Supporting Information

High Efficiency and High Open Circuit Voltage Quadruple-Junction Silicon Thin Film Solar Cells for Tomorrow's

Electronic Applications

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Fig. S1 AM1.5 illuminated *J-V* curves of a-SiGe: H single-junction thin-film solar cells (TFSCs) fabricated with low (dash line), medium (dash dot line), and high (solid line) germanium contents in a-SiGe: H absorbers.

The a-SiGe: H single-junction solar cells with low, medium, and high germanium contents were deposited on ZnO:Al (AZO) substrates, followed by highly-reflective boron doped ZnO (BZO)/Ag/Al back-reflectors. Fig. S1 demonstrates that by reducing the germanium content in the a-SiGe: H absorber, the open-circuit-voltages (V_{oc}) of a-SiGe: H single-junction TFSCs can be effectively tuned from 0.71 V to 0.81 V, accompanying with the increased fill factors and decreased short-circuit current densities (J_{sc}). Our results suggest the highly flexible tunability of a-SiGe: H third component cells in quadruple-junction (4J) TFSCs.



Fig. S2 AM1.5 illuminated *J-V* curves of μ c-Si: H single-junction TFSCs fabricated with low (solid line), medium (dash dot line), and high (dash line) silane (SiH₄) concentrations in absorbers. Corresponding device parameters are also noted in.

Fig. S2 displays the illuminated J-V curves of the µc-Si: H single-junction TFSCs

fabricated with low, medium, and high silane concentrations (SC) in the absorber. We can see that the V_{oc} of the µc-Si: H single-junction TFSCs is flexibly tunable from 0.485 V (low SC) to 0.559 V (high SC). This V_{oc} tunability of µc-Si: H bottom component cells offers more choices in our 4J TFSCs to meet the special requirements of various applications. Note that the J_{sc} values are also tunable from 26.0 mA/cm² (high SC) to 29.75 mA/cm² (low SC).



Fig. S3 (a) Atomic force microscope (AFM) image (scanning area: $20 \ \mu m \times 20 \ \mu m$) of the AZO substrate in a-SiC: H/a-Si: H/a-SiGe: H/ μ c-Si: H 4J TFSCs. (b) Extracted and fitted autocorrelation functions (ACF) to determine the lateral correlation length and roughness of the AZO surface morphology. Solid line: ACF extracted from the AFM image. Open circle: fitted ACF.

The correlation length and roughness of the AZO surface morphology were determined from the AFM image in Fig. S3a with the formula:

$$ACF(\tau_x) = \sigma_{rms}^2 e^{-\frac{\tau_x^2}{l_c^2}}$$

where σ_{rms} is the surface roughness, and l_c is the correlation length. The two parameters were estimated to characterize the typical lateral and vertical lengths of the AZO substrate.



Fig. S4 Surface inclination angle distribution of the employed AZO substrate in a-SiC: H/a-Si: H/a-SiGe: H/µc-Si: H 4J TFSC.



Fig. S5 Cross-sectional scanning electron microscope (SEM) images of the a-SiC: H 1J, a-SiC: H/a-Si: H 2J, and a-SiC: H/a-Si: H/a-SiGe: H 3J TFSCs.



BOGUS Area defined by back contact. No mask used. Temperature from Wahl surface probe.

Fig. S6 NREL measured 4J TFSC efficiency.