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Correlation between the Molecular Structures and Frequency-Dependent Dielectric Properties of Aromatic Polyimides in the 25–330 GHz Range

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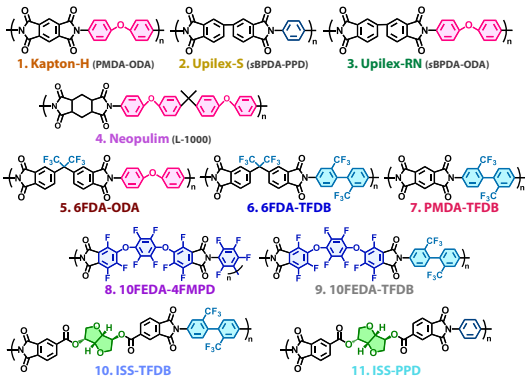
Introduction | The 5G mobile communication systems are currently operated in the sub-6 GHz to 39 GHz range, and it is expected that 6G systems will reach even higher frequencies, up to 300 GHz. With this technological advancement, the importance of polymer-based insulating materials featuring low dielectric constant (D_k) and low dielectric loss (D_f) has increased. Among these materials, polyimides (PIs) stand out due to their thermal stability, mechanical strength, and excellent dielectric properties. This study aims to analyze the dielectric dispersion of 11 different structured PIs (**Scheme 1**) across a wide frequency range from 25 GHz to 330 GHz, in order to establish new design guidelines for low-dielectric PIs suitable for 5G/6G mobile communications.



Scheme 1. Structures of PIs utilized in this study.

Experiment | Samples **1**–**4** were commercially available PIs, while the others **5**–**11** were obtained by thermal imidization of poly(amic acid) films. The frequency dependence of the D_k and D_f was measured using a Keysight Technologies N5290A network analyzer millimeter-wave system connected to Fabry-Perot resonators designed and manufactured by EM Labs Inc., with a frequency extension module.

Results and Discussion | **Fig. 1a** shows the D_k and D_f values of the PIs at four frequencies. The D_k of polymer materials is mainly determined by electronic polarization (P_e), while D_f is strongly influenced by dipolar orientational polarization (P_d). The D_k of all PIs decreased with increasing frequency, which can be attributed to the gradually decreasing contribution of P_d, making the D_k of each PI approach the square of its refractive index determined by P_e. Furthermore, PIs with a high fluorine content (**6**, **8**, **9**) exhibited significantly lower D_k in the GHz range compared to commercially available PIs (**1**, **2**, **3**), attributed to the reduced P_e of the PI main chain due to the introduction of plural fluorine atoms.

On the other hand, D_f showed an increasing trend with frequency, and the rate of increase (k_{Df}) was negatively correlated with the proportion of polar groups (*i.e.*, imide and ester groups) in each PI (*polar%*), as shown in **Fig. 1b**. While **6**, **8**, **9** exhibited significantly small D_f values at 30 GHz, **6** and **9** showed a larger k_{Df}, indicating relatively higher D_f at 330 GHz. PIs containing –CF_3 in the diamine moiety (**6**, **7**, **9**, **10**) showed a notable increase in D_f, suggesting the impact of a unique absorption in the THz region. Meanwhile, **1**, **2**, **3** maintained their low D_f even at 330 GHz, suggesting that the formation of a dense aggregate structure suppressed local oscillating motion, contributing to the reduction in D_f.

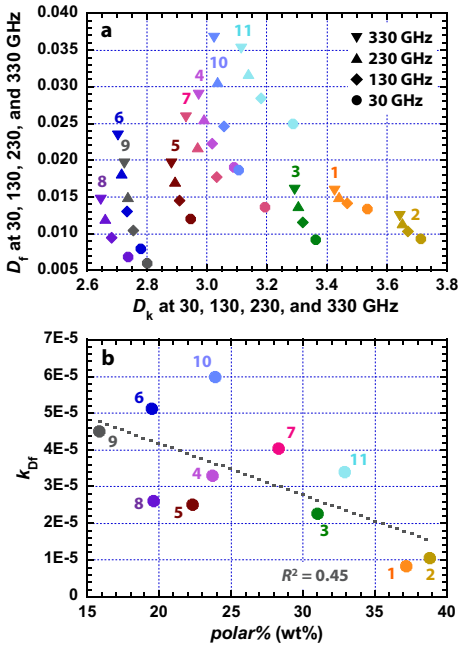


Fig. 1. (a) D_f *vs* D_k and (b) k_{Df} *vs polar%* of PIs.