

Old Materials for New Technology: Transparent Polyimides with High T_g and Low CTE

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The current rapid change of form factor in portable and wearable electronic devices such as mobile phones with unprecedented features demands the development of transparent and flexible displays as a key element for the realization of the next-generation devices. Whereas polymer materials are uniquely suited to meet the required technical demands, transparency to visible light, high glass transition temperature (T_g), and low coefficient of thermal expansion (CTE) of polymers have proved difficult to realize simultaneously. Metals and ceramics have low CTE values in the range of 0.5 - 25 ppm/ $^{\circ}$ C and maintain their physical dimensions over a wide range of temperatures. However, non-crosslinked organic polymers have relatively large thermal expansion and show high CTE values ranging from 30 to 400 ppm/ $^{\circ}$ C. Polymers with rigid backbones and strong secondary interchain interactions, like aromatic polyimides and polyamides, have low CTE values due to the effective chain packing aided by secondary interchain interactions which are known to suppress thermal expansion, especially when the polymer chains are aligned. However, their low solubility makes it difficult to process them into films, and aromatic polyimides are known to have a yellow hue due to the presence of aromatic rings in the backbone and the formation of charge transfer complexes between the electron-deficient dianhydride and electron-rich diamine units.

In order to tackle these challenges, we investigated the use of aromatic poly(amide-imide) as a new platform. We selected a rigid and strongly interacting aromatic poly(amide-imide) backbone and added sterically bulky trifluoromethyl groups to the two diamine groups. These groups could create a complex hydrogen bonding network in two different directions. This approach resulted in polymers with approximately 90% transparency and low CTE values as low as 4 ppm/ $^{\circ}$ C. We found that by controlling the fraction of asymmetrically positioned trifluoromethyl groups in the polymer backbone, the CTE values could be precisely adjusted in the range of 4-20 ppm/ $^{\circ}$ C without sacrificing high transparency. Structural analyses of the chain packing combined with density functional theory (DFT) calculations indicated that the asymmetrically positioned trifluoromethyl groups increase the interchain distance and the number of interchain hydrogen bonds, contributing to both low CTE value and high transparency without the formation of charge transfer complexes.

