Supplementary Information

Controlled solvothermal flow synthesis and spray-drying of fluorescent carbon-based nanomaterials based on phenylenediamines

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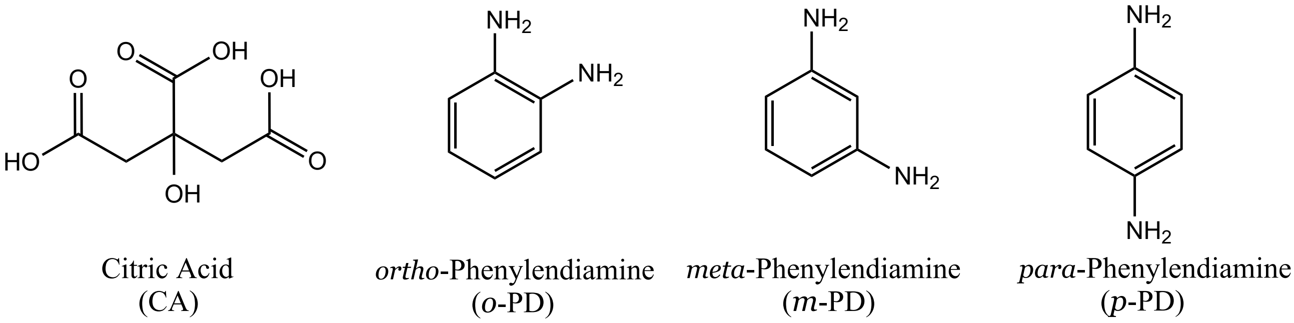

Figure S1: Molecule structures and abbreviations of the starting materials used for synthesis, comprising carboxylic acid and amines as functional groups.

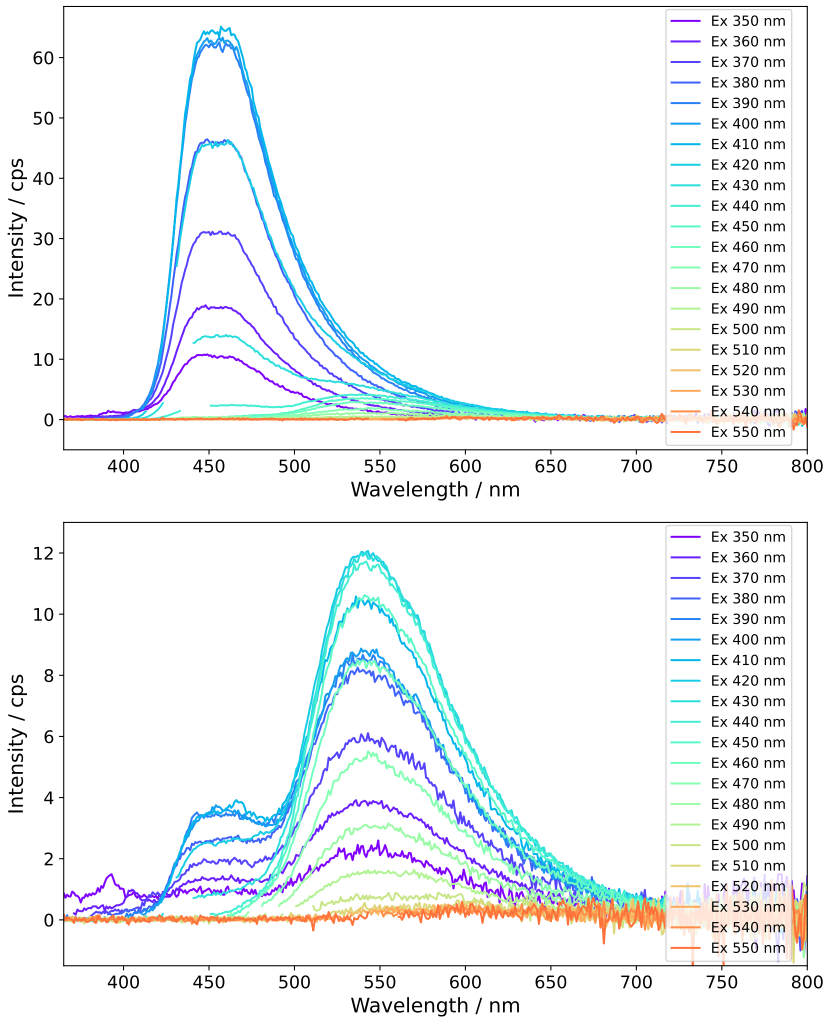

Figure S2: Emission spectra at consecutive excitation wavelengths (Ex) from 350 to 550 nm of the synthesis products using *o*-PD/CA as starting materials. Two different fluorescence maxima are observed: a primarily blue fluorophore (top) and primarily green fluorophore (bottom) which have overlapping excitation wavelength ranges.

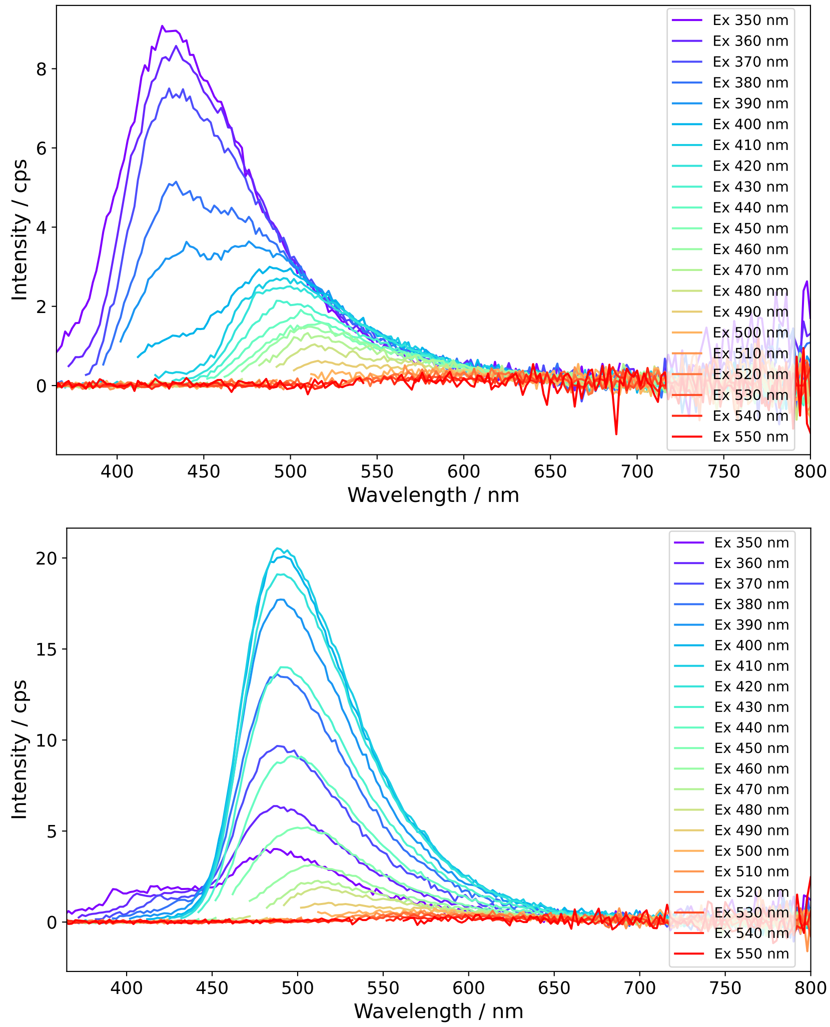

Figure S3: Emission spectra at consecutive excitation wavelengths (Ex) from 350 to 550 nm of the synthesis products using *m*-PD/CA as starting materials. Two different fluorescence maxima are observed: a primarily blue fluorophore (top) and primarily cyan fluorophore (bottom).

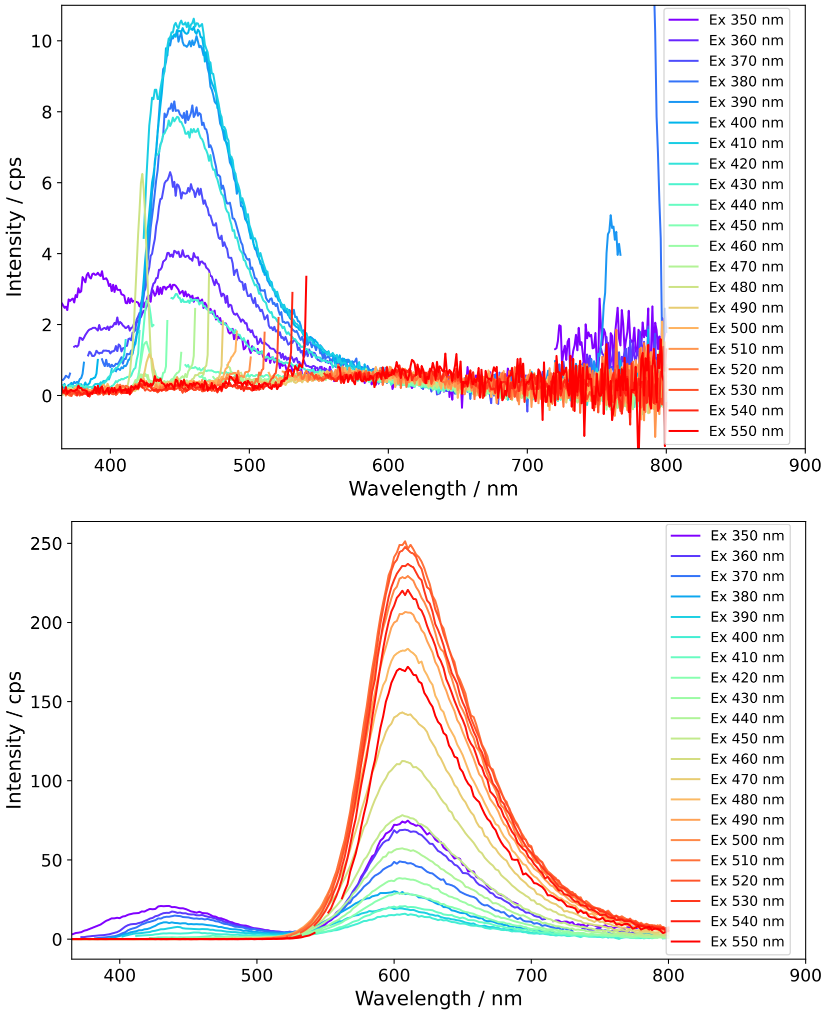

Figure S4: Emission spectra at consecutive excitation wavelengths (Ex) from 350 to 550 nm of the synthesis products using *p*-PD/CA as starting materials. Two different fluorescence maxima are observed: a primarily blue fluorophore (top) and primarily red fluorophore (bottom) which have overlapping excitation wavelength ranges.

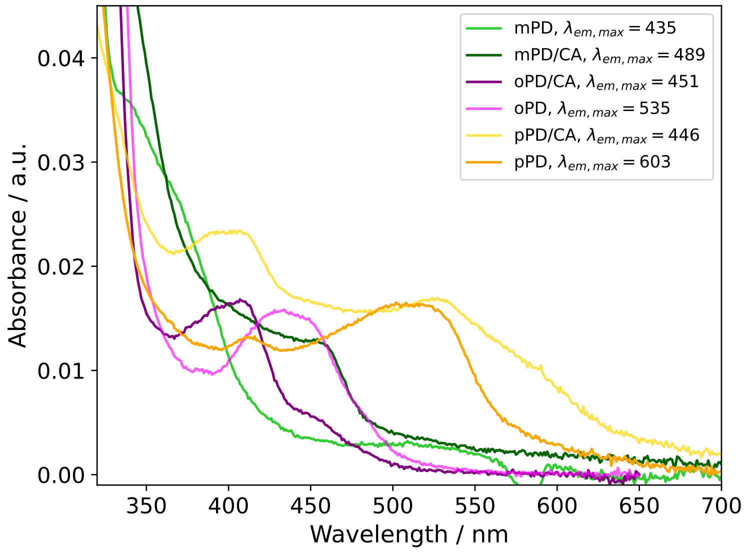

Figure S5: Absorbance spectra of the synthesis products with different emission wavelength maxima ($\lambda_{em,max}$) using *m*-, *o*-, *p*-PD/CA as starting materials. The wavelengths of the absorption maxima correspond to the optimal excitation ranges for fluorescence emission.

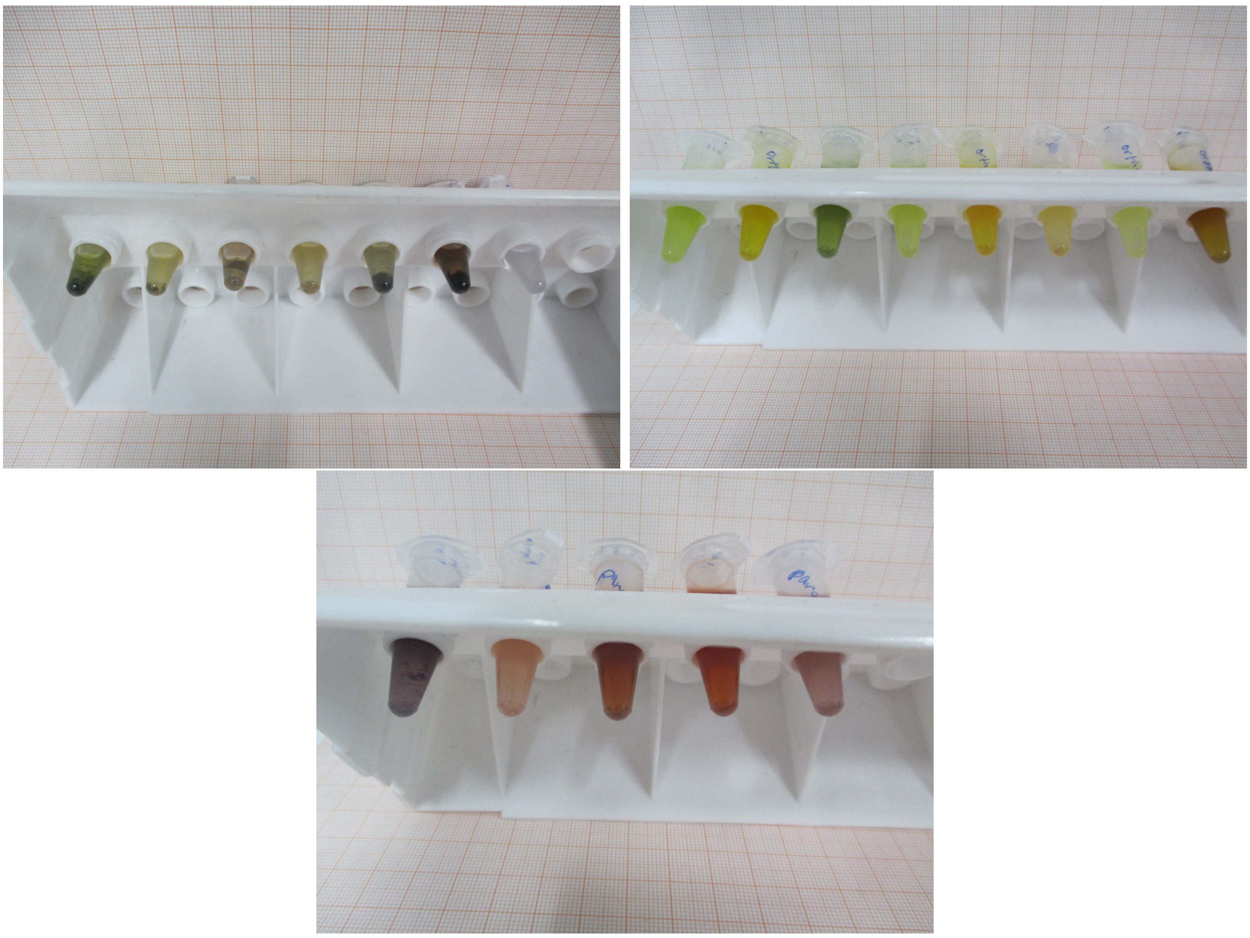

Figure S6: Sample materials after continuous solvothermal synthesis in ethanol using toluene as anti-solvent for precipitation of CBNs (top left *m*-PD/CA, top right *o*-PD/CA, bottom *p*-PD/CA).

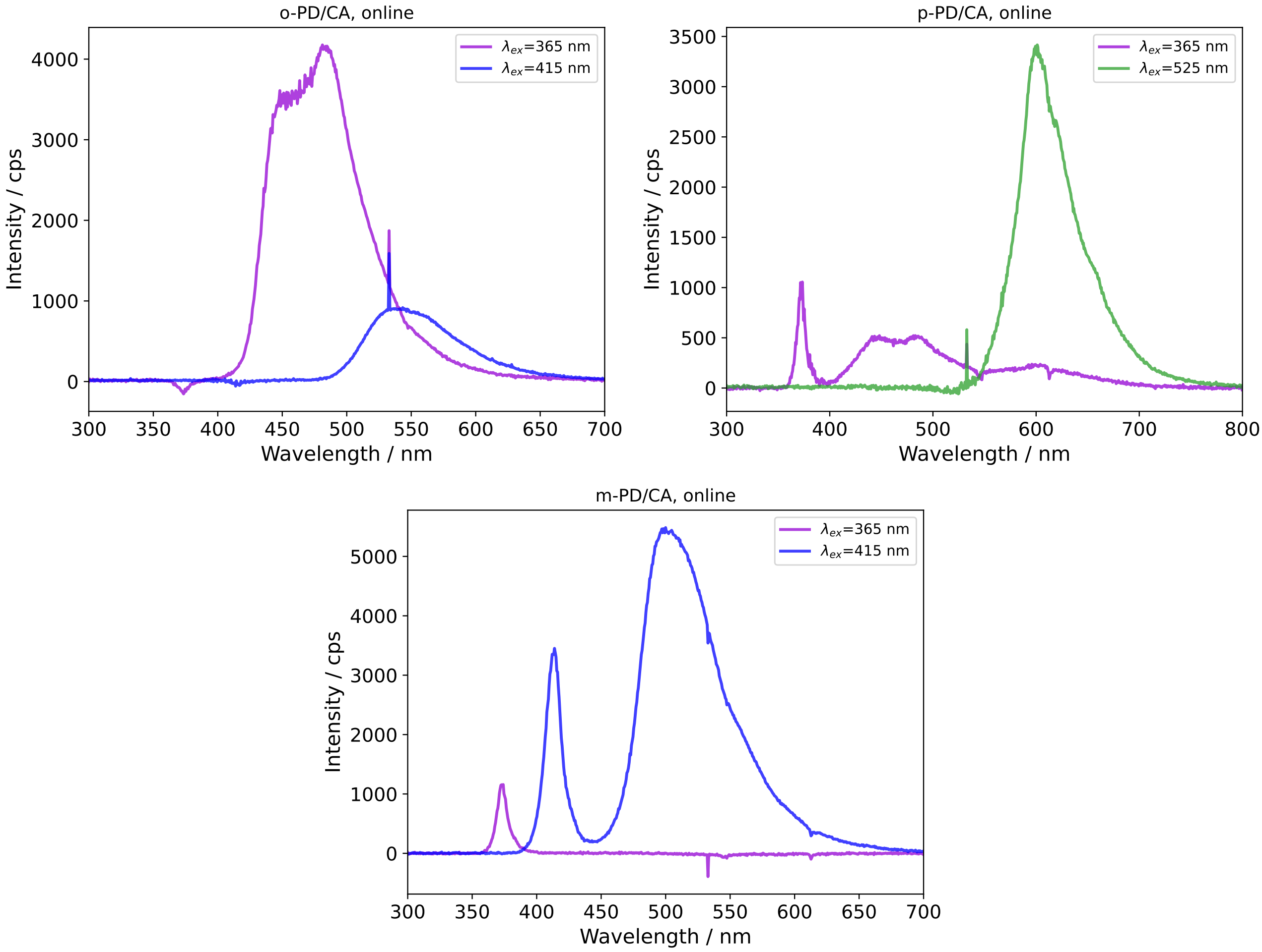

Figure S7: Online fluorescence spectra using *o*-, *p*-PD/CA as starting materials (top left and top right) and using *m*-PD/CA (bottom). Two excitation sources (λ_{ex}) are included in the reactor design to detect both synthesis products during continuous synthesis in an alternating scheme. Their wavelengths are partially visible in the sample spectrum at the respective wavelength positions. The spike at 540 nm is considered to be a measurement artifact since it is visible regardless of the sample material.

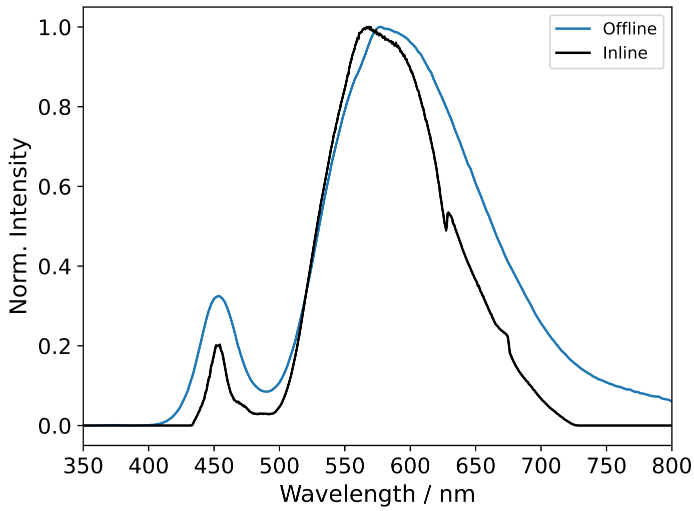

Figure S8: Fluorescence spectrum of a white LED obtained from the offline and inline spectrometer applied in this study.

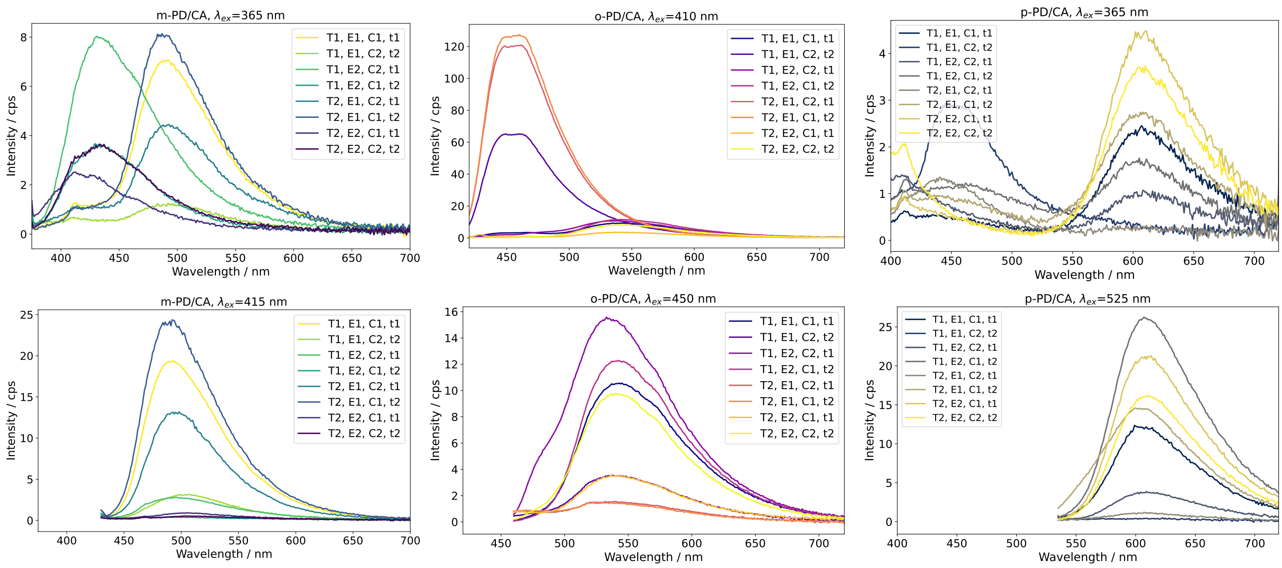

Figure S9: Fluorescence spectra of PD/CA-CBNs dispersions after continuous synthesis with different process parameter combinations (E, C, T, t). The titles above the diagrams detail the wavelength used to excite one of the two species in the PD/CA-systems: m-PD/CA (left top and bottom), o-PD/CA (middle top and bottom) and p-PD/CA (right top and bottom).

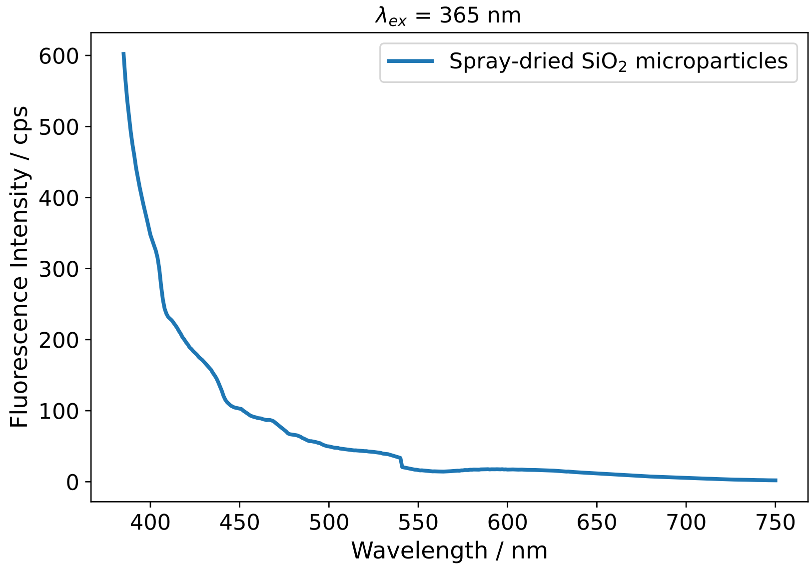

Figure S10: Fluorescence spectra of spray-dried SiO_2-nanoparticles used as blank data for mixtures with CBNs. The vertical signal progression at 541 nm is considered to be a measuring artifact from changing detectors within the spectrometer because it is visible in all sample materials regardless of their compositions.

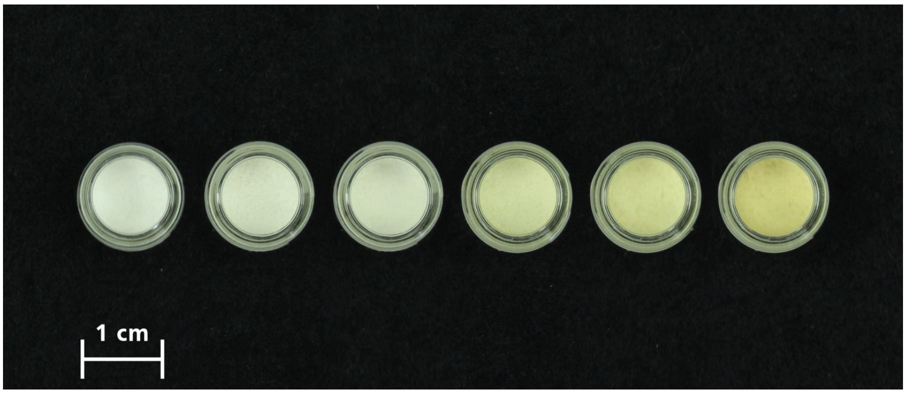

Figure S11: Photo image of spray-dried microparticles loaded with CBNs with decreasing dilution factor from left to right.

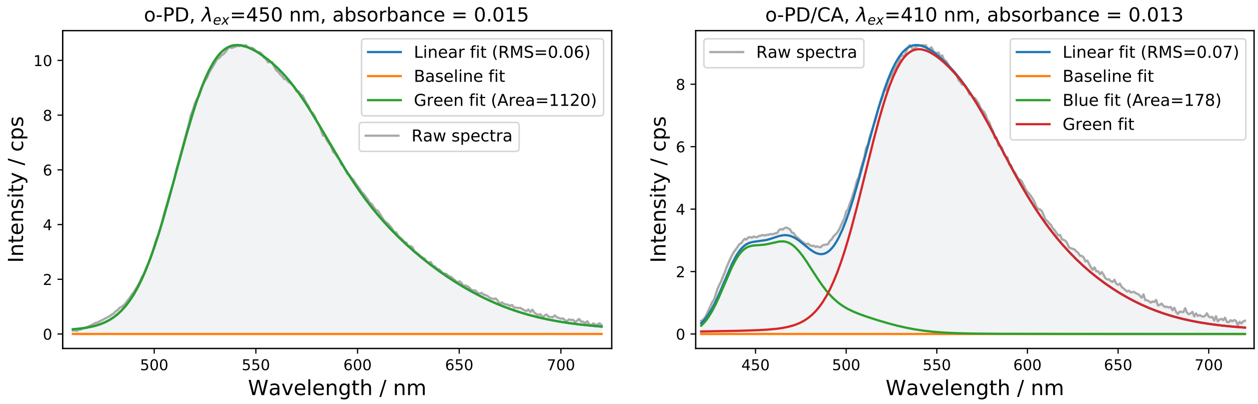

Figure S12: Raw spectra and deconvoluted mixed spectra of synthesis products using *o*-PD/CA as starting materials. The signals are obtained at comparable optical densities/absorbance at the excitation wavelength λ_{ex}=450 nm (left) and λ_{ex}=410 nm (right). The fit results are specified within brackets: residues (RMS) and surface integrals (Area).

Table S1: Deconvolution model summary for two emission bands within the *o*-PD/CA system.

Hard Model Summary for *o*-PD/CA	
RMS Residuals	0.0419768
Baseline	Linear
Component names	blue, green
Total components	2
Significant components	2
Total number of peaks	6
Peaks per component	3, 3
Degree of freedom (total free parameters)	28
Component fitting mode	W/B, Peak Positions
Component shift	No
Considered interacting peaks	10
Component weight threshold	0.001

Table S2: Deconvolution model summary for two emission bands within the m-PD/CA system.

Hard Model Summary for *m*-PD/CA	
RMS Residuals	0.558577
Baseline	Linear
Component names	blue, cyan
Total components	2
Significant components	2
Total peaks	4
Peaks per component	1,3
Degree of freedom (total free parameters)	24
Component fitting mode	W/B, Peak Positions
Component shift	No
Considered interacting peaks	10
Component weight threshold	0.001

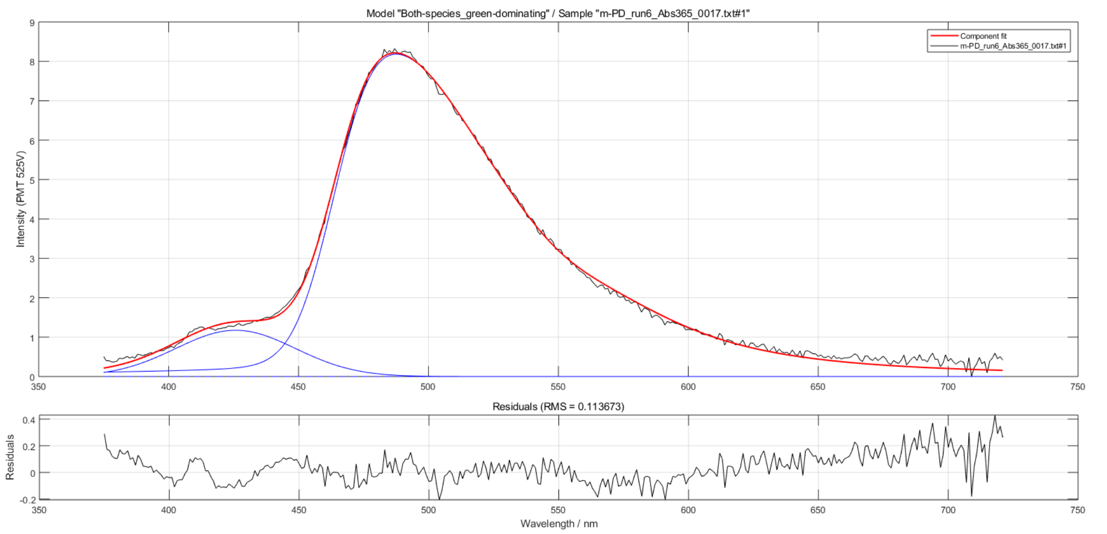

Table S3: Deconvolution model summary for two emission bands within the p-PD/CA system.

Hard Model Summary for *p*-PD/CA	
RMS Residuals	0.100157
Baseline	Linear
Component names	blue, red
Total components	2
Significant components	2
Total number of peaks	4
Peaks per component	2,2
Degree of freedom (total free parameters)	20
Component fitting mode	W/B, Peak Positions
Component shift	No
Considered interacting peaks	10
Component weight threshold	0.001

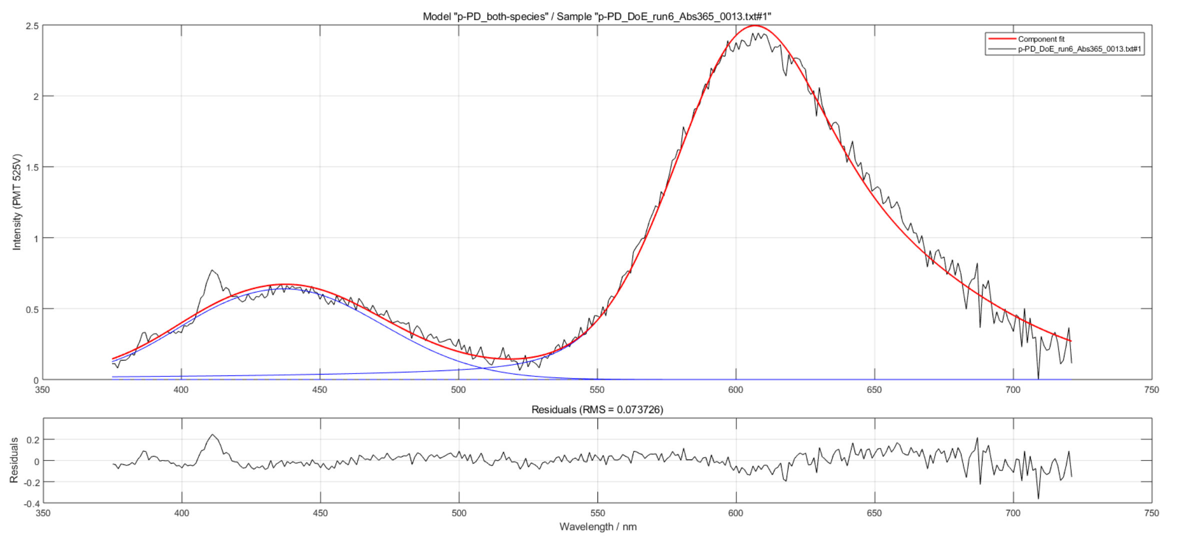

Table S4: ANOVA result table for the fluorescence sum intensity of the two species within the *m*-PD/CA system in correlation to the process parameters with high effect size. Note: P-values less than 0.0500 indicate model terms are statistically significant (df: Degree of Freedom).

***m*-PD/CA**					
Input	**df**	**Mean Square**	**F-Value**	**p-Value**	**Significance**
Model: Blue Species (435 nm)	3	172879	284	<0.0001	significant
factor E: CA (Omission)	1	466578	767	< 0.0001	significant
factor t: time (20 Min)	1	19801	33	0.0047	significant
factor Et/TC: 2-way interaction	1	32258	53	0.0019	significant
Residual	4	608	-	-	-
Fit statistic	**Mean**	**Std.Dev.**	**C.V / %**	**R^2**	Adjusted R^2
	315	25	8	0.995	0.992
Model: Cyan Species (489 nm)	3	1345842	12	0.0178	significant
factor E: CA (Addition)	1	2758901	25	0.0076	significant
factor C: Concentration	1	558625	5	0.0884	not significant
factor EC/Tt: 2-way interaction	1	720000	7	0.0636	not significant
Residual	4	111117	-	-	-
Fit statistic	**Mean**	**Std.Dev.**	**C.V / %**	**R^2**	Adjusted R^2
	746	333	45	0.900	0.826

Table S5: ANOVA result table for the fluorescence sum intensity of the two species within the *o*-PD/CA system in correlation to the process parameters with high effect size. Note: P-values less than 0.0500 indicate model terms are statistically significant (df: Degree of Freedom).

***o*-PD/CA**					
Input	**df**	**Mean Square**	**F-Value**	**p-Value**	**Significance**
Model: Blue Species (451 nm)	3	45535016	12	0.0179	significant
factor E: CA (Addition)	1	84714636	23	0.0090	significant
factor T: Temperature	1	25945206	7	0.0586	not significant
factor ET/tC: 2-way interaction	1	25945206	7	0.0586	not significant
Residual	4	3768564	-	-	-
Fit statistic	**Mean**	**Std.Dev.**	**C.V / %**	R^2	Adjusted R^2
	3254	1941	60	0.901	0.826
Input	**df**	**Mean Square**	**F-Value**	**p-Value**	**Significance**
Model: Green species (535 nm)	4	526687	10	0.0422	significant
factor E: CA (Omission)	1	775635	15	0.0299	significant
factor T: Temperature	1	909226	18	0.0243	significant
factor t: time	1	48205	0.9462	0.4024	not significant
factor EC/Tt: 2-way interaction	1	373680	7	0.0733	not significant
Residual	3	50946	-	-	-
Fit statistic	**Mean**	**Std.Dev.**	**C.V / %**	**R^2**	Adjusted R^2
	807	226	28	0.932	0.842

Table S6: ANOVA result table for the fluorescence sum intensity of the two species within the *p*-PD/CA system in correlation to the process parameters with high effect size. Note: P-values less than 0.0500 indicate model terms are statistically significant (df: Degree of Freedom).

***p*-PD/CA**					
Input	**df**	**Mean Square**	**F-Value**	**p-Value**	**Significance**
Model: Blue Species (446 nm)	4	9067	14	0.0273	significant
factor C: Concentration (10 mM)	1	22155	35	0.0098	significant
factor E: CA (Addition)	1	630	1	0.3938	not significant
factor t: time	1	5356	8	0.0627	not significant
factor EC/Tt: 2-way interaction	1	8128	13	0.0376	significant
Residual	3	639	-	-	-
Fit statistic	**Mean**	**Std.Dev.**	**C.V / %**	**R^2**	Adjusted R^2
	127	25	20	0.950	0.883
Input	**df**	**Mean Square**	**F-Value**	**p-Value**	**Significance**
Model: Red Species (603 nm)	3	1913760	22	0.0060	significant
factor C: Concentration (10 mM)	1	3508601	40	0.0031	significant
factor E: CA (Omission)	1	1700168	20	0.0114	significant
factor t: time	1	532512	6	0.0684	not significant
Residual	4	86728	-	-	-
Fit statistic	**Mean**	**Std.Dev.**	**C.V / %**	**R^2**	Adjusted R^2
	1187	295	25	0.943	0.900

Table S7: Composition ratio between CBNs and SiO_2-nanoparticles as spray drying feeds resulting in different dilution factors (DF).

Components	DF1001	DF201	DF101	DF21	DF11	DF5
15 nm SiO_2-nanoparticles / g	1	1	1	1	1	1
CBNs dispersion / ml	0.001	0.005	0.01	0.05	0.1	0.25
H_2O / g	6.66	6.66	6.65	6.61	6.56	6.41