

## Supporting Information

# Catalyst-Free Multicomponent Polymerization of Aldehyde, Amine and Trimethylsilyl Cyanide toward Poly( $\alpha$ -aminonitrile)s

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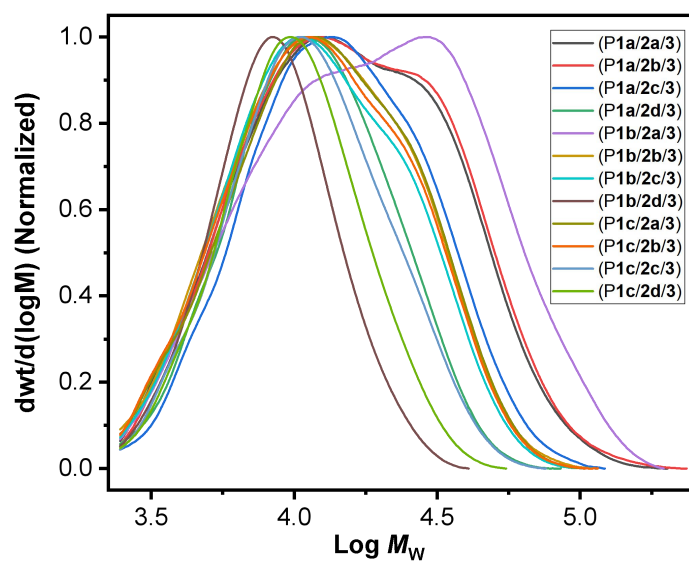
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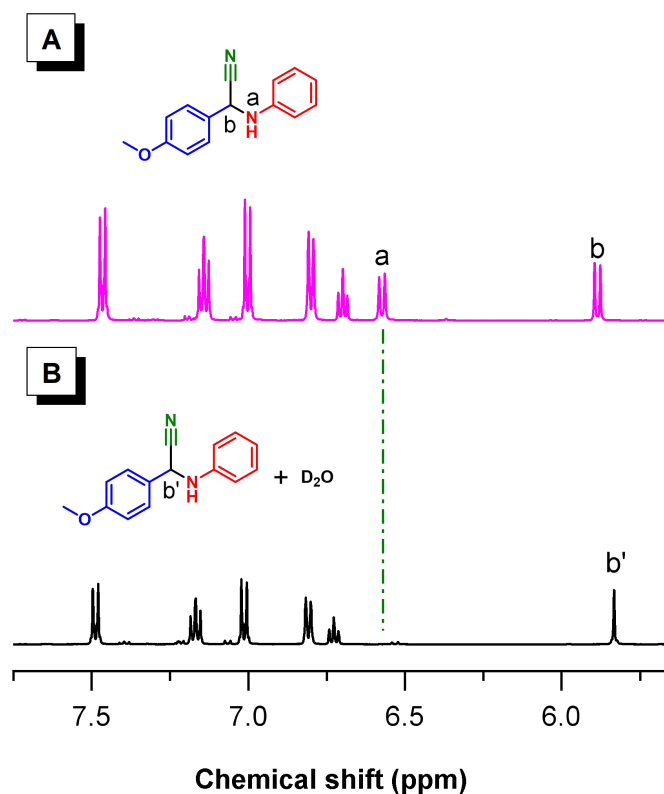
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**Figure S2.**  $^1\text{H}$  NMR spectra of (A) model compound and (B) model compound treated with  $\text{D}_2\text{O}$

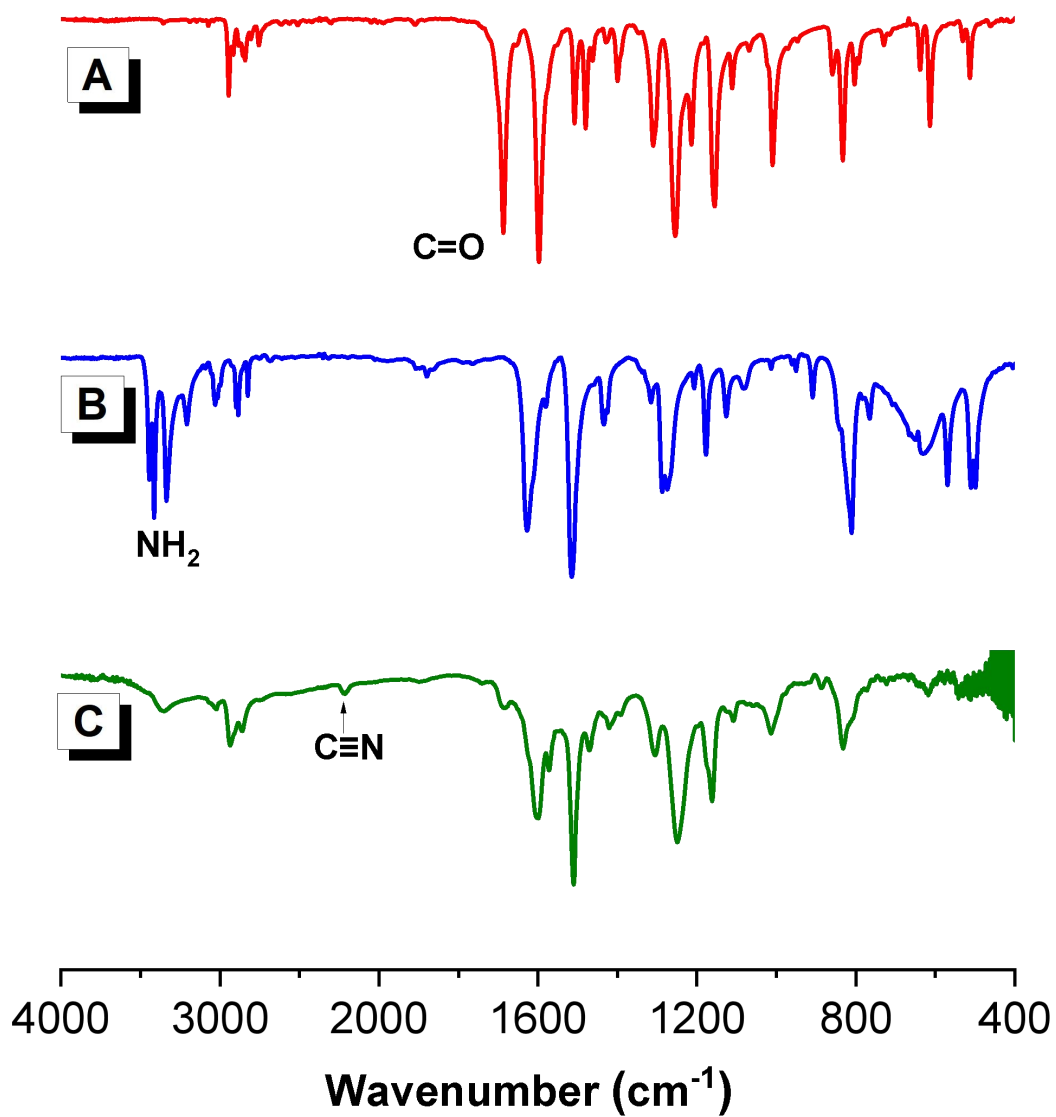


Figure S3. FT-IR spectra of (A) 1a, (B) 2b and (C) P1a/2b/3.

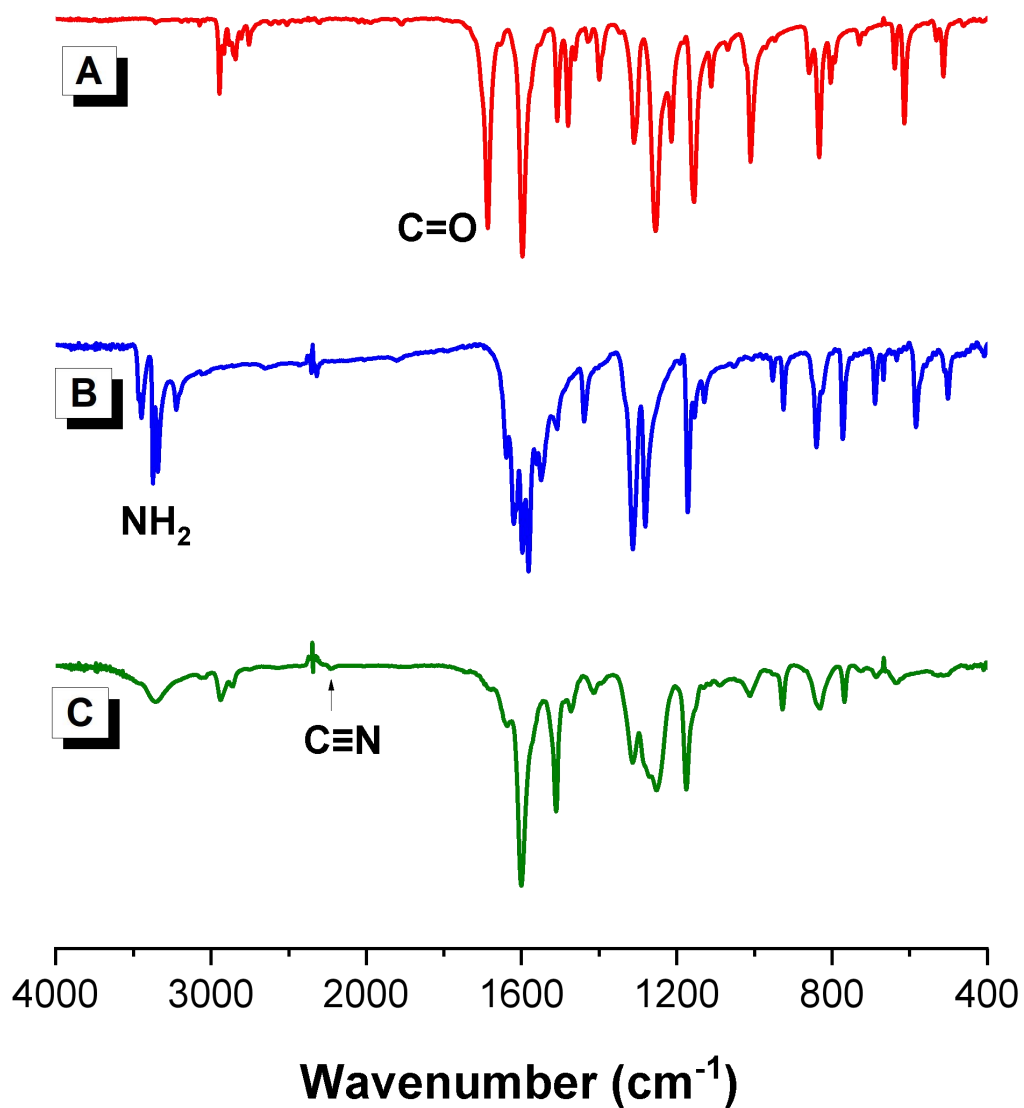


Figure S4. FT-IR spectra of (A) 1a, (B) 2c and (C) P1a/2c/3.

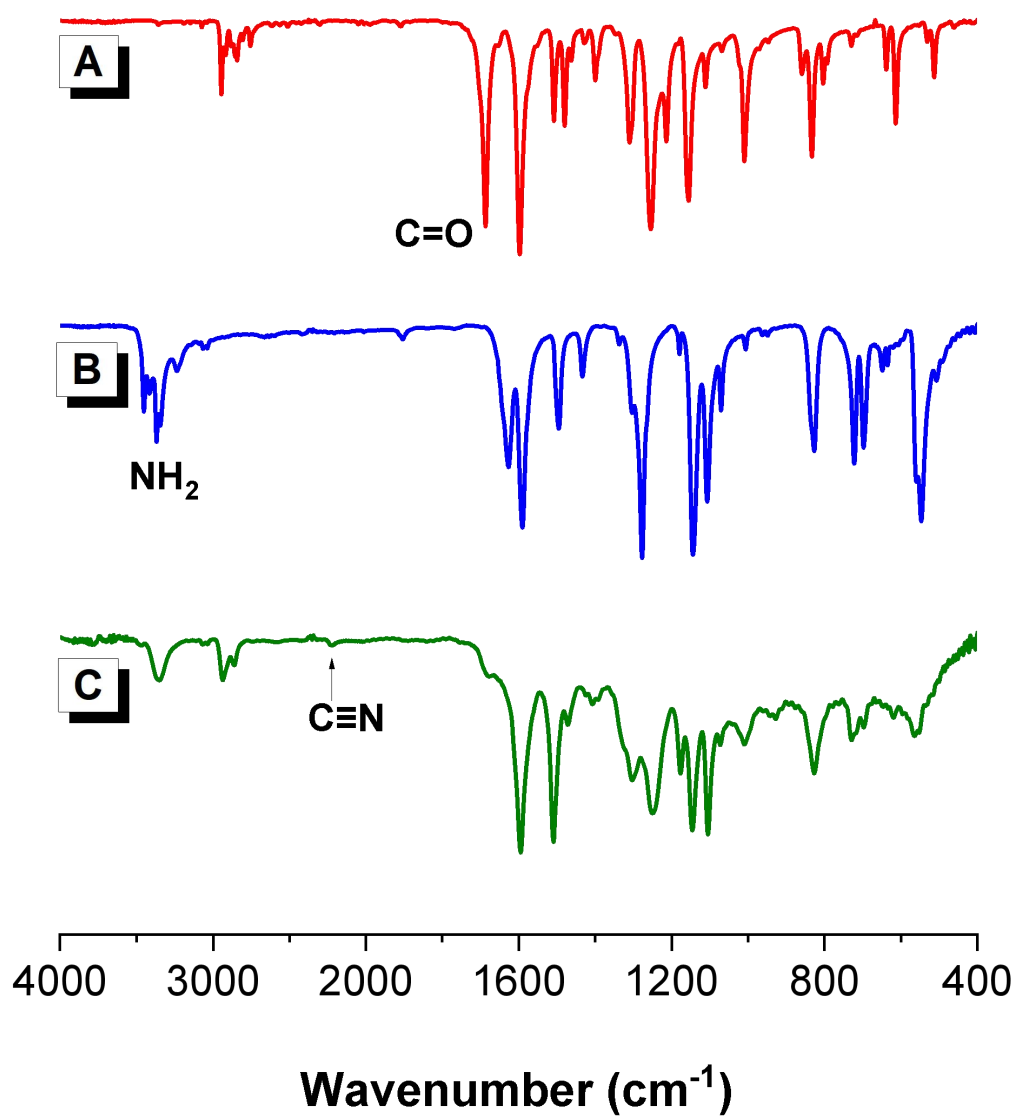


Figure S5. FT-IR spectra of (A) 1a, (B) 2d and (C) P1a/2d/3.

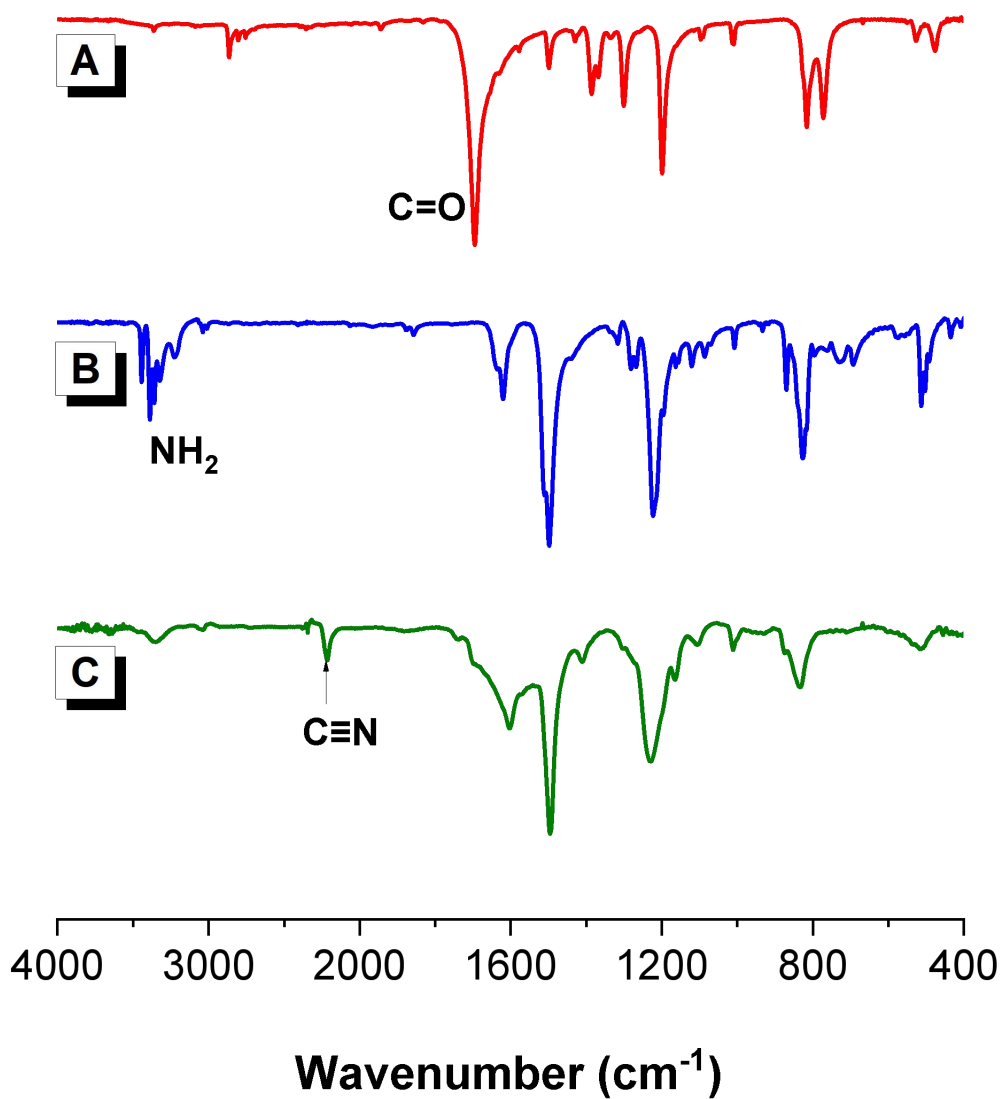


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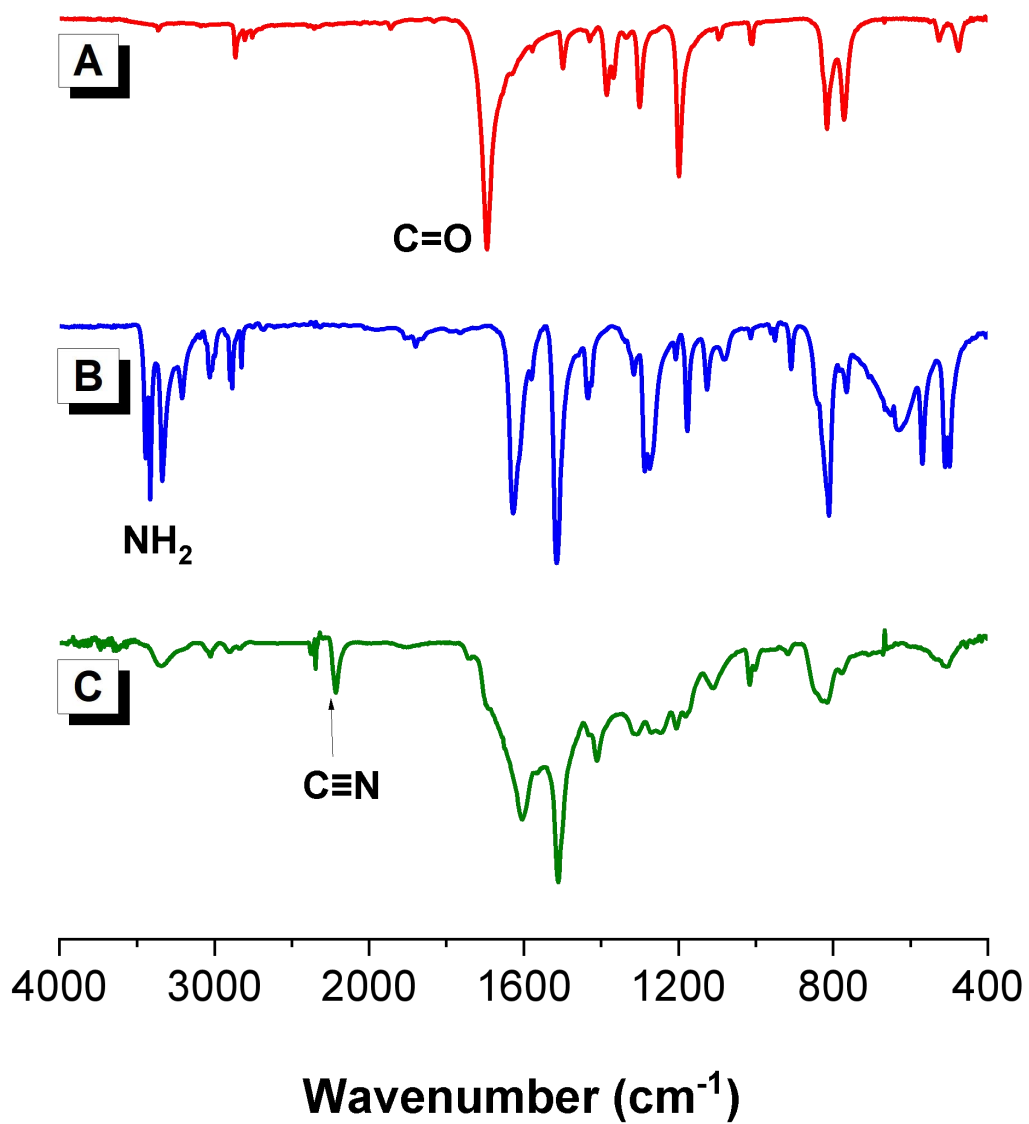


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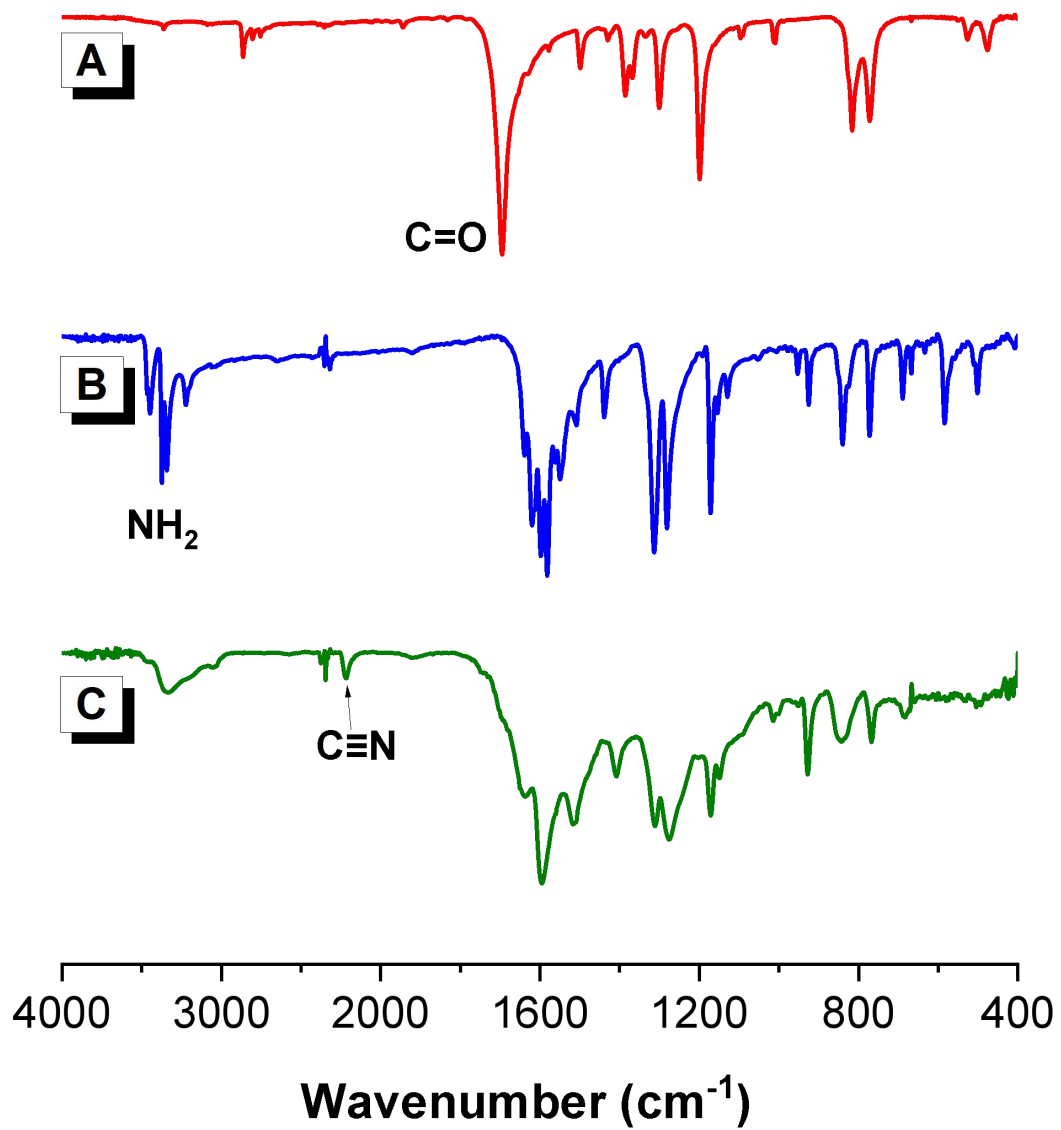


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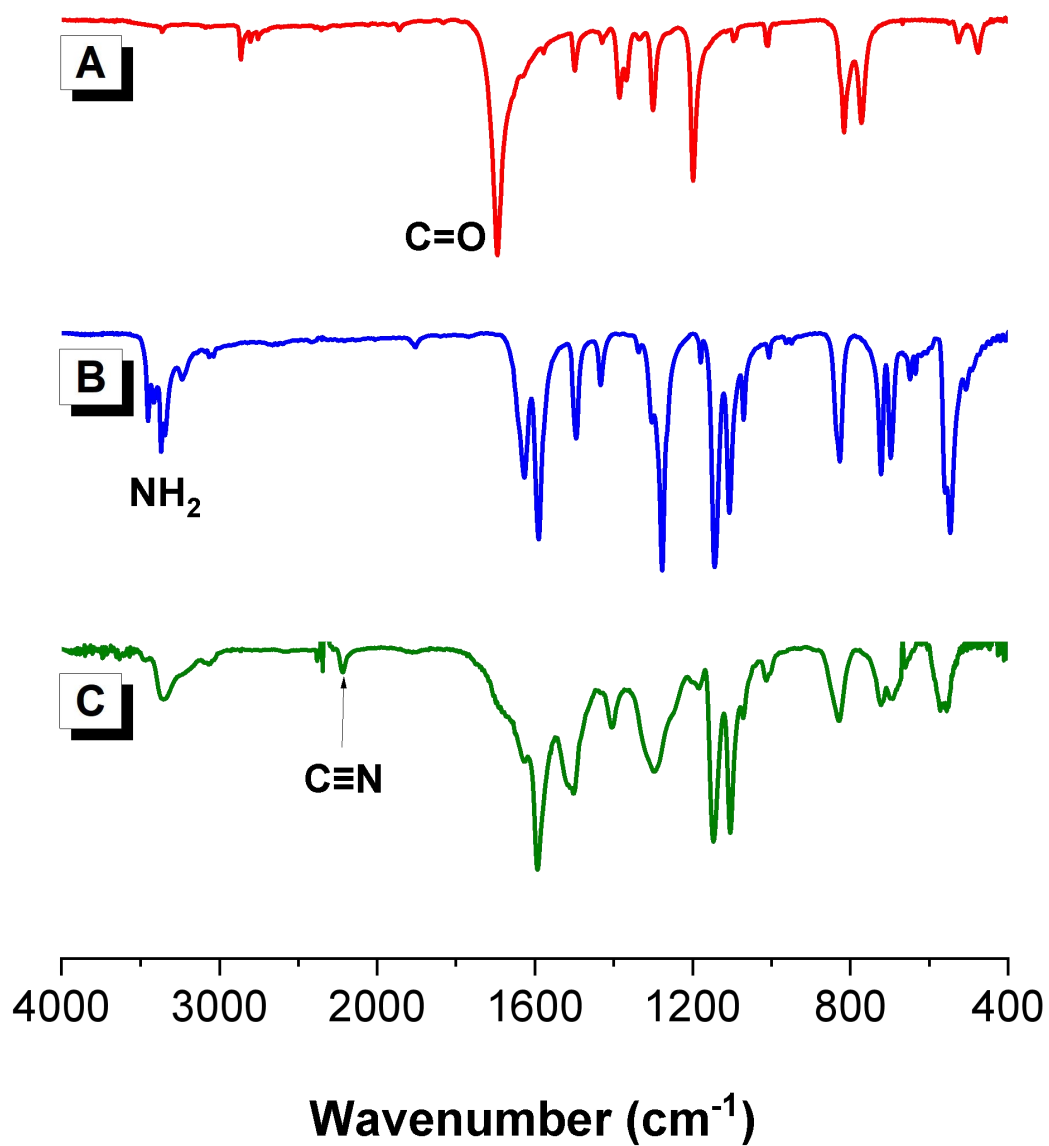


Figure S9. FT-IR spectra of (A) **1b**, (B) **2d** and (C) **P1b/2d/3**.

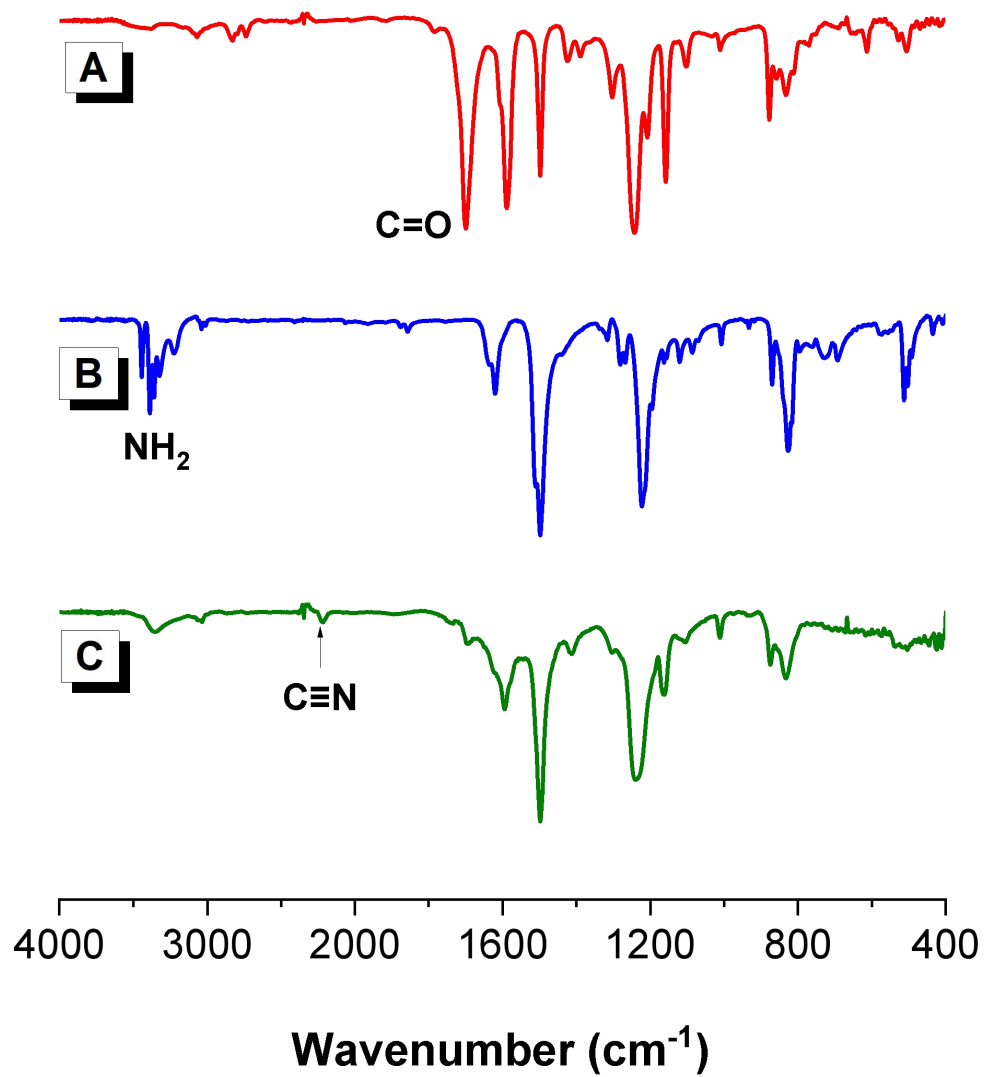


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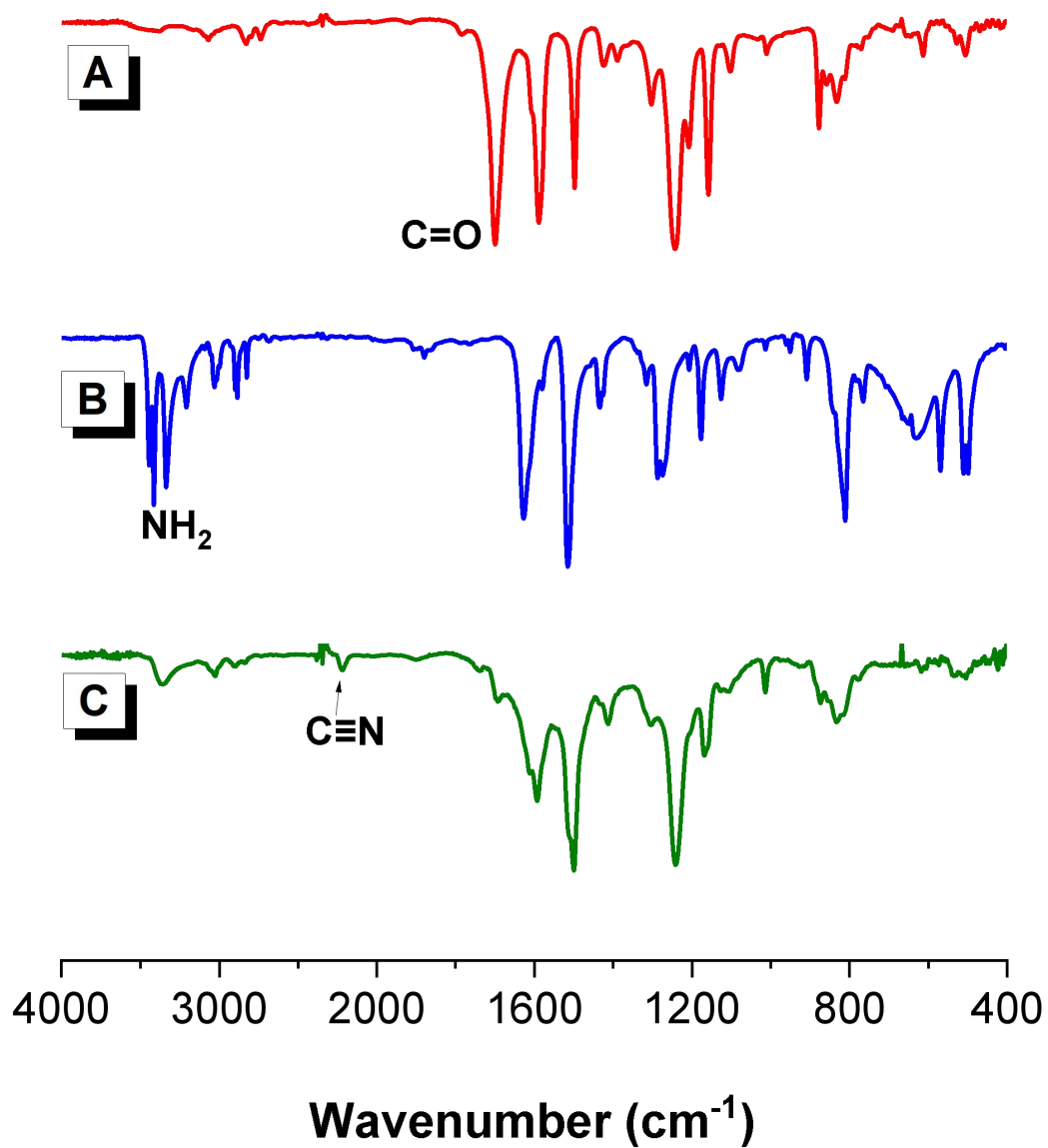


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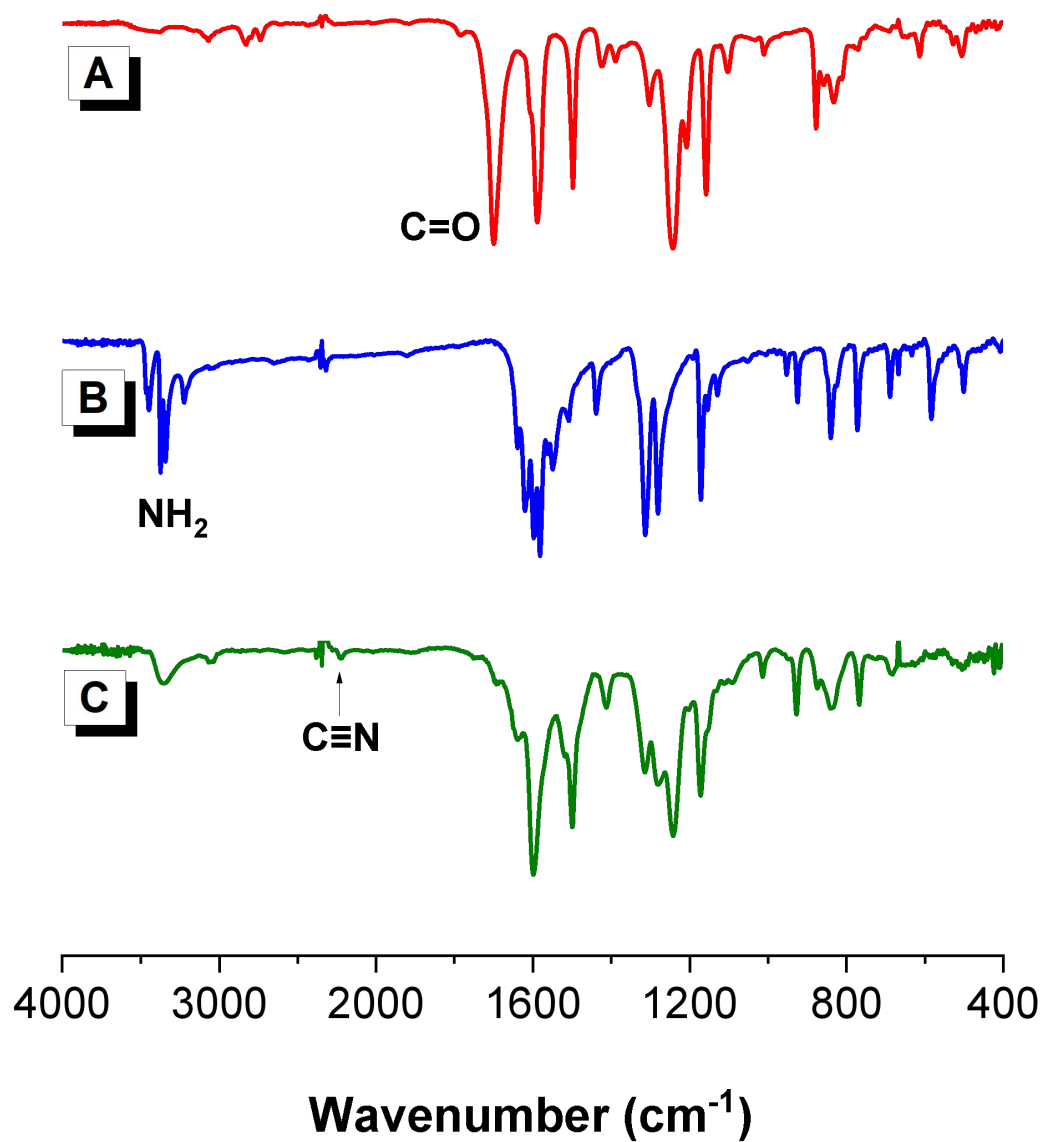


Figure S12. FT-IR spectra of (A) 1c, (B) 2c and (C) P1c/2c/3.

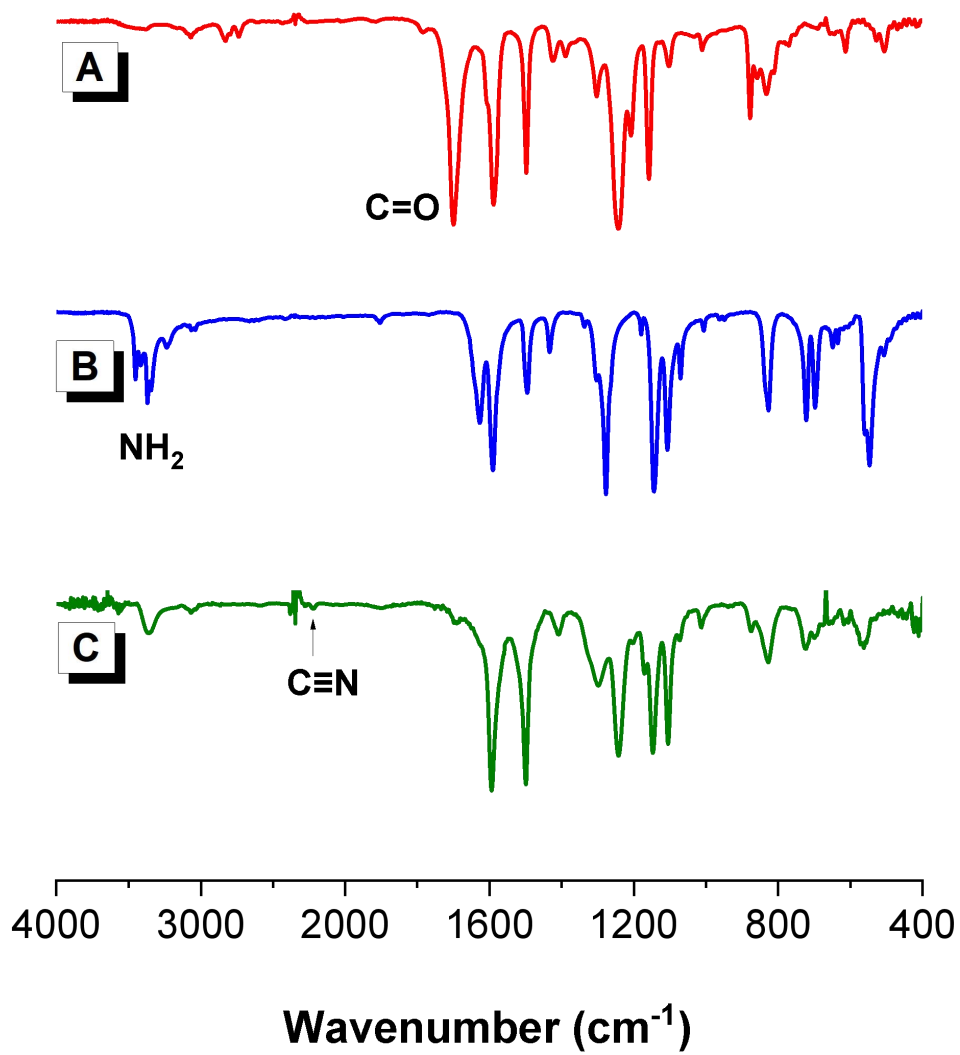
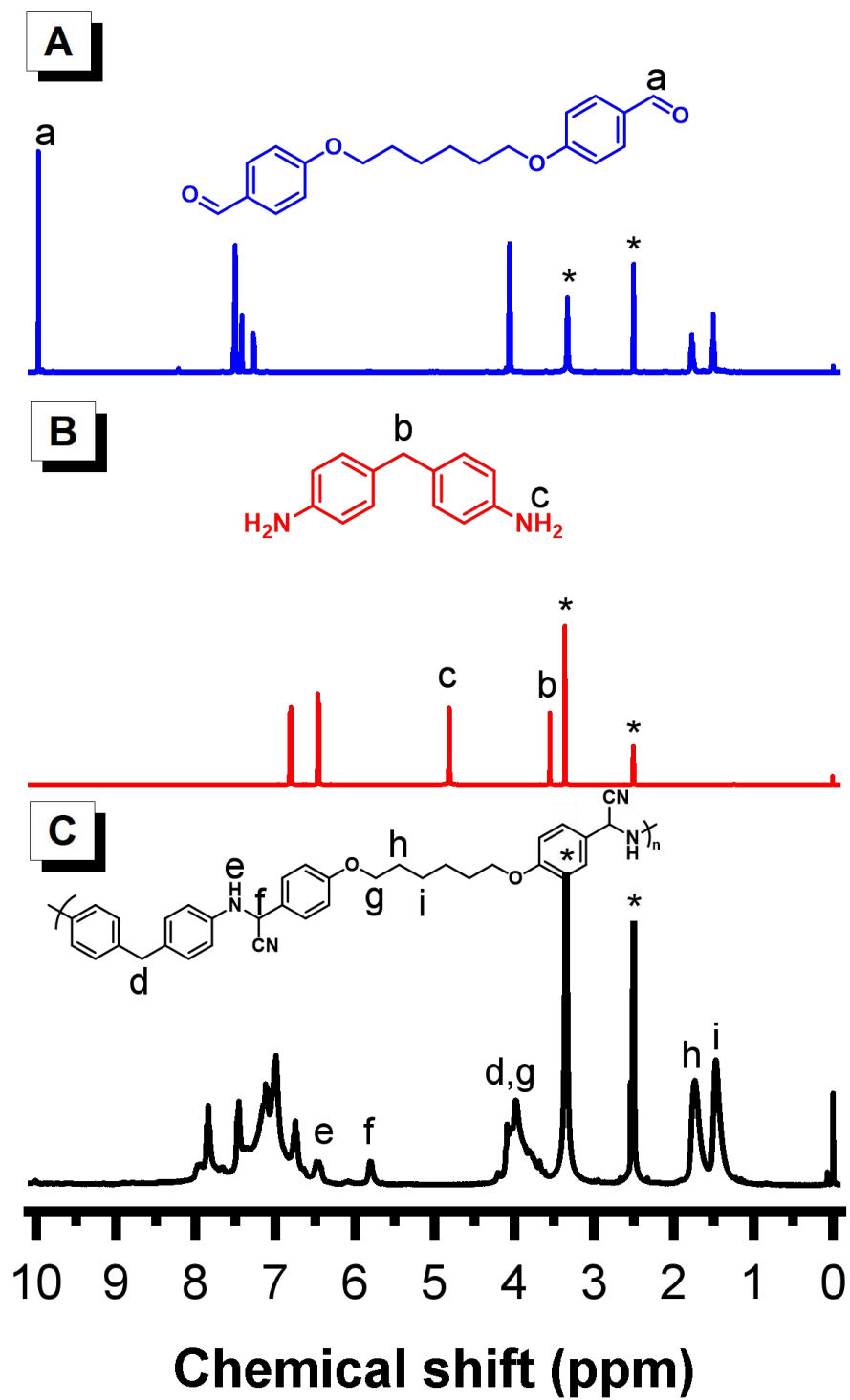
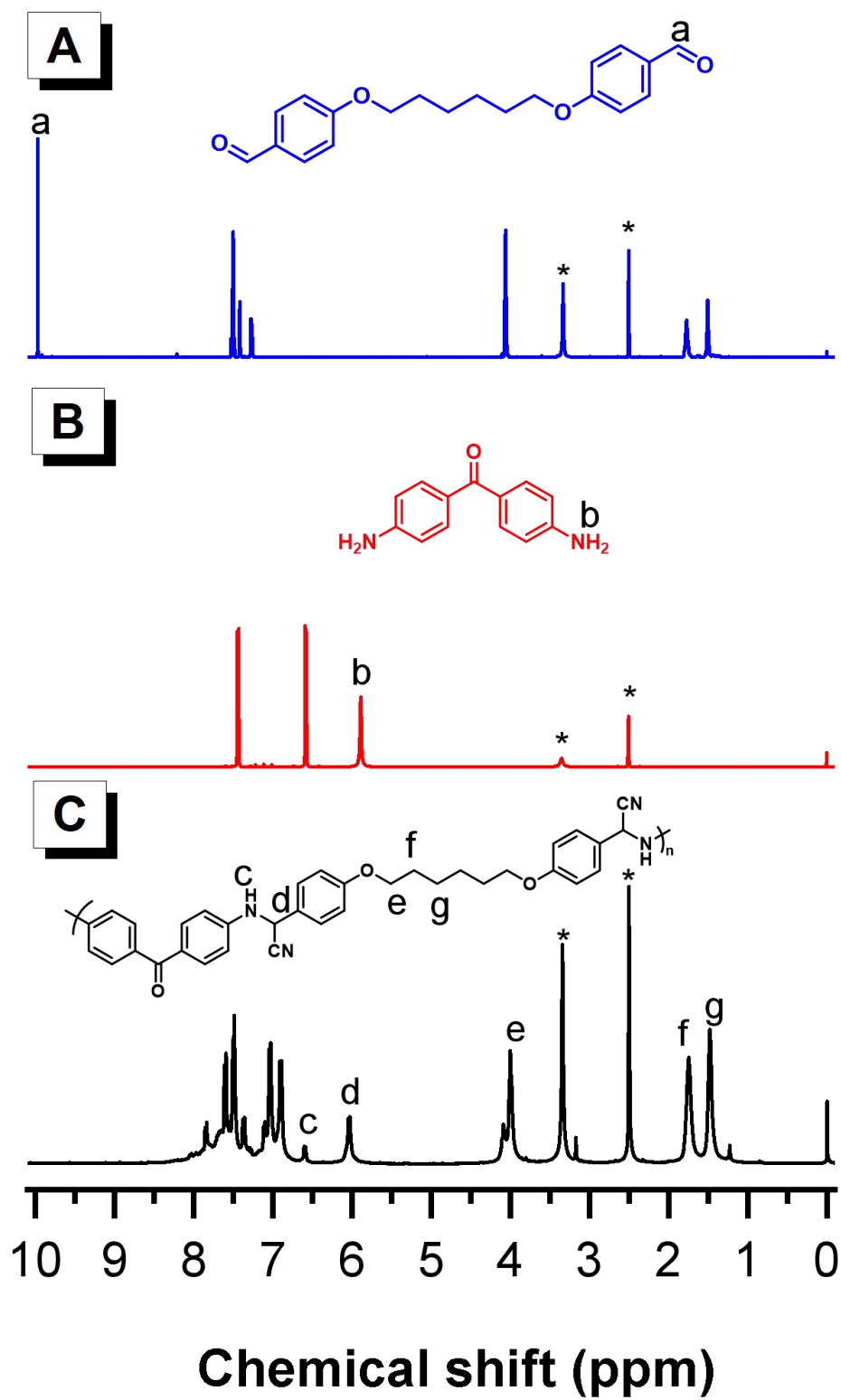


Figure S13. FT-IR spectra of (A) 1c, (B) 2d and (C) P1c/2d/3.

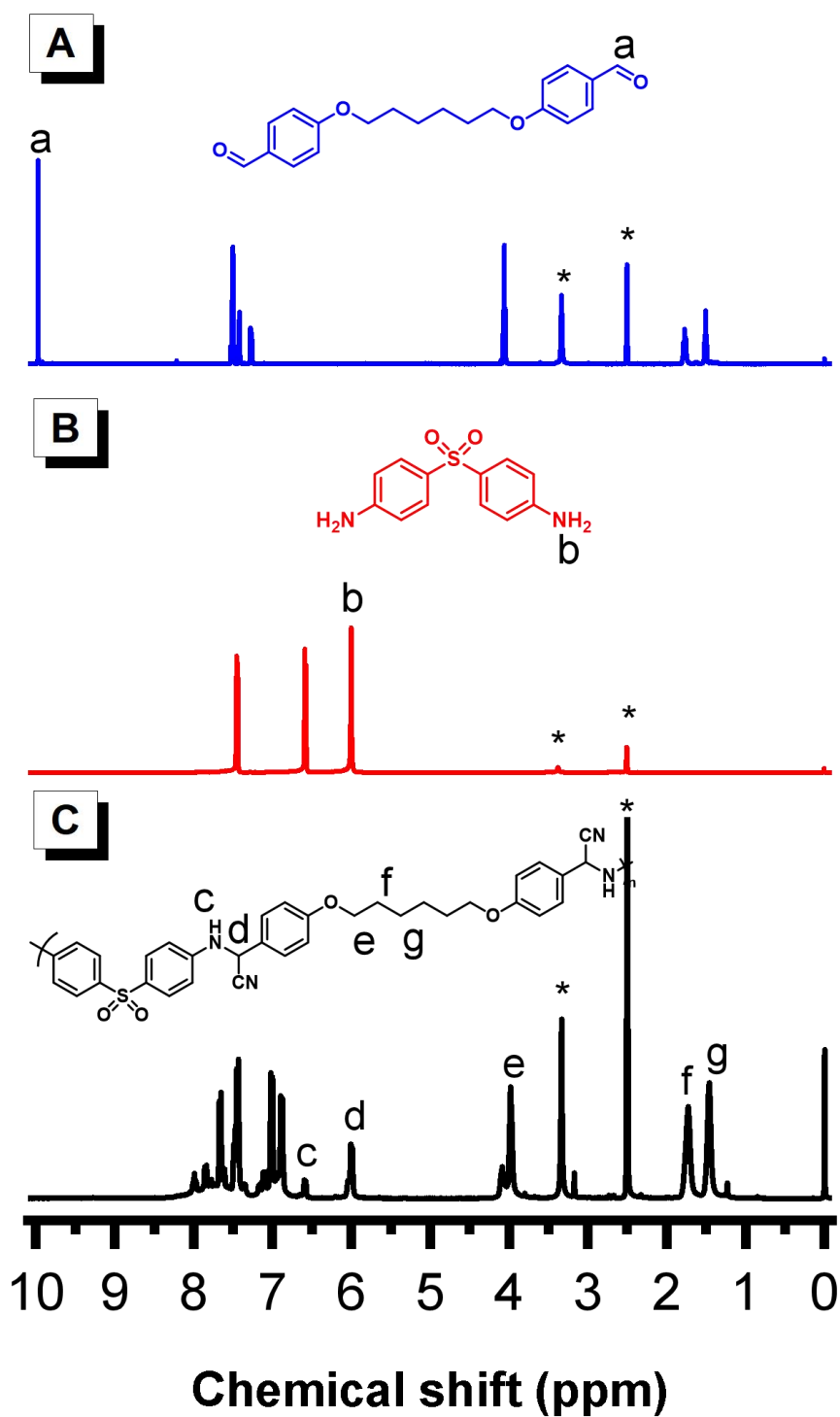


**Figure S14.**  $^1\text{H}$  NMR spectra of (A) **1a**, (B) **2b**, and (C) **P1a/2b/3** in  $\text{DMSO-}d_6$ . The solvent peaks are marked with asterisks.

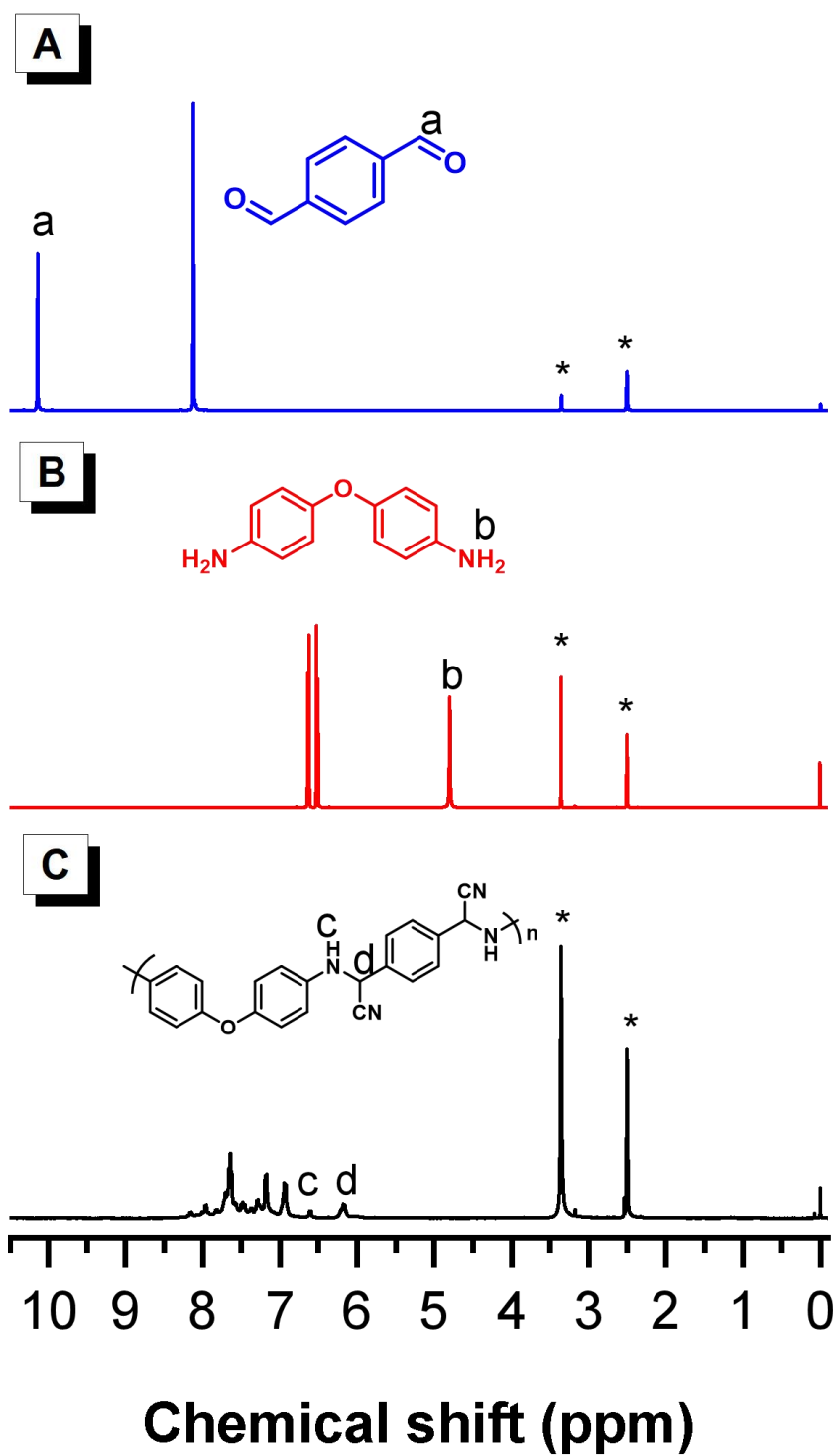




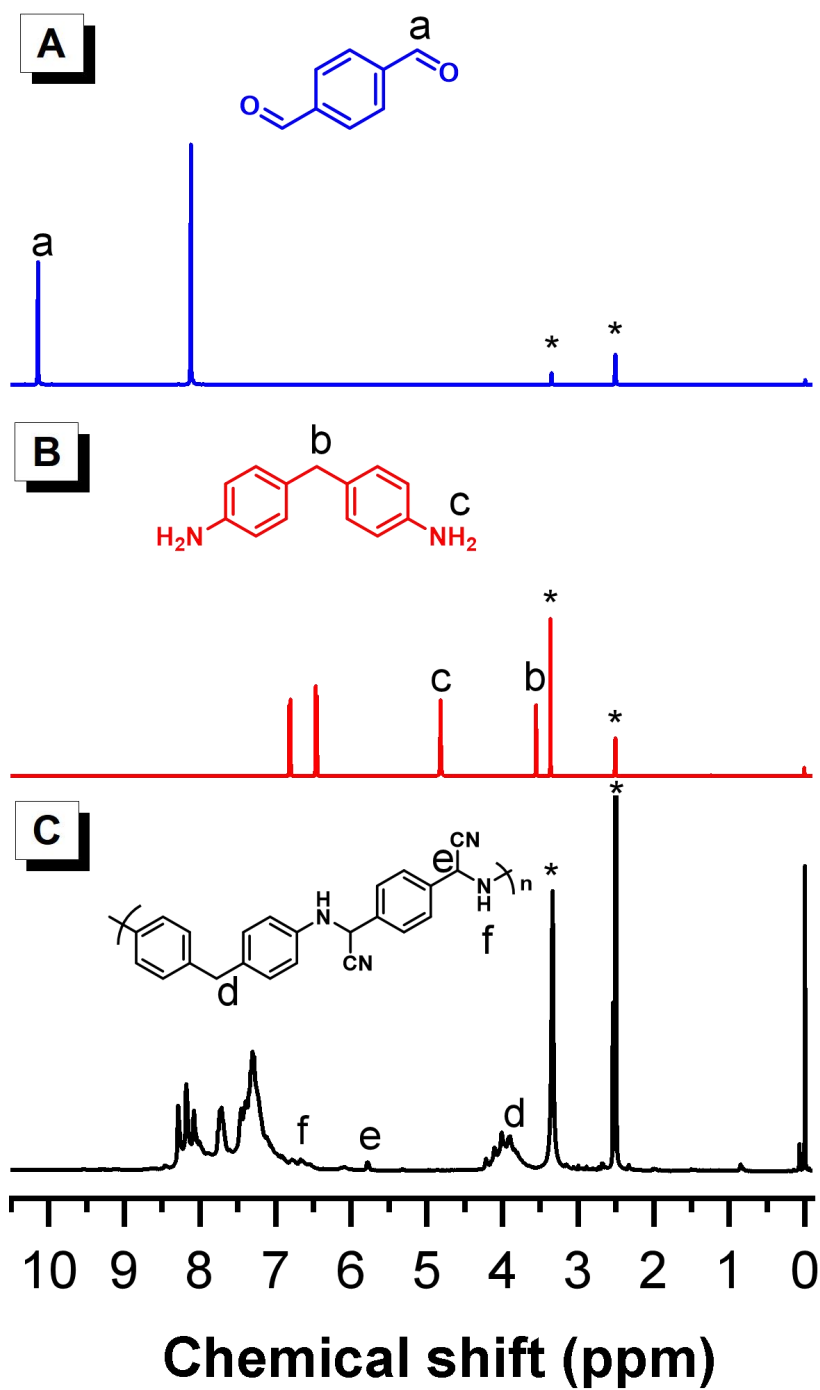
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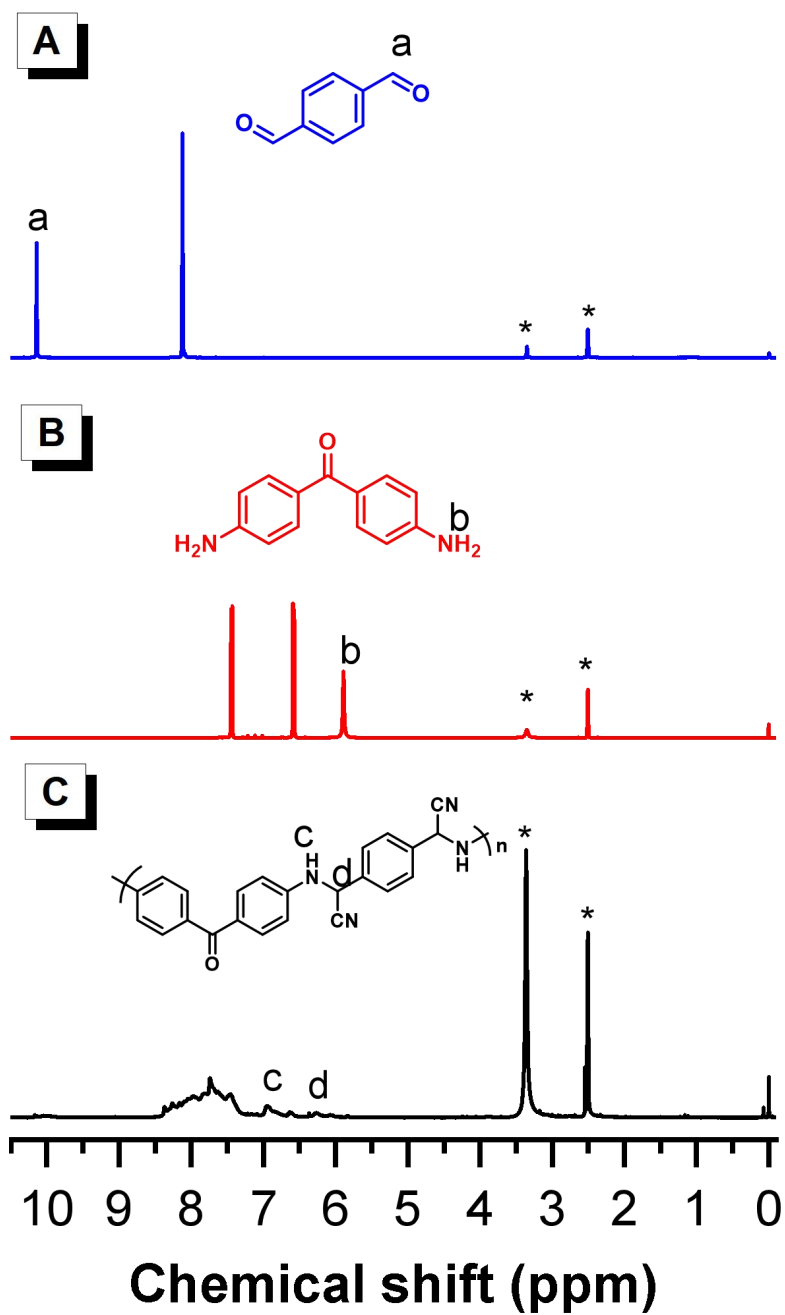
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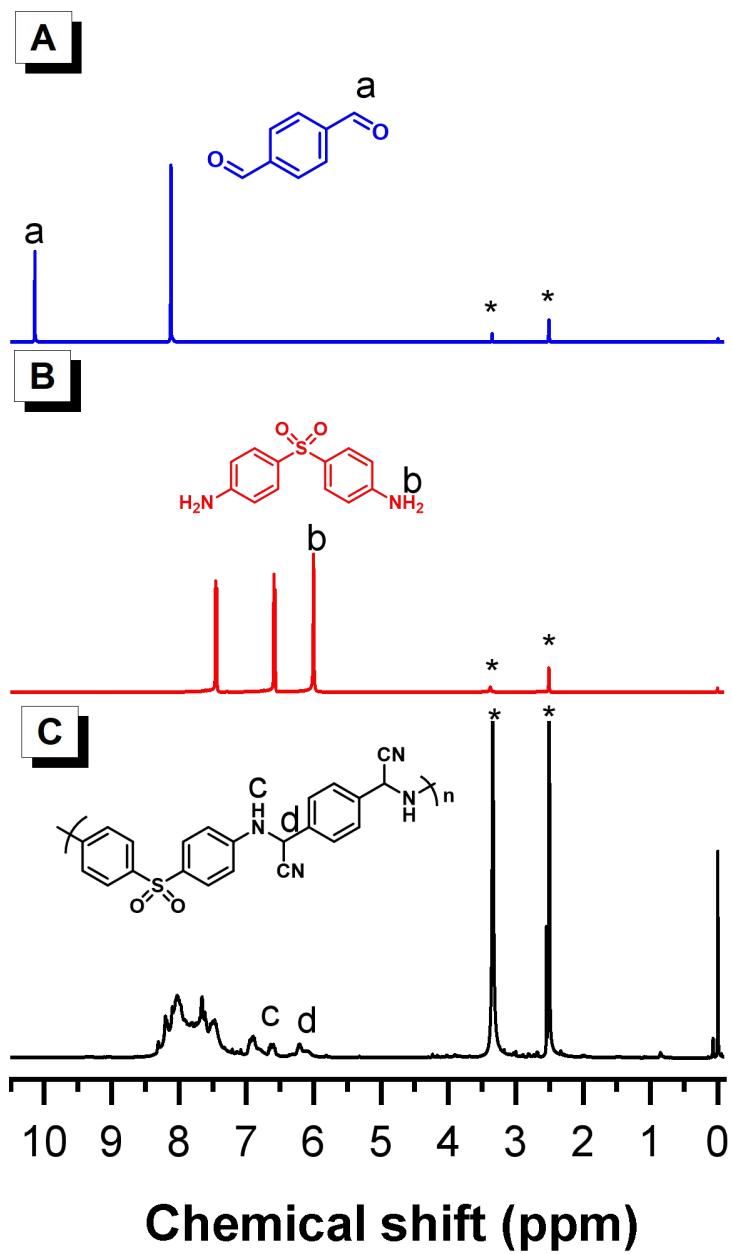
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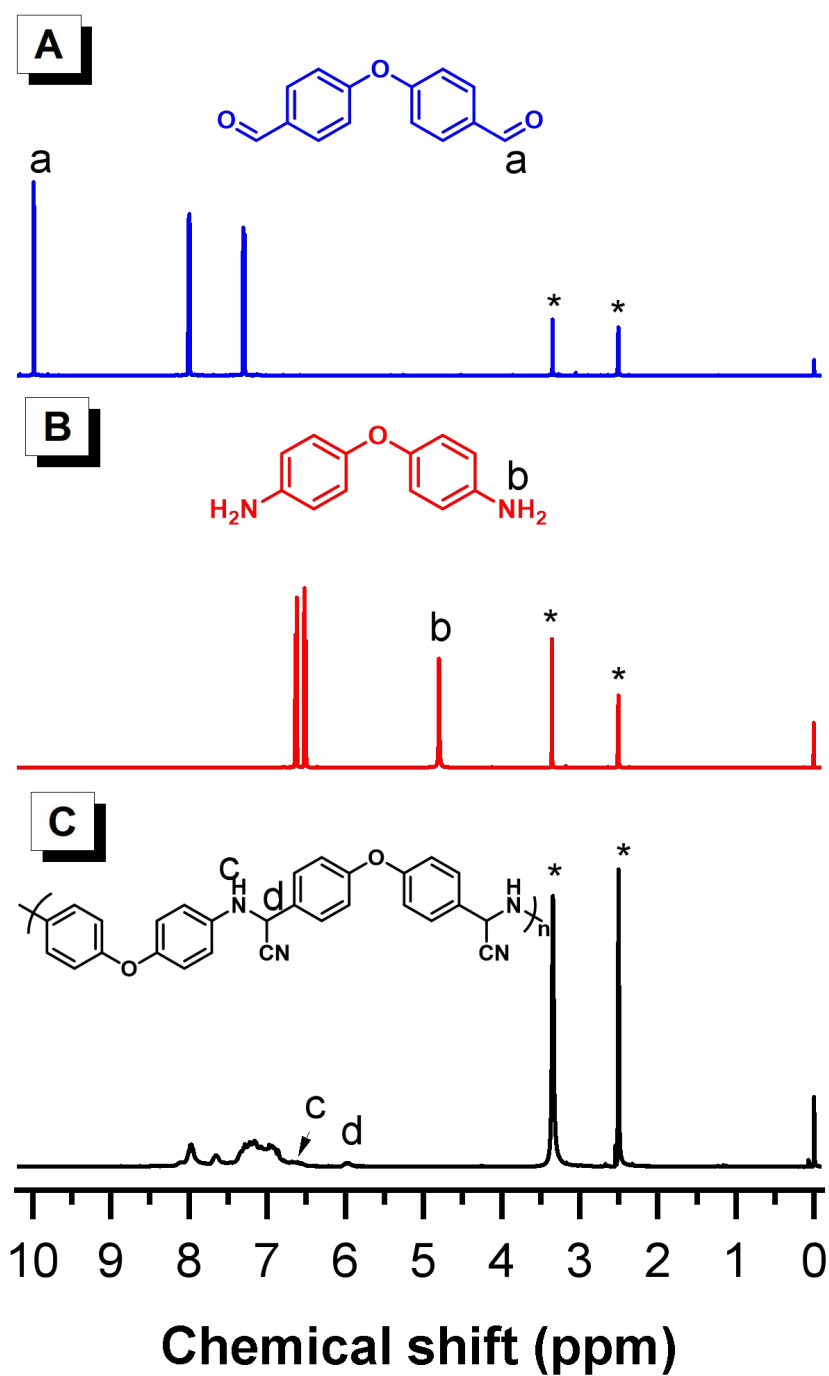
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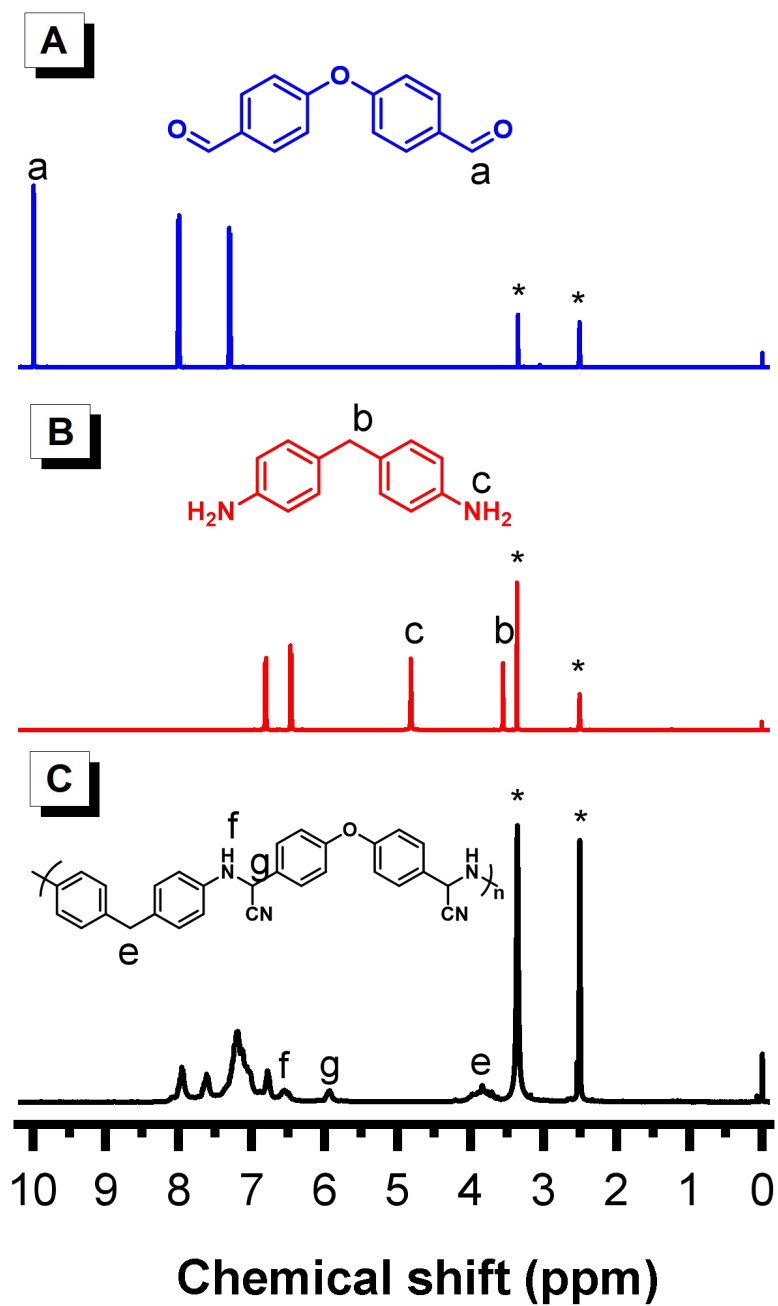
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**Figure S20.**  $^1\text{H}$  NMR spectra of (A) **1b**, (B) **2d**, and (C) **P1b/2d/3** in  $\text{DMSO-}d_6$ . The solvent peaks are marked with asterisks.

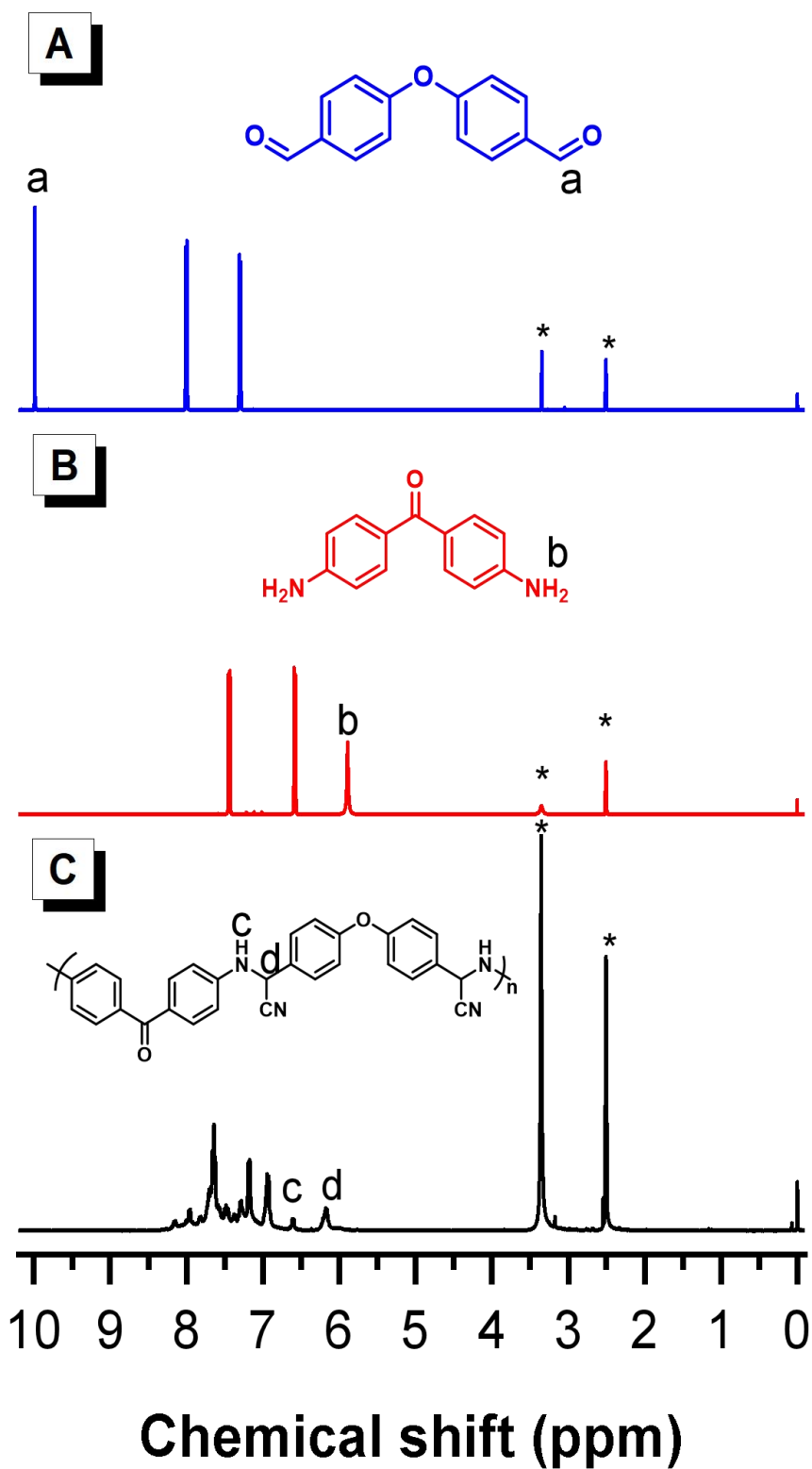


**Figure S21.**  $^1\text{H}$  NMR spectra of (A) **1c**, (B) **2a**, and (C) **P1c/2a/3** in  $\text{DMSO-}d_6$ . The solvent peaks are marked with asterisks.

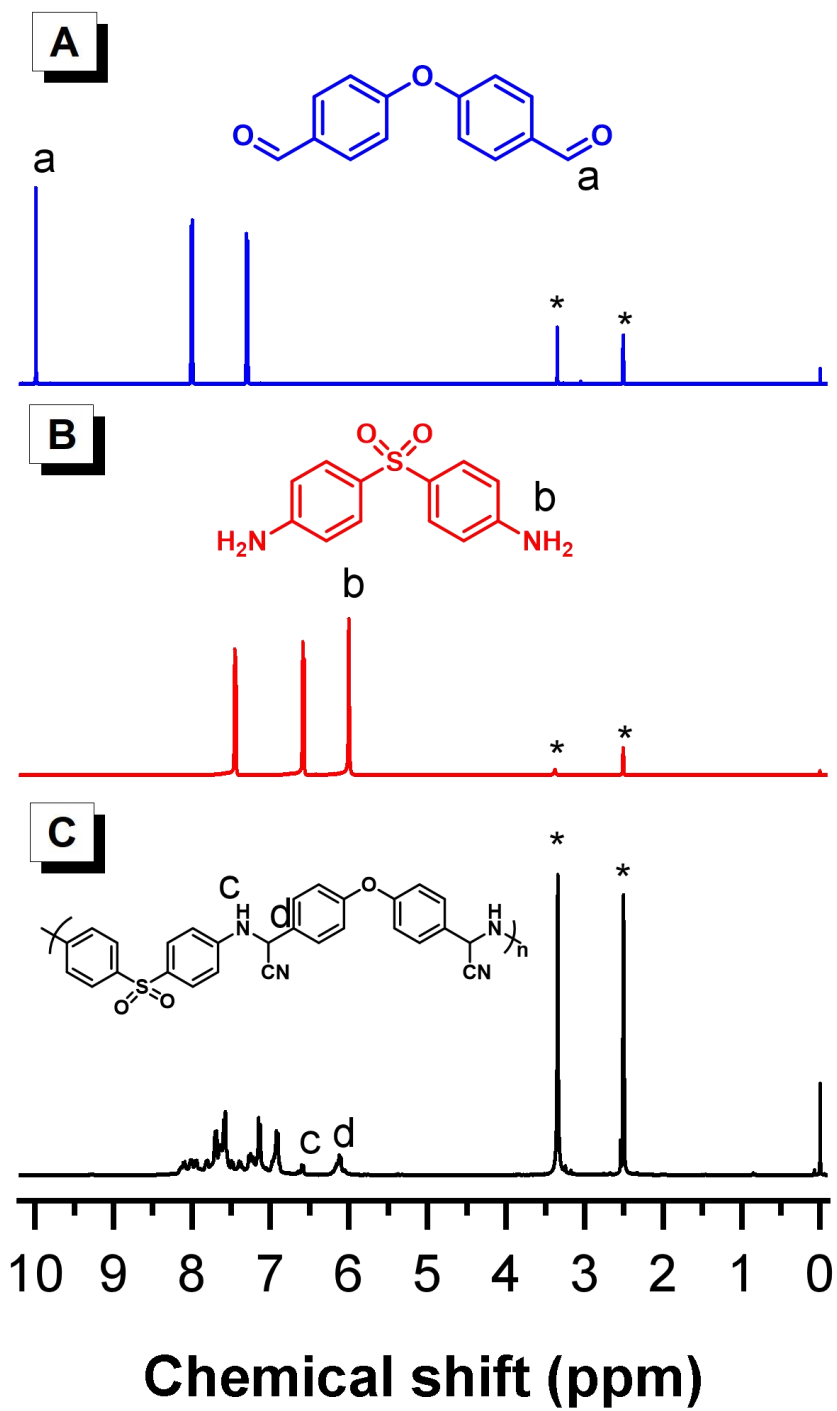


**Figure S22.**  $^1\text{H}$  NMR spectra of (A) **1c**, (B) **2b**, and (C) **P1c/2b/3** in  $\text{DMSO-}d_6$ . The solvent peaks are marked with asterisks.

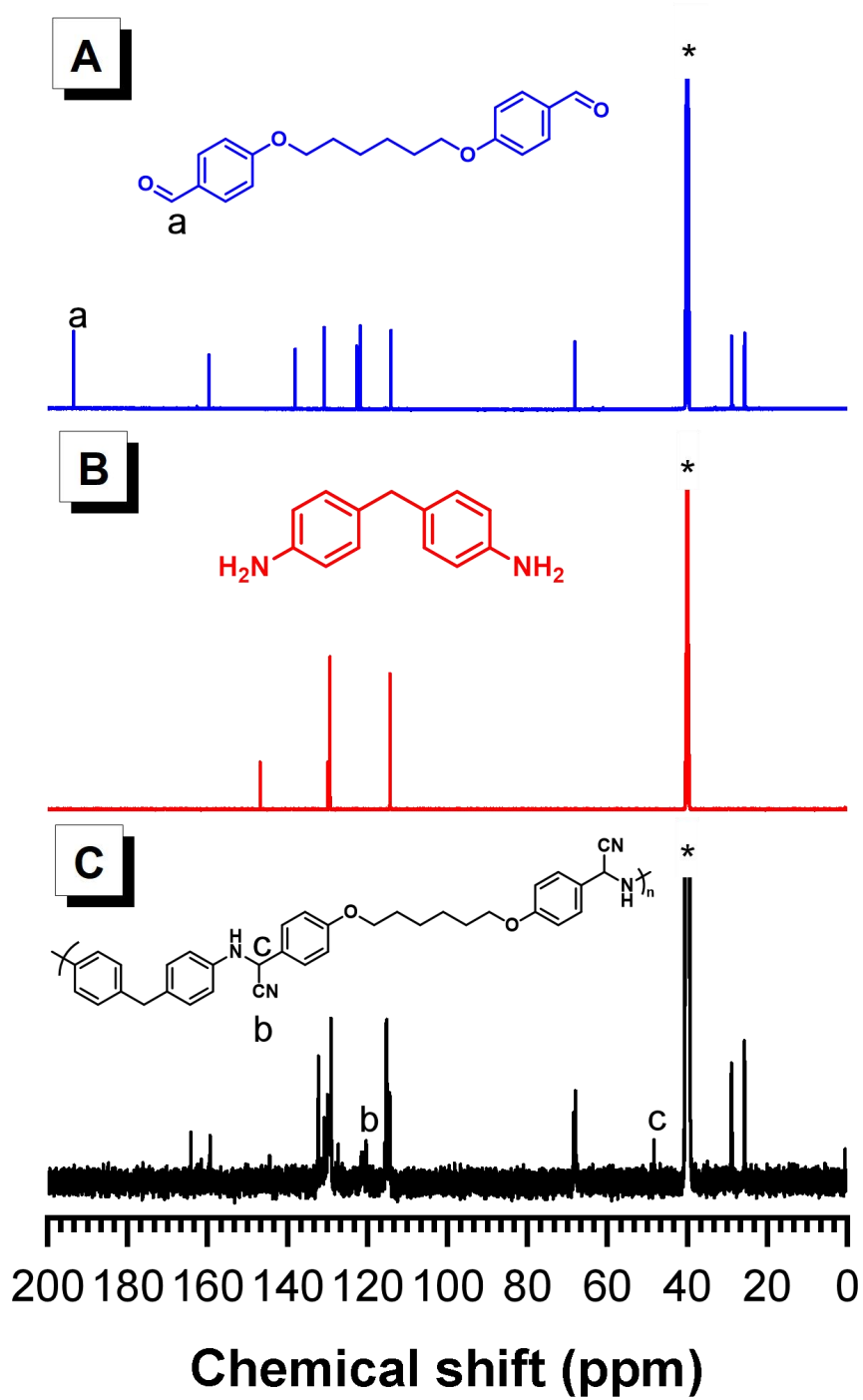




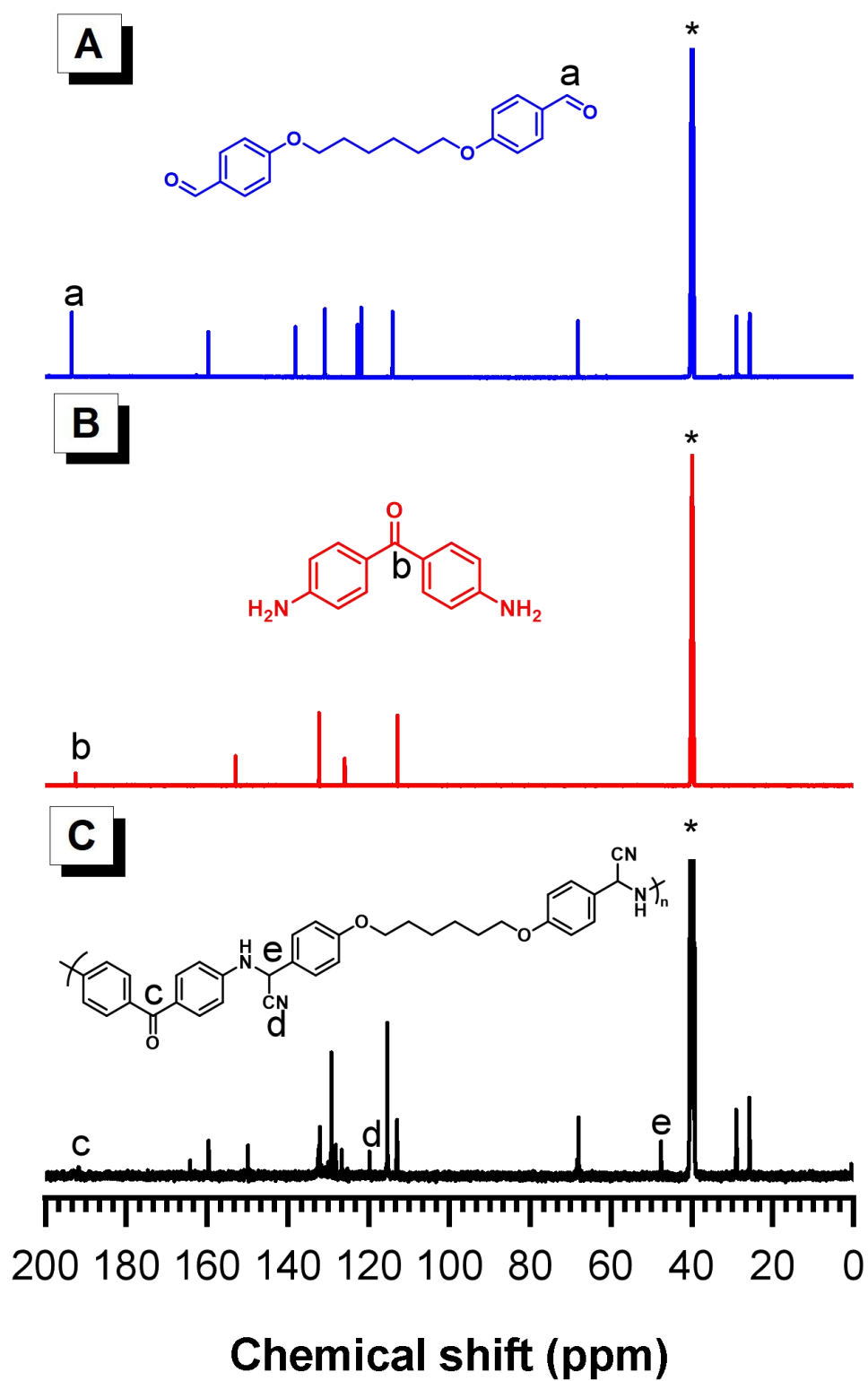
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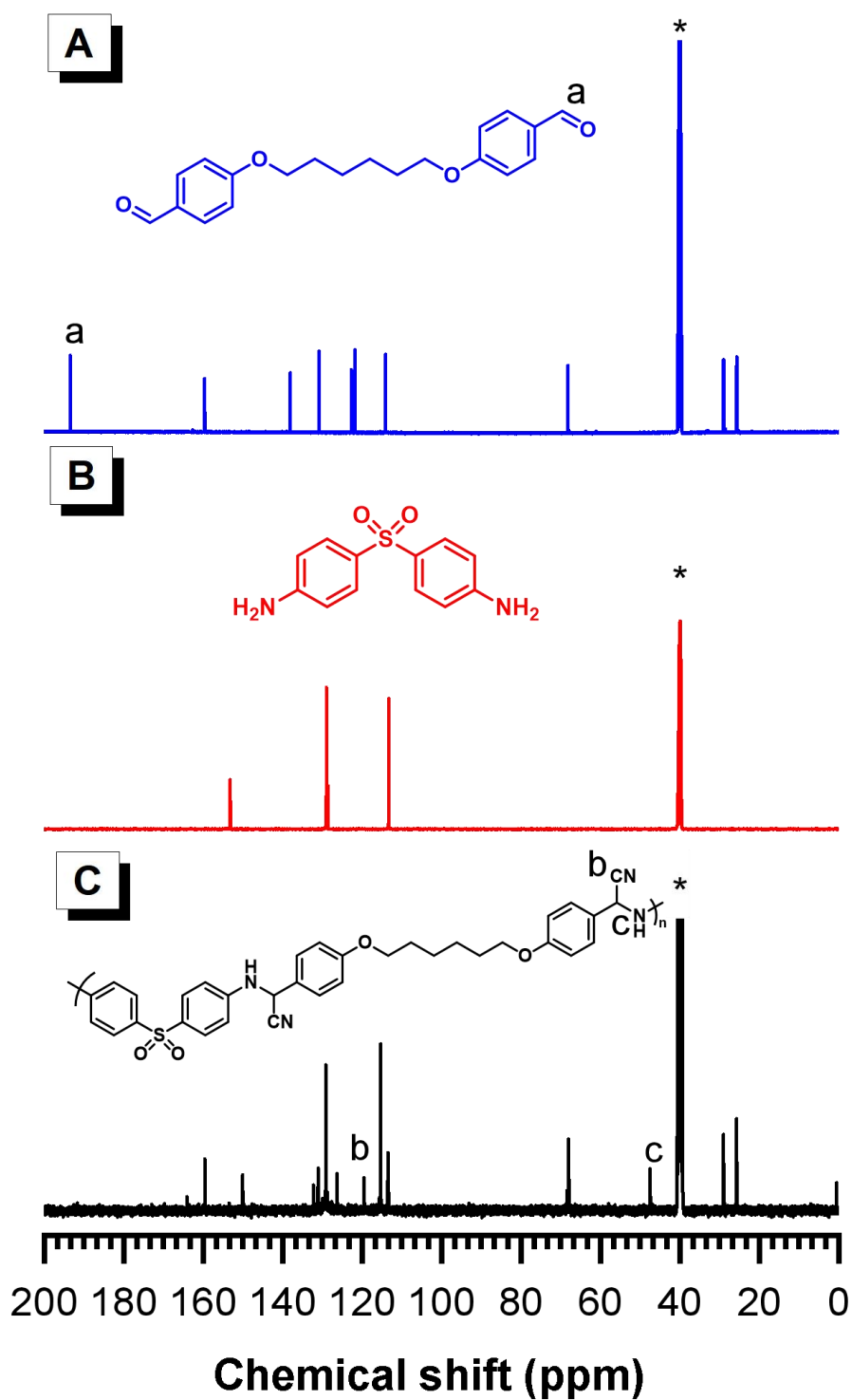
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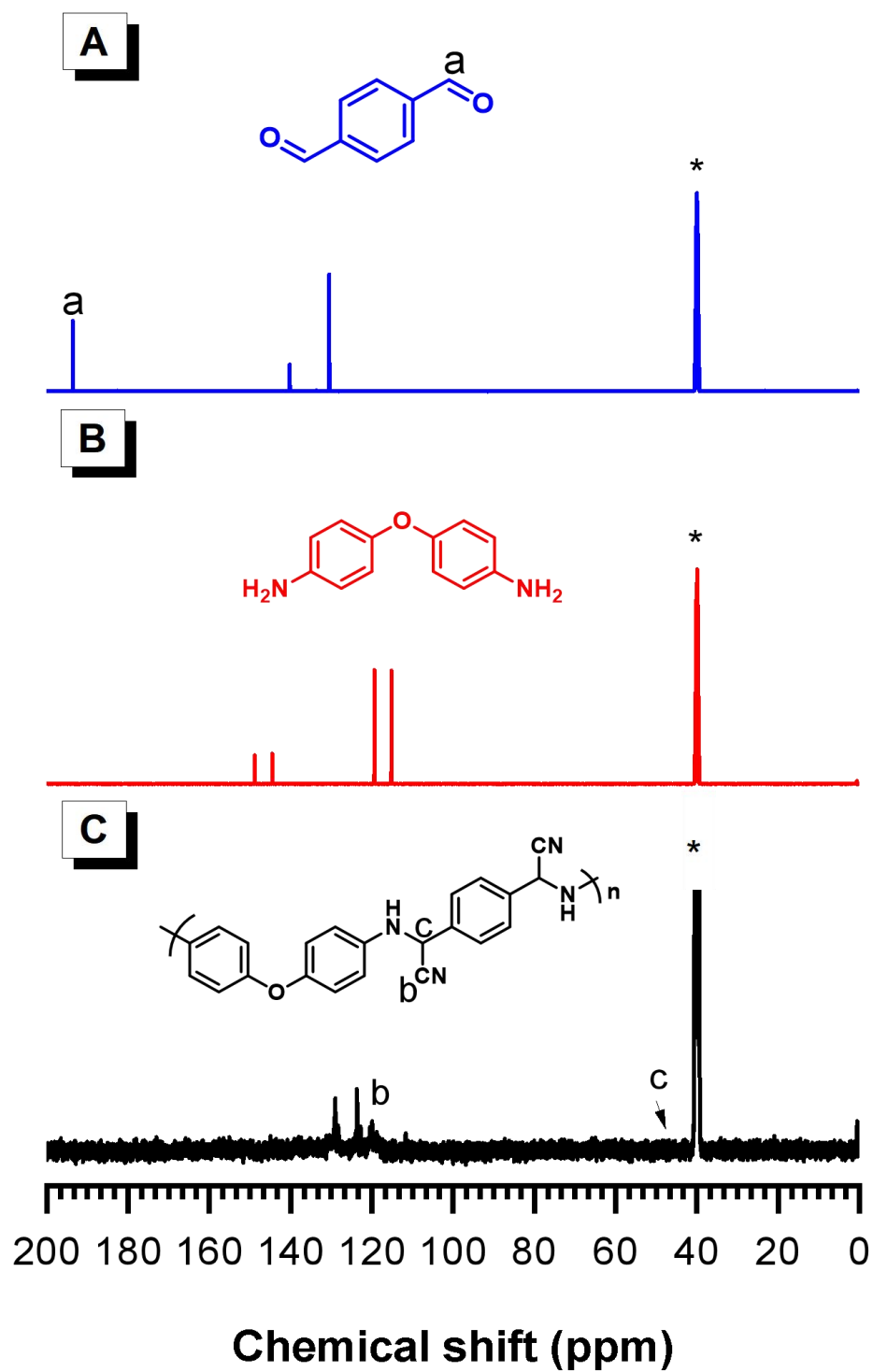
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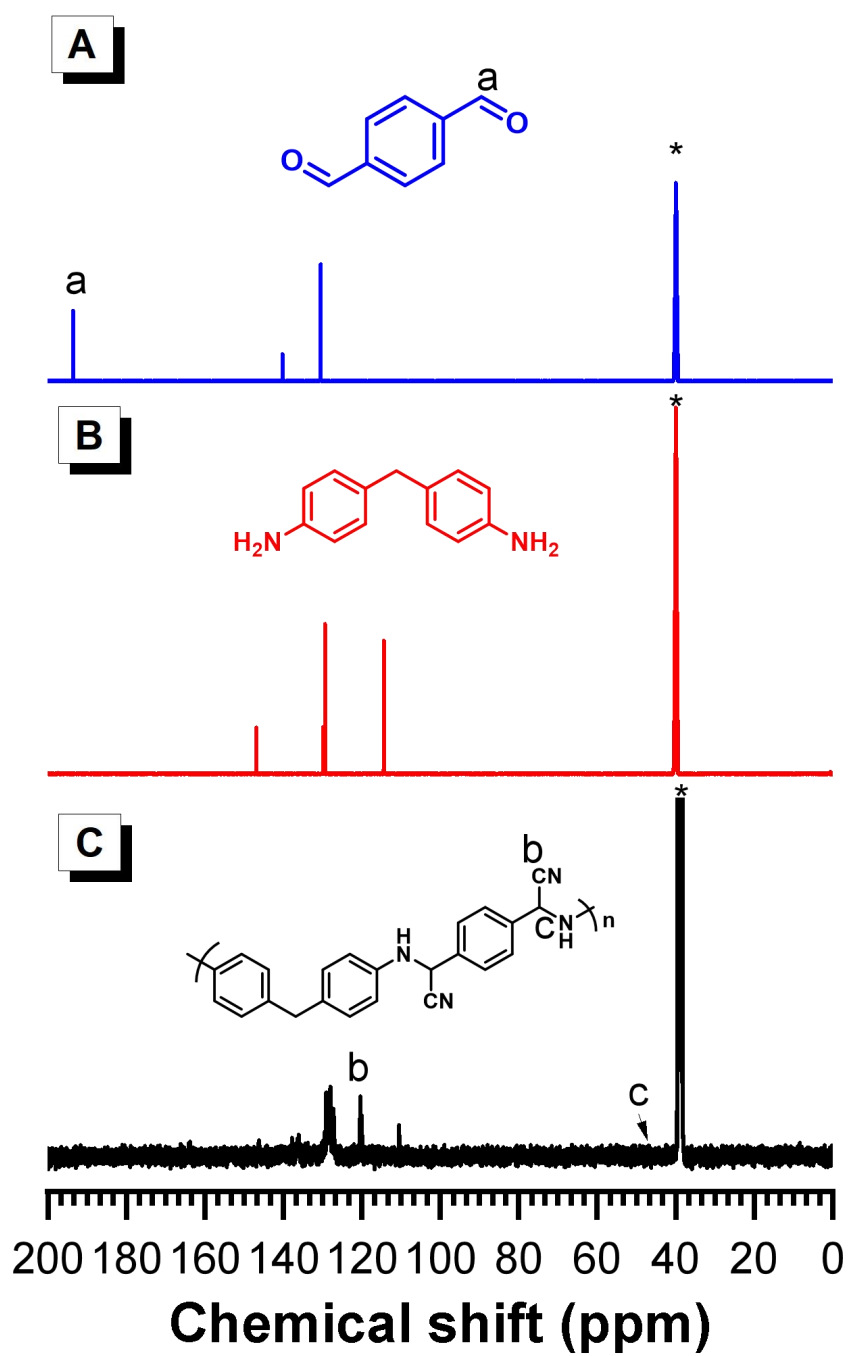
**Figure S26.**  $^{13}\text{C}$  NMR spectra of (A) **1a**, (B) **2c**, and (C) **P1a/2c/3** in  $\text{DMSO-}d_6$ . The solvent peaks are marked with asterisks.



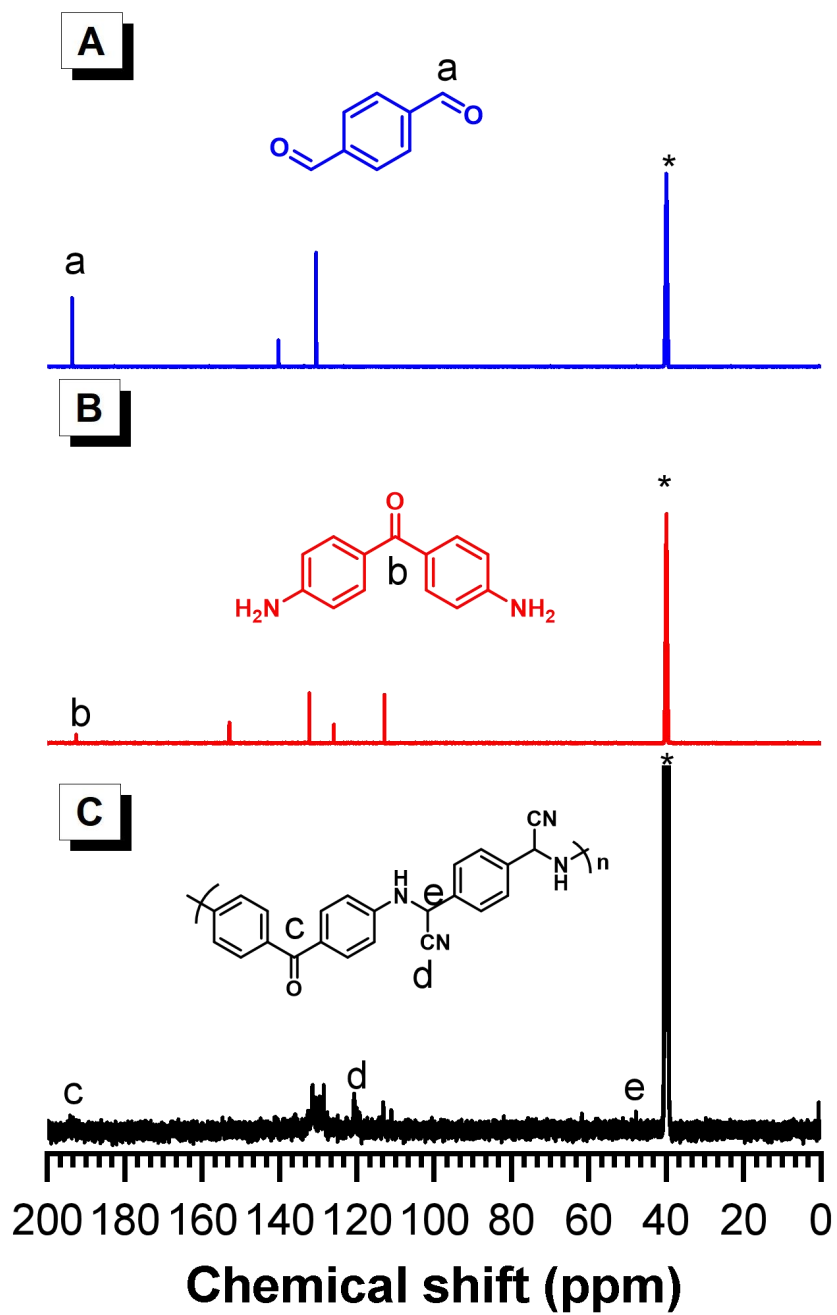
**Figure S27.**  $^{13}\text{C}$  NMR spectra of (A) **1a**, (B) **2d**, and (C) **P1a/2d/3** in  $\text{DMSO-}d_6$ . The solvent peaks are marked with asterisks.



**Figure S28.**  $^{13}\text{C}$  NMR spectra of (A) **1b**, (B) **2a**, and (C) **P1b/2a/3** in  $\text{DMSO-}d_6$ . The solvent peaks are marked with asterisks.

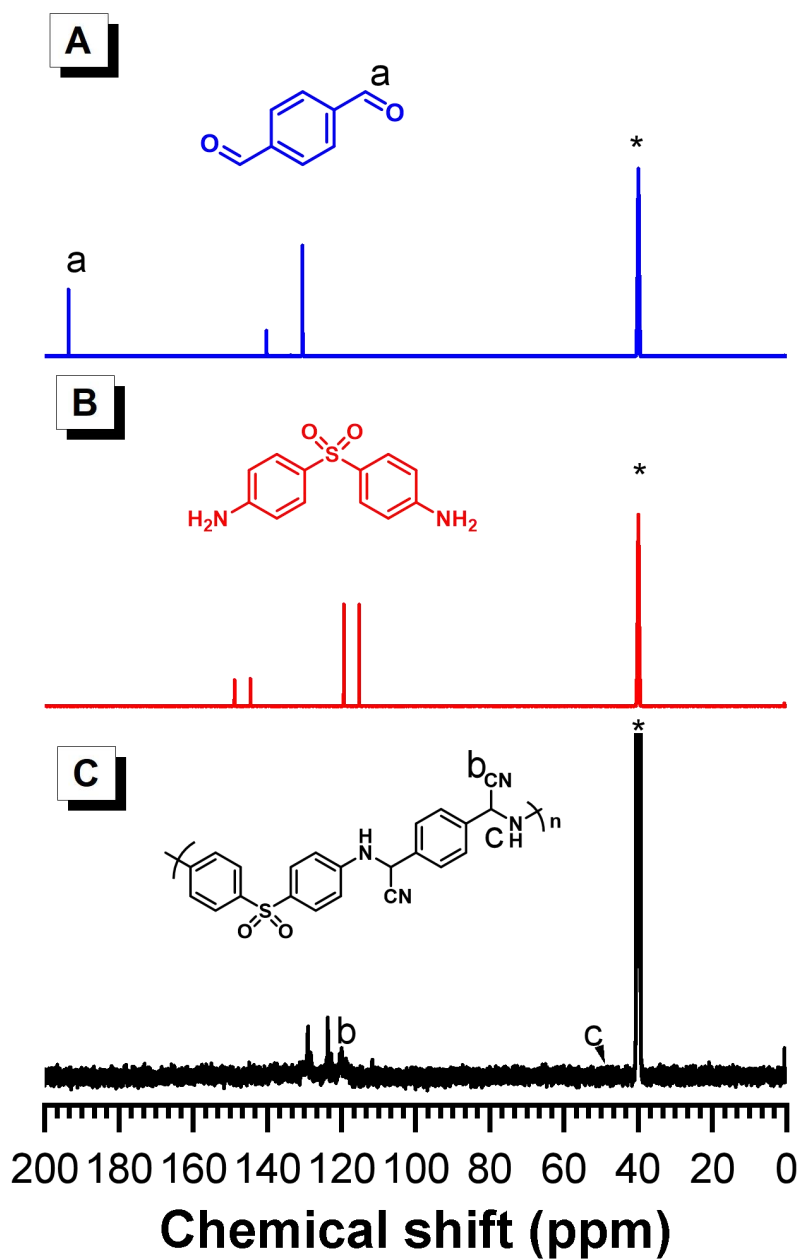


**Figure S29.**  $^{13}\text{C}$  NMR spectra of (A) **1b**, (B) **2b**, and (C) **P1b/2b/3** in  $\text{DMSO-}d_6$ . The solvent peaks are marked with asterisks.

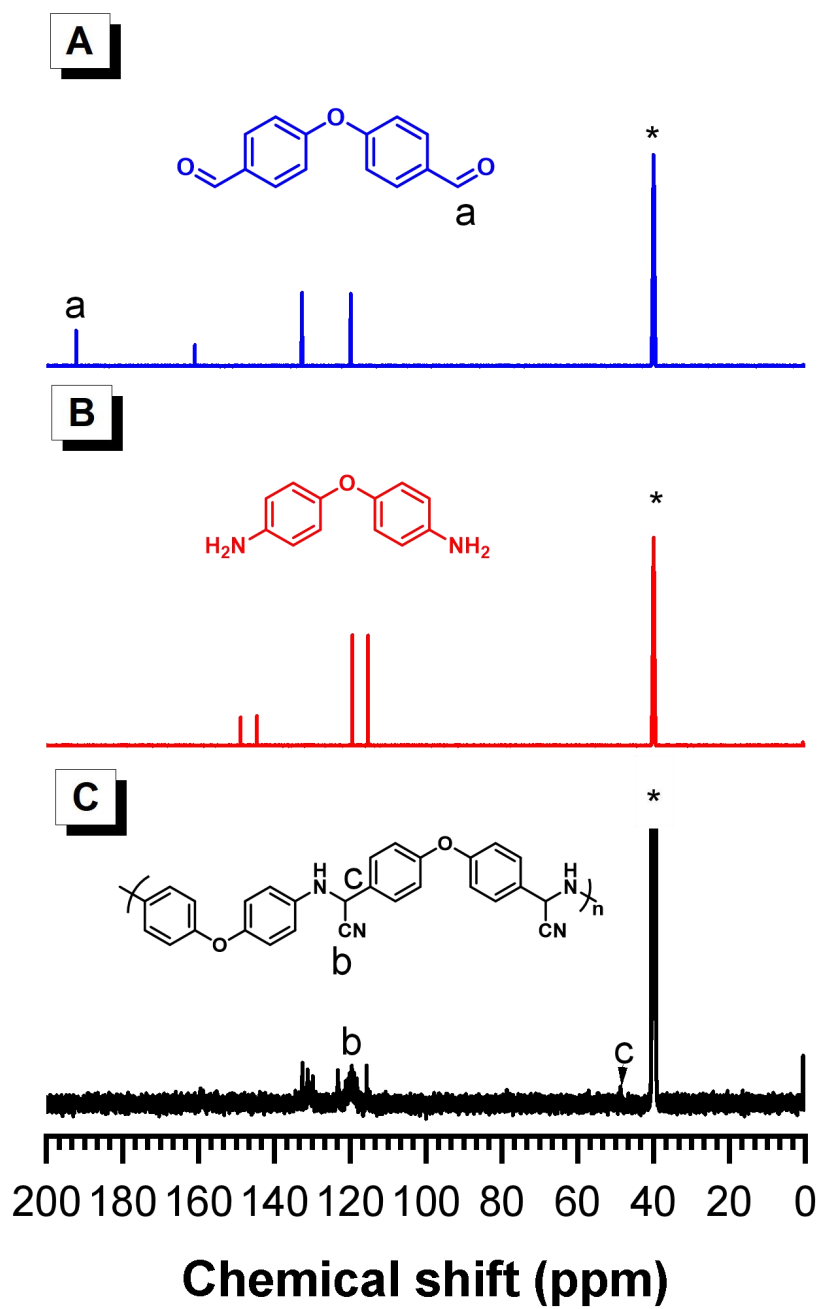


**Figure S30.**  $^{13}\text{C}$  NMR spectra of (A) **1b**, (B) **2c**, and (C) **P1b/2c/3** in  $\text{DMSO-}d_6$ . The solvent peaks are marked with asterisks.

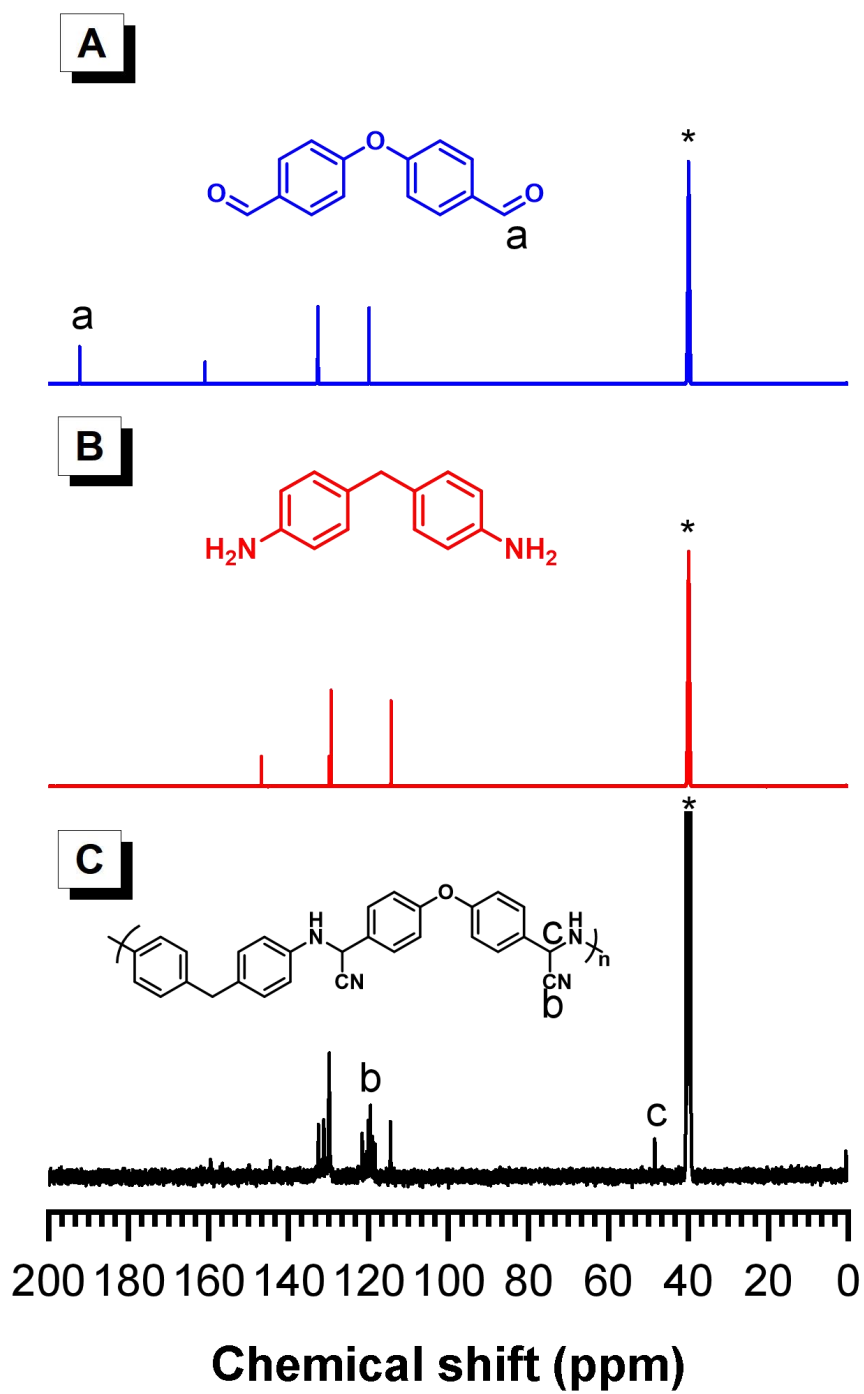




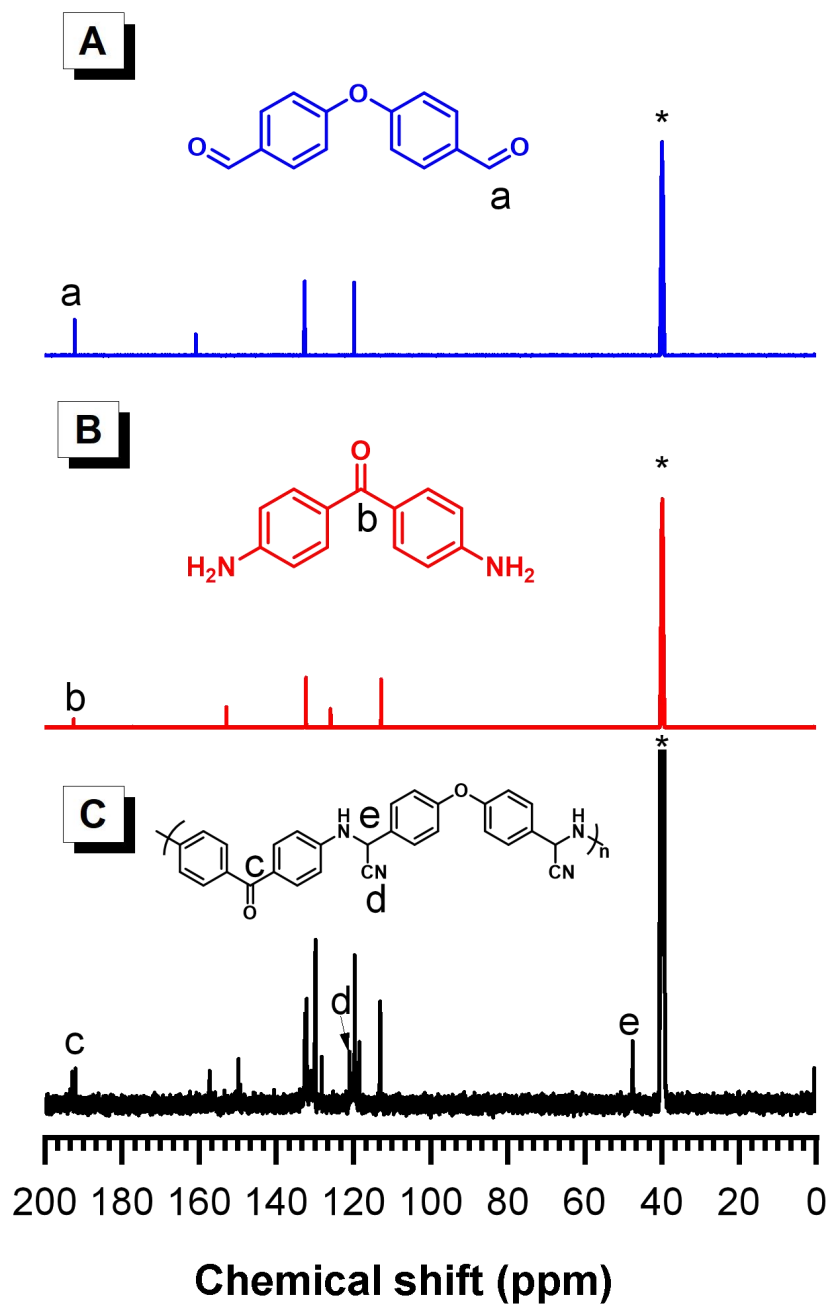
**Figure S31.**  $^{13}\text{C}$  NMR spectra of (A) **1b**, (B) **2d**, and (C) **P1b/2d/3** in  $\text{DMSO-}d_6$ . The solvent peaks are marked with asterisks.



**Figure S32.**  $^{13}\text{C}$  NMR spectra of (A) **1c**, (B) **2a**, and (C) **P1c/2a/3** in  $\text{DMSO-}d_6$ . The solvent peaks are marked with asterisks.



**Figure S33.**  $^{13}\text{C}$  NMR spectra of (A) **1c**, (B) **2b**, and (C) **P1c/2b/3** in  $\text{DMSO-}d_6$ . The solvent peaks are marked with asterisks.



**Figure S34.**  $^{13}\text{C}$  NMR spectra of (A) **1c**, (B) **2c**, and (C) **P1c/2c/3** in  $\text{DMSO-}d_6$ . The solvent peaks are marked with asterisks.

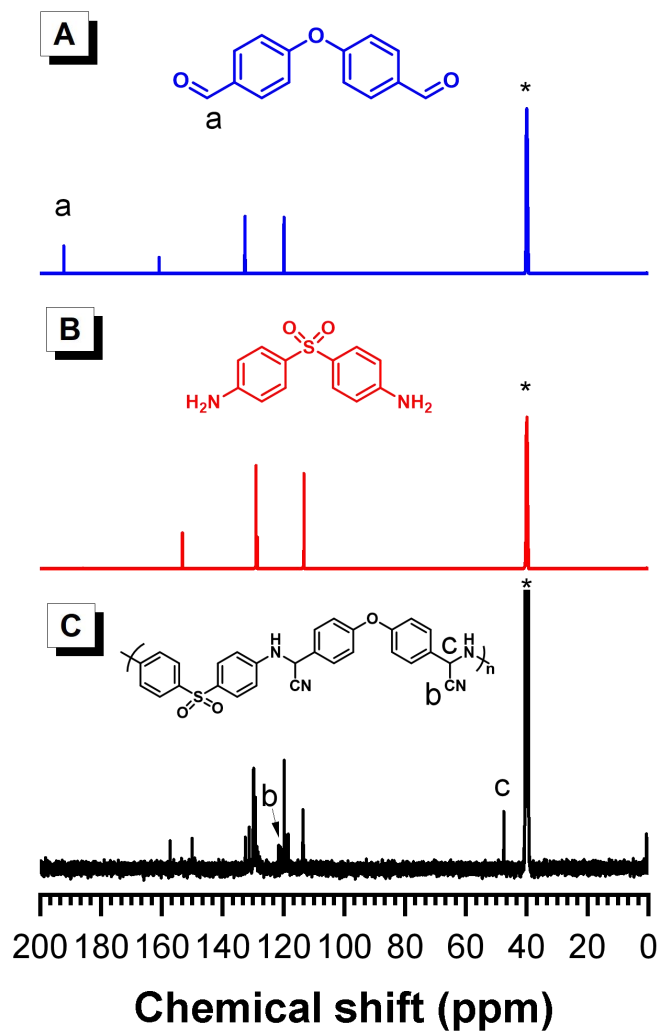


Figure S35.  $^{13}\text{C}$  NMR spectra of (A) **1c**, (B) **2d**, and (C) **P1c/2d/3** in  $\text{DMSO-}d_6$ . The solvent peaks are marked with asterisks.

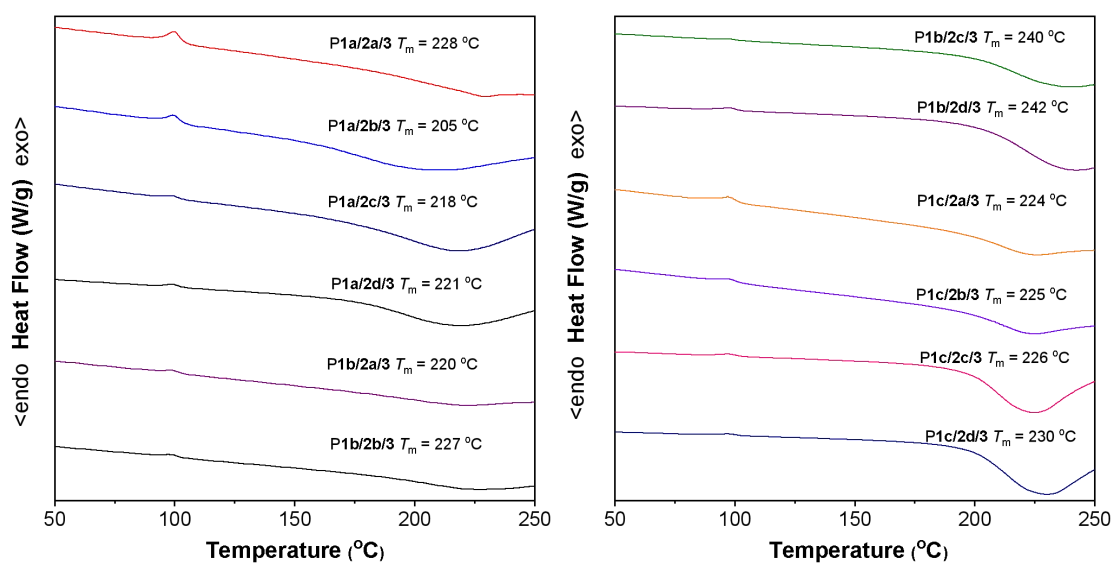
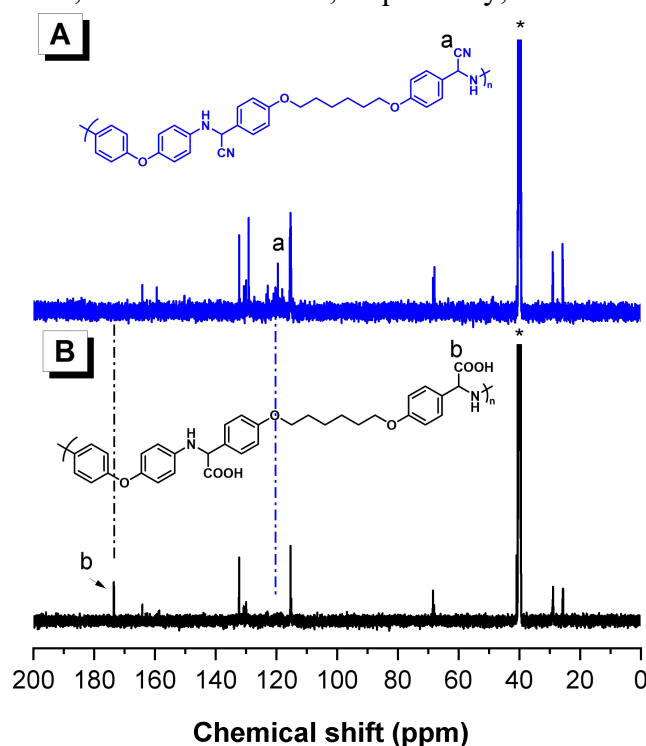


Figure S36. DSC curve of polymers.

**Table S1.** Refractive Indices, Abbé Numbers ( $\nu_D$ ), Revised Abbé Numbers ( $\nu'$ ), and Optical Dispersions ( $D$  and  $D'$ ) of Thin Films of **P1a/2/3**.<sup>a</sup>

Polymer	$n_{589}^b$	$n_{632.8}^c$	$\nu_D^d$	$D^d$	$\nu'^e$	$D'^e$
<b>P1a/2a/3</b>	1.7447	1.7322	11.2	0.0890	39.5	0.0253
<b>P1a/2b/3</b>	1.7006	1.6941	20.4	0.0491	75.7	0.0132
<b>P1a/2c/3</b>	1.7166	1.7049	11.5	0.0867	40.8	0.0245
<b>P1a/2d/3</b>	1.6793	1.6701	14.0	0.0714	50.4	0.0199

<sup>a</sup>Data taken from Figure 6B. <sup>b</sup> $n$  = RI value at 589 nm; <sup>c</sup> $n$  = RI value at 632.8 nm; <sup>d</sup> $\nu_D = (n_D - 1)/(n_F - n_C)$ , where  $n_D$ ,  $n_F$  and  $n_C$  are the RI values at wavelengths of Fraunhofer D, F and C spectral lines of 589.2, 486.1 and 656.3 nm, respectively;  $D = 1/\nu_D$ . <sup>e</sup> $\nu' = \text{Abbé numbers} = (n_{1319} - 1)/(n_{1064} - n_{1550})$ , where  $n_{1319}$ ,  $n_{1064}$  and  $n_{1550}$  are the RI values at wavelengths of 1319, 1064 and 1553 nm, respectively;  $D' = 1/\nu'$ .



**Figure S37.** <sup>13</sup>C NMR spectra of (A) **P1a/2a/3** and (B) postfunctionalized **P1a/2a/3** in DMSO-*d*<sub>6</sub>. The solvent and water peaks are marked with asterisks.

