both cells were operated for the same time the reused cell starts to have higher EROI after around half a year.

Continuing to focus on the reported efficiencies of recycled devices, the exact break-even points with pristine counterparts could be evaluated. The break-even points can be found in Fig. 3 where the green-contour lines cross the vertical dashed lines. For instance, Li et al., [19] report revived efficiency of 8.17 % for the Au-NiO structure, thus for a revived device to provide higher EROI, it would need to last a fraction of a year after revival. For both C- and NiO-PSC this would be negligible as well. In contrast, the reusing only the ZrO_2 -electrode from C-PSC required a longer time to break-even. The break-even point was after around 1/3 of a year of operation (based on efficiencies reported by Bogachuk et al., [21]) due to the energy investment for carbon layer remanufacturing. Altogether, EROI predictions combined with previously achieved efficiencies showed a good trajectory for revival or reuse of the ZrO_2-electrode, offering higher EROI than using only pristine devices.

3.2. Results of life cycle assessment

Following the ReCiPe 2016 midpoint methodology [42], the revival process of the NiO-based structure resulted in the highest potential impacts per kWh of delivered electricity. The biggest contributor was the nickel mesoporous layer, particularly the impact of the extraction and purification of nickel ore to produce pure nickel. AuNiO-PSC had highest impacts in freshwater and marine ecotoxicity, freshwater eutrophication and non-carcinogenic human toxicity, which is linked to the use of gold and the impacts of gold mining and purification, which surpasses the impacts of nickel production in the NiO-PSC system.

On the other hand, C-PSC and their reuse or revival offered the lowest impacts. This was mainly due to the use of carbon which had lower potential impacts than the nickel and gold used in the other two structures. Revival of the carbon structure offered lowest impacts per m^2 of the cell as compared to the reuse. The reason was that the carbon layer was preserved, thus avoiding the impacts of burning and redepositing carbon. However, to address potential losses in efficiency or decreased lifetime, the Global warming potential per kWh of electricity produced were analysed in following chapter.

Focusing on the ecotoxicity (freshwater, marine and terrestrial) and human toxicity impact (carcinogenic and non-carcinogenic) categories it was found that the lead counting perovskite did not contribute significantly towards these impact categories (see supplementary excel tables). For the metal-based structures mostly gold and nickel had the highest share followed by glass processing.

In general, the main impacts stemmed from the initial manufacturing, whereas the recycling had a comparatively low influence on the overall results (see the impact brake-down in supplementary excel table). The impacts were slightly increased for the reuse of the ZrO_2 electrode from C-PSC, as there is an increased need for materials to redeposit the carbon and to burn off the old carbon layer.

3.3. Cumulative carbon emissions

Even though solar cells are considered a renewable technology, they carry embodied carbon emissions. To provide a different perspective in the comparison of the three structures and the two end-of-life approaches. The cumulative Global Warming Potentials (GWP) are shown in Fig. 4 following similar logic as the EROI analysis.

At first, it becomes apparent that there is up to two orders of magnitude difference in possible impact between C-PSC (Fig. 4a and b) and NiO- or AuNiO-PSC (Fig. 4c and d). Generally, the lowest potential impacts stemmed from the revival of the carbon structure, while the ZrO_2-electrode reuse had slightly higher impacts. On the contrary, NiO-structure revival had the highest possible impact, mostly because of the emissions related to nickel in the screen-printed NiO electrode. Au-NiO-PSC fell in between the other two structures.

Analogous to EROI analysis, the break-even points with two pristine devices can be found and analysed in Fig. 4. Altogether, reviving PSCs led to lower global warming potential per kWh of electricity supposedly produced. On the other hand, ZrO_2-electrode reuse required over a year of operation to break even, assuming retention of the initial efficiency after reuse (see Fig. 4b – green contour line). This shortcoming was linked to the CO_2 release during decomposition of the carbon electrode and emissions related to the redeposition of the carbon electrode. Among the revived cells, NiO-PSC required seemingly the least time to overcome two pristine devices because the manufacturing GWP was much higher than the impacts related to the revival. On the contrary, the carbon revival required around 1/4 of a year to break even, as the impacts of revival (0.44 g CO_2-eq./kWh) were considerably lower relative to the impacts related to manufacturing (7.62 g CO_2-eq./kWh).

3.4. Sensitivity to prolonged lifetime and increased efficiency

Currently, the mesoporous PSCs suffer from short lifetimes and lower efficiencies compared to the state-of-the-art silicon solar panels; thus, early repair seemed advantageous [14]. However, prolonged lifetime and increased efficiency can be expected as the technology develops. Therefore, scenarios with prolonged lifetimes and increased efficiency were analysed (Supplementary Figs. S3–S5). If only the lifetime of the devices was increased, EROI increased, and the environmental impacts per produced kWh of electricity decreased. Same applies if the efficiency

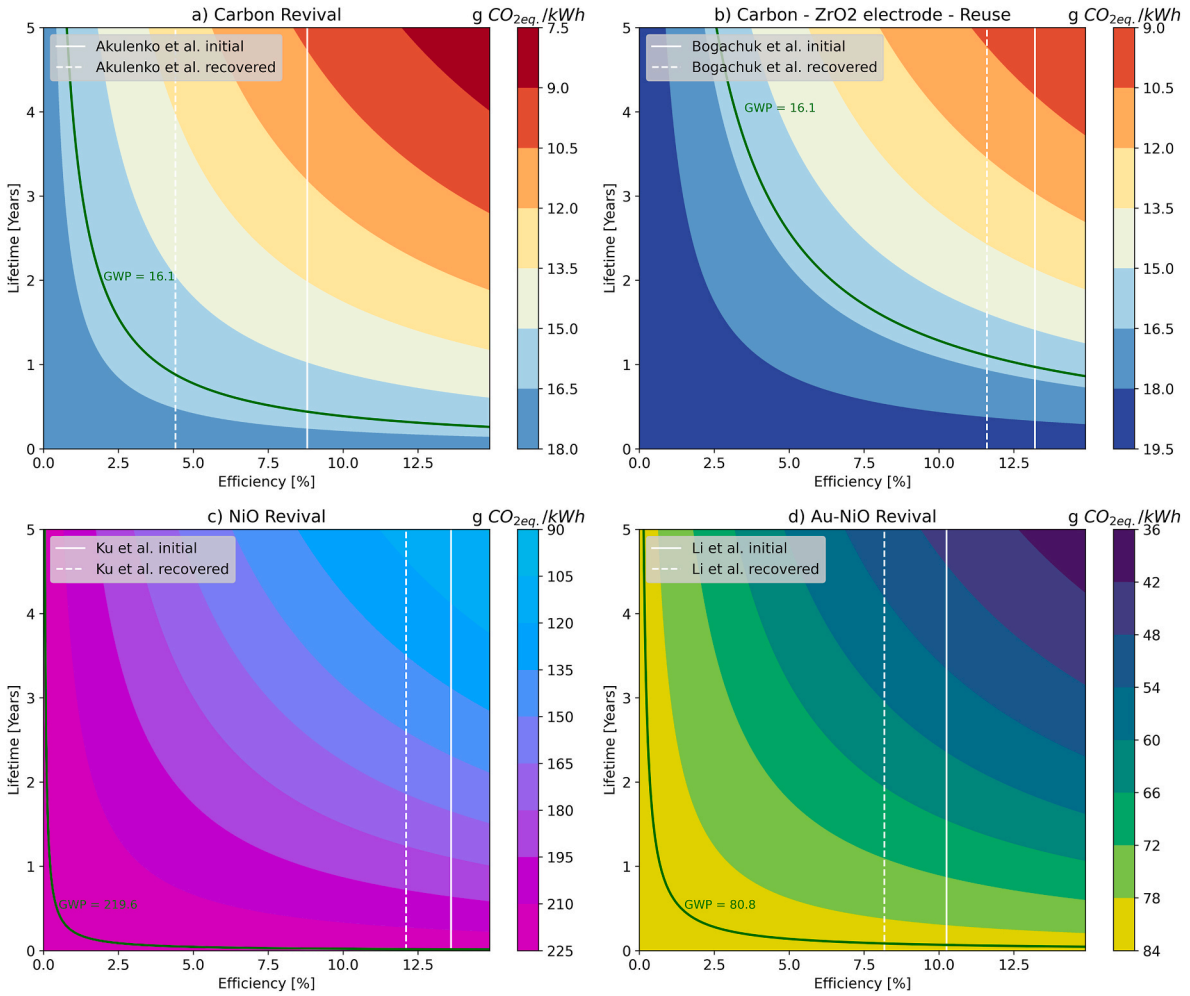

Fig. 4. Global warming potential shown as a function of efficiency and lifetime. a: C-PSC revival; b: C-PSC layer reuse; c: NiO-PSC structure revival; d: Au-NiO-PSC based structure revival (The green line in each of the plots highlights the maximum EROI value of a pristine device. The dashed white lines highlight efficiency after refurbishment or revival, while the white lines highlight initial reported efficiency).

would reach 25 %; then EROI goes up and environmental impacts go down. Such improvements were expected, because the cumulative electricity production is directly connected with device lifetime and efficiency.

What was interesting, though, was that the advantage of long-lasting well performing devices over two pristine devices grew linearly over time. For example, a revived carbon device can have an EROI 25 units higher than two pristine carbon devices during 5-year lifetime. If it lasts 25 years, the difference is up to 125 units. Thus, the benefit multiplies at the same rate as the lifetime increases (i.e., 5 times longer lifetime - 5 times higher EROI difference), mainly caused by the constant increase in produced electricity as the lifetime increases. Thus, strictly from an energy return and relative environmental impact point of view, revival or reuse would suit even long-lasting devices.

4. Discussion

The present work set out to compare revivable and re-manufacturable mesoporous PSC including Carbon-, NiO- and AuNiO-based cells. The comparison included not only different structures but also benchmarked the highest levels of recycling hierarchy [17]. For this benchmarking, the analysis included the concepts of EROI and LCA to illustrate the potential benefits and impacts respectively. Thus, this comparison aimed to direct the research towards more circular eco-design solutions [35,45].

Starting with EROI results, revival was more beneficial than reuse of the ZrO_2-electrode or manufacturing new cells. This benefit was evident because the revived devices required lower efficiencies and lifetimes to outperform pristine devices (See Fig. 3, where along the green contour line the efficiency-lifetime combinations can be deduced). This is mainly because the assumed net energy spent on revival was comparatively small (~0.1 kWh/m^2 of electricity). On the other hand, for C-PSC, reuse of the ZrO_2 electrode in the current state of research was a more viable option. This is because the reported efficiencies after revival were low [20], while the reuse of the ZrO_2-electrode reported higher second-life efficiencies [21]. Looking at the white vertical lines and their intersection with the upper edge of the graph (representing current knowledge of reuse and revival) in Fig. 3a and b, the reuse of ZrO_2-electrode would provide higher EROI in 5 years than the experimentally revived devices. On the contrary, due to the significantly lower energy consumption, the revival process would become more favourable once higher efficiencies of revived devices are achieved. From a strictly thermodynamic perspective, less materials were dissipated compared to ZrO_2-electrode reuse, thus avoiding unnecessary material degradation [16,46,47].

However, the cell efficiency after revival or refurbishment is only one side of the problem, while the degradation rate of the second-life devices is another. Given that the difference in maximum EROI between revival of C-PSC and the reuse of the ZrO_2-electrode is only around 2 units, degradation could play a decisive role. A steeper degradation rate after revival would erase the marginal benefit. The threshold difference in

lifetime degradation that would still justify revival over reuse of the ZrO_2 electrode was around 8%. This means that, in an example where the C-PSC after ZrO_2-electrode reuse does not degrade, the revived cell must retain at least 92% of its initial performance at the end of the second lifetime (assuming linear degradation and equal lifetimes) to be justifiable. A similar trend can be observed in Fig. 4, where the relative CO_2 emissions of the best-performing device after revival or reuse of the ZrO_2-electrode are close to each other. Therefore, experimentally verifying the exact degradation rates of second-life devices is required to determine if revival can outperform ZrO_2-electrode reuse in real-life settings.

Moving to solutions that could increase stability and reduce degradation, it is worth discussing encapsulation [13,48,49]. Even though it was not modelled herein, it is useful to consider how it would affect the EROI and LCA performance of the studied cells. Encapsulation, in the traditional sense—i.e., laminating with polymers such as ethylene-vinyl acetate (EVA) or polyolefin elastomer (POE)—will decrease EROI and increase environmental impacts. These increases would stem from additional processing and materials. Revival of encapsulated cells would require resealing the device and thus more energy inputs than reviving non-encapsulated cells. If the polymeric encapsulant must be removed completely to carry out revival, then the only moderate efficiency after revival might not justify the effort. In such cases, removing the polymeric layers and replacing the carbon electrode to achieve higher efficiency would be more beneficial [21]. To summarize, encapsulation will influence both EROI and LCA results, as encapsulation and its treatment during recycling will require additional resources.

Continuing with the prism of combined EROI and LCA, the importance of material choice becomes apparent. For instance, EROI of the reuse of the ZrO_2-electrode and revival of NiO-PSC were close to each other (see Fig. 3), however, the mostly material related GWP disadvantaged NiO-PSC significantly (see Fig. 4). It could be argued that a reduction of material use – especially nickel – provides some benefits as the impacts of the less nickel intensive Au-NiO-PSC were lower (see Fig. 4c and d). Yet, the impacts of Au-NiO-PSC were still 5-fold compared to the carbon-based devices (see Fig. 4a and b). Moreover, the reported AuNiO-PSC efficiency [19] hinders its EROI compared to all the other structures. Therefore, the relative decrease in GWP compared to NiO-PSC was outweighed by the lower EROI. Altogether the nickel containing cells would need to last significantly longer to outperform the C-PSC.

Additionally, it is worth comparing mesoporous PSCs with silicon solar panels as the state-of-the-art technology. Focusing just on GWP, Khan et al. [50] reported relative impacts in range between 6.6 and 54.8 g CO_2-eq per kWh, while Lunardi et al. [41] show GWP below 20 g CO_2-eq per kWh (using different impact assessment methodology). Even though both assumed lifetime of silicon solar panels in range from 25 to 40 years, much longer than the 5 + 5 years of PSC assumed herein, the carbon devices and Au-NiO devices could perform similarly to current silicon solar panels. Important caveat to this is that PSC in general struggle with stability [44], thus reaching 5 years of operation is difficult. This would significantly lower their competitiveness, in terms of environmental footprint, with Si-solar panels. On the other hand, the recycled carbon devices (see bottom of Fig. 4 a),b) where the GWP for C-PSC was around 19 g CO_2-eq per kWh) are competitive even if they fail early on. Thus, securing stable and revivable or remanufacturable (where part of the scaffold can be reused) structures becomes important to maintain potentially low GWP of PSCs.

Putting the EROI in context with other technologies, the perovskite revival or the ZrO_2-electrode reuse resulted in a higher overall EROI. As found across various studies, monocrystalline silicon solar panels have a mean EROI of 8.7 [51], which is significantly lower than perovskites studied herein. The closest to the EROI reported within present work may be CdTe solar panels, with an EROI of 34.2 [51]. Furthermore, Celik et al. [23] provided a comparison of tandem perovskite–perovskite and perovskite–silicon panels. They showed perovskite-silicon tandem EROI of 5.2 whereas this work presented mesoporous-PSC EROI between 17 (pristine Au-NiO-PSC, see Fig. 3) to 45 (revived C-PSC). The main reason for the difference between the present work and the referenced studies is reduced energy consumption during manufacturing. Furthermore, the present study used direct manufacturing electricity consumption to adhere to Arvesen & Hertwich [40] in contrast with primary energy demand used in both referred studies [23,51]. Recalculating to primary energy would lower the EROI; thus, the comparison here is indicative. The main goal of the present EROI calculation was to compare the different recycling strategies and cells against each other.

4.1. Sensitivity analysis

While searching for the energy and material consumptions, the authors came across significant variation in electricity consumption during, especially, the FTO-deposition. Values ranged from a maximum of 6.67 kWh/m^2 [52] down to 1.39 kWh/m^2 [21] (adopted herein) or even a minimum of 0.67 kWh/m^2 [26]. Therefore, the influence of the assumed electricity consumption during sputtering on GWP and EROI was analysed and is compared in Fig. 5.

Interestingly, the influence of a six-fold increase in electricity consumption during FTO sputtering resulted in only ~15% increase in GWP in the case of carbon revival, as shown in Fig. 5a. The same increase had a smaller influence on the ZrO_2-electrode reuse, resulting in a 10% GWP increase. In the case of ZrO_2-electrode reuse, the total CO_2 emissions are higher compared to revival due to the removal of the carbon layer thus electricity consumption had a smaller influence. The lowest impact was observed in NiO-PSC, less than one percent, due to the high GWP related to the use of nickel. Further reductions in electricity consumption had only a marginal effect, especially on the NiO- and Au-NiO-PSC (the minimum assumption in Fig. 5a).

However, looking at Fig. 5b the influence of the sputtering electricity consumption had higher influence on EROI. Increasing consumption lowered EROI by around 10% in each of the studied cases. Conversely, decreasing consumption provided an increase of around 2%, which showed that careful data collection is indeed important for EROI calculations. It also showed that when studying environmental impacts, electricity consumption had a lower influence, as material choices were more impactful. Nevertheless, the scale of changes in GWP is insignificant compared to the absolute differences show in Fig. 4 (from 7.5 to 100 g of CO_2 eq. in the most optimistic cases), thus the comparison remains robust. Similar conclusion can be drawn for EROI, as the effect was similar across the different cases in Fig. 5b.

4.2. Study limitations and future impacts

As solar cell revival research still has a low technical readiness level, this study omits a technoeconomic comparison of these technologies. There are too many uncertainties about processing and the costs of large-scale revival. Therefore, the present research focused on showing the sustainability advantages of higher levels of recycling. These advantages are still evident despite the electricity consumption during manufacturing and recycling being more conservative compared to the most optimistic values (see appendix for literature comparison). Furthermore, the recovery of elements like nickel or gold could be modelled to obtain further insights. However, this would require robust experimental data on the element-level recycling of mesoporous PSCs.

Moreover, encapsulation is left aside, which would play a role in manufacturing and recycling. Reusing parts of the of encapsulated cells was indeed showcased [21], but not for revival. The main expected impact is a lower EROI due to additional manufacturing steps and, thus, increased energy investment and life cycle impacts. The deviation between the work of Bogachuk et al. [21] and present study in the manufacturing impacts of the carbon-based structure is around 3 kg-CO_2 eq. per m^2 (18 kg CO_2-eq compared to 15 kg CO_2-eq respectively). Which shows that the relative impacts of encapsulated devices would

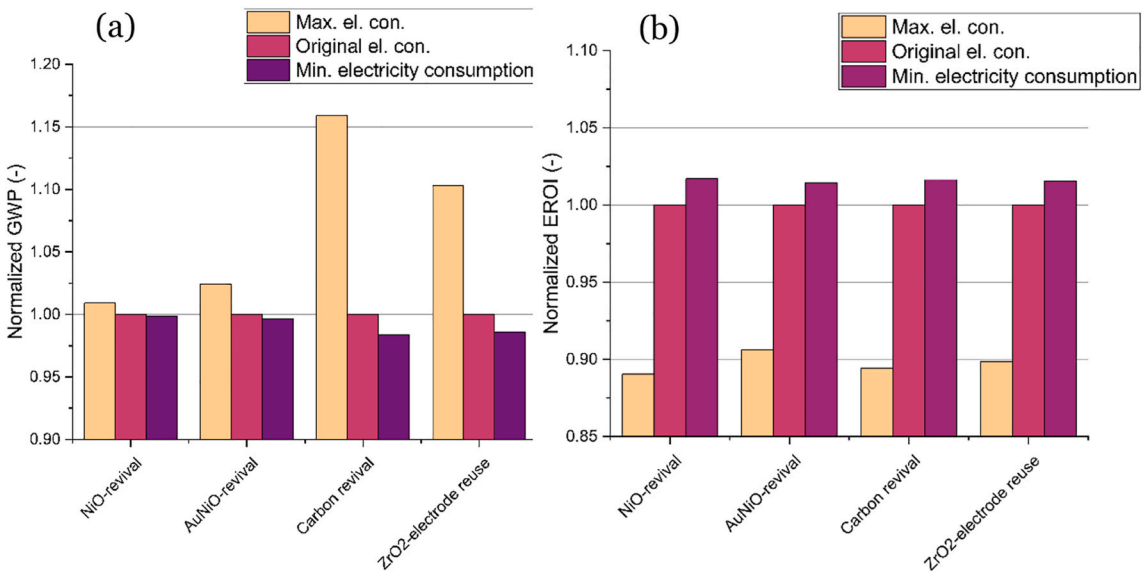

Fig. 5. Sensitivity analysis. a: GWP to changes in FTO sputtering electricity consumption; b: EROI to changes in FTO sputtering electricity consumption. Both are normalized to the original electricity consumption.

increase, yet not dramatically.

A general outlook on the future development of PSC revival raises the question of its suitability for short-lived novel technologies versus long-lived mature technologies. Revival appears similarly realistic for both short- and long-lived devices when judged solely by current results (Figs. 3 and 4; Supplementary Fig. S3–S5). At an early development stage, revival might enable the technology to break into the market. Later, it might facilitate the safe management of toxic materials such as lead [12] and potentially enable cost savings. However, the main obstacle to reviving mature technologies may be technological advances in the technologies themselves—for instance, the increasing efficiency of ever-evolving solar cells [34]. Thus, reviving obsolete technology might create lock-in with, for example, less efficient devices. Therefore, it would be valuable to revisit current research in a few years' time to re-assess whether revival remains valuable for PSCs amid the PSC-research advances.

Building on the present findings, it would be important to focus on showcasing encapsulation that supports revival. Furthermore, optimizing the carbon structures and their revival processes could provide significant benefits in terms of EROI and life cycle impacts. It would be especially valuable to study the degradation and performance of second-life devices in more holistic experimental studies as it remains an unknown parameter. Most importantly, providing experimental evidence of recycled-cell degradation would strengthen the comparison presented here. While the race for high efficiency and stable perovskites is ongoing, there is still room to focus on upscaling the most sustainable approaches.

5. Conclusion

Herein presented research set out to compare the two highest levels of recycling – revival and reuse of components. For this comparison the mesoporous perovskite solar cells were used. Mesoporous C-, NiO- and Au-NiO-PSCs were modelled from manufacturing to recycling and second use utilizing HSC chemistry and OpenLCA complemented by Ecoinvent database. These tools together with previous experimental findings were combined to model four recycling systems. The results were analysed using EROI analysis together with life cycle assessment. Present conclusions were drawn solely for the studied mesoporous PSCs and are not directly transferable to planar PSC.

On the purely electric-energy bases of EROI, it was shown that the highest level of recycling – revival is indeed more favourable. For the carbon-based cells reuse of the cells achieved higher return of energy compared to the remanufacturing – reuse of the ZrO_2 electrode. However, the difference was rather small, thus the second life efficiency loss could make the revival less beneficial. Moreover, for the three studied structure all achieved higher return compared to manufacturing pristine devices. This higher return was linked to comparatively low energy expenditure during revival. Despite EROI being important there was a significant difference in the materials used.

To understand the impacts of the materials used in the mesoporous PSCs a cradle to end-of-second-life LCA was conducted. This revealed that C-PSC had two orders of magnitude lower GWP than NiO-PSC. The NiO-PSC cell had the highest potential impacts in almost all the impact categories and that mainly because of the use of nickel. Even reducing the use of nickel in the Au-NiO-PSC failed to close the gap to C-PSC. This indicated that across different mesoporous cell architectures the material choices dictate the environmental impacts. Focusing on C-PSC, interestingly burning away the Carbon electrode and its redeposition (in ZrO_2-elecrode reuse) did not have significant negative impact compared to the revival of C-PSC. Thus, investigating the material related impacts revealed that lower level of recycling might be less impactful than revival of material intensive PSC structures.

In summary this work highlights the importance of targeting higher levels of recycling as it outperforms virgin manufacturing. Strictly from environmental and energy return point of view the revival could provide most benefits. However, if the efficiency drops significantly after revival, reuse at the current state of knowledge proved to be more advantageous. The lifetime and degradation rate of second life devices will influence which recycling strategy becomes more viable. Thus, building on this work the future studies could set out to improve efficiency and stability after revival and provide techno-economic insights, which limits present research. Moreover, extending this framework to encapsulated cells and other types of perovskite cells would bring additional value.

Most importantly, the PSC are in early stage of commercialization thus there is still an opportunity to choose a recyclable design. Based on present findings targeting low impact-revivable cells – like mesoporous carbon perovskite solar cells would align perovskite research closely with circular economy principles. Present analysis showed that mesoporous PSC are already designed so that they could be revived and reused with significant energy and environmental benefits.

CRediT authorship contribution statement

Šimon Jech: Writing – original draft, Visualization, Methodology, Investigation, Conceptualization. **Mahboubeh Hadadian:** Writing – review & editing, Methodology, Formal analysis. **Kati Miettunen:** Writing – review & editing, Project administration, Methodology, Funding acquisition, Conceptualization. **Annukka Santasalo-Aarnio:** Writing – review & editing, Supervision, Methodology, Funding acquisition, Conceptualization.

Declaration of generative AI and AI-assisted technologies in the writing process

During the preparation of this work the authors used Aalto AI Assistant (GPT 4.o language model) to check for grammatical errors and improve readability. After using this tool/service, the authors reviewed and edited the content as needed and take full responsibility for the content of the published article.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Simon Jech reports financial support was provided by Research Council of Finland. Mahboubeh Hadadian reports financial support was provided by Research Council of Finland. Kati Miettunen reports financial support was provided by Research Council of Finland. Annukka Santasalo-Aarnio reports financial support was provided by Research Council of Finland. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.solmat.2026.114315.

Data availability

The data used for present study is in supplementary materials and in the linked repository (link in supplementary file).

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