

## Synthesis and Memory Behavior Study of Functional Copolyimides Containing Pyrene Group

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In recent years, functional polyimide-based resistive type memory device has become a remarkable area due to their outstanding advantages such as low cost potential, large storage capacity, high mechanical strength, excellent chemical and thermal stability and potential application in three-dimensional stacking structure devices [1-3]. However, the relationship between the chemical structure of polyimide active layer and the corresponding memory behavior is still not well-known.

Targeting for an even deeper understanding and producing new-generation flexible electronics with anticipant memory behavior, a series of functional copolyimides, which were denoted as BAPF<sub>X</sub>DAPAP<sub>100-X</sub>-PI (in which X represented the molar ratio of BAPF and equaled to 0, 50, 80, 90, 95, 99 and 100), were prepared from the dianhydride, 3,3',4,4'-diphenylsulfonetetracarboxylic dianhydride (DSDA) and the diamines, 9,9'-bis(4-aminophenyl)fluorene (BAPF) and N,N-bis(4-aminophenyl)aminopyrene (DAPAP).

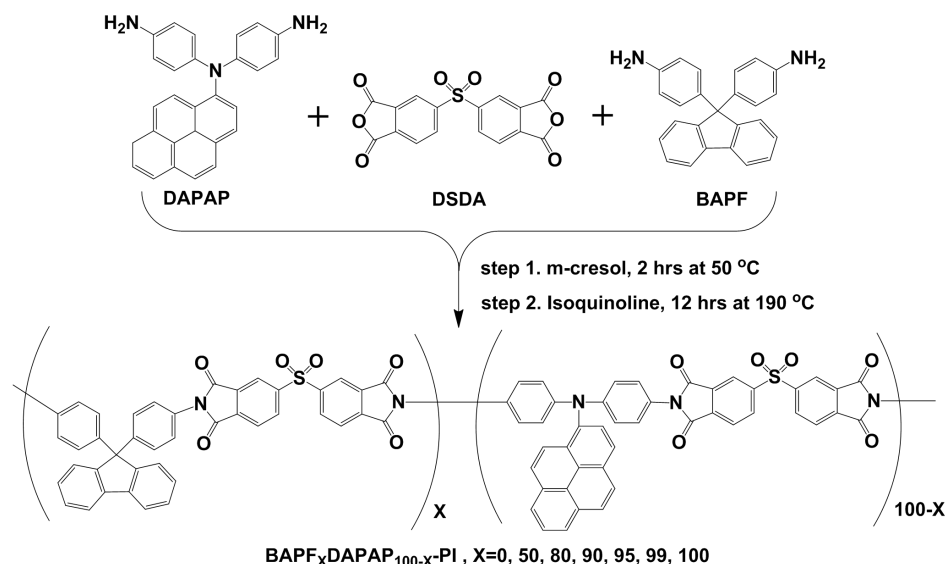


Figure 1. Synthesis route of BAPF<sub>X</sub>DAPAP<sub>100-X</sub>-PI with X equals to 0, 50, 80, 90, 95, 99 and 100.

The sandwich memory devices using the copolyimides as the active layer, the pre-cleaned ITO substrate as the bottom electrode and Au as the top electrode, were fabricated elaborately and exhibited tunable electrical bistability from nonvolatile write once read many times (WORM) (X=0, 50, 80, 90) to volatile static random access memory (SRAM) (X=95, 99, 100) with the increasement of BAPF composition. The molecular simulation results show that at the threshold voltage, an electron in BAPF<sub>0</sub>DAPAP<sub>100</sub>-PI transits from the HOMO to the LUMO+2 within the electron donor, overcoming a smaller energy gap of 3.32 eV, thus forming a more sable excited state compared to that of BAPF<sub>100</sub>DAPAP<sub>0</sub>-PI, resulting in two different memory behaviors of the corresponding copolyimides. As can be envisaged, the concept of structural design through copolymerization of various diamines and dianhydride demonstrated in present study may provide an alternative approach to modulate memory behaviors, thus expanding the application of polyimide memory electronics in information technology.

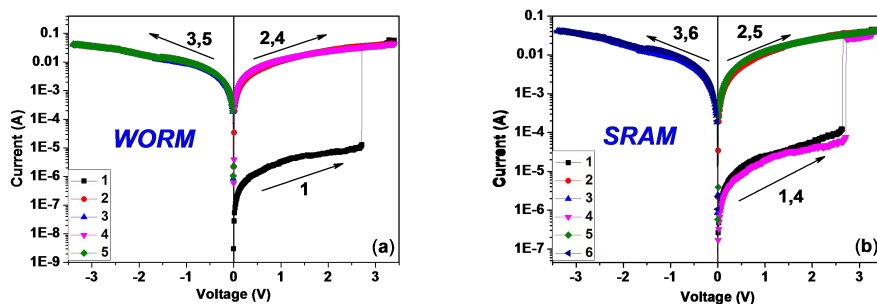


Figure 2. Two typical Current-Voltage (I-V) curves of sandwich memory devices with (a) BAPF<sub>0</sub>DAPAP<sub>100</sub>-PI and (b) BAPF<sub>100</sub>DAPAP<sub>0</sub>-PI as the active layer.

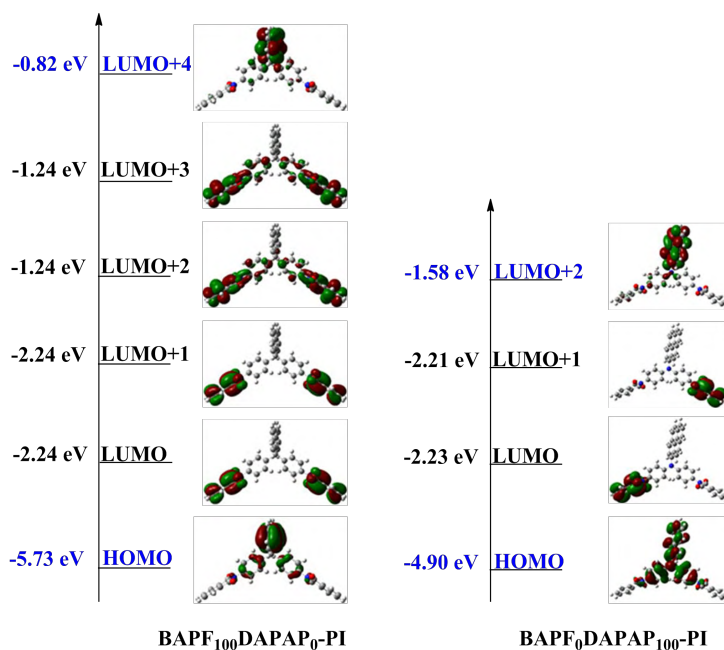


Figure 3. Molecular simulation results of BAPF<sub>100</sub>DAPAP<sub>0</sub>-PI and BAPF<sub>0</sub>DAPAP<sub>100</sub>-PI.

### References:

- [1] Q. D. Ling, F. C. Chang, Y. Song, C. X. Zhu, *Journal of America Chemistry Society*, 2006, 128, 27: 48732-73.
- [2] Q. D. Ling, D. J. Liaw, E. Y. H. Teo, C. X. Zhu, D. S. H. Chan, E. T. Kang, K. G. Neoh, *Polymer*, 2007, 48, 18: 5182-5201.
- [3] T. Kurosawa, T. Higashihara, M. Ueda, *Polymer Chemistry*, 2013, 4, 1: 16-30.