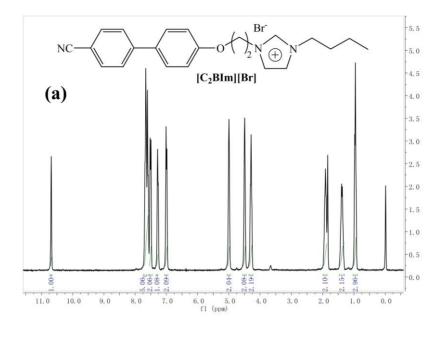
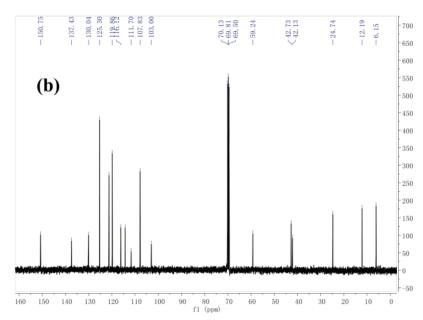
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## High Performance All-Solid-State Dye-Sensitized Solar Cells Based on Cyanobiphenyl-Functionalized Imidazolium-Type Ionic Crystals

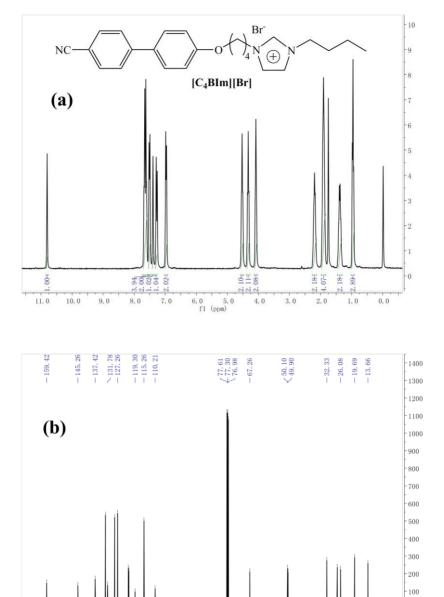
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**Figure S1.** Full (a)  $^{1}$ H- and (b)  $^{13}$ C-NMR spectra for [C<sub>2</sub>BIm][Br].



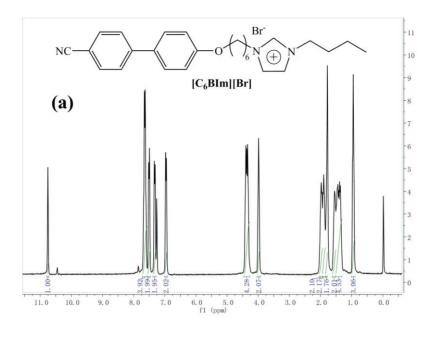
**Figure S2.** Full (a)  $^{1}$ H- and (b)  $^{13}$ C-NMR spectra for [C<sub>4</sub>BIm][Br].

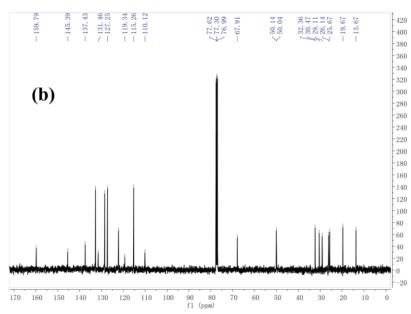
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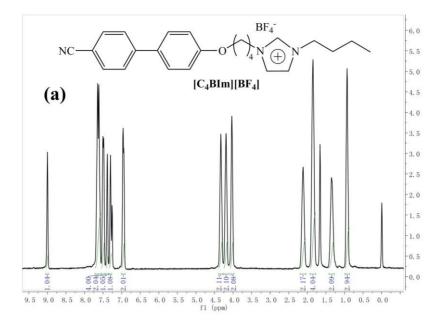
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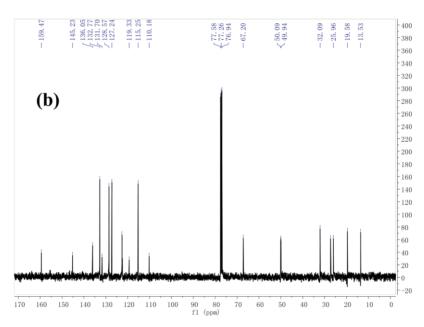
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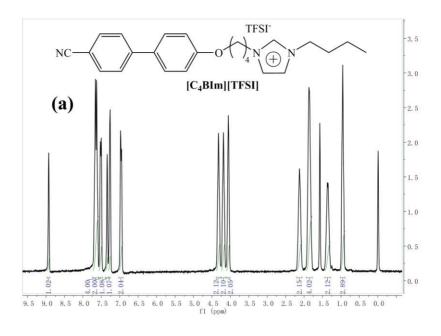


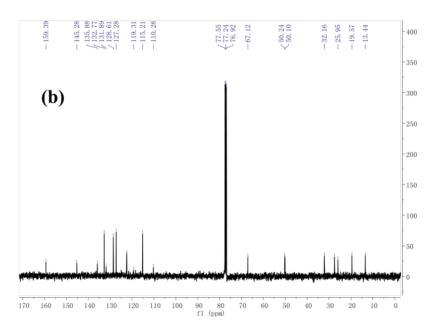
**Figure S3.** Full (a)  $^{1}$ H- and (b)  $^{13}$ C-NMR spectra for [C<sub>6</sub>BIm][Br].



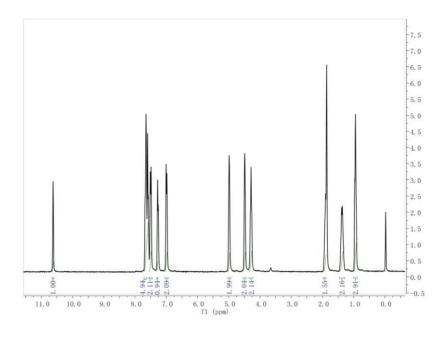


**Figure S4.** Full (a)  $^{1}$ H- and (b)  $^{13}$ C-NMR spectra for [C<sub>4</sub>BIm][BF<sub>4</sub>].





**Figure S5.** Full (a) <sup>1</sup>H- and (b) <sup>13</sup>C-NMR spectra for [C<sub>4</sub>BIm][TFSI].



**Figure S6.** Full  $^{1}$ H-NMR spectra for [C<sub>2</sub>BIm][Br] after be heated at 150  $^{\circ}$ C for 0.5 h under a N<sub>2</sub> atmosphere.

**Table S1.** Device performances of DSSCs with different electrolytes at 55 °C under different sun light intensity illumination.

Device	Electrolytes <sup>[a]</sup>	J <sub>sc</sub> [mA/cm <sup>2</sup> ]	V <sub>oc</sub> [V]	FF	PCE [%]	Light Intensity [mW/cm <sup>2</sup> ]
I	EMII/I <sub>2</sub>	10.89	0.575	0.626	3.92	100
		6.35	0.557	0.690	4.88	50
		1.69	0.527	0.717	4.26	15
II	EMII/I <sub>2</sub> /[C <sub>2</sub> BIm][Br]	13.88	0.536	0.581	4.33	100
		8.20	0.502	0.623	5.13	50
		2.08	0.467	0.681	4.41	15
III	EMII/I <sub>2</sub> /[C <sub>4</sub> BIm][Br]	14.57	0.568	0.617	5.11	100
		8.52	0.554	0.679	6.41	50
		2.22	0.522	0.711	5.49	15
IV	$EMII/I_2/[C_6BIm][Br]$	14.41	0.561	0.614	4.96	100
		8.45	0.546	0.668	6.16	50
		2.20	0.519	0.705	5.37	15
V	$EMII/I_2/[C_4BIm][BF_4]$	14.33	0.542	0.592	4.60	100
		8.39	0.534	0.645	5.78	50
		2.18	0.495	0.702	5.05	15
VI	EMII/I <sub>2</sub> /[C <sub>4</sub> BIm][TFSI]	14.08	0.540	0.588	4.47	100
		8.27	0.522	0.636	5.49	50
		2.10	0.489	0.691	4.73	15

 $<sup>^{\</sup>rm a}$  20 wt% [C\_nBIm][X] doped EMII electrolytes containing 0.1 M  $I_2.$