Simulations of non-monolithic tandem solar cell configurations for electrolytic fuel generation

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Supporting Information

S1. Spectral Data



Fig. S1. Spectral data vs. time of day, showing the (a) power density and (b) photon flux vs. wavelength, and the corresponding total spectral power density vs. time of day.

S2. MAPbI₃/Si for Water-splitting.



Fig. S2. Daily performance variation for the four-terminal configuration $MAPbI_3(x2)/Si(x3)$ tandem PV. Current density vs. voltage for (a) the top layer of $MAPbI_3(x2)$ and (b) the bottom layer of top-cell filtered Si (x3). (c) Hourly variation in the solar-to-hydrogen efficiency of the two-terminal (2T) tandem (MAPbI_3/Si), the combined four-terminal (4T) tandem (MAPbI_3(x2)/Si(x3)), the four-terminal top layer only (MAPbI_3(x2)), and the four-terminal bottom layer only (Top-cell Filtered Si (x3)). Efficiencies are for each device driving a PEM water electrolyzer (dashed line with circles in a and b).

S3. 1.74 eV Perovskite/Si for Water-splitting.



Fig. S3. Daily performance variation for the configurations of 1.74 eV/Si tandem PV. Current density vs. voltage for (a) the two-terminal 1.74 eV/Si with one electrolyzer and (b) the top layer of the four-terminal configuration using 1.74 eV (x3) with two electrolyzers in series. (c) Hourly variation in the solar-to-hydrogen efficiency of the top layer 1.74 eV(x3) driving two electrolyzers, the bottom layer of top-cell filtered Si(x3) driving one electrolyzer, and the combined four-terminal tandem performance (4T 1.74 eV(x3)[x2 Electrolyzer]/Si(x3)).

S4. Effects of Width and Series Resistance.



Fig. S4. Effect of series resistance on the daily performance variation for the four-terminal configuration top layer of 1.74 eV (x3) driving two electrolyzers in series, with a unit PV width, w, of (a) 1 cm, (b) 2 cm, and (c) 3 cm.



S5. Non-monolithic Tandem Configurations for CO₂ Reduction

Fig. S5. Example schematics of non-monolithic tandem configurations for Cu-catalyzed CO₂ reduction. (a) Twoterminal (1.74 eV/Si) (x2). (b) Four-terminal 1.74 eV (x3) with Si (x6). To target specific operating conditions of maximum faradaic efficiency for a desired product, MEA areas were varied independently for each PV section. Not to scale.

S6. Non-monolithic Tandem Configurations for CO₂ Reduction Targeting Formic Acid Production.



Fig. S6. Daily performance variation for configurations of 1.74 eV/Si tandem PV for Cu-catalyzed CO₂ reduction targeting formic acid production. Current density per MEA area (J_{MEA}) vs. voltage for (a) the two-terminal (1.74 eV/Si) (x2) ($A_{PV} = 0.25$) and (b) the top layer of the four-terminal configuration using 1.74 eV (x3) ($A_{PV} = 0.38$), and (c) the bottom layer of the four-terminal configuration using top-cell filtered Si (x6) ($A_{PV} = 0.75$).

S7. Non-monolithic Tandem Configurations for CO₂ Reduction Targeting Ethylene Production.



Fig. S7. Daily performance variation for configurations of 1.74 eV/Si tandem PV for Cu-catalyzed CO₂ reduction targeting ethylene production. Current density per MEA area (J_{MEA}) vs. voltage for (a) the two-terminal (1.74 eV/Si) (x3) ($A_{PV} = 0.67$) and (b) the top layer of the four-terminal configuration using 1.74 eV (x4) ($A_{PV} = 0.88$), and (c) the bottom layer of the four-terminal configuration using top-cell filtered Si (x7) ($A_{PV} = 1.61$).



S8. Charge Distribution to Products for Non-monolithic Tandem Configurations for CO₂ Reduction.

Fig. S8. Cumulative daily moles of electrons per PV area directed to the reaction products resulting from configurations of 1.74 eV/Si tandem PV driving Cu-catalyzed CO₂ reduction. (a) Two-terminal (2T) (1.74 eV/Si) (x2) tandem and four-terminal (4T) 1.74 eV (x3)/Si (x6) tandem targeting formic acid production. (b) Two-terminal (2T) (1.74 eV/Si) (x1.74 eV/Si) (x3) tandem and four-terminal (4T) 1.74 eV (x4)/Si (x7) tandem targeting ethylene production.