

Supplementary Materials: The Pivotal Role of Benzimidazole in Improving the Thermal and Dielectric Performance of Upilex-Type Polyimide

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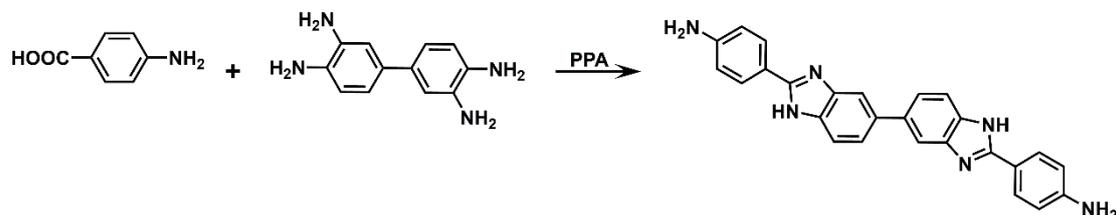
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The synthesis and characterization of 4,4'-[5,5'-bi-1H-benzimidazole]-2,2'-diylbis-benzenamine (BB)

The synthesis procedure was shown in Scheme S1 as follows: 160 g Poly(phosphoric acid), 10.7 g 3,3'-diaminobenzidine (50 mmol) and 13.7 g 4-aminobenzoic acid (100 mmol) were added to a dried 500 mL three-neck flask equipped with a thermometer, condenser, and a mechanical stirrer. The mixture was heated to 200 °C and reacted at this temperature for 12 h. Then cooled to 60 °C and poured into ice-cold water with rapid stirring. Finally, the precipitate formed was soaked in a 5% sodium hydrogen carbonate solution overnight. The crude product was purified by chromatography eluting with ethyl acetate/ethanol (3:1, v/v) to afford a light yellow solid. The crude product dissolved DMSO and then precipitated in water to obtain a white solid. ¹H NMR (500 MHz, DMSO-*d*₆) δ 12.51 (s, 2H), 7.88 (d, *J* = 8.5 Hz, 4H), 7.73 (s, 2H), 7.57 (d, *J* = 7.6 Hz, 2H), 7.47 (dd, *J* = 8.3, 1.5 Hz, 2H), 6.70 (d, *J* = 8.6 Hz, 4H), 5.63 (s, 4H).



Scheme S1. Synthesis of BB.

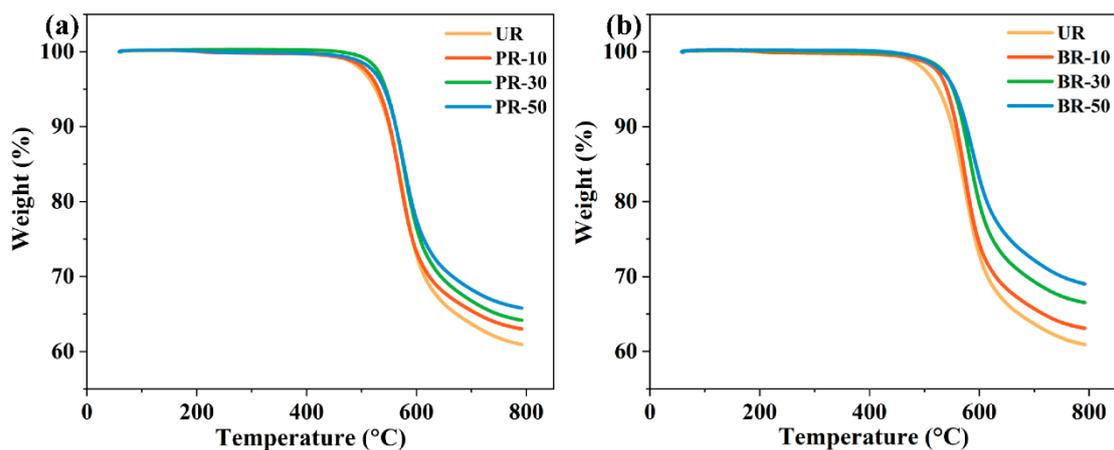


Figure S1. TGA curves of PI films (a) series PR and (b) series BR.

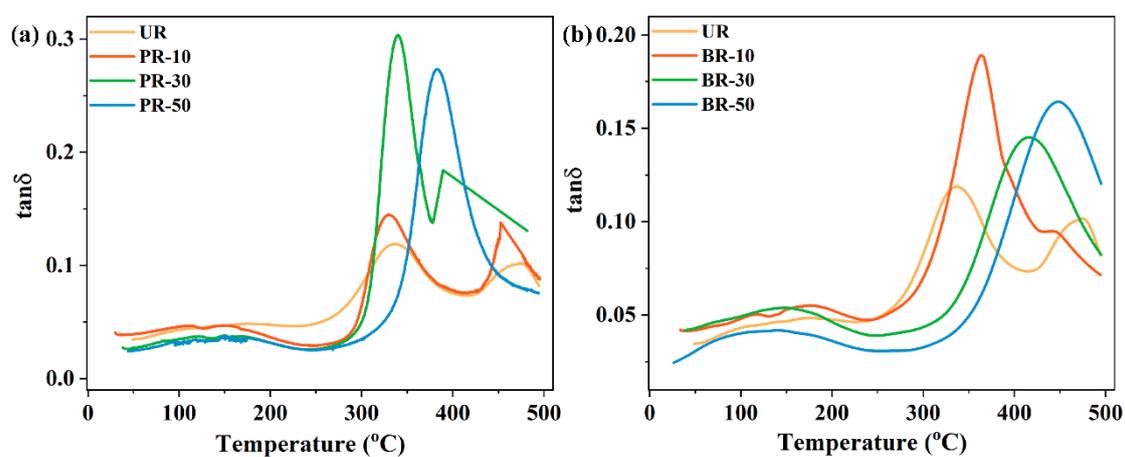


Figure S2. DMA curves of PI films (a) series PR and (b) series BR.

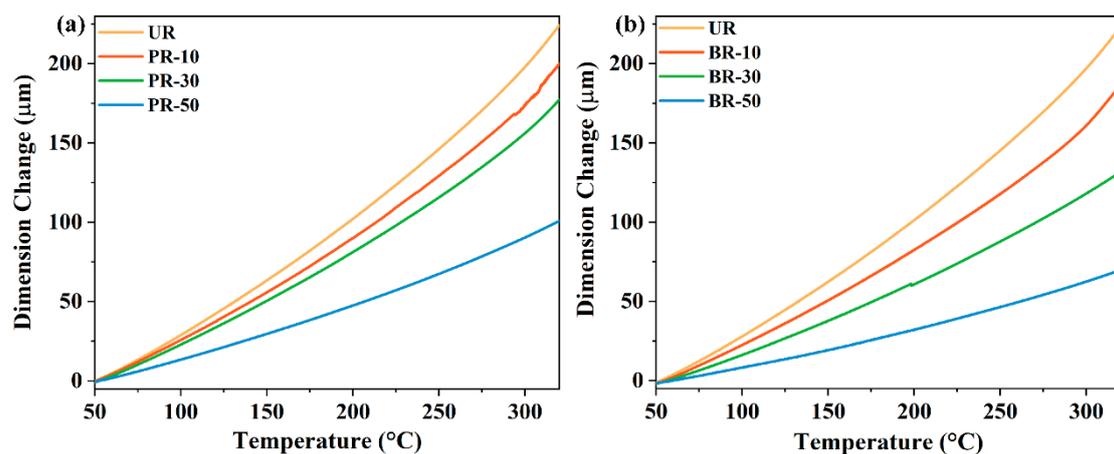


Figure S3. TMA curves of PI films (a) series PR and (b) series BR.

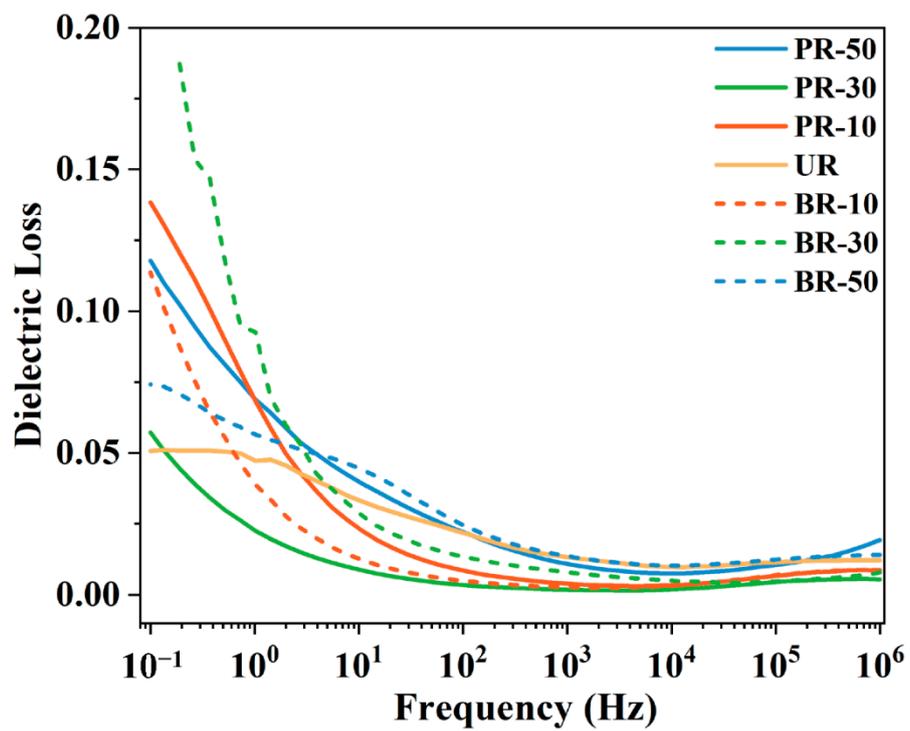


Figure S4. Frequency-scan broadband dielectric loss spectroscopy of polyimides.