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Effects of Chemical Structure, Molecular Chain Orientation, and Aggregation State on the Thermo-optic Coefficients of Polyimide Films Measured by Optical Interferometry

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ABSTRACT:
Polyimides (PIs) exhibit excellent properties such as high thermal stability, chemical stability and mechanical strength. In case of optical applications such as thin film waveguides and micro lens, precise control of thermo-optic (TO) coefficient (*dn/dT*) is required. Optical interferometry is one of the powerful methods to measure TO coefficients of optically transparent thin films. In this study, the effects of the chemical structure, molecular orientation, and aggregation state of PI films on the TO coefficient were investigated by near-infrared optical interferometry. We synthesized PIs from the same dianhydride moiety (*s*BPDA) and eight kinds of diamines having linear or bent structures. In-plane and out-of-plane coefficients of thermal expansion ($CTE_{//}$, $CTE_{\perp}$) were evaluated by thermal mechanical analysis and optical interferometry, respectively, and coefficient of volumetric thermal expansion (CVE) was estimated by $CVE = 2\ CTE_{//} + CTE_{\perp}$. According to the Lorentz-Lorentz equation, average TO coefficient (dn_{av}/dT) could be increased with increasing the CVE and n_{av}. However, clear correlations were not observed among the TO coefficient, $CTE_{//}$, CVE and n_{av}. On the other hand, the TO coefficient showed positive correlations with $CTE_{\perp}$ and the degree of main chain orientation ($|P_{200}|$). The PI films having rigid and linear diamine moiety showed large TO coefficients due to their large values of $CTE_{\perp}$ and $|P_{200}|$. This result strongly suggests that TO coefficient measured by optical interferometry is not directly related to the average TO coefficient but to the anisotropy between the in-plane ($dn_{//}/dT$) and out-of-plane ($dn_{\perp}/dT$) directions.

Keywords: Thermo-optic coefficient, Polyimide, Refractive index, Optical interferometry, Molecular orientation