

Supporting Information

Design of White Circularly Polarized Luminescence with High g_{lum} Across Entire Visible Regions by Dual-Loop Active Learning

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As a materials scientist, advise on selecting red, green, and blue fluorescent polymers for high-CRI white light emission. Recommend 20 specific polymers with high thermal stability, high quantum yield, and liquid-crystal-like characteristics. For each, briefly explain its role in achieving high-CRI white light.

Blue-Emitting Fluorescent Polymers

1. Polyfluorene (PFO)
2. Poly(p-phenylene vinylene) (PPV) derivatives
3. Poly(3,6-carbazole) (PCDTBT)
4. Poly(9,9-di-n-octylfluorene-alt-bis-N,N'-(4-butylphenyl)-bis-N,N'-phenyl-1,4-phenylenediamine) (PFB)

Green-Emitting Fluorescent Polymers

1. Poly(2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene) (MEH-PPV)
2. Poly(9,9-dioctylfluorene-co-benzothiadiazole) (F8BT)
3. Poly[(9,9-dioctylfluorenyl-2,7-diyl)-alt-(4,7-bis(2-thienyl)-2,1,3-benzothiadiazole)-5,5'-diyl] (PFO-DBT)
4. Poly(p-phenylene ethynylene) (PPE) derivatives

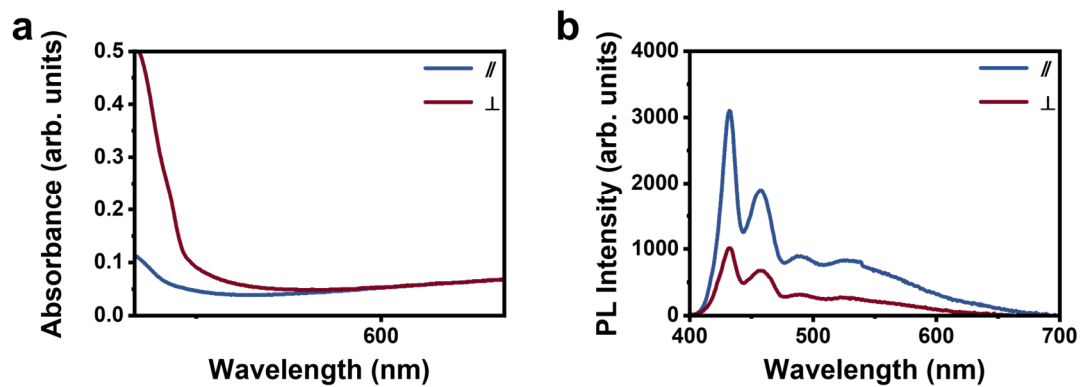
Red-Emitting Fluorescent Polymers

1. Poly(2,7-fluorene-alt-5,5'-(4',7'-di-2-thienyl-2',1',3'-benzothiadiazole)) (PF-DBT)
2. Poly[(9,9-dioctylfluorenyl-2,7-diyl)-alt-(4,7-bis(2-thienyl)-2,1,3-benzothiadiazole)-5,5'-diyl] (PFO-DBT)
3. Poly(1,4-phenylenevinylene-co-2,5-dioctyloxy-1,4-phenylenevinylene) (PPV-DO)
4. Poly(9,9-dioctylfluorene-alt-4,7-di(thiophen-2-yl)benzo[c][1,2,5]thiadiazole) (PF-TBT)

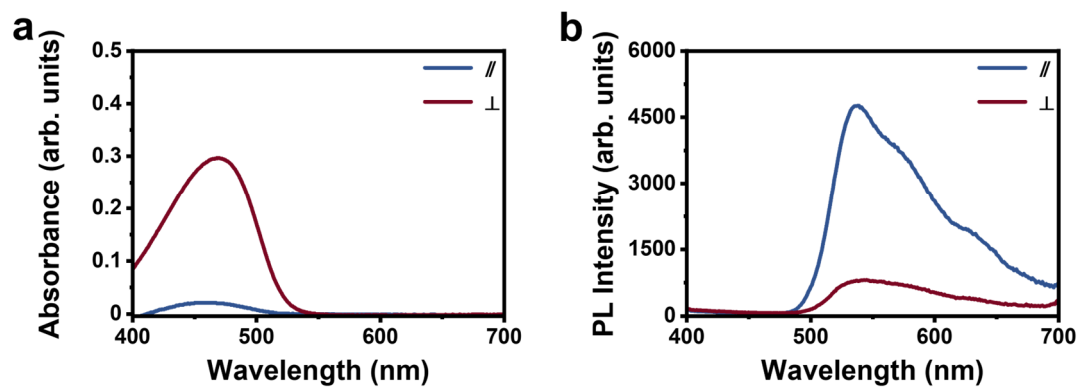


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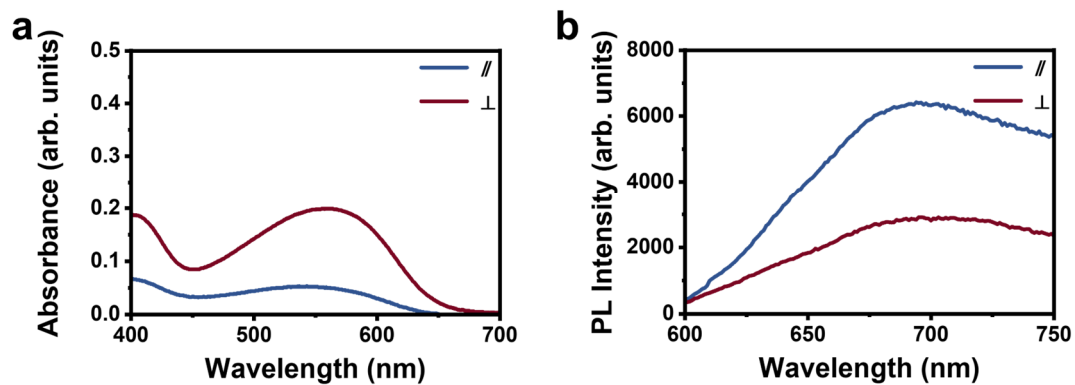
Supplementary Figure 1 | Schematic diagram of fluorescent molecules recommended by the large language models.



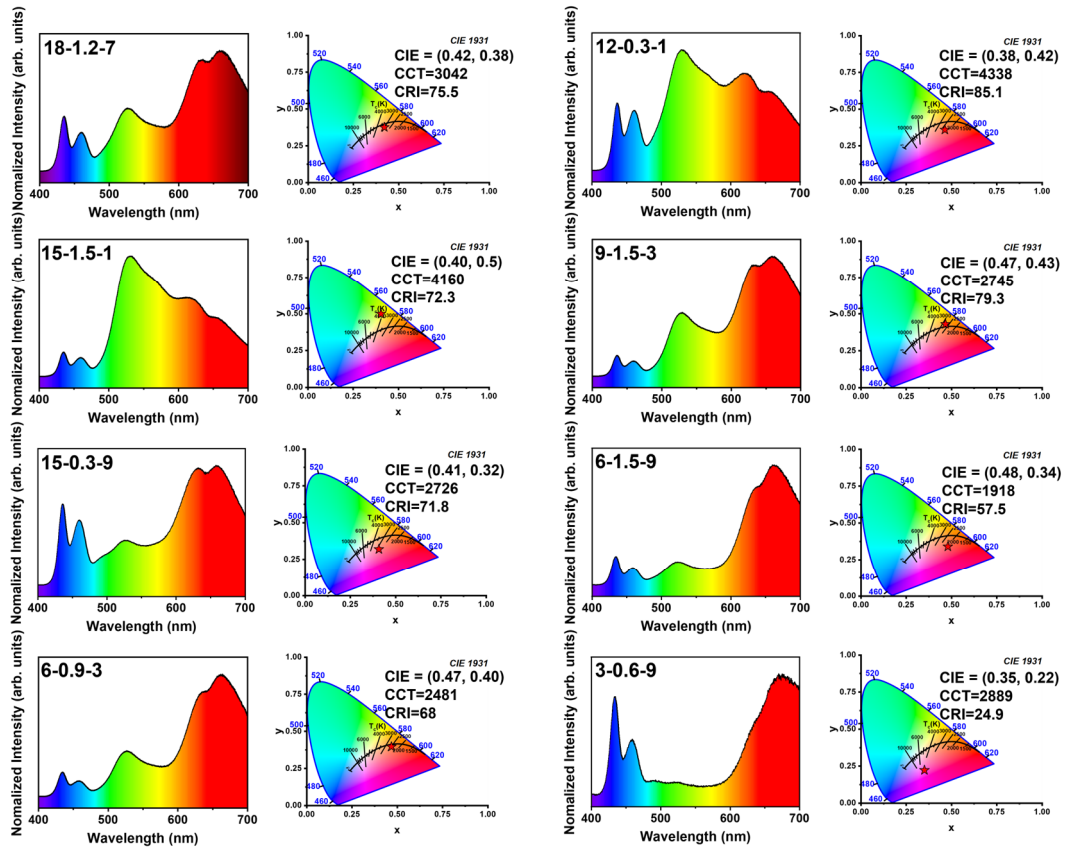
Supplementary Figure 2 | The optical properties of oriented PFO films. a Polarized UV-vis absorption spectra and **(b)** polarized fluorescence spectra of oriented PFO films ($\lambda_{\text{ex}}=365$ nm).



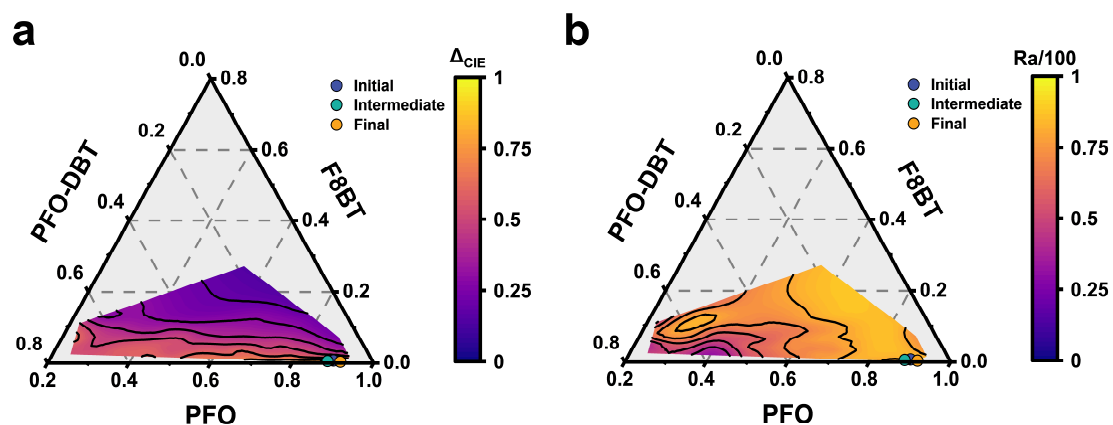
Supplementary Figure 3 | The optical properties of oriented F8BT films. a Polarized UV-vis absorption spectra and **(b)** polarized fluorescence spectra of oriented F8BT films (λ_{ex} =365 nm).



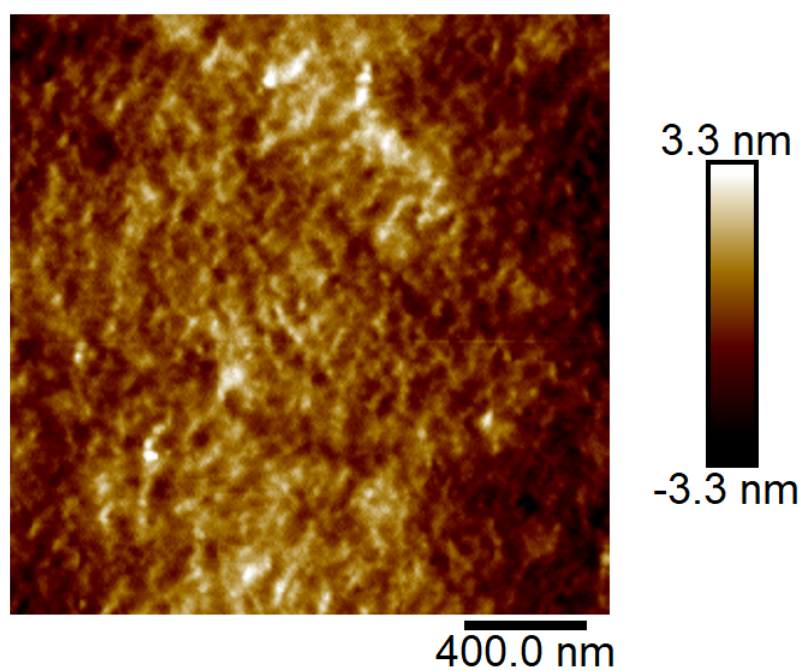
Supplementary Figure 4 | The optical properties of oriented PFO-DBT. a Polarized UV-vis absorption spectra and **(b)** polarized fluorescence spectra of oriented PFO-DBT films (λ_{ex} =400 nm).



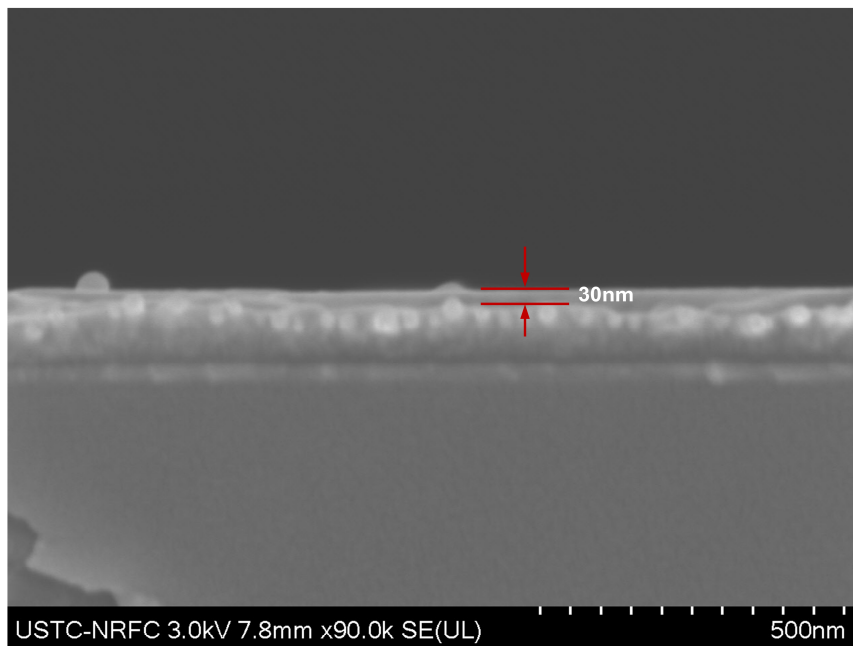
Supplementary Figure 5 | A subset of data sources of ternary system used to train the model. The luminescence spectra and the corresponding CIE coordinates, CRI and CCT.



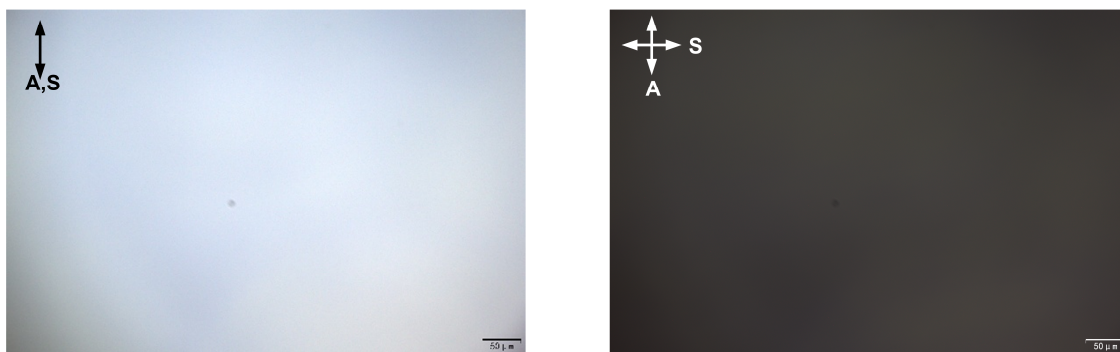
Supplementary Figure 6 | Active learning guides design of high performance WLE of ternary system. Representation of active learning path traversed through copolymer chemical space for (a) Δ_{CIE} and (b) $Ra/100$. The chemical space is represented as a ternary diagram with coordinates providing the fraction of incorporation of PFO, F8BT, and PPFO-DBT in WLE films. Colored circles indicate the mean composition of WLE films proposed during a given iteration.



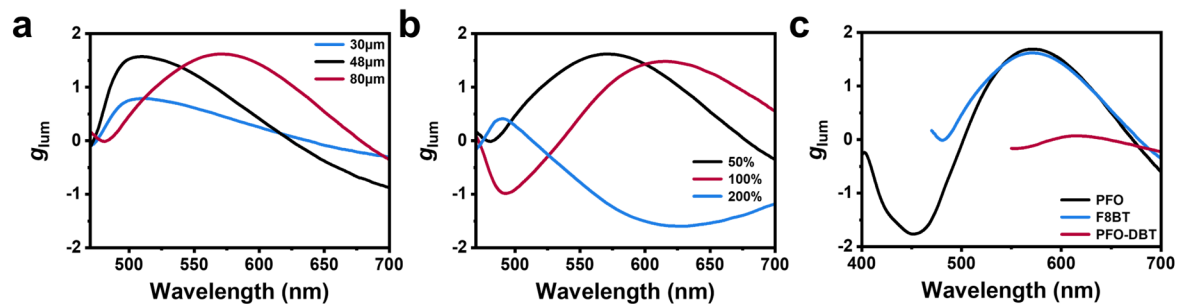
Supplementary Figure 7 | AFM images of the surface morphology of the emissive layer in the twisted-stacking hybrid film.



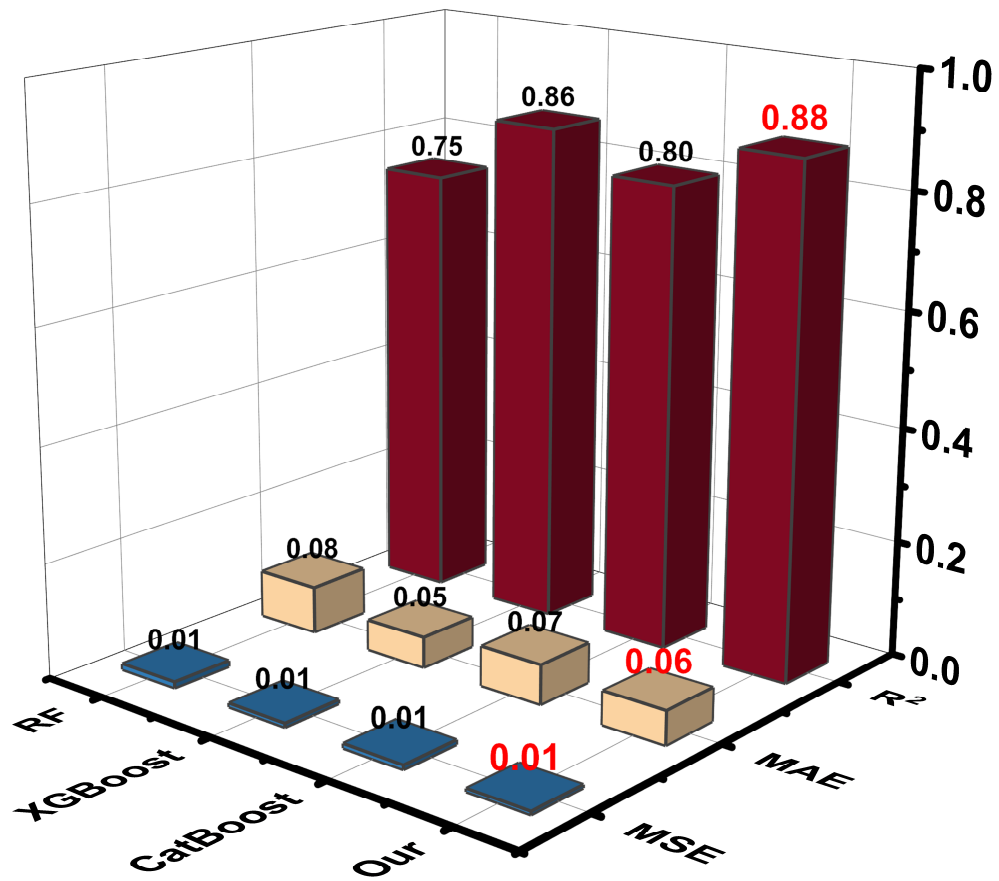
Supplementary Figure 8 | SEM images of the emissive layer in the twisted-stacking hybrid film.



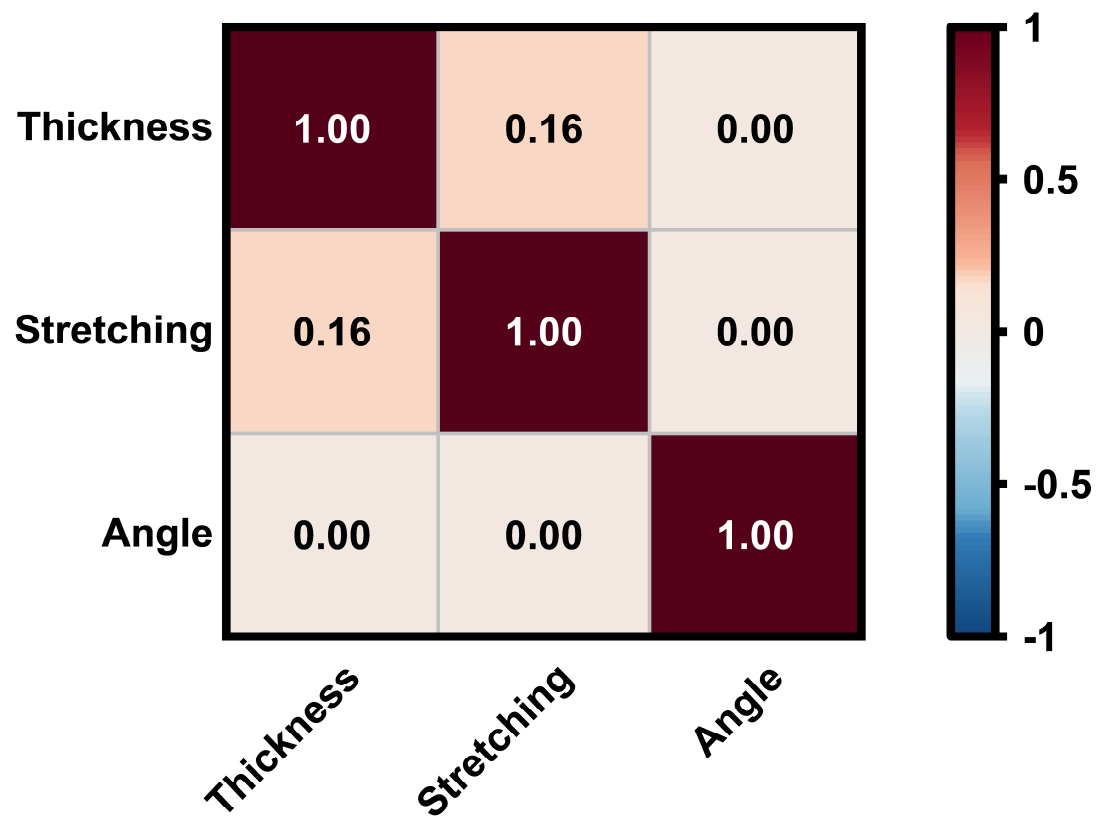
Supplementary Figure 9 | POM images of the emissive layer in the twisted-stacking hybrid film. The polarization direction for the polarizer is A (analyzer). The orientation direction of the hybrid film is S. Scale bars: 50 μ m.



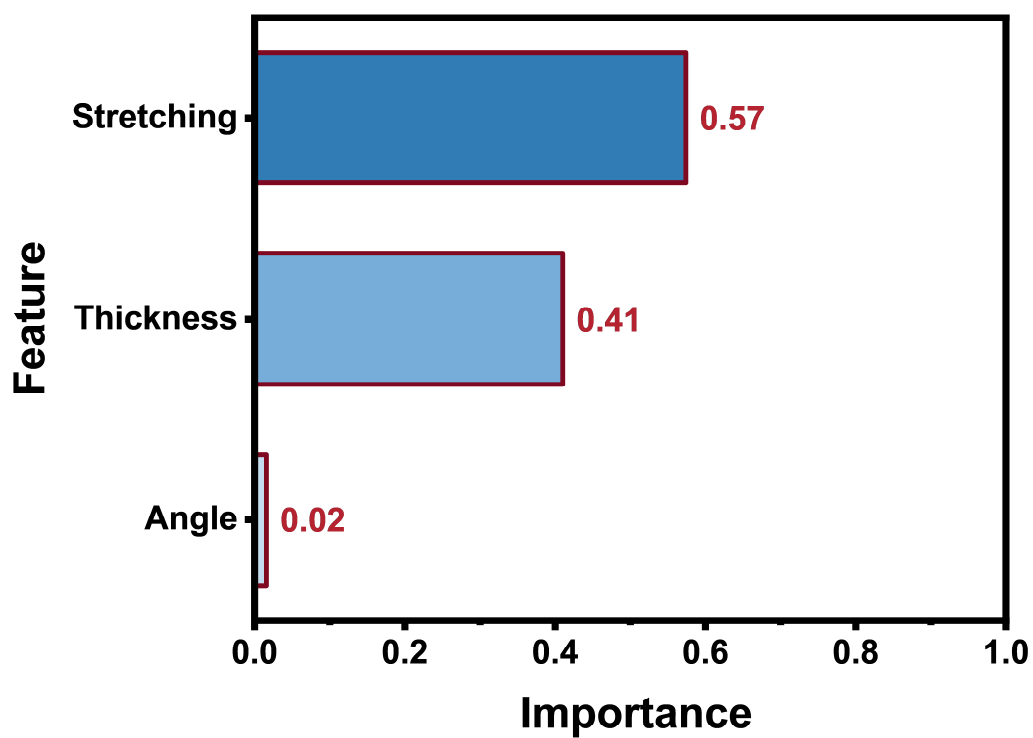
Supplementary Figure 10 | The variation of CPL spectra of chiroptical films with different parameters. The spectral lines of g_{lum} versus wavelength for heterostructure bilayer chiroptical films with different (a) thickness and (b) stretching degree of PVA layer, and (c) molecular selection of the highly oriented emissive layer.



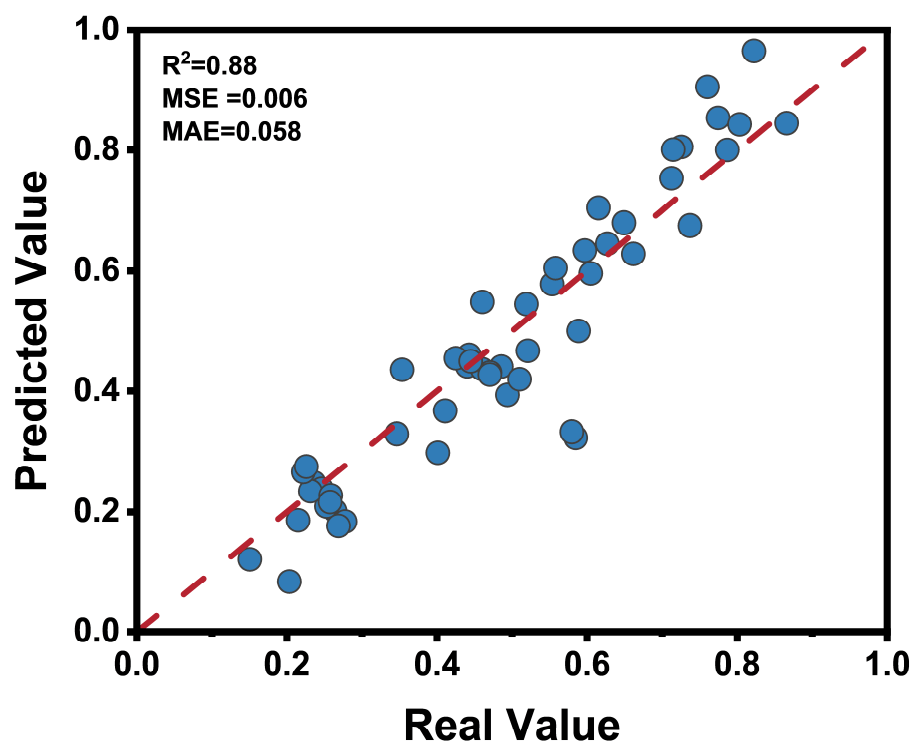
Supplementary Figure 11 | Active learning regression model. Mean square error, mean absolute error and coefficient of determination histogram of the models.



Supplementary Figure 12 | The correlation heat map of the feature. Correlation analysis among thickness, stretching agree, and angle parameters.



Supplementary Figure 13 | Feature-sorted histogram. Importance comparison of stretching, thickness and angle features.



Supplementary Figure 14 | Fitting performance of the TabPFN model in ternary systems.
Scatter plots of experimental and predicted results by the TabPFN model.



First, analyze the existing spectrum and determine why white light is not produced. Then, based on your expertise, provide solutions to address this issue. The final output should include 20 common small molecules or polymer additives, along with explanations of how each additive contributes to enabling the fluorescent dye to emit white light.

Cyan/Green-Blue Emitters (to bridge PFO and F8BT)

1. TTB
2. BCzVBi
3. C545T
4. Ir(mpim)₃
-

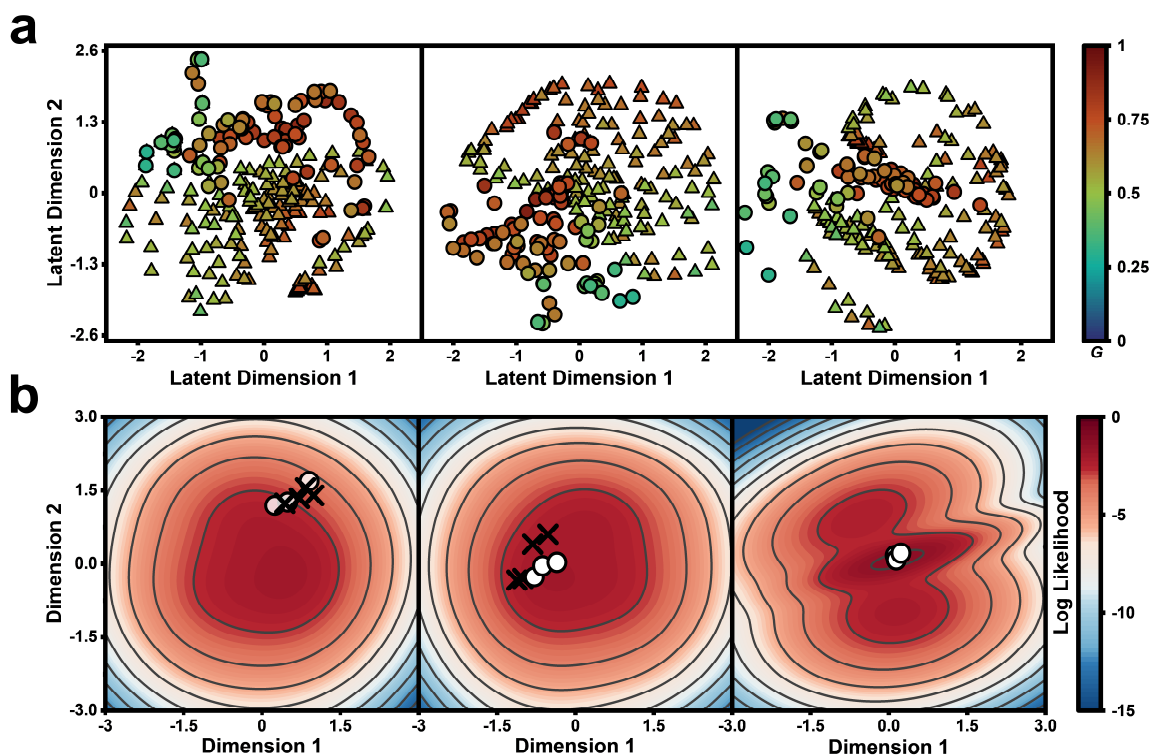
Category 2: Yellow/Orange Emitters (to balance Green-Red ratio and broaden the peak)

1. Rubrene
2. Coumarin 6
3. Ir(ppy)₃
4. MEH-PPV
-

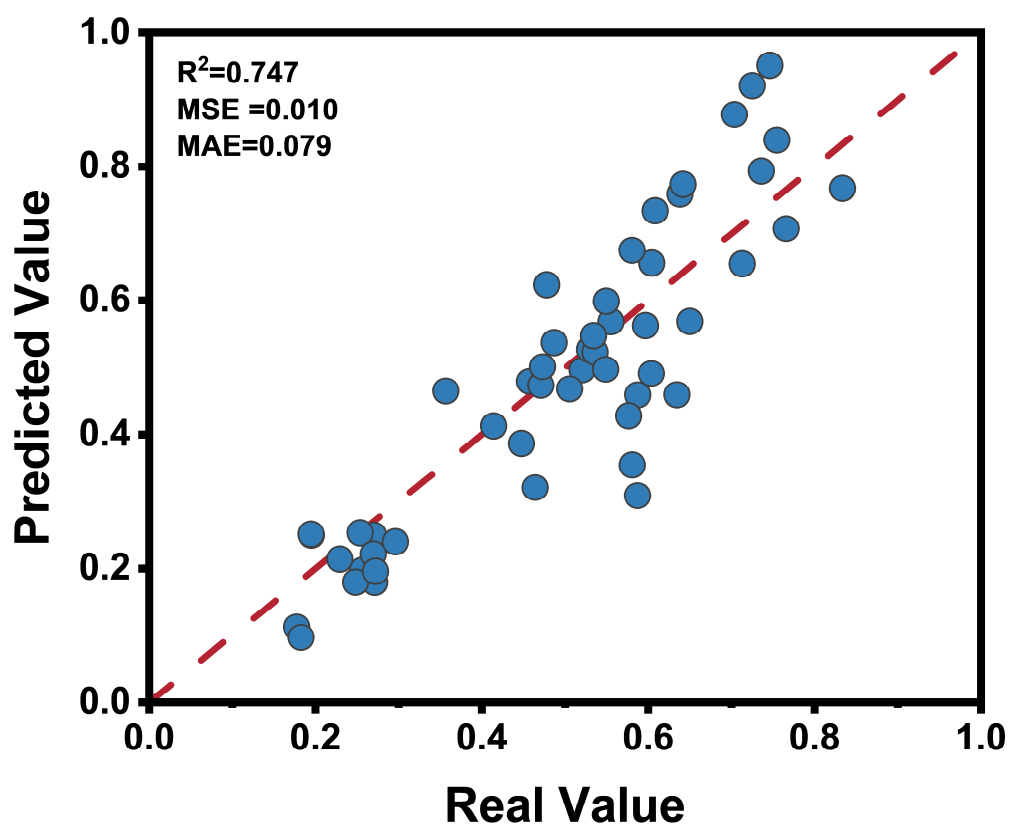


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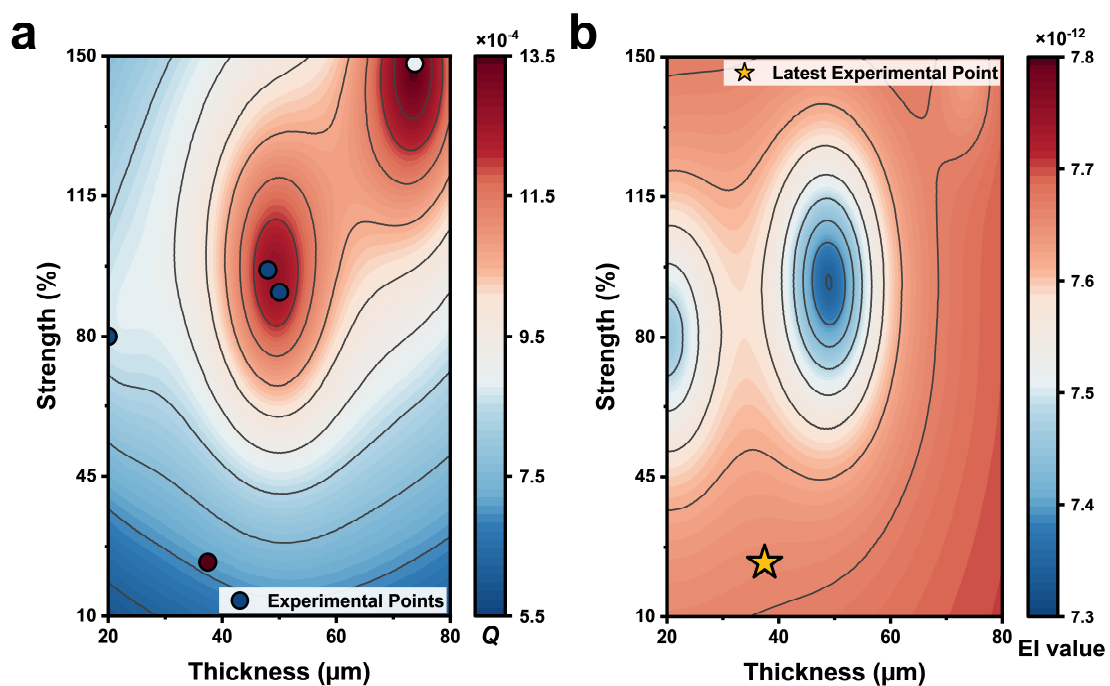
Supplementary Figure 15 | Schematic diagram of fluorescent molecules recommended by the large language models.



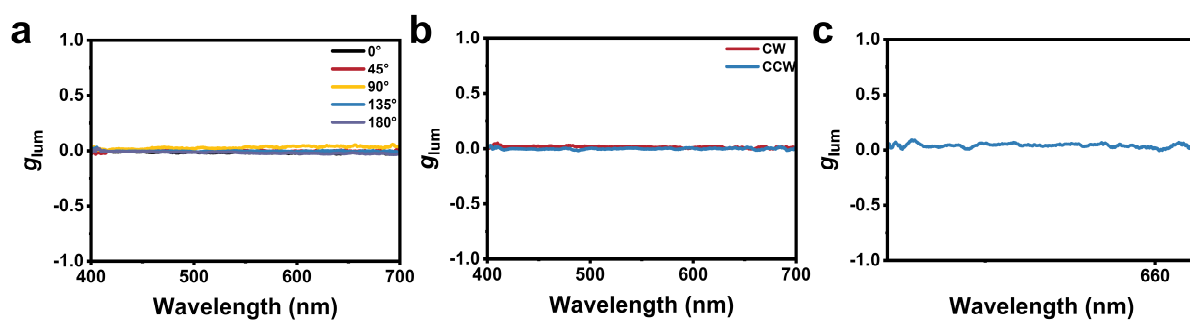
Supplementary Figure 16 | Iterations of High-Performance WCPL generation in the Quaternary Systems. (a) WAE latent space and (b) GMM-modeled density of the different iteration stages. The colors of the data points in the latent space denote their corresponding G . The new candidates are marked by crosses, and the best ranked compositions are marked by circles.



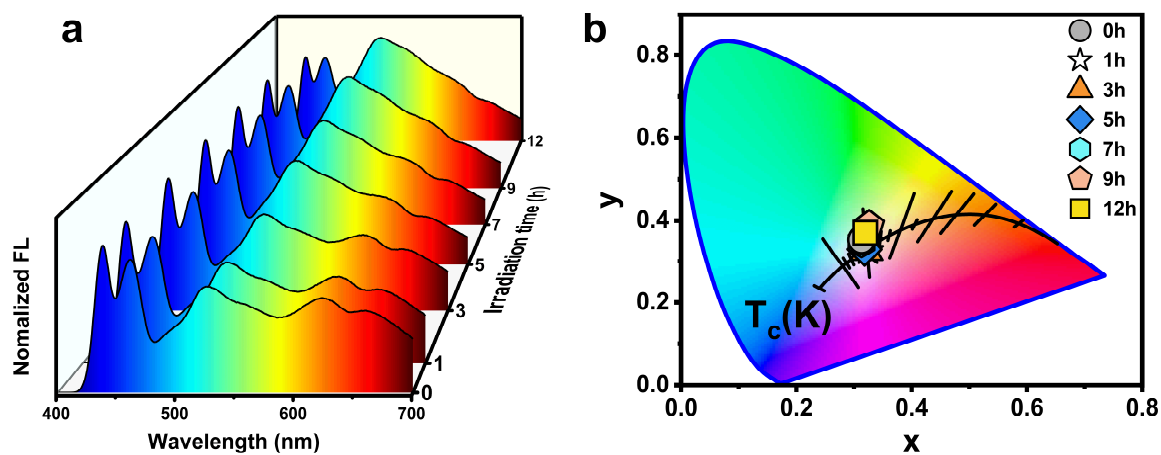
Supplementary Figure 17 | Fitting performance of the TabPFN model in Quaternary systems. Scatter plots of experimental and predicted results by the TabPFN model.



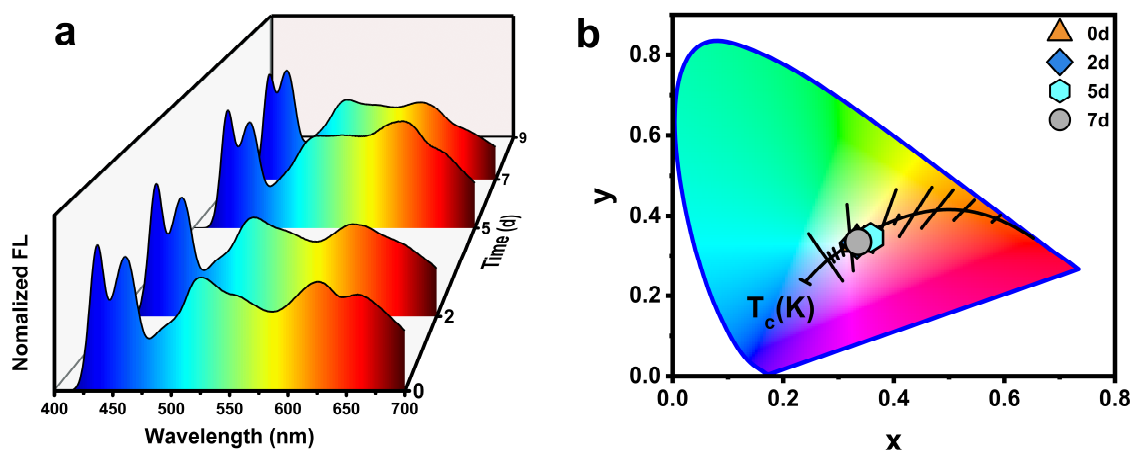
Supplementary Figure 18 | Visualization of 2D Parameter Space in Bayesian Optimization for Quaternary Systems. **a** Gaussian-process-predicted mean objective Q across film thickness-strength space, with contour lines and coloured circles for sampled experimental points. **b** Heatmap of expected improvement (EI) acquisition function; yellow star marks the latest proposed experimental point.



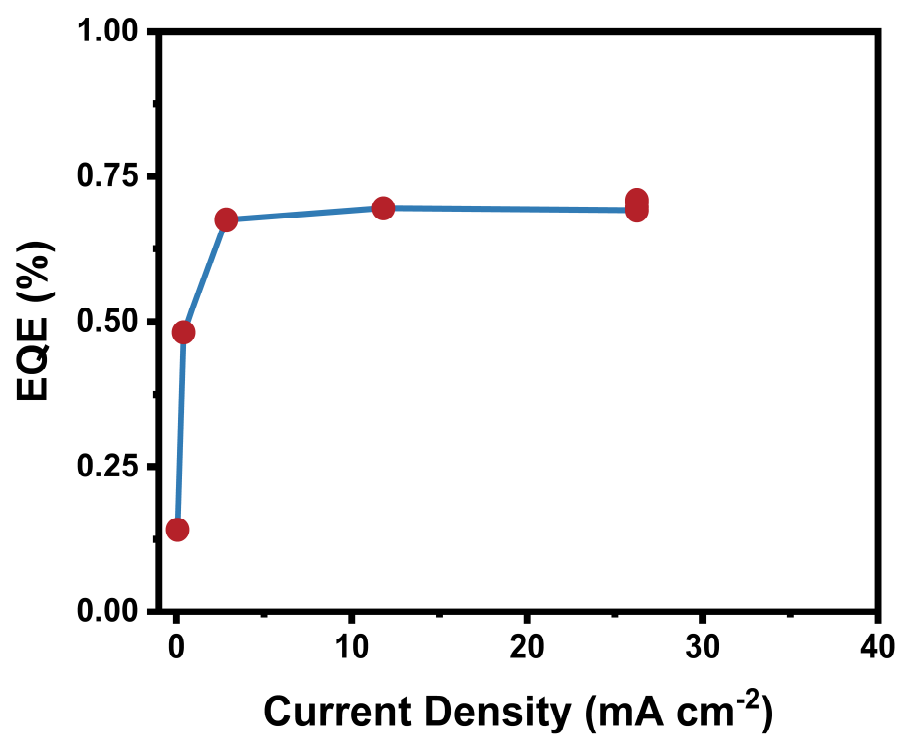
Supplementary Figure 19 | The variation of CPL spectra of chiroptical films with different parameters. The g_{lum} value of (a) the emissive layer alone, (b) the emissive layer combined with an unstretched (isotropic) PVA layer and (c) the emissive layer combined with a stretched PVA layer but at a twist angle of 0°.



Supplementary Figure 20 | Long-term UV irradiation photostability of the WCPL conversion film under continuous UV-LED excitation for 12 h. Time-dependent evolution of (a) normalized PL emission intensity and (b) CIE 1931 chromaticity coordinates of the film recorded at irradiation durations of 0,1,3,5,7,9, and 12 h.



Supplementary Figure 21 | Environmental stability of the WCPL conversion film. (a) PL spectra and (b) CIE chromaticity coordinates of samples stored under ambient conditions at room temperature for different durations.



Supplementary Figure 22 | The performance of the CPW-PL device. EQE vs current density curves.

Supplementary Table 1 | A comparison of the $|g_{lum}|$ and M values of representative WCPL materials and complexes documented in the literature and this study.

<i>Category</i>	<i>g_{lum}</i>	<i>Ra</i>	<i>M</i>	<i>Ref</i>
<i>Chiral organic-inorganic hybrid</i>	8×10^{-3}	93.4	7.5×10^{-3}	1
	1.56×10^{-2}	96.9	1.5×10^{-2}	2
	0.01	91	9.1×10^{-3}	3
	1.64×10^{-2}	81.2	1.3×10^{-2}	4
<i>Chiral polymer</i>	6.2×10^{-2}	98	6.1×10^{-2}	5
	1.4×10^{-2}	89	1.2×10^{-2}	6
	2.19×10^{-2}	97	2.1×10^{-2}	7
	1.8	90	1.62	<i>This work</i>
<i>Chiral liquid crystals</i>	1.88	82	1.5	8
	1	83	0.83	9
	0.46	94	0.43	10
<i>Chiral inorganic</i>	1.8×10^{-3}	81	1.5×10^{-3}	11
	0.01	88	8.8×10^{-3}	12
	2.32×10^{-2}	78	1.8×10^{-2}	13
	5×10^{-4}	89	4.5×10^{-4}	14
<i>Chiral organic molecules</i>	5×10^{-3}	60	3×10^{-3}	15
	3×10^{-3}	73	2.2×10^{-3}	16

Supplementary Table 2 | The composition and performance of the samples for ternary systems that were tested during the optimization process. The formulations are ranked from best to worst according to their performance in the optimization experiments.

Formulation			Performance			
PFO	F8BT	PFO-DBT	x	CIE y	CCT	Ra
80	0.3	7	0.36	0.35	4528	87
80	0.5	10	0.37	0.36	4262	88
90.4	0.7	9.6	0.37	0.35	4124	84
90.3	1	9.7	0.37	0.38	4209	89
89.9	1	10.1	0.38	0.38	4085	89
80	0.5	7	0.35	0.39	4977	88
92.7	0.8	7.3	0.36	0.39	4595	90
91.1	0.8	8.9	0.38	0.39	4196	90
12	0.3	3	0.38	0.36	3759	82
91.9	1	8.1	0.37	0.40	4424	91
92.3	1	7.7	0.37	0.40	4561	89
87.9	0.3	12.1	0.36	0.31	4111	71
18	0.3	5	0.41	0.36	3216	87
18	0.3	7	0.40	0.35	3135	82
15	0.3	7	0.41	0.35	2896	82
12	0.3	5	0.39	0.32	3205	69
18	0.3	9	0.40	0.31	2894	72
88.6	0.5	11.4	0.37	0.30	3377	65
15	0.6	9	0.42	0.35	2845	79
15	0.6	7	0.42	0.37	2932	84
12	0.3	1	0.38	0.42	4338	85
15	0.3	9	0.41	0.32	2726	72
18	0.9	3	0.41	0.41	3587	87
3	0.9	9	0.39	0.30	2763	69
6	0.3	3	0.40	0.34	3151	67
48	1	10	0.42	0.41	3405	86
15	0.9	5	0.43	0.40	3081	86

18	0.3	1	0.37	0.43	4498	80
15	0.9	7	0.42	0.37	2930	78
12	0.6	3	0.43	0.40	3110	85
18	0.6	3	0.43	0.40	3090	85
18	0.6	9	0.44	0.36	2616	82
18	1.2	5	0.42	0.39	3170	80
18	0.6	7	0.44	0.36	2655	81
18	0.9	5	0.41	0.38	3187	75
18	1.2	7	0.42	0.38	3042	75
15	0.9	9	0.43	0.36	2742	76
9	0.3	3	0.44	0.37	2740	79
12	0.3	9	0.39	0.30	2824	61
78.2	3	18.9	0.42	0.42	3385	85
12	0.6	9	0.41	0.32	2768	63
15	1.5	9	0.44	0.38	2826	80
38.1	1.2	10.7	0.43	0.39	2948	78
12	0.6	5	0.42	0.36	2988	66
12	0.9	7	0.43	0.36	2798	71
18	1.2	9	0.42	0.36	2861	69
3	0.9	5	0.46	0.38	2516	87
12	0.3	7	0.42	0.32	2510	66
75.1	3.2	21.1	0.43	0.41	3103	81
12	0.9	9	0.43	0.35	2655	70
87	1.9	11.1	0.40	0.45	3955	86
12	1.2	9	0.42	0.35	2889	65
15	1.5	7	0.45	0.41	2888	85
3	1.2	9	0.43	0.32	2319	69
3	0.9	7	0.45	0.36	2361	80
12	0.6	7	0.44	0.35	2520	72
3	0.3	1	0.45	0.42	3010	88
18	1.5	3	0.43	0.44	3409	87
18	1.5	5	0.43	0.39	2972	76
3	0.3	3	0.44	0.36	2608	72

12	0.9	5	0.44	0.39	2751	77
18	0.9	9	0.42	0.34	2679	62
6	0.3	5	0.41	0.31	2672	57
6	0.9	5	0.42	0.36	2907	65
15	0.3	1	0.38	0.45	4413	78
9	0.3	5	0.44	0.34	2405	68
6	0.6	1	0.43	0.45	3526	87
15	1.2	3	0.42	0.45	3570	87
6	0.6	3	0.44	0.39	2858	75
12	1.2	5	0.45	0.40	2727	80
62.1	2.5	35.4	0.43	0.35	2613	65
12	1.2	7	0.45	0.38	2603	76
3	1.2	7	0.47	0.38	2385	82
3	0.6	3	0.45	0.38	2658	75
9	0.3	7	0.44	0.33	2244	67
18	1.5	7	0.44	0.38	2757	70
9	0.9	3	0.45	0.40	2743	78
9	0.6	3	0.46	0.40	2557	81
18	1.5	9	0.44	0.38	2671	69
3	0.6	1	0.45	0.45	3165	87
18	0.9	7	0.44	0.36	2569	64
3	1.2	5	0.48	0.40	2410	86
6	0.6	5	0.43	0.34	2511	59
3	0.3	5	0.42	0.30	2371	54
12	0.6	1	0.41	0.46	3837	82
9	0.6	5	0.46	0.38	2413	73
3	0.6	5	0.45	0.36	2344	68
6	0.9	1	0.44	0.46	3290	86
12	1.5	5	0.46	0.40	2638	73
12	1.5	3	0.46	0.43	2864	81
9	0.9	5	0.46	0.38	2496	71
9	0.9	1	0.44	0.47	3381	87
9	0.3	9	0.44	0.31	2080	60

9	1.2	9	0.42	0.33	2525	52
12	1.5	7	0.46	0.38	2480	68
6	1.2	3	0.47	0.42	2644	81
12	0.9	1	0.41	0.48	3907	81
6	0.6	7	0.43	0.31	2333	51
9	1.2	3	0.47	0.41	2592	77
9	1.2	1	0.45	0.47	3343	86
9	1.5	3	0.47	0.43	2745	79
6	1.2	5	0.47	0.38	2331	70
9	0.9	9	0.45	0.33	2150	59
18	0.6	1	0.40	0.48	4157	75
18	0.9	1	0.39	0.48	4339	75
18	1.2	1	0.39	0.48	4348	75
6	0.9	7	0.44	0.33	2238	54
3	0.3	7	0.44	0.30	2039	53
12	1.5	1	0.43	0.48	3622	83
15	0.6	1	0.39	0.48	4313	73
15	0.9	1	0.40	0.48	4108	76
3	0.6	7	0.47	0.34	2064	63
6	1.2	1	0.47	0.46	2869	87
9	1.5	5	0.47	0.39	2497	68
9	1.5	1	0.44	0.48	3546	84
3	0.9	1	0.47	0.47	3019	87
12	1.2	1	0.43	0.48	3737	80
9	0.6	7	0.48	0.35	2000	66
9	0.6	9	0.46	0.32	1978	58
6	0.9	3	0.47	0.40	2481	68
6	1.5	1	0.47	0.47	2899	87
9	1.2	5	0.48	0.39	2284	70
6	0.3	7	0.38	0.26	2669	38
3	0.3	9	0.45	0.30	1952	53
6	1.2	9	0.46	0.34	2066	58
12	1.5	9	0.46	0.36	2235	59

9	1.5	7	0.47	0.37	2290	62
6	1.5	3	0.49	0.42	2335	78
6	0.9	9	0.43	0.30	2120	45
6	1.5	7	0.49	0.37	2089	66
3	1.5	1	0.50	0.45	2547	84
18	1.5	1	0.40	0.50	4161	74
9	0.9	7	0.48	0.36	2049	61
6	1.2	7	0.48	0.35	2024	61
3	1.5	5	0.49	0.37	2050	63
9	1.2	7	0.48	0.36	2047	61
15	1.5	1	0.40	0.50	4160	72
6	1.5	5	0.50	0.38	2053	69
15	1.2	1	0.41	0.50	4006	73
6	1.5	9	0.48	0.34	1918	58
3	1.5	3	0.50	0.40	2158	71
9	1.5	9	0.47	0.34	2007	54
3	1.5	9	0.42	0.27	1966	39
6	0.6	9	0.41	0.26	2016	31
3	1.5	7	0.51	0.36	1775	60
6	0.3	9	0.39	0.24	2158	25
3	0.6	9	0.35	0.22	2889	25

Note: All values in the formulation are given as relative volume parts, defining the volume ratio of each component in the blend.

Supplementary Table 3 | The composition and performance of the samples for quaternary systems that were tested during the optimization process. The formulations are ranked from best to worst according to their performance in the optimization experiments.

Formulation				Performance			
PFO	F8BT	PFO-DBT	RUB	CIE		CCT	Ra
				x	y		
85	0.5	10	8	0.33	0.33	5720	90
85	0.5	12	10	0.33	0.33	5482	84
75	0.5	10	5	0.35	0.36	4938	91
80	0.5	12	8	0.35	0.34	4696	86
82	0.5	15	10	0.34	0.33	5017	82
82.4	0.5	12.2	5.4	0.35	0.35	4830	86
70	0.5	14	10	0.35	0.34	4773	84
72	0.5	12	8	0.35	0.34	4733	85
80	0.5	15	10	0.35	0.34	4692	85
82	0.5	10	10	0.34	0.37	5280	89
80	0.5	10	8	0.36	0.36	4580	89
20	0.3	2	1	0.36	0.38	4682	91
20	0.5	2	2	0.35	0.38	4860	89
75	0.5	15	10	0.36	0.35	4304	82
18	0.3	3	2	0.35	0.32	4648	75
21	0.3	2	2	0.35	0.38	5002	88
83.8	0.5	12.5	3.7	0.35	0.32	4474	75
42	1	10	2	0.36	0.38	4485	90
70	0.5	16	10	0.36	0.33	4241	77
20	0.3	4	2	0.33	0.30	5368	74
70	0.5	16	16	0.37	0.37	4131	86
12	0.3	3	2	0.34	0.31	5021	70
80	0.5	18	10	0.36	0.32	4200	75
65	0.5	15	10	0.38	0.36	3989	82
86.5	0.5	6.3	7.2	0.33	0.39	5505	83
37	1	8.8	4.1	0.39	0.37	3806	87
37	1	8.4	3.8	0.39	0.38	3835	88
18	0.3	2	7	0.33	0.39	5663	80

18	0.3	3	5	0.38	0.33	3595	78
31	1	10	16	0.38	0.39	4255	89
23	0.5	2	2	0.40	0.38	3514	90
25	0.5	2	2	0.35	0.40	5037	84
36	1	11	1.9	0.39	0.36	3605	82
73.1	0.5	17.9	9	0.37	0.32	3894	69
25	0.2	1	1	0.34	0.39	5391	78
18	0.3	3	3	0.32	0.29	6547	69
25.6	1.8	13.6	8.9	0.40	0.38	3475	88
38	1	4.6	4.9	0.38	0.41	4189	89
37	1	3.7	8.4	0.37	0.40	4555	84
12	0.3	3	5	0.39	0.33	3392	74
70	0.5	14	8	0.40	0.36	3446	78
12	0.3	3	3	0.34	0.30	5143	63
37	1	4.4	7.4	0.38	0.41	4208	87
40	1	3.7	4.9	0.37	0.41	4411	85
40	1	7.7	1.9	0.37	0.42	4574	85
38	1	4.2	7.4	0.38	0.42	4301	86
14.7	1	19.8	14.6	0.40	0.33	2977	74
42	1	4.5	2.6	0.39	0.42	4150	88
25	0.3	1	1	0.33	0.41	5737	72
27	1.5	8	10	0.42	0.40	3385	88
26.3	2.3	18.8	2.5	0.42	0.38	3053	84
20	1	2	2	0.39	0.43	4119	87
23	0.5	1.5	2	0.36	0.43	4745	80
37	1	2.7	9.4	0.37	0.43	4599	80
26.2	2	10.3	11.4	0.42	0.41	3448	87
32	1	6	10	0.37	0.43	4569	80
83	0.5	5.9	11.1	0.34	0.43	5244	74
40	1.3	3.7	4.9	0.38	0.44	4327	82
14.3	1.3	8	30	0.42	0.43	3541	88
20	0.3	1	1.5	0.33	0.42	5554	70
38	1	5	5	0.37	0.44	4519	80

33	1	3.5	12.8	0.38	0.44	4382	81
17.5	2.8	13.3	16.3	0.44	0.41	2967	88
12.4	1.6	8	33	0.44	0.42	3012	91
12	0.4	2	7	0.40	0.44	4020	85
32	1	5.6	11.2	0.38	0.44	4488	78
17.5	2.5	13.3	16.3	0.44	0.40	2860	81
20	0.5	1	1	0.35	0.44	4984	73
25.5	2.5	17.4	4.6	0.45	0.39	2654	81
17.5	2.5	7	16	0.44	0.45	3258	88
38	1	4.6	6.8	0.36	0.45	4838	73
30	1	3.2	16.5	0.35	0.45	5210	66
20.8	6	20.9	2.2	0.47	0.41	2525	82
23.3	7.3	14.3	5.1	0.48	0.43	2567	85
9.4	7.8	18.7	14.1	0.49	0.40	2292	77
15.5	9.2	18.8	6.5	0.49	0.42	2369	80
12.4	1.6	2	33.8	0.39	0.47	4235	62
12.4	1.6	2.2	33.8	0.55	0.40	1722	90
14.1	15.2	13.7	7	0.50	0.41	2199	73
12.6	11	3.5	22.9	0.52	0.45	2268	85
10.6	3.2	18.6	17.6	0.53	0.36	1690	74
16.7	16.3	14.1	3	0.51	0.42	2220	75
7	14.2	10.8	18	0.52	0.39	1895	71
10.7	1.3	18.9	19.1	0.55	0.35	1480	75
15.2	2.3	7.1	25.4	0.57	0.36	1407	81
7.2	16.6	10.9	15.3	0.54	0.40	1841	68
5.6	10.1	17.3	16.9	0.55	0.38	1617	73
14.3	1.3	1	34	0.38	0.52	4574	54
8	14.3	23.3	4.4	0.53	0.36	1622	55
10.4	3.2	16.4	19.9	0.59	0.35	1248	79
3.8	9.6	28.6	7.9	0.54	0.35	1534	49
2	12	24.2	11.8	0.55	0.35	1482	50

Note: All values in the formulation are given as relative volume parts, defining the volume ratio of each component in the blend.

Supplementary references

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