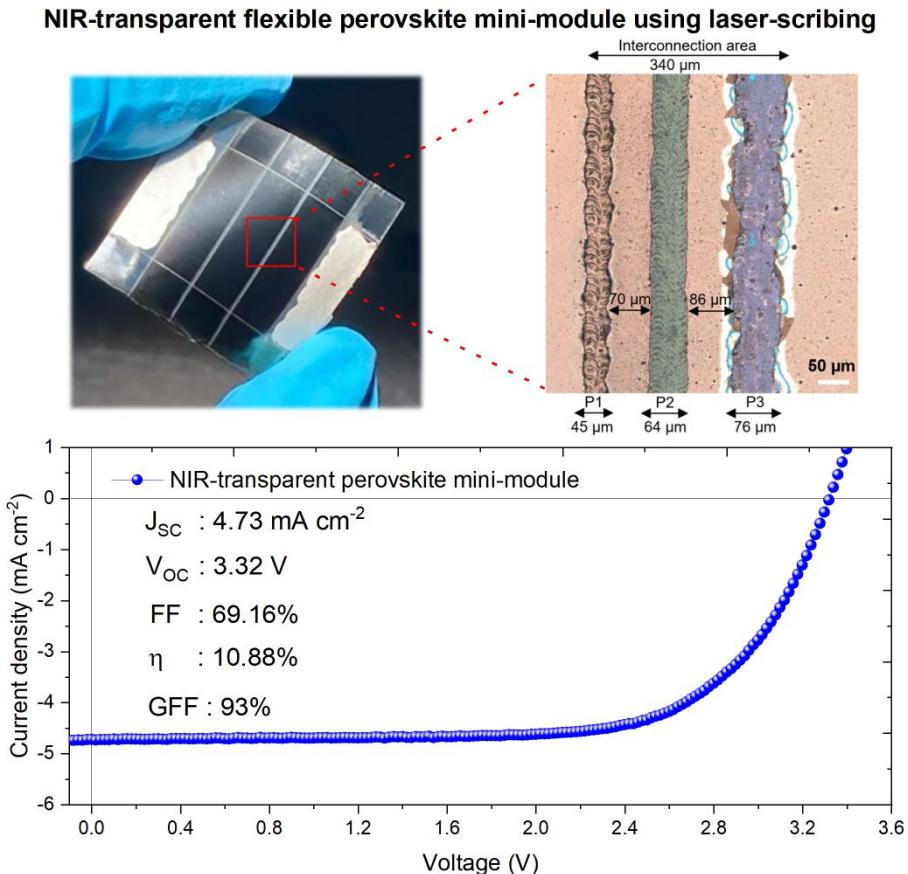


Laser patterned flexible perovskite-CIGS tandem mini-module with over 18% efficiency

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Flexible NIR-transparent perovskite mini-module with an efficiency of 10.88% and a geometric fill factor (GFF) of over 93% is developed using an all laser-based patterning approach. In combination with a CIGS flexible mini-module, a proof of concept flexible 4T tandem mini-module with 18.4% efficiency is demonstrated.

Supporting Information

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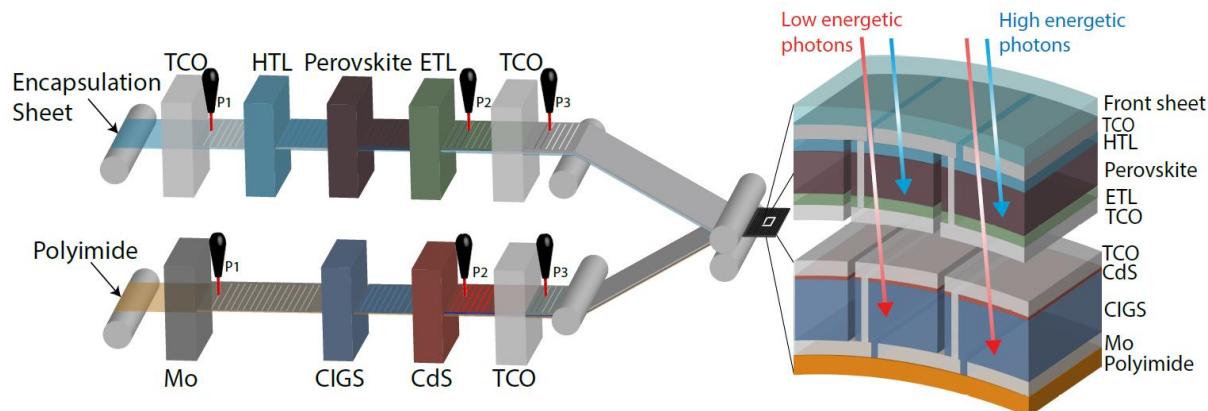


Figure S1. Schematic representation of vision of future manufacturing production unit of flexible perovskite-CIGS tandem module.

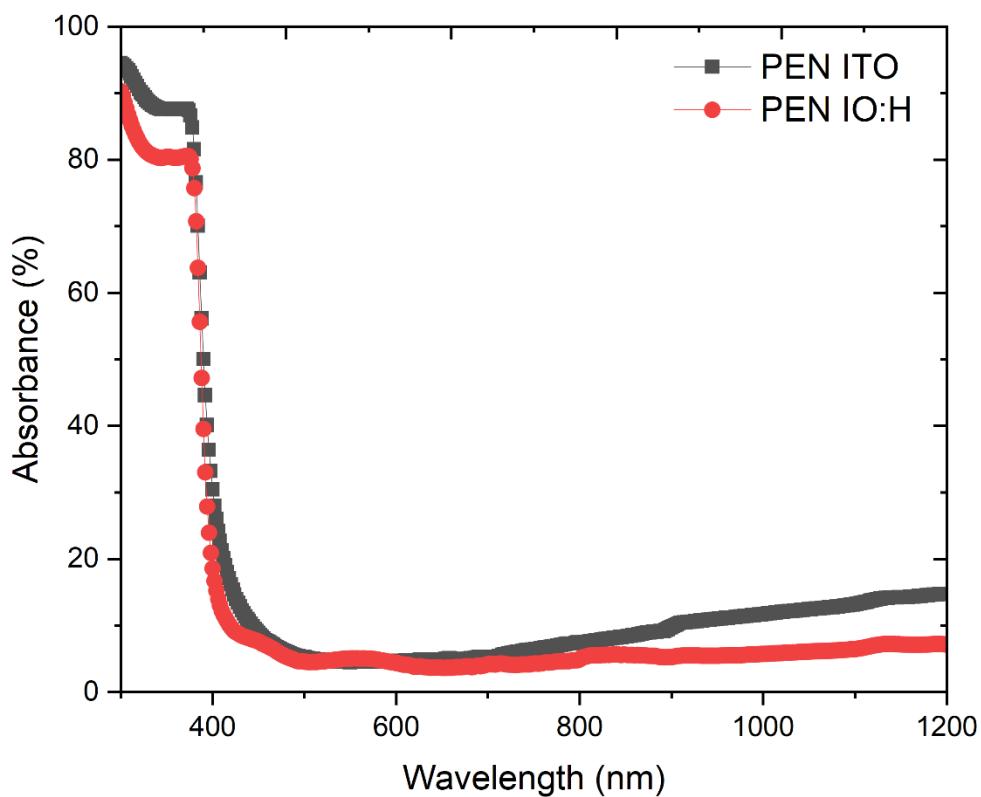


Figure S2. Absorbance of commercial ITO and IO:H coated flexible PEN substrate.

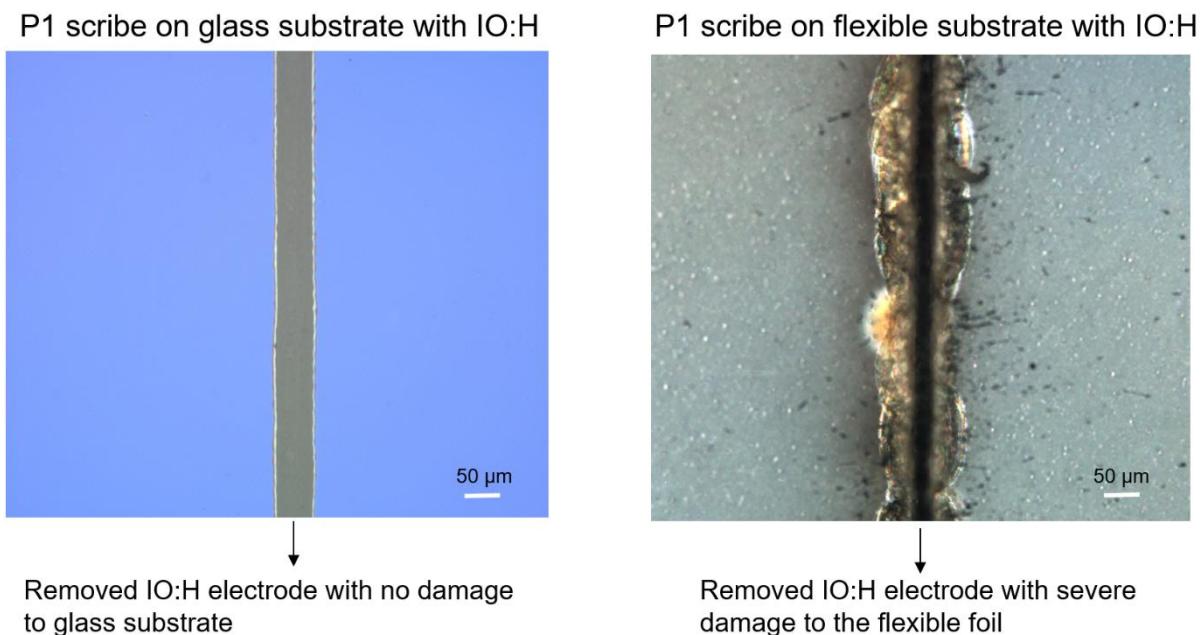


Figure S3. P1 scribe comparison of (a) Rigid, and (b) Flexible substrate with IO:H electrode. A laser fluence of 2.4 J/cm^2 with 12 repetitions is used to carry out P1 scribe. The laser pulse induce damages to the flexible foil making them unsuitable for further processing.

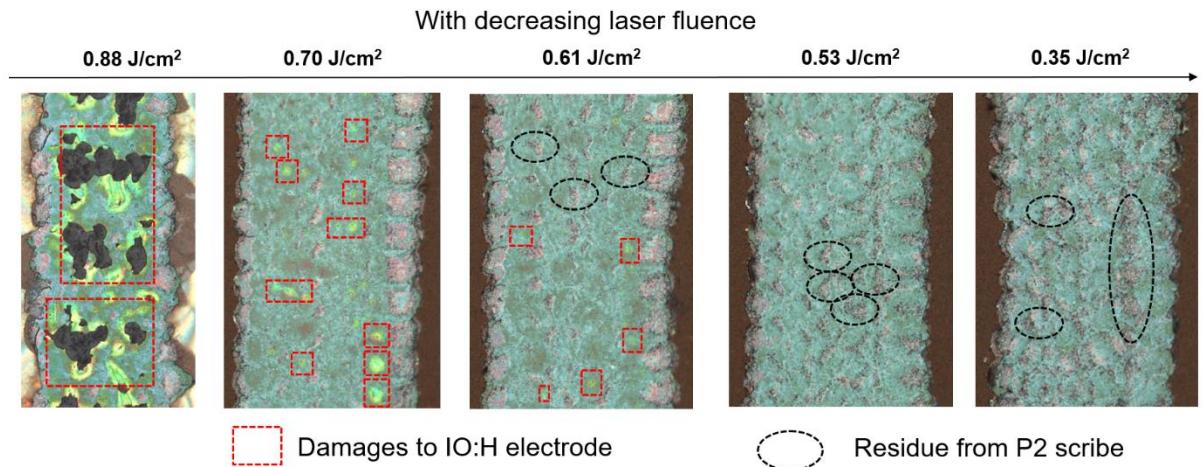


Figure S4. Confocal image of sample with different laser fluence for P2 scribing optimization. The elliptical areas indicate the residue after P2 scribing and the boxes represent ablation of bottom IO:H electrode.

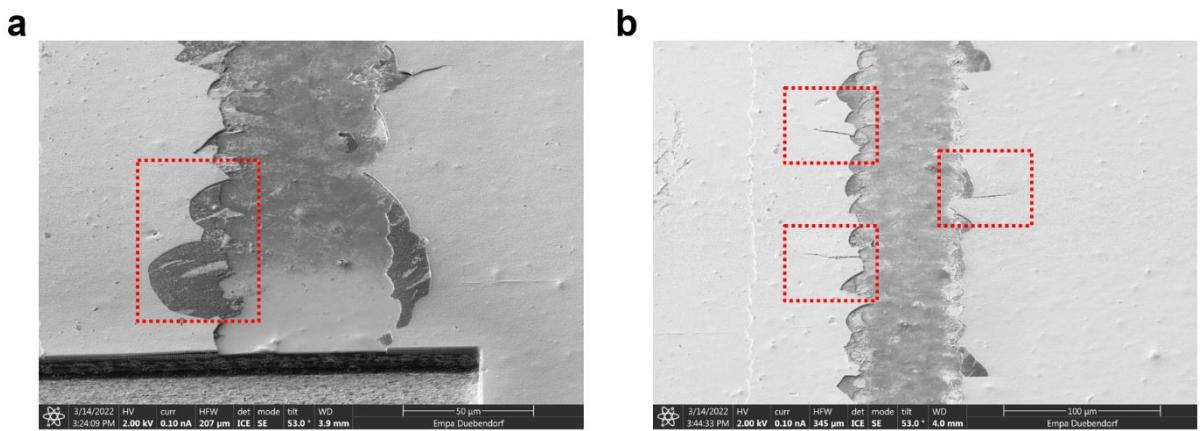


Figure S5. FIB SEM image showing the (a) delamination of the IZO electrode, and (b) extend of rupture cross section after P3 scribing.

Table S1. Summary of photovoltaic parameters of best performing NIR-transparent perovskite solar cells and mini-modules without any ARC coating.

Flexible solar cell/mini-module	V_{OC} (V)	J_{SC} (mA cm^{-2})	FF (%)	PCE (%)	Area (cm^2)
NIR-transparent perovskite solar cell	1.05	14.53	71.94	10.98	0.09
NIR-transparent perovskite mini-module	3.20	4.71	69.84	10.56	2.20

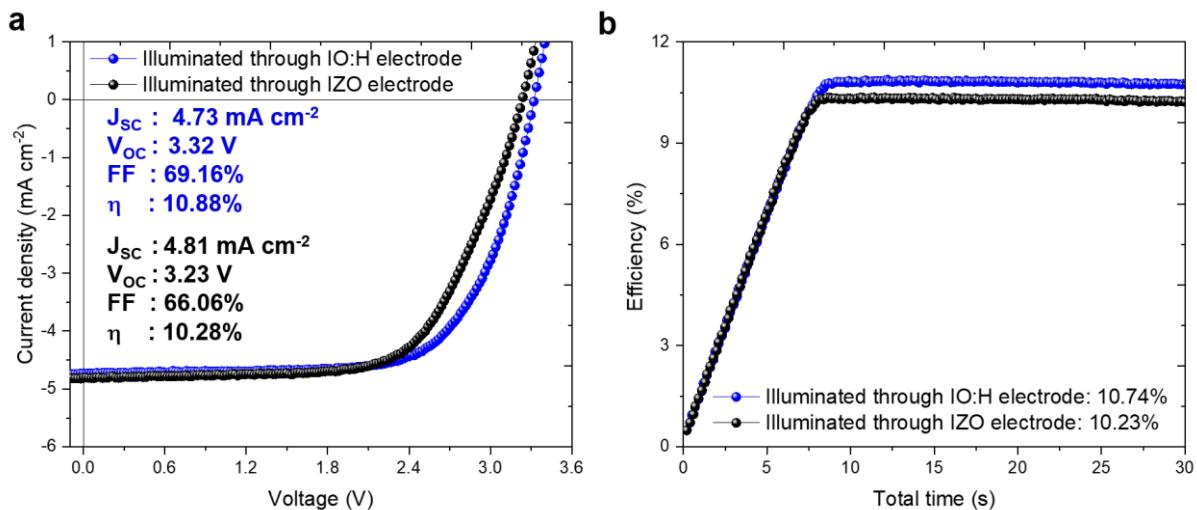


Figure S6. (a) JV and (b) MPP efficiency characteristics comparison of NIR-transparent perovskite solar mini-module illuminated through IO:H (blue) and IZO (black) electrode.

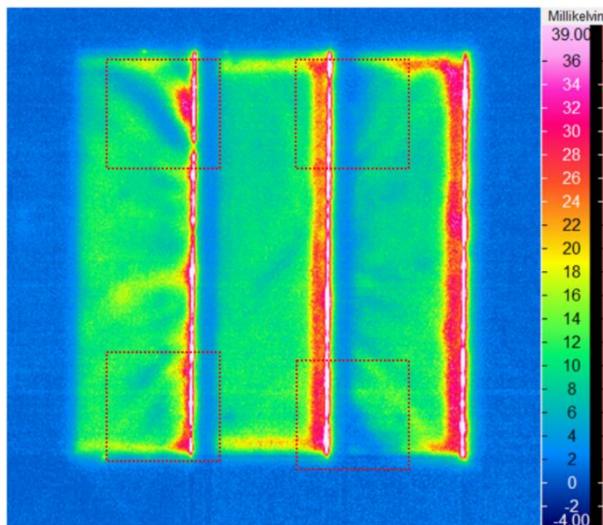


Figure S7. Dark lock-in thermography image of perovskite mini-module under high injection forward bias condition. As a guide to the eye, the thermal scale is attached to the image.

Figure S7 shows the dark-lock in thermography image of a mini-module probed at a voltage exceeding their V_{oc} under forward bias condition under a specific lock-in frequency. Under this condition, the diode current flows through the semiconductor with certain series resistance and results in heat dissipation (Joule's heating effect). The emitted thermal radiation from the surface is then captured by a thermal infrared camera to provide the thermal emissive profile of the sample. The lock-in thermography confirms the presence of 3 subcells that are monolithically interconnected. The red box indicates the region near the scribe lines were the a non-uniform coating in the form of streaky lines are visible. The scribing lines act as disruption to flow of the solvent in the spin-coating process. These streaky patterns are more profound at the

exterior region of the sample where the outward centrifugal force is maximum.

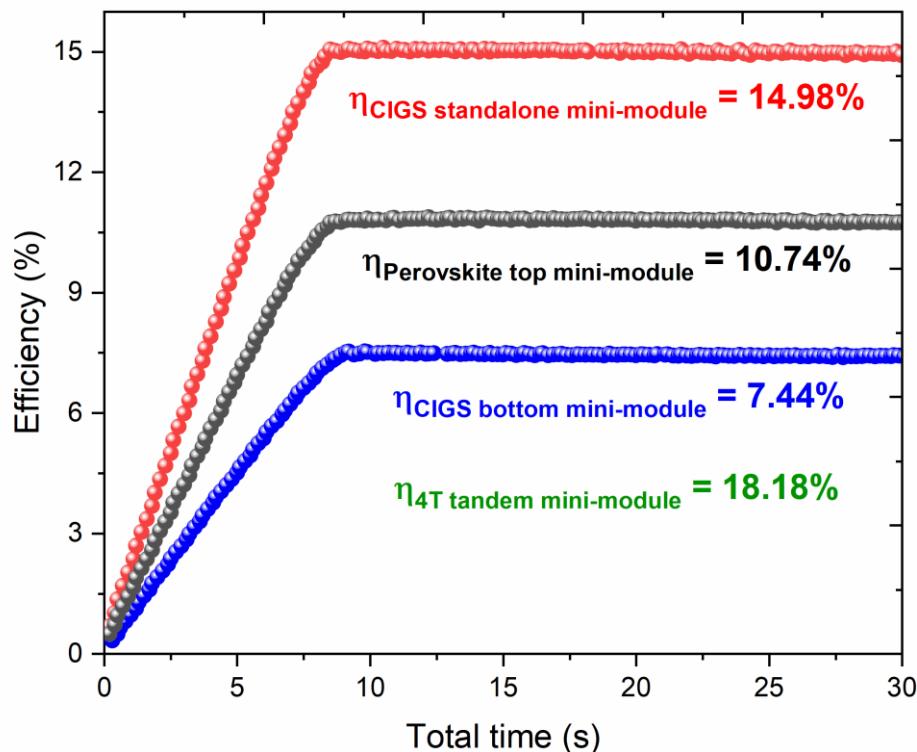


Figure S8. MPP efficiency characteristics of stand-alone CIGS mini-module, and perovskite and CIGS mini-module in 4T tandem configuration.

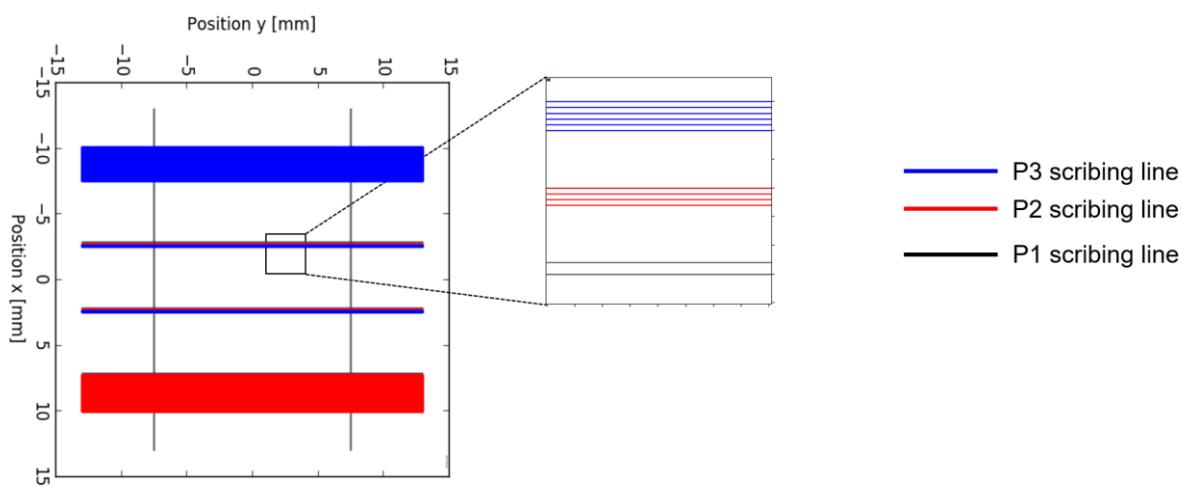


Figure S9. Laser scribing layouts for NIR-transparent perovskite mini-module on flexible substrate. A magnified view of the interconnection region is provided to view the P1 (black), P2 (red) and P3 (blue) interconnection coordinates. A fiducial marker based alignment system (represented with a black crosshair) is used as the reference point to carry out P1, P2 and P3 scribing with improved precision.

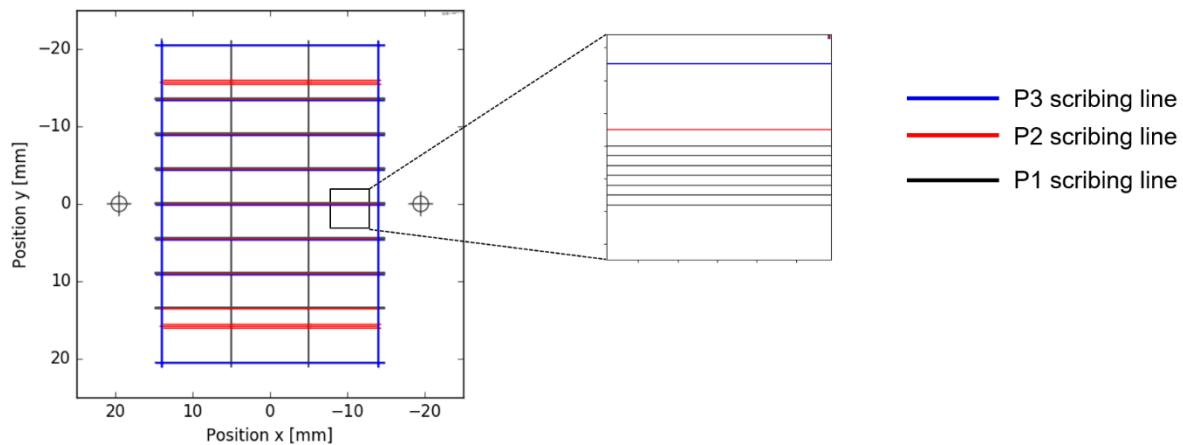


Figure S10. Laser scribing layouts for CIGS mini-module on flexible substrate. A magnified view of the interconnection region is provided to view the P1 (black), P2 (red) and P3 (blue) interconnection coordinates. Similar to perovskite mini-module, a fiducial marker system (with a black cross hair) is used as the reference point to carry out P1, P2 and P3 scribing.

Table S2. Summary of perovskite-CIGS 4T Tandem solar cells/mini-modules published in literature.

Institute	J _{SC} a) (mA cm ⁻²)	V _{OC} (V)	FF (%)	PCE (%)	Tandem PCE (%)	Area (cm ²)	Year	Reference
Stanford U.	17.5/10.9	1.02/0.68	71.0/78.8	12.7/5.9	18.6	0.39/0.39	2014	[1]
Empa/EPFL	16.7/14.4	1.03/0.66	70.3/77.4	12.1/7.4	19.5	0.28/0.21	2015	[2]
Empa	17.4/12.7	1.10/0.66	73.6/74.9	14.1/6.3	20.5	0.52/0.21	2015	[3]
Empa	19.1/15.3	1.12/0.43	75.4/73	16.1/4.8	20.9	0.28/0.21	2016	[4]
Empa	19.1/12.1	1.12/0.67	75.4/73.6	16.1/6.0	22.1	0.28/0.21	2016	[4]
U. Washington	15.8/17.0	1.07/0.48	74/62.0	12.5/5.1	18.7	NA/0.11	2017	[5]
U. Washington	15.8/12.6	1.07/0.61	74/68.2	12.5/5.2	18.8	NA/0.11	2017	[5]
SinBeRISE/NTU	20.1/15.2	0.98/0.47	78.1/64.6	15.3/4.0	20.7	0.09/0.16	2017	[6]
Empa #	16.1/12.6	1.08/0.63	68.5/77.3	11.9/6.1	18.2	0.15/0.21	2017	[7]
Imec/KIT	9.7/11.2	6.31/2.58	72/76	11.7/5.8	17.8	3.76/3.76	2017	[8]
ANU	21.8/13	1.14/0.62	74/72	18.4/5.8	23.9	0.3/0.5	2018	[9]
Empa #	18.7/12.0	1.06/0.65	68.9/71.6	13.7/5.6	19.6	0.15/0.21	2018	[10]
Imec	11.1/10.6	7.10/2.68	73/75.6	14.4/5.7	20.1	4/3.8	2018	[11]
NREL	19.6/15.6	1.14/0.72	76.8/79.2	17.1/8.8	25.9	0.06/0.42	2019	[12]
Empa	19.2/18.6	1.12/0.58	75.2/74.2	16.1/8.0	24.1	0.57	2019	[13]
Empa/Solliance	20.8/19.4	1.03/0.56	79.8/74.2	17.2/8.1	25.0	0.57	2019	[13]
ZSW/KIT				15.3/8.0	23.3	0.24/0.50	2019	[14]
ZSW/KIT				15.0/8.0	23	0.50/0.50	2019	[14]
KIT	19.7/13.6	1.16/0.71	78.7/78.1	18.0/7.5	25.0	0.22/0.50	2020	[15]
Empa	22.9/17.5	1.09/0.56	70.6/73.7	17.62/7.3	24.6	0.3/0.51	2020	[16]
Soochow University #	16.3/12.8	1.18/0.63	77.5/75.5	15.02/6.0	21.06	0.07	2020	[17]
Empa/EPFL	24.5/14.3	1.12/0.55	78.2/72.4	21.4/5.7	27.1	0.09/0.57	2022	[18]
This work #	4.73/3.18	3.32/3.53	69.2/66.9	10.9/7.52	18.4	2.20/2.03	2022	This work

a) The PV parameters of the top and bottom cells are separated by a slash.

represents work carried out on flexible substrate.

References

1. Bailie, C.D., Christoforo, M.G., Mailoa, J.P., Bowring, A.R., Unger, E.L., Nguyen, W.H., Burschka, J., Pellet, N., Lee, J.Z., Grätzel, M., Noufi, R., Buonassisi, T., Salleo, A., and McGehee, M.D. *Energy Environ. Sci.*, 2015, 8, 956–963.

2. Kranz, L., Abate, A., Feurer, T., Fu, F., Avancini, E., Löckinger, J., Reinhard, P., Zakeeruddin, S.M., Grätzel, M., Buecheler, S., and Tiwari, A.N. *J. Phys. Chem. Lett.*, 2015, 6, 2676–2681.
3. Fu, F., Feurer, T., Jäger, T., Avancini, E., Bissig, B., Yoon, S., Buecheler, S., and Tiwari, A.N. *Nat. Commun.*, 2015, 6, 1–9.
4. Fu, F., Weiss, T.P., Fu, F., Feurer, T., Weiss, T.P., Pisoni, S., Avancini, E., Andres, C., Buecheler, S., and Tiwari, A.N. *Nat. Energy*, 2016, 1.
5. Uhl, A.R., Yang, Z., Jen, A.K.Y., and Hillhouse, H.W. *J. Mater. Chem. A*, 2017, 5, 3214–3220.
6. Guchhait, A., Dewi, H.A., Leow, S.W., Wang, H., Han, G., Suhaimi, F. Bin, Mhaisalkar, S., Wong, L.H., and Mathews, N. *ACS Energy Lett.*, 2017, 2, 807–812.
7. Pisoni, S., Fu, F., Feurer, T., Makha, M., Bissig, B., Nishiwaki, S., Tiwari, A.N., and Buecheler, S. *J. Mater. Chem. A*, 2017, 5, 13639–13647.
8. Paetzold, U.W., Jaysankar, M., Gehlhaar, R., Ahlsweide, E., Paetel, S., Qiu, W., Bastos, J., Rakocevic, L., Richards, B.S., Aernouts, T., Powalla, M., and Poortmans, J. *J. Mater. Chem. A*, 2017, 5, 9897–9906.
9. Shen, H., Duong, T., Peng, J., Jacobs, D., Wu, N., Gong, J., Wu, Y., Karuturi, S.K., Fu, X., Weber, K., Xiao, X., White, T.P., and Catchpole, K. *Energy Environ. Sci.*, 2018, 11, 394–406.
10. Pisoni, S., Carron, R., Moser, T., Feurer, T., Fu, F., Nishiwaki, S., Tiwari, A.N., and Buecheler, S. *NPG Asia Mater.*, 2018, 10, 1076–1085.
11. Jaysankar, M., Paetel, S., Debucquoy, M., Ahlsweide, E., Gehlhaar, R., and Poortmans, J. *2018 IEEE 7th World Conf. Photovolt. Energy Conversion, WCPEC 2018 - A Jt. Conf. 45th IEEE PVSC, 28th PVSEC 34th EU PVSEC*, 2018, 3584–3587.
12. Kim, D.H., Muzzillo, C.P., Tong, J., Palmstrom, A.F., Larson, B.W., Choi, C., Harvey, S.P., Glynn, S., Whitaker, J.B., Zhang, F., Li, Z., Lu, H., van Hest, M.F.A.M., Berry,

- J.J., Mansfield, L.M., Huang, Y., Yan, Y., and Zhu, K. *Joule*, 2019, 3, 1734–1745.
13. Feurer, T., Carron, R., Torres Sevilla, G., Fu, F., Pisoni, S., Romanyuk, Y.E., Buecheler, S., and Tiwari, A.N. *Adv. Energy Mater.*, 2019, 9, 2–7.
14. Schultes, M., Helder, T., Ahlswede, E., Aygüler, M.F., Jackson, P., Paetel, S., Schwenzer, J.A., Hossain, I.M., Paetzold, U.W., and Powalla, M. *ACS Appl. Energy Mater.*, 2019, 2, 7823–7831.
15. Gharibzadeh, S., Hossain, I.M., Fassl, P., Nejand, B.A., Abzieher, T., Schultes, M., Ahlswede, E., Jackson, P., Powalla, M., Schäfer, S., Rienäcker, M., Wietler, T., Peibst, R., Lemmer, U., Richards, B.S., and Paetzold, U.W. *Adv. Funct. Mater.*, 2020, 30.
16. Jiang, Y., Feurer, T., Carron, R., Sevilla, G.T., Moser, T., Pisoni, S., Erni, R., Rossell, M.D., Ochoa, M., Hertwig, R., Tiwari, A.N., and Fu, F. *ACS Nano*, 2020, 14, 7502–7512.
17. Li, S., Wang, C., Zhao, D., An, Y., Zhao, Y., Zhao, X., and Li, X. *Nano Energy*, 2020, 78, 105378.
18. Zhang, C., Chen, M., Fu, F., Zhu, H., Feurer, T., Tian, W., Zhu, C., Zhou, K., Jin, S., Zakeeruddin, S.M., Tiwari, A.N., Padture, N.P., Grätzel, M., and Shi, Y. *Energy Environ. Sci.*, 2022.