

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

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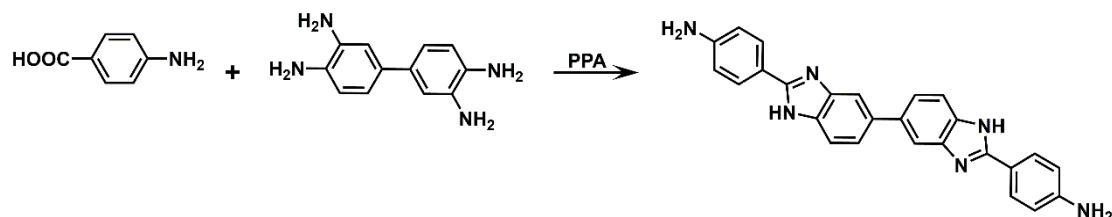
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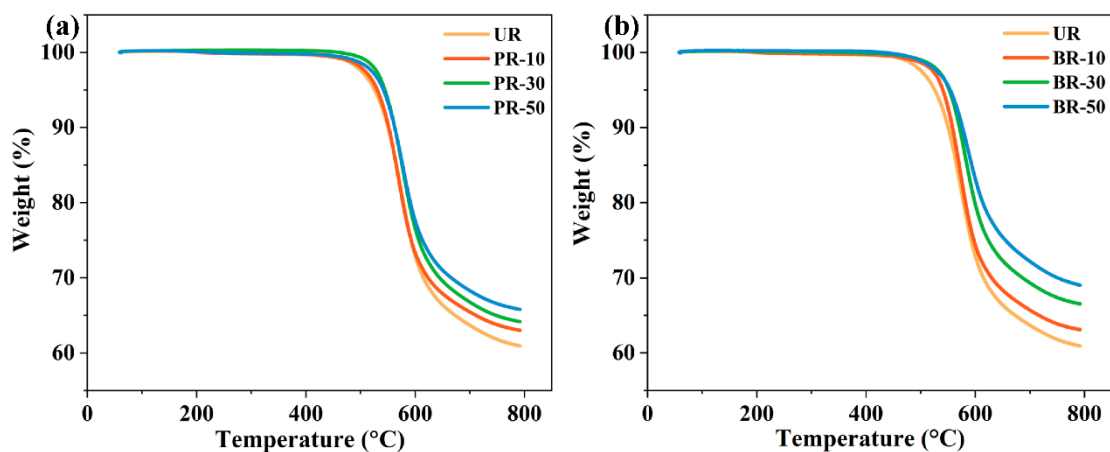
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## The synthesis and characterization of 4,4'-[5,5'-bi-1H-benzimidazole]-2,2'-diylbis-benzenamine (BB)

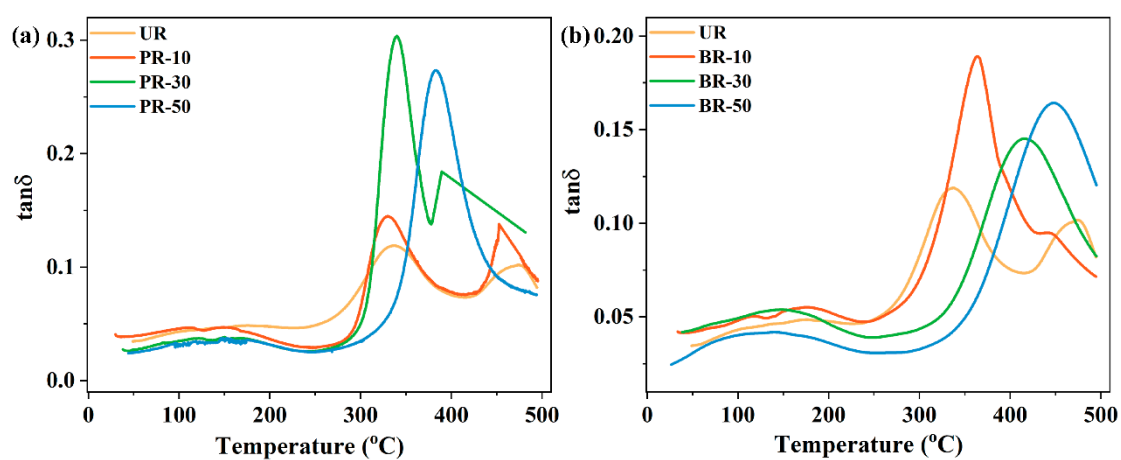
The synthesis procedure was shown in Scheme S1 as follows: 160 g Poly(phosphoric acid), 10.7 g 3,3'-diaminobenzidine (50 mmol) and 13.7 g 4-aminobenzoic acid (100 mmol) were added to a dried 500 mL three-neck flask equipped with a thermometer, condenser, and a mechanical stirrer. The mixture was heated 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. <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>) 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).



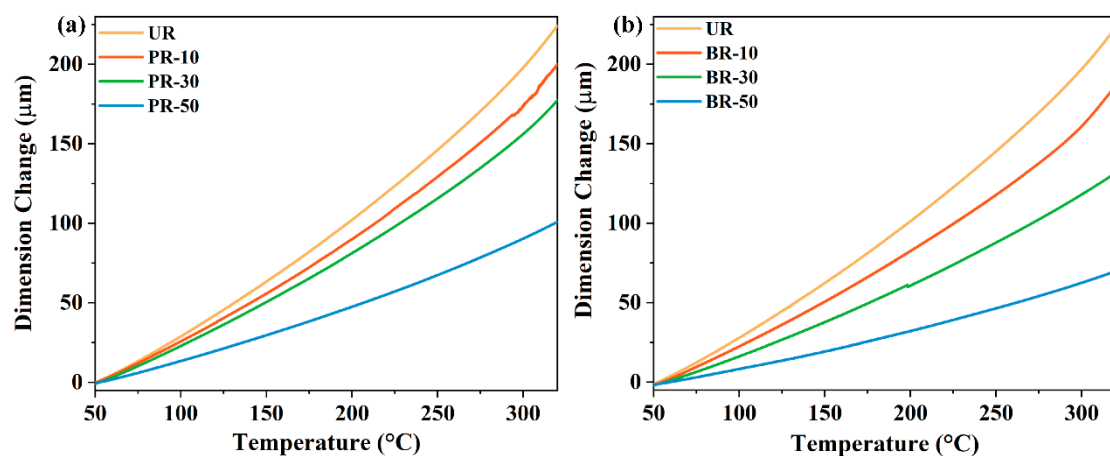
Scheme S1. Synthesis of BB.



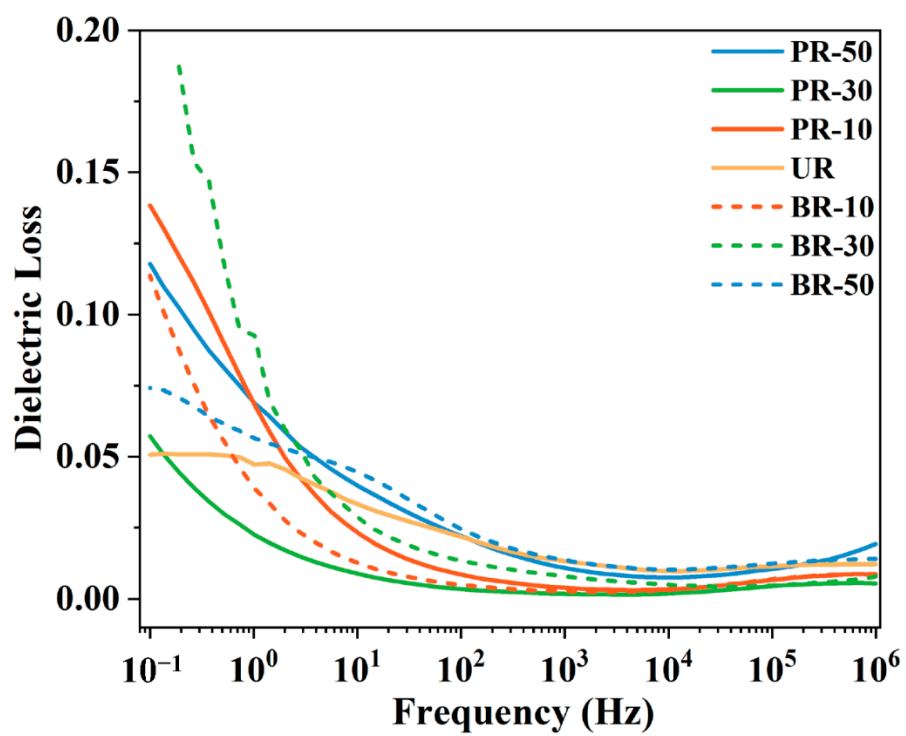
**Figure S1.** TGA curves of PI films (a) series PR and (b) series BR.



**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.