Synthesis and Dielectric Properties of Polyimides Including Double-Decker Silsesquioxane (DDSQ) in the Main Chain

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Abstract : In recent years, the expansion of the Internet and advancements in the performance of electronic technology device has driven the demand for higher capacity and faster speeds in communication technologies. Beyond the traditional fourth-generation (4G) frequency bands, the advent of fifth-generation (5G) communication technology has brought into play new frequency bands in the millimeter wave region, notably at 20 GHz or higher. Consequently, developing heat-resistant materials with low dielectric properties for next-generation wireless communication devices has become crucial. Such materials are key to achieving faster communication speeds, enabling the connection of numerous devices simultaneously, and minimizing data processing delays.

In this study, we synthesized polyimides (PIs, figure 1) incorporating a double-decker-shaped silsesquioxane (DDSQ, figure 2) with a high silicon content in the main chain. The degree of polymerization of poly(amic acid)s was estimated using viscometry measurements and size exclusion chromatography (SEC) measurements, revealing viscosities ranging from approximately 0.2 to 0.5 and weight average molecular weights (M_w) from approximately 20,000 to 60,000. These PIs were successfully synthesized, resulting in self-supporting films with high thermal resistance. Dielectric property measurements at 10 GHz and 20 GHz demonstrated that the unique bulky molecular structure of DDSQ significantly contributes to achieving the low dielectric constants (D_k) around 2.80 and low loss tangent (D_f) around 0.003, while the D_k and D_f of fully aromatic PI (PMDA, pyromellitic dianhydride/ODA, 4,4'-oxydianiline) at 10 GHz were 3.1 and 0.014, respectively. The refractive indices were also lower than those of PI (PMDA/ODA), suggesting that high molecular size and low polarizability of DDSQ lead to an increase in molar volume and a decrease in overall PI polarization. The molecular mobility may be suppressed due to the rigidity of the molecular chains of DDSQ-including PIs, potentially resulting in a lower D_f. This investigation highlights the promising potential of PIs including DDSQ in the main chain for use in the next-generation wireless communication devices.

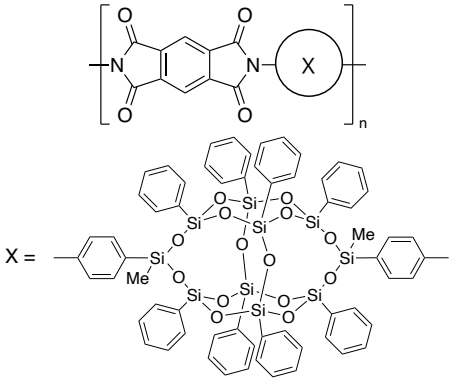

Figure 1. Chemical structure of double-decker silsesquioxane (DDSQ)-based polyimide

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