Supporting Information

Photoresponsive Supramolecular Architectures Based on Polypeptide Hybrids

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CONTENT

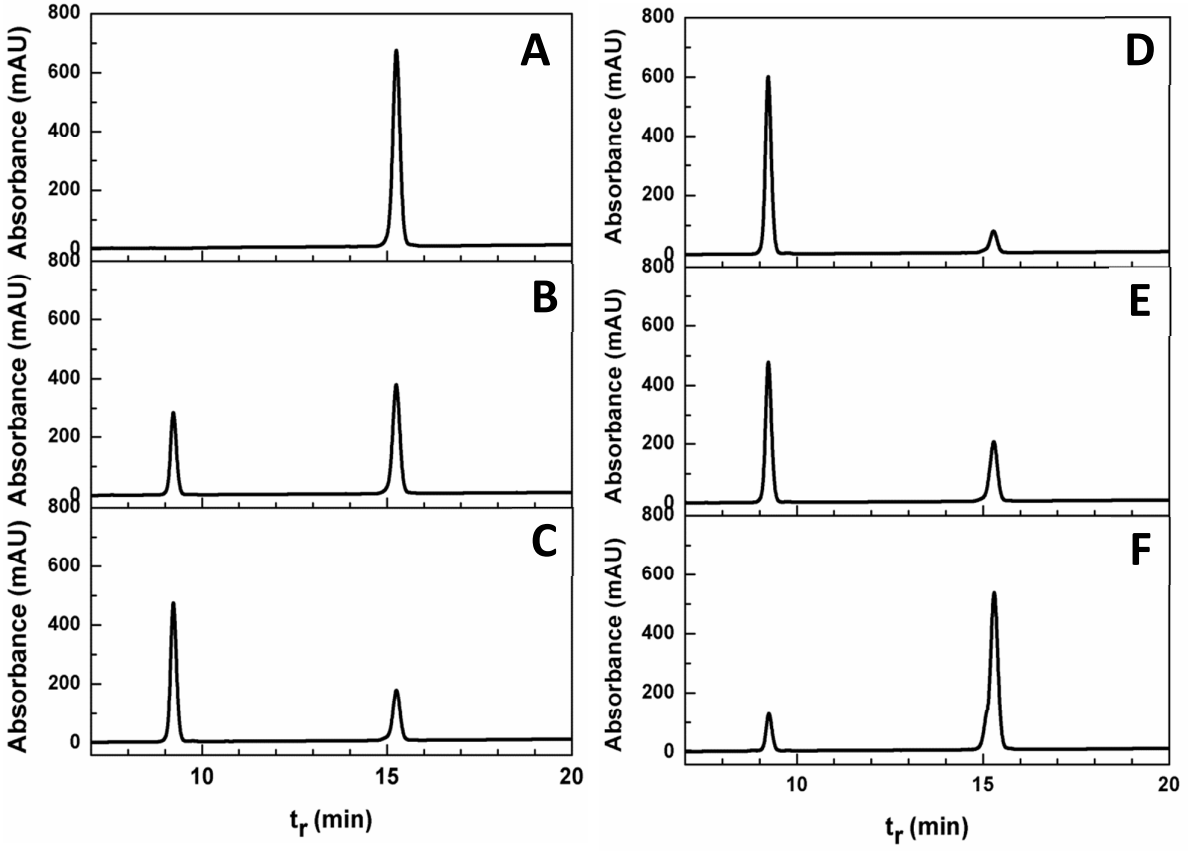

Figure S1: HPLC profiles of the reversible photoisomerization of H-*bis*AzoPhe-OMe (**5**): *trans* form (**A**), and after irradiation at 365 nm for, respectively, 10 s (**B**), 20 s (**C**), and 40 s (**D**). The isomerization reaches its photostationary state (nearly quantitative conversion to the *cis* form) after 40 s. Further irradiation (up to 1 h) at 365 nm led to an identical HPLC profile as in **D**.

HPLC profiles obtained starting from the *cis* form (**D**) upon irradiation at 420 nm for 20 s (**E**) and 60 s (**F**). Also in this case the isomerization to the *trans* form is nearly quantitative . After irradiation at 420 nm for 1 h an identical HPLC profile as in (**F**) was obtained.

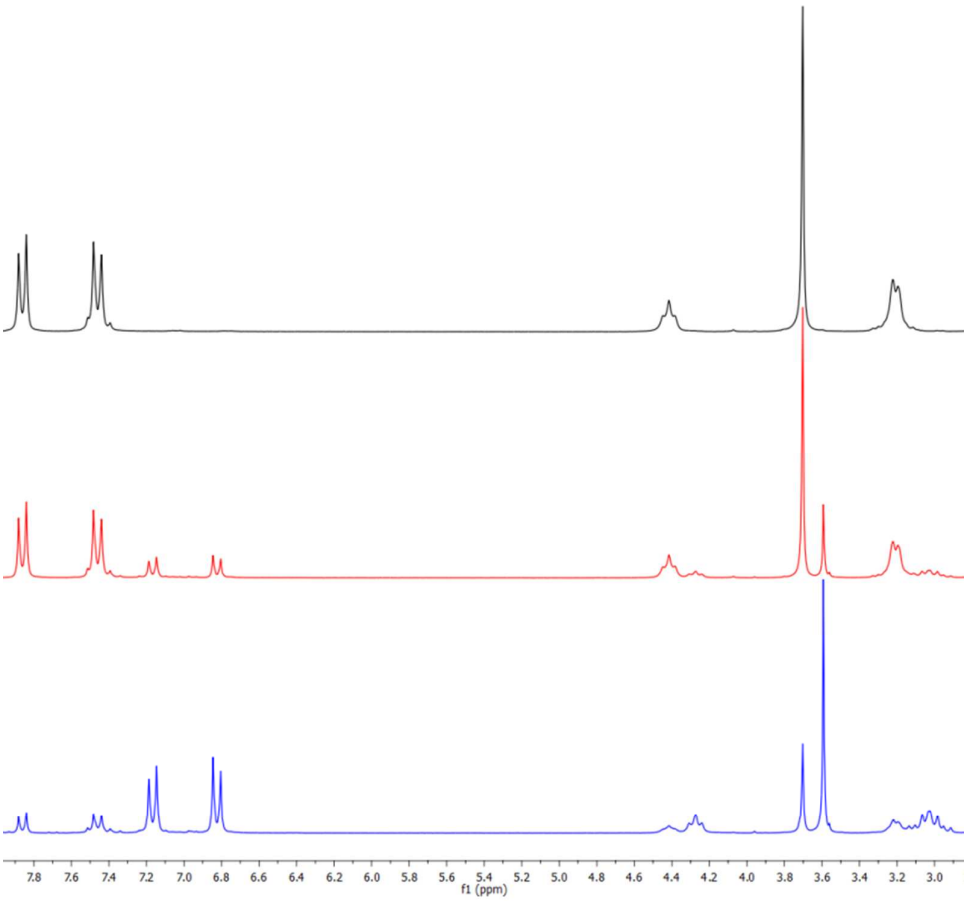

Figure S2: ^{1}H NMR spectra of (**5**) in DMSO, d_6 in the *trans* form (black) and after different times of irradiation at 365 nm: 20 s (red) and 40 s (blue).

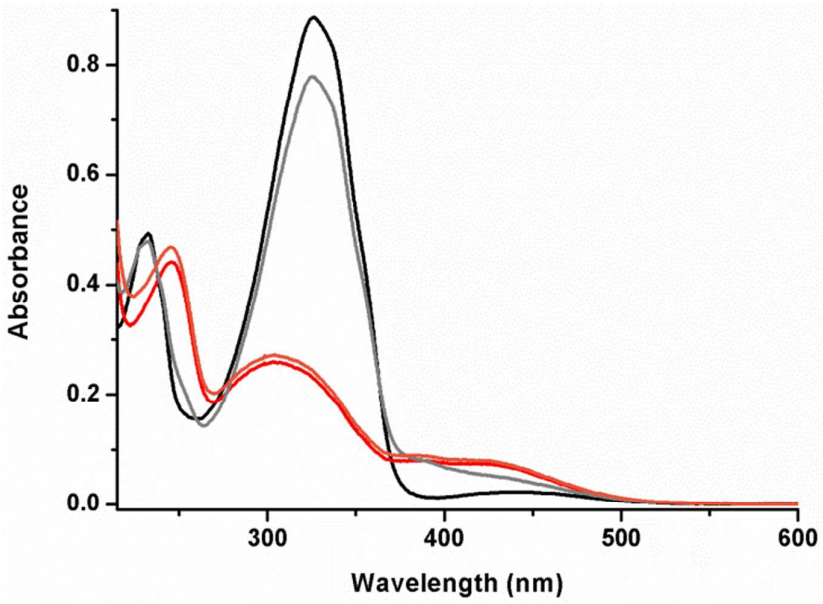

Figure S3: UV-Vis absorption spectra of (**5**) in MeOH showing the reversible *trans*/*cis* isomerization for two cycles (black lines, *trans* form; red lines, *cis* form).

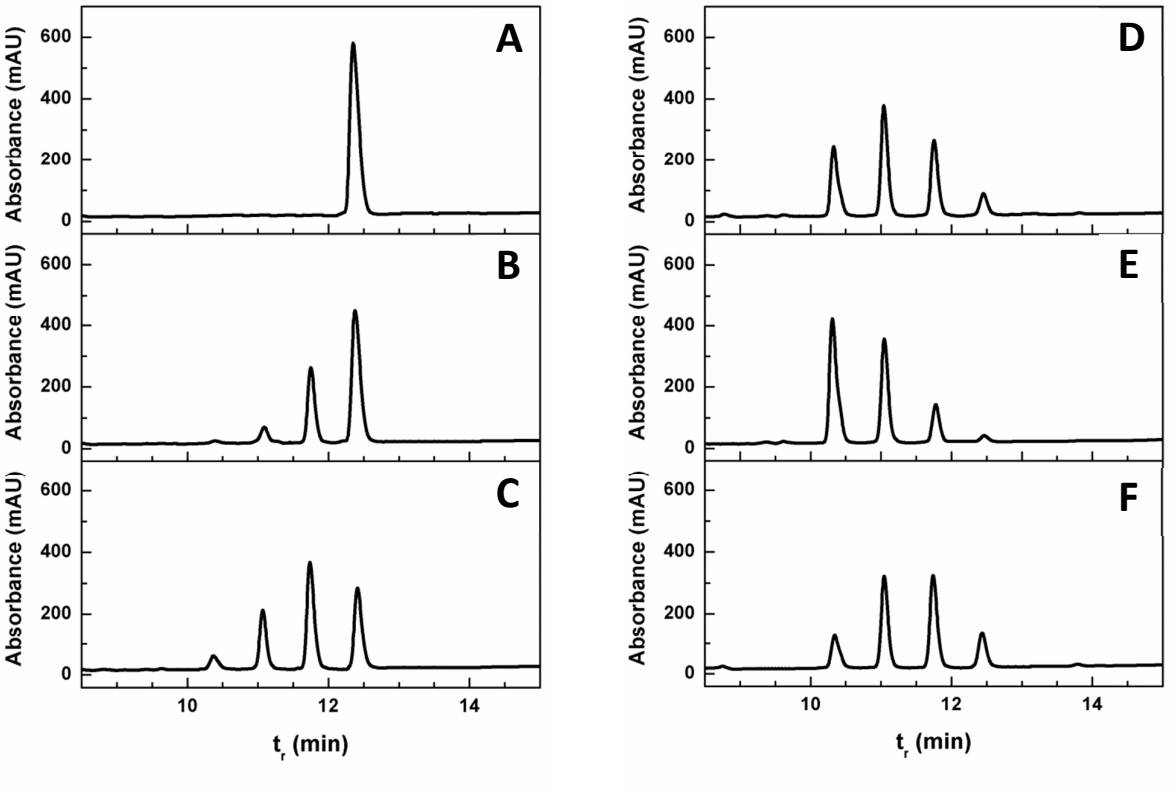

Figure S4: HPLC profiles of the reversible photoisomerization of compound (**11**): all *trans* form (**A**), and after irradiation at 365 nm for, respectively, 15 s (**B**), 30 s (**C**), 1 min (**D**), and 2 min (**E**). The isomerization reaches its photostationary state after 2 min. Longer irradiation times at 365 nm led to an identical HPLC profile as in **E**. The equilibrium profile after 1 h irradiation at 365 nm followed by 12 h in the dark is given in (**F**).

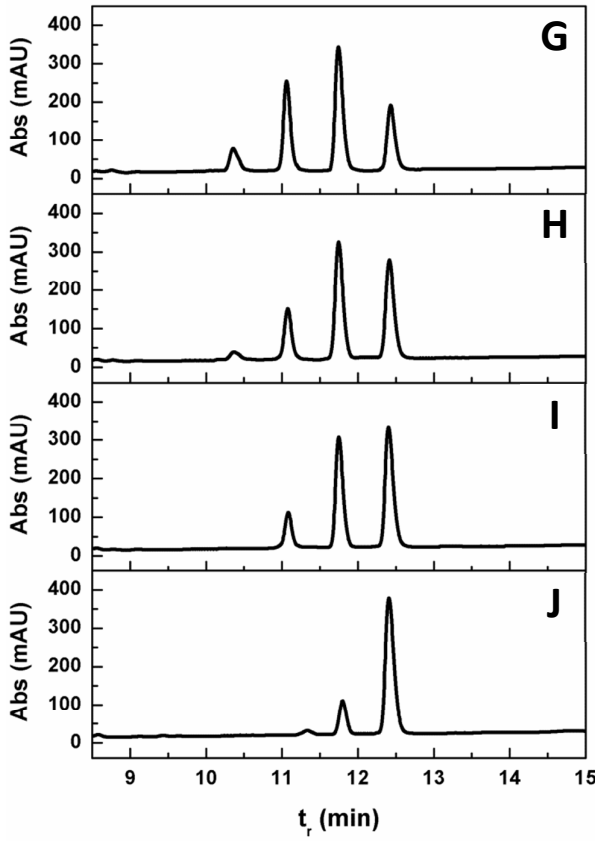

Figure S5: HPLC profiles of the photoisomerization of compound (**11**), starting from the equilibrium form (Figure S4, **F**), and irradiated at 420 nm for, respectively, 20 s (**G**), 40 s (**H**), 80 s (**I**), and 30 min (**J**).

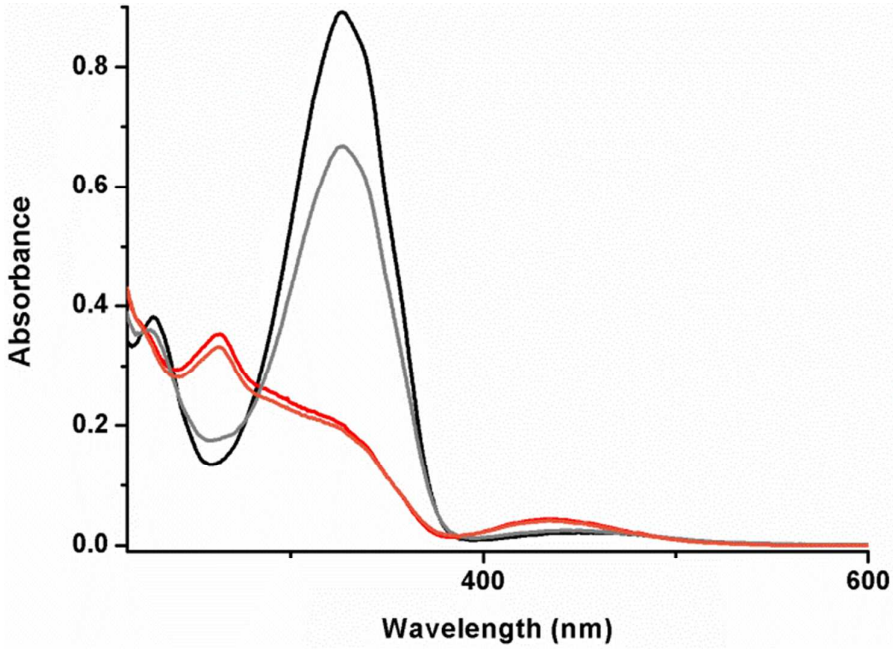

Figure S6: UV-Vis absorption spectra of compound (**11**) in MeOH showing the reversible *trans/cis* isomerization for two cycles (black lines, *trans* form; red lines, *cis* form).

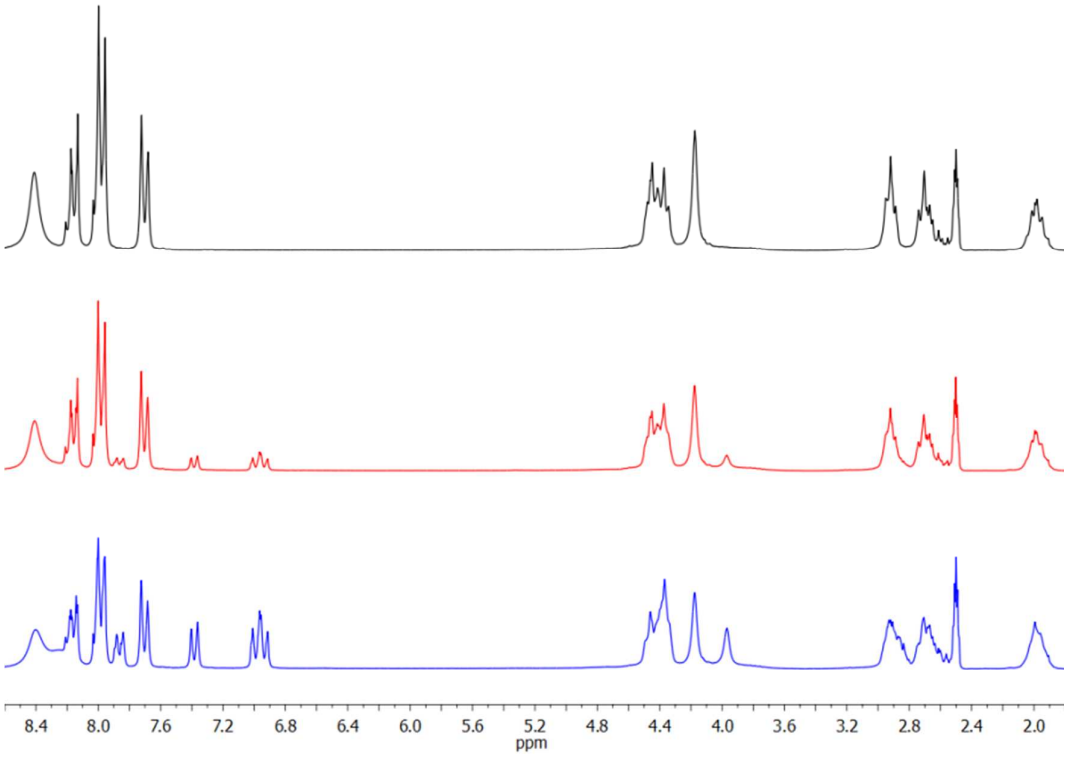

Figure S7: ^{1}H NMR spectra of compound (**11**) in DMSO, d_6 in the *trans* form (black) and after different times of irradiation at 365 nm: 30 s (red) and 1 min (blue).

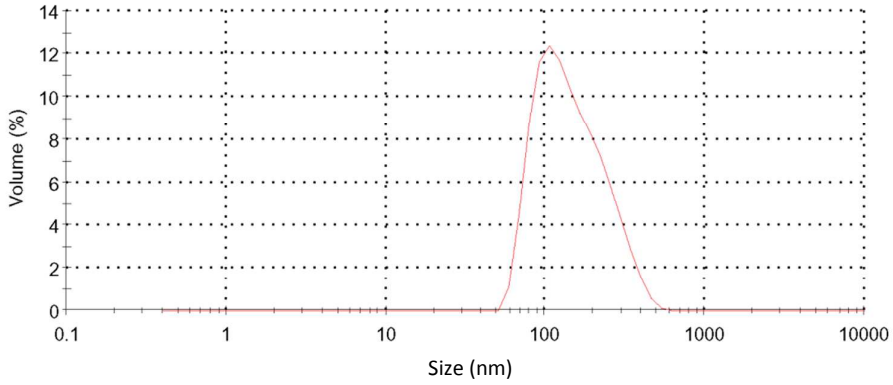

Figure S8: DLS profile of CF-encapsulated vesicles (see Figure 5, upper part, in the manuscript) from C_2-symmetry PBLG in water.

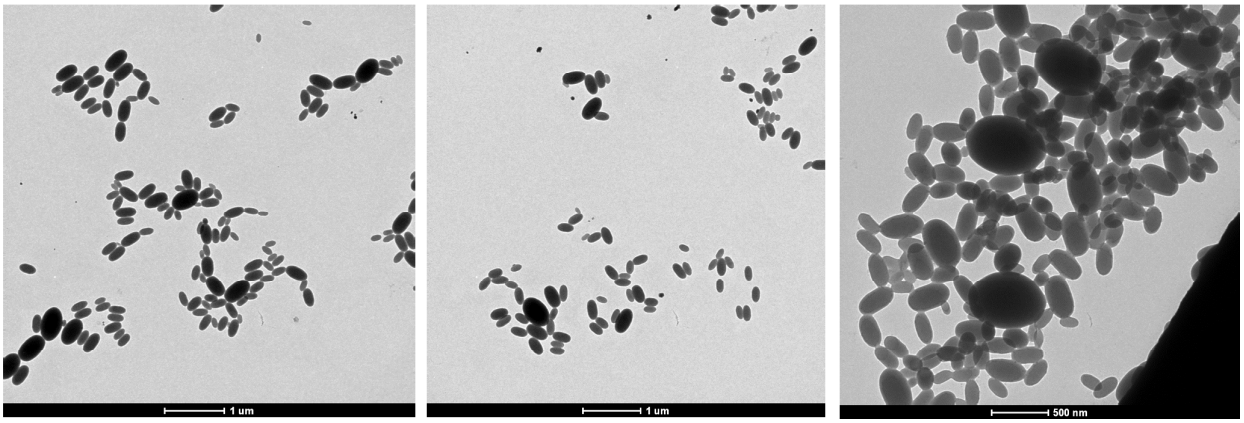

Figure S9: TEM images of self-assembled nanostructures from high MW C_3-symmetry PBLG under different times of irradiation at 365 nm (left, non-irradiated microstructures; center, after 30 min irradiation; right, after 3 h irradiation).

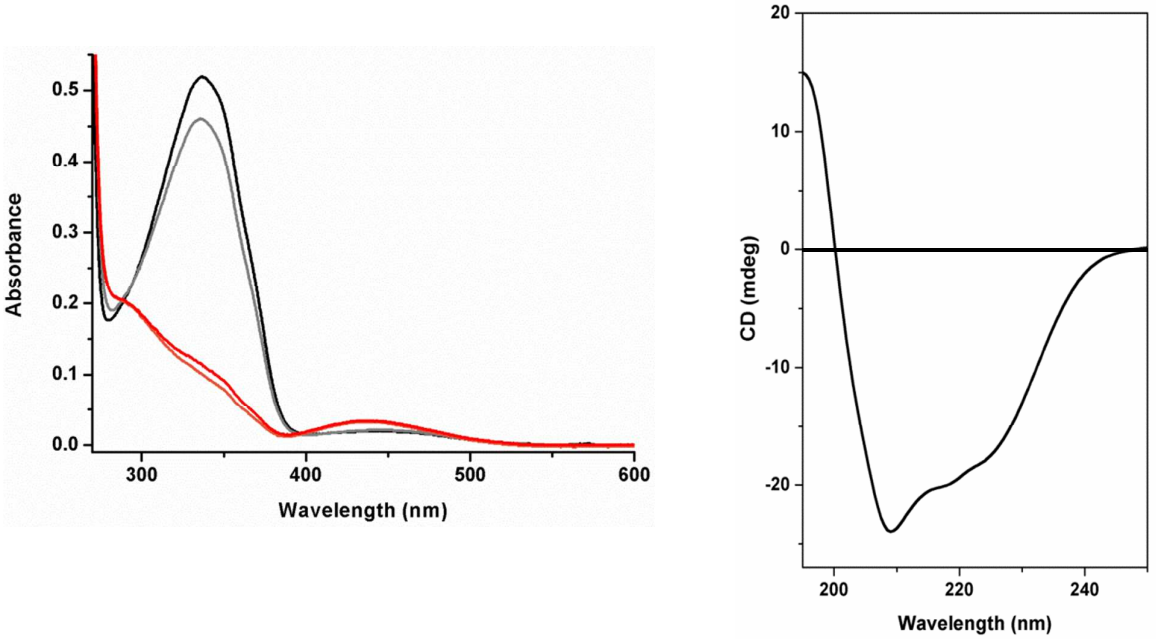

Figure S10: Left: UV-Vis absorption spectra of C_3-symmetry PBLG in DMF solution showing the reversible *trans*/*cis* isomerization for two cycles (black lines, *trans* form; red lines, *cis* form). Right: ECD spectrum of C_3-symmetry PBLG in HFIP solution.

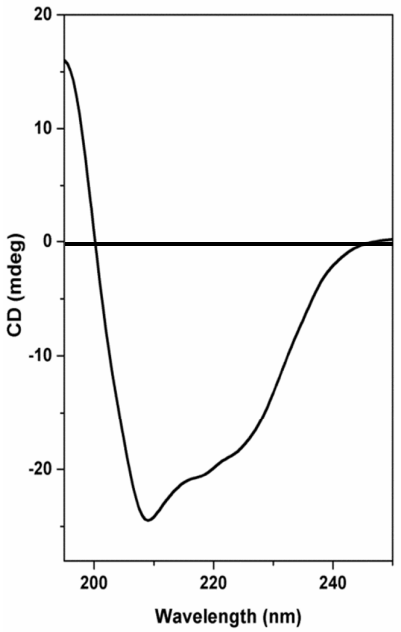

Figure S11: ECD spectrum of C_3-symmetry PBLG (high MW) in HFIP solution.

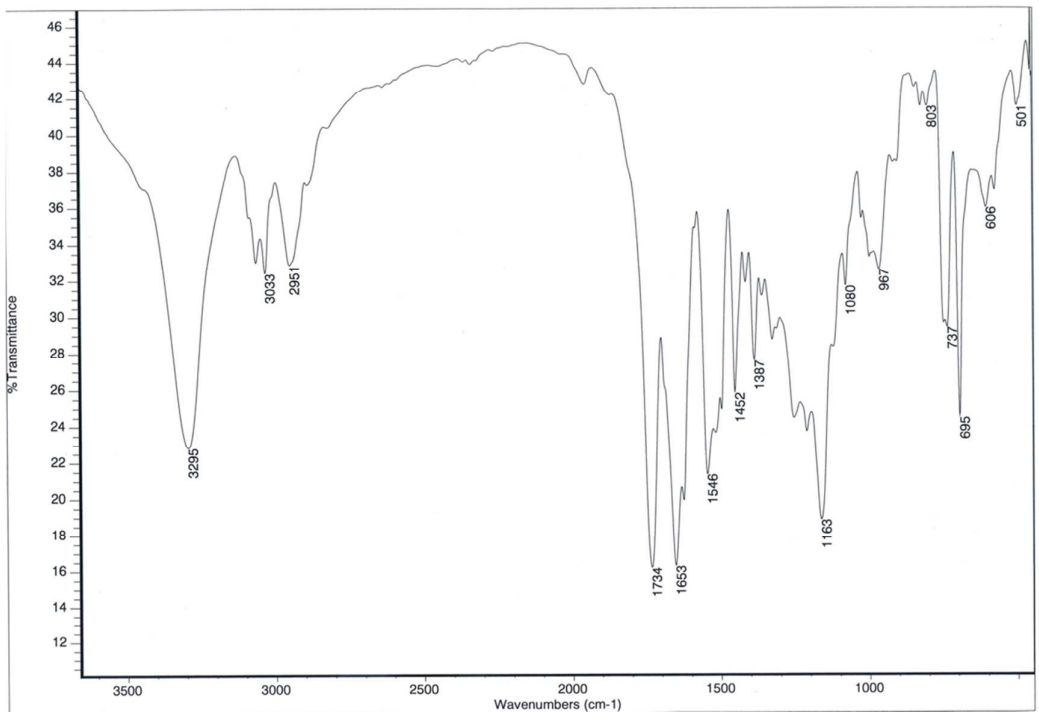

Figure S12: FT-IR absorption spectrum of C_2-symmetry PBLG in KBr disk.

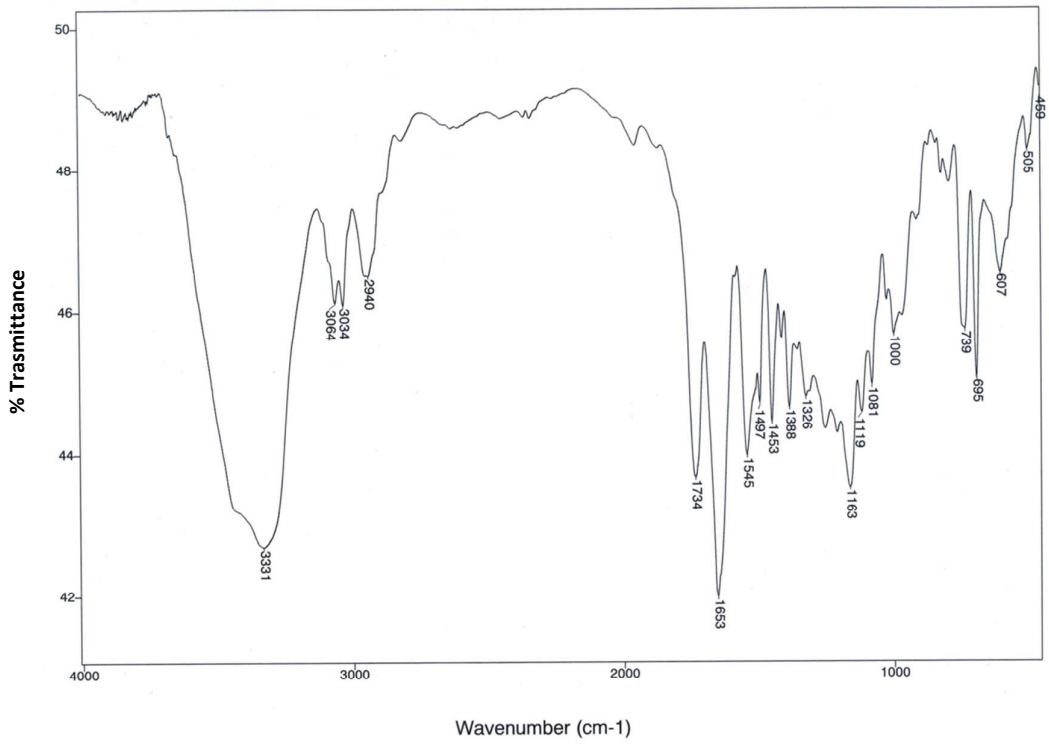

Figure S13: FT-IR absorption spectrum of C_3-symmetry PBLG in KBr disk.

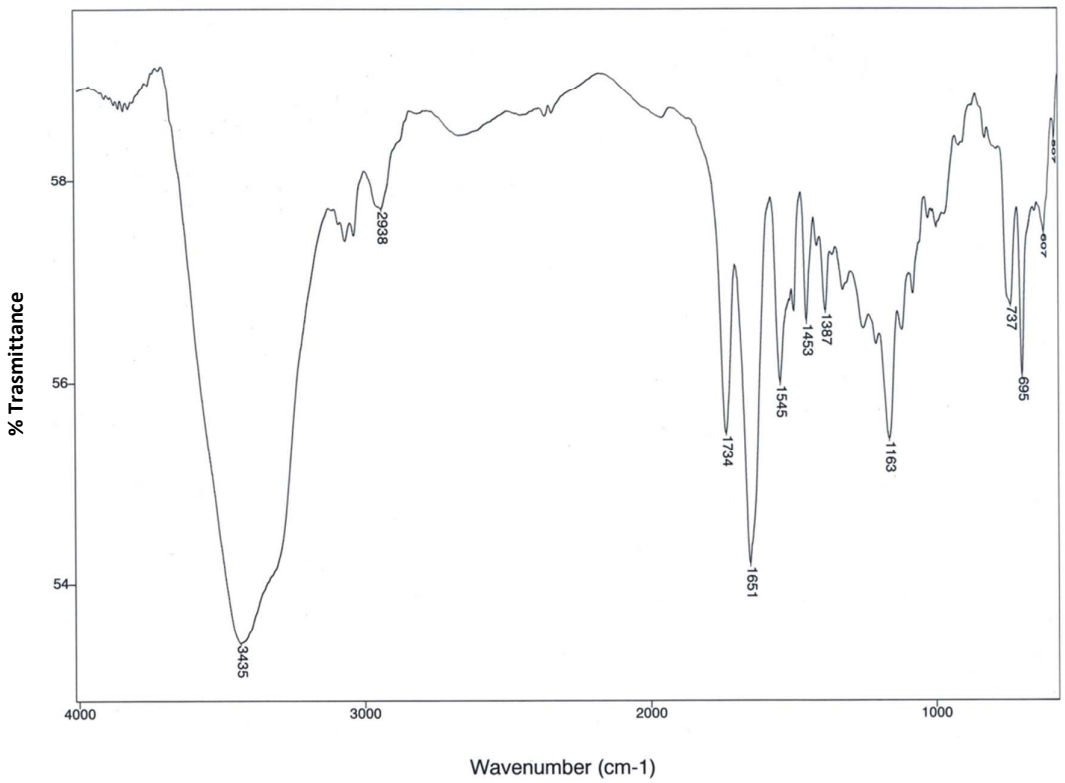

Figure S14: FT-IR absorption spectrum of C_3-symmetry PBLG (high MW) in KBr disk.

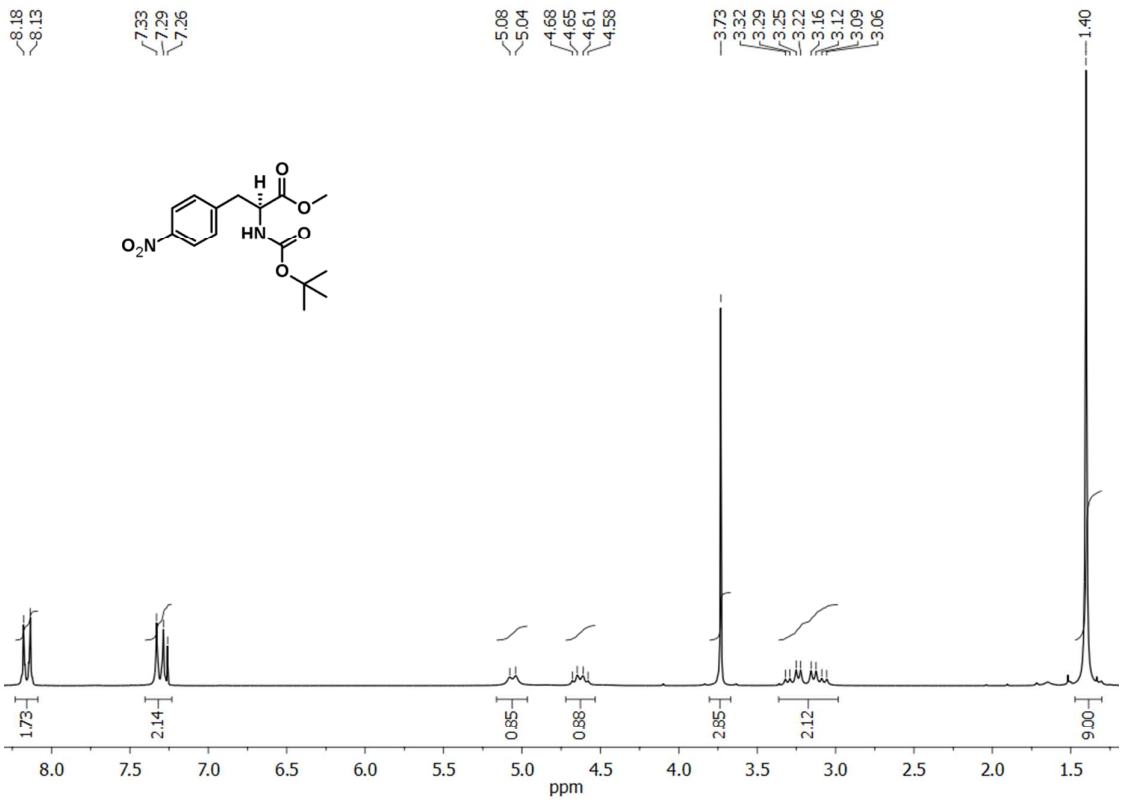

Figure S15: ^{1}H NMR spectrum of compound (**1**) in $CDCl_3$.

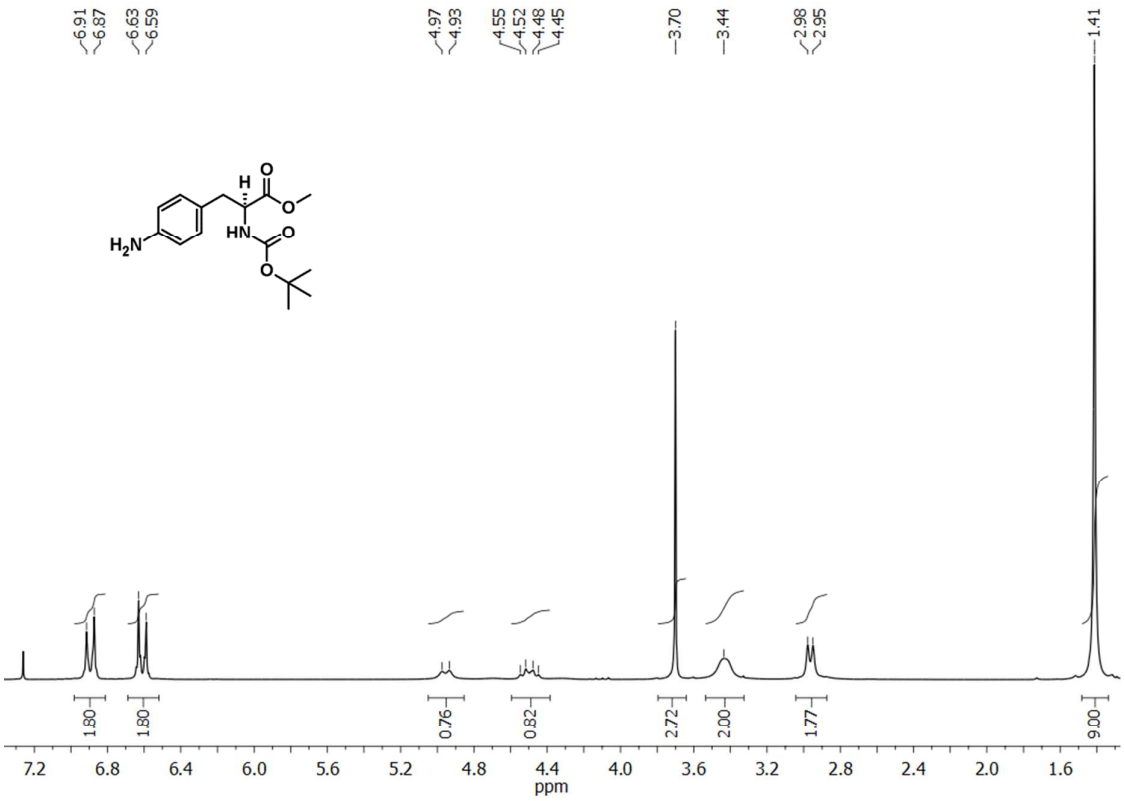

Figure S16: ^{1}H NMR spectrum compound of (**3**) in $CDCl_3$.

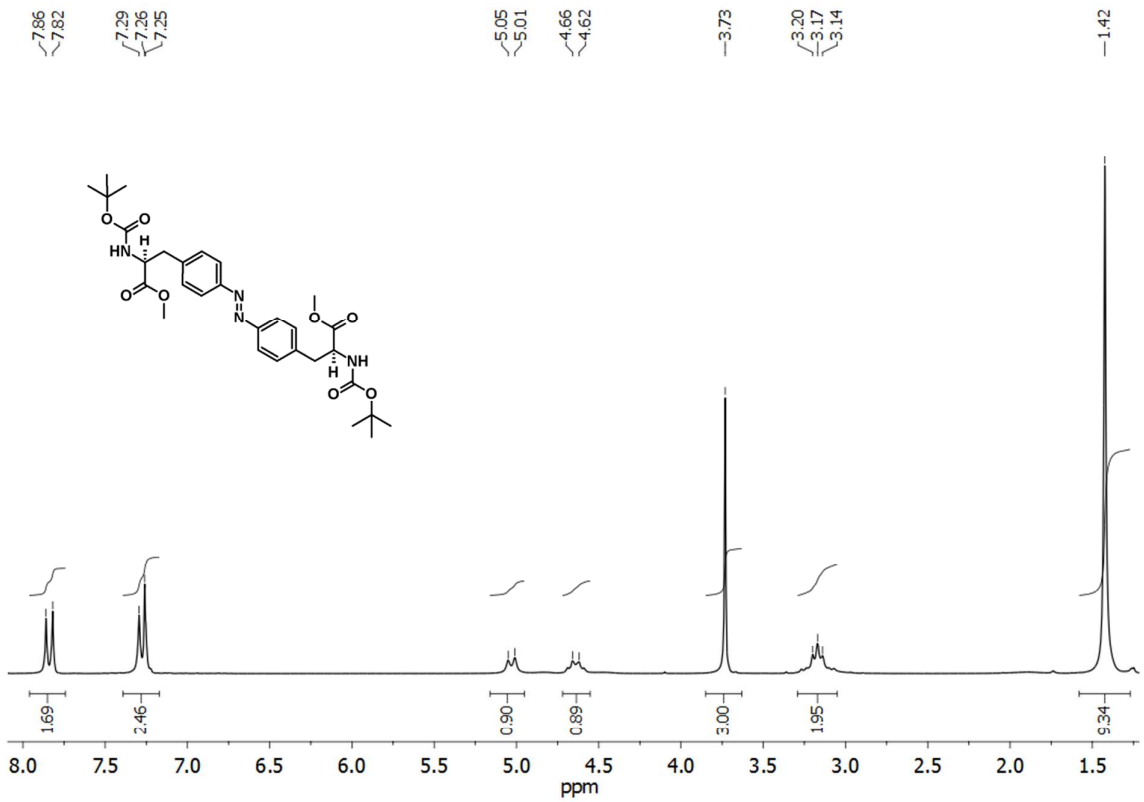

Figure S17: ^{1}H NMR spectrum of compound (**4**) in $CDCl_3$.

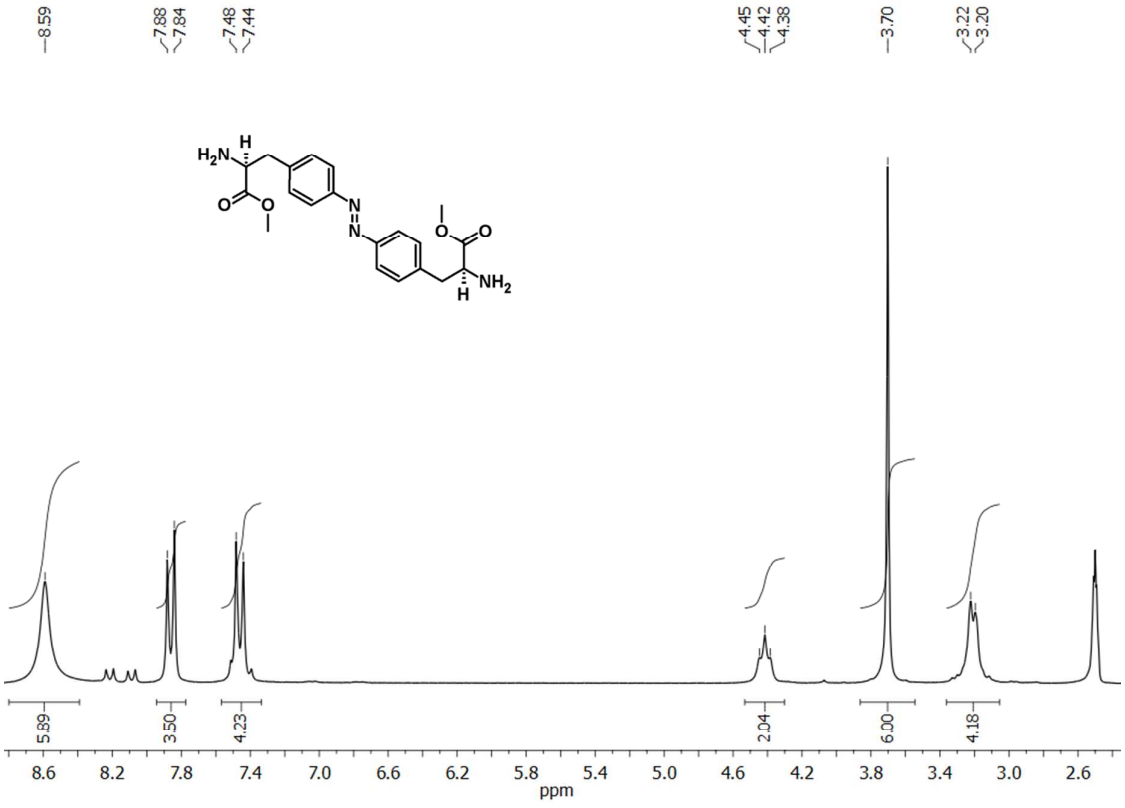

Figure S18: ^{1}H NMR spectrum of compound (**5**) in DMSO, d_6.

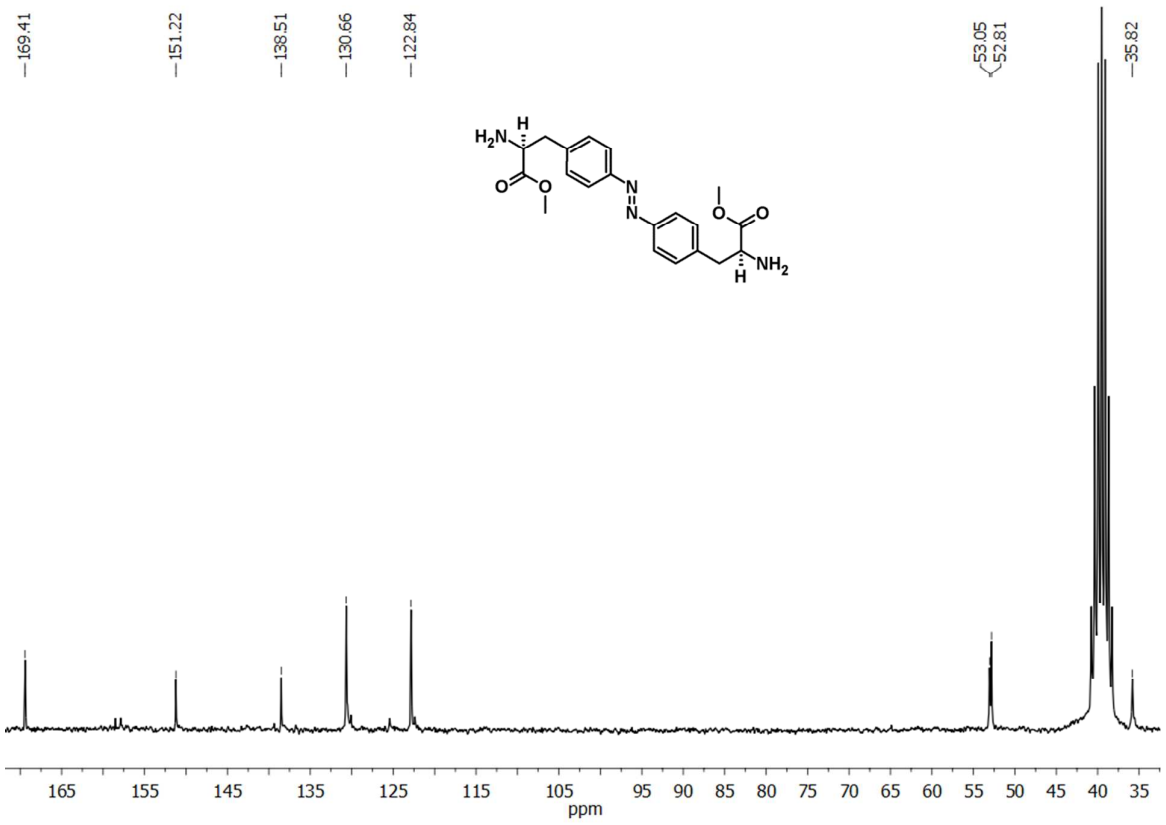

Figure S19: ^{13}C NMR spectrum of compound (**5**) in DMSO, d_6.

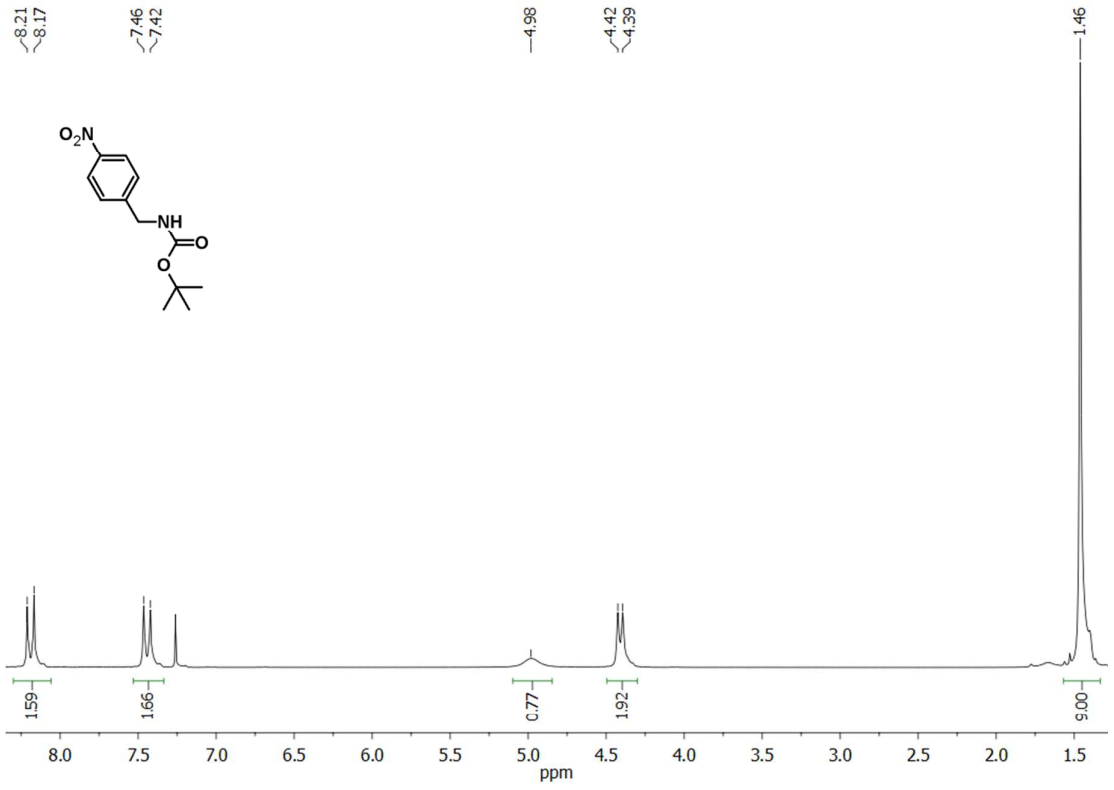

Figure S20: ^{1}H NMR spectrum of compound (**6**) in $CDCl_3$.

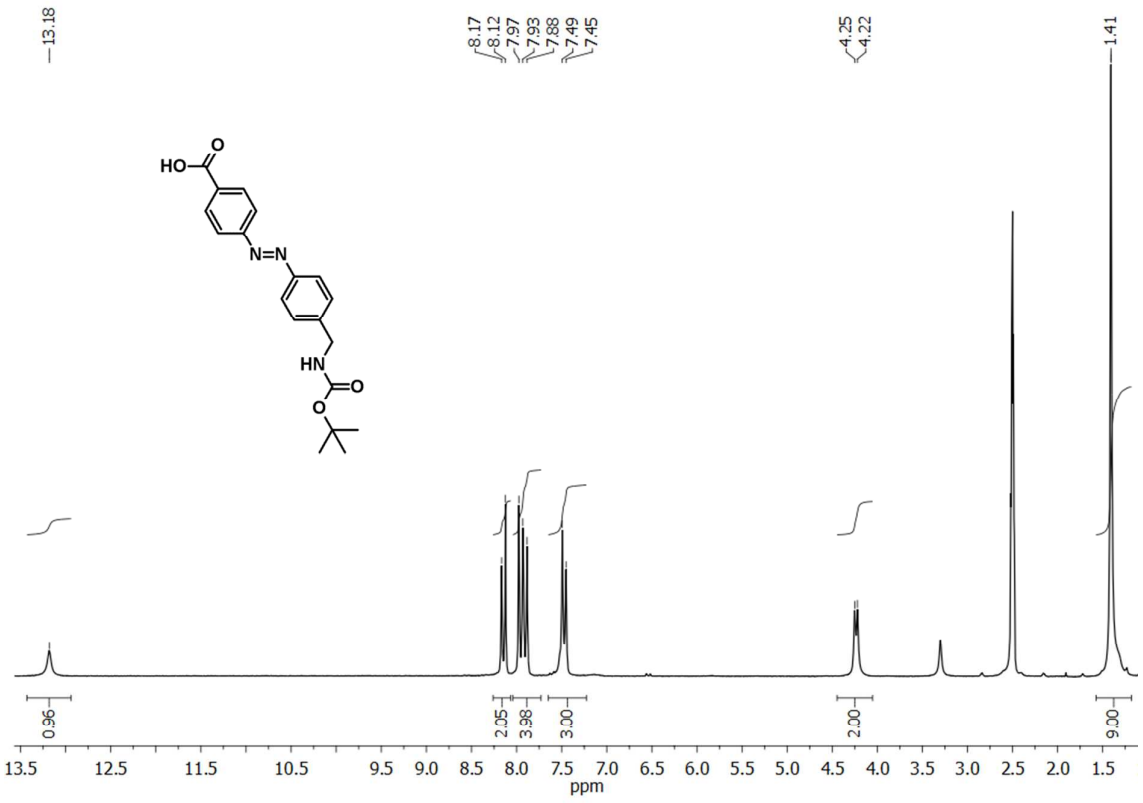

Figure S21: ^{1}H NMR spectrum of compound (**8**) in DMSO, d_6.

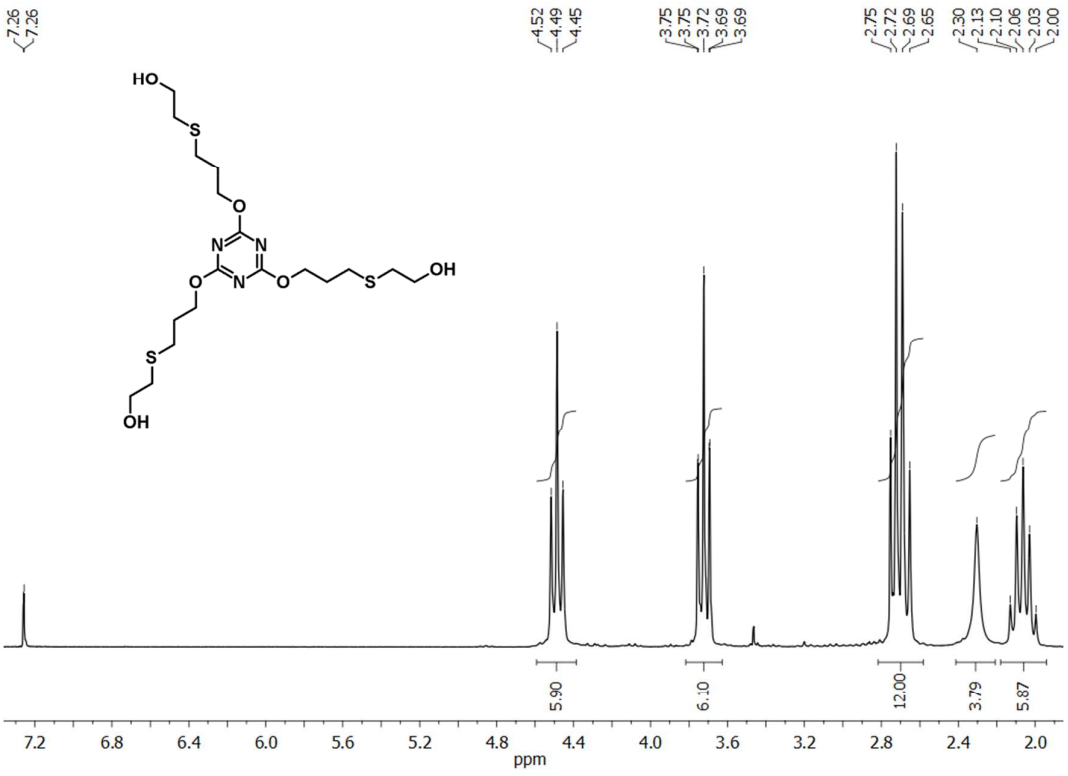

Figure S22: ^{1}H NMR spectrum of compound (**9**) in $CDCl_3$.

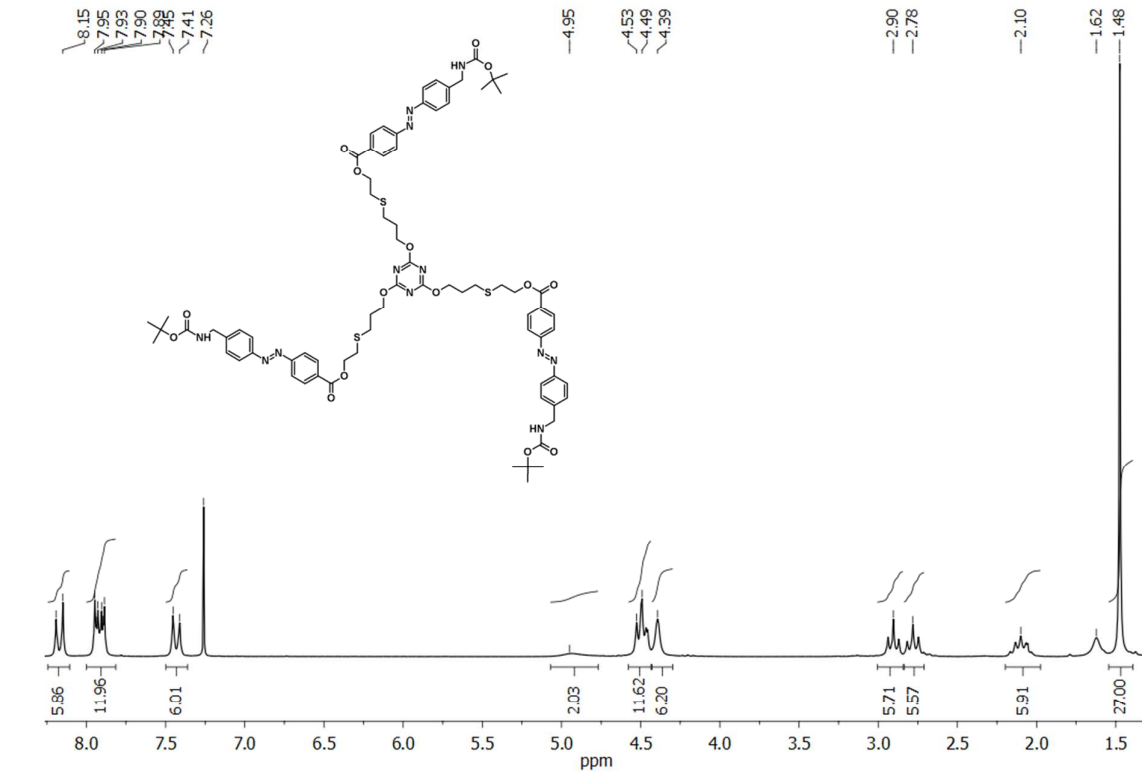

Figure S23: ^{1}H NMR spectrum of compound (**10**) in $CDCl_3$.

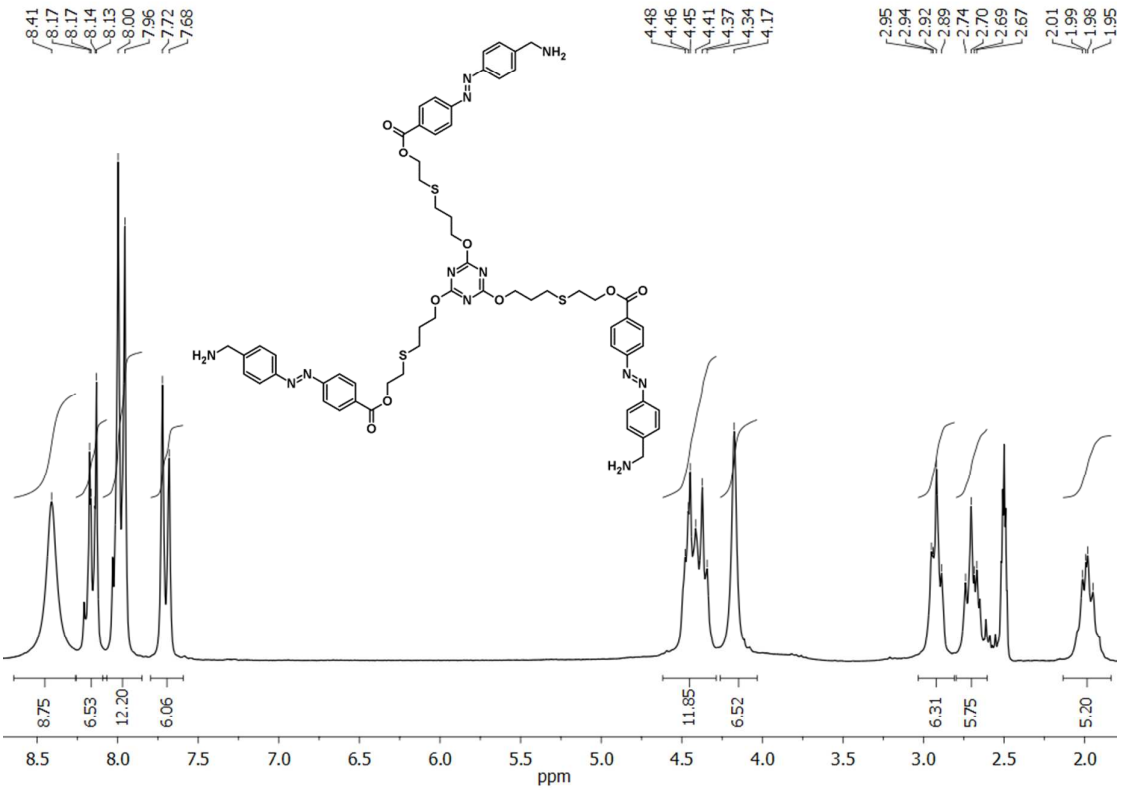

Figure S24: ^{1}H NMR spectrum of compound (**11**) in DMSO, d_6.

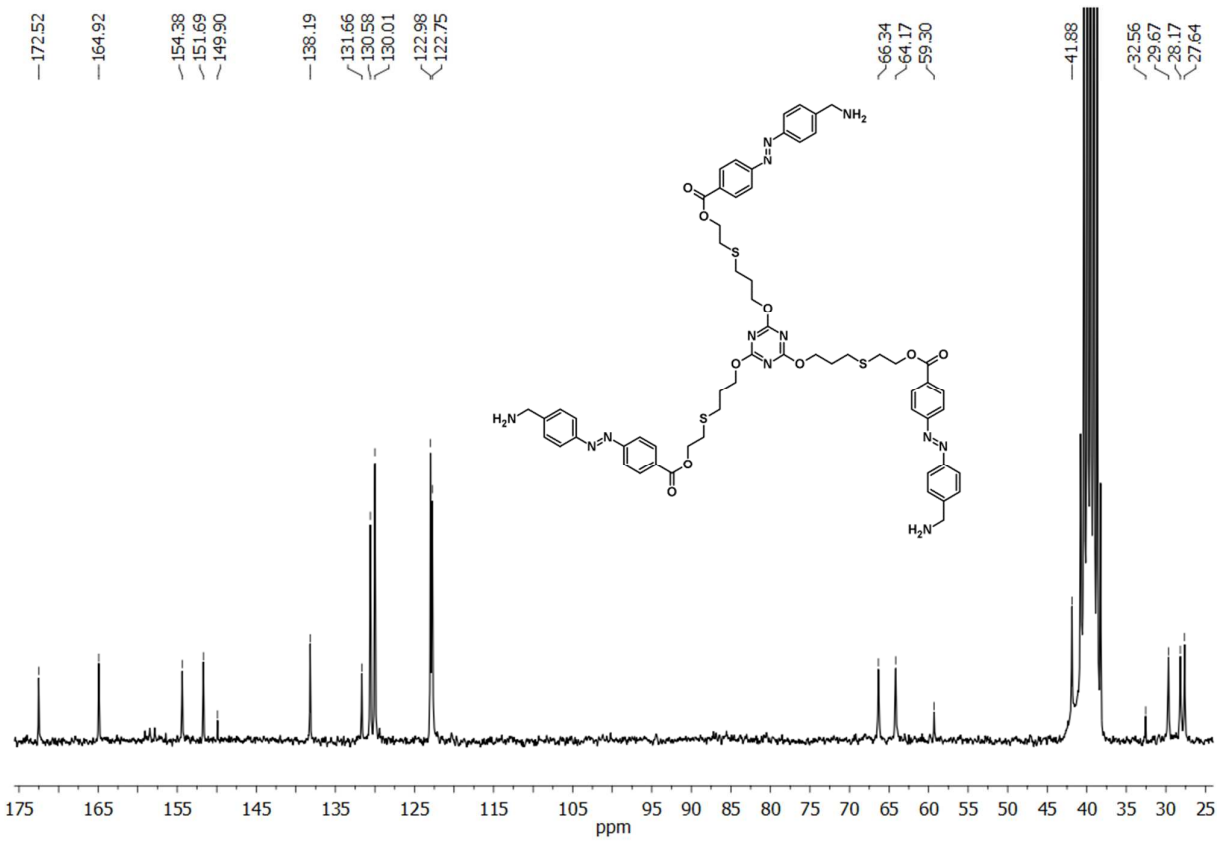

Figure S25: ^{13}C NMR spectrum of compound (**11**) in DMSO, d_6.

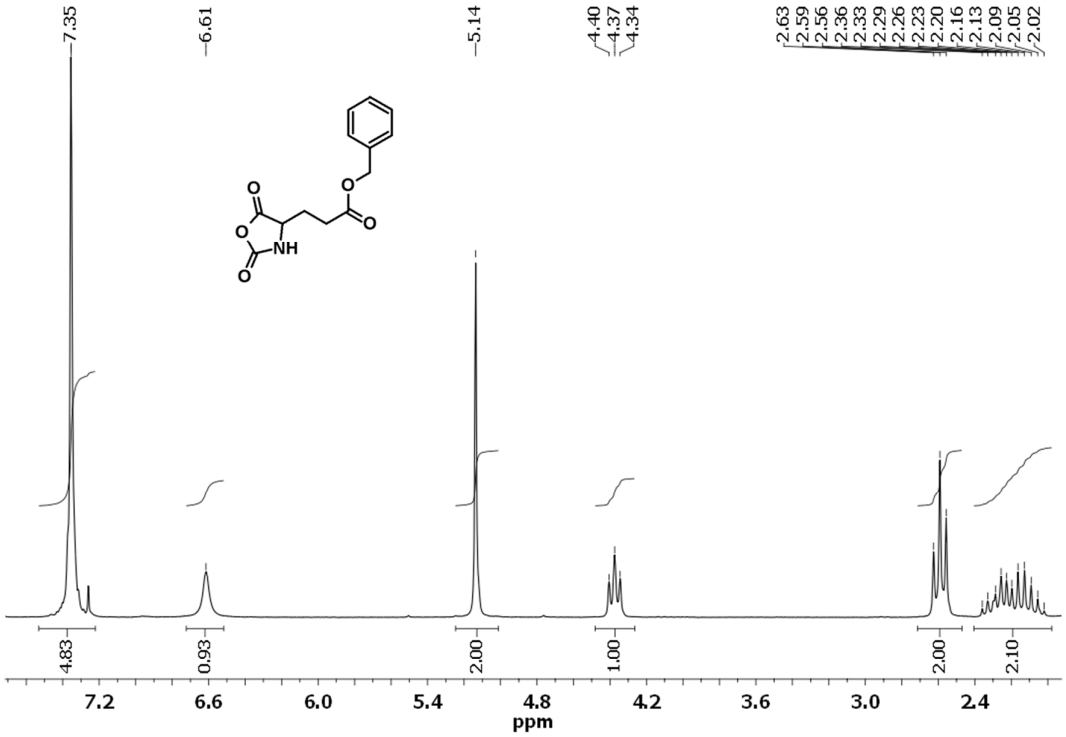

Figure S26: ^{1}H-NMR spectrum of BLG-NCA in $CDCl_3$.