Electronic supplementary information (ESI) for

Achieving Over 4% Efficiency for SnS/CdS Thin-Film Solar Cells by Improving Heterojunction Interface Quality

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Fig. S1. (a) Transmittance spectra, (b) bandgap extraction, and (c) electrical properties of the AZO films grown at different temperatures from room temperature to 400 °C.



Fig. S2. *J*–*V* characteristics of the SnS/CdS TFSCs at different AZO film deposition temperatures of (a) 150 °C, (b) 200 °C, (c) 250 °C, (d) 300 °C, and (e) 350 °C.



Fig. S3. Cross-sectional SEM images of the SnS/CdS TFSCs with AZO films deposited at (a) 150 °C and (b) 300 °C.



Fig. S4. Full SIMS depth profiles of the TFSCs with AZO deposited at (a) 250 and (b) 350 °C without Al top contact. (c) The selected profiles for Sn, S, and Cd ions. (d) The shifted profiles for matching the initial detection of S, Sn, and Cd signals to evaluate the Cd diffusion into SnS absorbers. Slight Cd diffusion into SnS absorber is noted for the 350 °C device.



Fig. S5. (a) J-V characteristics and (b) the efficiency retention of SnS/CdS TFSCs at various AZO film deposition temperatures of 150–350 °C. To test the long-term stability of the TFSCs, the device performance was measured again after almost two years.



Fig. S6. (a) Temperature dependence of V_{oc} for TFSCs with various AZO deposition temperatures at 150 to 350 °C. (b) The extracted activation energy as a function of AZO deposition temperature.



Fig. S7. Plan-view SEM images of the SnS absorbers annealed in (a) vacuum condition (~10⁻² Torr), and (b) 20 Torr in Ar ambient, respectively. Partial evaporation of SnS surface is observed for (a) vacuum condition.



Fig. S8. Transmittance spectrum of the AZO layer grown in a home-made sputter at room temperature; the inset shows the bandgap extraction plot ($E_g = 3.58 \text{ eV}$). The average transmittance for 400 – 700 nm was 89.0%. The film thickness was ~380 nm and the sheet resistance was 28 ohm/sq. with resistivity of 1.066×10^{-3} ohm cm.



Fig. S9. *J*–*V* characteristics of the TFSCs with (a) as-deposited SnS/CdS without any junction annealing, (b) annealed SnS at 300 °C in Ar ambient and as-deposited CdS without any junction annealing, and (c) as-deposited SnS/CdS with additional junction annealing at 300 °C in Ar ambient. Here, the annealing duration and pressure was fixed at 1 h and 20 Torr, respectively, and in total, six cells were fabricated. The highest efficiencies achieved for these devices were 2.04%, 1.61%, and 4.20%, respectively.



Fig. S10. (a) *J*–*V* and (b) EQE certifications of the champion cell ($\eta = 4.225\%$) measured at KIER in Korea.



Fig. S11. (a) Reflectance spectrum of the full device without a top metal grid. Transmittance, reflectance, and absorbance spectra for (b) glass/i-ZnO/AZO, (c) glass/FTO, and (d) glass/FTO/CdS. Figure (b) was used for calculating absorption in the transparent electrodes (i-ZnO/AZO). Figure (c) and (d) were used for calculating absorption in CdS buffer layer.

AZO film deposition Temperature (°C)	RT	100	200	300	400
Thickness (nm)	494	491	406	435	405
$R_{ m sheet}$ ($\Omega/ m sq.$)	55.1	16.0	7.8	11.7	21.9
Resistivity (Ω cm)	2.72×10^{-3}	7.86 × 10 ⁻⁴	3.17 × 10 ⁻⁴	5.09 × 10 ⁻⁴	8.87 × 10 ⁻⁴
Carrier concentration (cm ⁻³)	2.2×10^{20}	$5.4 imes 10^{20}$	$9.6 imes 10^{20}$	$6.2 imes 10^{20}$	6.4×10^{20}
Mobility (cm ² V ⁻¹ s ⁻¹)	9.8	14.5	20.6	19.5	10.9

Table S1. Electrical properties of AZO thin films grown at different temperatures.

AZO film deposition temperature (°C)	$G_{ m sh}$ (mS cm ⁻²)	$R_{ m s}$ (Ω cm ²)	A	J ₀ (mA cm ⁻²)
150	2.430	0.491	1.228	2.23×10^{-2}
200	2.795	0.534	1.215	1.27×10^{-2}
250	2.214	0.441	1.424	1.66 × 10 ⁻³
300	1.073	0.707	1.244	3.63×10^{-3}
350	0.289	0.710	1.241	2.88×10^{-3}

Table S2. Simulated diode parameters for SnS/CdS TFSCs at various AZO film deposition temperatures from 150 °C to 350 °C.