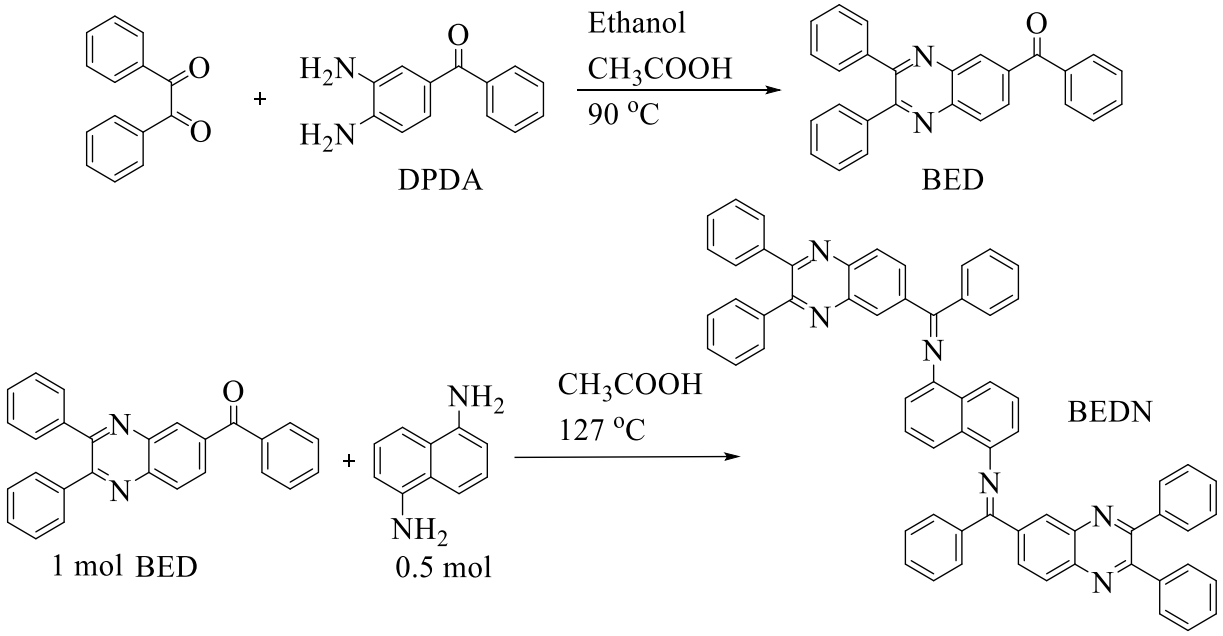
Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supplementary Information for

Sequential Condensation Route as a Versatile Platform for Low Cost and Efficient Hole Transport Material in Perovskite Solar Cell



Scheme 1. Molecular structure and synthetic routes for BEDN.

S1. NMR images of products:

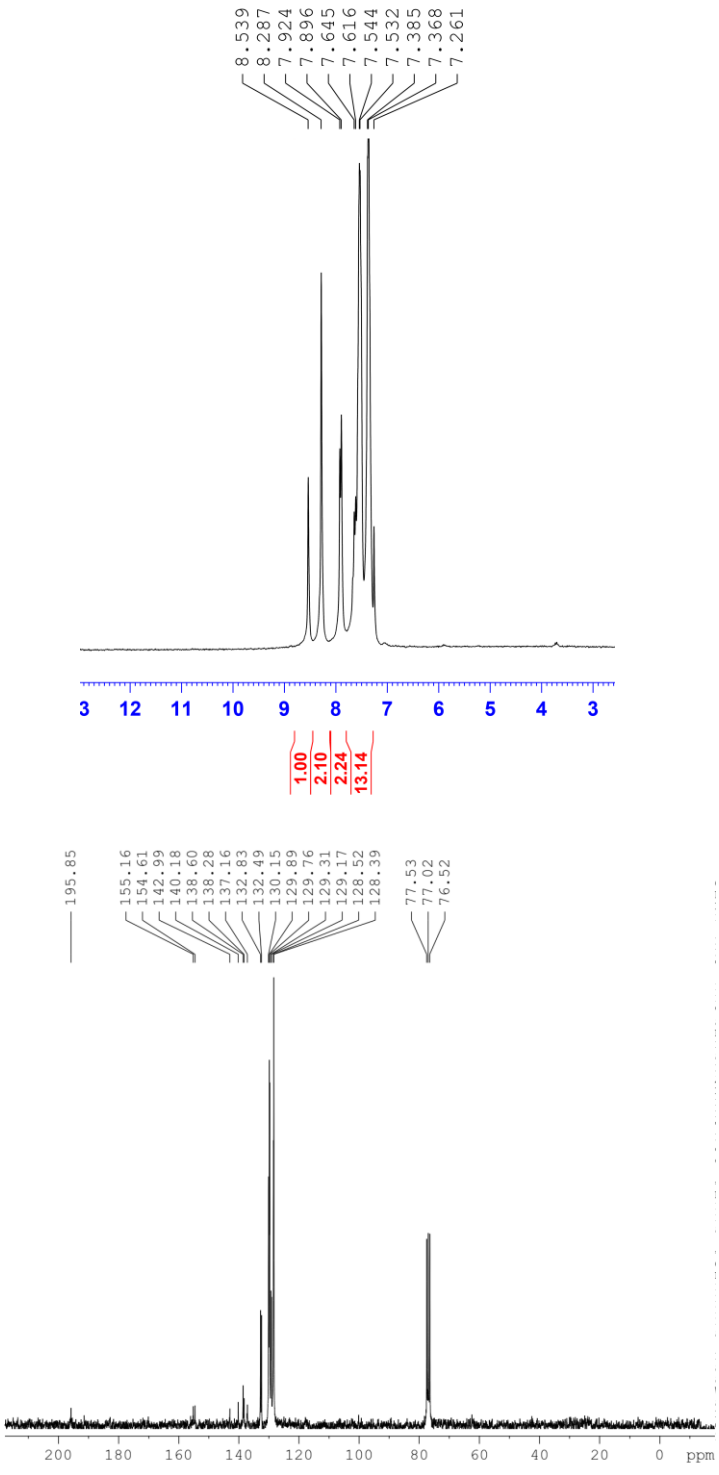


Figure S1. ^{1}H/ ^{13}C NMR of **BEDN** in $CDCl_3$ solvent.

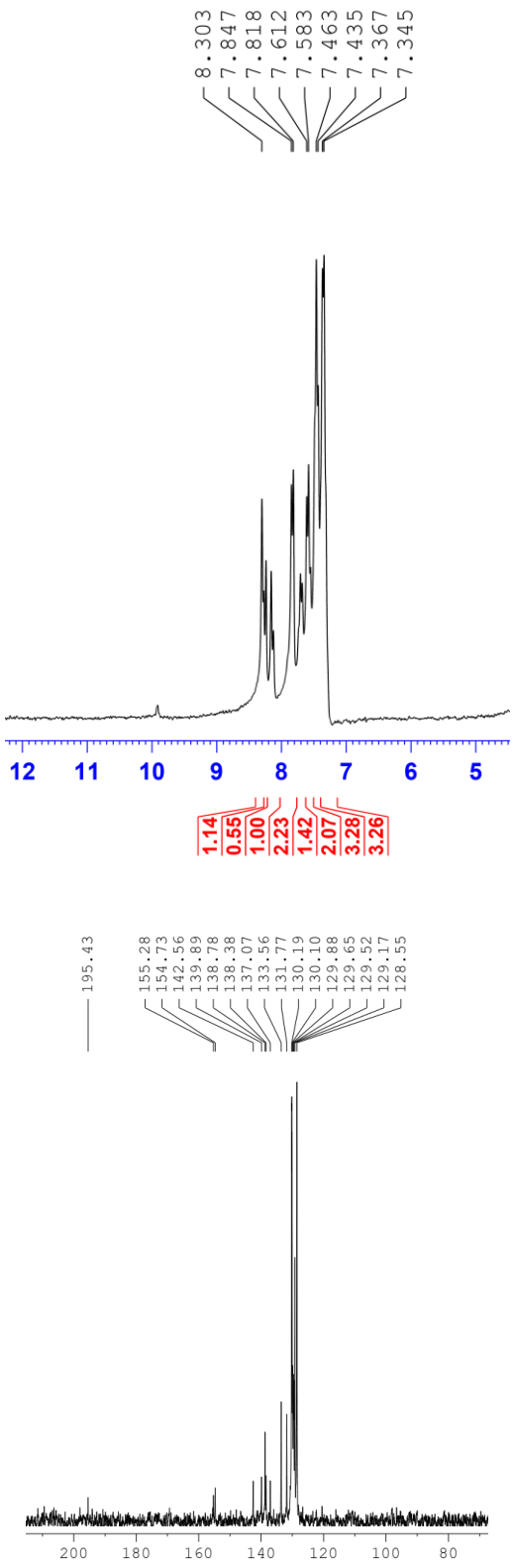
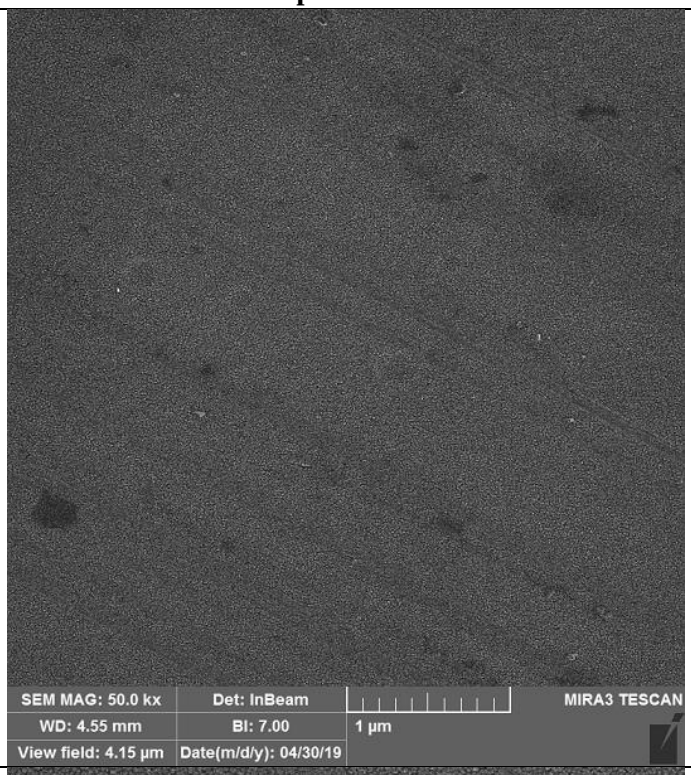
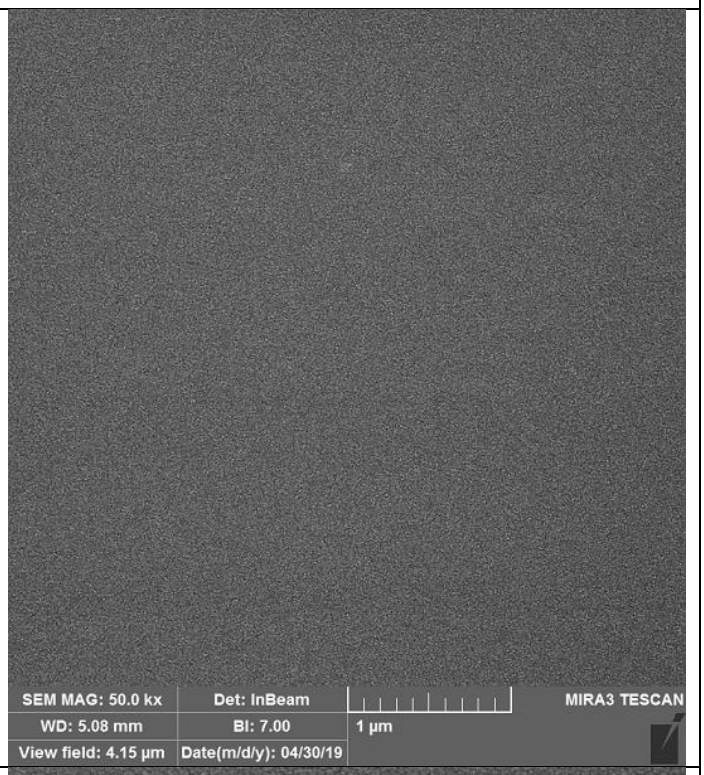
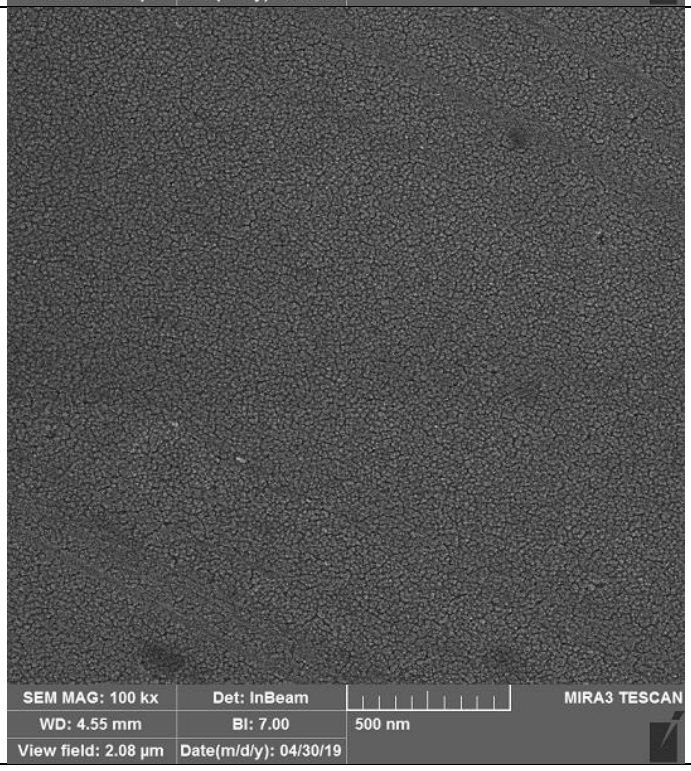
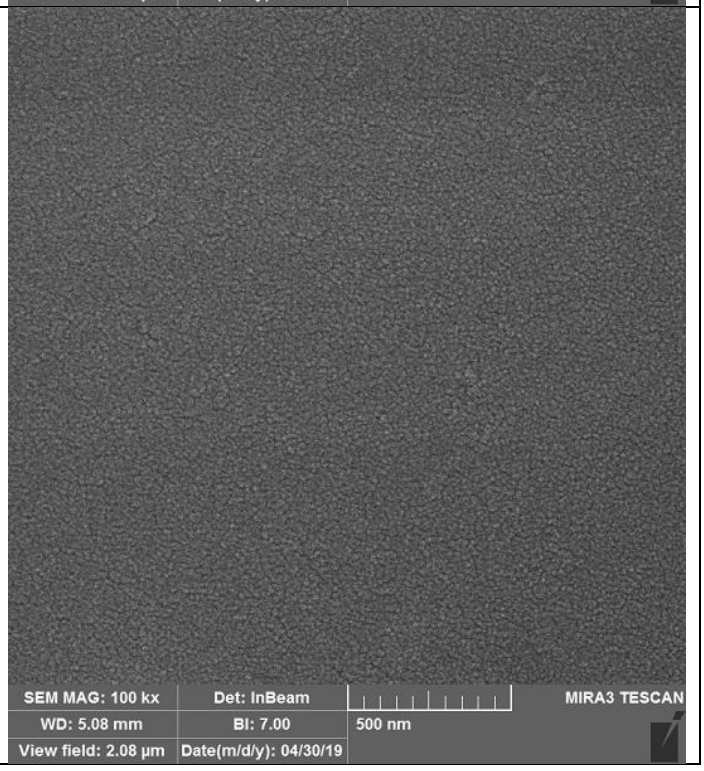


Figure S2. ^{1}H/ ^{13}C NMR of **BEDN** in $CDCl_3$ solvent.

	Glass/spiro-OMeTAD	Glass/ BEDN
1 µm		
500 nm		

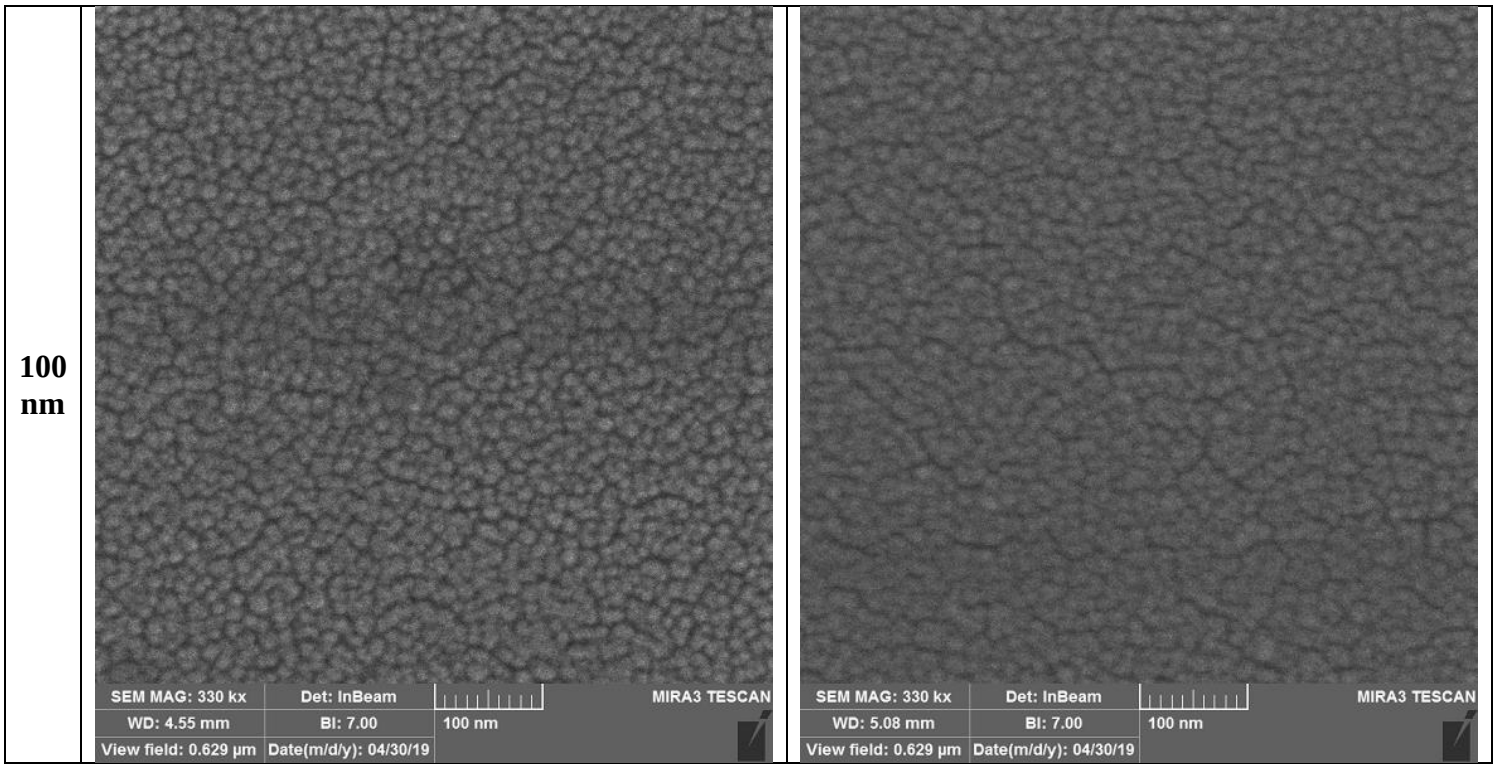
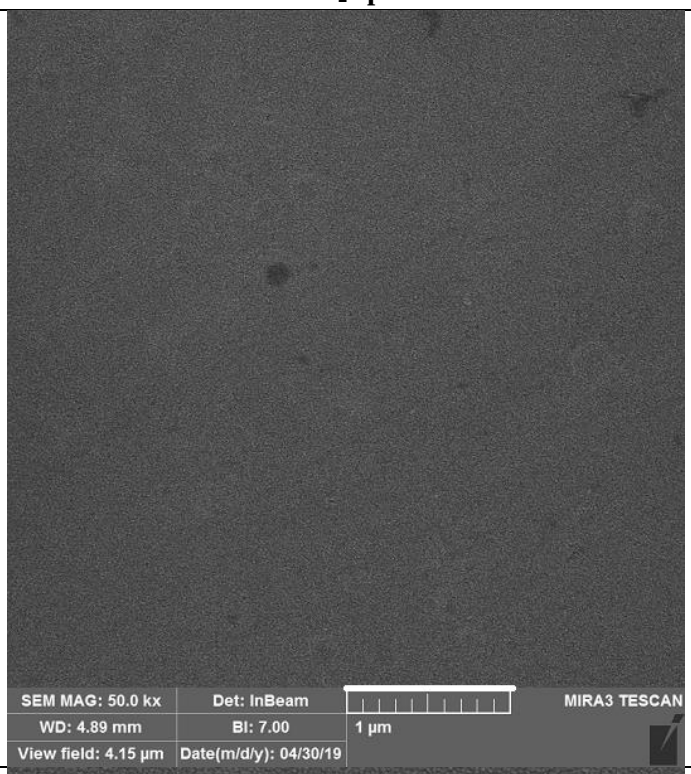
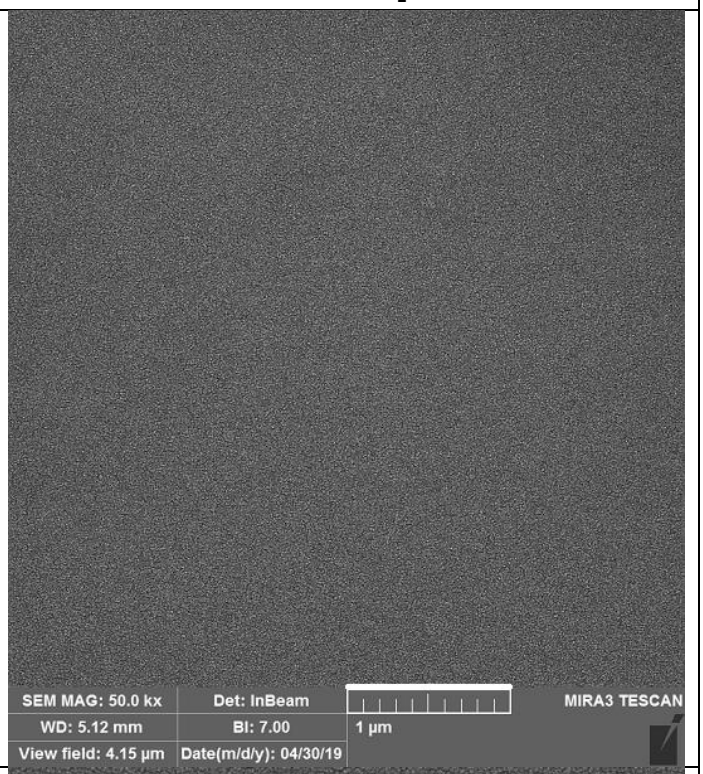
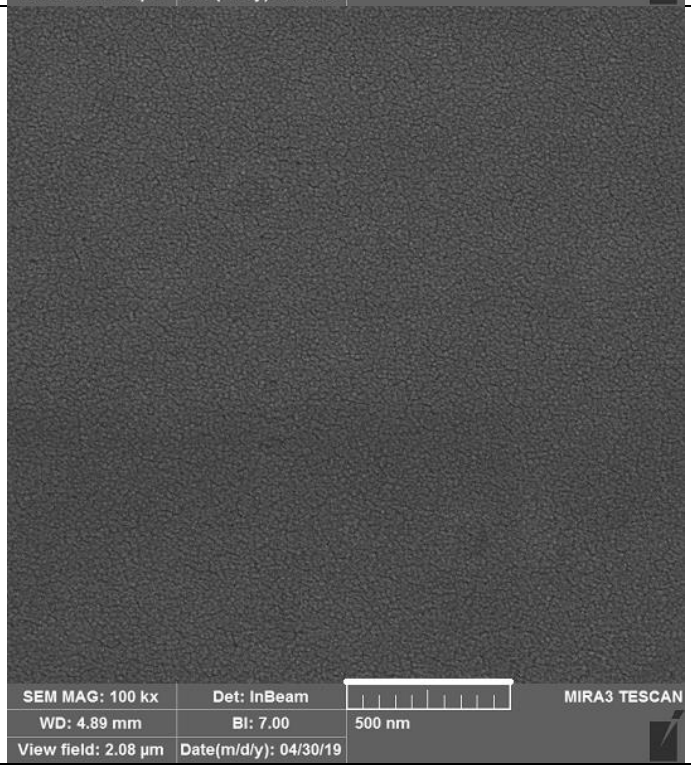
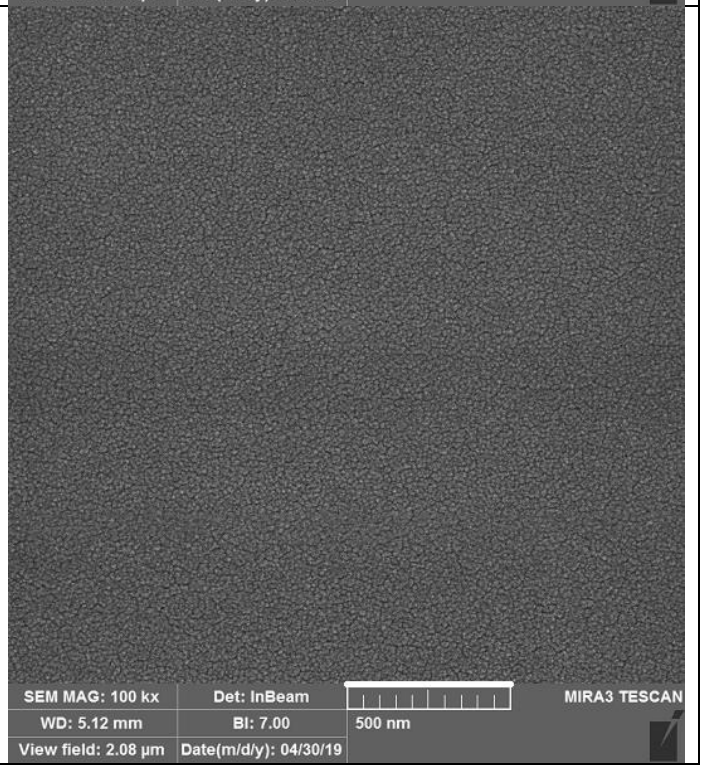


Figure S3. FE-SEM images of the top view of the cells with structures Glass/spiro-OMeTAD (left) and Glass/ BEDN (right).

	Glass/meso-TiO_2/spiro-OMeTAD	Glass/meso-TiO_2/BEDN
1 µm		
500 nm		

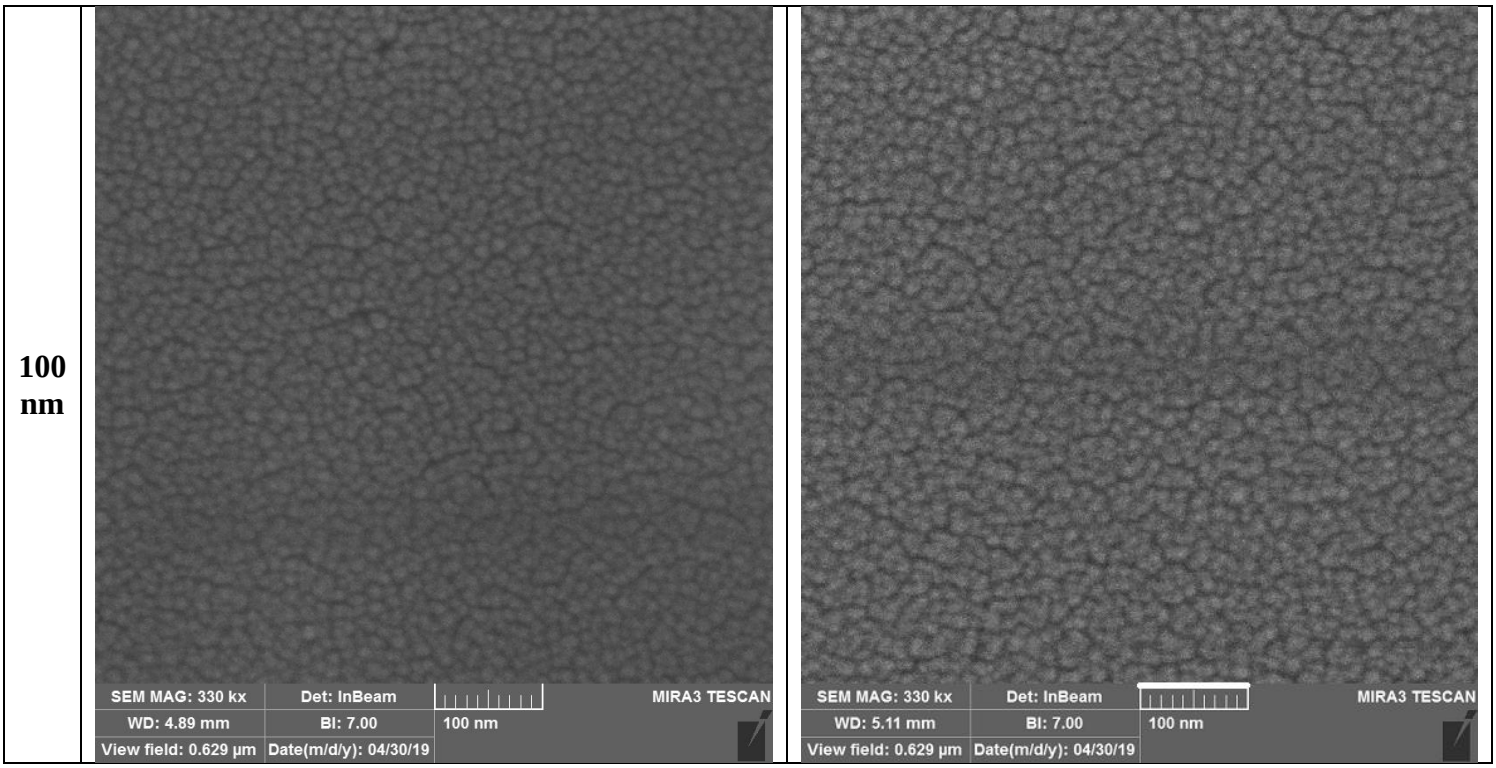
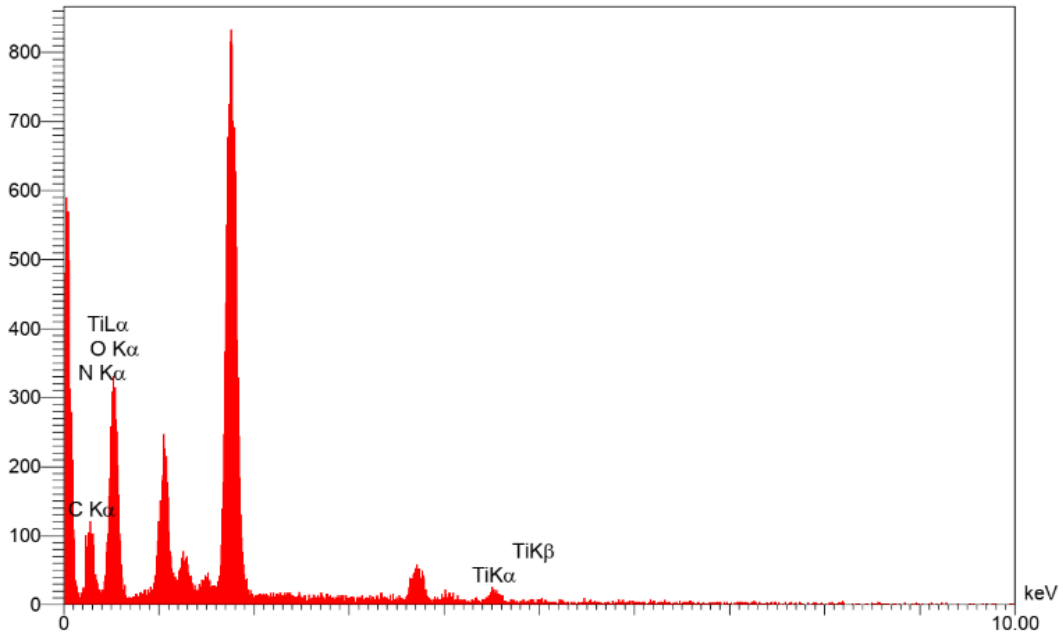


Figure S4. FE-SEM images of the top view of the cells with structure Glass/meso-TiO_2/spiro-OMeTAD (left) and Glass/meso-TiO_2/BEDN (right) .

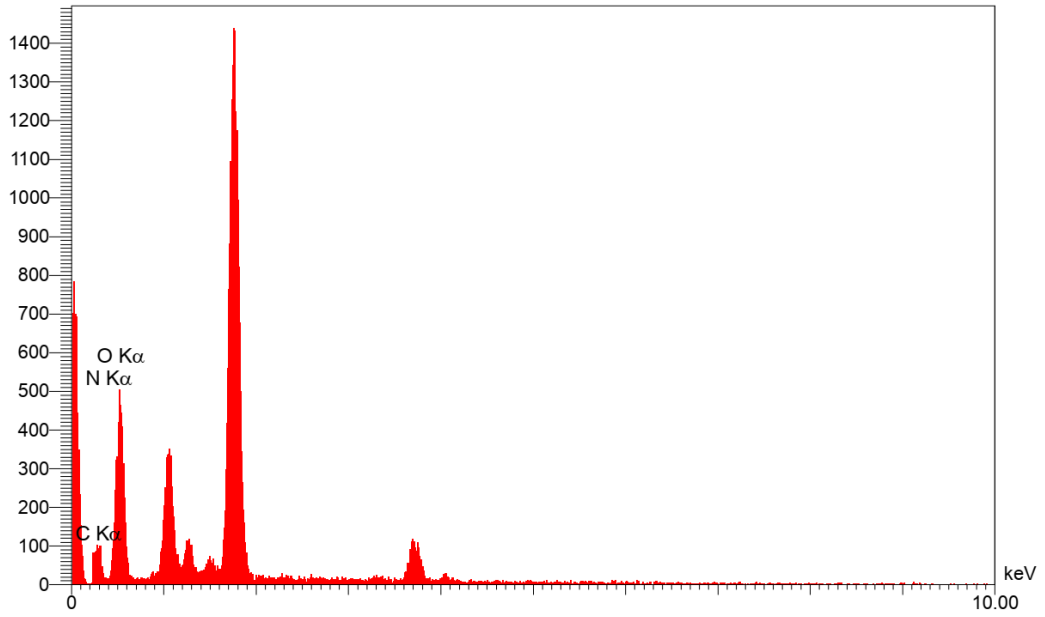
Glass/meso-TiO_2/spiro-OMeTAD



Quantitative Results

Elt	XRay	Int	Error	K	Kratio	W%	A%	ZAF	Formula	Ox%	Cat#
C	Ka	7.4	2.7611	0.2325	0.1074	21.29	26.67	0.5043		0.00	0.00
N	Ka	0.3	2.7611	0.0101	0.0047	1.62	1.74	0.2867		0.00	0.00
O	Ka	45.1	2.7611	0.7327	0.3384	75.64	71.14	0.4470		0.00	0.00
Ti	Ka	1.5	0.1950	0.0247	0.0114	1.45	0.46	0.7867		0.00	0.00
				1.0000	0.4618	100.00	100.00			0.00	0.00

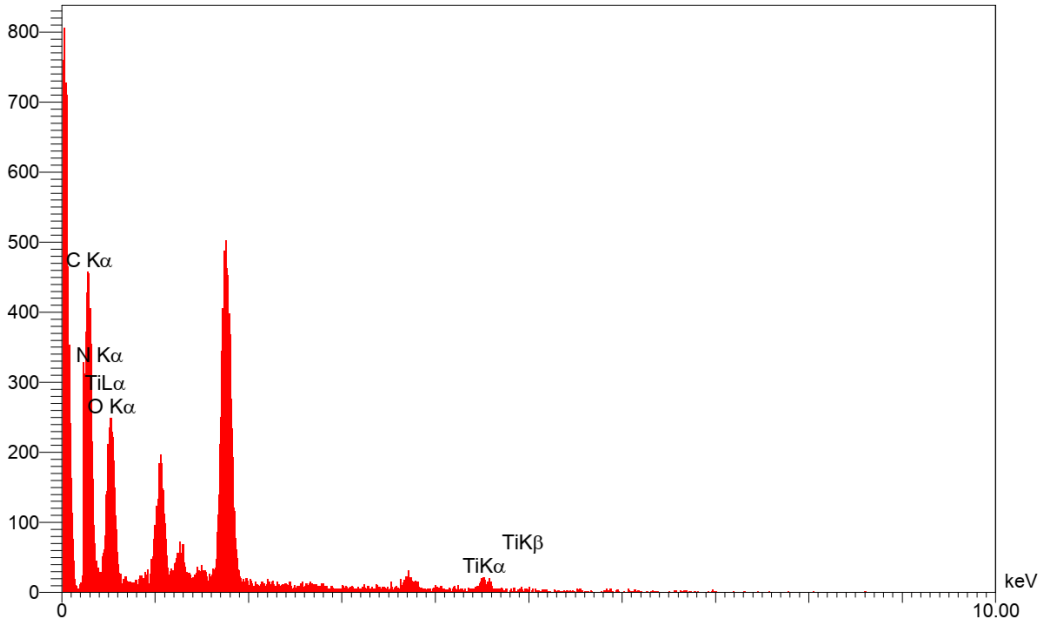
Glass/spiro-OMeTAD



Quantitative Results

Elt	XRay	Int	Error	K	Kratio	W%	A%	ZAF	Formula	Ox%	Cat#
C	Ka	29.0	4.3299	0.2507	0.1076	21.09	26.07	0.5101		0.00	0.00
N	Ka	3.5	4.3299	0.0370	0.0159	5.49	5.82	0.2892		0.00	0.00
O	Ka	159.5	4.3299	0.7123	0.3057	73.42	68.12	0.4161		0.00	0.00
				1.0000	0.4292	100.00	100.00			0.00	0.00

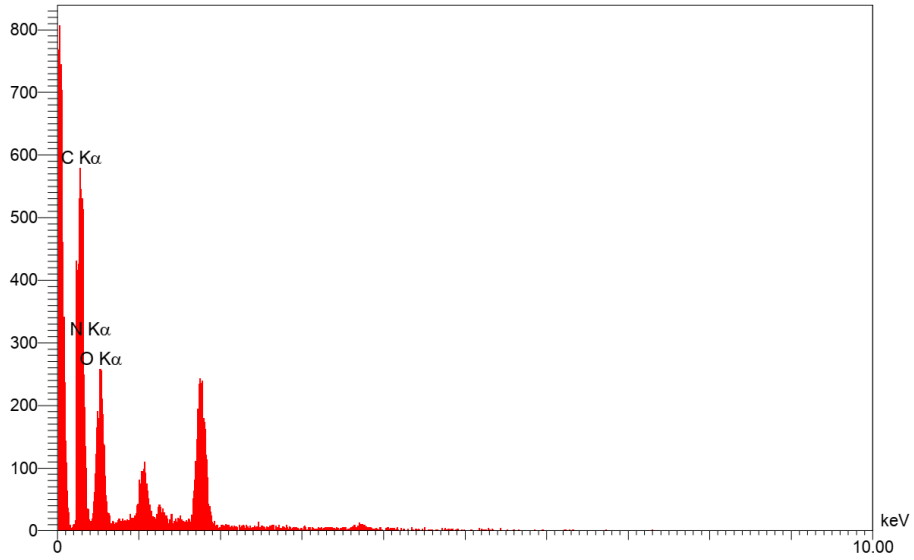
Glass/meso-TiO_2/BEDN



Quantitative Results

Elt	XRay	Int	Error	K	Kratio	W%	A%	ZAF	Formula	Ox%	Cat#
C	Ka	69.2	24.5607	0.6840	0.2919	47.38	54.69	0.6160		0.00	0.00
N	Ka	0.7	24.5607	0.0084	0.0036	2.56	2.53	0.1399		0.00	0.00
O	Ka	56.5	24.5607	0.2888	0.1232	49.03	42.48	0.2513		0.00	0.00
Ti	Ka	3.5	0.3933	0.0189	0.0080	1.04	0.30	0.7768		0.00	0.00
				1.0000	0.4267	100.00	100.00			0.00	0.00

Glass/BED N



Quantitative Results

Elt	XRay	Int	Error	K	Kratio	W%	A%	ZAF	Formula	Ox%	Cat#
C	Ka	160.3	11.1188	0.7718	0.3346	51.57	58.17	0.6488		0.00	0.00
N	Ka	3.4	11.1188	0.0201	0.0087	6.81	6.58	0.1281		0.00	0.00
O	Ka	83.6	11.1188	0.2081	0.0902	41.62	35.25	0.2167		0.00	0.00
				1.0000	0.4336	100.00	100.00			0.00	0.00

Figure S5. EDS images of the top view of the cells with structure Glass/meso-TiO_2/HTM and Glass/meso-TiO_2/HTM.

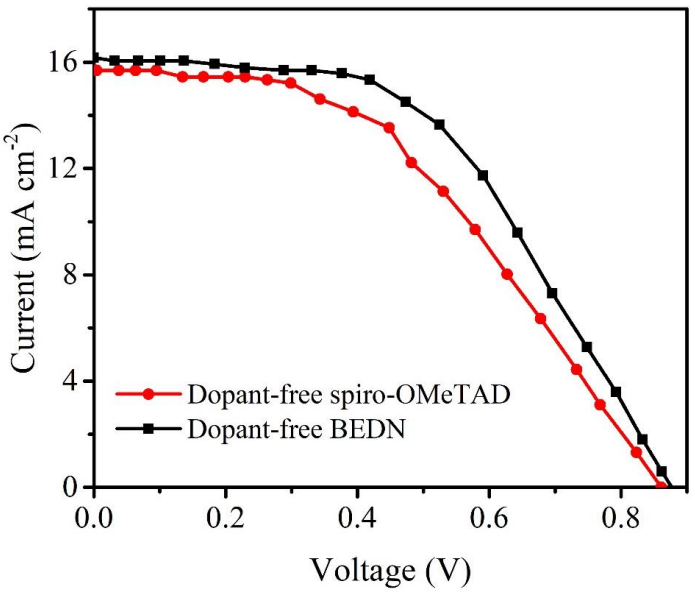


Figure S6. J-V curves of PSCs based on dopant-free BEDN and dopant-free spiro-OMeTAD HTMs.

Table S1. The photovoltaic parameters derived from J-V curves for the PSCs based on dopant-free HTMs.

HTMs	VOC [V]	JSC [mA cm^{-2}]	FF [%]	PCE [%]
Dopant-free BEDN	0.88	16.12	50	7.10
Dopant-free spiro-OMeTAD	0.86	15.72	44	5.94

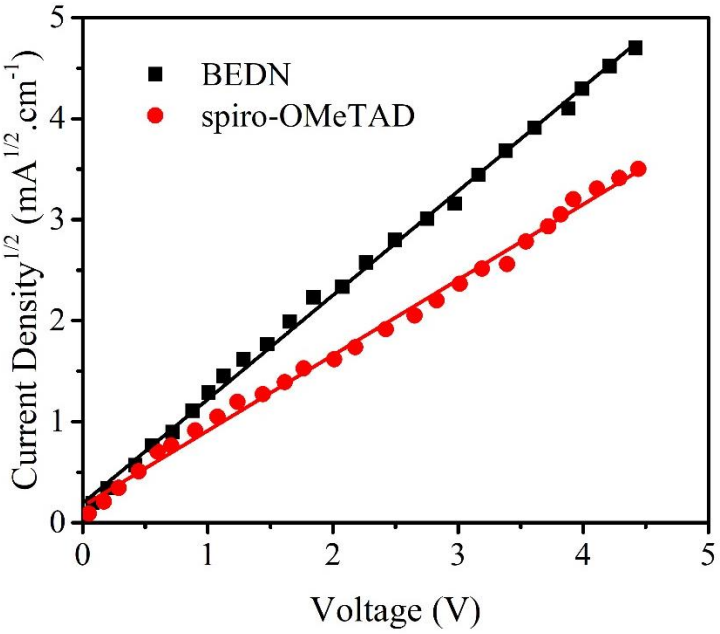


Figure S7. $J^{1/2}$-V curves of dopant-free BEDN and spiro-OMeTAD based devices with a architecture of ITO/PEDOT:PSS/HTMs/Au.

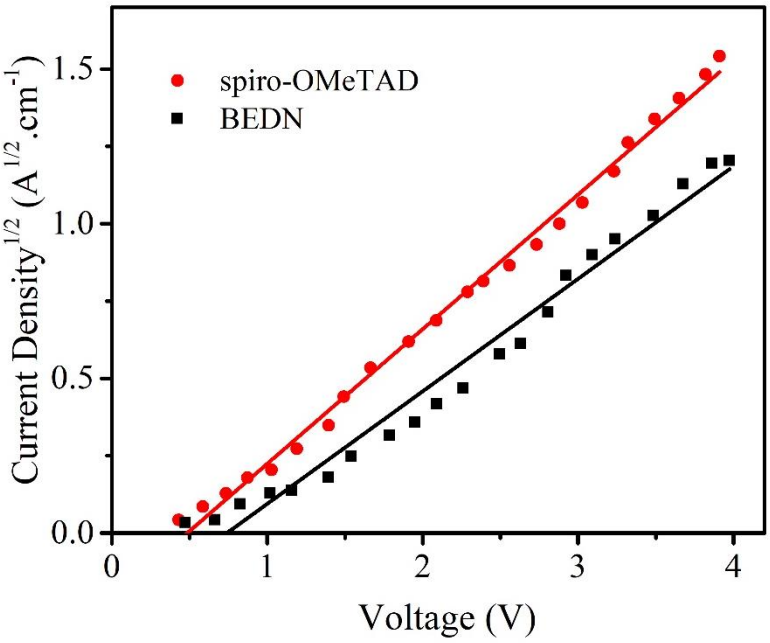


Figure S8. $J^{1/2}$-V curves of BEDN and spiro-OMeTAD based devices with a architecture of ITO/PEDOT:PSS/HTMs:Dopants/Au.

S2. Synthesis cost estimation of 1 gram BEDN

We estimated the synthesis cost of 1 gram BEDN according to the cost models of Petrus et al.[1] and Osedach et al.[2] The price of the materials used has been obtained from Merck, Sigma Aldrich, DeJong companies. We compared the price of 1 gram of this new HTM with the price of 1 gram of spiro-OMeTAD, which is reported in the literature.[3-5]

Table S2. Materials quantities and cost for the synthesis of BEDN.

Chemical	Weight reagent (g)	Weight solvent (g)	Weight workup (g)	Price of Chemical ($/kg)	Material cost ($/g product)	Cost per step ($/step)
3,4-Diaminobenzophenone	0.422			580	0.24	
benzil	0.42			350	0.151	
Ethanol	-	39.45	-	15.23	0.6	0.98
Ethanol	-	-	15.78	9.53	0.15	
CH_3COOH	-	2.1	-	40.12	0.08	
1,5-Diaminonaphthalene	0.078	-	-	1460	0.11	
CH_3COOH	-	5		40.12	0.2	0.4
CH_3COOH			10	10	0.1	
Total	0.92	46.55	25.78	-	-	1.38

References:

1. M. Petrus, T. Bein, T. Dingemans and P. Docampo, *J. Mater. Chem. A*, 2015, **3**, 12159-12162.
2. T. P. Osedach, T. L. Andrew and V. Bulović, *Energy Environ. Sci.*, 2013, **6**, 711-718.
3. B. Xu, D. Bi, Y. Hua, P. Liu, M. Cheng, M. Grätzel, L. Kloo, A. Hagfeldt and L. Sun, *Energy Environ. Sci.*, 2016, **9**, 873-877.
4. M. Saliba, S. Orlandi, T. Matsui, S. Aghazada, M. Cavazzini, J.-P. Correa-Baena, P. Gao, R. Scopelliti, E. Mosconi and K.-H. Dahmen, *Nat. Energy*, 2016, **1**, 15017.
5. C. H. Teh, R. Daik, E. L. Lim, C. C. Yap, M. A. Ibrahim, N. A. Ludin, K. Sopian and M. A. M. Teridi, *J. Mater. Chem. A*, 2016, **4**, 15788-15822.