Electronic Supplementary Information:

A series of AB_2-type second-order nonlinear optical (NLO) polyaryleneethynylenes: using different end-capped spacers with adjustable bulk to achieve high NLO coefficients

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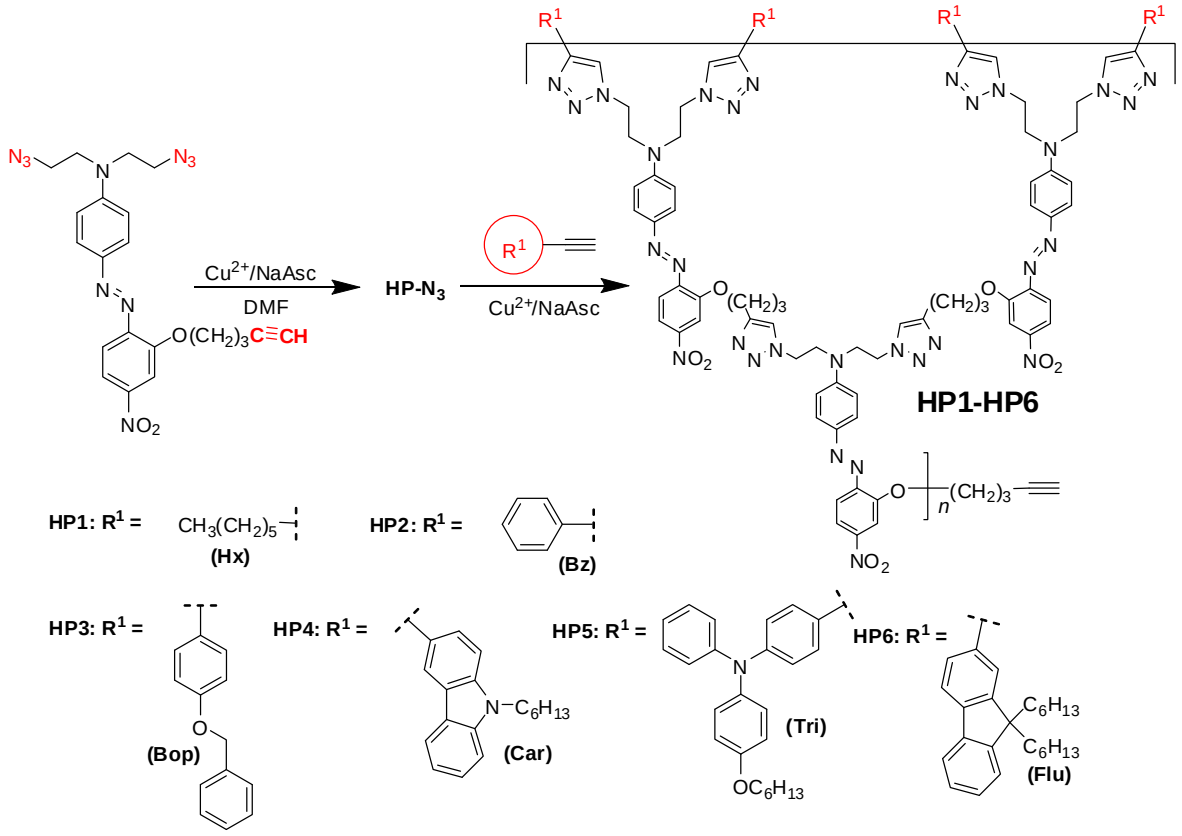


Chart S1 The structure and synthesis of **HP1-HP6**.

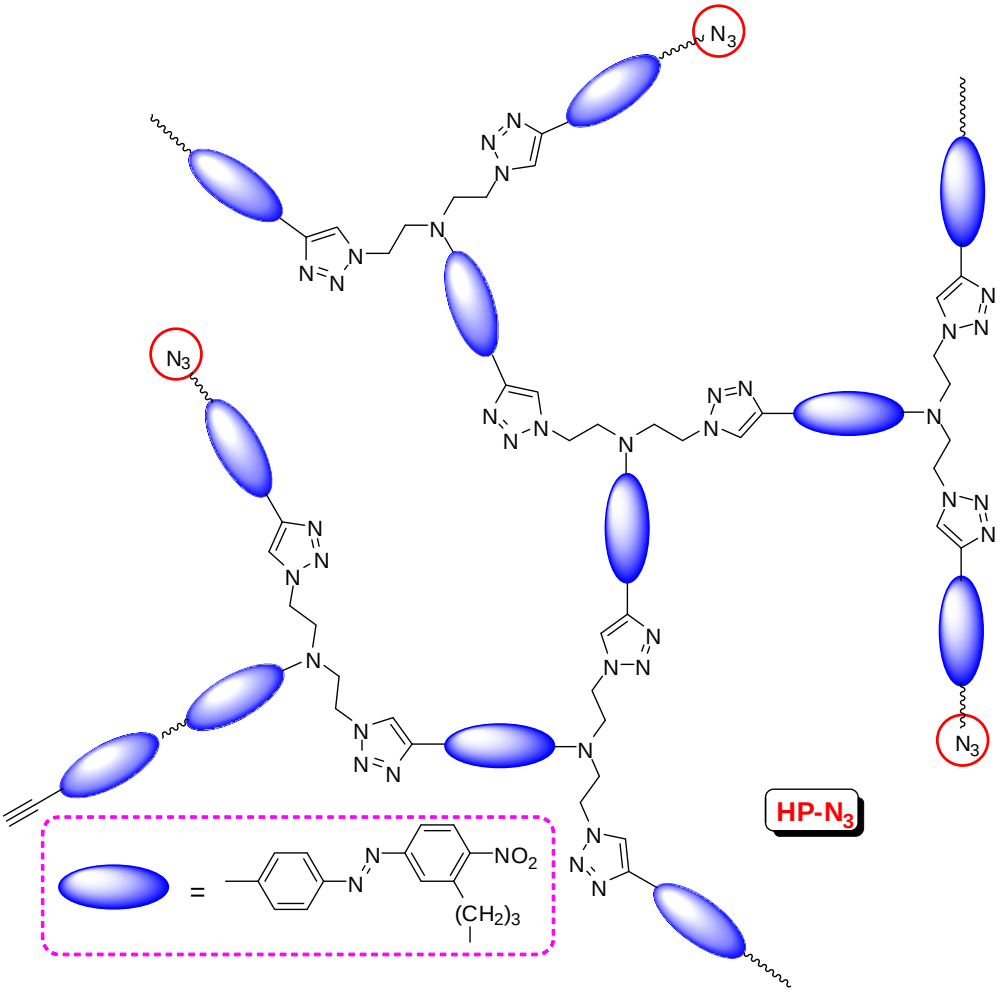
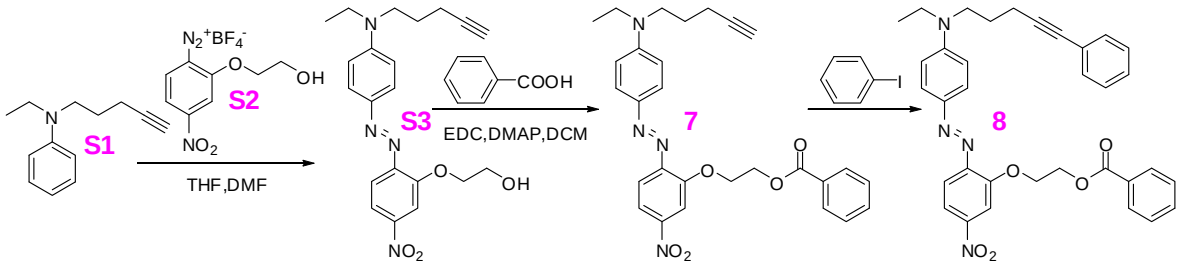


Chart S2 The structure of **HP-N_3**.



Scheme S1 The synthesis of model molecules **7** and **8**.

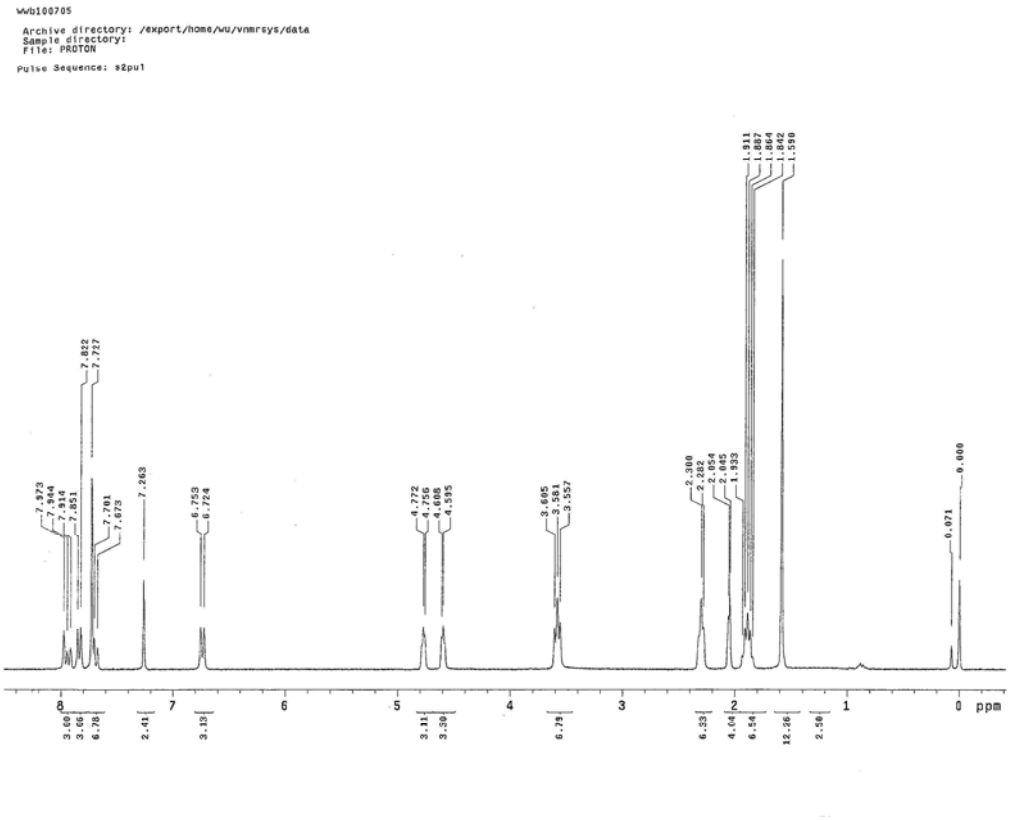


Fig. S1 ^{1}H NMR spectrum of AB_2-type monomer **3** in chloroform-*d*.

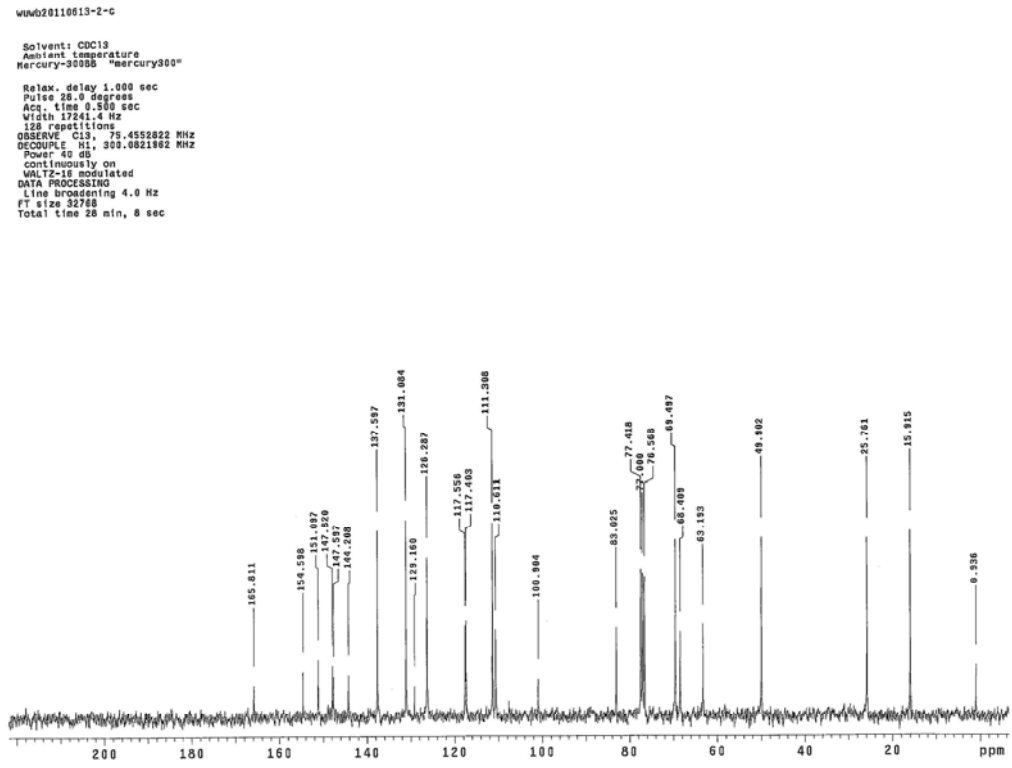


Fig. S2 ^{13}C NMR spectrum of AB_2-type monomer **3** in chloroform-*d*.

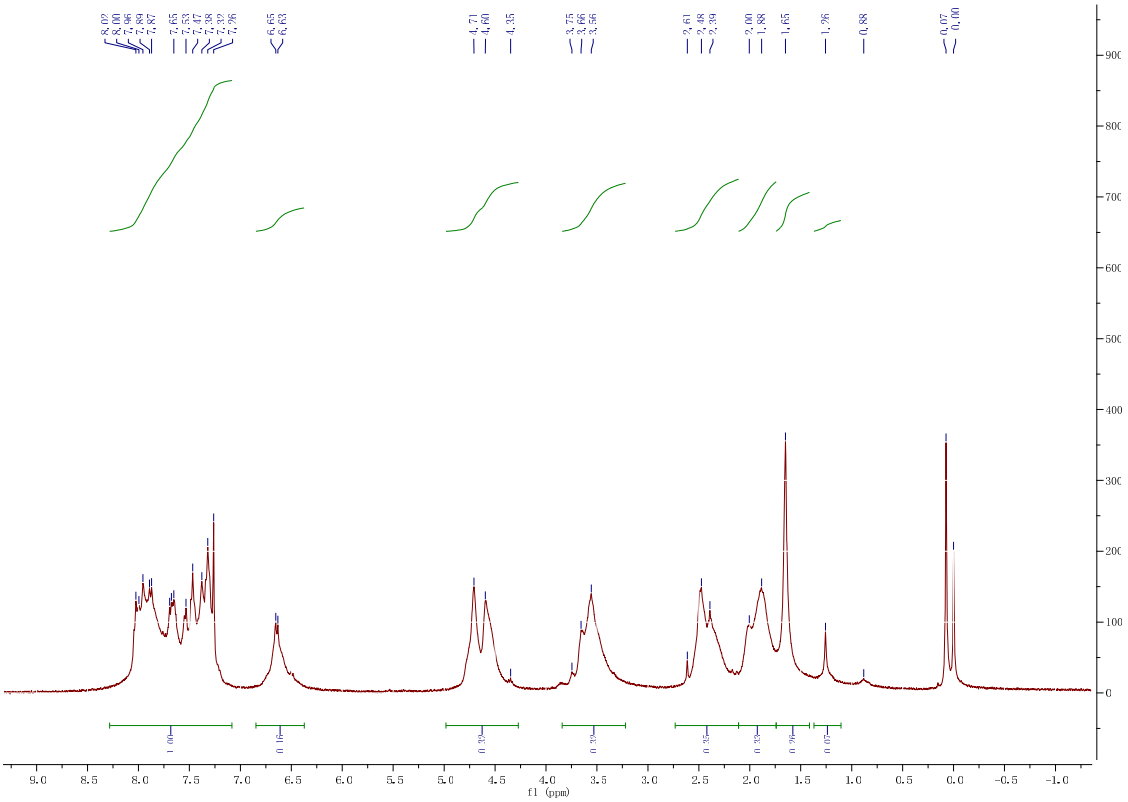


Fig. S3 ^{1}H NMR spectrum of hyperbranched polymer **P1** in chloroform-*d*.

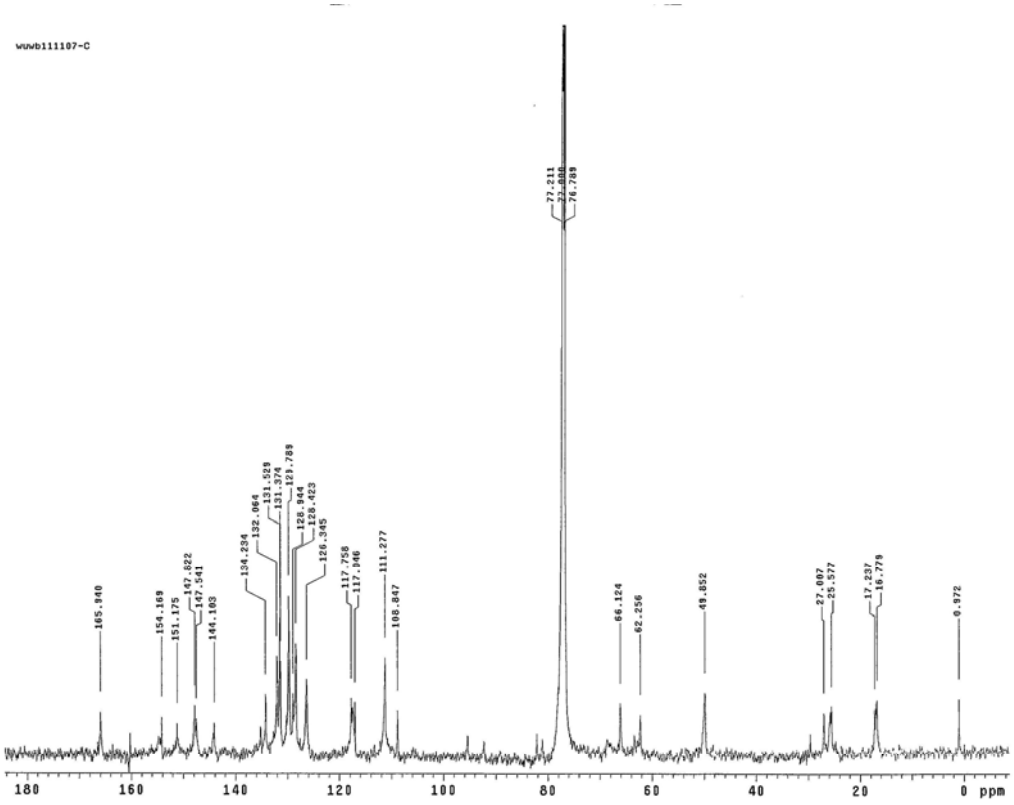


Fig. S4 ^{13}C NMR spectrum of hyperbranched polymer **P1** in chloroform-*d*.

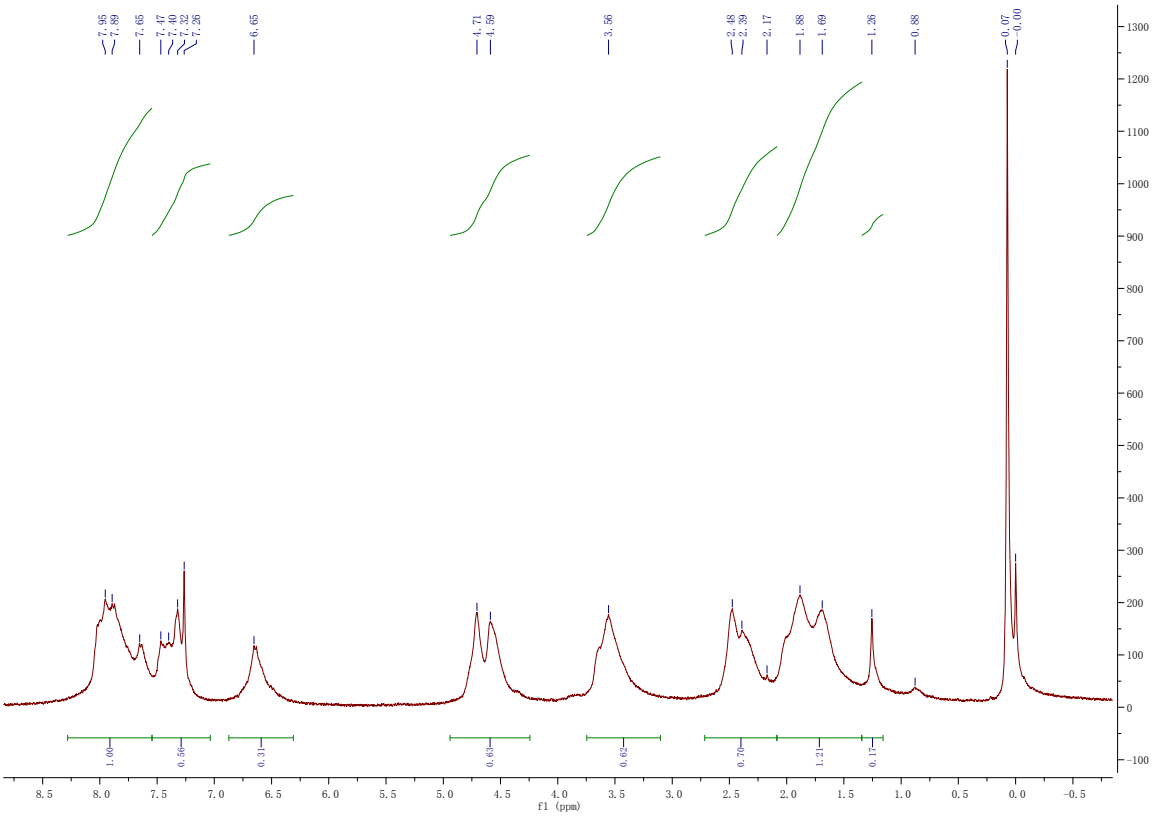


Fig. S5 ^{1}H NMR spectrum of hyperbranched polymer **P2** in chloroform-*d*.

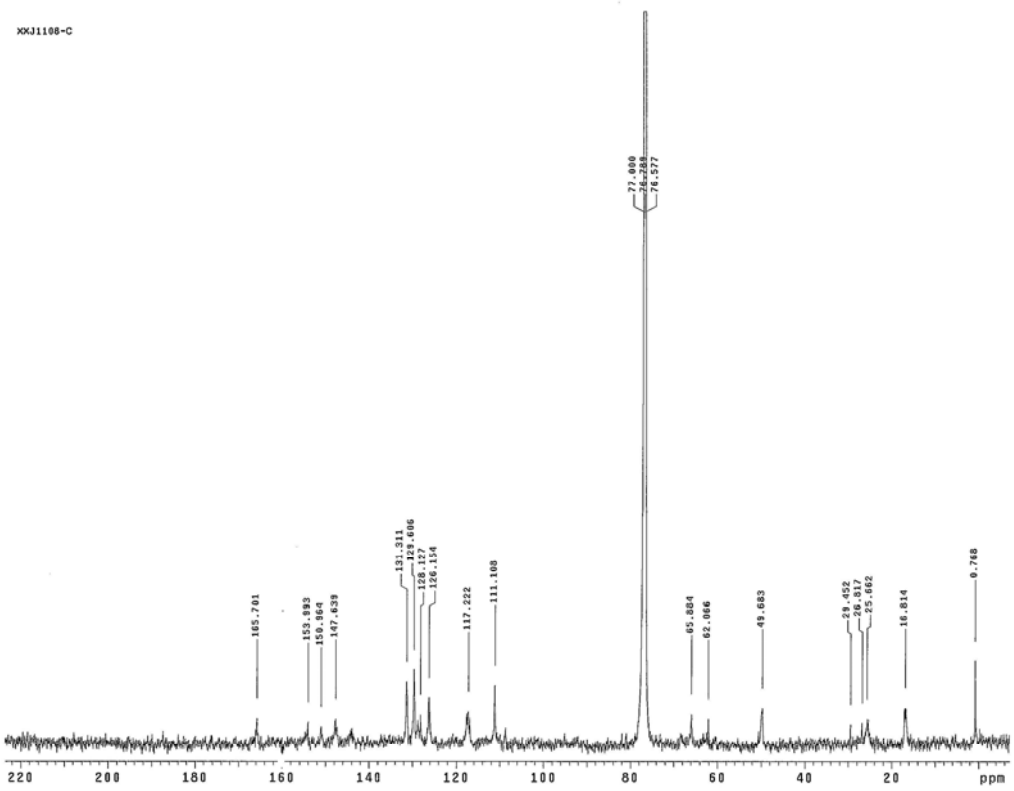


Fig. S6 ^{13}C NMR spectrum of hyperbranched polymer **P2** in chloroform-*d*.

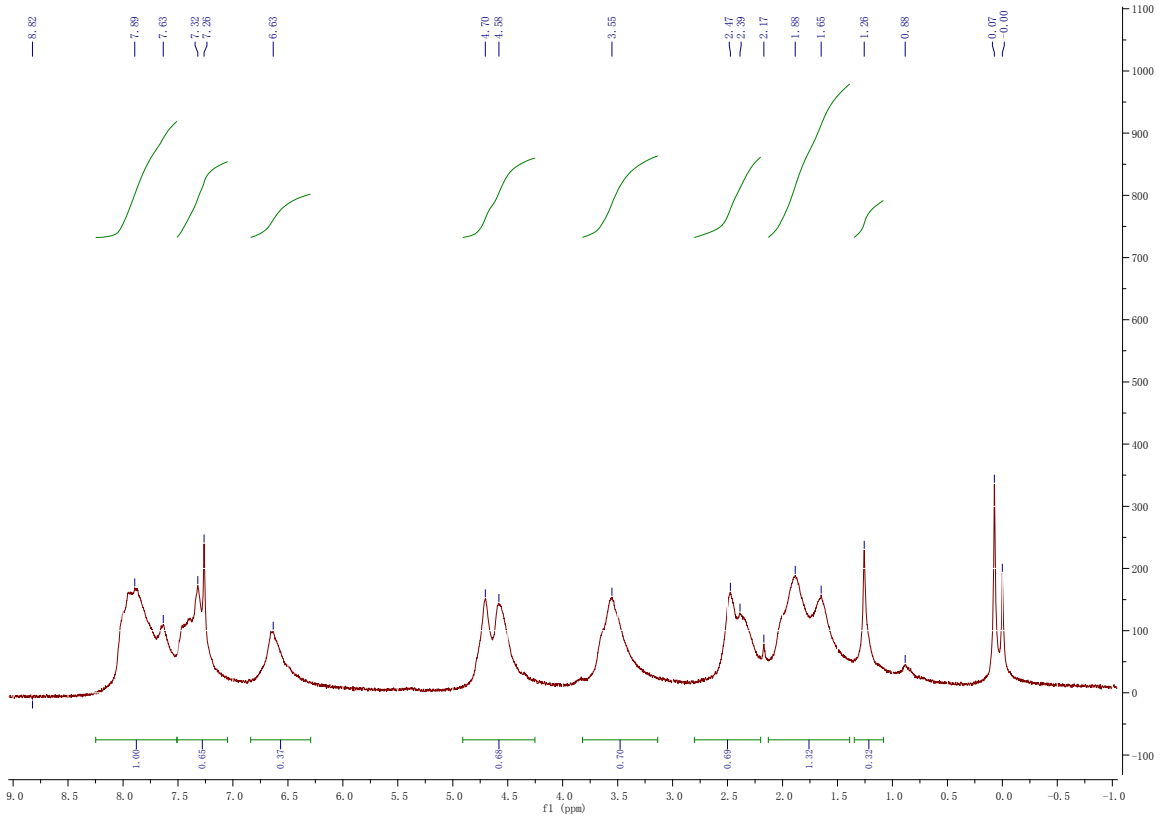


Fig. S7 ^{1}H NMR spectrum of hyperbranched polymer **P3** in chloroform-*d*.

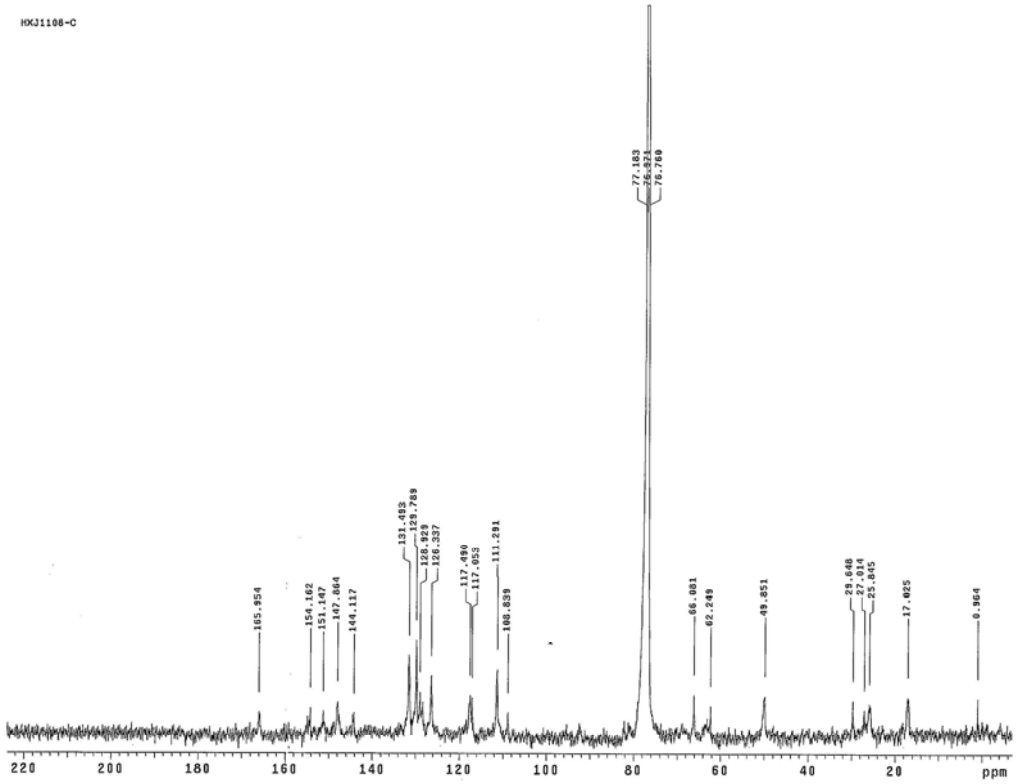


Fig. S8 ^{13}C NMR spectrum of hyperbranched polymer **P3** in chloroform-*d*.

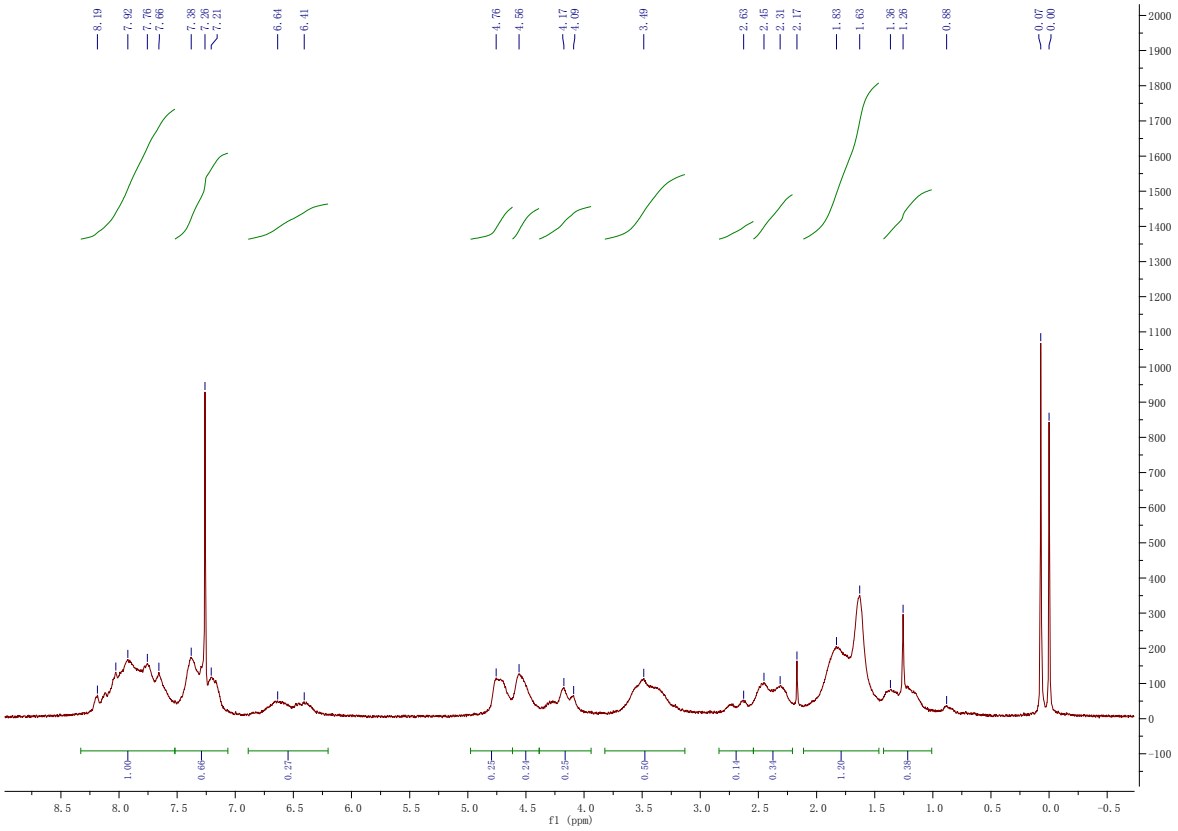


Fig. S9 ^{1}H NMR spectrum of hyperbranched polymer **P4** in chloroform-*d*.

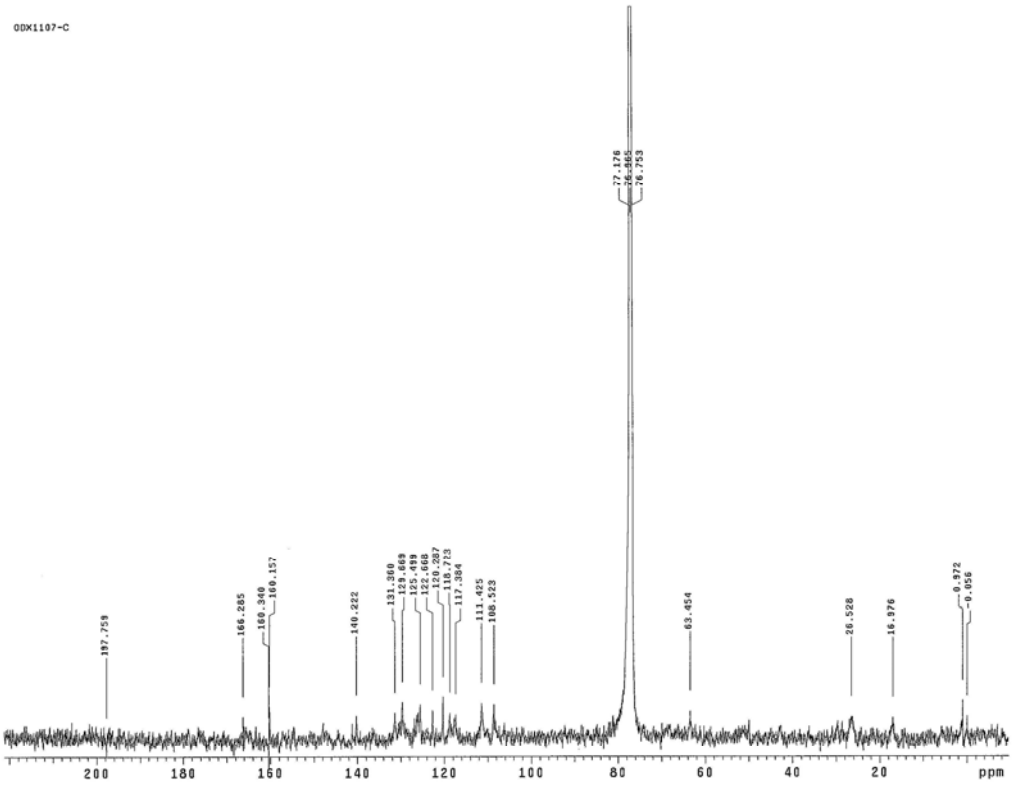


Fig. S10 ^{13}C NMR spectrum of hyperbranched polymer **P4** in chloroform-*d*.

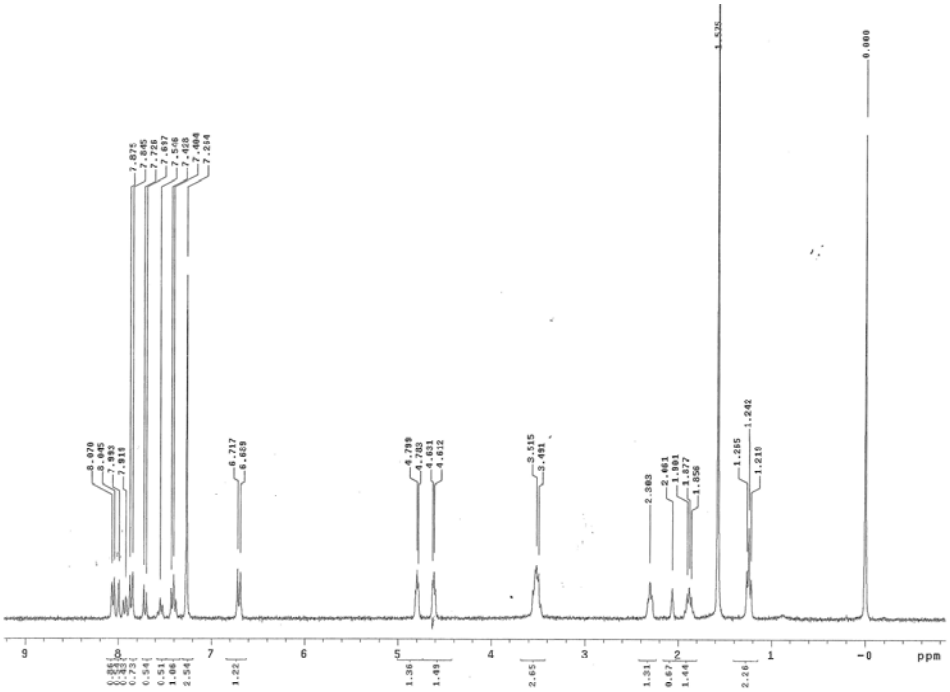


Fig. S11 ^{1}H NMR spectrum of compound **7** in chloroform-*d*.

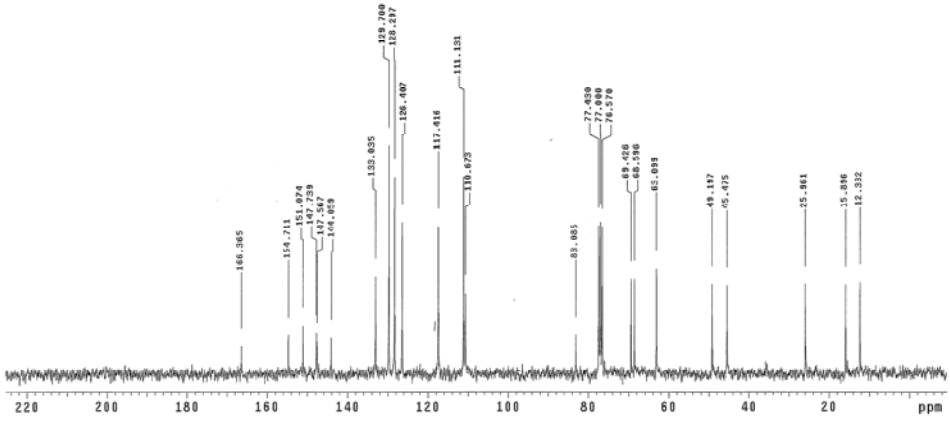


Fig. S12 ^{13}C NMR spectrum of compound **7** in chloroform-*d*.

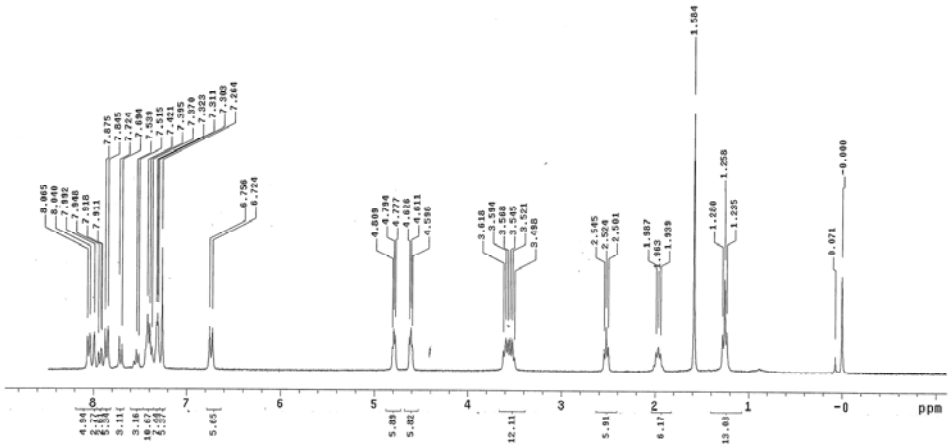


Fig. S13 ^{1}H NMR spectrum of compound **8** in chloroform-*d*.

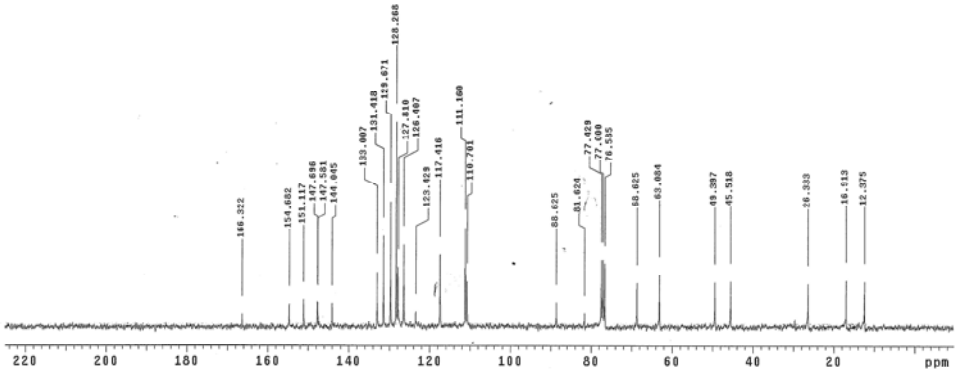


Fig. S14 ^{13}C NMR spectrum of compound **8** in chloroform-*d*.

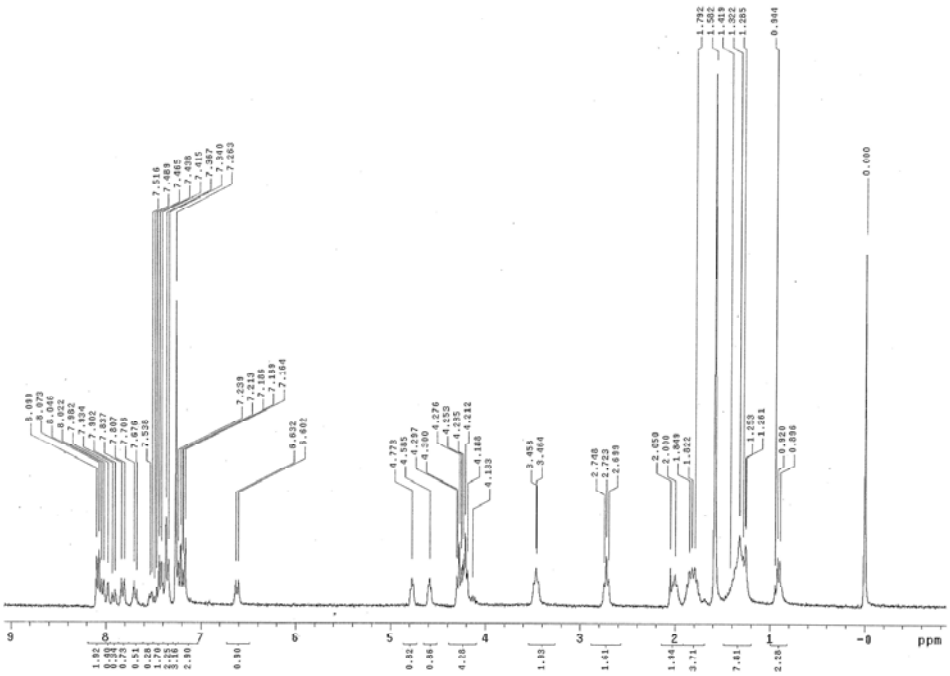


Fig. S15 ^{1}H NMR spectrum of compound **10** in chloroform-*d*.

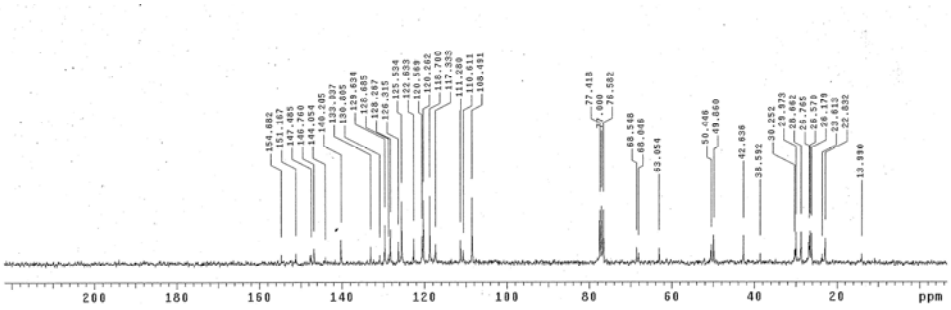


Fig. S16 ^{13}C NMR spectrum of compound **10** in chloroform-*d*.

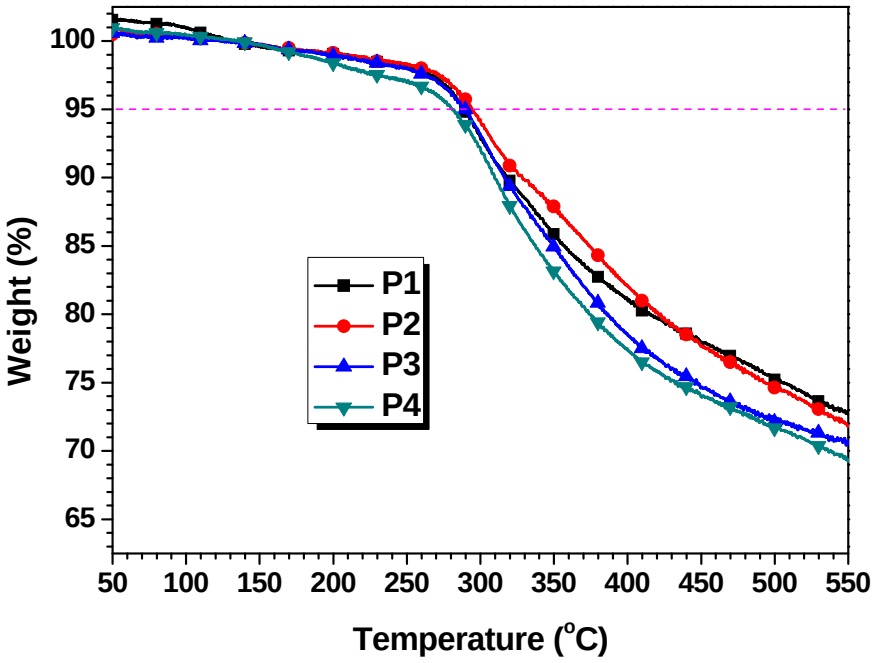


Fig. S17 TGA thermograms of polymers **P1**-**P4**, measured in nitrogen at a heating rate of 10 °C/min.

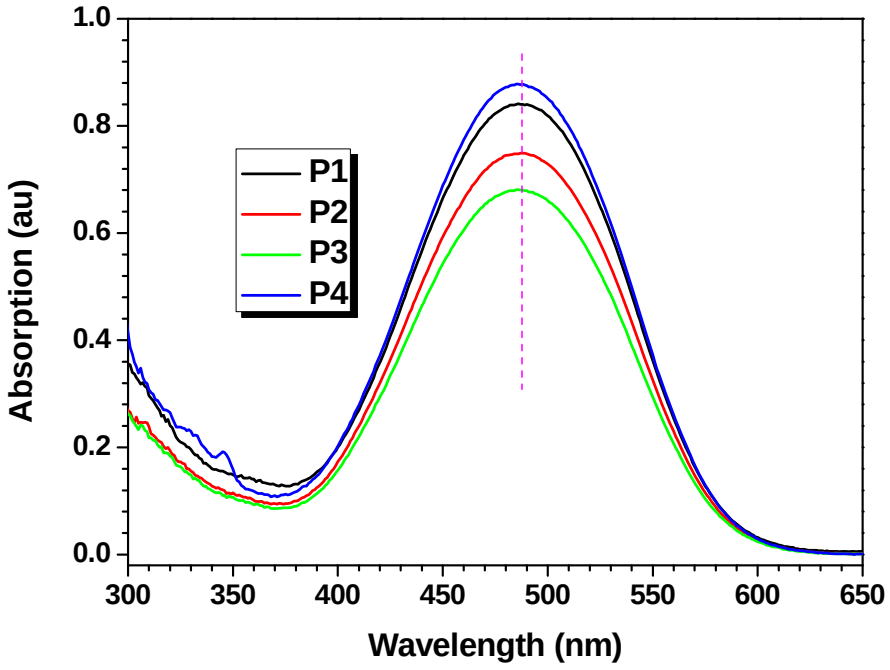


Fig. S18 UV-vis spectra of polymers **P1**-**P4** in THF (0.02 mg/mL).

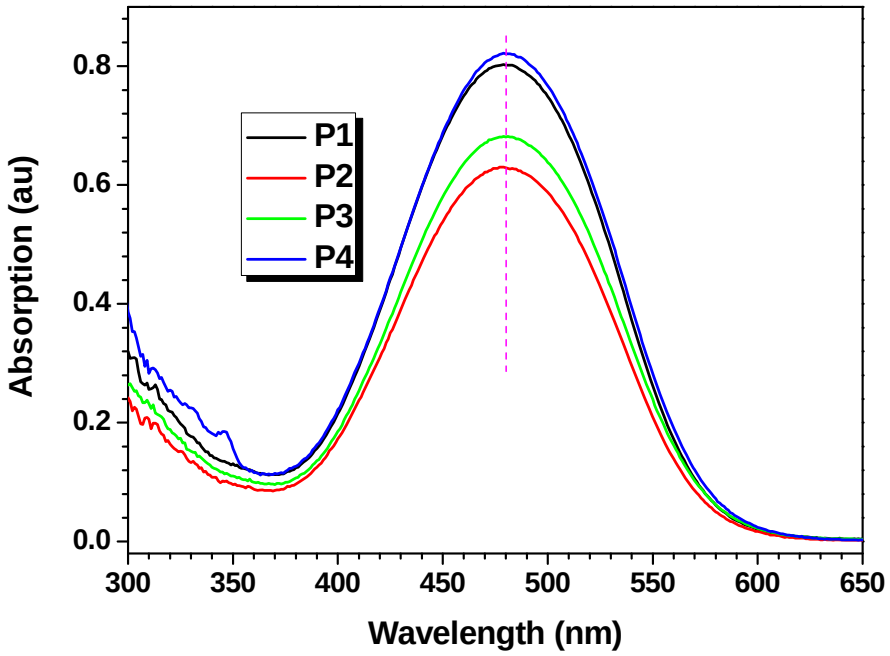


Fig. S19 UV-vis spectra of polymers **P1**-**P4** in 1,4-dioxane (0.02 mg/mL).

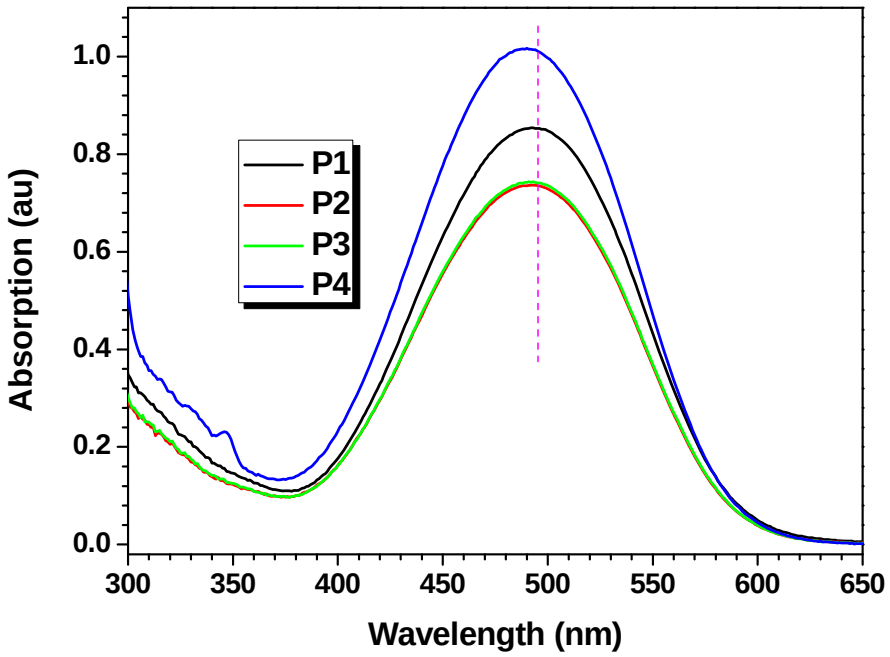


Fig. S20 UV-vis spectra of polymers **P1**-**P4** in chloroform (0.02 mg/mL).

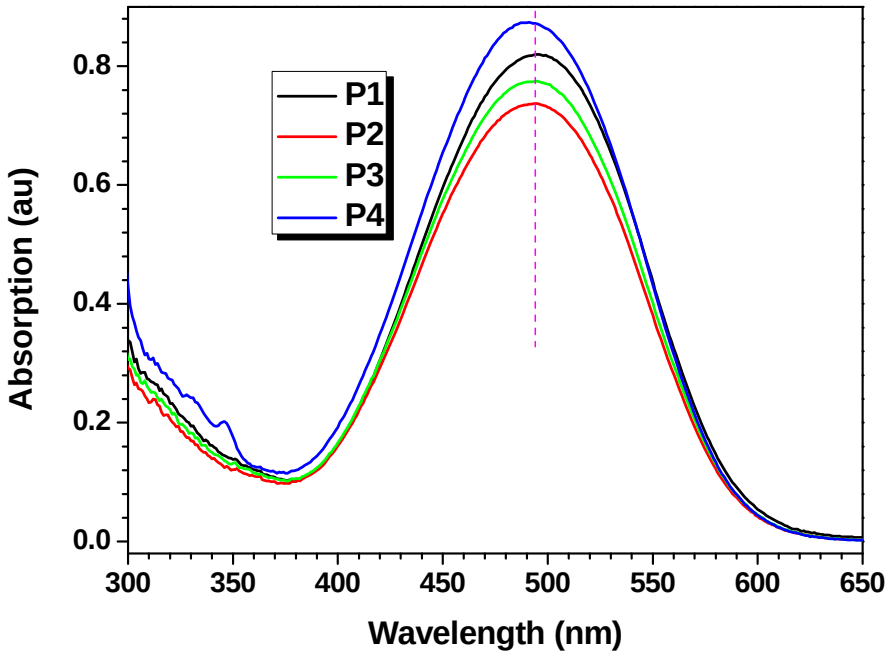


Fig. S21 UV-vis spectra of polymers **P1**-**P4** in dichloromethane (0.02 mg/mL).

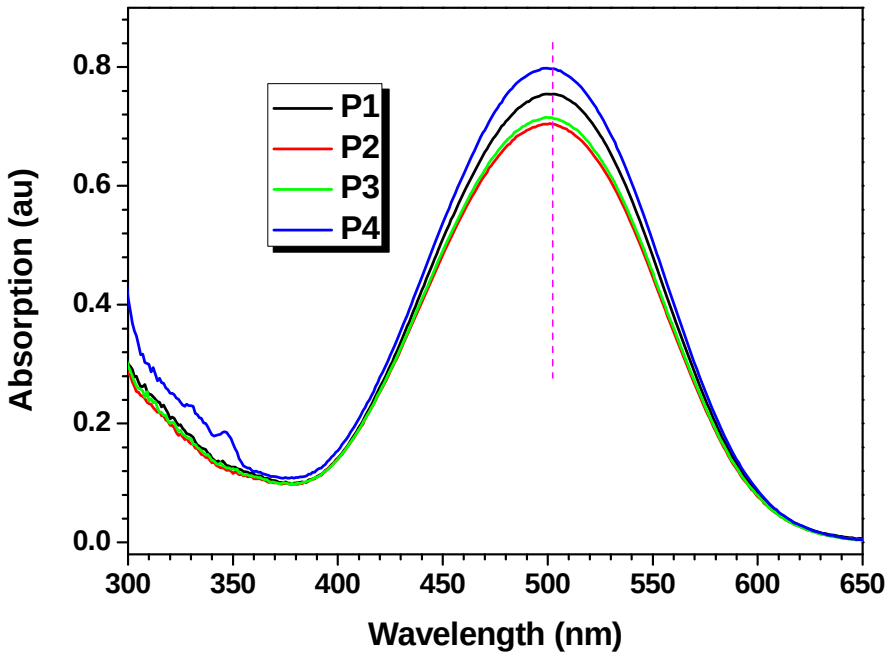


Fig. S22 UV-vis spectra of polymers **P1**-**P4** in DMF (0.02 mg/mL).

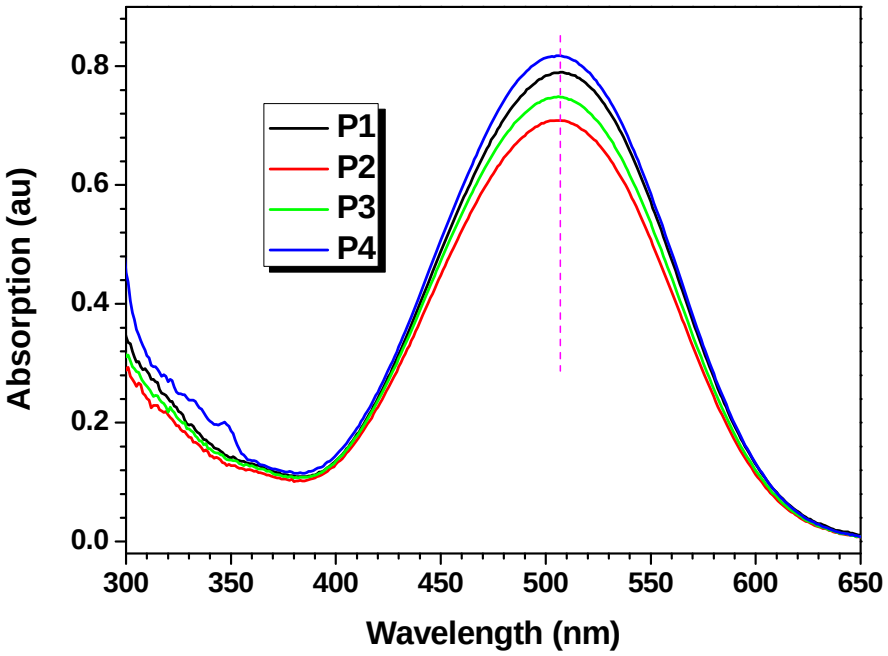


Fig. S23 UV-vis spectra of polymers **P1**-**P4** in DMSO (0.02 mg/mL).

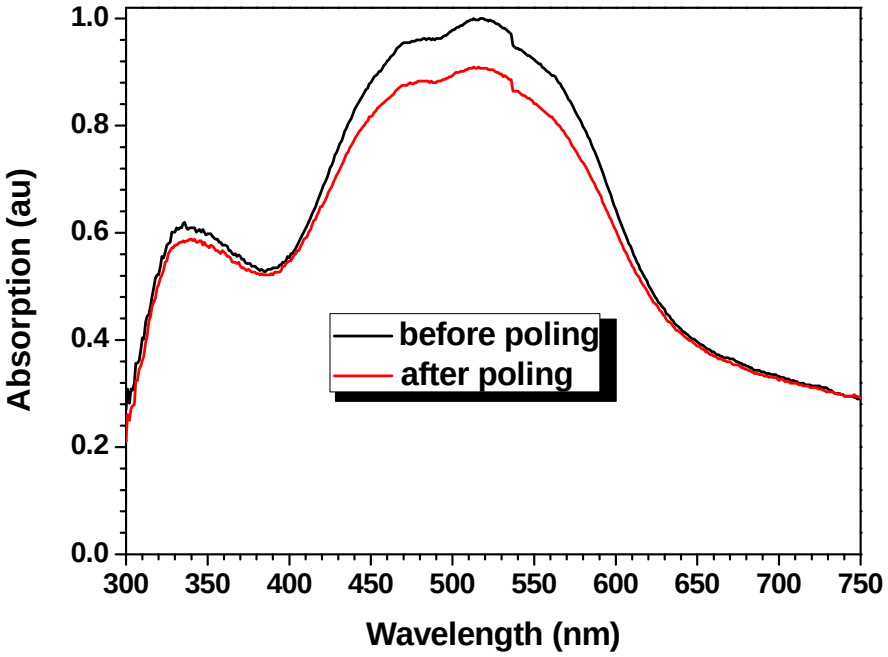


Fig. S24 Absorption spectra of the film of **P1** before and after poling.

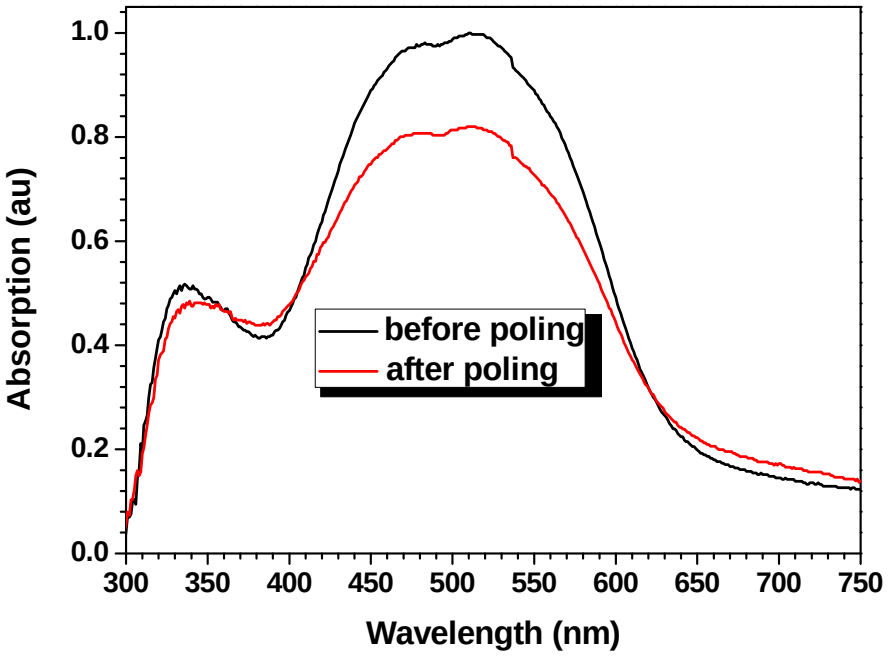


Fig. S25 Absorption spectra of the film of **P2** before and after poling.

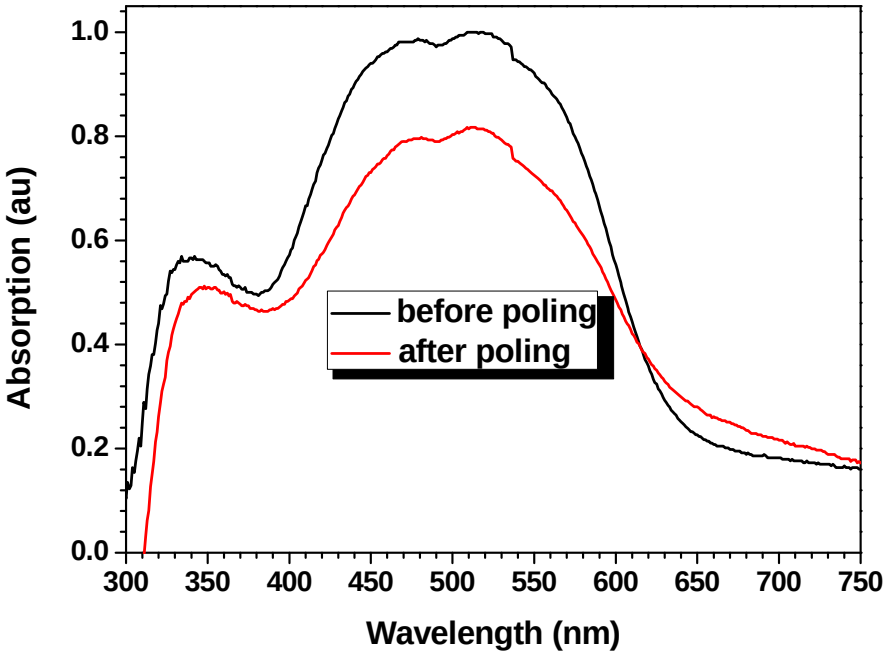


Fig. S26 Absorption spectra of the film of **P3** before and after poling.

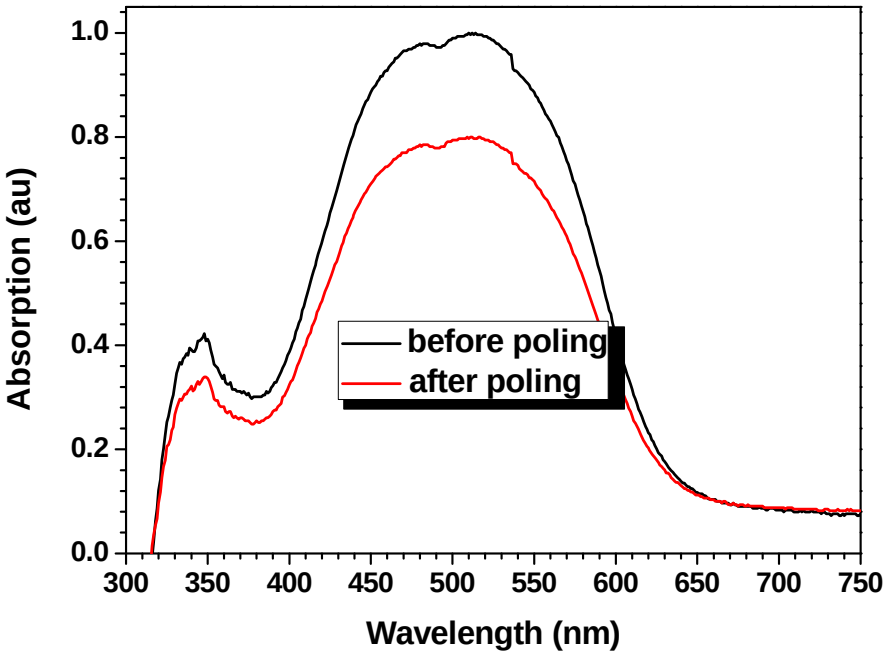


Fig. S27 Absorption spectra of the film of **P4** before and after poling.