Electronic Supplementary Material (ESI) for Sustainable Energy & Fuels.

Supporting information

Scenario-based recycling strategies for Perovskite-Silicon Tandem Solar Cells: a harmonized Life Cycle Assessment study.

Mercy Jelagat Kipyator[1,2], Federico Rossi[1,3], Luigi Vesce[4], Aldo di Carlo[4,5], Riccardo Basosi[1,6,7], Maria Laura Parisi[1,6,7], Adalgisa Sinicropi[1,6,7*]

[1] University of Siena, R^{2}ES Lab, Department of Biotechnology, Chemistry and Pharmacy, Via Aldo Moro, 2, Siena, Italy. E-mail: adalgisa.sinicropi@unisi.it

[2]Scuola Superiore Studi Pavia IUSS(Istituto Universitario di Studi Superiori),Piazza della Vittoria, 15,27100,Pavia,Italy

[3] University of Florence, Department of Industrial Engineering, Via Santa Marta, 3, Florence, Italy

[4] CHOSE - Centre for Hybrid and Organic Solar Energy (CHOSE), Department of Electronic Engineering, University of Rome Tor Vergata, via del Politecnico 1, 00133 Rome, Italy

[5] ISM-CNR, Istituto di Struttura della Materia, Consiglio Nazionale delle Ricerche Via del Fosso del Cavaliere 100, 00133 Rome, Italy

[6] CSGI, Center for Colloid and Surface Science, Via della Lastruccia 3, 50019 Sesto Fiorentino, Italy

[7] Institute of Chemistry of Organometallic Compounds (CNR-ICCOM), Via Madonna del Piano 10, 50019 Sesto Fiorentino, Italy

1. MATERIALS COMPOSITION

As stated in the main text a tandem solar cell consists of two semicells which are combined in a multijunction configuration where each cell absorbs different portions of the solar spectrum. Different materials can make tandem solar cells. In this study, we investigated the environmental performance of a perovskite-silicon tandem solar cell where the perovskite is employed as the top semi-cell and the silicon as the bottom cell. Table S1 shows the most common materials used by the scientific community to make a tandem solar cell, while Table S2 shows the tandem solar cells investigated in this study with details on layers, materials and deposition process.

Table S1. Common materials used in making monolithic tandem solar cells[1,2].

Top cell (Perovskite) Layer	
Transparent conductive oxide (TCO)	Indium tin oxide (ITO), fluorine tin oxide (FTO), indium zinc oxide (IZO), and Aluminium-doped zinc oxide (AZO).
Electron transport layer (ETL)	titanium dioxide (TiO_2), zinc oxide (ZnO), aluminium oxide (Al_2O_3), tin dioxide (SnO_2), PCBM, and fullerene-C60 (C60)
Absorber	The perovskite absorber layer has an ABX_3 structure. A can be made of methylammonium (MA), formamidinium (FA) or caesium (Cs); B is composed of lead (Pb) or tin (Sn); X can be chlorine (Cl), bromine (Br) or iodine (I) Common active layer composition is $MAPbI_3$, $MAPbI_xBr_{3-x}$, $MAFAPbI_3$, $CsMAFAPbI_3$, $RbCsFAPbI_3$, $FAPbI_3$, $RbFAPbI_3$
Hole transport layer (HTL)	Spiro-OMeTAD (2,2′,7,7′ -Tetrakis [N, N-di(4-methoxyphenyl)amino]-9,9′ -spirobifluorene), s PTAA (poly(triaryl amine)), P3HT (Poly(3-hexylthiophene-2,5-diyl)), Copper Thiocyanate (CuSCN) and Nickel Oxide (NiO_x)
Recombination junction	Zinc-tin oxide (ZTO), ITO, IZO, Si layers such as n-nc-SiOx/ITO, n++/p++ semiconductors like a-Si: H, p-nc-Si:H/n-nc-Si: H, n-nc-Si:H/p-nc-Si: H, or amorphous SiO_x
Bottom cell	Silicon solar cells such as Silicon heterojunction silicon (HIT), Passivated emitted rear contact (PERC) with 21-24% efficiency, Copper indium gallium selenide (CIGS)
Back Contact	gold (Au), silver (Ag), aluminium (Al), nickel (Ni), copper (Cu), Al/MoOx, ZnO/Al, and Cr2O3/C

Table S2. Tandem solar cell architectures investigated with details on materials, layers and deposition process.

TSC	**L1**[3]		**L2**[3]		**L3**[3] **(inverted structure)**		**C**[4]		**I**[5]	
ENCAPSULATION	Glass-PET		Glass PET		Glass-PET		Glass-PET		Glass-PET	
Layer	Material	Deposition process	Material	Deposition process	Material	Deposition process	Material	Deposition process	Material	Deposition process
Front grid	Au	Metal evaporation	Ag	Metal evaporation	Al	Metal evaporation	Ag	Screen printing	Ag	Screen printing
Top layer	ITO/MoO_3	Sputtering	ITO/MoO_3	Sputtering	ITO/ZnO	Sputtering	ITO/MoO3	Sputtering	ITO	Sputtering
HTM	SpiroOmetad	Spin coating	SpiroOmetad	Spin coating	PEDOT: PSS	Spin coating	SpiroOmetad	Spin coating	NiO_2	Sputtering
ABSORBER	$MAPbI_3$	Spin coating	$MAPbI_3$	Spin coating	MAPbI3	Spin coating	$MAPbI_3$	Spin coating	$MAPbI_3$	PbI- thermal evaporation MAI-Slot die coating
ETL	TiO_2	Screen printing Sintering	TiO_2	Screen printing Sintering	PCBM	Solution processing	PCBM	Solution processing	SnO2	Sputtering
Tunnel junction	ITO	Sputtering	ITO	Sputtering	ITO	Sputtering	ZnO: In	Sputtering	n, p)-u-c-Si	Plasma-enhanced chemical vapour deposition (PECVD)
Bottom cell	HIT Si	PECVD	HIT Si	PECVD	Si(n)/Si(p)		HIT Si	PECVD	HIT Si	PECVD
Back contact layer	ITO	Sputtering	ITO	Sputtering	ITO	Sputtering	ITO	Sputtering	ITO	Sputtering
Rear contact	Ag	Screen printing	Ag	Screen printing	Al	Screen printing	Ag	Screen printing	Ag	Sputtering

The tables above report the materials making up the tandem solar cells. These materials have been modelled grounding on the ecoinvent database[6]. Accordingly, some of the materials included in the life cycle inventory are modelled by the database considering a recycling input rate. Indeed according to ecoinvent processes, Gold (Gold {GLO}| market for | Cut-off, S) and silver (Silver {GLO}| market for | Cut-off, S) respectively have recycling input rates of 5.5% and 23%). On the other hand, Zinc Oxide (Zinc oxide {RER}| production | Cut-off, U) and Nickel Oxide (Oxygen + Nickel, class 1 {GLO}| market for nickel, class 1 | Cut-off, S) have recycling input rates of 85% and 34% respectively.

2. DEGRADATION RATE AND LIFETIME

The total energy produced by a tandem solar cell over its lifetime is influenced by its performance in the operating environment and the degradation rate[7]. Degradation can occur both linearly and non-linearly, depending on the underlying mechanism causing the degradation. Linear degradation occurs when the degradation rate is constant over time and the performance of the PV module gradually decreases. Non-linear degradation occurs when the rate of degradation changes over time, resulting in faster or slower performance degradation[8]. There is particularly uncertainty on the rate of degradation of the Perovskite cell whose stability is still a challenge. Basing our estimations on the values provided by Qian et al. [7]we investigate the effect of different degradation rates on the environmental performance of the tandem solar cell, i.e. if they were to maintain 80% of their initial power conversion efficiency after 5, 10, 15, 20 and 25 years of operation. Table S3 shows the estimations of the degradation rates for the perovskite solar cell based on the assumption that the higher the degradation rate the lower the lifetime of the perovskite solar cell.

Table S3. Different degradation rates of the perovskite solar cell

Monolithic	Mechanically stacked
0.9%- 25 years	*1.3%- 25 years*
1.3%-20 years	*1.63%-20 years*
1.7%-15 years	*2.2%-15 years*
2.3%-10 years	*3.3%-10 years*
4.5%-5 years	*6.5%-5 years*

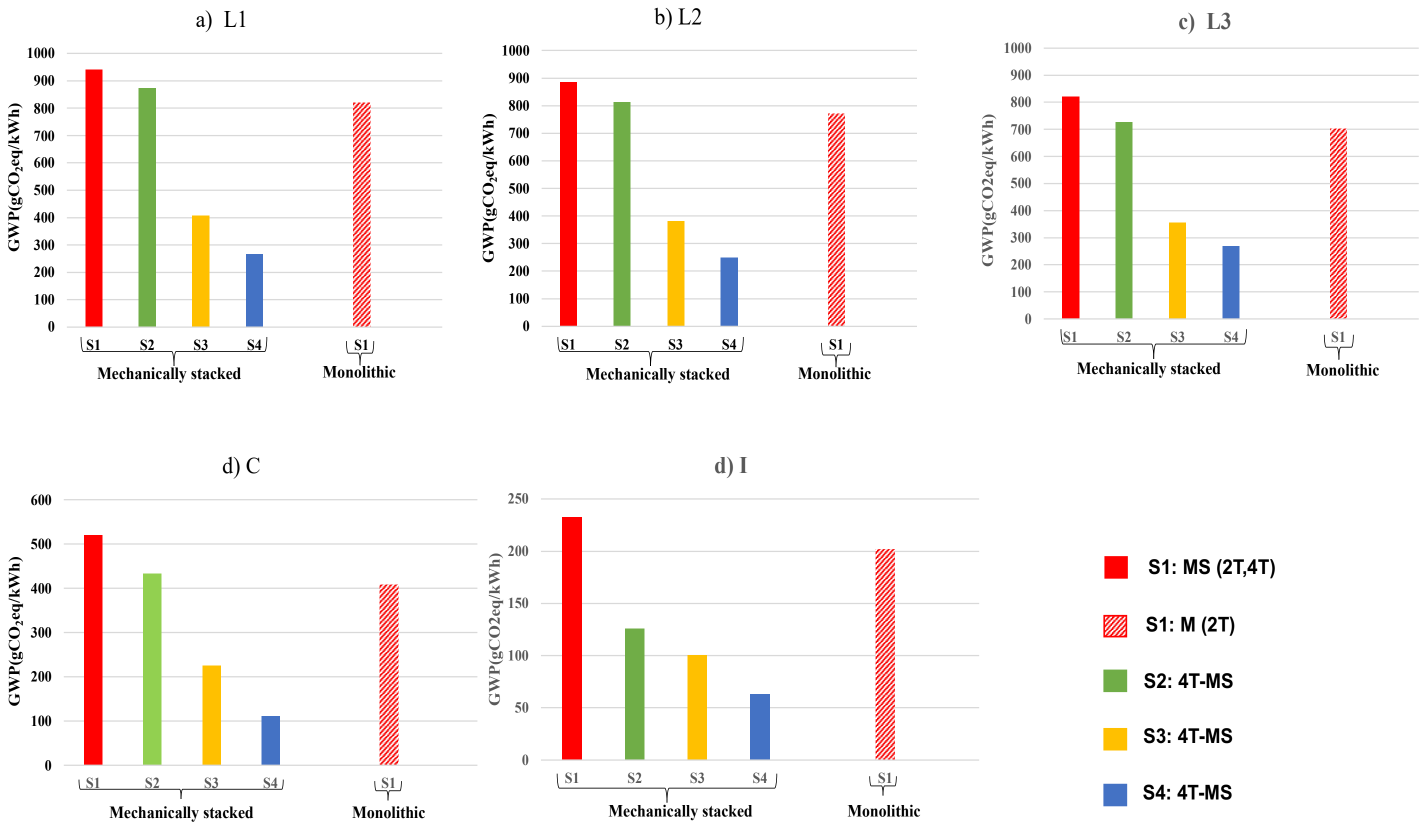

Figure S1: Comparison of global warming potential (GWP) of the recycling scenarios for the five PSK/c-Si tandem cells.MS represents mechanically stacked configurations while M represents monolithic configuration.

Similar to the results obtained with constant values of degradation rate (0.9% for PSK in 2T-M and 1.3% in 2T,4T-MS), the environmental impact shown in Figure S1 can be ranked in the following order (sorted from most to least impactful): S1 > S2 >S3 > S4. However, the absolute values of the GWP in this case are higher because there are higher efficiency losses. The most impactful strategy is S1 in both MS (2T,4T) and 2T-M cells: because of the short lifespan of the PSK semi-cell, in S1 the TSC shall be replaced 5 times, including the substitution of c-Si semi-cell that would still be highly productive. S2, which involves the periodic replacement of the PSK cell, shows better environmental performance for tandem cells than S1; however, in a situation where the PSK cells reach their end of life and do not affect the functionality of silicon solar cell, retaining the PSK cell in the tandem system (S3) seems to be beneficial for the eco-profile of TSC cells. Nevertheless, taking into account that S3 could involve a drop in the efficiency of the tandem cell, this result shows that S4 could be the most sustainable recycling scenario for tandem cells. A sensitivity analysis was also done taking into account the different degradation rates (table S3). As shown in Figure S2, regardless of different degradation rates, a lifespan extension results in a reduction of GWP since the increase in lifespan contributes to enhanced energy performance. GWP mitigation is particularly strong for those strategies that imply the replacement of the TSC (S1) or of the PSK semi-cell (S2) because, with an increase in lifespan, the GWP for manufacturing the semi-cells reduces.

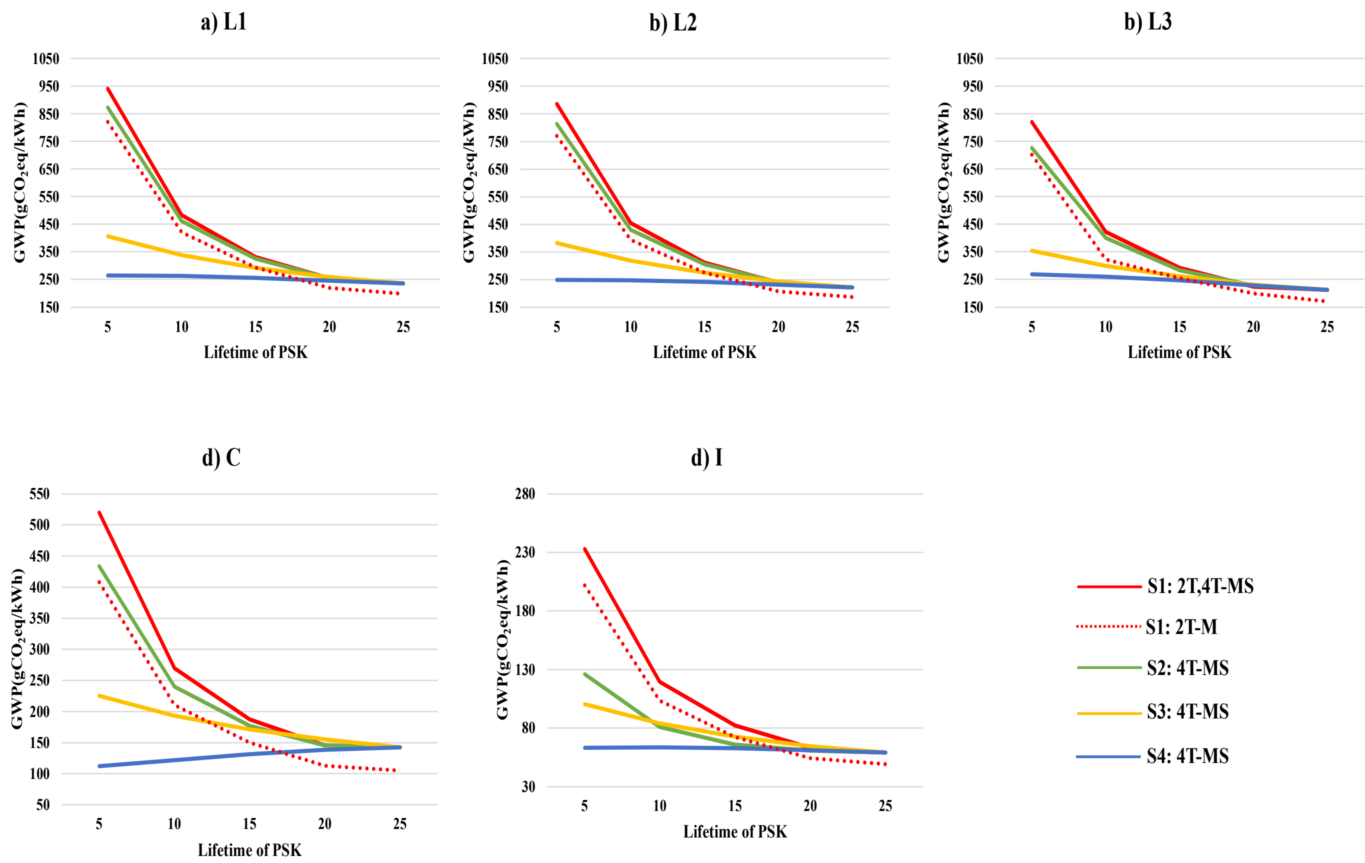

Figure S2. Sensitivity analysis of the effect of lifespan on GWP of the five PSK/c-Si cells with different PSK layer lifespan (5, 10, 15, 20, 25 years). The full lines correspond to the recycling scenarios of mechanically stacked tandems (2T and 4T) while the dotted lines correspond to monolithic tandem cells (2T).

REFERENCES

1. H. Li and W. Zhang, *Chemical Reviews*, 2020, **120**, 9835–9950.

2. E. Leccisi and V. Fthenakis, *Progress in Energy*, 2020, **2**, 032002–032002.

3. M. Monteiro Lunardi, A. Wing Yi Ho-Baillie, J. P. Alvarez-Gaitan, S. Moore and R. Corkish, *Prog. Photovolt: Res. Appl.*, 2017, **25**, 679–695.

4. I. Celik, A. B. Phillips, Z. Song, Y. Yan, R. J. Ellingson, M. J. Heben and D. Apul, *Energy & Environmental Science*, 2017, **10**, 1874–1884.

5. R. Itten and M. Stucki, *Energies*, 2017, **10**, 841.

6. G. Wernet, C. Bauer, B. Steubing, J. Reinhard, E. Moreno-Ruiz and B. Weidema, *Int J Life Cycle Assess*, 2016, **21**, 1218–1230.

7. J. Qian, M. Ernst, N. Wu and A. Blakers, *Sustainable Energy and Fuels*, 2019, **3**, 1439–1447.

8. T. Rahman, A. Mansur, M. Hossain Lipu, Md. Rahman, R. Ashique, M. Houran, R. Elavarasan and E. Hossain, *Energies*, 2023, **16**, 3706.