Supplementary Information

Opportunities and Challenges for Emerging Inorganic Chalcogenide-Silicon Tandem Solar Cells

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Note 1

Previous studies on metal-chalcogenide-based thin-film solar cells with multi-layered structures have shown that surface roughness is a critical factor in achieving high PCE. A rough surface can lead to a high recombination rate due to the formation of defects and defect clusters or cause shunt current loss through shunting paths in thin-film solar cells. To address this issue, various strategies have been explored, including doping, compositional ratio control, optimization of post-annealing parameters, additives in the precursor solution, crystal orientation control, surface etching, and the introduction of seed or interfacial layers, as summarized in **Table S1**.

Table S1 The large area roughness of chalcogenides; CIGS, CZTSSe, and $Sb_2(S,Se)_3$ by various deposition processes.

Chalcogenide materials	**Deposition process**	**Average Roughness (nm)**	**Control strategy**	**Device performance (%)**	**refs**
CIGS	Sputtered metallic precursor and post-annealing	200	Surface etching	10	1
	Three stage co-evaporation	90	Surface etching	14	2
	Three stage co-evaporation	80	Modification of initial stage	.	3
	Hydrazine-based precursor and post-annealing	8.6	Sb doping	12.3	4
	Spin coated precursor and post-annealing	60	.	3.4	5
	Three stage co-evaporation	44	Alkali doping	.	6
	PLD-based precursor and	25	.	7.25	7,8

	post-annealing				
	Three stage co-evaporation	55	Control of composition	17.4	9
	Three stage co-evaporation	34.9	.	12.3	10
Kesterite	Co-electrodeposited metallic precursor and post-annealing	140	Modification of metallic precursor	8.64	11
	Sputtered metallic precursor and post-annealing	150	Deposition of overlayer	10.1	12
	Co-electrodeposited metallic precursor and post-annealing	153	Control of metallic precursor	9.5	13
	Solution-based precursor and post-annealing	105	Al doping	9.25	14
	Co-electrodeposited metallic precursor and post-annealing	322	.	9.4	15
	Sputtered metallic precursor and post-annealing	117	Control of compositional ratio	9.7	16
	Sputtered precursor and post-annealing	49.5	Deposition of overlayer	10.2	17
	Solution-based precursor and post annealing	55.7	Sb doping	13.11	18
	Solution-based precursor and post-annealing	67	Mg doping	9.0	19
	Spin-coated precursor and post-annealing	68.2	Ti doping	12.07	20
	Solution-based precursor and post-annealing	50.27	Ag doping	11.23	21
$Sb_2(S,Se)_3$	Hydrothermal	31.6	Surface etching	10	22

Vapor thermal deposition	10.7	Vapor atmosphere	7.27	[23]
Chemical bath deposition	38.5	Treatment of oxygen plasma	6.7	[24]
Spin coating-based precursor and post-annealing	6	Ag doping	7.73	[25]
Hydrothermal	28	Addition of ethanol in precursor	10.75	[26]
Chemical bath deposition	18.7	.	8.06	[27]
Hydrothermal	20.8	Additive in precursor solution	9.94	[28]
Hydrothermal	15.5	Addition of $BaBr_2$ in precursor solution	10.12	[29]
Hydrothermal and CSS process	5.3	Addition of see layer	7.4	[30]
CSS	17	.	4.27	[31]
Spin coating -based precursor and post-annealing	27.10	Control of crystal orientation	9.21	[32]

Table S2. The device parameters for representative emerging absorber/Si TSCs

Device structure	Tandem types	E_g (eV) (Top /bottom)	V_{oc} (V)	J_{sc} (mA /cm^2)	FF (%)	PCE (%)	Theoretical PCE Limit (%)	Challenges and Solutions
CZTS/Si[33]	2T	1.5/ 1.12	0.899	6.9	17.6	1.10	32-35	Band tailing, Voc-Deficit – Needs interface passivation and defect control
CZTS/Si[33]			0.948	6.3	58.3	3.5		Low doping density, back-contact losses
CZTS/Si[34]			0.997	8.1	47.2	3.9		KCN etching and alkali doping improve Voc
CZTS/Si[35]			1.083	10.8	58.6	6.80		
CGS/Si[35]	2T	1.7/1.12	1.30	8.01	49.32	5.13	35-40	Stability under light exposure needs improvement
CGS/Si[36]			1.32	12.3	59.4	9.7		
$Sb_2(S,Se)_3$/Si[37]	4T	1.5/1.12	0.526/ 0.492	23.5/ 139	57/67.5	7.05/ 4.61 (11.66)	28-32	Large Voc-deficit due to deep trap states
Sb_2S_3/Si[38]	2T	1.71/1.1	0.367	0.01	28	.	28-32	Growth control needed to avoid phase segregation
GaInP/GaAs/Si[39] (stacked)	4T		2.52/0.681	13.6/11.0	87.5/78	35.9	40-45	Expensive III-IV material growth, needs cost reduction
GaInP/GaInAsP/Si[39] (bonded)	2T		3.30	12.7	86.0	36.1	40-45	High processing costs, scalability challenge
PSK/Si[40]	2T	.	1.96	20.76	83.0	33.9	35-40	Stability and

								lead toxicity concerns
PSK/PSK[41]	2T	1.25 /1.78	2.159	16.59	78.9	28.2	35-40	Stability and lead toxicity concerns
PSK/CIGSe[42]	2T	.	1.76	19.24	72.9	24.2	30-35	Needs interface engineering for stability
PSK/CIGSe[43]	4T	1.63 /1.00	1.24/0.57	21.5/19.3	81.9/73.7	21.8 /8.1 (29.9)	30-35	Long-term stability still a challenge

Table S3 Ratio of different device parameters of champion chalcogenide and high-performance PV to their theoretical limit (S-Q).

Absorber	**PCE/ $PCE_{(S-Q)}$**	**V_{oc}/ $V_{oc(S-Q)}$**	**J_{sc}/ $J_{sc(S-Q)}$**	**FF/ $FF_{(S-Q)}$**	**V_{oc}×FF/ V_{oc}×$FF_{(S-Q)}$**
$AgBiS_2$	0.31	0.50	0.76	0.82	0.41
$AgBiS_2$	0.27	0.44	0.81	0.77	0.34
CGSe	0.39	0.76	0.70	0.74	0.56
CIGS	0.55	0.72	0.97	0.79	0.57
CIS	0.37	0.55	0.86	0.79	0.44
CZGS	0.26	0.53	0.79	0.62	0.33
CZTS	0.42	0.59	0.86	0.76	0.45
CZTSSe	0.46	0.63	0.87	0.85	0.53
CZTSSe-Ag	0.43	0.66	0.80	0.81	0.53
CZTSSe-Ge	0.37	0.60	0.74	0.83	0.50
GeS	0.05	0.37	0.37	0.34	0.13
SnS	0.15	0.34	0.69	0.62	0.21
$Sb_2(S,Se)_3$	0.30	0.51	0.75	0.79	0.40
$Sb_2(S,Se)_3$	0.33	0.52	0.85	0.75	0.39

$Sb_2(S,Se)_3$	0.34	0.53	0.86	0.74	0.39
Sb_2S_3	0.28	0.52	0.80	0.66	0.35
Sb_2S_3	0.28	0.47	0.82	0.71	0.33
Sb_2Se_3	0.32	0.48	0.87	0.77	0.37
Sb_2Se_3	0.24	0.42	0.82	0.70	0.30
Si-C	0.83	0.86	0.96	1.00	0.86
GaAs	0.88	0.97	0.93	0.97	0.94
Perovskite	0.84	0.95	0.95	0.94	0.89
CdTe	0.69	0.79	0.96	0.90	0.72
CIGSe	0.71	0.87	0.88	0.93	0.80

Note 2

The excellent research and reviews for the PCE degradations, mechanism, and long-term stability results in CIGS-based thin film solar cells have been extensively investigated.[44–46] In contrast, the PCE degradations and demonstrating mechanisms for the kesterite- and $Sb_2(S,Se)_3$ are lacking. Thus, the detailed PEC degradation investigations and mechanisms for kesterite- and $Sb_2(S,Se)_3$-based thin film solar cells should be studied to realize early commercialization.

Table S4 The stability failure of chalcogenide-based SC; CIGS, CZTSSe, and $Sb_2(S,Se)_3$, respectively.

Chalcogenide materials	Cell or module information	Stability and atmosphere	Failures	refs
	Cells	Less than 10% after 480 h under damp and heat conditions (85% humidity and 85 °C)	Degradation of buffer and TCO, migration of alkali elements	47
	Mini module	Less than 20% after 1000	Degradation of	48

CIGS		h under encapsulation and damp and heat condition	buffer and TCO, migration of alkali elements	
	Cells	Less than 30% after 800 h under damp and heat condition	migration of alkali elements, Increase the electrical resistivity	49
	Cells	Less than 5% after 336 h under encapsulation and damp and heat condition	migration of alkali-elements	50
	Cells	0% after 50 h under air atmosphere	migration of alkali-elements	51
	Cells	Less than 30% after 800 h under damp and heat condition	Degradation of buffer and TCO	52
	Commercial modules	Less than 5% after 120 h under damp and heat condition	Leakage current density from shunting path	53
	Field module	Less than 15% after 750 h under damp and heat conditions and negative 1000 V biases applied.	Charge carrier concentration or lifetime reduction, TCO corrosion	54
	Mini module on K-rich borosilicate glass	No degradation after 1600 h under damp and heat conditions and positive 1000 V biases applied.	.	55
Kesterite	Cells	Less than 20% after 5000 h under inside box with N_2	.	56
	Cells	Less than 20% after 7200 h under air atmosphere	Degradation of TCO	57
	Cells	Less than 40% after 95 h under heat conditions (105 °C)	.	58

	Cells	Less than 5% after 720 h under air atmosphere	.	59
	Flexible Cells	No degradation after 168 h under ambient light and 80 % humidity		60
	Cells	Less than 10 % after 2400 h under ait atmosphere	Instability for organic HTL	61
$Sb_2(S,Se)_3$	Cells	Less than 5% after 2160 h under air atmosphere	.	62
	Cells	Less than 30% after 100 h under damp and heat condition	.	63
	Cells	Less than 5% after 1440 h under air atmosphere	.	64
	Cells	Less than 5% after 360 h under heat conditions (85 °C) and N_2 atmosphere	.	65
	Cells	No degradation after 500 h under air atmosphere	.	66
	Cells	Less than 20% after 140 h under 85 °C and air atmosphere	Degradation of HTL	67
	Cells	Less than 5% after 960 h under air atmosphere	Instability for organic HTL	68

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