**Supporting Information**

Synthesis of Superheat-Resistant Polyimides with High *T*g and Low Coefficient of Thermal Expansion by Introduction of Strong Intermolecular Interaction

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**Contents**

Figure S1. 1H NMR spectrum of the synthesized **DP** in DMSO-*d*6.

Figure S2. 13C NMR spectrum of the synthesized **DP** in DMSO-*d*6.

Figure S3. TOF-MS mass spectrum of **DP**.

Figure S4. 1H NMR spectra of PAAs solution (a) **PAA-a** (***m:n***=0:100), (b) **PAA-b** (***m:n***=0:100) and (c) PAA (***m:n***=100:0) in DMSO-*d*6.

Figure S5. ATR spectra of (a)PI-a and (b)PI-b films.

Figure S6. ATR spectra of C=N band for (a)PI-a and (b)PI-b films.

Figure S7. The geometry optimization of diamine structure.

Figure S8. TGA curves of (a)PI-a and (b)PI-b films.

Figure S9. DMA curves of (a)PI-a and (b)PI-b films.

Figure S10. TMA curves of (a)PI-a and (b)PI-b films.

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**Figure S1.** 1H NMR spectrum of the synthesized **DP** in DMSO-*d*6.

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**Figure S2.** 13C NMR spectrum of the synthesized **DP** in DMSO-*d*6.

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**Figure S3.** TOF-MS mass spectrum of **DP**.

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**Figure S4.** 1H NMR spectra of PAAs solution (a) **PAA-a** (***m:n***=0:100), (b) **PAA-b** (***m:n***=0:100) and (c) PAA (***m:n***=100:0) in DMSO-*d*6.

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**Figure S5.** ATR spectra of (a)**PI-a** and (b)**PI-b** films.

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**Figure S6.** ATR spectra of C=N band for (a)**PI-a** and (b)**PI-b** films.

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The geometry optimization was carried out using Chem3D Pro 14.0 under MM2 force field module. Then through the distance measurement method to obtain the distance of two atoms and the angle of backbone was calculated according to eq:

**Figure S7.** The geometry optimization of diamine structure.

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**Figure S8.** TGA curves of (a)**PI-a** and (b)**PI-b** films.

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**Figure S9.** DMA curves of (a)**PI-a** and (b)**PI-b** films.

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**Figure S10.** TMA curves of (a)**PI-a** and (b)**PI-b** films.