## **Supplementary Information**

## Statistic regression strategy: Mixture Design

For the purpose of regression,  $X_1$ ,  $X_2$ , and  $X_3$  are denoted as the ratio of PEIE, TOAB, and PVP, respectively. The current density, light intensity, and current efficiency of the PLED from experimental results are listed together with the design matrix as shown in Table s1. The experimental results were regressed into polynomial form via SAS. The regression equations for the current density, light intensity, and current efficiency of the PLED are of the following form:

$$J=690X_{1}+550X_{2}+299X_{3}+584X_{1}X_{2}+595X_{2}X_{3}-2498X_{1}X_{2}X_{3}+337X_{1}X_{2}(X_{1}-X_{2})$$
  
+1684X\_{1}X\_{3}(X\_{1}-X\_{3})-962X\_{2}X\_{3}(X\_{2}-X\_{3})+10346X\_{1}X\_{1}X\_{2}X\_{3} (1) (1)

 $L=47454X_{1}+39176X_{2}+19892X_{3}+28545X_{1}X_{2}+16729X_{1}X_{3}+52457X_{2}X_{3}+378500X_{1}X_{3}(X_{1}-X_{3})-58824X_{2}X_{3}(X_{2}-X_{3})+553647X_{1}X_{1}X_{2}X_{3}+60801X_{1}X_{2}X_{3}X_{3}$ (2)

$$CE=6.92X_{1}+7.18X_{2}+6.66X_{3}-2.05X_{1}X_{2}+4.89X_{1}X_{3}+2.31X_{2}X_{3}-4.77X_{1}X_{2}(X_{1}-X_{2})+9.22X_{1}X_{3}(X_{1}-X_{3})-105.39X_{1}X_{1}X_{2}X_{3}+150.88X_{1}X_{2}X_{3}X_{3}$$
(3)

$$L=1199.05X_{1}+1289.01X_{2}+1386.63X_{3}-847.68X_{1}X_{2}-$$

$$748.43X_{1}X_{3}+1467.29X_{1}X_{2}X_{3}+766.84X_{1}X_{2}(X_{1}-X_{2})+1561.18X_{1}X_{3}(X_{1}-X_{3})+1354.34X_{2}X_{3}(X_{2}-X_{3})+27151X_{1}X_{2}X_{3}X_{3}$$
(4)

 $CE=11.64X_{1}+12.33X_{2}+11.88X_{3}+2.89X_{2}X_{3}+11.13X_{1}X_{2}(X_{1}-X_{2})-10.33X_{1}X_{3}(X_{1}-X_{3})+47.24X_{1}X_{2}X_{2}X_{3}+71.83X_{1}X_{2}X_{3}X_{3}$ (5)

$$\begin{split} PE &= 11.25X_1 + 13.48X_2 + 11.58X_3 - 7.82X_1X_2 - 5.84X_1X_3 - \\ & 4.29X_2X_3 + 46.99X_1X_2X_3 + 15.26X_1X_2(X_1 - X_2) - 5.10X_1X_3(X_1 - X_3) + 58.62X_1X_2X_3X_3(6) \end{split}$$

$$J=7.65X_{1}+7.88X_{2}+6.86X_{3}+8.77X_{1}X_{2}+6.35X_{1}X_{3}+3.55X_{2}X_{3}-11.35X_{1}X_{3}(X_{1}-X_{3})-13.21X_{2}X_{3}(X_{2}-X_{3})+49.57X_{1}X_{1}X_{2}X_{3}-257.37X_{1}X_{2}X_{3}X_{3}$$
(7)

 $CE=12.43X_{1}+12.77X_{2}+14.03X_{3}-7.62X_{1}X_{2}-7.73X_{1}X_{3}-11.60X_{1}X_{2}X_{3}+6.28X_{1}X_{2}(X_{1}-X_{2})+12.3X_{1}X_{3}(X_{1}-X_{3})+16.05X_{2}X_{3}(X_{2}-X_{3})+275.26X_{1}X_{2}X_{3}X_{3}$ (8)

 $PE=9.38X_{1}+10.72X_{2}+7.72X_{3}+15.60X_{1}X_{2}+22.52X_{1}X_{3}+27.69X_{2}X_{3}+27.61X_{1}X_{2}(X_{1}-X_{2})-29.20X_{2}X_{3}(X_{2}-X_{3})-303.16X_{1}X_{1}X_{2}X_{3} \tag{9}$ 

For checking the statistic significant of this regression equations, the analysis of variance (ANOVA) for current density, light intensity, current efficiency, and power efficiency at various conditions are shown from Tables s4 to s12. The test statistics, F is defined as F= MSR/MSE where MSR and MSE are the mean square of regression and mean square error from the analysis of variance, respectively. MSR can be obtained by dividing the sum of squares of regression with the degrees of freedom (d.f.). The statistically significant regression model is obtained when the calculated value of F is greater than that of the table F (P - 1, v, 1 -  $\alpha$ ) value, where v = N - P is the degrees of freedom of error, N is the number of experiments, and P is the parameters. F (P - 1, v, 1 -  $\alpha$ ) is the F value at the  $\alpha$  probability level. After checking with F value and R<sup>2</sup>, all these three regression equations are statistic significant. The compositions versus current density, light intensity, current efficiency, and power efficiency contour plots were drawn by using MATLAB as shown in Fig. S2.

		composition				
EIL	<b>x</b> <sub>1</sub>	x <sub>2</sub>	X3	J	L	CE
No.	(PEIE)	(TOAB)	(PVP)	$(mA/cm^2)$	$(cd/m^2)$	(cd/A)
1	1	0	0	667	47500	7.11
2	0	1	0	540	39100	7.20
3	0	0	1	292	19800	6.76
4	1/3	1/3	1/3	672	53200	7.90
5	1/2	1/2	0	691	50700	7.33
6	0	1/2	1/2	590	42600	7.19
7	1/2	0	1/2	447	37500	8.38
8	2/3	1/6	1/6	881	59800	6.77
9	1/6	2/3	1/6	579	45900	7.91
10	1/6	1/6	2/3	440	38000	8.62
11	3/4	1/4	0	845	50300	5.63
12	1/4	3/4	0	713	46500	6.52
13	3/4	0	1/4	783	51200	6.52
14	1/4	0	3/4	271	22700	8.38
15	0	3/4	1/4	498	38600	7.75
16	0	1/4	3/4	551	40000	7.24

**Table S1** Design matrix and experimental results of PEIE-TOAB-PVP Ternarydevices at 7V.

		composition				
EIL	<b>X</b> 1	x <sub>2</sub>	X3	L	CE	PE
No.	(PEIE)	(TOAB)	(PVP)	$(cd/m^2)$	(lm/W)	(lm/W)
1	1	0	0	1220	12	11.54
2	0	1	0	1322	12.5	12.44
3	0	0	1	1409	12	11.68
4	1/3	1/3	1/3	1481	12.7	12.8
5	1/2	1/2	0	1129	12	11.44
6	0	1/2	1/2	1271	12.5	12.08
7	1/2	0	1/2	1151	12	10.45
8	2/3	1/6	1/6	1310	12.6	11.66
9	1/6	2/3	1/6	1345	13.4	11.16
10	1/6	1/6	2/3	1447	14.6	12.22
11	3/4	1/4	0	1074	12.6	11.02
12	1/4	3/4	0	961	10.6	9.47
13	3/4	0	1/4	1019	10.5	10.6
14	1/4	0	3/4	1225	12.3	9.38
15	0	3/4	1/4	1449	12.9	11.71
16	0	1/4	3/4	1251	12.5	11.05

**Table S2** Design matrix and experimental results of PEIE-TOAB-PVP Ternary devices at 10 mA/cm<sup>2</sup>.

**Table S3** Design matrix and experimental results of PEIE-TOAB-PVP Ternarydevices at 1000  $cd/m^2$ .

		composition				
EIL	<b>x</b> <sub>1</sub>	x <sub>2</sub>	X3	J	CE	PE
No.	(PEIE)	(TOAB)	(PVP)	$(mA/cm^2)$	(cd/A)	(lm/W)
1	1	0	0	7.39	12.74	12.12
2	0	1	0	7.74	13.5	13.5
3	0	0	1	6.72	14.63	13.13
4	1/3	1/3	1/3	7.2	14.16	13.89
5	1/2	1/2	0	9.38	12.02	11.73
6	0	1/2	1/2	7.98	13.46	12.43
7	1/2	0	1/2	8.24	12.07	10.82
8	2/3	1/6	1/6	8.45	12.63	12.39
9	1/6	2/3	1/6	7.3	13.34	11.97
10	1/6	1/6	2/3	6.77	14.2	13.51

11	3/4	1/4	0	9.87	10.88	11.02
12	1/4	3/4	0	9.96	9.65	9.47
13	3/4	0	1/4	9.73	10.47	10.6
14	1/4	0	3/4	8	12.09	10
15	0	3/4	1/4	7.49	13.96	12.53
16	0	1/4	3/4	9.18	11.69	11.09

**Table S4** The analysis of variance for the fit of current densities of PLED at 7V.

Source	d.f.	Sum of	Mean square	F value
		squares		
Model	10	6048036	604804	224
Error	6	16222	2704	
Total	16	6064258		

 $R^2 = 0.9973$ 

Table S5 The analysis of variance for the fit of light intensities of PLED at 7V.

Source	d.f.	Sum of squares	Mean square	F value
Model	10	30843229163	86.63	270.33
Error	6	1.923	0.32	
Total	16	868.25		

 $R^2 = 0.9999$ 

Table S6 The analysis of variance for the fit of current efficiency of PLED at 7V.

Source	d.f.	Sum of squares	Mean square	F value
Model	10	866.325	3084322916	12413
Error	6	1490837	248473	
Total	16	30844720000		

 $R^2 = 0.9978$ 

Source	d.f.	Sum of squares	Mean square	F value
Model	10	25498507	2549851	499.62
Error	6	30621	5103.53	
Total	16	25529128		

**Table S7** The analysis of variance for the fit of light intensities of PLED at 10  $mA/cm^2$ .

 $R^2 = 0.9988$ 

**Table S8** The analysis of variance for the fit of current efficiencies of PLED at 10  $mA/cm^2$ .

Source	d.f.	Sum of	Mean square	F value
		squares		
Model	8	2453	307	755
Error	8	3	0.4	
Total	16	2457		

 $R^2 = 0.9987$ 

**Table S9** The analysis of variance for the fit of power efficiency of PLED at 10  $mA/cm^2$ .

Source	d.f.	Sum of squares	Mean square	F value
Model	10	2087.74	208.77	359.66
Error	6	3.48	0.58	
Total	16	2091.23		

 $R^2 = 0.9983$ 

**Table S10** The analysis of variance for the fit of current density of PLED at 1000  $cd/m^2$ .

Source	d.f.	Sum of squares	Mean square	F value
Model	10	1094.95	109.49	283.06
Error	6	2.32	0.39	
Total	16	1097.27		

 $R^2 = 0.9979$ 

Source	d.f.	Sum of squares	Mean square	F value
Model	10	2562.06	256.20	243.36
Error	6	6.32	1.05	
Total	16	2568.37		

**Table S11** The analysis of variance for the fit of current efficiency of PLED at 1000  $cd/m^2$ .

**Table S12** The analysis of variance for the fit of power efficiency of PLED at 1000  $cd/m^2$ .

Source	d.f.	Sum of squares	Mean square	F value
Model	9	2687.52	298.61	30.75
Error	7	67.97	9.71	
Total	16	2755.49		

 $R^2 = 0.9753$ 



Fig. S1 FT-IR spectra for PEIE, PVP, and PEIE : PVP



**Fig. S2** Contour line plots of (a) light intensity (cd/m<sup>2</sup>) and (b) current efficiency (cd/A) at 7V; (c) light intensity (cd/m<sup>2</sup>) and (d) power efficiency (lm/W) at 10 mA/cm<sup>2</sup>; (e) current density (mA/cm<sup>2</sup>) and (f) current efficiency (cd/A) at 1000 cd/m<sup>2</sup>.

## Confirmation of statistic model via experimental results

Since the statistic significant has been checked by ANOVA, the model still needs to be consistent with the experimental results. Three compositions (PEIE: PVP: TOAB for A = 0.6 :0.2 :0.2, B = 0.4 : 0.35 : 0.25, and C = 0.25 : 0.5 :0.25) as the checking points has been chosen for fabricating devices. The L-V curves for A to C and contour plot with three checking points are shown in Fig. S3. The light intensities respectively are 61025, 55166, and 43025 cd/m<sup>2</sup> for A, B, and C compositions. The excellent match

between statistic and experimental results confirms the statistic significant of this model.



**Fig. S3** L-V curves with the three checking points A, B, C. Insert: Contour plot of light intensity with three checking points.

## **Deconvolution of XPS spectrum**

The parameters for C=O and C-O-H are acquired from the deconvolution of XPS O 1s core level spectrum for PEIE and PVP as shown in Fig. S4. The deconvolution of XPS spectrum for Binary film using two components shows mismatch with the raw data.



**Fig. S4** Deconvolution of XPS O 1s core level spectrum for (a) PEIE (b) PVP (c) Binary with two components.