Supplementary Information (SI) for Polymer Chemistry.
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Supplementary Information

Naphthalimide Derivative-based Photoinitiating Systems for Multi-Wavelength Visible Light Photopolymerization and 3D Printing

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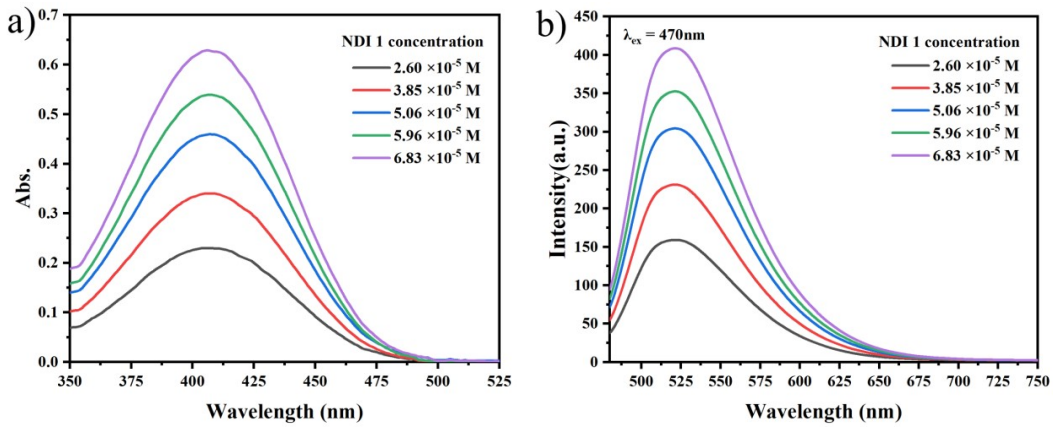


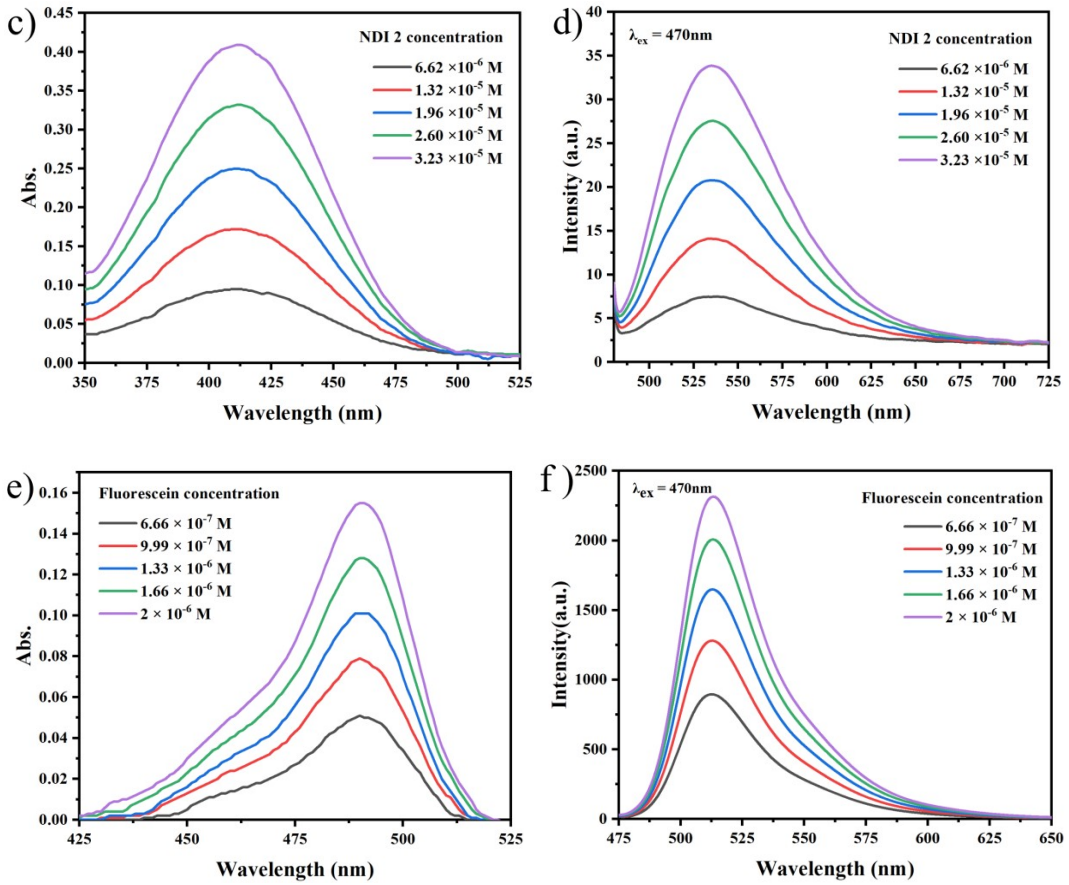
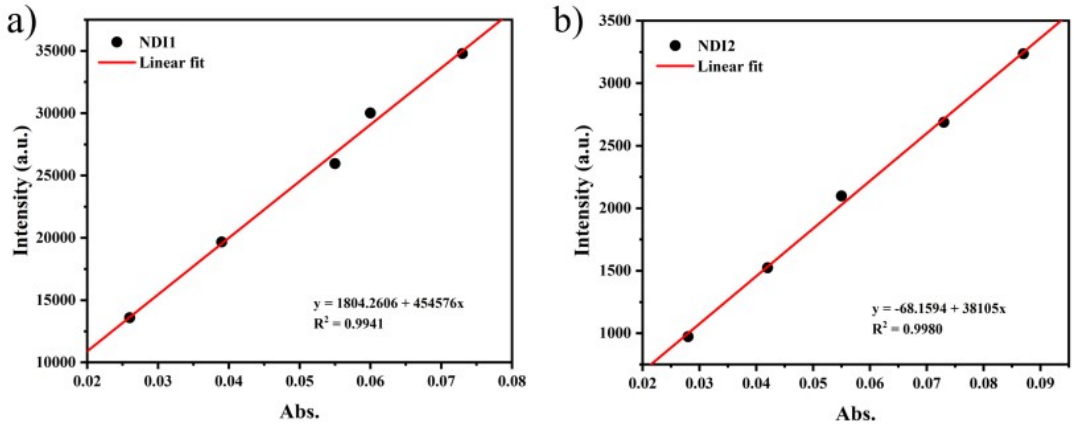


Figure S1. a) UV-vis absorption spectra and b) fluorescence emission spectra (λ_{ex} = 470 nm) of NDI 1 in acetonitrile with different concentrations; c) UV-vis absorption spectra and d) fluorescence emission spectra (λ_{ex} = 470 nm) of NDI 2 in acetonitrile with different concentrations; e) UV-vis absorption spectra and f) fluorescence emission spectra (λ_{ex} = 470 nm) of fluorescein in 0.1 M NaOH with different concentrations.



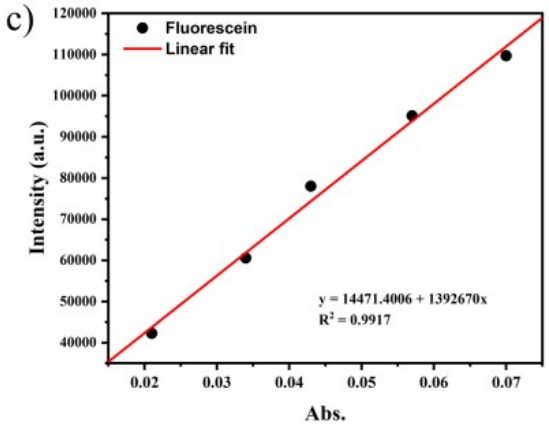


Figure S2. Integrated spectral fluorescence intensity with different absorbances of a) NDI 1； b) NDI 2; c) fluorescein.

Table S1. The absorbances (A) and corresponding integrated spectral fluorescence intensities (I) linear fit slopes (Grad) of solutions with different concentrations, and the fluorescence quantum yields (QY) of studied photoinitiators.

		1	2	3	4	5	Grad	QY
NDI 1	A	0.026	0.039	0.055	0.06	0.073	454576	0.25
	I	13590.845	19667.165	25958.52	30017.835	34794.78		
NDI 2	A	0.028	0.042	0.055	0.073	0.087	38105	0.021
	I	972.87	1523.905	2098.2	2688.28	3235.93		
fluorescein	A	0.021	0.034	0.043	0.057	0.07	1392670	0.91[a]
	I	42273.11	60577.855	78026.57	95120.67	109709.675		

[a] Standard fluorescent quantum yield of fluorescein in 0.1 M NaOH solution with an excitation wavelength at 470 nm.

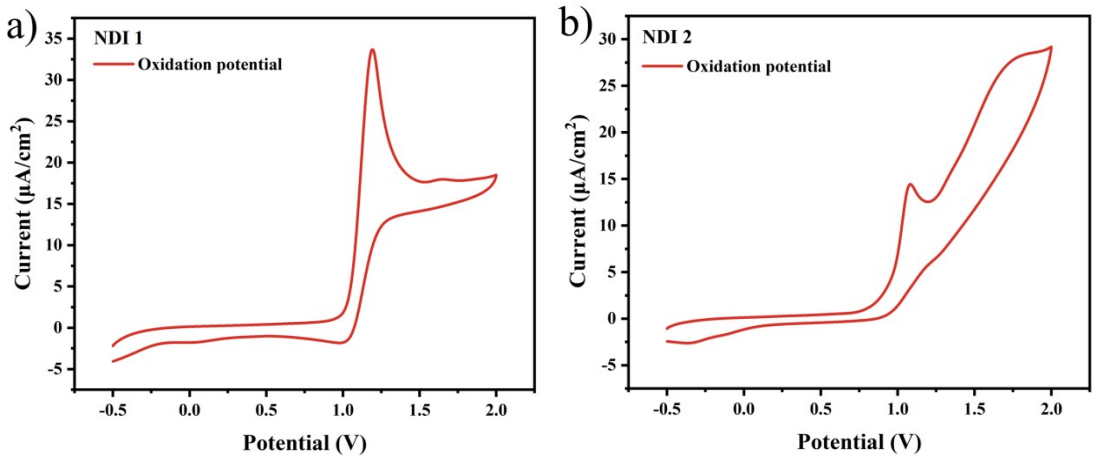


Figure S3. Cyclic voltammetry of electrochemical reactions in acetonitrile against saturated calomel electrode. a) NDI 1; b) NDI 2.

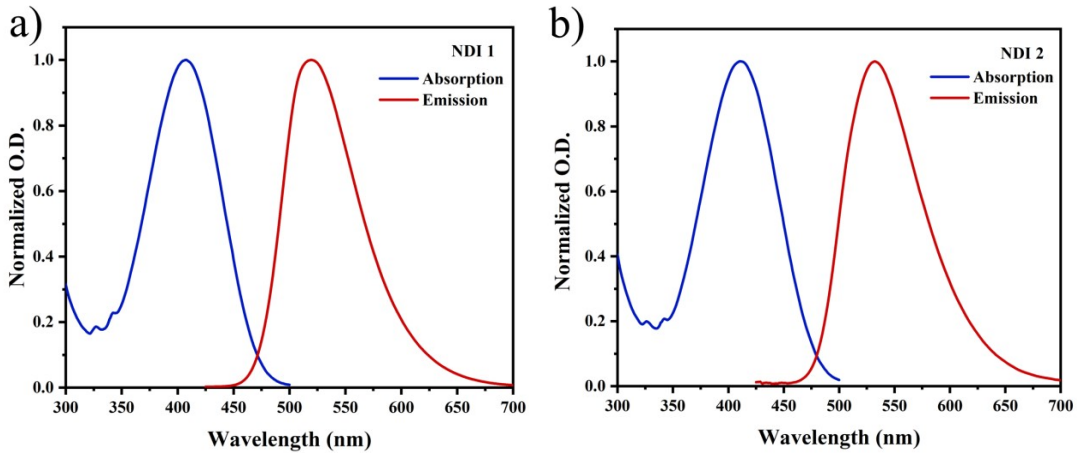


Figure S4. Normalized UV-vis absorption and fluorescence spectra of a) NDI 1; b) NDI 2.

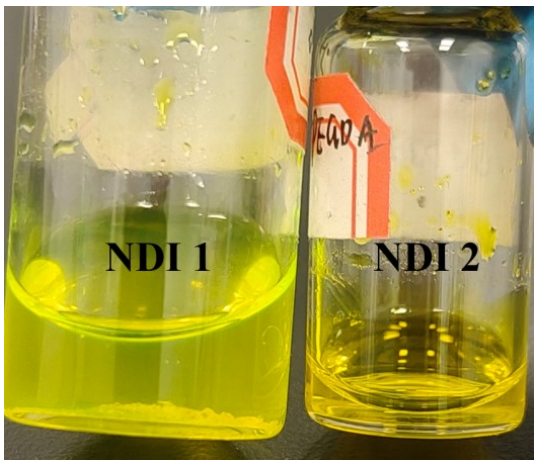
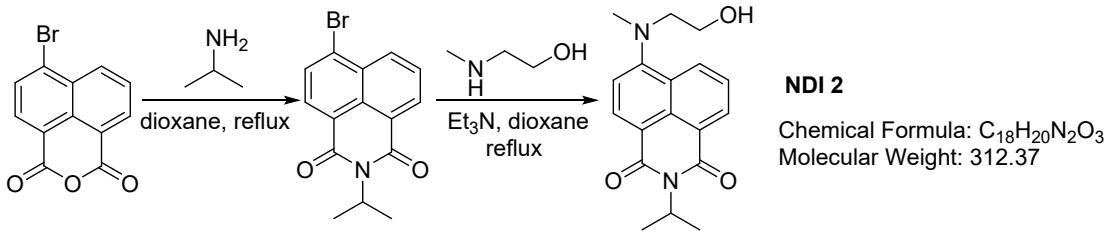


Figure S5. The solubility of NDI 1 (left) and NDI 2 (right) in PEGDA 700 at a concentration of 0.5 wt%.

Synthesis of 4-[N-methyl-N-(2-hydroxyethyl)amino]-N-isopropyl-1,8-naphthalimide (NDI 2).



4-Bromo-1,8-naphthalic anhydride (1.4 g, 5.0 mmol) was dissolved in 1,4-dioxane (20 mL) in a round-bottom flask equipped with a magnetic stirrer and a condenser. Isopropyl amine (1.7 mL, 20 mmol) was added and the mixture was heated under

reflux, and the reaction progress was monitored by TLC until the complete reaction of anhydride. The solvent was removed under reduced pressure. The crude product (4-Bromo-N-isopropylnaphthalimide) was purified by silica column chromatography using n-heptane/acetone (3:1, v/v) as the eluent. Yield: 1.2 g, 78%.

4-Bromo-N-isopropylnaphthalimide (0.6 g, 1.9 mmol) was dissolved in 1,4-dioxane (50 mL) in a round-bottom flask equipped with a magnetic stirrer and a condenser. 2-(Methylamino)ethanol (1.2 mL, 16 mmol) and triethylamine (1.4 mL, 10.0 mmol) were then added. The mixture was heated under reflux and the reaction progress was monitored by TLC. After completion, the solvent was removed under reduced pressure. The crude product was purified by column chromatography on silica gel using n-heptane/acetone (1:1, v/v) as the eluent. Yield: 0.45 g (76%).

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.58 (d, J = 8.6 Hz, 1H), 8.52 (d, J = 7.3 Hz, 1H), 8.45 (d, J = 8.1 Hz, 1H), 7.68 – 7.62 (m, 1H), 7.23 (d, J = 8.1 Hz, 1H), 5.41 (p, J = 7.0 Hz, 1H), 3.95 (s, 2H), 3.47 (t, J = 5.5 Hz, 2H), 3.05 (s, 3H), 1.57 (d, J = 7.0 Hz, 6H).

TOF-MS (m/z) calcd for $C_{18}H_{20}N_2O_3$ $[M+H]^+$ 313.1547 (100%), found: 313.1543.

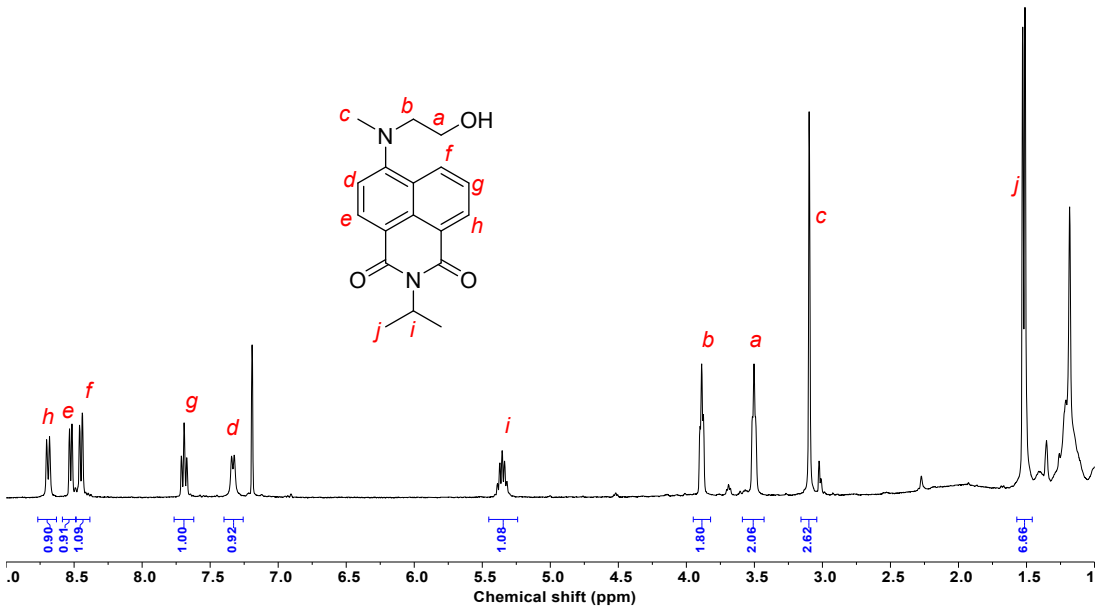


Figure S6. ^{1}H NMR spectrum of 4-[N-methyl-N-(2-hydroxyethyl)amino]-N-isopropyl-1,8-naphthalimide (NDI 2).

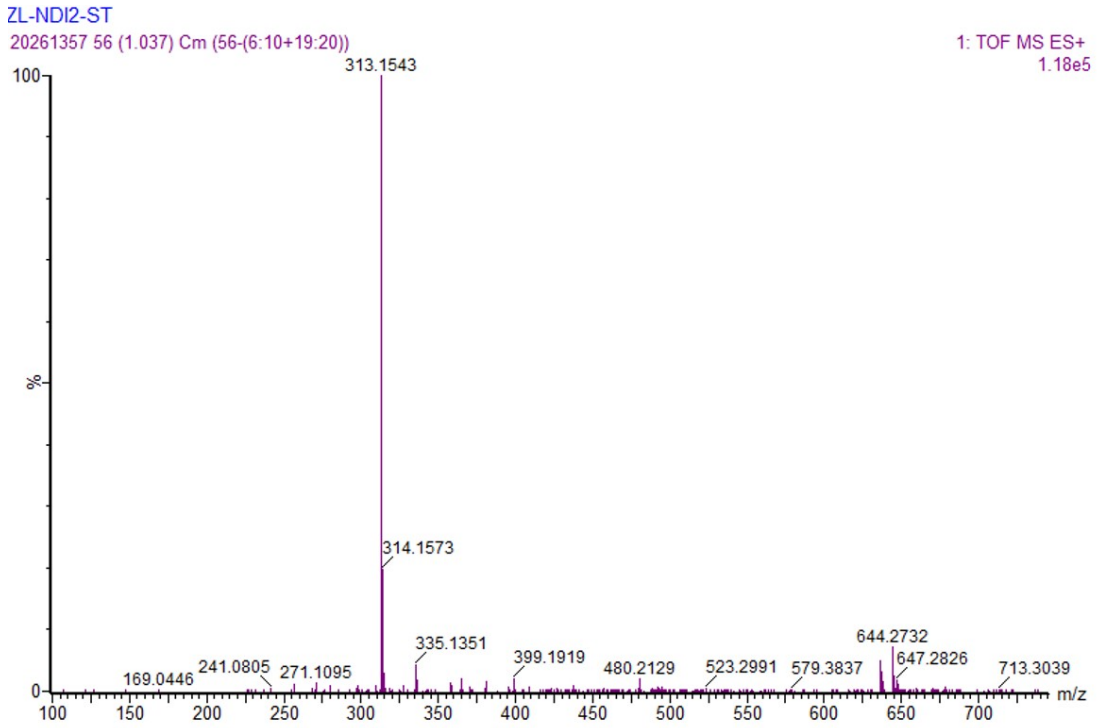
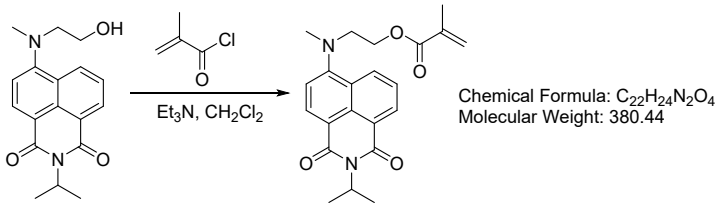


Figure S7. TOF mass spectrum of NDI 2.

Synthesis of 4-[N-methyl-N-(2-methacryloyloxyethyl)amino]-N-isopropyl-1,8-naphthalimide (NDI 1).



Dichloromethane (50 mL), NDI 2 (0.31 g, 1.0 mmol), and triethylamine (0.41 mL, 3.0 mmol) were mixed in a round bottom flask. Methacryloyl chloride (0.2 mL, 2.0 mmol) dissolved in dichloromethane (5 mL) was added dropwise to the mixture at 0 °C using an addition funnel. The resulting mixture was stirred at this temperature for 30 min, then allowed to warm to room temperature and stirred for an additional 10 h. Upon the completion of NDI 2, the reaction was quenched with water (5 mL). The organic layer was subsequently washed with saturated $NaHCO_3$ solution (3 × 10 mL). The

organic phase was dried over anhydrous Na_2SO_4, filtered, and concentrated under reduced pressure. NDI 1 was isolated by silica column chromatography using hexane/acetone (3:1, v/v) as the eluent. Yield: 0.26 g (56%).

^{1}H NMR (400 MHz, $CDCl_3$) δ 8.56 (d, J = 7.3 Hz, 1H), 8.49 (dd, J = 7.7, 5.9 Hz, 2H), 7.71 – 7.64 (m, 1H), 7.26 (d, J = 17.8 Hz, 1H), 6.00 (s, 1H), 5.55 (s, 1H), 5.44 (p, J = 6.9 Hz, 1H), 4.47 (t, J = 5.5 Hz, 2H), 3.67 (t, J = 5.5 Hz, 2H), 3.12 (s, 3H), 1.89 (s, 3H), 1.60 (d, J = 6.9 Hz, 6H).

^{13}C NMR (101 MHz, $CDCl_3$) δ 167.52, 164.78, 164.28, 156.34, 135.86, 132.08, 131.00, 130.21, 130.05, 126.61, 125.90, 125.57, 123.80, 116.51, 115.65, 61.45, 56.89, 46.46, 41.86, 19.81, 18.86.

TOF-MS (m/z) calcd for $C_{22}H_{24}N_2O_4$ $[M+H]^+$ 381.1809 (100%), found: 381.1801.

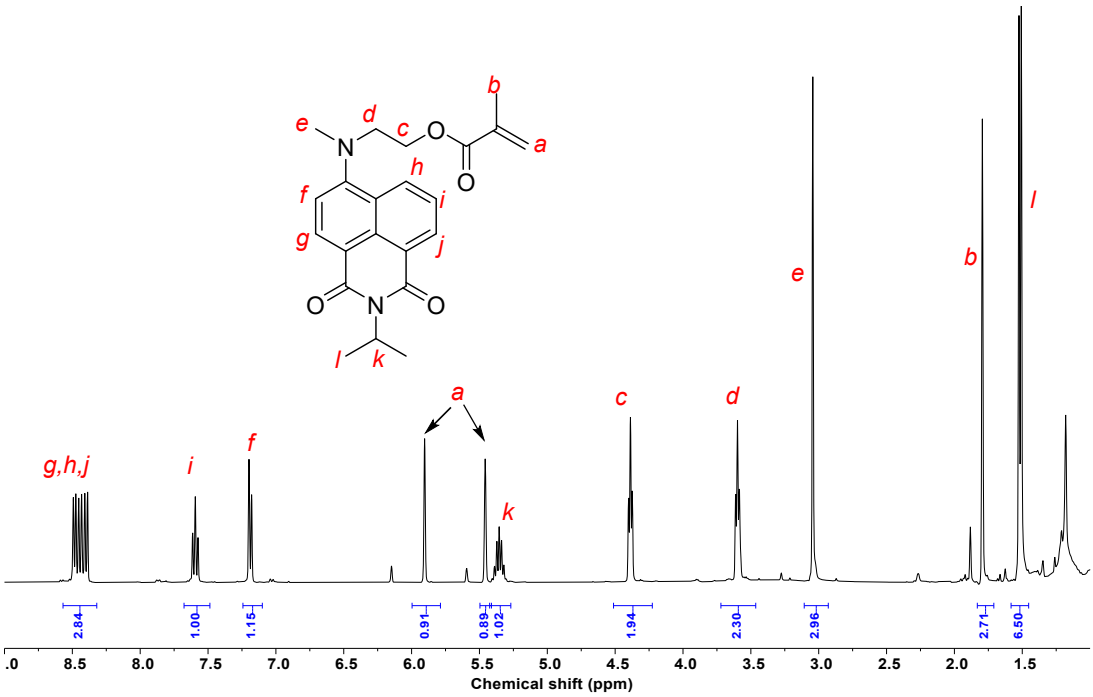


Figure S8. ^{1}H NMR spectrum of 4-[N-methyl-N-(2-methacryloyloxyethyl)amino]-N-isopropyl-1,8-naphthalimide (NDI 1).

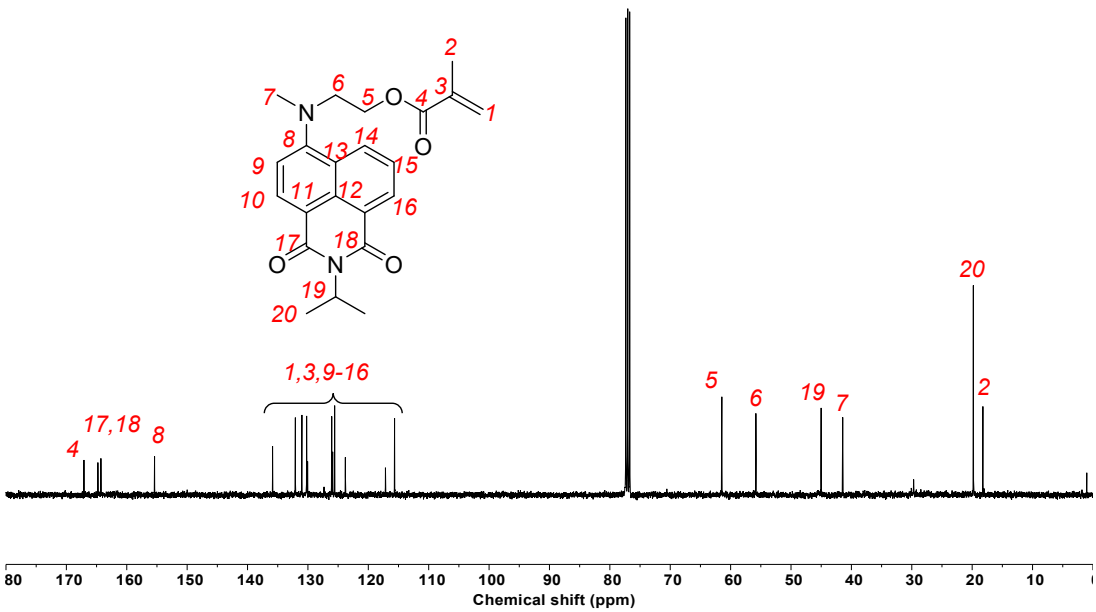


Figure S9. ^{13}C NMR spectrum of 4-[N-methyl-N-(2-methacryloyloxyethyl)amino]-N-isopropyl-1,8-naphthalimide (NDI 1).

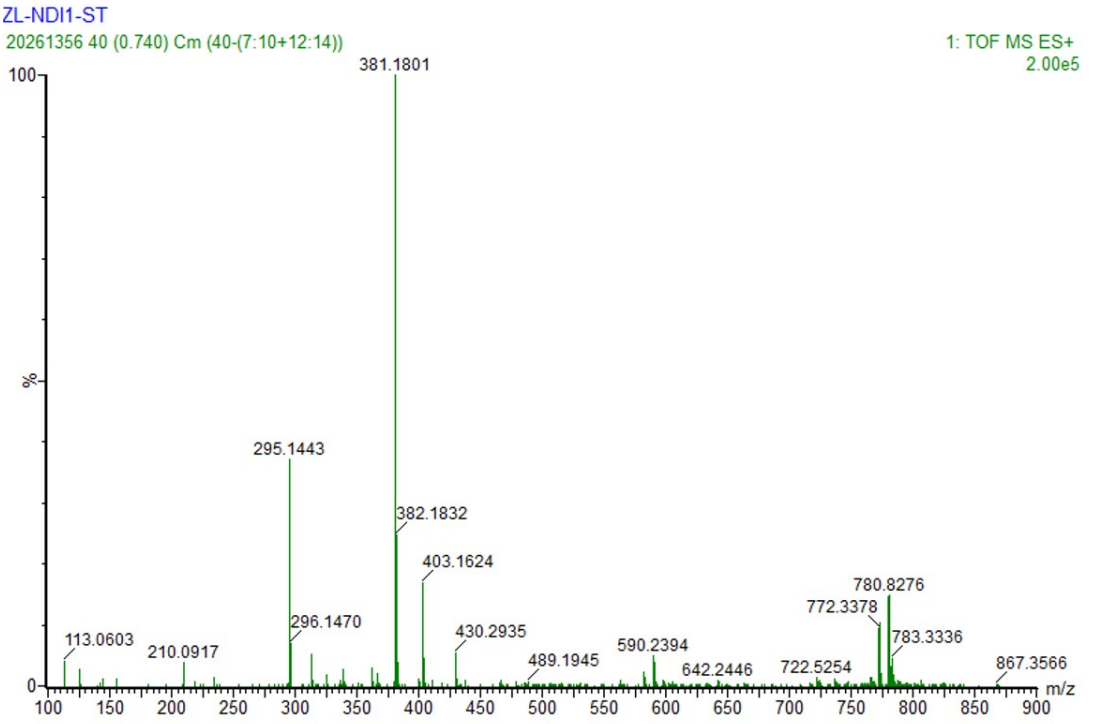


Figure S10. TOF mass spectrum of NDI 1.