Supporting Information

Simplifying Complexity: Integrating Color Science for Predictable Full-color and On-demand Persistent Luminescence Using Industrial Disperse Dyes

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Materials and methods

Poly (tetramethylene ether glycol) (PTMG, Mn=2000 g/mol, J&K Chemicals) and Isophorone diisocyanate (IPDI, 98%, J&K Chemicals), dibutyltin dilaurate (DBTDL, 95%, Aladdin), N-methyl diethanolamine (MDEA, 99%, Aladdin), 1,4-butanediol (BDO, 99%, Macklin), and Tetraacetylethylenediamine (TAED, 97%, Aladdin), N, N'-diacetylethylenediamine (DAED, 97%, Macklin) were used as received. Other solvents were used as received without further purification except for specifying otherwise.
Fluorescence spectra, phosphorescence spectra, and ultralong lifetimes were performed on the Edinburgh FLS1000. Excitation-fluorescence mapping and excitation-phosphorescence mapping were measured using Hitachi F-4700. The X-ray photoelectron spectroscopy (XPS) was obtained using a Shimadzu AXIS SUPRA+ spectrometer. Powder X-Ray diffraction (PXRD) patterns was measured using a Bruker D8 Advance diffractometer. Fourier transform infrared spectra (FTIR) were recorded on the IRTracer-100. UV-vis spectra were measured on a UV-2600i UV-Vis spectrophotometer.

Synthetic procedures

Synthesis of Polyurethane (PU). PTMG (30 g, 0.015 mmol) dried in vacuum for 1 h at 70 °C was added to a four-necked round-bottom flask. IPDI was added drop-wise into the reactor allowing reaction at 85 °C with DBTDL as catalyst under water-free environment for 2 h. After cooling to 50 °C, MDEA (1.19 g, 0.01 mol) dissolved in acetone was dropped into the reactor and the mixture was stirred for 2 h at 50°C to obtain the prepolymer. Then, BDO (0.45 g, 0.005 mol) as chain extender was added to the prepolymer and the mixture was stirred for 2 h at 65 °C. The TAED doped polymer films were prepared according to the description in the literature[52]. The molecular structure formula of polyurethane is shown in Scheme S1.
Synthesis of PU-TAED Films and PU-TAED PU-TAED Balls. Taking PU@TAED@Dye-R-0.1% as an example: After the reaction was completed, TAED (2.94 g, 0.013 mol) dissolved in acetone was dropped into the reactor and the mixture was stirred for 2 h at 70 °C. Then, C.I. Disperse red 145 (0.1% of monomer mass) dissolved in acetone was dropped into the reactor to obtain TAED-doped polyurethane (PU@TAED@Dye-R-0.1%). The PU@TAED@Dye-R-0.1% polymer was washed in ethanol with ultrasonic for 1 min. After that, the product was dried in vacuum to give a red film. In addition, PU balls were prepared using two hemispherical polytetrafluoroethylene (PTEF) molds. PU@TAED@Dye-R-0.1% polymer was poured into the two hemispherical molds separately. They were dried at 50 °C until the solvent content reached about 5%. Then, by combining the two hemispheres together, they were dried at 50 °C for 2 hours to obtain PU balls.

References

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57. Zhou, Z., Ji, H. & Yang, X. Illumination correction of dyed fabric based on extreme learning machine with improved ant lion optimizer. *Color Res. Appl*., **47**, 1065–1077 (2022).
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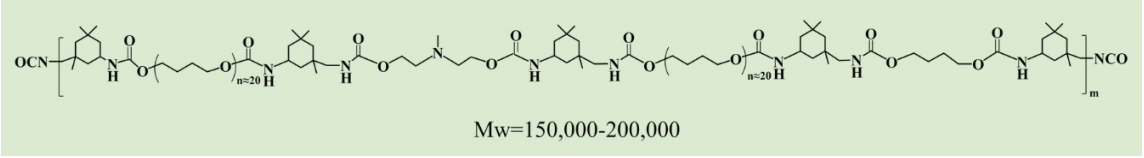


Scheme S1. The molecular structure formula and molecular weight of polyurethane (PU).

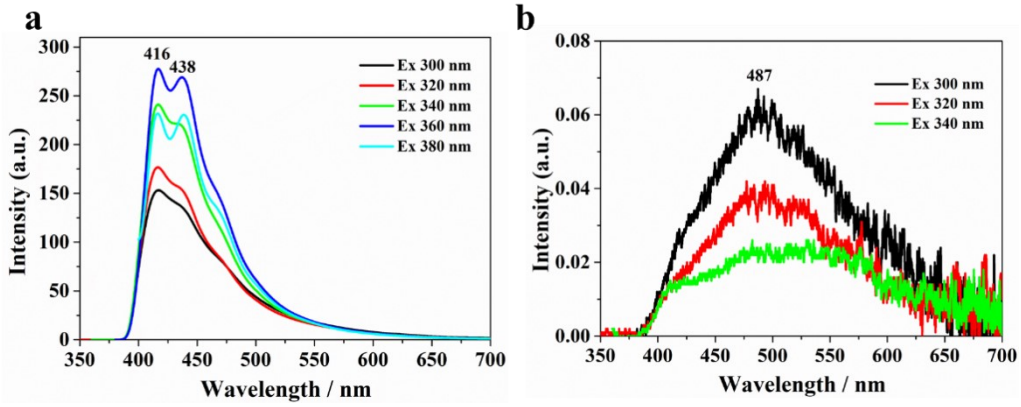
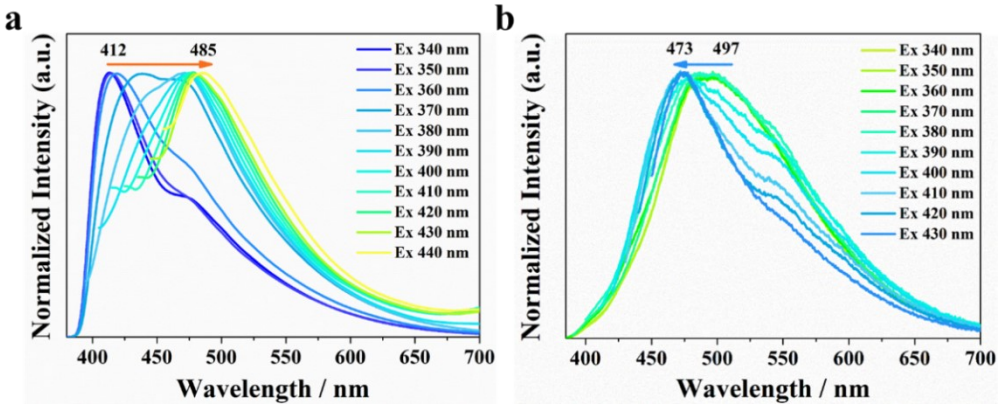


Figure S1. (a) Prompt PL emission spectra and (b) Delayed PL emission spectra of DAED with different excitation wavelengths.



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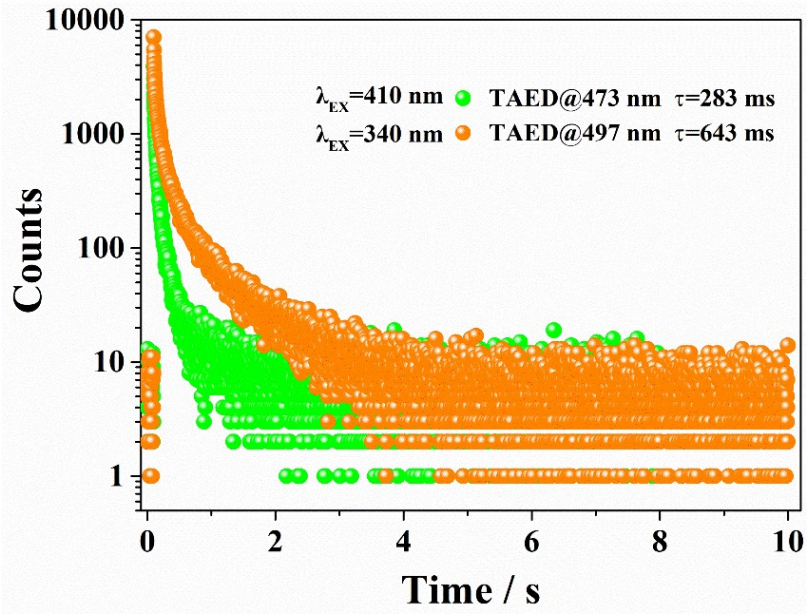

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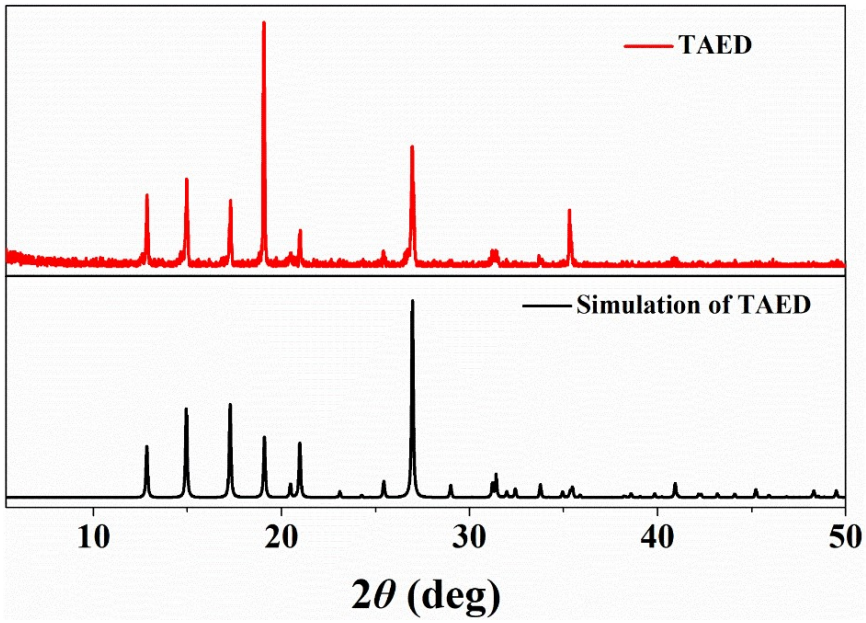

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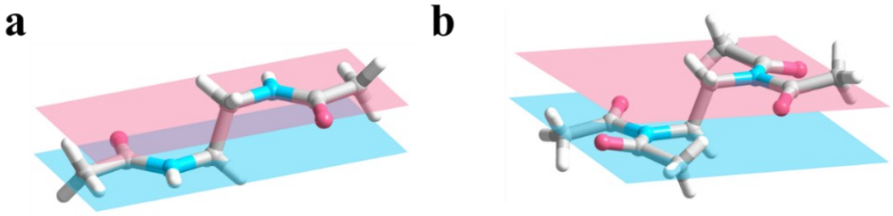


Figure S5. The crystal structures of DAED and TAED.

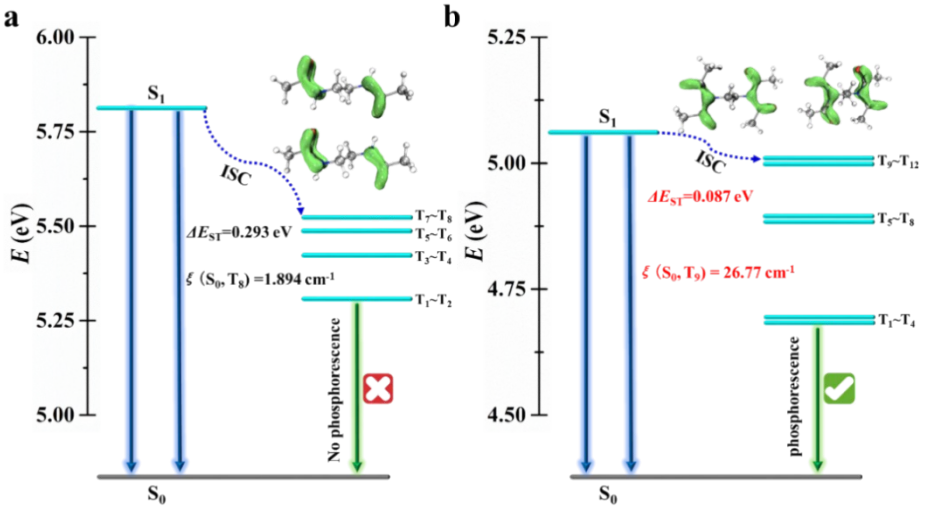
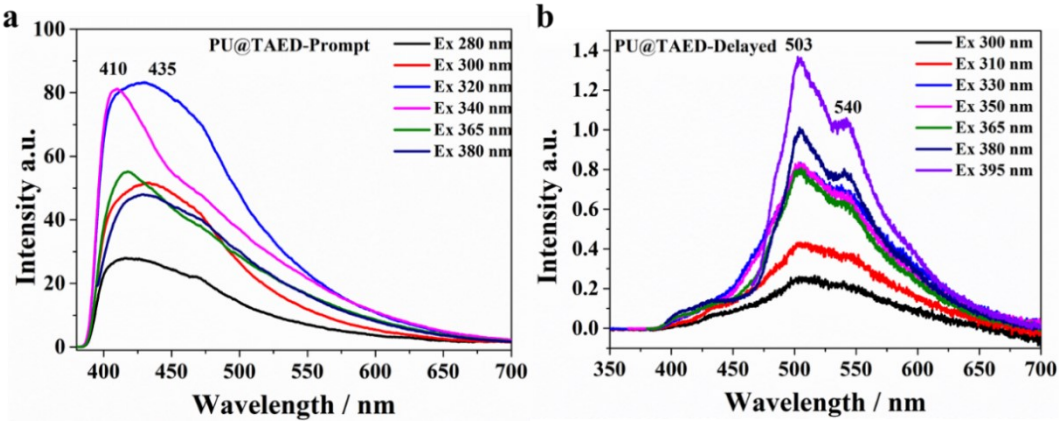


Figure S6. (a) Diagrams of the calculated energy levels and the possible ISC channels, as well as the SOC constants between singlet and triplet excited sates of DAED and TAED dimers, respectively. The insets are the dimers molecular conformations and π electron distribution map.



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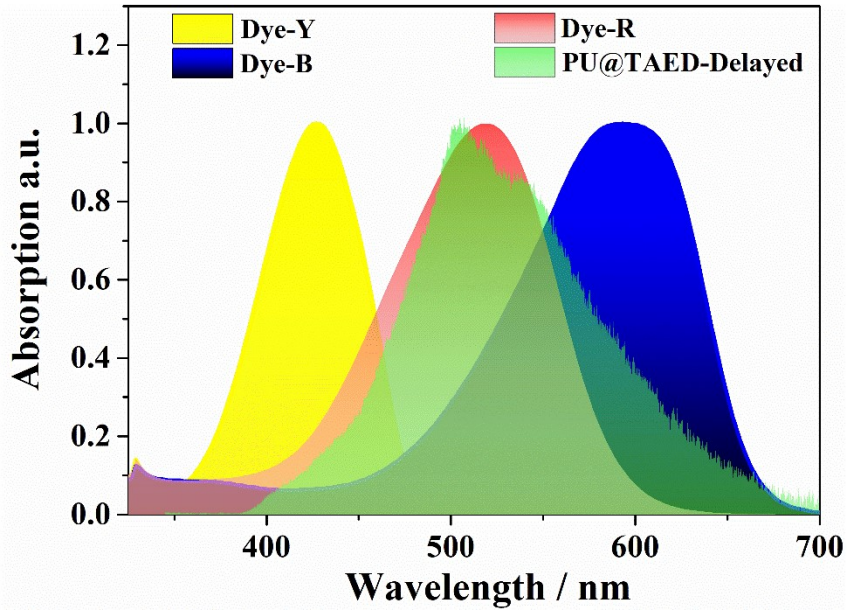

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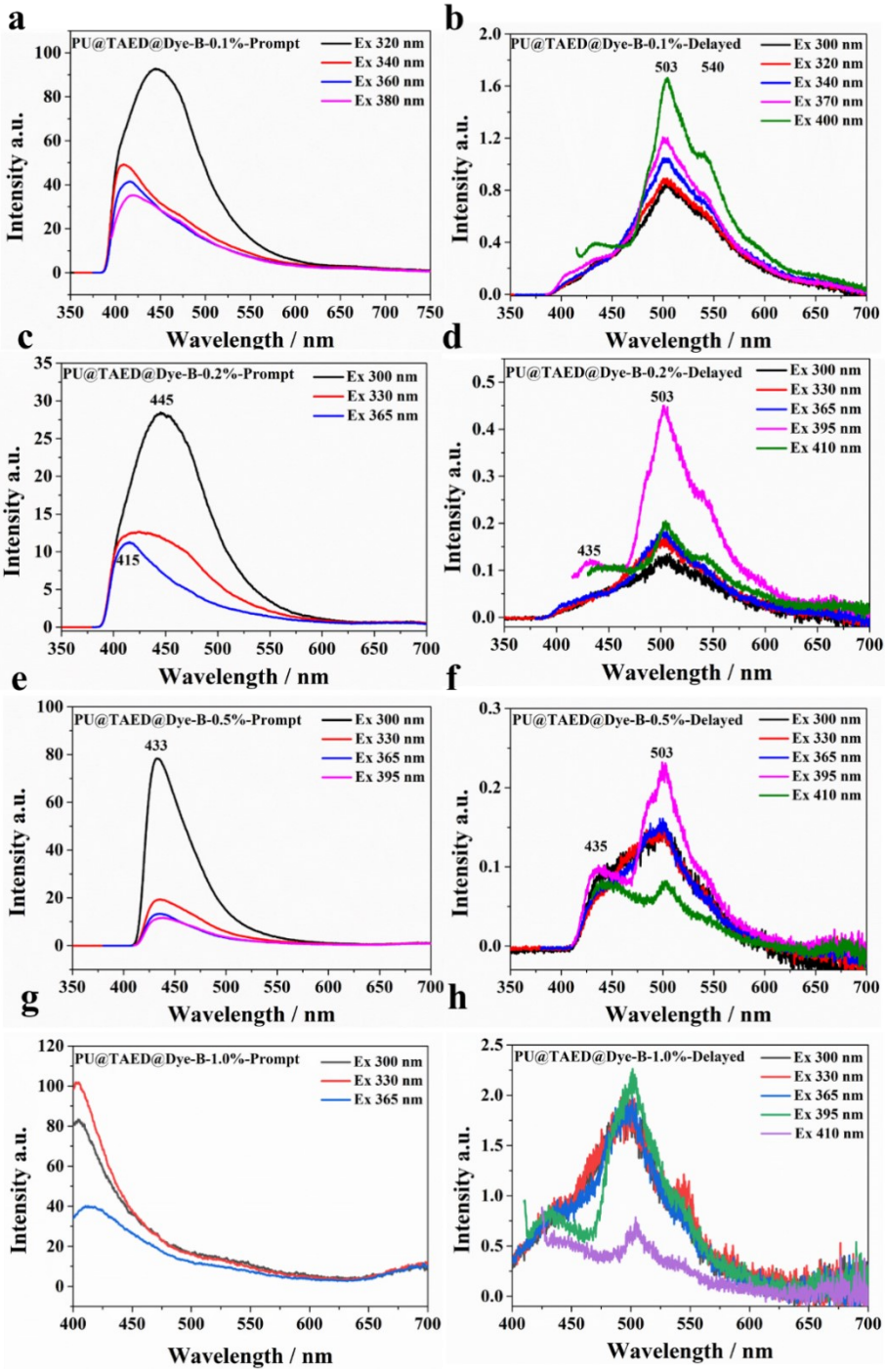

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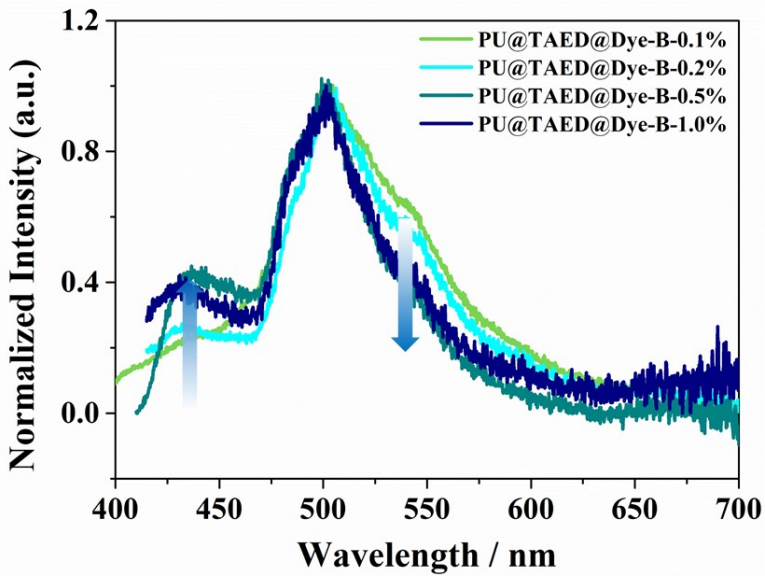

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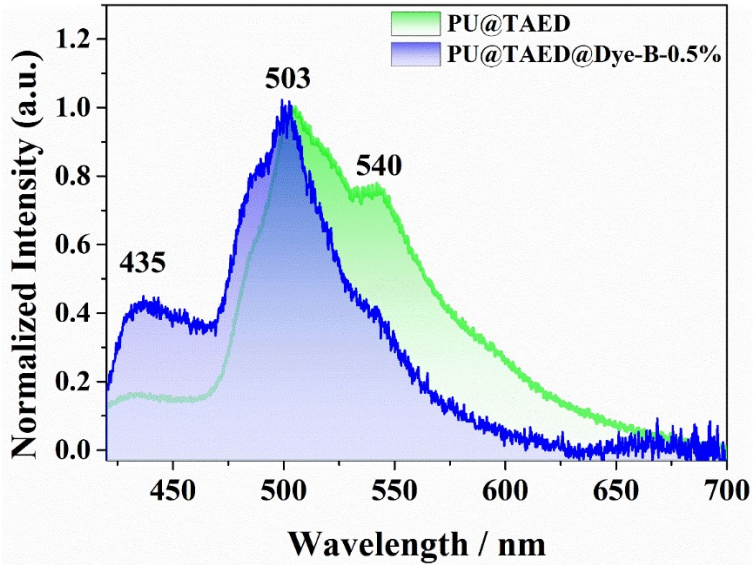

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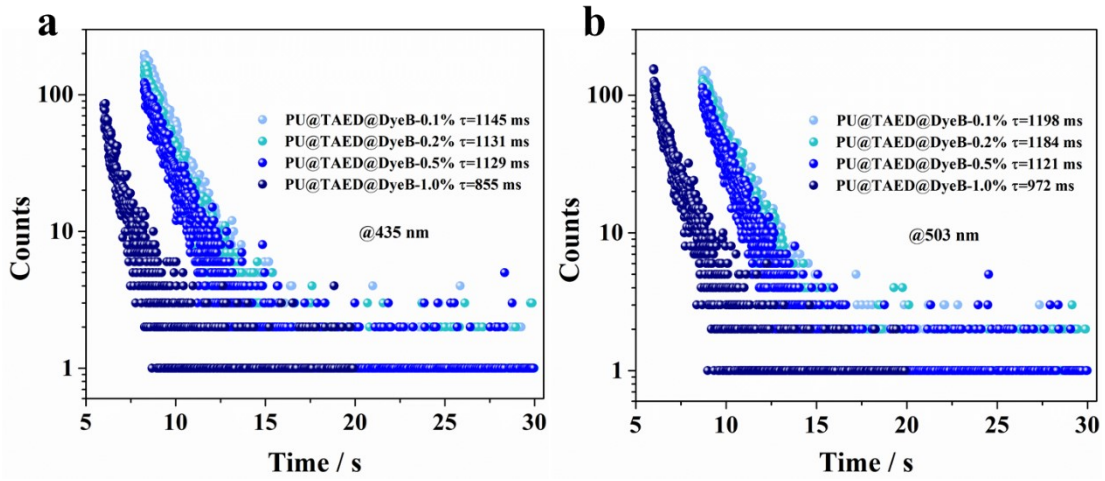
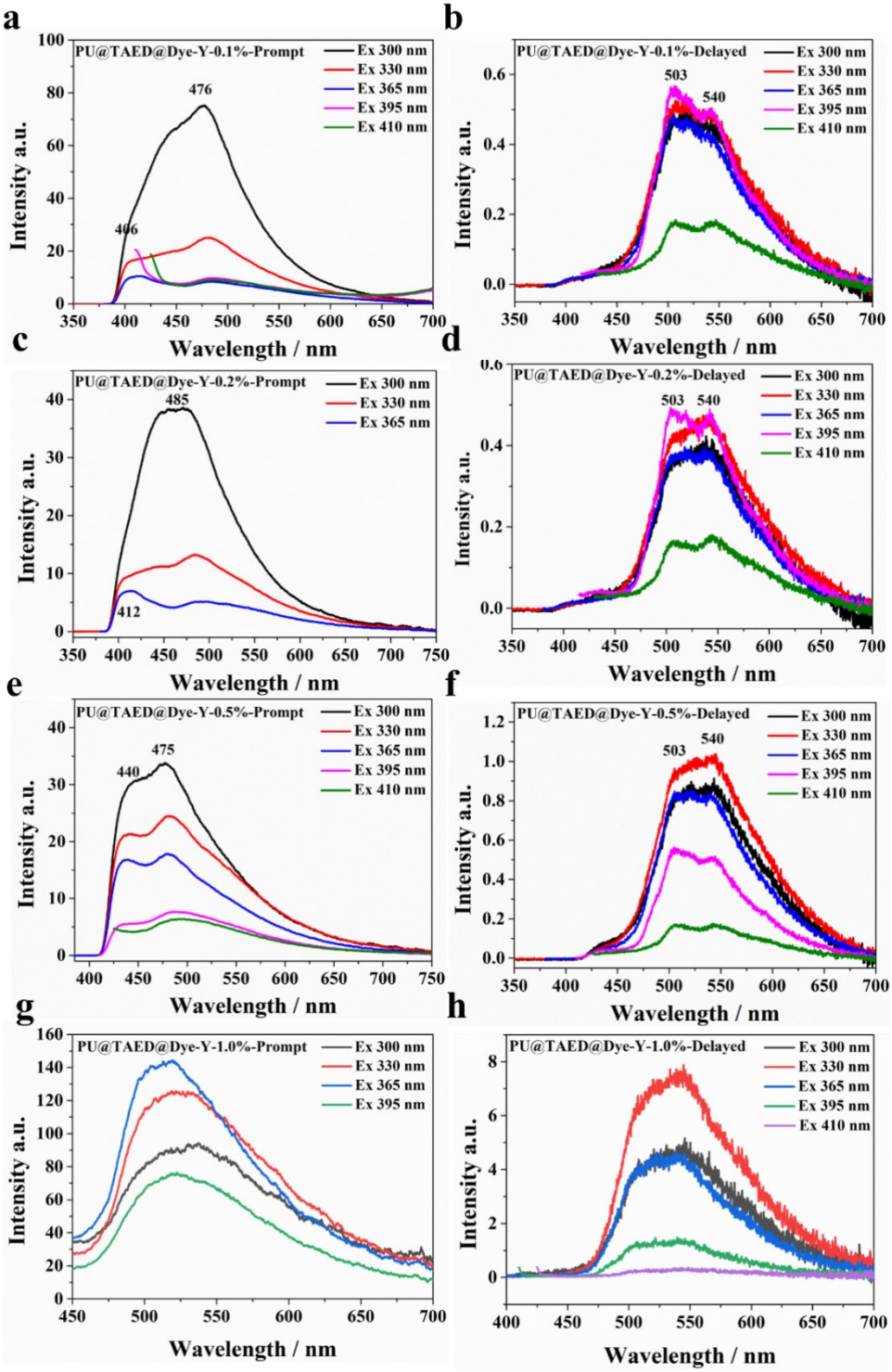


Figure S12. Decay curves obtained at room temperature for PU@TAED@Dye-B (0.1%, 0.2%, 0.5% and 1.0%)@435 nm and 503 nm.



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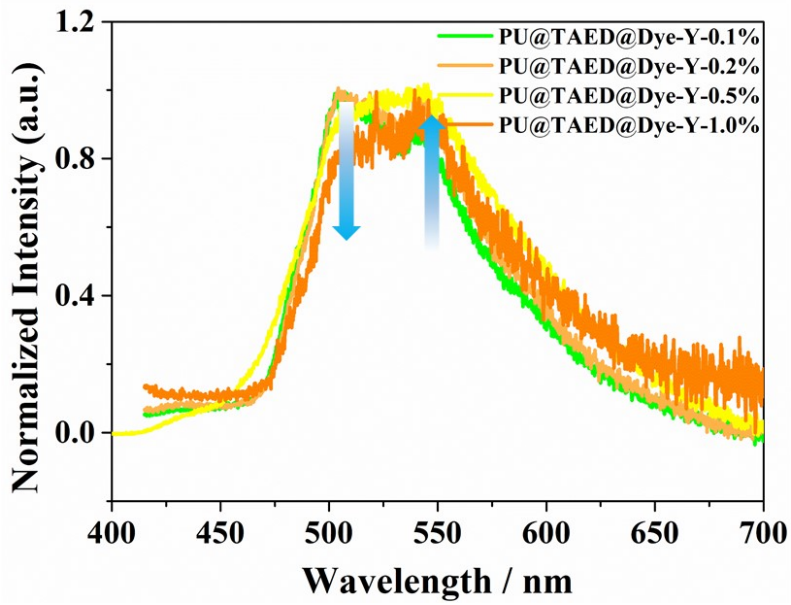

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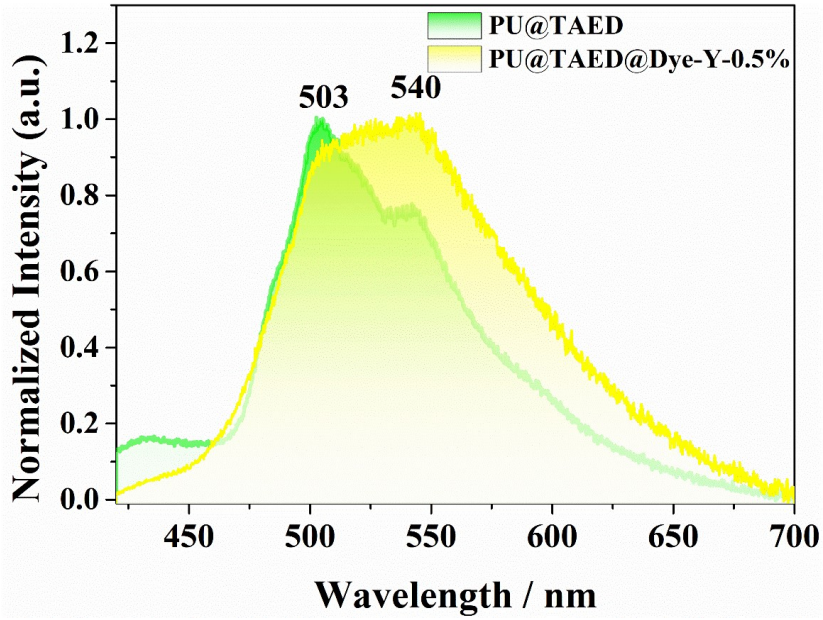
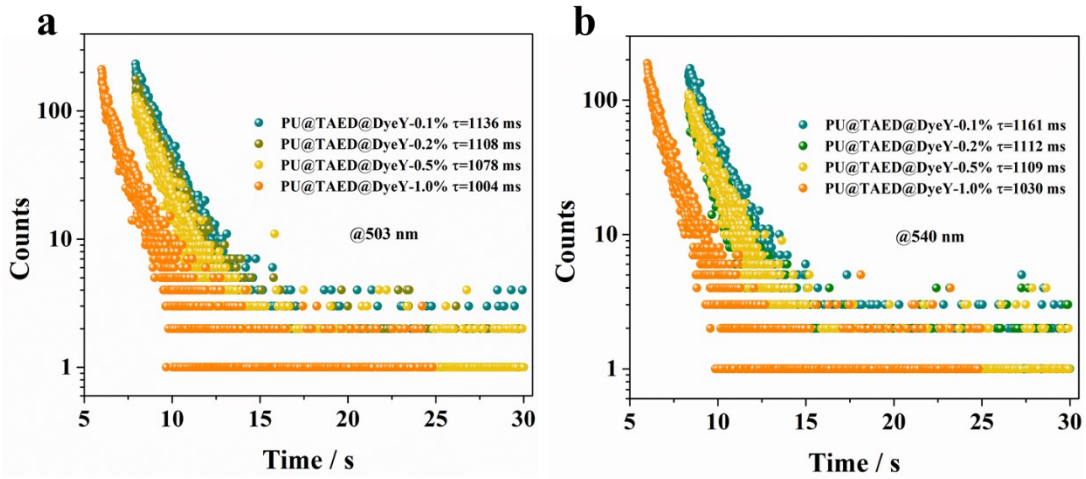


Figure S15. Delayed PL emission spectra of PU@TAED and PU@TAED@Dye-Y-0.5%.



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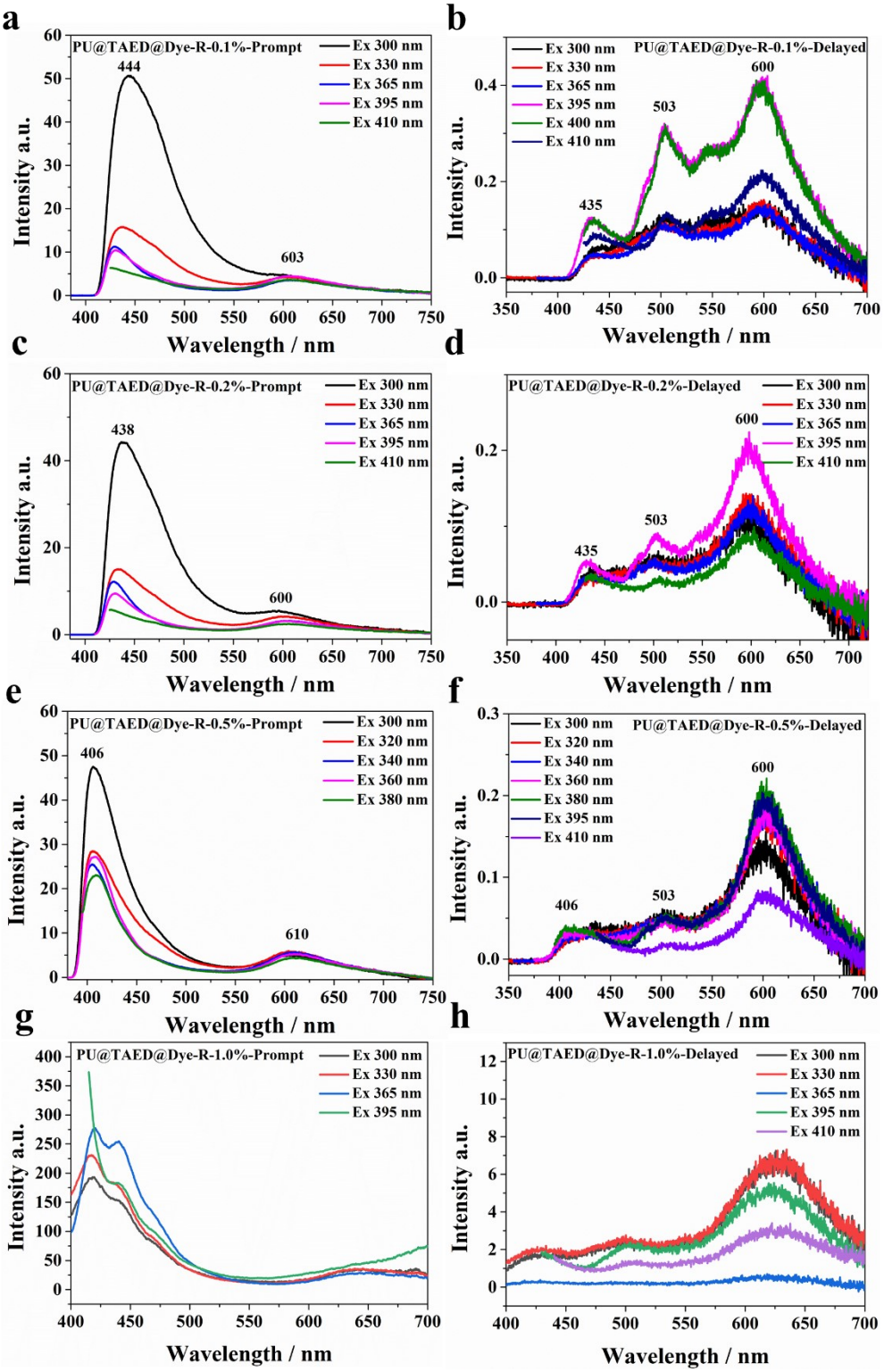
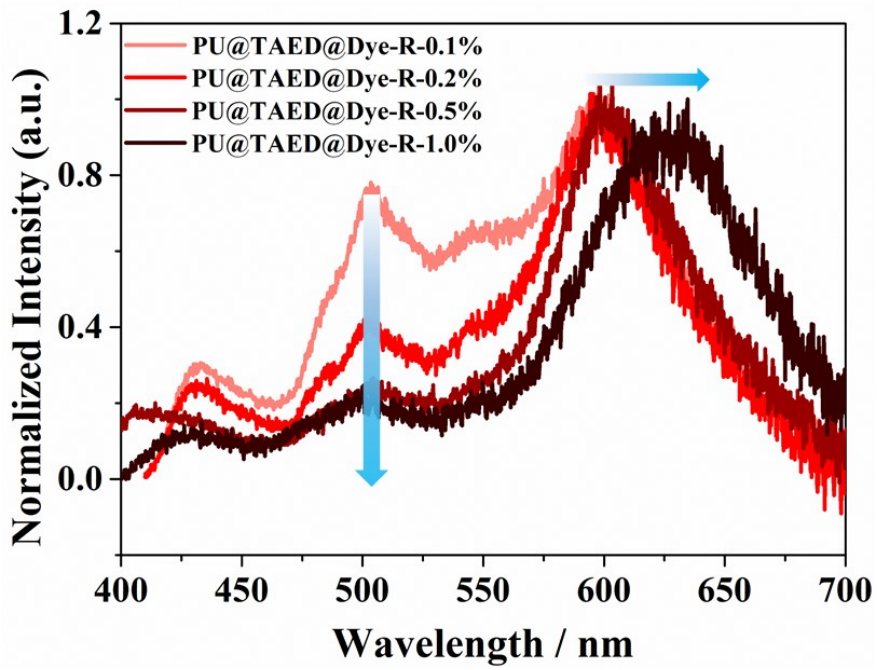


Figure S17. (a, c, e) Prompt and (b, d, f) delayed emission spectra of PU@TAED@Dye-R (0.1%, 0.2% ,0.5% and 1.0%) with different excitation wavelengths.



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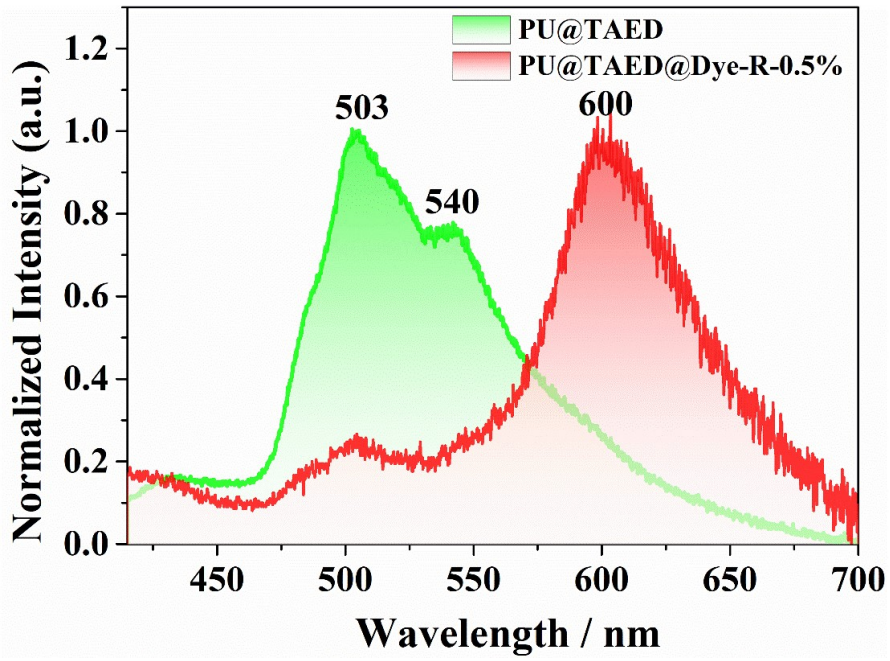

Figure S19. Delayed PL emission spectra of PU@TAED and PU@TAED@Dye-R-0.5%.

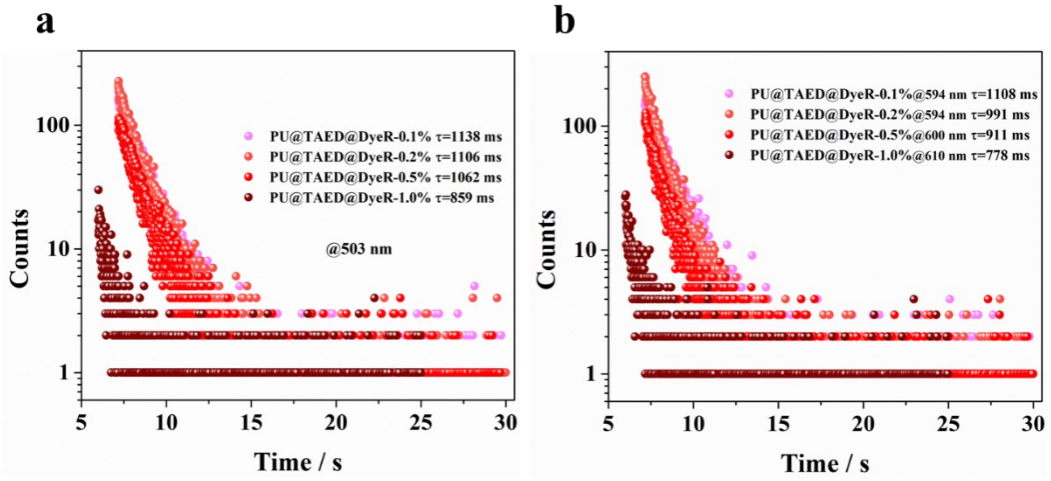

Figure S20. Decay curves obtained at room temperature for PU@TAED@Dye-R (0.1%, 0.2%, 0.5% and 1.0%)@503 nm and 594, 600, 610 nm.

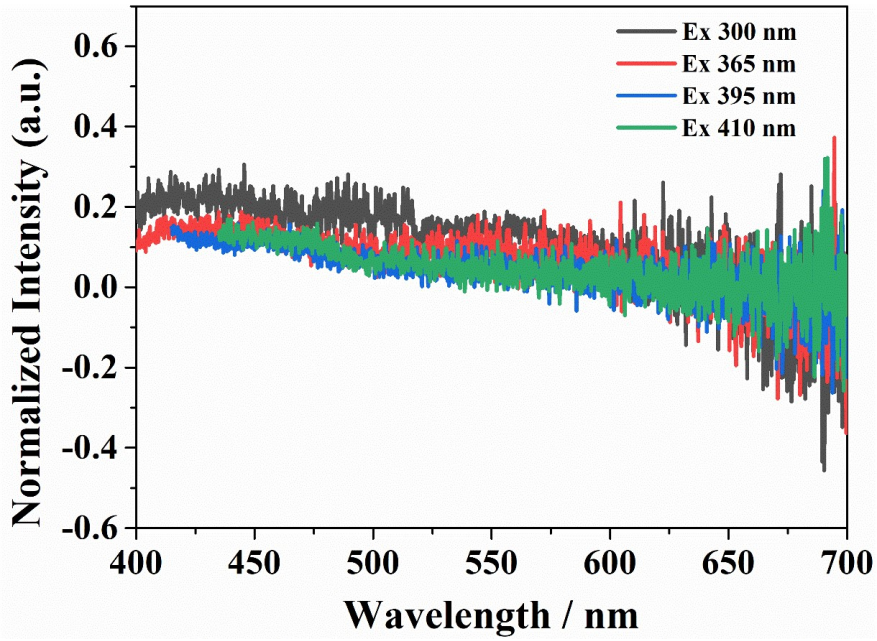

Figure S21. Delayed PL emission spectra of PU.

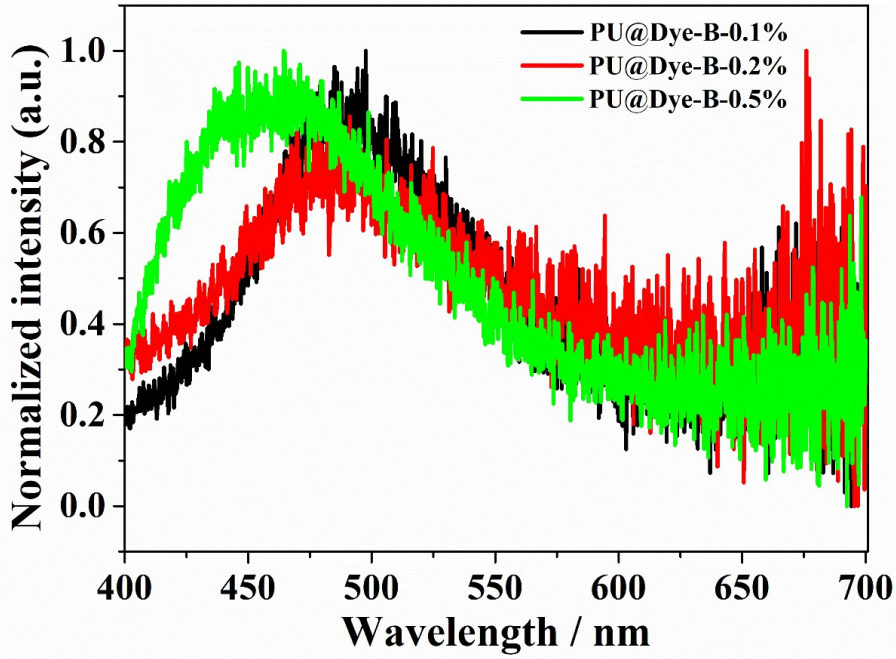

Figure S22. Delayed PL emission spectra of PU@Dye-B-0.1%, PU@Dye-B-0.2% and PU@Dye-B-0.5%.

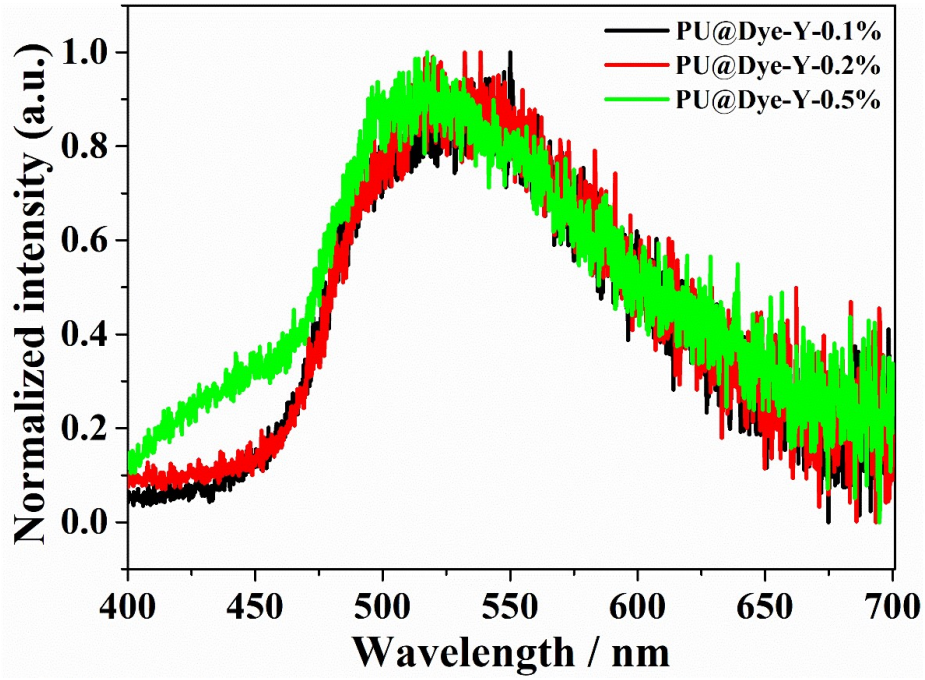

Figure S23. Delayed PL emission spectra of PU@Dye-Y-0.1%, PU@Dye-Y-0.2% and PU@Dye-Y-0.5%.

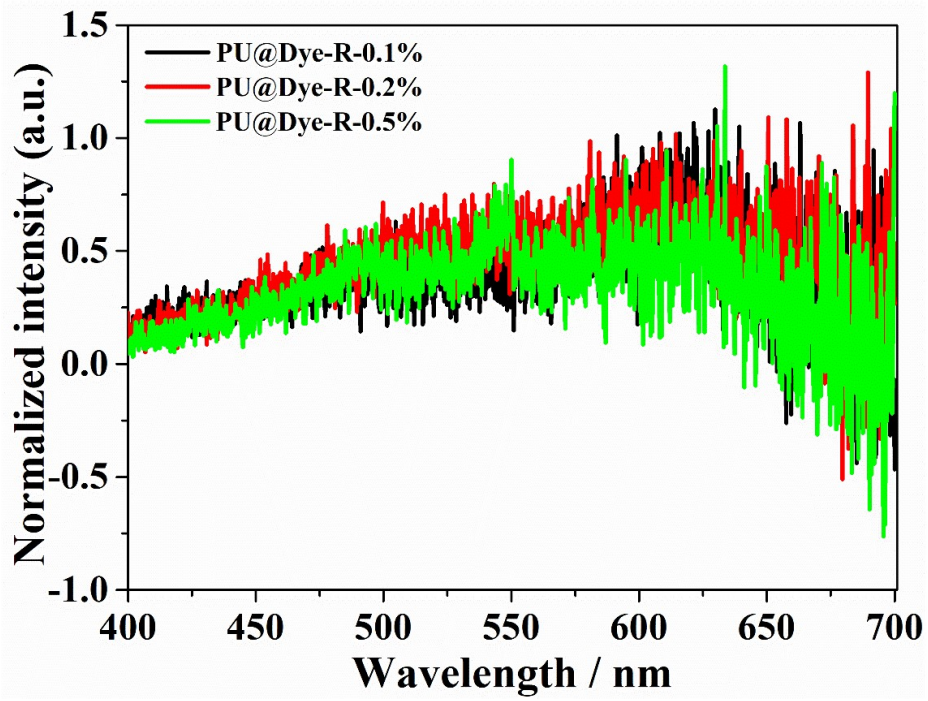

Figure S24. Delayed PL emission spectra of PU@Dye-R-0.1%, PU@Dye-R-0.2% and PU@Dye-R-0.5%.

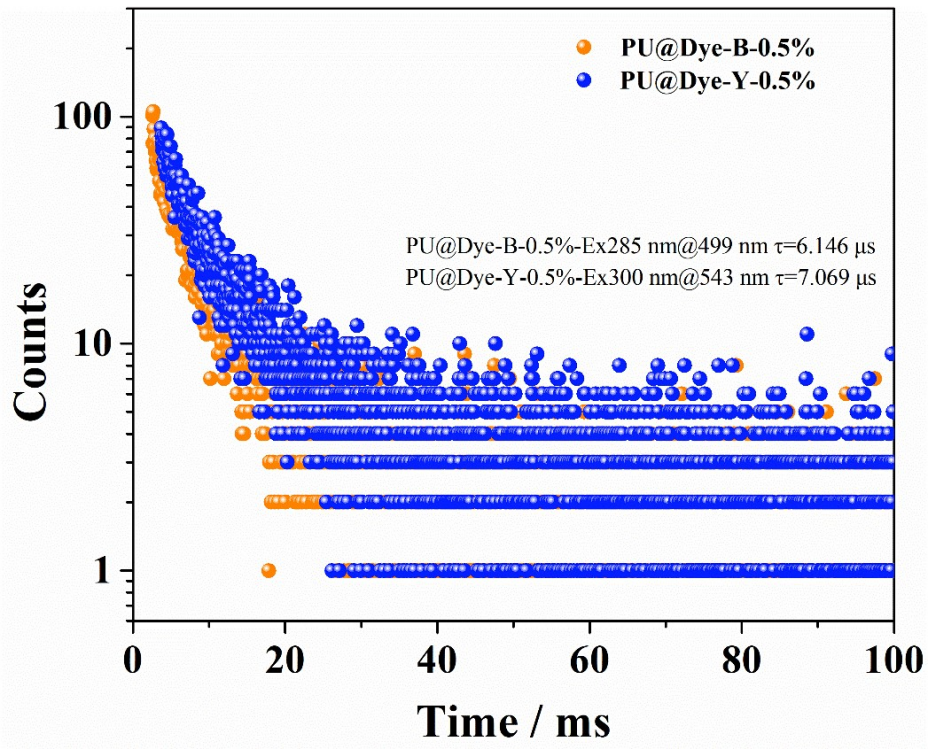

Figure S25. Decay curves obtained at room temperature for PU@Dye-B-0.5% and PU@Dye-Y-0.5%.

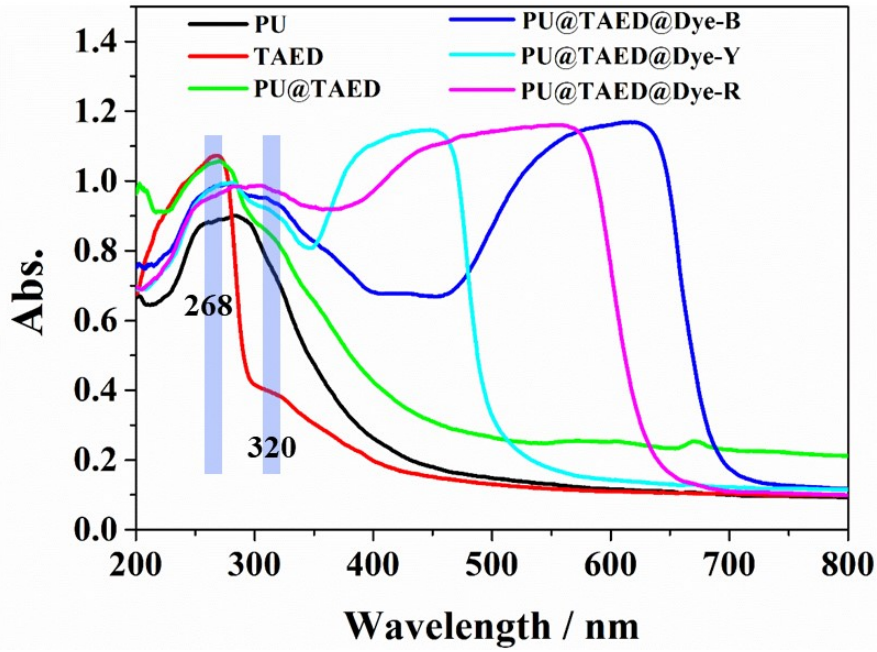

Figure S26. Absorption spectra of PU, TAED, PU@TAED, PU@TAED@Dye-B, PU@TAED@Dye-Y and PU@TAED@Dye-R.

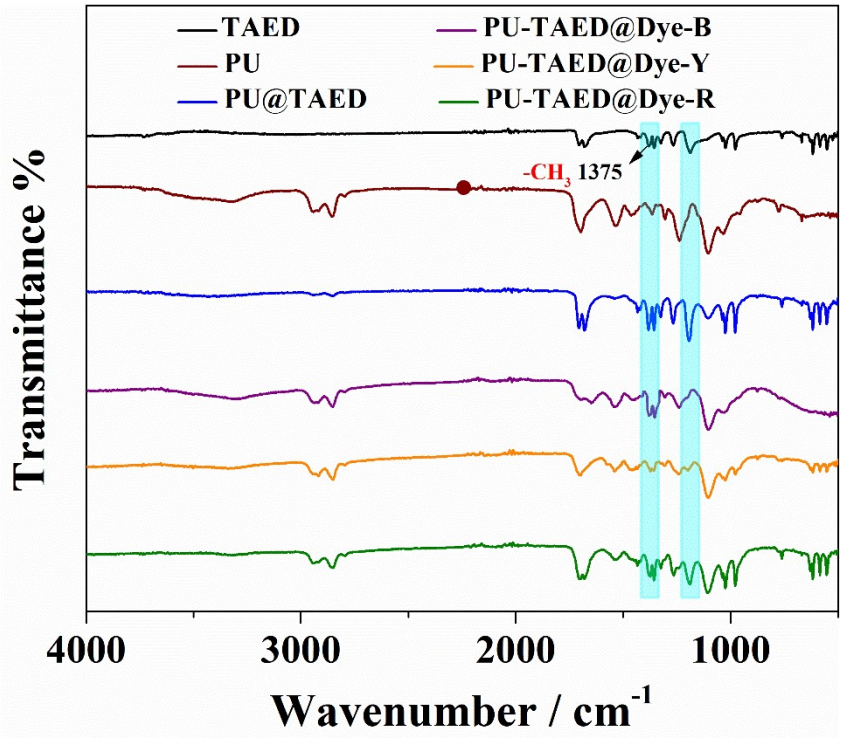

Figure S27. The FT-IR spectra of TAED, PU, PU@TAED, PU@TAED@Dye-B, PU@TAED@Dye-Y, PU@TAED@Dye-R.

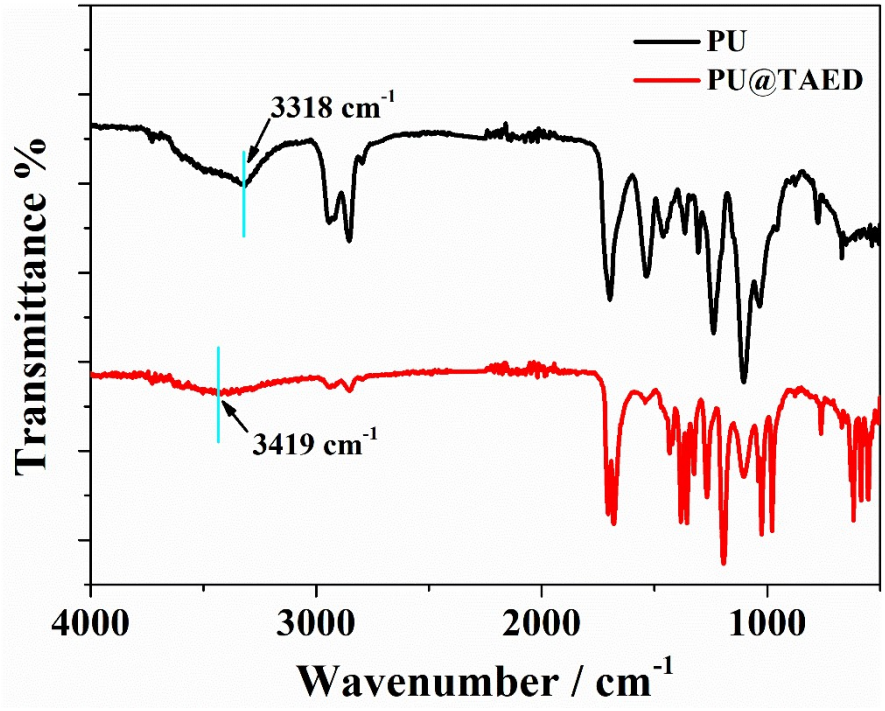

Figure S28. The FT-IR spectra of PU and PU@TAED.

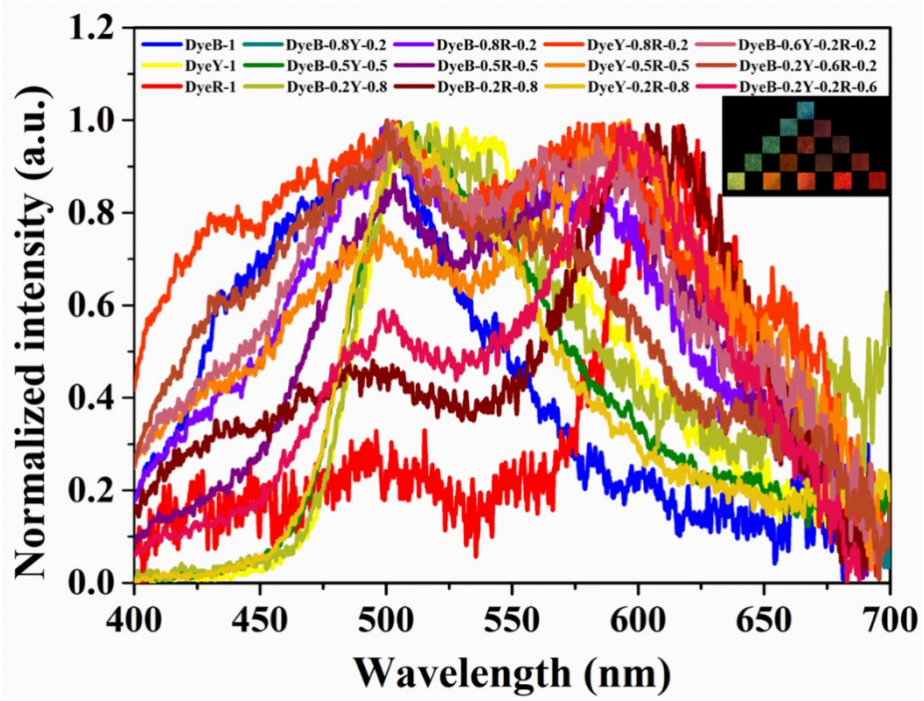

Figure S29. Delayed PL emission spectra of the predictable phosphorescence multi-color system.

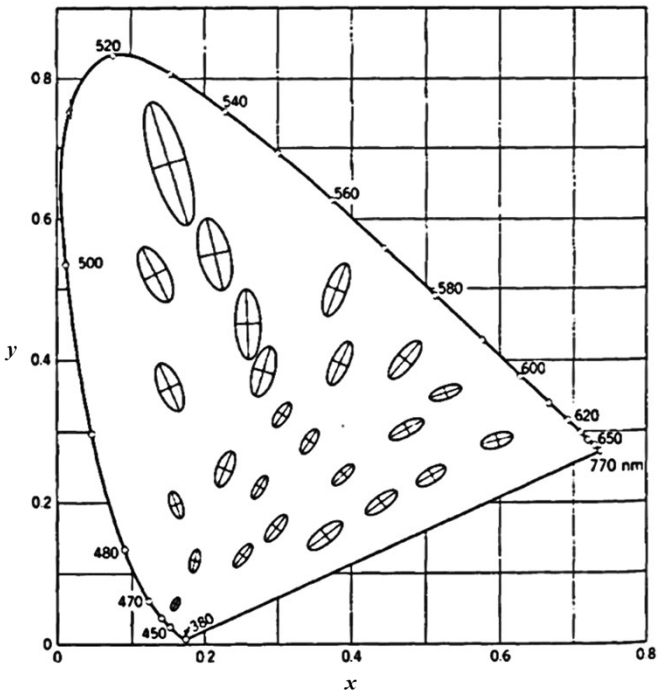

Figure S30. MacAdam ellipses: areas in the CIE chromaticity diagram corresponding to ten times the standard deviation of color matching for one observer.

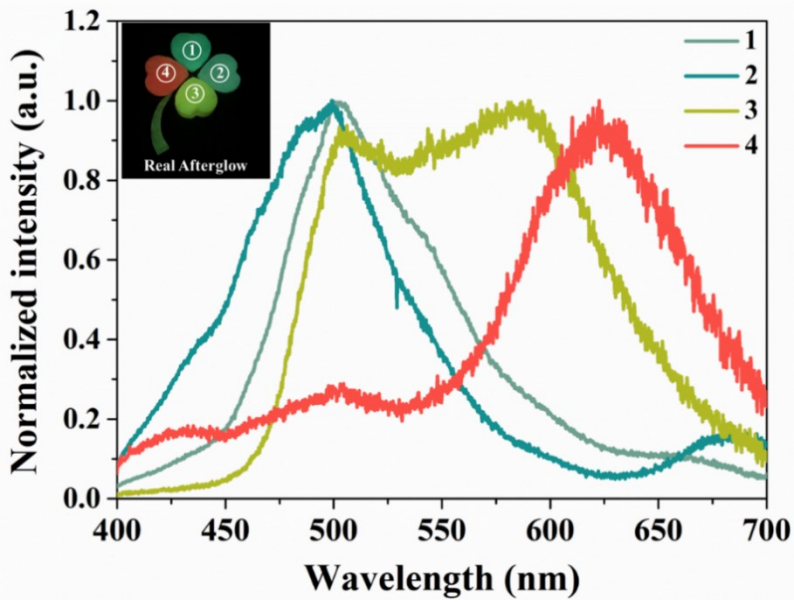

Figure S31. Delayed PL emission spectra of four-leaf clover label.

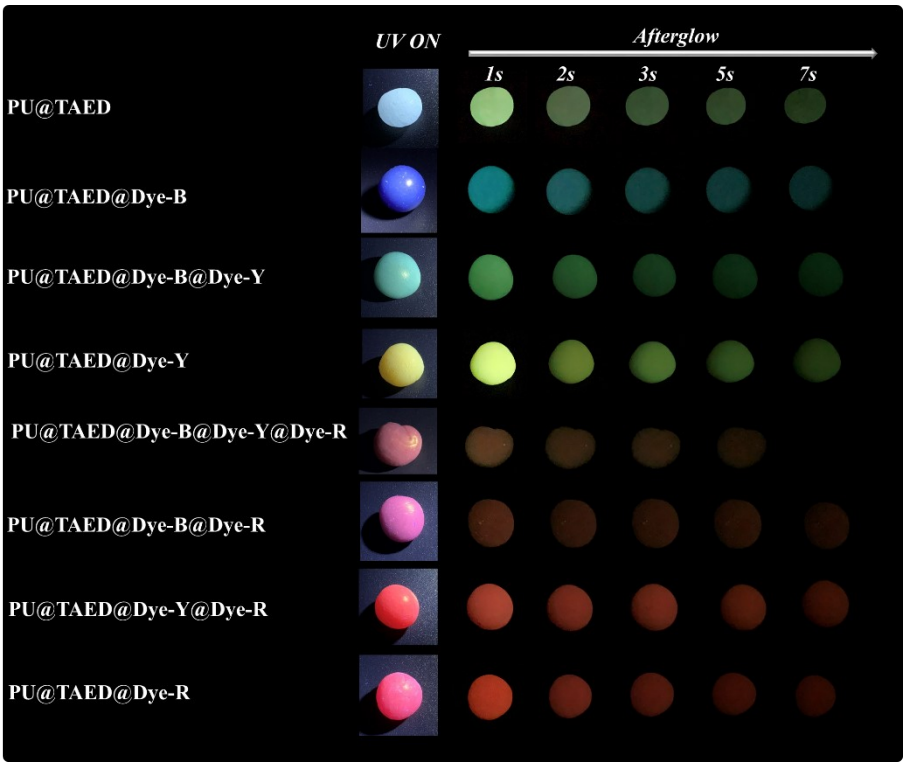

Figure S32. Afterglow photographs of PU@TAED, PU@TAED@Dye-B, PU@TAED@Dye-Y, PU@TAED@Dye-R, PU@TAED@Dye-B@Dye-Y, PU@TAED@Dye-B@Dye-R, PU@TAED@Dye-Y@Dye-R and PU@TAED@Dye-B@Dye-Y@Dye-R under different UV 365 nm.

Table S1. The density of C=O bond in DAED and TAED.

Sample name	Cell volume	Z	Density of C=O
DAED	196.6	1	98
TAED	567.473	2	70.9

Table S2. The comparison of the well-developed doping dyes systems.

Reference Number	Prediction accuracy /%	Literature
55	80.00	Personalized saliency prediction using color spaces[1]
56	66.67	Towards a reliable prediction of the aquatic toxicity of dyes
57	78.88	Evaluation model of color difference for dyed fabrics based on the Support Vector Machine
58	79.85	Reactive Blue 21 Exhaustion Degree Investigated Using the Surface Response Methodology as an Auxiliary Tool in Cotton Dyeing

59	87.1	Illumination correction of dyed fabric based on extreme learning machine with improved ant lion optimizer
60	80.2	Attribute analysis and modeling of color harmony based on multi-color feature extraction in real-life scenes
This work	88.89	

Table S3. The fitting parameters for phosphorescence lifetimes.

Compound	λ_{ex} (nm)	λ_{em} (nm)	τ_1 (ms)	A_1 (%)	τ_2 (ms)	A_2 (%)	<τ> (ms)	χ^2
TAED	410	473	112	65.99	615	34.01	283	1.317
	340	497	300	46.85	946	53.15	643	1.308
PU@TAED	340	503	481	7.98	1230	92.02	1170	1.038
	340	540	163	0.14	1181	99.86	1180	1.131
PU@TAED@Dye-B-0.5%	300	435	366	1.39	1141	98.61	1130	1.175
	340	503	327	9.41	1202	90.59	1120	1.165
	340	540	116	4.51	906	95.49	870	1.005
PU@TAED@Dye-Y-0.5%	340	503	200	2.17	1100	97.83	1080	1.295
	340	540	34	8.94	1205	91.06	1100	1.189
PU@TAED@Dye-Y-0.5%	340	503	208	6.95	1124	93.05	1060	1.249
	340	540	42	2.14	960	97.86	940	1.287
	395	600	198	7.35	967	92.65	910	1.276
PU@TAED@Dye-B-0.1%	300	435	421	6.16	1193	93.84	1145	1.173
	340	503	132	0.30	1201	99.70	1198	1.241
PU@TAED@Dye-B-0.2%	300	435	302	1.56	1144	98.44	1131	1.280
	340	503	49	0.23	1187	99.77	1184	1.293
PU@TAED@Dye-B-0.5%	300	435	290	5.82	1181	94.18	1129	1.176
	340	503	411	11.97	1218	88.03	1121	1.208
PU@TAED@Dye-B-1.0%	300	435	109	6.65	909	93.35	855	1.235
	340	503	165	4.68	1012	95.32	972	1.275
PU@TAED@Dye-Y-	340	503	228	2.20	1156	97.80	1136	1.281

0.1%	340	540	599	6.36	1199	93.64	1161	1.216
PU@TAED@Dye-Y-	340	503	338	8.46	1179	91.54	1108	1.283
0.2%	340	540	469	6.62	1158	93.38	1112	1.196
PU@TAED@Dye-Y-	340	503	136	4.31	1120	95.69	1078	1.146
0.5%	340	540	147	1.04	1118	98.96	1109	1.290
PU@TAED@Dye-Y-	340	503	431	16.19	1115	83.81	1004	1.277
1.0%	340	540	303	16.47	1173	83.53	1030	1.014
PU@TAED@Dye-R-	340	503	16	0.59	1145	99.41	1138	1.287
0.1%	395	594	293	8.18	1181	91.82	1108	1.225
PU@TAED@Dye-Y-	340	503	341	9.96	1191	90.04	1106	1.128
0.2%	395	594	11	0.85	999	99.15	991	1.139
PU@TAED@Dye-R-	340	503	138	8.02	1143	91.98	1062	1.237
0.5%	395	600	101	3.90	944	96.10	911	1.301
PU@TAED@Dye-R-	340	503	141	11.87	956	88.13	859	1.162
1.0%	395	610	104	14.47	892	85.53	778	1.313
PU@Dye-B-0.5%	285	499	0.0025	26.88	0.0075	73.12	0.006	1.138
	300	543	0.0010	12.69	0.0079	87.31	0.007	1.236