

## Supporting Information

### **Solution Processable Carbazole-Benzophenone Derivatives Bipolar Hosts Enabling Highly-efficient Green TADF Organic LEDs**

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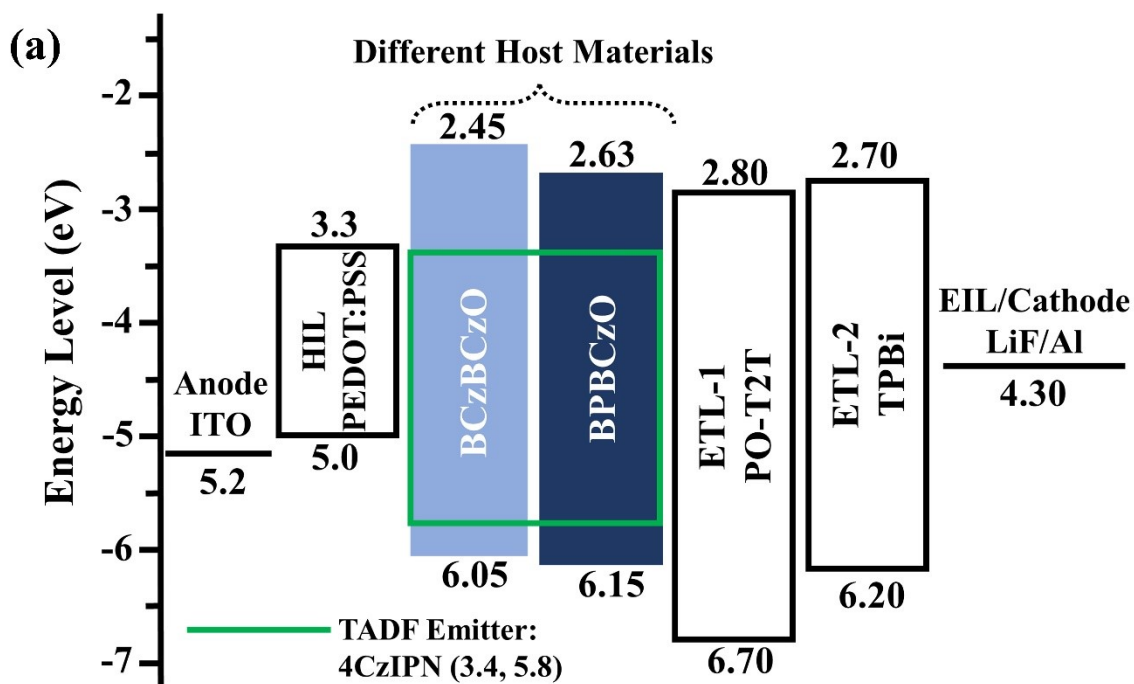


Figure S1. Energy-level diagrams of green TADF organic LEDs with newly synthesized carbazole-benzophenone derivatives, i.e. **BCzBCzO** and **BPBCzO** hosts.

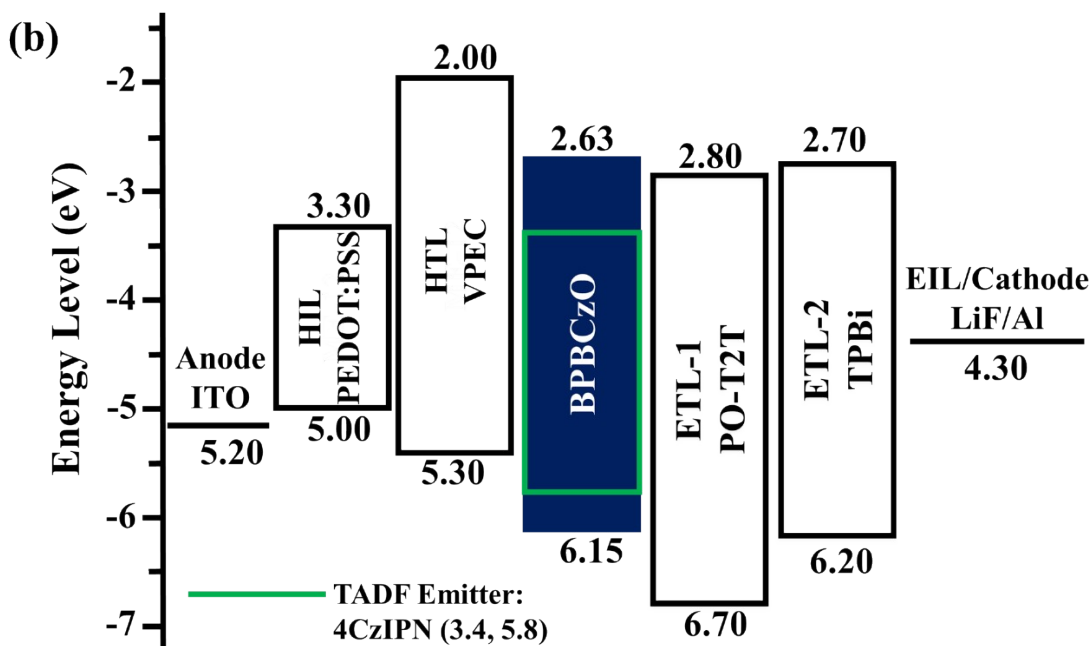


Figure S2. Energy-level diagrams of green organic LEDs (a) without and (b) with crosslinkable hole-transporting materials, i.e. VPEC, and newly synthesized carbazole-benzophenone derivatives, i.e. **BPBCzO** host.

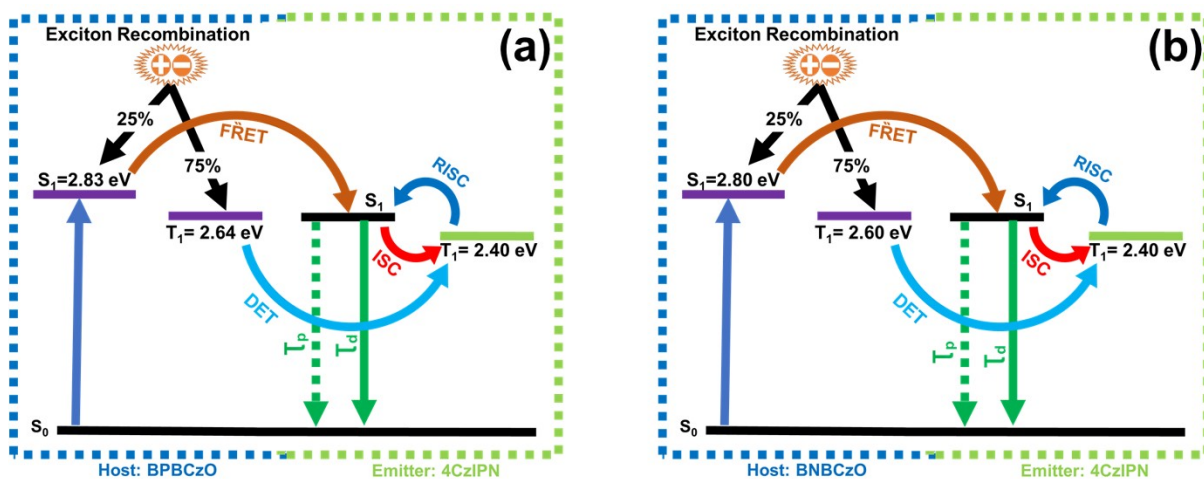


Figure S3. (a-b) Energy-transfer routes from host-to-guest in **BPBCzO** and **BCzBCzO**.

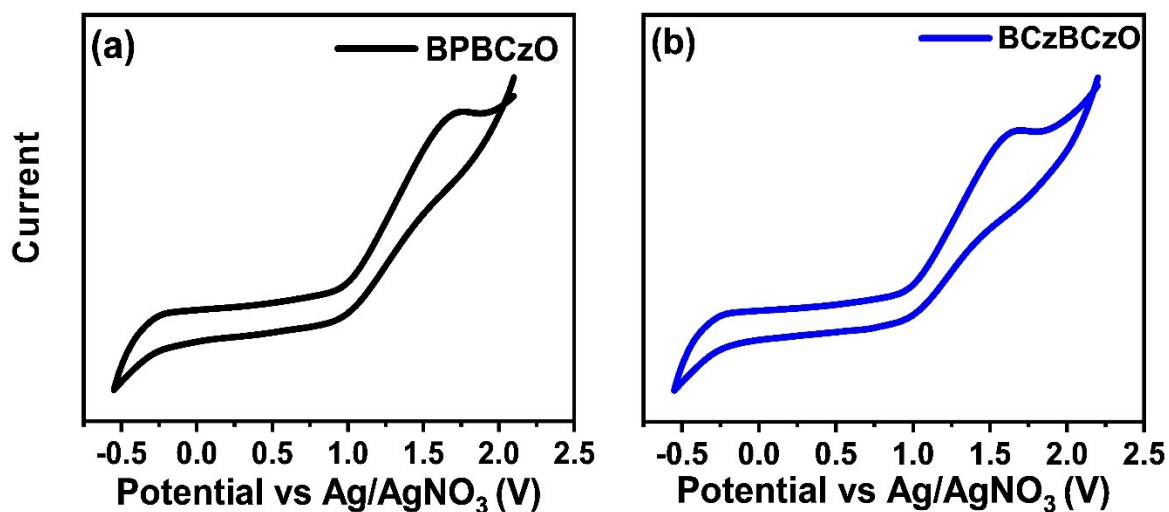


Figure S4. Cyclic voltammograms of newly synthesized carbazole-benzophenone derivatives, (a) **BPBCzO** and (c) **BCzBCzO**, were measured in DCM solution.

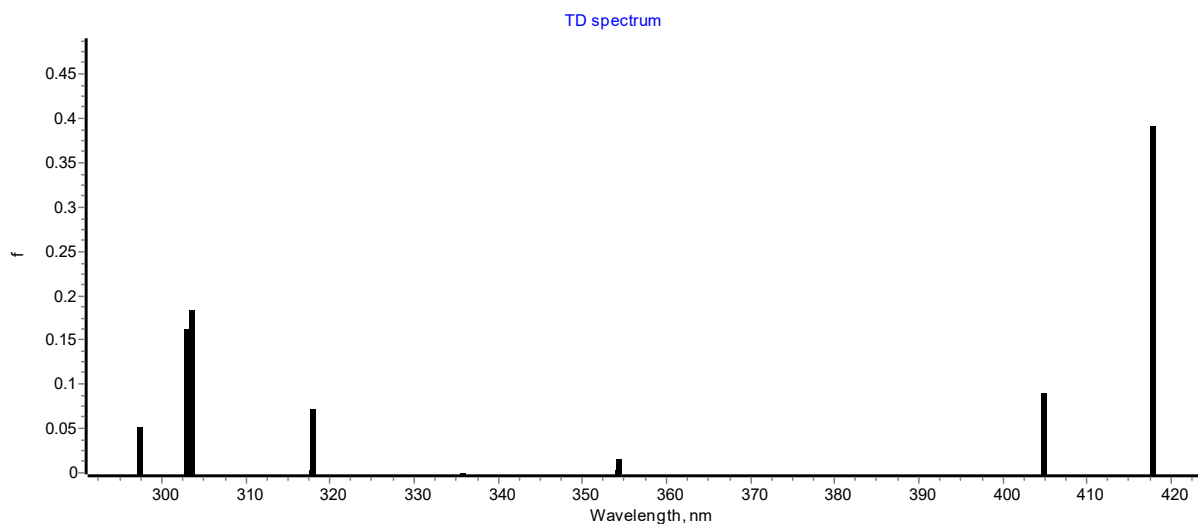


Figure S5. TD spectrum of the **BPBCzO** molecule.

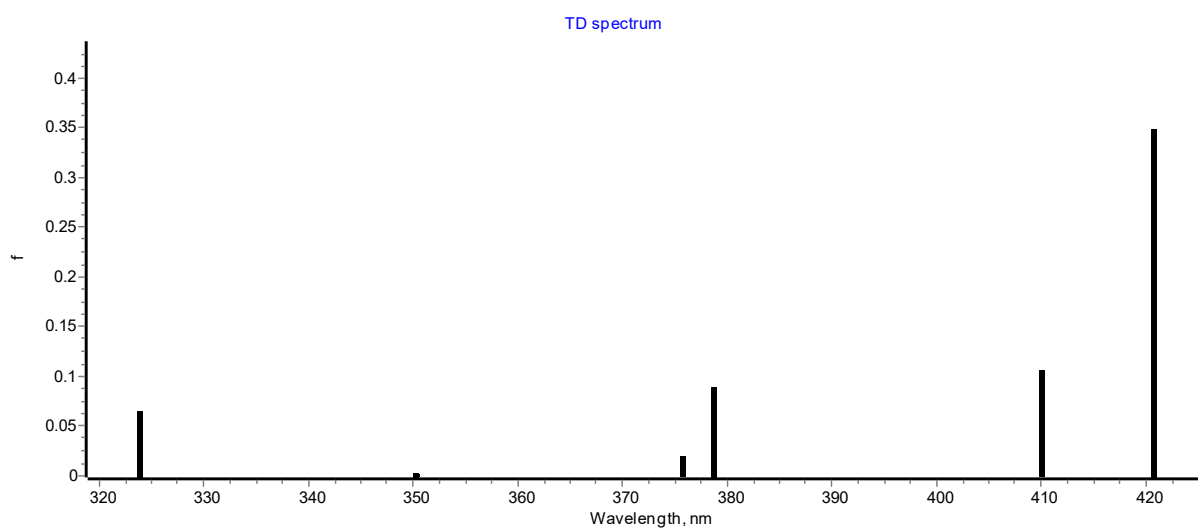


Figure S6. TD spectrum of the **BCzBCzO** molecule.

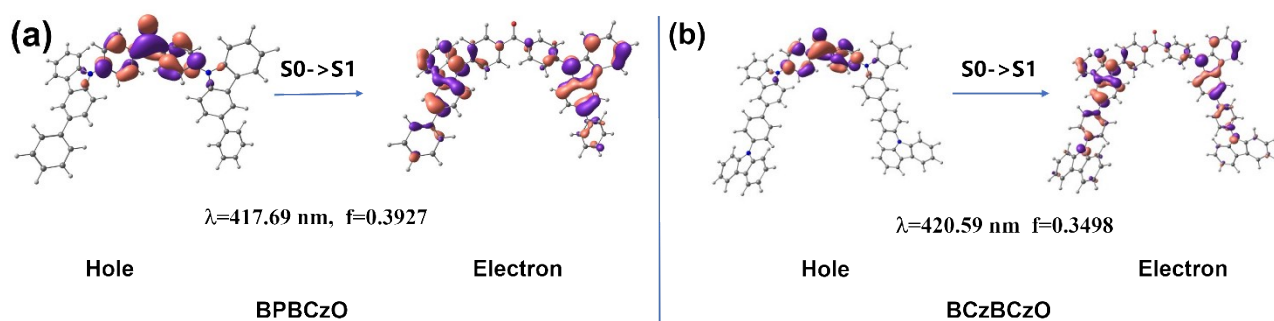
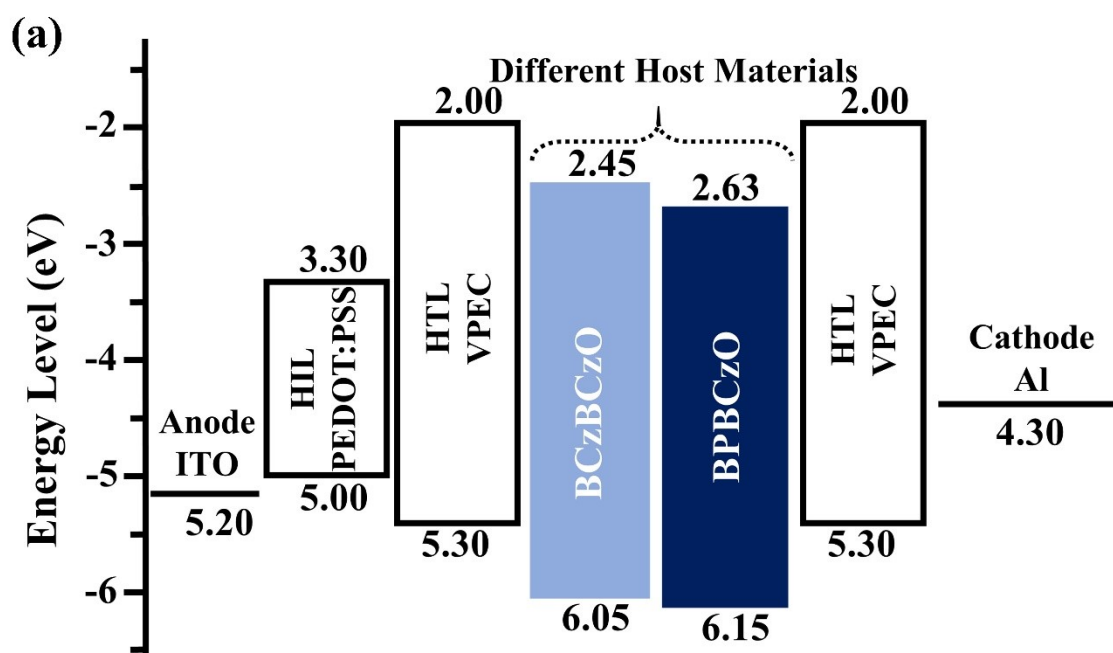


Figure S7. The NTO diagrams of  $S_0 \rightarrow S_1$  transition with their significant oscillator strengths for (a) **BPBCzO** and (b) **BCzBCzO**, respectively.

Table S1. TD-DFT analysis of **S0**->**S1** transitions (Transition energy, MO involved, and oscillator strength).

Molecule	Transition Energy/Wavelength	Nature of transitions  MOs involved Coefficients	oscillator strength  (f)
<b>BPBCzO</b>	2.9683eV/417.69 nm	HOMO->LUMO  0.69948	0.3927
<b>BCzBCzO</b>	2.4979 eV/420.59 nm	HOMO->LUMO 0.62921 HOMO-2 ->LUMO -0.30940	0.3498



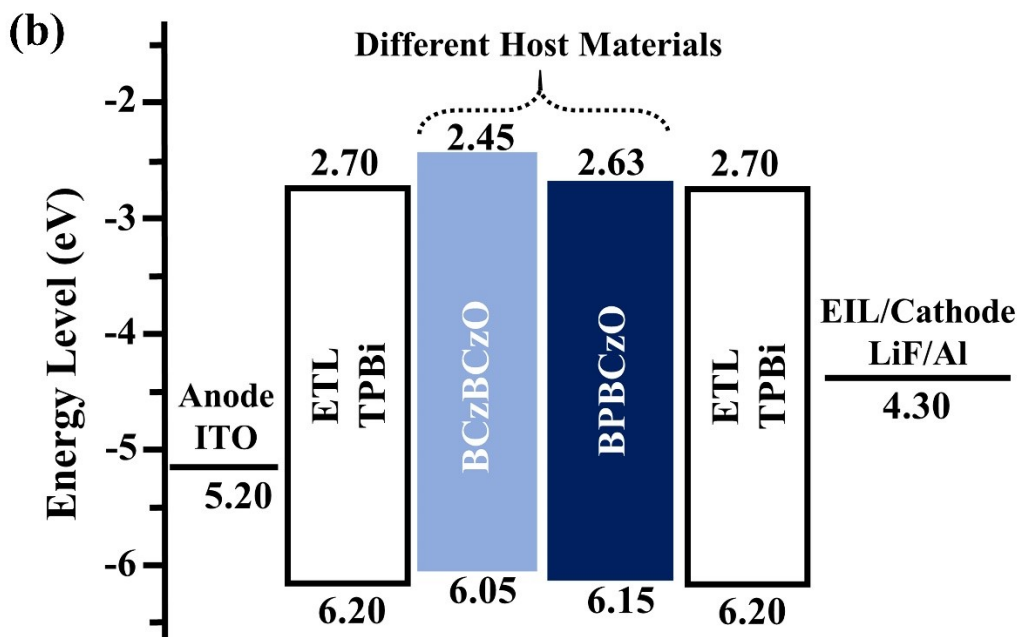


Figure S8. Schematic diagram of single carrier devices such as (a) hole-only device and (b) electron-only device fabricated with newly synthesized host materials, i.e. **BPBCzO** and **BCzBCzO**. VPEC and TPBi were used as an electron blocking layer and hole blocking layer in hole-only devices and electron-only devices, respectively.

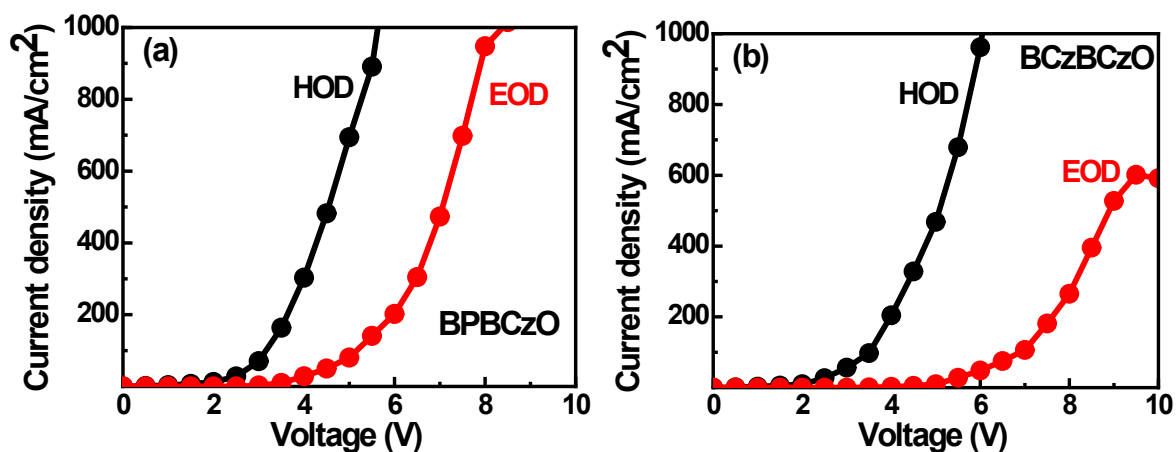
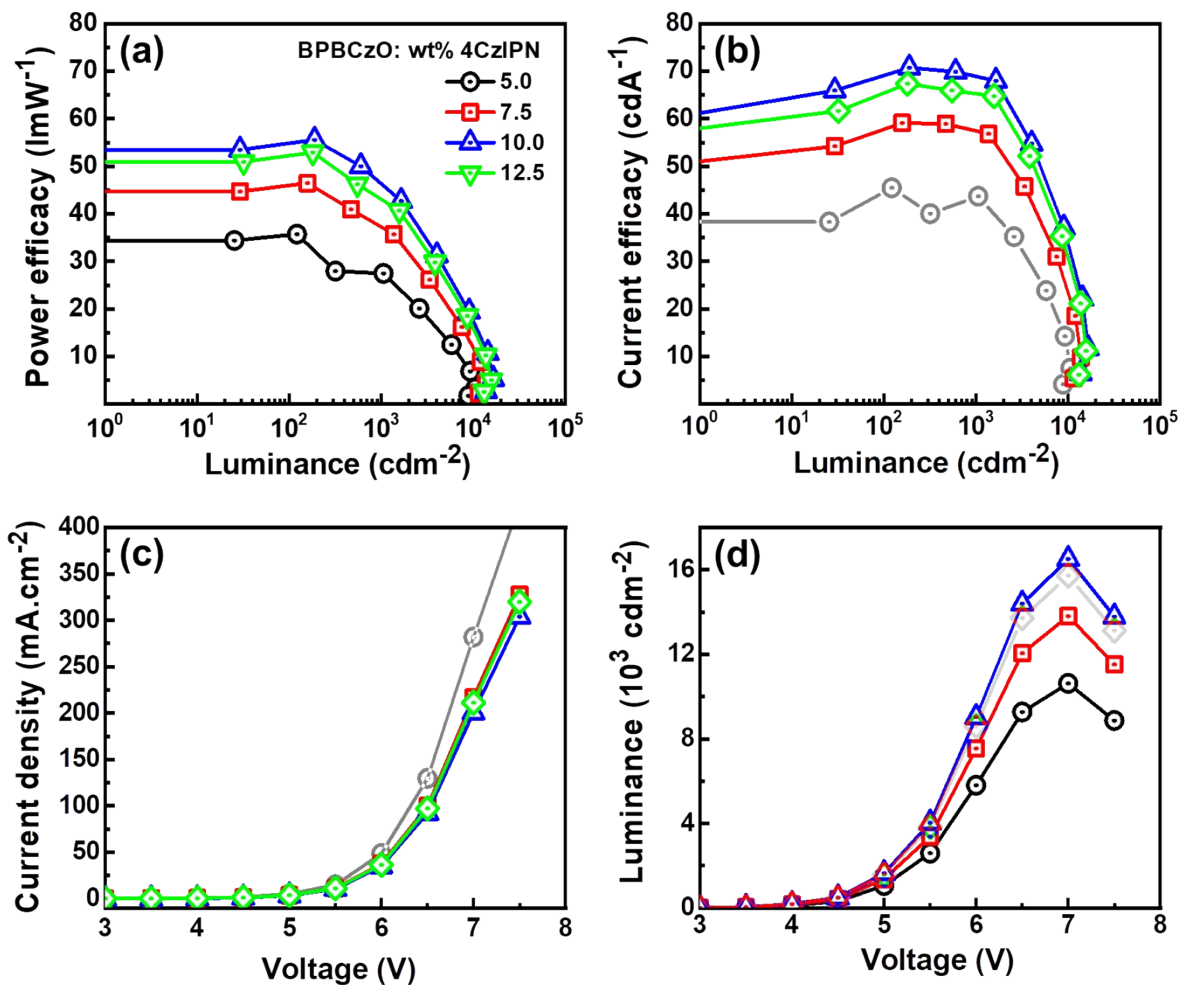
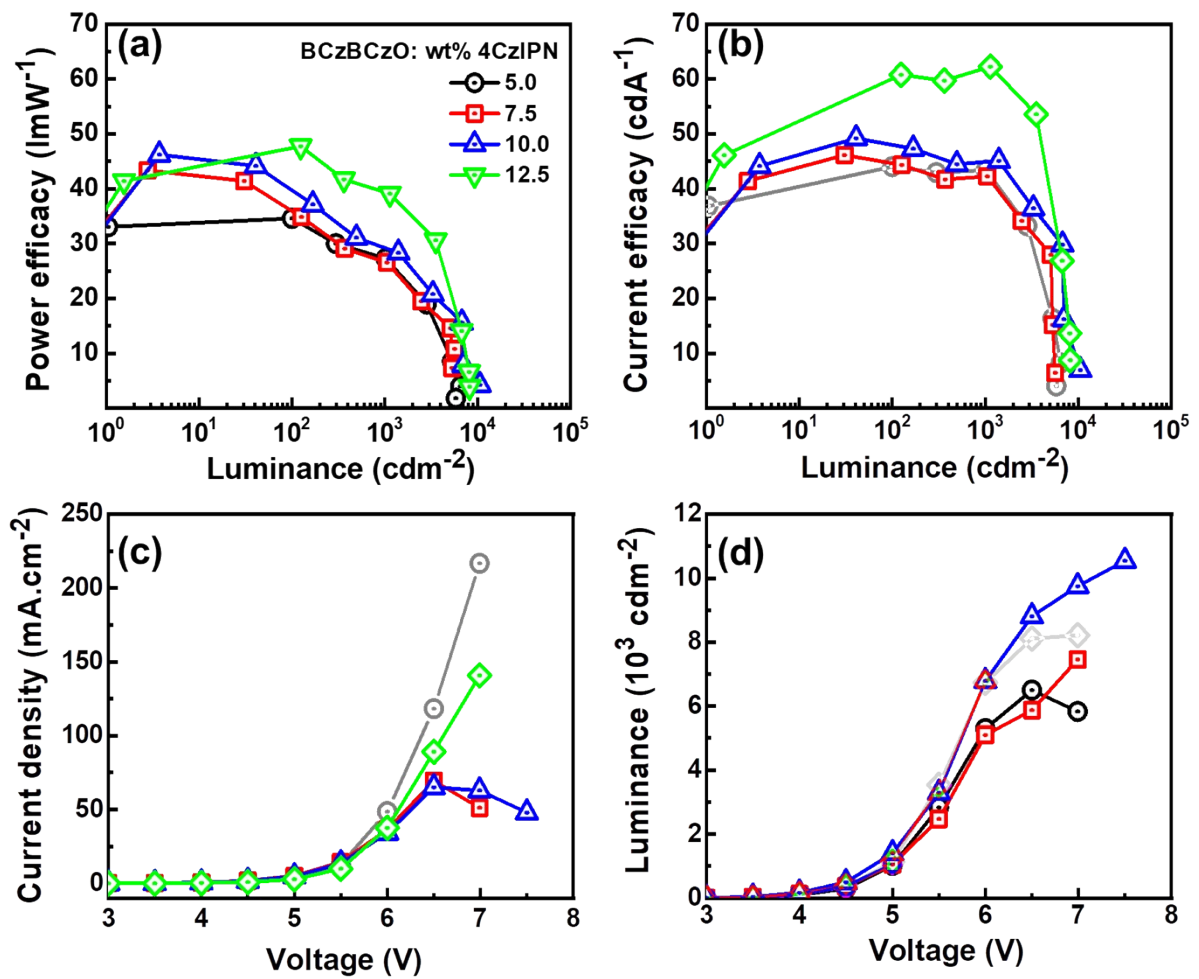


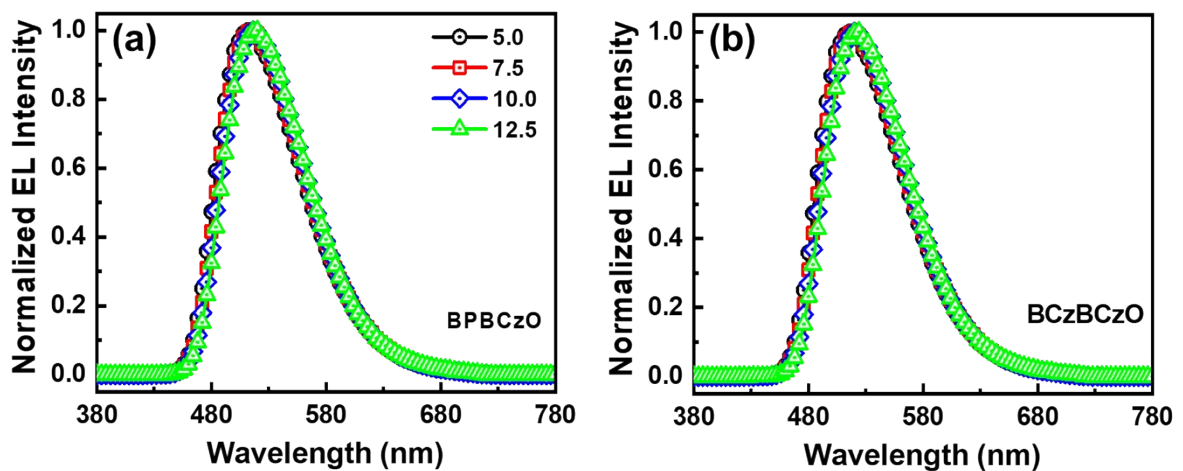
Figure S9. Current density characteristics of hole-only (black line plots) and electron-only devices (red line plots) fabricated with (a) **BPBCzO** and (b) **BCzBCzO**.



**Figure S10.** Effects of doping concentration on (a) power efficacy vs luminance, (b) current efficacy vs luminance, (c) current density vs voltage, and (d) luminance vs voltage results of the BPBCzO based green TADF OLED.



**Figure S11.** Effects of doping concentration on (a) power efficacy vs luminance, (b) current efficacy vs luminance, (c) current density vs voltage, and (d) luminance vs voltage results of the BCzBCzO based green TADF OLED.

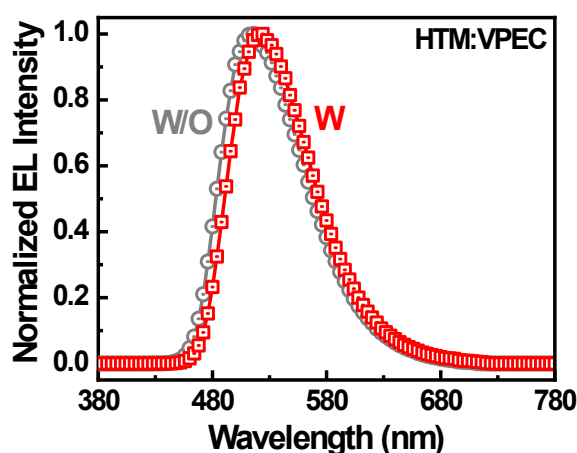


**Figure S12.** Effects of doping concentration on EL spectra of (a) BPBCzO and (d) BCzBCzO based green TADF organic LEDs.



**Table S1:** Carbazole-benzophenone derivatives-based green TADF organic LED performance at various dopant concentrations.

Hosts	Dopant Concen. (wt%)	V <sub>on</sub> (V)	Power efficacy (lm W <sup>-1</sup> )			Current efficacy (cd A <sup>-1</sup> )			External quantum efficiency (%)			CIE <sub>xy</sub> @100 cdm <sup>-2</sup>	L <sub>max</sub> cdm <sup>-2</sup>
			Max.	100 cdm <sup>-2</sup>	1,000 cdm <sup>-2</sup>	Max.	100 cdm <sup>-2</sup>	1,000 cdm <sup>-2</sup>	Max.	100 cdm <sup>-2</sup>	1,000 cdm <sup>-2</sup>		
BCzBCzO	5.0	3.5	34.6	34.6	27.4	44.0	44.0	43.4	9.3	9.3	9.0	(0.26,0.56)	6,501
	7.5	3.3	43.3	36.7	26.8	46.1	44.8	42.2	14.3	14.0	13.1	(0.28,0.57)	7,900
	10.0	2.7	46.2	40.9	29.5	49.2	48.3	44.8	15.3	15.0	14.0	(0.29,0.57)	10,540
	12.5	3.0	47.7	46.5	39.6	62.3	57.9	61.8	13.3	12.2	13.2	(0.29,0.57)	8,213
BPBCzO	5.0	3.0	35.7	35.4	27.5	45.4	43.8	43.4	14.2	13.7	13.6	(0.27,0.57)	10,620
	7.5	3.0	46.4	45.6	35.9	59.1	54.8	54.9	18.4	17.1	17.2	(0.28,0.57)	13,800
	10.0	3.0	55.6	54.3	43.1	70.7	64.1	64.8	23.2	21.0	21.3	(0.28,0.57)	16,500
	12.5	3.1	52.9	51.8	41.0	67.4	61.4	61.9	21.0	19.1	19.3	(0.28,0.57)	15,720



**Figure S13.** EL spectra of BPBCzO based TADF organic LEDs fabricated with (red line) and without (grey line) VPEC.

#### Cartesian coordinates of the optimized structures of BPBCzO

6	7.662807617	-3.595291164	-2.053823890
6	8.723133724	-2.677605404	-1.990359371
6	8.535335968	-1.417856566	-1.433928102
6	7.275948568	-1.078707121	-0.929558275

6	6.220357063	-2.021314282	-0.988021111
6	6.400380041	-3.282347350	-1.559155734
6	6.737131720	0.118370279	-0.318269138
6	5.374043095	-0.140719097	-0.038838473
7	5.064850395	-1.443424066	-0.444445216
6	7.293310873	1.353993053	0.015860606
6	6.506255126	2.333204071	0.630057091
6	5.152714599	2.038826514	0.908540090
6	4.575326673	0.817052567	0.587393054
6	3.799272976	-2.071982088	-0.327221849
6	2.656892999	-1.455967498	-0.846604587
6	1.414667997	-2.065786455	-0.712843221
6	1.295274497	-3.314039172	-0.088045147
6	2.452057699	-3.936756818	0.401796579
6	3.688409850	-3.317314588	0.305039819
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6	-1.295231597	-3.314035568	0.087408965
6	-1.414408060	-2.065734579	0.712154428
6	-2.656573260	-1.455878692	0.846270628
6	-3.799150307	-2.071910983	0.327338209
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6	-2.452207647	-3.936765364	-0.401989426
7	-5.064643667	-1.443294132	0.444950592

6	-5.373961231	-0.140661652	0.039143754
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6	-6.506377472	2.333075952	-0.630017858
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6	-8.386948000	3.762999341	-1.494602295

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1	-9.935403129	5.057856297	-2.224492649
1	-8.978345698	2.866571272	-1.642938505

**Cartesian coordinates of the optimized structure of BCzBCzO**

6	9.760234188	5.107380604	0.161672167
6	8.334186964	5.386952691	1.900496867
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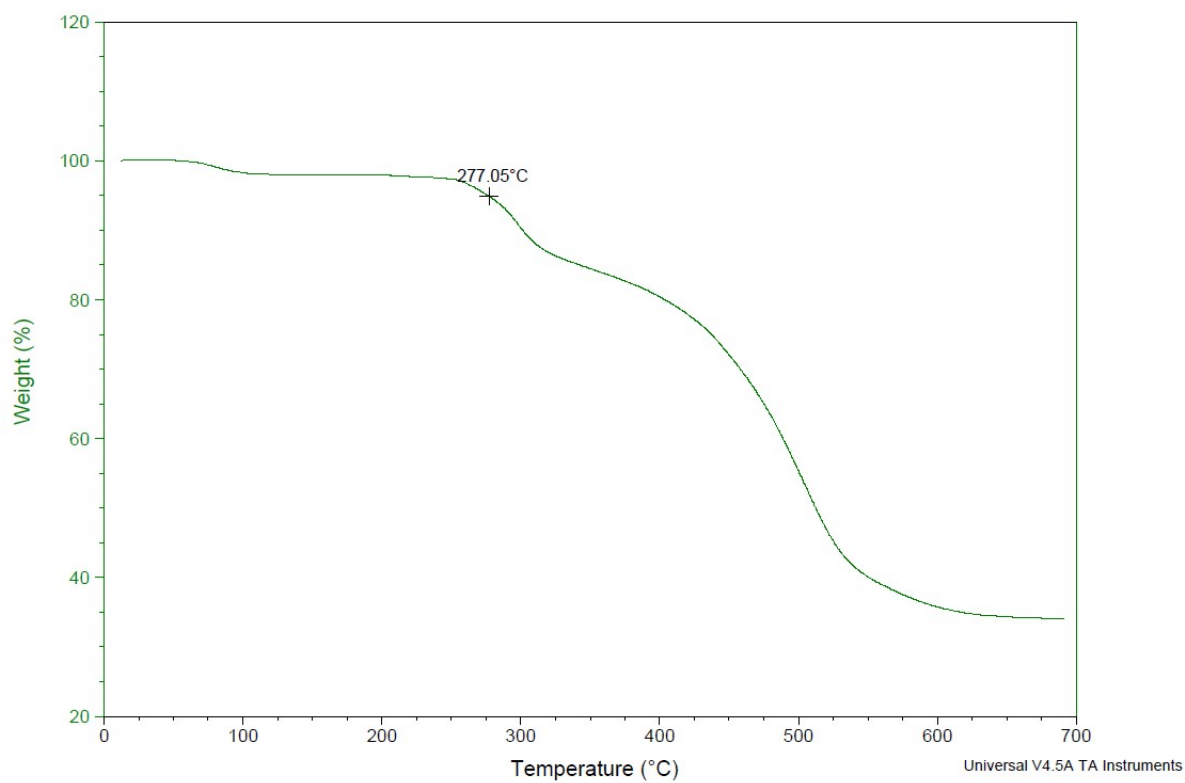
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6	7.948532216	7.433525540	3.733906933
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6	9.124188959	6.561942141	1.826076606
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1	-9.374772801	-5.929829916	3.449691288
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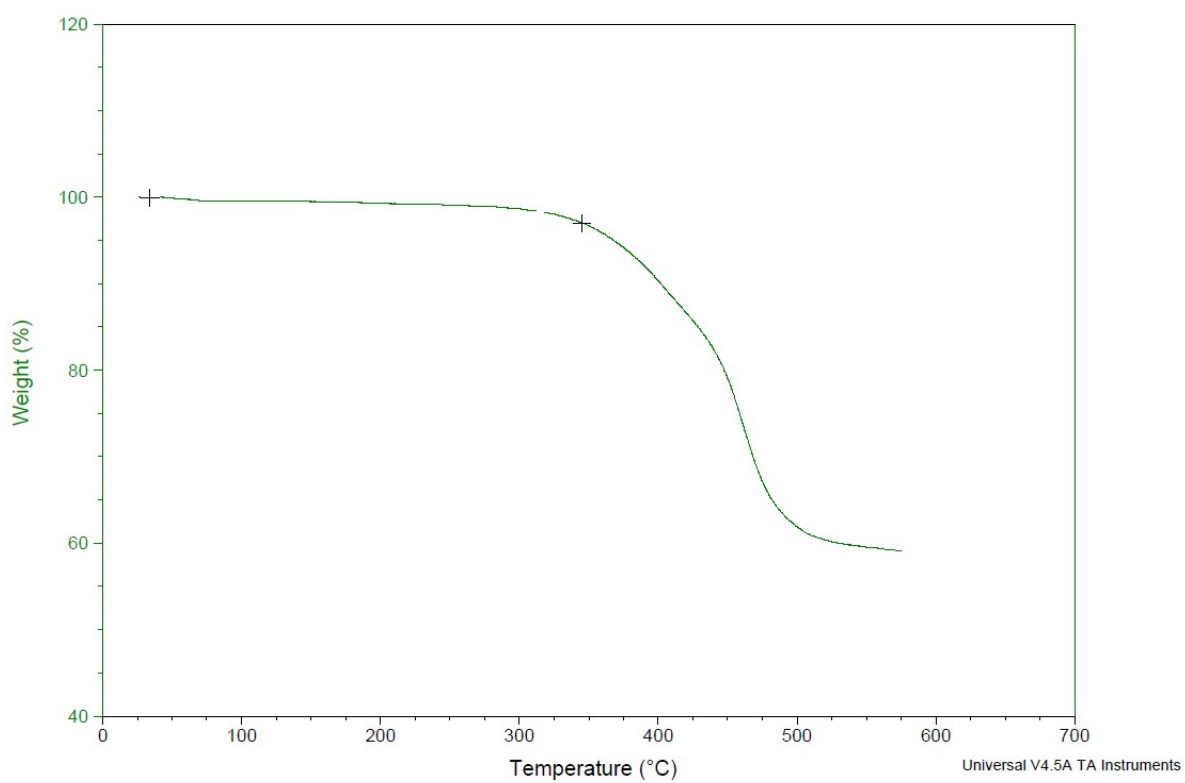


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1	-4.621436162	-6.821056480	-0.209688751
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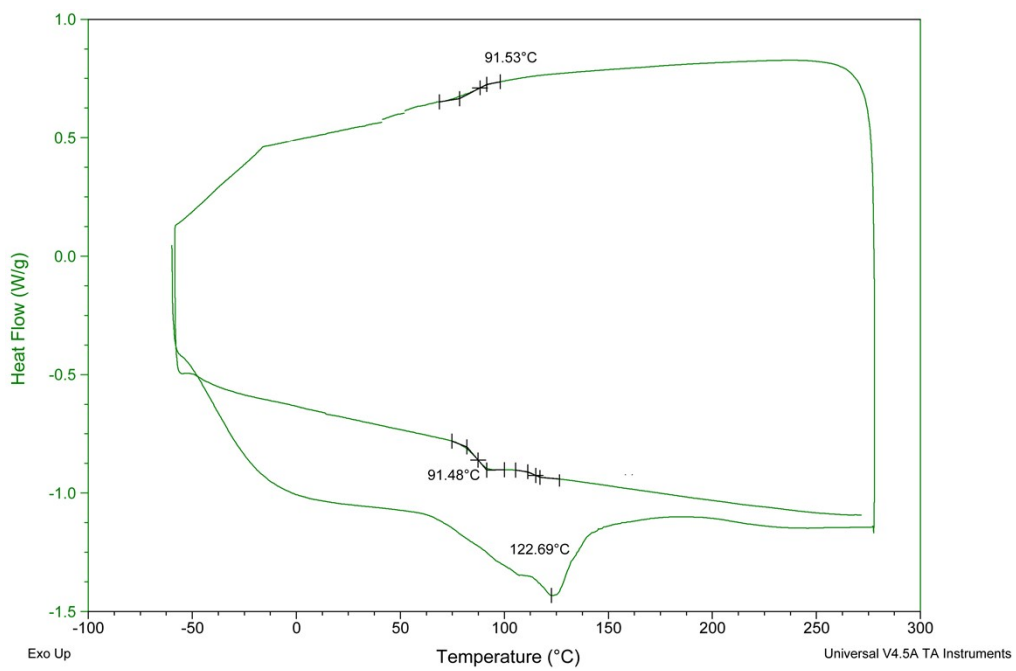
1	-9.517748292	8.494342035	-2.709615061
1	-7.780364237	8.222672014	-4.457468279
1	-6.432084479	6.154714848	-4.575651842
1	-6.768423717	4.318786919	-2.950724292
1	-10.233136188	3.660232241	1.380525476
1	-11.989642962	5.097362918	2.367121565
1	-12.502885256	7.318450550	1.411750522
1	-11.252093705	8.161890609	-0.555511533
1	7.780362756	8.222394937	4.457799189
1	9.517463700	8.494391513	2.709718272
1	11.251544154	8.162272887	0.555337952
1	12.502176846	7.319083922	-1.412139726
1	11.989135058	5.097952295	-2.367502441
1	10.232979817	3.660526051	-1.380705576



**Figure S14.** TGA curve of compound **BPBCzO**



**Figure S15.** TGA curve of compound **BCzBCzO**



**Figure S16.** DSC curves of compound **BPBCzO**. Heating rate: 10 °C/min.