

## Approaching truly sustainable solar cells by the use of water and cellulose derivatives

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### SUPPORTING INFORMATION

**Table S1 – Ionic conductivity at ambient temperature of gel electrolytes containing NaI 0.50 M and different amount of CMC.**

CMC (wt%)	Run	Z (Ω)	$\sigma$ (S cm <sup>-1</sup> )	CMC (wt%)	Run	Z (Ω)	$\sigma$ (S cm <sup>-1</sup> )
3	1	0.63	1.03×10 <sup>-2</sup>	12.5	1	0.67	9.70×10 <sup>-3</sup>
	2	0.5	1.30×10 <sup>-2</sup>		2	0.62	1.05×10 <sup>-2</sup>
5	1	0.75	8.66×10 <sup>-3</sup>	15	1	0.56	1.16×10 <sup>-2</sup>
	2	0.55	1.18×10 <sup>-2</sup>		2	0.65	9.99×10 <sup>-3</sup>
7.5	1	0.55	1.18×10 <sup>-2</sup>	20	1	0.6	1.08×10 <sup>-2</sup>
	2	0.64	1.02×10 <sup>-2</sup>		2	0.5	1.30×10 <sup>-2</sup>
10	1	0.58	1.12×10 <sup>-2</sup>	10*	1	0.81	8.02×10 <sup>-3</sup>
	2	0.51	1.27×10 <sup>-2</sup>				

\* Blank sample containing CMC 10 wt% in deionized water without NaI.

**Table S2 – Ionic conductivity at different temperatures of gel electrolytes containing NaI 0.50 M and different amounts of CMC.**

<i>T</i> (°C)	CMC 5 wt%		CMC 10 wt%		CMC 20 wt%	
	<i>Z</i> (Ω)	<i>σ</i> (S cm <sup>-1</sup> )	<i>Z</i> (Ω)	<i>σ</i> (S cm <sup>-1</sup> )	<i>Z</i> (Ω)	<i>σ</i> (S cm <sup>-1</sup> )
70	0.54	1.20×10 <sup>-2</sup>	0.49	1.33×10 <sup>-2</sup>	0.57	1.14×10 <sup>-2</sup>
60	0.59	1.10×10 <sup>-2</sup>	0.56	1.16×10 <sup>-2</sup>	0.57	1.14×10 <sup>-2</sup>
50	0.66	9.84×10 <sup>-3</sup>	0.59	1.10×10 <sup>-2</sup>	0.58	1.12×10 <sup>-2</sup>
40	0.65	9.99×10 <sup>-3</sup>	0.59	1.10×10 <sup>-2</sup>	0.59	1.10×10 <sup>-2</sup>
30	0.67	9.70×10 <sup>-3</sup>	0.59	1.10×10 <sup>-2</sup>	0.62	1.05×10 <sup>-2</sup>
20	0.65	9.99×10 <sup>-3</sup>	0.62	1.05×10 <sup>-2</sup>	0.64	1.02×10 <sup>-2</sup>
10	0.71	9.15×10 <sup>-3</sup>	0.68	9.55×10 <sup>-3</sup>	0.67	9.70×10 <sup>-3</sup>
5	0.78	8.33×10 <sup>-3</sup>	0.73	8.90×10 <sup>-3</sup>	0.77	8.44×10 <sup>-3</sup>
-10	0.93	6.99×10 <sup>-3</sup>	0.88	7.38×10 <sup>-3</sup>	0.95	6.84×10 <sup>-3</sup>
-25	12	5.41×10 <sup>-4</sup>	17.8	3.65×10 <sup>-4</sup>	1834.9	3.54×10 <sup>-6</sup>

**Table S3 – Best performances of aqueous DSSCs at different CMC levels corresponding to data points in Fig. 4.**

CMC (wt%)	Sample		Day		$V_{oc}$ (V)		$J_{sc}$ (mA cm <sup>-2</sup> )		FF (%)		PCE (%)	
	Na	K	Na	K	Na	K	Na	K	Na	K	Na	K
3.5	Na35B	K35A	6	6	0.356	0.474	0.53	2.28	51	62	0.1	0.67
4.5	Na45A	K45A	6	0	0.439	0.456	1.98	2.21	60	63	0.52	0.63
5.5	Na55A	K55B	0	0	0.437	0.453	2.33	2.61	60	61	0.61	0.72
6.5		K65A		1		0.467		2.33		61		0.66
7.5	Na75A	K75B	6	0	0.433	0.448	1.76	2.15	59	61	0.45	0.59
0	B2*	K55A**	2	12	0.543	0.488	2.10	2.70	60	62	0.68	0.81
0	B1*	K55B**	2	12	0.536	0.450	1.20	2.78	58	59	0.37	0.74

\* Liquid NaI 4.5 M + I<sub>2</sub> 50 mM

\*\* Liquid KI 5.5 M + I<sub>2</sub> 50 mM

**Table S4 – Evolution of photovoltaic parameters of aqueous DSSCs sensitized with D131:CDCA = 1:1.18 and filled with NaI 4.50 M, I<sub>2</sub> 50 mM and CMC 3.5 wt% (cell Na35A) or KI 5.50 M, I<sub>2</sub> 50 mM and CMC 3.5 wt% (cell K35B) stored at RT under dark condition.**

Na35A						K35B					
Time (d)	Run	V <sub>oc</sub> (mV)	J <sub>sc</sub> (mA cm <sup>-2</sup> )	FF (%)	PCE (%)	Time (d)	Run	V <sub>oc</sub> (mV)	J <sub>sc</sub> (mA cm <sup>-2</sup> )	FF (%)	PCE (%)
0	1	363	0.45	51	0.08	0	1	314	0.32	42	0.04
	2	356	0.44	50	0.08		2	313	0.30	44	0.04
	3	353	0.45	50	0.08		3	313	0.30	44	0.04
6	1	364	0.59	48	0.10	6	1	358	0.52	46	0.09
	2	345	0.59	44	0.09		2	356	0.56	47	0.09
	3	337	0.58	41	0.08		3	357	0.57	48	0.10
13	1	394	0.73	56	0.16	13	1	376	0.80	47	0.14
	2	385	0.74	56	0.16		2	369	0.81	47	0.14
	3	382	0.75	56	0.16		3	370	0.82	48	0.14
20	1	390	0.76	56	0.17	20	1	390	1.04	49	0.20
	2	386	0.77	56	0.17		2	391	1.05	50	0.20
	3	382	0.78	56	0.17		3	391	1.05	50	0.21
29	1	388	0.80	57	0.18	28	1	428	1.30	60	0.33
	2	383	0.81	57	0.18		2	432	1.29	60	0.33
	3	376	0.82	56	0.17		3	432	1.27	59	0.32

**Table S5 – Evolution of photovoltaic parameters of aqueous DSSCs sensitized with D131:CDCA = 1:1.18 and filled with NaI 4.50 M, I<sub>2</sub> 50 mM and CMC 3.5 wt% (cell Na35B) or KI 5.50 M, I<sub>2</sub> 50 mM and CMC 3.5 wt% (cell K35A) stored at 60 °C under dark conditions after 6 days of storage at RT.**

Na35B							K35A						
Time (d)	Run	V <sub>oc</sub> (mV)	J <sub>sc</sub> (mA cm <sup>-2</sup> )	FF (%)	PCE (%)	Time @60 °C (d)	Time (d)	Run	V <sub>oc</sub> (mV)	J <sub>sc</sub> (mA cm <sup>-2</sup> )	FF (%)	PCE (%)	Time @60 °C (d)
0	1	348	0.40	49	0.07	0	0	1	429	1.19	62	0.32	0
	2	341	0.40	48	0.07			2	426	1.18	62	0.31	
	3	340	0.41	48	0.07			3	423	1.19	62	0.31	
6	1	369	0.53	51	0.10	0	1	1	440	1.14	63	0.32	0
	2	360	0.53	51	0.10			2	436	1.13	63	0.31	
	3	356	0.53	51	0.10			3	432	1.12	63	0.30	
7	1*	305	1.11	43	0.15	1	6	1	481	2.28	62	0.68	0
	2*	324	1.12	44	0.16			2					
	3*	333	1.13	44	0.17			3	474	2.28	62	0.67	
8	1	393	1.12	47	0.21	1	7	1*	383	1.57	57	0.34	1
	2	389	1.13	47	0.21			2*	401	1.64	59	0.39	
	3	386	1.13	46	0.20			3*	409	1.68	59	0.41	
13	1*	263	0.35	46	0.04	6	8	1	475	1.90	63	0.57	1
	2*	282	0.36	48	0.05			2	472	1.89	63	0.56	
	3*	289	0.37	49	0.05			3	468	1.89	63	0.55	
14	1	399	1.35	51	0.27	6	13	1*	379	1.28	57	0.28	6
	2	396	1.36	51	0.27			2*	397	1.33	59	0.31	
	3	391	1.37	51	0.27			3*	404	1.35	59	0.32	
20	1*	264	0.29	46	0.04	12	14	1	470	1.54	63	0.45	6
	2*	280	0.30	48	0.04			2	464	1.53	63	0.44	
	3*	287	0.30	48	0.04			3	461	1.51	62	0.43	
21	1	389	1.19	50	0.23	12	20	1*	366	1.03	56	0.21	12
	2	387	1.19	50	0.23			2*	387	1.07	57	0.24	
	3	384	1.19	50	0.23			3*	395	1.08	58	0.25	

<b>29</b>	<b>1</b>	377	1.02	46	0.18	<b>20</b>	<b>21</b>	<b>1</b>	453	1.22	61	0.33	<b>12</b>
	<b>2</b>	375	1.01	46	0.17			<b>2</b>	449	1.18	61	0.32	
	<b>3</b>	373	1.01	46	0.17			<b>3</b>	446	1.16	61	0.31	
							<b>28</b>	<b>1*</b>	371	0.90	54	0.18	<b>19</b>
							<b>2*</b>	386	0.91	55	0.19		
							<b>3*</b>	392	0.91	55	0.20		
							<b>29</b>	<b>1</b>	499	1.79	62	0.55	<b>19</b>
							<b>2</b>	495	1.77	61	0.54		
							<b>3</b>	491	1.74	61	0.52		
							<b>36</b>	<b>1</b>	445	1.04	59	0.27	<b>19</b>
							<b>2</b>	440	1.01	59	0.26		
							<b>3</b>	438	0.99	59	0.25		

\* measured at ~60 °C

**Table S6 – Evolution of photovoltaic parameters of aqueous DSSCs sensitized with D131:CDCA = 1:1.18 and filled with NaI 4.50 M, I<sub>2</sub> 50 mM and CMC 4.5 wt% (cell Na45A) or KI 5.50 M, I<sub>2</sub> 50 mM and CMC 4.5 wt% (cell K45B) stored at RT under dark conditions.**

Na45A						K45B					
Time (d)	Run	$V_{oc}$ (mV)	$J_{sc}$ (mA cm <sup>-2</sup> )	FF (%)	PCE (%)	Time (d)	Run	$V_{oc}$ (mV)	$J_{sc}$ (mA cm <sup>-2</sup> )	FF (%)	PCE (%)
0	1	427	1.38	56	0.33	0	1	326	0.32	43	0.04
	2	421	1.40	56	0.33		2	328	0.33	44	0.05
	3	418	1.42	55	0.33		3	333	0.35	46	0.05
6	1	443	1.92	59	0.51	6	1	394	0.72	49	0.14
	2	442	1.96	60	0.52		2	389	0.77	50	0.15
	3	439	1.98	60	0.52		3	387	0.79	50	0.15
13	1	440	1.84	59	0.48	13	1	406	1.09	51	0.23
	2	438	1.91	60	0.50		2	405	1.09	52	0.23
	3	434	1.92	60	0.50		3	401	1.07	52	0.23
20	1	441	1.88	60	0.49	20	1	429	1.33	58	0.33
	2	439	1.91	61	0.51		2	428	1.33	59	0.34
	3	435	1.93	61	0.51		3	425	1.33	59	0.33
29	1	436	1.89	60	0.49	28	1	408	1.17	51	0.25
	2	433	1.93	61	0.51		2	405	1.18	52	0.25
	3	431	1.94	61	0.51		3	405	1.19	52	0.25

**Table S7 – Evolution of photovoltaic parameters of aqueous DSSCs sensitized with D131:CDCA = 1:1.18 and filled with NaI 4.50 M, I<sub>2</sub> 50 mM and CMC 4.5 wt% (cell Na45B) or KI 5.50 M, I<sub>2</sub> 50 mM and CMC 4.5 wt% (cell K45A) stored at 60 °C under dark conditions after 6 days of storage at RT.**

Na45B							K45A						
Time (d)	Run	V <sub>oc</sub> (mV)	J <sub>sc</sub> (mA cm <sup>-2</sup> )	FF (%)	PCE (%)	Time @60 °C (d)	Time (d)	Run	V <sub>oc</sub> (mV)	J <sub>sc</sub> (mA cm <sup>-2</sup> )	FF (%)	PCE (%)	Time @60 °C (d)
0	1	409	1.28	57	0.30	0	0	1	465	2.23	63	0.65	0
	2	404	1.30	56	0.29			2	460	2.22	63	0.64	
	3	400	1.30	55	0.29			3	456	2.21	63	0.63	
6	1	421	1.68	60	0.42	0	1	1	443	1.13	63	0.32	0
	2	419	1.71	60	0.43			2	439	1.13	63	0.31	
	3	416	1.72	60	0.43			3	437	1.13	63	0.31	
7	1*	353	2.22	58	0.45	1	6	1	475	1.97	63	0.59	0
	2*	369	2.29	59	0.50			2	469	1.97	63	0.58	
	3*	379	2.34	60	0.53			3	390	1.26	58	0.29	
8	1	435	2.48	62	0.67	1	7	1*	405	1.32	60	0.32	1
	2	431	2.48	62	0.66			2*	412	1.35	61	0.34	
	3	428	2.47	62	0.65			3*	473	1.52	63	0.46	
13	1*	362	1.63	57	0.33	6	8	1	470	1.51	64	0.45	1
	2*	371	1.67	57	0.35			2	465	1.51	63	0.44	
	3*	376	1.69	58	0.37			3	386	1.06	58	0.24	
14	1	426	1.74	60	0.45	6	13	1*	399	1.09	59	0.26	6
	2	421	1.74	60	0.44			2*	407	1.11	60	0.27	
	3	417	1.73	60	0.43			3*	472	1.25	64	0.38	
20	1*	355	1.32	55	0.26	12	14	1	469	1.23	63	0.37	6
	2*	362	1.34	56	0.27			2	465	1.23	63	0.36	
	3*	367	1.35	56	0.28			3	381	0.88	57	0.19	
21	1	421	1.45	58	0.36	12	20	1*	395	0.89	58	0.20	12
	2	415	1.43	58	0.34			2*	401	0.90	58	0.21	
	3	410	1.41	58	0.34			3*	459	1.02	62	0.29	



<b>29</b>	<b>1</b>	409	1.21	56	0.28	<b>20</b>	<b>21</b>	<b>1</b>	455	0.99	62	0.28	<b>12</b>
	<b>2</b>	404	1.19	55	0.27			<b>2</b>	451	0.97	62	0.27	
	<b>3</b>	400	1.18	55	0.26			<b>3</b>	384	0.80	57	0.17	
							<b>28</b>	<b>1*</b>	397	0.81	57	0.18	<b>19</b>
							<b>2*</b>	403	0.81	58	0.19		
							<b>3*</b>	451	0.90	60	0.24		
							<b>29</b>	<b>1</b>	450	0.88	60	0.24	<b>19</b>
							<b>2</b>	447	0.87	60	0.23		
							<b>3</b>	451	0.88	60	0.24		
							<b>36</b>	<b>1</b>	449	0.86	60	0.23	<b>19</b>
							<b>2</b>	446	0.85	60	0.23		
							<b>3</b>	469	1.23	63	0.37		

\* measured at ~60 °C

**Table S8 – Evolution of photovoltaic parameters of aqueous DSSCs sensitized with D131:CDCA = 1:1.18 and filled with NaI 4.50 M, I<sub>2</sub> 50 mM and CMC 5.5 wt% (cell Na55A) or KI 5.50 M, I<sub>2</sub> 50 mM and CMC 5.5 wt% (cell K45B) stored at RT under dark conditions.**

Na55A						K45B					
Time (d)	Run	V <sub>oc</sub> (mV)	J <sub>sc</sub> (mA cm <sup>-2</sup> )	FF (%)	PCE (%)	Time (d)	Run	V <sub>oc</sub> (mV)	J <sub>sc</sub> (mA cm <sup>-2</sup> )	FF (%)	PCE (%)
0	1	443	2.30	59	0.60	0	1	461	2.60	60	0.73
	2	440	2.32	60	0.61		2	458	2.61	61	0.72
	3	437	2.33	60	0.61		3	453	2.61	61	0.72
6	1	442	2.17	60	0.58	1	1	458	2.31	60	0.63
	2	441	2.19	61	0.59		2	456	2.32	61	0.64
	3	438	2.20	61	0.59		3	453	2.34	61	0.64
13	1	430	1.58	59	0.40	6	1	449	1.87	59	0.50
	2	427	1.61	59	0.41		2	448	1.90	60	0.51
	3	424	1.63	59	0.41		3	446	1.91	61	0.52
20	1	438	2.04	61	0.54	13	1	438	1.64	59	0.42
	2	436	2.06	61	0.55		2	438	1.68	60	0.44
	3	432	2.08	61	0.55		3	436	1.70	60	0.45
29	1	441	2.13	61	0.58	20	1	430	1.48	58	0.37
	2	435	2.14	61	0.57		2	429	1.50	59	0.38
	3	431	2.14	62	0.57		3	427	1.52	60	0.39
						28	1	431	1.39	58	0.35
							2	431	1.42	59	0.36
							3	428	1.44	60	0.37
						36	1	428	1.36	58	0.34
							2	429	1.39	59	0.35
							3	427	1.40	60	0.36

**Table S9 – Evolution of photovoltaic parameters of aqueous DSSCs sensitized with D131:CDCA = 1:1.18 and filled with NaI 4.50 M, I<sub>2</sub> 50 mM and CMC 5.5 wt% (cell Na55B) or KI 5.50 M, I<sub>2</sub> 50 mM and CMC 5.5 wt% (cell K55A) stored at 60 °C under dark condition after 6 days of storage at ambient temperature.**

Na55B							K55A						
Time (d)	Run	V <sub>oc</sub> (mV)	J <sub>sc</sub> (mA cm <sup>-2</sup> )	FF (%)	PCE (%)	Time @60 °C (d)	Time (d)	Run	V <sub>oc</sub> (mV)	J <sub>sc</sub> (mA cm <sup>-2</sup> )	FF (%)	PCE (%)	Time @60 °C (d)
0	1	445	2.10	58	0.56	0	0	1	452	2.13	57	0.55	0
	2	442	2.12	59	0.55			2	449	2.13	57	0.55	
	3	438	2.14	59	0.55			3	445	2.12	57	0.54	
6	1	439	1.90	59	0.49	0	1	1	448	2.12	57	0.54	0
	2	437	1.92	60	0.50			2	448	2.15	58	0.56	
	3	435	1.93	60	0.50			3	447	2.16	58	0.56	
7	1*	388	2.10	58	0.48	1	6	1	437	1.65	55	0.40	0
	2*	404	2.18	59	0.52			2	438	1.68	57	0.42	
	3*	412	2.22	59	0.54			3	436	1.70	58	0.43	
8	1	455	2.60	62	0.74	1	7	1*	362	1.40	55	0.28	1
	2	451	2.60	62	0.73			2*	380	1.49	58	0.33	
	3	447	2.59	62	0.72			3*	389	1.55	59	0.35	
13	1*	382	1.77	59	0.40	6	8	1	447	1.72	60	0.46	1
	2*	391	1.79	59	0.41			2	446	1.73	61	0.47	
	3*	397	1.81	59	0.42			3	443	1.72	61	0.47	
14	1	450	1.93	62	0.54	6	13	1*	400	1.81	57	0.41	6
	2	445	1.91	61	0.52			2*	416	1.89	59	0.46	
	3	441	1.90	61	0.51			3*	424	1.93	60	0.49	
20	1*	381	1.49	58	0.33	12	14	1	494	2.18	62	0.67	6
	2*	389	1.49	58	0.34			2	490	2.15	62	0.66	
	3*	394	1.50	58	0.34			3	486	2.14	62	0.64	
21	1	372	0.38	59	0.08	12	20	1*	396	1.59	57	0.36	12
	2	368	0.37	59	0.08			2*	415	1.65	58	0.40	
	3	364	0.37	59	0.08			3*	424	1.69	59	0.42	

<b>29</b>	<b>1</b>	434	1.40	56	0.34	<b>20</b>	<b>21</b>	<b>1</b>	494	1.94	61	0.58	<b>12</b>
	<b>2</b>	430	1.36	55	0.32			<b>2</b>	490	1.92	61	0.57	
	<b>3</b>	428	1.35	55	0.32			<b>3</b>	485	1.90	61	0.56	
							<b>28</b>	<b>1*</b>	415	1.55	57	0.36	<b>19</b>
							<b>2*</b>	432	1.60	58	0.40		
							<b>3*</b>	437	1.62	58	0.41		
							<b>29</b>	<b>1</b>	488	1.77	59	0.51	<b>19</b>
							<b>2</b>	487	1.75	59	0.50		
							<b>3</b>	483	1.73	59	0.49		
							<b>36</b>	<b>1</b>	490	1.76	59	0.50	<b>19</b>
							<b>2</b>	488	1.74	58	0.49		
							<b>3</b>	484	1.73	58	0.49		

\* measured at ~60 °C

**Table S10 – Evolution of photovoltaic parameters of aqueous DSSCs sensitized with D131:CDCA = 1:1.18 and filled with KI 5.50 M, I<sub>2</sub> 50 mM and CMC 6.5 wt% (cell K65B) stored at RT under dark conditions.**

<b>K65B</b>					
<b>Time (d)</b>	<b>Run</b>	<b>V<sub>oc</sub> (mV)</b>	<b>J<sub>sc</sub> (mA cm<sup>-2</sup>)</b>	<b>FF (%)</b>	<b>PCE (%)</b>
<b>0</b>	<b>1</b>	461	2.08	57	0.55
	<b>2</b>	452	2.09	56	0.53
	<b>3</b>	447	2.09	56	0.53
<b>1</b>	<b>1</b>	470	2.17	60	0.61
	<b>2</b>	464	2.16	60	0.60
	<b>3</b>	459	2.16	60	0.59
<b>6</b>	<b>1</b>	464	2.02	60	0.56
	<b>2</b>	462	2.04	60	0.57
	<b>3</b>	459	2.04	60	0.57
<b>13</b>	<b>1</b>	469	1.93	60	0.55
	<b>2</b>	466	1.94	61	0.55
	<b>3</b>	462	1.95	61	0.55
<b>20</b>	<b>1</b>	467	1.86	60	0.52
	<b>2</b>	463	1.87	61	0.52
	<b>3</b>	459	1.87	60	0.52
<b>28</b>	<b>1</b>	466	1.80	60	0.50
	<b>2</b>	464	1.82	61	0.51
	<b>3</b>	461	1.82	61	0.51
<b>36</b>	<b>1</b>	471	1.76	60	0.50
	<b>2</b>	466	1.77	61	0.50
	<b>3</b>	462	1.77	61	0.50

**Table S11 – Evolution of photovoltaic parameters of aqueous DSSCs sensitized with D131:CDCA = 1:1.18 and filled with KI 5.50 M, I<sub>2</sub> 50 mM and CMC 6.5 wt% (cell K65A) stored at 60 °C under dark condition after 6 days of storage at ambient temperature.**

<b>K65A</b>						
<b>Time (d)</b>	<b>Run</b>	<b>V<sub>oc</sub> (mV)</b>	<b>J<sub>sc</sub> (mA cm<sup>-2</sup>)</b>	<b>FF (%)</b>	<b>PCE (%)</b>	<b>Time @60 °C (d)</b>
<b>0</b>	<b>1</b>	470	2.13	60	0.60	<b>0</b>
	<b>2</b>	457	2.11	59	0.57	
	<b>3</b>	451	2.11	59	0.56	
<b>1</b>	<b>1</b>	475	2.34	61	0.67	<b>0</b>
	<b>2</b>	470	2.33	61	0.67	
	<b>3</b>	467	2.33	61	0.66	
<b>6</b>	<b>1</b>	475	2.16	61	0.63	<b>0</b>
	<b>2</b>	472	2.16	61	0.63	
	<b>3</b>	469	2.17	61	0.62	
<b>7</b>	<b>1*</b>	401	1.83	59	0.43	<b>1</b>
	<b>2*</b>	414	1.90	61	0.48	
	<b>3*</b>	421	1.95	61	0.50	
<b>8</b>	<b>1</b>	413	1.85	59	0.45	<b>1</b>
	<b>2</b>	426	1.90	60	0.48	
	<b>3</b>	434	1.94	61	0.51	
<b>13</b>	<b>1*</b>	481	2.14	63	0.65	<b>6</b>
	<b>2*</b>	478	2.14	64	0.65	
	<b>3*</b>	474	2.13	63	0.64	
<b>14</b>	<b>1</b>	505	2.18	64	0.70	<b>6</b>
	<b>2</b>	498	2.16	64	0.69	
	<b>3</b>	495	2.15	64	0.68	
<b>20</b>	<b>1*</b>	429	1.68	60	0.43	<b>12</b>
	<b>2*</b>	440	1.71	60	0.45	
	<b>3*</b>	444	1.73	60	0.46	
<b>21</b>	<b>1</b>	499	1.94	63	0.61	<b>12</b>
	<b>2</b>	495	1.91	62	0.59	
	<b>3</b>	488	1.90	62	0.58	
<b>28</b>	<b>1*</b>	420	1.56	59	0.39	<b>19</b>
	<b>2*</b>	420	1.56	59	0.39	
	<b>3*</b>	439	1.61	60	0.42	
<b>29</b>	<b>1</b>	487	1.76	61	0.53	<b>19</b>
	<b>2</b>	484	1.73	61	0.51	
	<b>3</b>	481	1.72	61	0.50	
<b>36</b>	<b>1</b>	495	1.74	62	0.53	<b>19</b>
	<b>2</b>	491	1.71	61	0.51	
	<b>3</b>	486	1.69	61	0.50	

\* measured at ~60 °C

**Table S12 – Evolution of photovoltaic parameters of aqueous DSSCs sensitized with D131:CDCA = 1:1.18 and filled with NaI 4.50 M, I<sub>2</sub> 50 mM and CMC 7.5 wt% (cell Na75A) or KI 5.50 M, I<sub>2</sub> 50 mM and CMC 7.5 wt% (cell K75B) stored at ambient temperature under dark condition.**

Na75A						K75B					
Time (d)	Run	$V_{oc}$ (mV)	$J_{sc}$ (mA cm <sup>-2</sup> )	FF (%)	PCE (%)	Time (d)	Run	$V_{oc}$ (mV)	$J_{sc}$ (mA cm <sup>-2</sup> )	FF (%)	PCE (%)
0	1	420	1.52	53	0.34	0	1	456	2.13	61	0.59
	2	420	1.54	53	0.34		2	452	2.14	61	0.59
	3	419	1.55	54	0.35		3	448	2.15	61	0.59
6	1	437	1.73	59	0.44	1	1	459	1.88	61	0.53
	2	438	1.75	59	0.45		2	455	1.89	61	0.53
	3	433	1.76	59	0.45		3	451	1.89	61	0.52
13	1	439	2.07	60	0.55	6	1	437	1.51	59	0.39
	2	437	2.10	61	0.56		2	437	1.54	60	0.41
	3	434	2.12	61	0.56		3	435	1.55	61	0.41
20	1	422	1.54	59	0.39	13	1	440	1.42	59	0.37
	2	418	1.54	60	0.39		2	439	1.45	60	0.38
	3	416	1.57	59	0.39		3	436	1.46	61	0.39
29	1	426	1.55	59	0.39	20	1	438	1.36	59	0.35
	2	421	1.57	60	0.40		2	436	1.40	60	0.37
	3	417	1.58	60	0.39		3	432	1.40	61	0.37
						28	1	437	1.30	59	0.34
							2	436	1.32	60	0.35
							3	435	1.33	61	0.35
						36	1	438	1.25	59	0.33
							2	437	1.28	60	0.34
							3	434	1.30	60	0.34

**Table S13 – Evolution of photovoltaic parameters of aqueous DSSCs sensitized with D131:CDCA = 1:1.18 and filled with NaI 4.50 M, I<sub>2</sub> 50 mM and CMC 7.5 wt% (cell Na75B) or KI 5.50 M, I<sub>2</sub> 50 mM and CMC 7.5 wt% (cell K75A) stored at 60 °C under dark condition after 6 days of storage at ambient temperature.**

Na75B							K75A						
Time (d)	Run	V <sub>oc</sub> (mV)	J <sub>sc</sub> (mA cm <sup>-2</sup> )	FF (%)	PCE (%)	Time @60 °C (d)	Time (d)	Run	V <sub>oc</sub> (mV)	J <sub>sc</sub> (mA cm <sup>-2</sup> )	FF (%)	PCE (%)	Time @60 °C (d)
0	1	381	0.58	52	0.11	0	0	1	443	2.090	56	0.52	0
	2	376	0.60	52	0.12			2	418	0.885	57	0.21	
	3	376	0.62	51	0.12			3	414	0.889	57	0.21	
6	1	430	1.52	59	0.38	0	6	1	421	0.647	60	0.16	0
	2	429	1.54	58	0.39			2	420	0.655	60	0.17	
	3	427	1.55	58	0.39			3	417	0.661	60	0.17	
7	1*	388	2.10	58	0.48	1	7	1*	389	1.28	57	0.29	1
	2*	404	2.18	59	0.52			2*	401	1.34	59	0.32	
	3*	412	2.22	59	0.54			3*	409	1.38	59	0.34	
8	1	470	2.44	62	0.71	1	8	1	392	1.18	59	0.27	1
	2	467	2.44	62	0.71			2	403	1.22	60	0.29	
	3	463	2.44	62	0.70			3	409	1.24	60	0.31	
13	1*	418	2.03	59	0.50	6	13	1*	473	1.56	63	0.46	6
	2*	426	2.06	59	0.52			2*	469	1.56	63	0.46	
	3*	429	2.08	59	0.53			3*	465	1.56	62	0.45	
14	1	475	2.22	61	0.64	6	14	1	477	1.42	63	0.42	6
	2	472	2.21	61	0.63			2	473	1.40	63	0.41	
	3	469	2.20	60	0.62			3	469	1.38	63	0.40	
20	1*	400	1.74	56	0.39	12	20	1*	390	1.04	58	0.24	12
	2*	408	1.76	57	0.41			2*	403	1.07	59	0.25	
	3*	413	1.78	57	0.42			3*	410	1.08	59	0.26	
21	1	461	1.91	57	0.50	12	21	1	466	1.21	61	0.35	12
	2	459	1.89	57	0.50			2	461	1.16	61	0.33	
	3	456	1.88	57	0.49			3	456	1.15	61	0.32	



<b>29</b>	<b>1</b>	448	1.66	49	0.37	<b>19</b>	<b>28</b>	<b>1*</b>	403	0.93	57	0.21	<b>19</b>
	<b>2</b>	445	1.65	50	0.37			<b>2*</b>	410	0.93	58	0.22	
	<b>3</b>	443	1.64	51	0.37			<b>3*</b>	415	0.93	58	0.23	
							<b>29</b>	<b>1</b>	458	1.05	59	0.28	<b>19</b>
						<b>2</b>		453	1.02	59	0.27		
						<b>3</b>		449	1.00	59	0.26		
							<b>36</b>	<b>1</b>	458	1.04	59	0.28	<b>19</b>
						<b>2</b>		453	1.02	59	0.27		
						<b>3</b>		449	0.99	59	0.26		

\* measured at ~60 °C

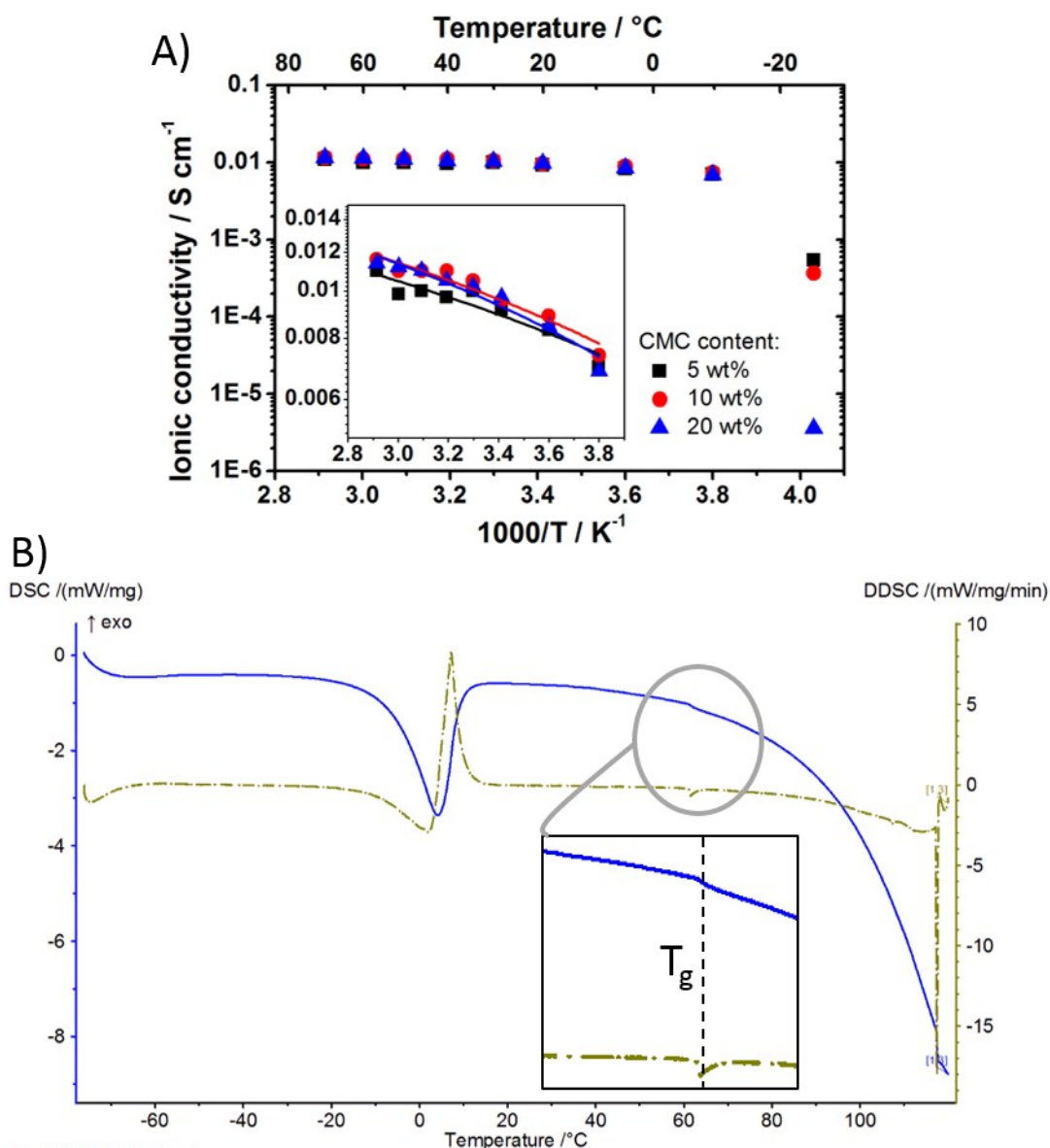
**Table S14 – List of the weak aspects of 100% aqueous electrolytes present in the literature with respect to the hydrogel proposed in this work.**

Components	$J_{sc}$ (mA cm <sup>-2</sup> )	$V_{oc}$ (V)	FF	PCE (%)	PCE loss (%)	Ref.	Weak points with respect to this work
NaI 0.10 M and I <sub>2</sub> 10 mM	1.92	0.498	0.55	0.529	-	[1]	Not stable. Less efficient.
LiI 0.10 M, I <sub>2</sub> 10 mM, 0.5 wt% κ-carrageenan and HNO <sub>3</sub> (pH 2)	2.69	0.442	0.48	0.586	-	[2]	Not stable. Less efficient. Strongly acidic.
KI 0.50 M and I <sub>2</sub> 25 mM	2.14	0.44	0.64	0.6	-	[3]	Not stable. Less efficient.
KI 0.50 M and I <sub>2</sub> 25 mM	3.61	0.47	0.65	1.1	50% (75 d, dark, RT)	[3]	Not stable.
PMII 2.0 M, I <sub>2</sub> 50 mM, GuSCN 0.10 M, TBP 0.50 M and Triton X-100 1%	2.28	0.66	0.79	1.16	-	[4]	Not stable. It contains petroleum-derived and toxic additives. Based on our experience, TBP is not soluble in H <sub>2</sub> O [5]
KI 4.0 M, I <sub>2</sub> 20 mM, HNO <sub>3</sub> (pH 4) and BMPP (saturated)	3.97	0.481	0.68	1.34	-	[6]	Not stable.
[Co(bpy) <sub>3</sub> ]Cl <sub>2</sub> 0.13 M, [Co(bpy) <sub>3</sub> ]Cl <sub>3</sub> 40 mM, NMBI 0.80 M	3.4	0.63	0.68	1.4	25% (200 h, 1 sun, RT)	[7]	Less stable. It contains heavy metals and petroleum-derived/toxic additives.
Acrylic acid : Gelatin : (NH <sub>4</sub> ) <sub>2</sub> S <sub>2</sub> O <sub>8</sub> = 10:1:0.08 polymerized at 80 °C, then swollen in LiI 0.50 M and I <sub>2</sub> 50 mM	4.3	0.59	0.71	1.80	20% (2 h, dark)	[8]	Less stable. It requires a thermal treatment.
NaI 2.0 M, I <sub>2</sub> 20 mM, GuSCN 0.50 M and CDCA until saturation	4.91	0.62	0.64	1.88	-	[9]	Not stable.
TEMPOL 1.0 M and NaBF <sub>4</sub> 1.0 M	4.5	0.69	0.64	2.1	-	[10]	Not stable.
PMII 2.0 M, I <sub>2</sub> 50 mM, GuSCN 0.10 M, TBP 0.50 M	3.78	0.68	0.82	2.1	-	[4]	Not stable. It contains petroleum-derived and toxic additives. Based on our

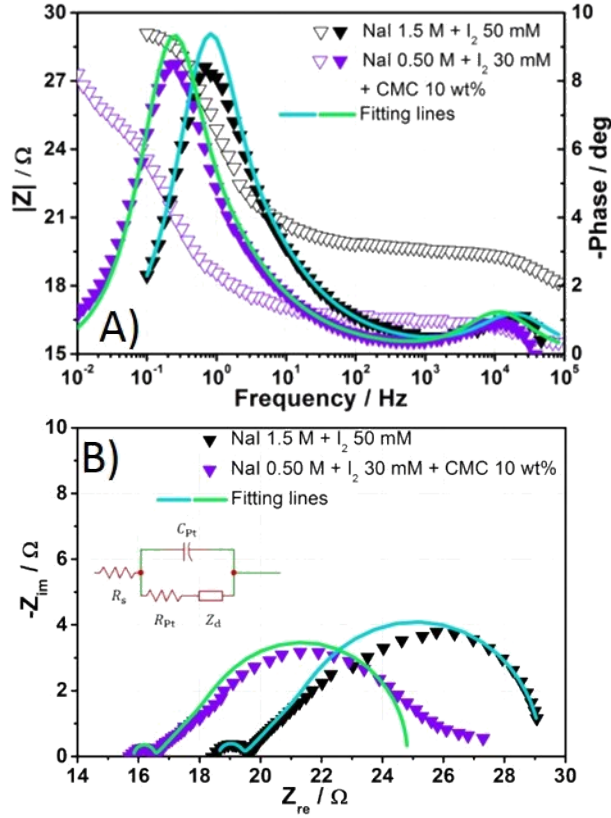
and Triton X-100 1%							experience, TBP is not soluble in H <sub>2</sub> O [5].
KI 4.0 M, I <sub>2</sub> 20 mM and CDCA until saturation	4.86	0.600	0.76	2.20	-	[11]	Not stable.
PMII 2.0 M, I <sub>2</sub> 50 mM, GuSCN 0.10 M, TBP 0.50 M and Triton X-100 1%	4.7	0.74	0.69	2.4	-	[12]	Not stable. It contains petroleum-derived and toxic additives. Based on our experience, TBP is not soluble in H <sub>2</sub> O [5].
TT-EMI <sup>+</sup> 0.20 M, DTT 0.20 M, TBP 0.50 M and 1% Triton X-100	7.2	0.65	0.55	2.6	37% (4 h, 1 sun, RT)	[13]	Not stable. It contains petroleum-derived and toxic additives. Based on our experience, TBP is not soluble in H <sub>2</sub> O [5].
NaI 2.0 M, I <sub>2</sub> 20 mM, GuSCN 0.50 M and CDCA until saturation	7.34	0.59	0.63	2.64	-	[9]	Not stable.
KI 2.0 M, I <sub>2</sub> 10 mM and CDCA until saturation	6.85	0.650	0.67	3.01	-	[11]	Not stable.
NaI 2.0 M, I <sub>2</sub> 20 mM, GuSCN 1.0 M, HNO <sub>3</sub> (pH 3) and CDCA until saturation	8.5	0.59	0.63	3.08	-	[9]	Not stable. Strongly acidic.
GuI 8.0 M, I <sub>2</sub> 20 mM, CDCA (sat.) and SiO <sub>2</sub> (5 wt%)	8.20	0.75	0.51	3.13	37% (1200 h, outdoor)	[14]	Lower amount of renewable components. Lack of thermal stability test.
[Co(phen) <sub>3</sub> ]Cl <sub>2</sub> 0.13 M, [Co(phen) <sub>3</sub> ]Cl <sub>3</sub> 40 mM, NMBI 0.80 M	6.3	0.75	0.73	3.4	45% (200 h, 1 sun, RT)	[7]	Less stable. It contains heavy metals and petroleum-derived/toxic additives.
TT-EMI <sup>+</sup> 0.20 M, DTT 0.60 M, TBP 0.50 M and 1% Triton X-100	9.5	0.61	0.59	3.5	-	[13]	Not stable. It contains petroleum-derived and toxic additives.
NaI 2.0 M, I <sub>2</sub> 0.20 M, GuSCN 0.10 M and FC-134 0.2 wt%	10.97	0.53	0.68	3.96	63% (50 d, 1 sun, RT, UV filter)	[15]	Less stable.

GuI 8.0 M, I <sub>2</sub> 20 mM and CDCA until saturation	10.02	0.61	0.67	4.06	-	[9]	Not stable.
K <sub>4</sub> Fe(CN) <sub>6</sub> 0.40 M, K <sub>3</sub> Fe(CN) <sub>6</sub> 40 mM, KCl 0.10 M, Trizma-HCl buffer 50 mM (pH 8) and Tween 20 0.1%	7.2	0.76	0.75	4.1	50% (2 h)	[16]	Not stable.
[Co(bpy) <sub>3</sub> ] <sup>2+</sup> 0.20 M, [Co(bpy) <sub>3</sub> ] <sup>3+</sup> 40 mM, NMBI 0.70 M, PEG 300 1% and Gelatin 15%	7.9	0.70	0.74	4.1	25% (120 d, dark)	[17]	It contains heavy metals and petroleum-derived/toxic additives.
[Co(bpy) <sub>3</sub> ] <sup>2+</sup> 0.20 M, [Co(bpy) <sub>3</sub> ] <sup>3+</sup> 40 mM, NMBI 0.70 M and PEG 300 1%	8.3	0.68	0.72	4.2	10% (90 d, dark)	[18]	It contains heavy metals and petroleum-derived/toxic additives.
TEMPO 0.15 M, TEMPOBF <sub>4</sub> 50 mM, LiClO <sub>4</sub> 0.10 M and NMBI 0.20 M	5.78	0.955	0.75	4.14	-	[19]	Not stable.
[Co(phen) <sub>3</sub> ]Cl <sub>2</sub> 0.13 M, [Co(phen) <sub>3</sub> ]Cl <sub>3</sub> 60 mM, NMBI 0.80 M	8.6	0.75	0.72	4.8	45% (200 h, 1 sun, RT)	[7]	Less stable. It contains heavy metals and petroleum-derived/toxic additives.
TEMPO 0.40 M, NOBF <sub>4</sub> 0.40 M, LiI 0.10 M, I <sub>2</sub> 50 mM, DMPII 0.60 M, GuSCN 0.10 M and Tween 20 0.1%	9.56	0.77	0.67	4.96	-	[20]	Not stable.
[Co(bpy) <sub>3</sub> ] <sup>2+</sup> 0.20 M, [Co(bpy) <sub>3</sub> ] <sup>3+</sup> 40 mM, NMBI 0.70 M NMBI and PEG 300 1%	9.8	0.69	0.74	5	Gain 2% (2 d, dark)	[18]	Less stable. It contains heavy metals and petroleum-derived/toxic additives.
[Co(bpy-pz) <sub>3</sub> ]Cl <sub>2</sub> 0.13 M, [Co(bpy-pz) <sub>3</sub> ]Cl <sub>3</sub> 60 mM, NMBI 0.80 M	81	0.90	0.76	5.5	-	[7]	Not stable. It contains heavy metals and petroleum-derived/toxic additives.
[Co(bpy) <sub>3</sub> ](NO <sub>3</sub> ) <sub>2</sub> 0.20 M, [Co(bpy) <sub>3</sub> ](NO <sub>3</sub> ) <sub>3</sub> 40 mM,	10.17	0.821	0.68	5.64	0% (500 h, dark)	[21]	It contains heavy metals and petroleum-derived/toxic additives.

NMBI 0.70 M and PEG 300 1 wt%							
JC-IL 0.40 M and NOBF <sub>4</sub> 0.40 M	9.87	0.88	0.68	5.97	-	[22]	Not stable.



**Fig. S1. A)** Arrhenius plot showing the ionic conductivity data in the temperature range of  $-25/+70^{\circ}\text{C}$ , for aqueous NaI 0.50 M gels containing different amounts of CMC. The corresponding  $R^2$  values are 0.87 (5 wt%), 0.93 (10 wt%) and 0.95 (20 wt%).; **B)** Differential scanning calorimetry (DSC) trace of the 10 wt% CMC-containing electrolyte: a  $T_g$  value equal to  $+62^{\circ}\text{C}$  emerges. The  $T_g$  is above the standard operating temperature of DSSCs, therefore we collected almost all ionic conductivity data below the  $T_g$ . This means that the VTF behavior cannot be invoked, since it is related to semirandom motion of short polymer segments occurring when electrolytes work in a temperature exceeding the  $T_g$ . Thus, we likely attribute the ionic conductivity vs. temperature trend to a mixed Arrhenius / Non-Arrhenius behaviour.



**Fig. S2. A)** Bode (modulus = open triangles; phase = filled triangles) and **B)** Nyquist plots of a dummy cell filled with aqueous NaI 0.50 M, I<sub>2</sub> 30 mM and CMC 10 wt%. The spectra of a dummy cell filled with aqueous NaI 1.5 M + I<sub>2</sub> 50 mM are shown for comparison. The spectra were fitted according to Eq. 2 and the equivalent circuit is shown in panel B. according to:

$$Z(\omega) = R_s + \frac{1}{\frac{1}{(R_{ct} + Z_d)} + T_{Pt}(i\omega)^{\varphi_{Pt}}} \quad (\text{Eq. S1})$$

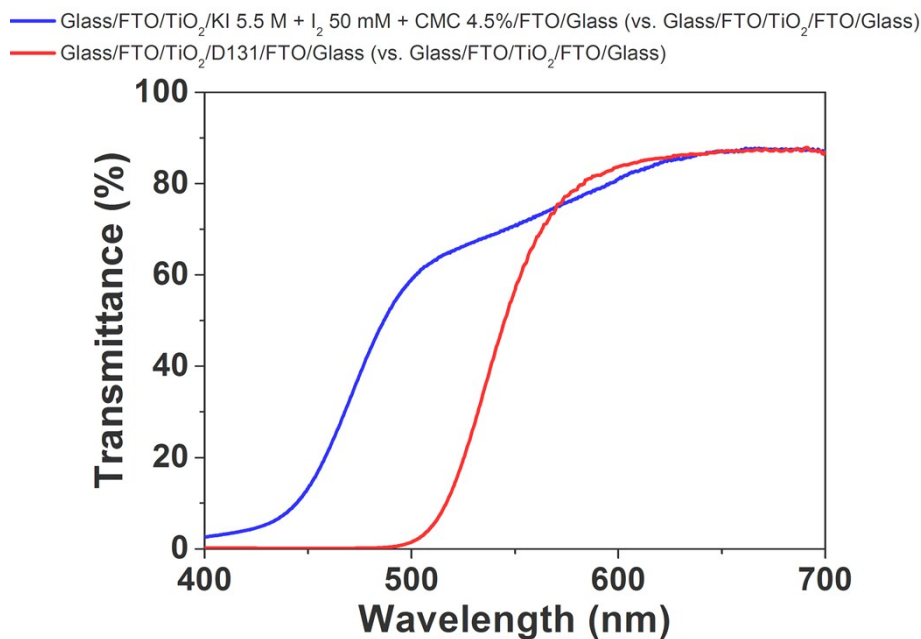
where  $R_s$  is the series resistance,  $R_{ct}$  is the charge transfer resistance at the TiO<sub>2</sub>/electrolyte interface,  $Z_d$  is the impedance due to ionic diffusion,  $\omega$  is the frequency of the applied small-amplitude modulated potential,  $T_{Pt}$  and  $\varphi_{Pt}$  are the parameters of the constant phase element (CPE) used to describe the double layer capacitance at the Pt/electrolyte interface ( $C_{Pt}$ , accounting for the corresponding depressed semicircle in the Nyquist plot). The constraint  $0.90 \leq \varphi_{Pt} \leq 1$  was applied and the double layer capacitance was calculated from the CPE as follows [23]:

$$C_{Pt} = \varphi_{Pt} \sqrt{\frac{T_{Pt}}{R_{Pt}(\varphi_{Pt} - 1)}} \quad (\text{Eq. S2})$$

where  $R_{Pt}$  is the AC resistance to charge transfer at the Pt/electrolyte interface.







**Fig. S3.** Transmittance of: i) The hydrogel electrolyte proposed in this work (blue curve); ii) The metal-free sensitizer chosen for DSSCs (red curve). It emerges the typical absorption in the near-UV region due to the iodine-based redox couple, and the absorption at low wavelength typical of a yellow dye as D131. This allows to obtain solar cells with a high transmittance (>70%) above 550 nm, which accounts for their suitability in building-integrated applications.

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## References

- [1] H. Zhang, Y. Wang, P. Liu, Y. Li, H. G. Yang, T. An, P. K. Wong, D. Wang, Z. Tang and H. Zhao, *Nano Energy*, 2015, **13**, 124-130.
- [2] M. Kaneko, T. Nomura and C. Sasaki, *Macromol. Rapid Commun.*, 2003, **24**, 444-446.
- [3] H. Saito, S. Uegusa, T. N. Murakami, N. Kawashima and T. Miyasaka, *Electrochemistry*, 2004, **72**, 310-316.
- [4] H. Choi, B. S. Jeong, K. Do, M. J. Ju, K. Song and J. Ko, *New J. Chem.*, 2013, **37**, 329-336.
- [5] F. Bella, S. Galliano, M. Falco, G. Viscardi, C. Barolo, M. Grätzel and C. Gerbaldi, *Chem. Sci.*, 2016, **7**, 4880-4890.
- [6] R. Cisneros, M. Beley, F. Lopicque and P. C. Gros, *Eur. J. Inorg. Chem.*, 2016, **2016**, 33-39.
- [7] H. Ellis, R. Jiang, S. Ye, A. Hagfeldt and G. Boschloo, *Phys. Chem. Chem. Phys.*, 2016, **18**, 8419-8427.
- [8] S. Zhang, G. Y. Dong, B. Lin, J. Qu, N. Y. Yuan, J. N. Ding and Z. Gu, *Sol. Energy*, 2016, **127**, 19-27.
- [9] C. Law, O. Moudam, S. Villarroja-Lidon and B. C. O'Regan, *J. Mater. Chem.*, 2012, **22**, 23387-23394.
- [10] R. Kato, F. Kato, K. Oyaizu and H. Nishide, *Chem. Lett.*, 2014, **43**, 480-482.
- [11] V. Leandri, H. Ellis, E. Gabrielsson, L. Sun, G. Boschloo and A. Hagfeldt, *Phys. Chem. Chem. Phys.*, 2014, **16**, 19964-19971.
- [12] C. H. Law, S. C. Pathirana, X. Li, A. Y. Anderson, P. R. F. Barnes, A. Listorti, T. H. Ghaddar and B. O'Regan, *Adv. Mater.*, 2010, **22**, 4505-4509.
- [13] H. Tian, E. Gabrielsson, P. W. Lohse, N. Vlachopoulos, L. Kloo, A. Hagfeldt and L. Sun, *Energy Environ. Sci.*, 2012, **5**, 9752-9755.
- [14] O. Moudam and A. El Gamouz, *Org. Electron.*, 2016, **36**, 7-11.
- [15] H. Zhang, L. Qiu, D. Xu, W. Zhang and F. Yan, *J. Mater. Chem. A*, 2014, **2**, 2221-2226.
- [16] T. Daeneke, Y. Uemura, N. W. Duffy, A. J. Mozer, N. Koumura, U. Bach and L. Spiccia, *Adv. Mater.*, 2012, **24**, 1222-1225.
- [17] W. Xiang, D. Chen, R. A. Caruso, Y. B. Cheng, U. Bach and L. Spiccia, *ChemSusChem*, 2015, **8**, 3704-3711.
- [18] W. Xiang, F. Huang, Y. B. Cheng, U. Bach and L. Spiccia, *Energy Environ. Sci.*, 2013,

---

6, 121-127.

[19] W. Yang, M. Söderberg, A. I. K. Eriksson and G. Boschloo, *RSC Adv.*, 2015, **5**, 26706-26709.

[20] R. Y. Y. Lin, T. M. Chuang, F. L. Wu, P. Y. Chen, T. C. Chu, J. S. Ni, M. S. Fan, Y. H. Lo, K. C. Ho and J. T. Lin, *ChemSusChem*, 2015, **8**, 105-113.

[21] C. Dong, W. Xiang, F. Huang, D. Fu, W. Huang, U. Bach, Y. B. Cheng, X. Li and L. Spiccia, *Angew. Chem. Int. Ed.*, 2014, **53**, 6933-6937.

[22] R. Y. Y. Lin, F. L. Wu, C. T. Li, P. Y. Chen, K. C. Ho and J. T. Lin, *ChemSusChem*, 2015, **8**, 2503-2513.

[23] J. Macdonald and E. Barsoukov, *Impedance spectroscopy: theory experiment and applications. 2<sup>nd</sup> edition*, Hoboken (New Jersey): John Wiley & Sons, Inc; 2005.