Electronic Supplementary Material (ESI) for Sustainable Energy & Fuels.

Supplementary Information:

Interface Engineering at Hole Transport Layer/Perovskite with Low band gap 2D-Carbon Nitrides For Solar Cells Fabrication

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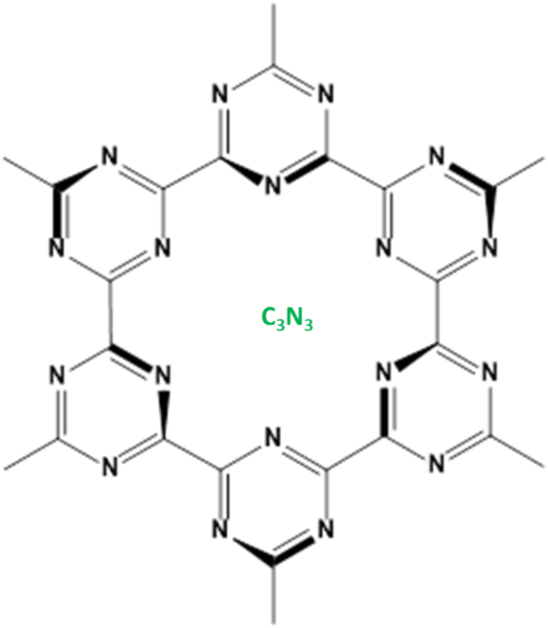

Figure S1: Structure of C_3N_3

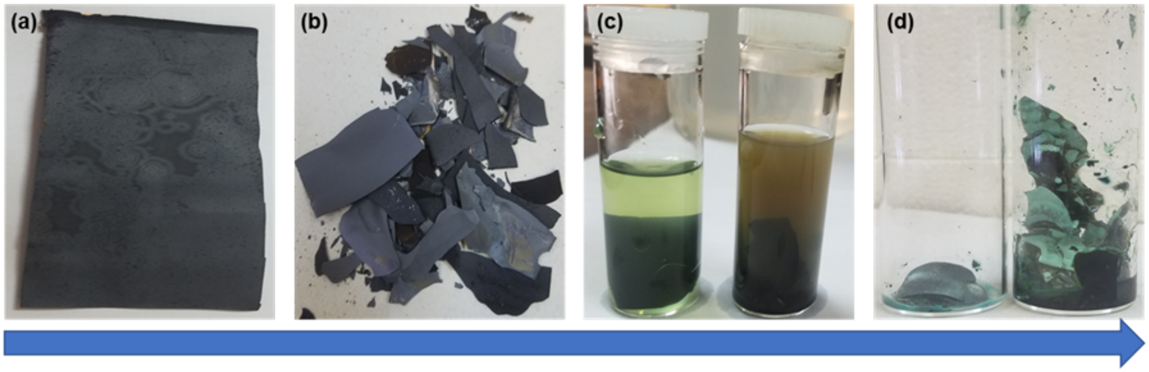

Figure S2: Digital micrograph of the as prepared C_3N_3 before and after washing.

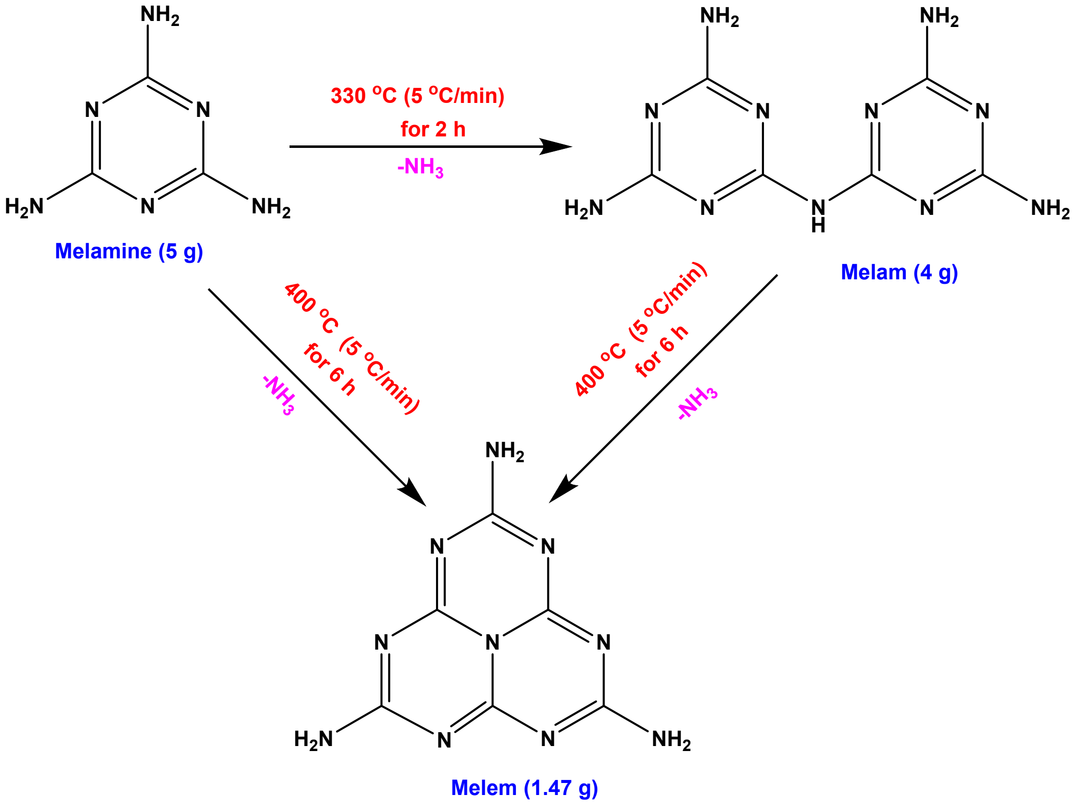

Figure S3: Two steps chemical reaction and structural presentation for the synthesis of melem.

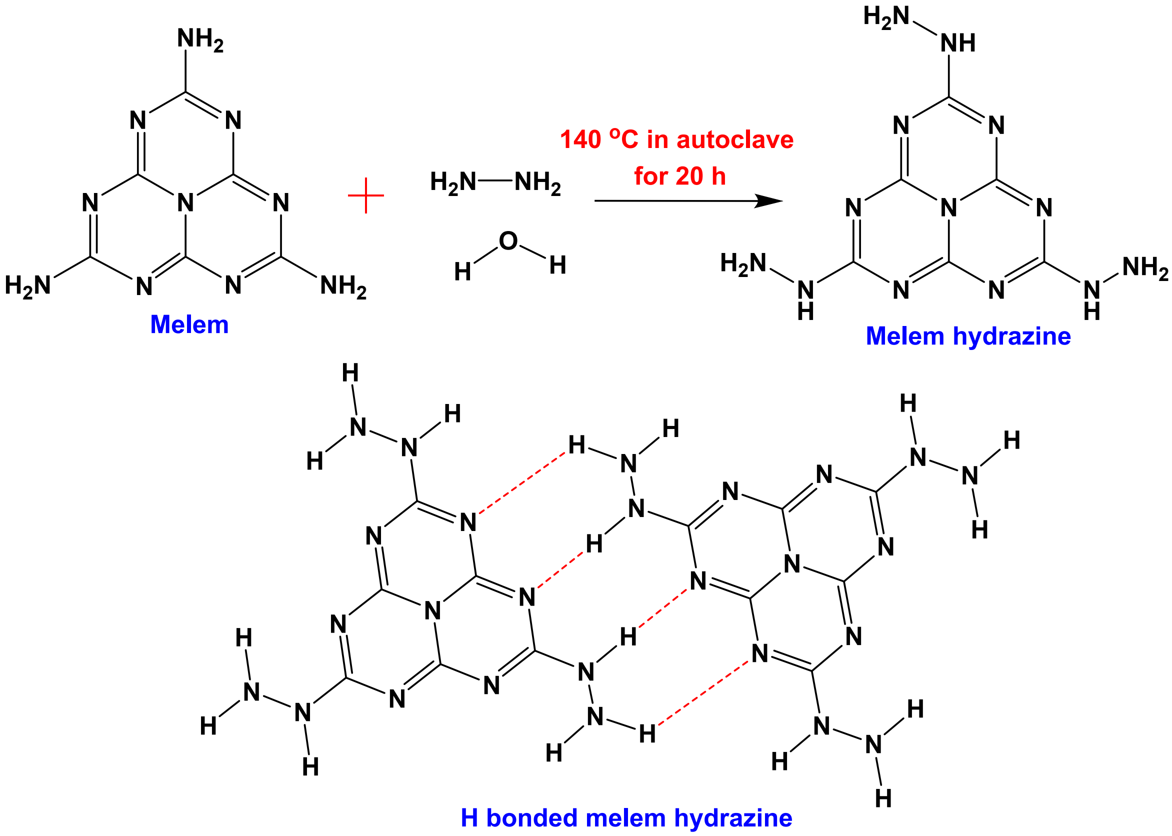

Figure S4: Chemical reaction and structural presentation of melem, melem hydrazine, H-bonded melem hydrazine.

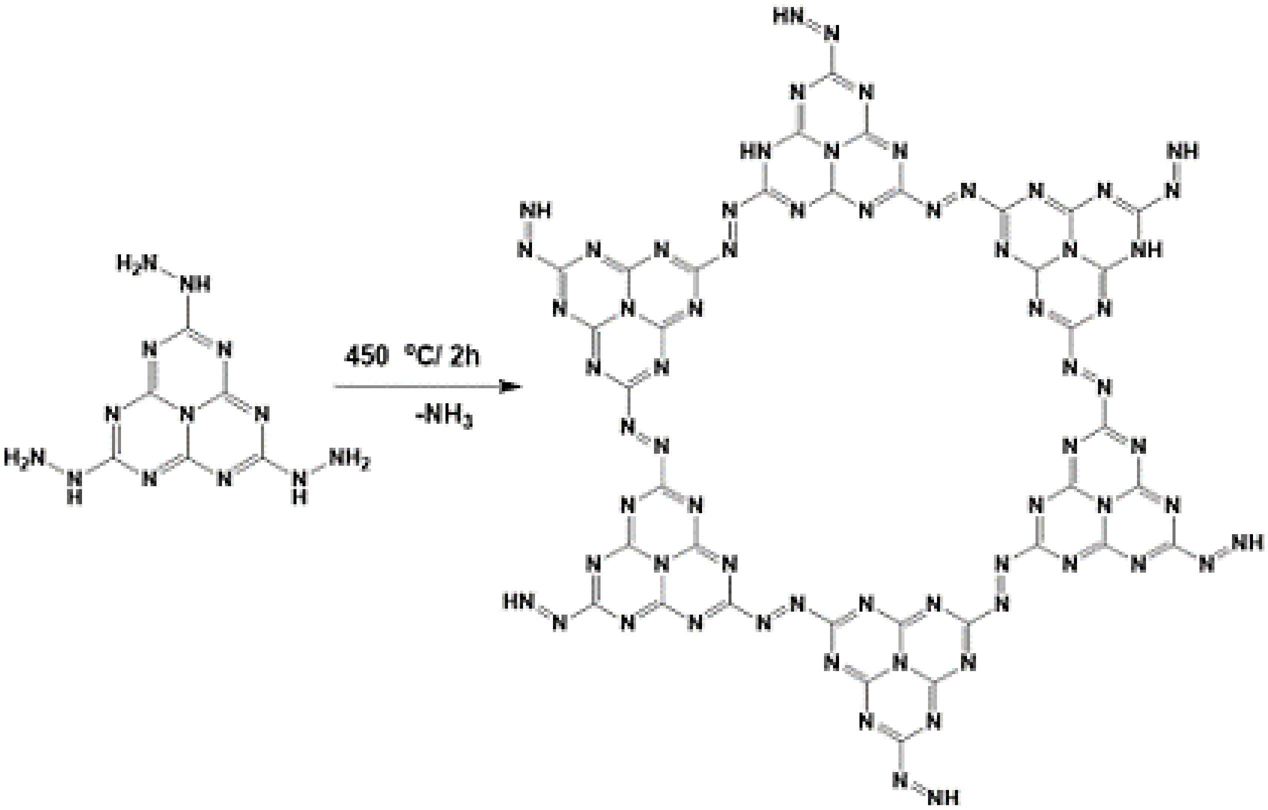

Figure S5: Structure of C_3N_5

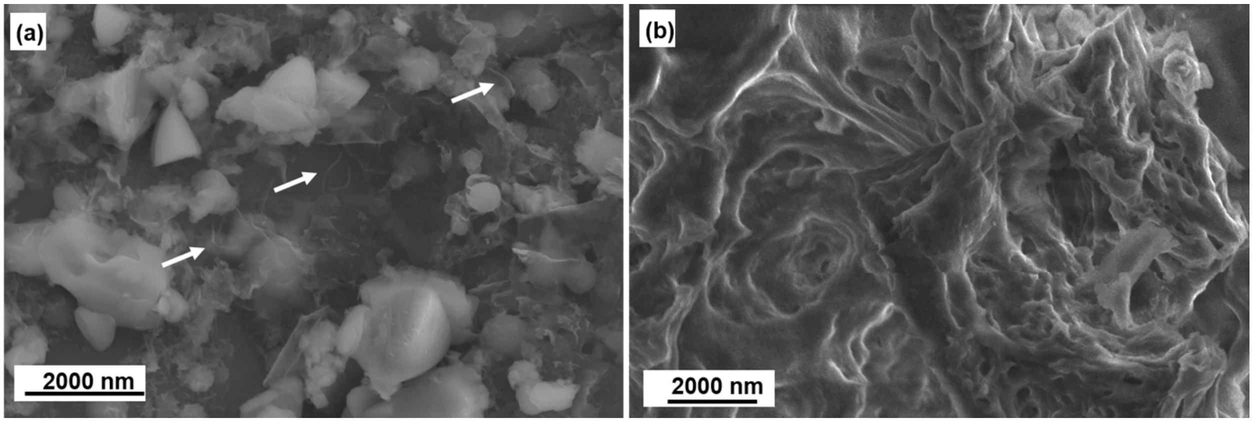

Figure S6: SEM image indicating the 2D layers structure of; (a) C_3N_3 and (b) C_3N_5.

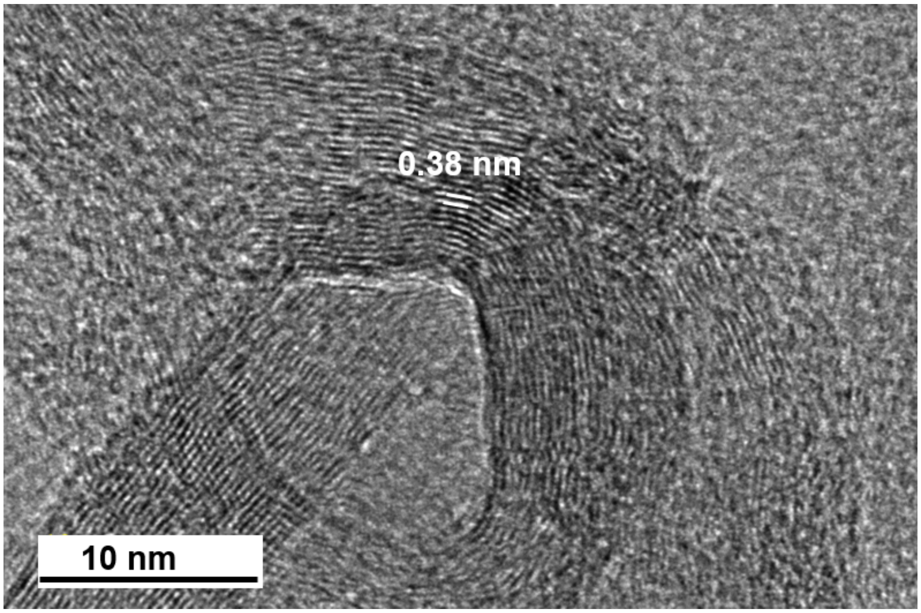

Figure S7: High-resolution TEM image indicating the crystalline nature of C_3N_3 before washing with mineral acids.

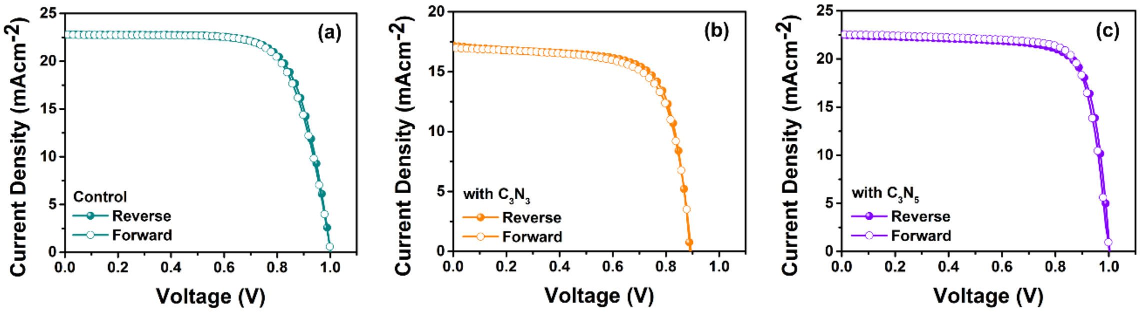

Figure S8: Reverse and forward scan *J-V* curves under simulated AM 1.5G illumination of (a) control, (b) with C_3N_3, and (c) with C_3N_5 based PSCs.

Table S1. Photovoltaic statistics of the devices without the interfacial layer.

Device	V_{oc} (mV)	J_{sc} (mAcm^{-2})	FF (%)	PCE (%)
1	1007.3	22.76	72.46	16.61
2	995.6	22.47	72.35	16.18
3	1004.7	22.54	71.15	16.11
4	1009.9	22.65	72.17	16.51
5	1001.9	22.79	73.07	16.69
Statistics	1003.88±4.9	22.64±0.12	72.24±0.62	16.42±0.23

Table S2. Photovoltaic statistics of the devices with C_3N_3 interfacial layer.

Device	V_{oc} **(mV)**	J_{sc} **(mAcm^{-2})**	**FF (%)**	**PCE (%)**
1	860.29	16.71	68.77	9.89
2	888.30	16.79	70.71	10.55
3	899.31	16.86	70.84	10.74
4	896.15	17.44	70.19	10.97
5	890.16	17.21	71.74	10.99
6	865.29	17.26	71.69	10.71
Statistics	883.2±14.9	17.04±0.27	70.66±1.00	10.64±0.37

Table S3. Photovoltaic statistics of the devices with C_3N_5 interfacial layer.

Device	V_{oc} **(mV)**	J_{sc} **(mAcm^{-2})**	**FF (%)**	**PCE (%)**
1	999.63	22.40	75.98	17.02
2	998.57	22.24	76.41	16.97
3	1000.85	22.69	75.13	17.06
4	1003.48	22.52	75.95	17.17
5	1003.29	22.39	75.78	17.02
6	1003.15	22.43	74.26	16.71
Statistics	1001.5±1.9	22.45±0.14	75.59±0.70	16.99±0.14

Table S4. R_s, R_{sh}, and HF were extracted from the *J-V* curves of PSCs.

Device	**Scan direction**	R_s **(Ω)**	R_{sh} **(kΩ)**	**HF (%)**
Control	Reverse	52.33	42.78	1.44
	Forward	62.48	34.71	
with C_3N_3	Reverse	61.19	11.49	3.75
	Forward	70.17	8.91	
with C_3N_5	Reverse	45.97	47.07	-0.85
	Forward	42.12	73.17	