

## Supporting information for

### **Tailoring the properties of semi-aromatic copolyimides through structural manipulation towards energy-storage applications**

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#### **Table of content**

**Table S1.** Performance of commercial polyimides and of some developed polyimide films.

**Table S2.** Co-monomer ratio of copolyimides coPI1–coPI9.

**Table S3.** Solubility of copolyimides coPI1–coPI9.

**Figure S2.** WAXD patterns of copolyimides coPI1–coPI9.

**Figure S3.** AFM topography 3D images of copolyimides coPI1–coPI9.

**Figure S4.** DSC curves of copolyimides coPI1–coPI9.

**Figure S5.** The evolution of dielectric constant with frequency at room temperature for copolyimides incorporating (a) J-600, (b) J-900 and (c) J-2000.

**Figure S6.** The evolution of dielectric loss with frequency at room temperature for copolyimides based on (a) J-600, (b) J-900 and (c) J-2000.

**Figure S7.** The evolution of conductivity with frequency at room temperature for copolyimides containing (a) J-600, (b) J-900 and (c) J-2000.

#### **References**

**Table S1.** Performance of commercial polyimides and of some developed polyimide films.

| Material            | T <sub>g</sub><br>(°C) | ε <sub>r</sub><br>(25°C, 1 kHz) | tan δ<br>(25°C) | E <sub>b</sub><br>(MV/m) | U <sub>e</sub><br>(J/cm <sup>3</sup> ) | Reference |
|---------------------|------------------------|---------------------------------|-----------------|--------------------------|--|-----------|
| Ultem®              | 217                    | 3.2–3.1                         | ~0.3            | 439 (150°C)              | 1.14                                   | 1         |
| Kapton®             | 360                    | 3.3–2.8                         | 0.1             | 154–303<br>(7.6–127 μm)  | 0.59                                   | 2         |
| Upilex-S            | 355                    | 3.3                             | 0.1             | 272                      | -                                      | 3         |
| BTDA-<br>HK511      | 78                     | 7.8                             | 0.555           | 676                      | 15.77                                  | 4         |
| BTDA-<br>HK25-HDA   | 150                    | 4.77                            | <0.01           | 775                      | 12                                     | 5         |
| PI-PEI<br>(50%-50%) | 248                    | 3                               | ~0.17           | 1000                     | 8                                      | 6         |
| ODPA-pDS            | 290                    | 5.98                            | 0.00373         | 536                      | 7.60                                   | 7         |
| BTDA-<br>BPBPA      | 296                    | 7.2                             | 0.038           | 295                      | 2.77                                   | 8         |
| BTDA-2CN            | 325                    | 4.8                             | 0.00157         | 219                      | 1.02                                   | 9         |

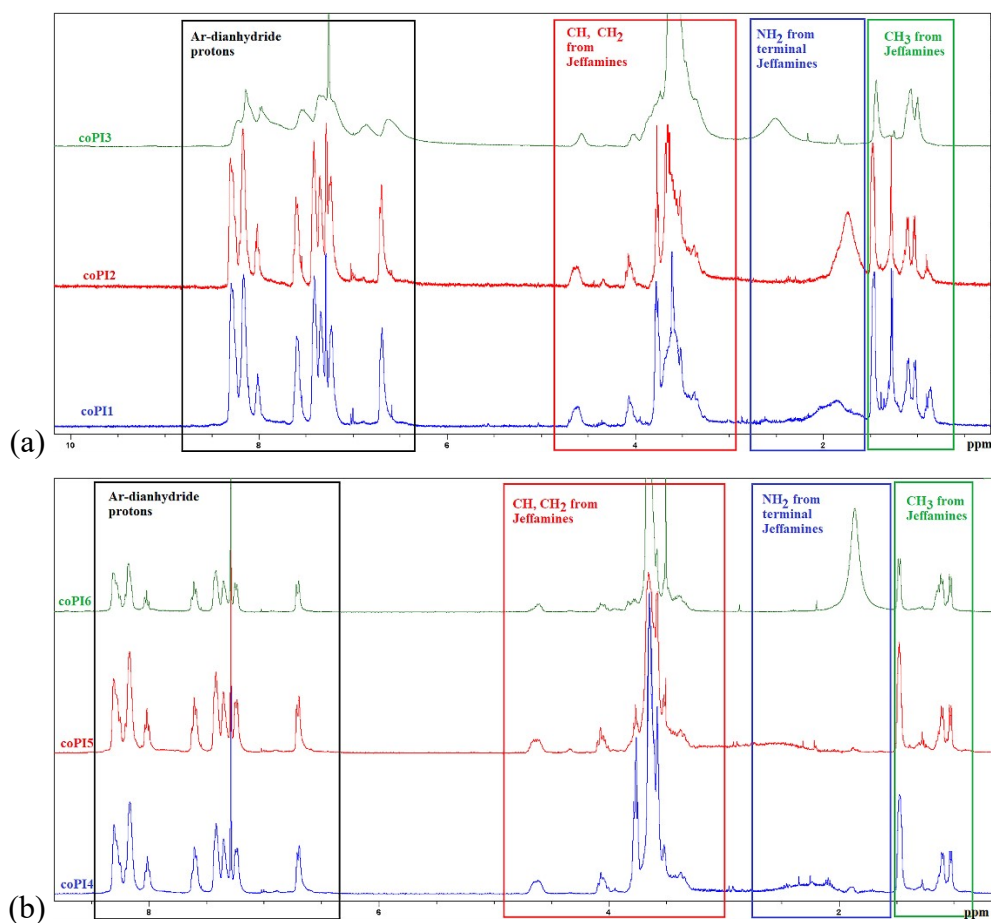
**Table S2.** Co-monomer ratio of copolyimides coPI1–coPI9.

| Copoly-<br>imides | Targeted molar ratio of<br>the structural units |        | Experimental molar ratio of<br>the structural units<br>from <sup>1</sup> H-NMR |        | Experimental wt. % ratio<br>of structural units<br>from <sup>1</sup> H-NMR |        |
|-------------------|---|--------|--|--------|--|--------|
|                   | Ar-BTDA   | J-BTDA | Ar-BTDA  | J-BTDA | Ar-BTDA  | J-BTDA |
| coPI1             | 0.7   | 0.3    | 0.65   | 0.35   | 56.31  | 43.69  |
| coPI2             | 0.7   | 0.3    | 0.64   | 0.36   | 47.71  | 52.29  |
| coPI3             | 0.7   | 0.3    | 0.79   | 0.21   | 50.24  | 49.76  |
| coPI4             | 0.6   | 0.4    | 0.55   | 0.45   | 44.93  | 55.07  |
| coPI5             | 0.6   | 0.4    | 0.59   | 0.41   | 42.30  | 57.70  |
| coPI6             | 0.6   | 0.4    | 0.63   | 0.37   | 31.25  | 68.75  |
| coPI7             | 0.5   | 0.5    | 0.46   | 0.54   | 36.82  | 63.18  |
| coPI8             | 0.5   | 0.5    | 0.43   | 0.57   | 27.72  | 72.28  |
| coPI9             | 0.5   | 0.5    | 0.58   | 0.42   | 26.89  | 73.11  |

**Table S3.** Solubility of copolyimides coPI1–coPI9.

| Copoly-imide | Solvent           |     |     |     |     |      |      |     |
|--------------|-------------------|-----|-----|-----|-----|------|------|-----|
|              | CHCl <sub>3</sub> | THF | ACN | DCM | DMF | DMAc | DMSO | NMP |
| coPI1        | ++                | ++  | ±   | ++  | ++  | ++   | ++   | ++  |
| coPI2        | ++                | ++  | ±   | ++  | ++  | ++   | ++   | ++  |
| coPI3        | ±                 | ++  | –   | ±   | ++  | ++   | ++   | ++  |
| coPI4        | ++                | ++  | ±   | ++  | ++  | ++   | ++   | ++  |
| coPI5        | ++                | ++  | ±   | ++  | ++  | ++   | ++   | ++  |
| coPI6        | ++                | ++  | ±   | ++  | ++  | ++   | ++   | ++  |
| coPI7        | ++                | ++  | ±   | ++  | ++  | ++   | ++   | ++  |
| coPI8        | ++                | ++  | ±   | ++  | ++  | ++   | ++   | ++  |
| coPI9        | ++                | ++  | ±   | ++  | ++  | ++   | ++   | ++  |

++ soluble at room temperature; ± partially soluble



**Figure S1.** <sup>1</sup>H-NMR spectra of copolyimides (a) coPI1–coPI3 obtained from a molar ratio between aromatic diamine and aliphatic diamine of 0.7:0.3, and (b) coPI4–coPI6 obtained from a molar ratio between aromatic diamine and aliphatic diamine of 0.6:0.4.

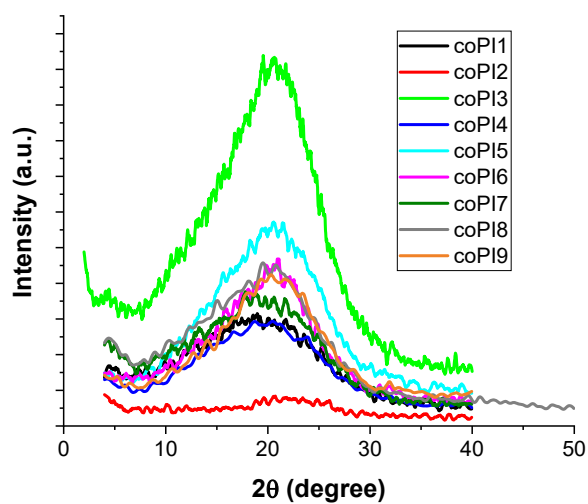


Figure S2. WAXD patterns of copolyimides coPI1–coPI9.

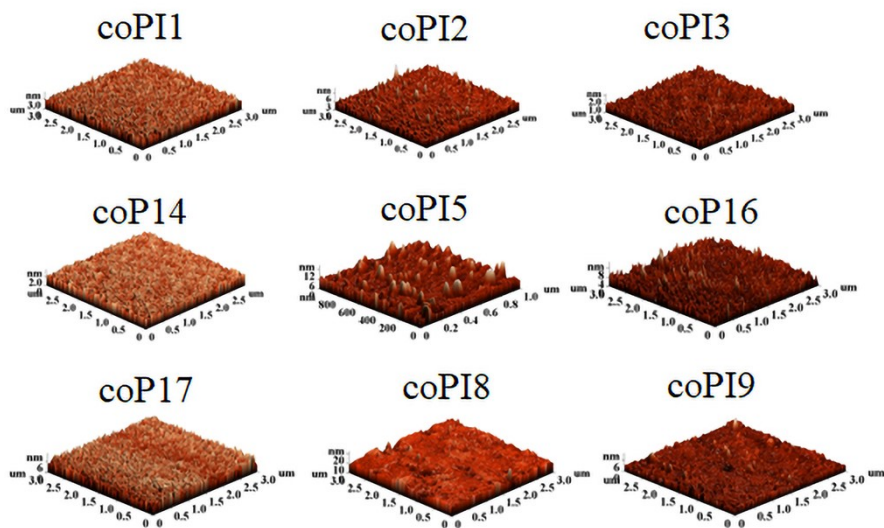


Figure S3. AFM topography 3D images of copolyimides coPI1–coPI9.

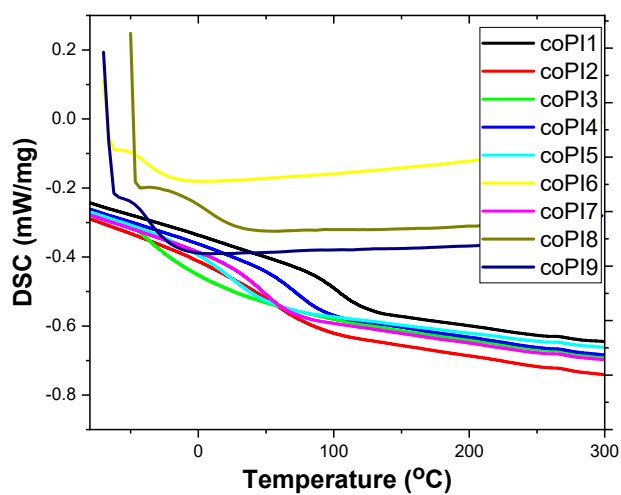


Figure S4. DSC curves of copolyimides coPI1–coPI9.

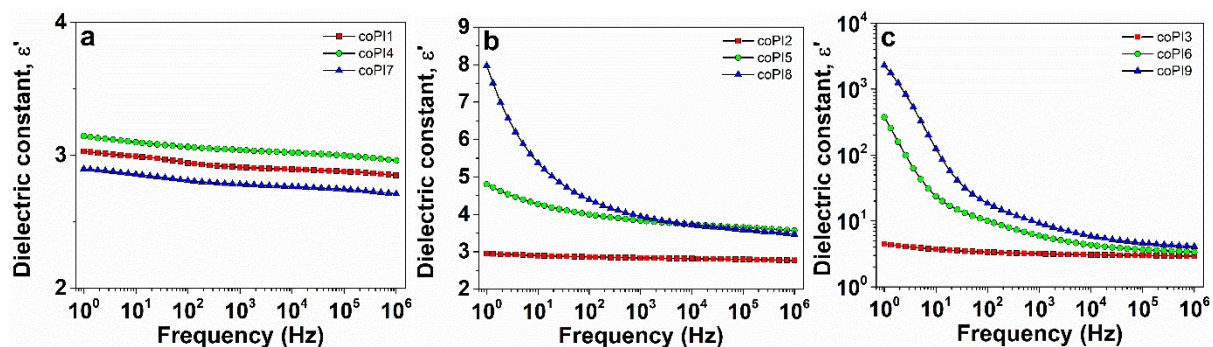


Figure S5. The evolution of dielectric constant with frequency at room temperature for copolyimides incorporating (a) J-600, (b) J-900 and (c) J-2000.

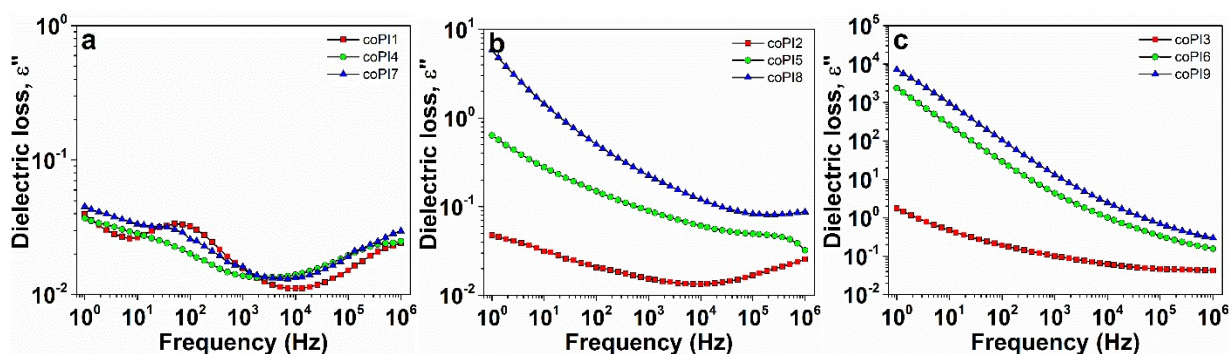


Figure S6. The evolution of dielectric loss with frequency at room temperature for copolyimides based on (a) J-600, (b) J-900 and (c) J-2000.

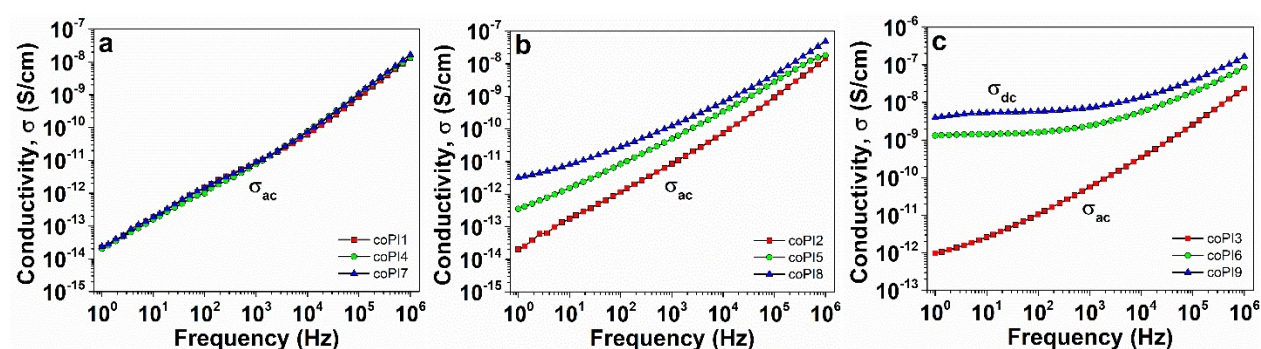


Figure S7. The evolution of conductivity with frequency at room temperature for copolyimides containing (a) J-600, (b) J-900 and (c) J-2000.

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