Liquid Crystal Polymer (LCP) for Next-Generation High-Speed Communication Applications

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Liquid crystal polymer (LCP) is a thermoplastic resin with low dielectric constant (relative permittivity) and dissipation factor in the high-frequency range and is expected to be applied to the next-generation high-speed communication applications. Based on our accumulated technologies, new LCP resins/compounds for these applications have been studied and some products with unique and excellent properties have been successfully developed. In this article, details of developed materials designed for films and connectors, which are promising applications of LCPs with low dielectric properties, are reported.

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Introduction

Next-generation high-speed, large-capacity communication systems will enable ultra-high-speed, ultra-low latency, and multiple simultaneous connections. As a result, they are expected to realize unprecedented new services and comfortable communication environments, such as remote medical care, improved autonomous driving, and immersive movies and sports viewing. Achieving such applications requires consideration of the system environment as well as the development of materials with low -transmission loss properties. This is because next-generation high-speed communication will be performed in high-frequency bands, which increases transmission loss compared to previous systems. Therefore, the resin materials used in circuit boards are required to have even lower dielectric constants and dielectric dissipation factors. Examples of materials that can fulfill these requirements include FR-4 (epoxy resin), polyimide resin, fluororesin, and liquid crystal polymers (LCP)1); however, a polymer material that excels in all aspects, such as dielectric properties, processability, and dimensional stability, remains elusive. Therefore, improvements to existing resins and the development of new resins are nascent.

LCP is a resin that has liquid crystal molecules known as mesogens bonded by ester bonds in the main skeleton, and has the properties of a highly aromatic and rigid primary structure, which is reflected when performing injection molding: when the resin is subjected to external forces, such as shear force or elongation stress during melting, it is strongly aligned in the direction of those forces, maintaining the orientation state when cooled and solidified (**Fig. 1**). For convenience, LCP is classified into types I, II, and III based on its heat resistance. **Fig. 2** presents these classifications along with the representative molecular structures. Only type I can be used with lead-free solder (approximately 260 °C), which is a crucial factor in applicability to electronic circuit components.

Additionally, as its name suggests, LCP is characterized by exhibiting liquid crystallinity in the molten state, and a unique phenomenon that is not observed in conventional polymers, that is, a large increase in the amount of transmitted light when heated under a polarized microscope, is observed in the melt state. As previously mentioned, the molecular chains of LCP are easily oriented through the application of shear force when melted, so the mechanical strength varies greatly based on the molding conditions (injection pressure, temperature, *etc.*). A thinner molded product results in a higher proportion of the skin layer, which increases the mechanical strength, while the strength perpendicular to the resin flow direction decreases; consequently, the material exhibits strong anisotropy. Thus, the anisotropy caused by the molecular orientation of LCP is both

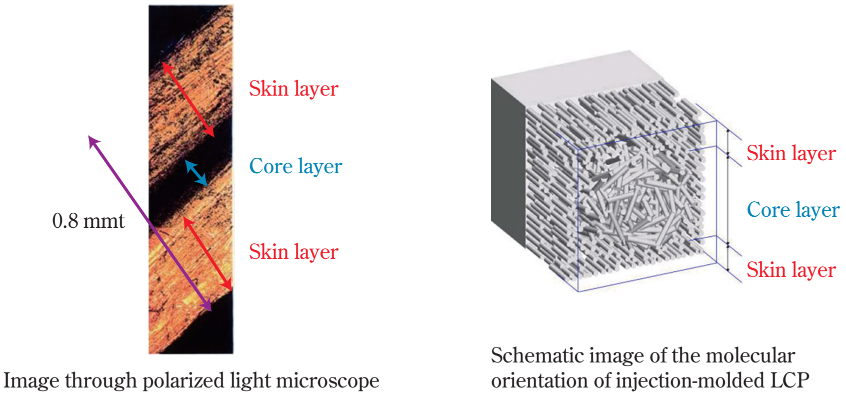


Fig. 1 Skin-core structure of LCP

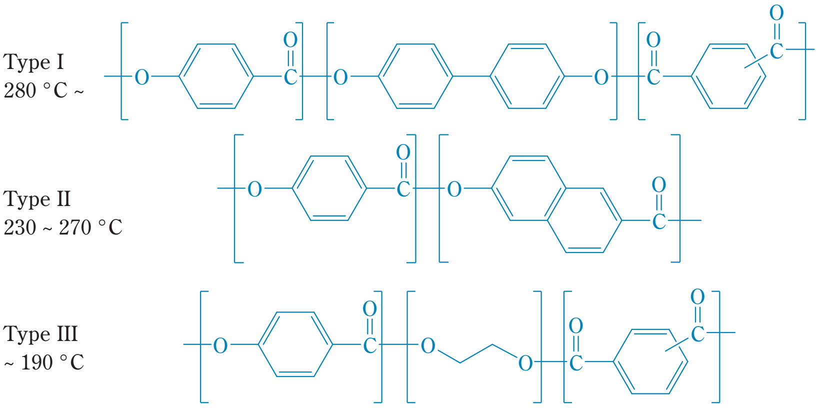


Fig. 2 Typical structure and corresponding soldering heat resistance temperature of LCP Type I - Ⅲ

an advantage and disadvantage. Therefore, fillers such as glass fiber (GF) are added to alleviate this anisotropy and optimize the molding conditions to use LCP in precision parts, such as connectors, relays, bobbins, and switches, which are commonly used in the in electrical, electronics, and information and communications fields. The excellent electrical properties, dimensional stability, and heat resistance of LCP have also attracted attention in recent years, and there has been active development of films and fibers using LCP.

Our company began its LCP business in the 1970s, and the technology that we have cultivated to date was used as a basis to investigate resins and compounds that could be used for the above purposes. Through our efforts, we developed an LCP material that can be used in a variety of parts, which is reported in this paper.

Development of LCP for melt extrusion

A representative type II LCP contains only two types of hydroxycarboxylic acids, *p*-hydroxybenzoic acid (HBA) and 2-hydroxy-6-naphthoic acid (HNA). It is a simple two-component system and has been studied in both academic and industrial contexts, being marketed as "Vectra" A series by Celanese Corp. The LCP contains only two types of hydroxycarboxylic acids, so the sequence is easily randomized and is less likely to produce a repeated sequence of HBA units with high-melting points that tend to degrade the physical properties; thus, the LCP has excellent moldability and mechanical properties$^{2)}$. As previously mentioned, the resin has a high degree of sequence randomness. Therefore, it is believed that the physical properties can be easily compared owing to differences in the original monomer composition. Accordingly, we synthesized LCPs with different ratios of HBA and HNA and evaluated their physical properties. **Fig. 3** shows the change in dielectric dissipation factor, and **Fig. 4** shows the temperature dependence of melt viscosity. As shown in **Fig. 3**, the dielectric dissipation factor decreases as the HNA ratio increases, with a minimum value found at approximately 75%.

The temperature dependence of the melt viscosity, particularly the viscosity change at low temperatures, was found to change based on the composition, and it is

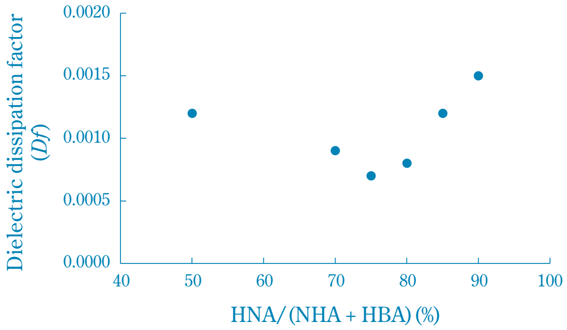


Fig. 3 Variation of dielectric dissipation factor of HNA/HBA copolymers as a function of HNA content

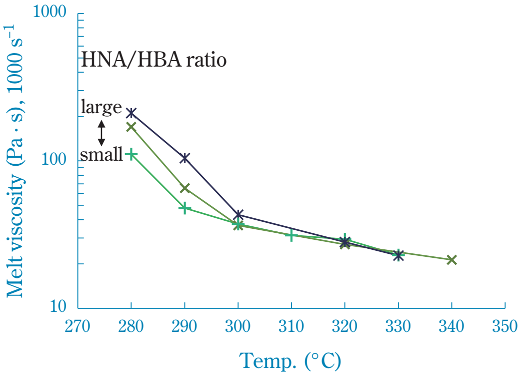


Fig. 4 Temperature dependence of melt viscosity of HNA/HBA copolymers with different HNA/HBA ratios

expected that this will impact moldability and film-forming properties (**Fig. 4**).

As mentioned in the Introduction, low transmission loss is a key performance requirement for electronic materials in next-generation high-speed communication applications. As shown in **Fig. 5**, transmission loss can be divided into conductor loss (α_c) that occurs mainly in signal lines made of metal materials and dielectric loss (α_d) that occurs in dielectrics such as LCP, and dielectric loss is expressed as the product of the dielectric constant (*Dk*) to the power of 1/2 and the dielectric dissipation factor (*Df*). Therefore, we focused on keeping the dielectric dissipation factor of LCP as low as possible and developed two types of LCP with excellent processability, which are introduced here.

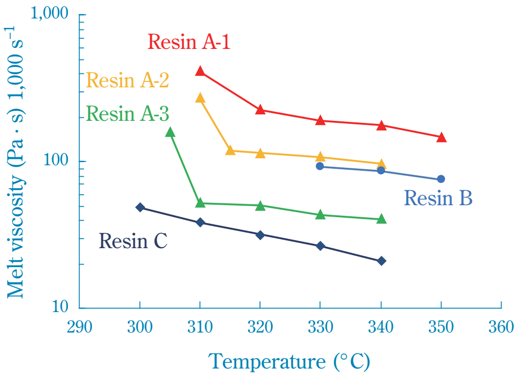
As presented in **Table 1**, these two types of LCP (resin A and resin B) have excellent dielectric dissipation factors and solder heat resistance. In particular, the dielectric dissipation factor is half the value of the LCP that is currently used by LCP film manufacturers (resin C), which makes it a material with potential to greatly reduce transmission loss. Additionally, for resin A, we created three different viscosity ranges, considering the processing method used by film manufacturers (**Fig. 6**).

$$\text{Transmission loss } \alpha = \text{Conductor loss } \alpha_c + \text{Dielectric loss } \alpha_d$$

$$\alpha_d \propto \sqrt{\varepsilon} \times \tan\delta$$

$$(\varepsilon\text{: } Dk\text{, } \tan\delta\text{: } Df)$$

Fig. 5 Transmission loss formula



Test conditions: JIS K7199, A2 method
Inner diameter of capillary die = 0.5 mm
Length of capillary die = 10 mm

Fig. 6 Temperature dependence of melt viscosity of low dielectric dissipation factor resins A and B

Table 1 Physical properties of low dielectric dissipation factor resins A and B

		Resin A	Resin B	Resin C
Melting point (T_m)	°C	315	323	280
Melt viscosity (T_m + 20 °C)	Pa · s	100	85	50
Soldering heat resistance temperature	°C	275	280	230
Water absorption rate	%	< 0.05	< 0.05	< 0.05
Relative permittivity (*Dk*)*		3.5	3.6	3.3
Dielectric dissipation factor (*Df*)*		0.001	< 0.001	0.002

*: Split cylinder resonator method, 10 GHz, 23 °C, 50%RH

Currently, we are providing these lineups to film manufacturers to evaluate the film processability and physical properties of the obtained films.

Development of soluble LCP

As previously mentioned, LCP has a tendency to be easily oriented; thus, problems such as the anisotropy of the film in the vertical and horizontal directions during melt-casting and the surface layer and interlayer delamination caused by shear forces during film formation need to be resolved. Therefore, our company innovatively developed a solvent-soluble LCP, considering that the solvent casting method used to prepare polycarbonate and polyamide films could be one such solution to the aforementioned problems. Solubilization of LCP in solvents has been previously considered, but the reported methods require dissolution in expensive fluorinated phenol solvents, making practical application difficult. However, fully utilizing the copolymerization technology of our company has enabled us to successfully develop an LCP that is soluble in general-purpose solvents. Casting a solution of this soluble LCP onto a support and drying the solvent at a low temperature of approximately 100–150 °C results in the LCP taking on an amorphous (non-crystalline) state and becoming a transparent film. Further heat treatment at a high temperature of at least 250 °C orients the liquid crystal, thereby resulting in a semi-transparent to opaque film with no anisotropy (**Fig. 7**). The LCP film obtained via the above process has excellent dielectric properties, solder heat resistance, and thermal conductivity, which enables its application to flexible printed circuit boards (FPCs) and facilitates its development into heat dissipation substrates and rigid substrates via pre-pregs impregnated with glass cloth and other materials by utilizing the fact that the LCP film can be highly filled with filler by dissolving it in solution3)–7).

High performance of soluble LCP

As previously mentioned, our company has successfully solubilized LCP and developed cast films, and has already started deploying the results in specific applications, such as speaker diaphragms, FPCs, and heat dissipation substrates. This technology reflects the present circumstances, in which material development for high-frequency communication substrates is actively underway in conjunction with the spread of next-generation high-speed communication technology. As part of these efforts, our company is striving to further improve the functionality of soluble LCP.

The global spread of next-generation high-speed communication faces challenges such as delays in the development of millimeter wave infrastructure, a lack of services and use cases, and difficulties in area expansion. However, high-speed, large-capacity communication using the millimeter wave band offers a wide bandwidth that enables a large amount of data to be transmitted at once, and substrate materials with low dielectric dissipation factors play a pivotal role in the development of hardware that can achieve such communication. To this end, soluble LCP is believed to be a promising material that meets existing requirements.

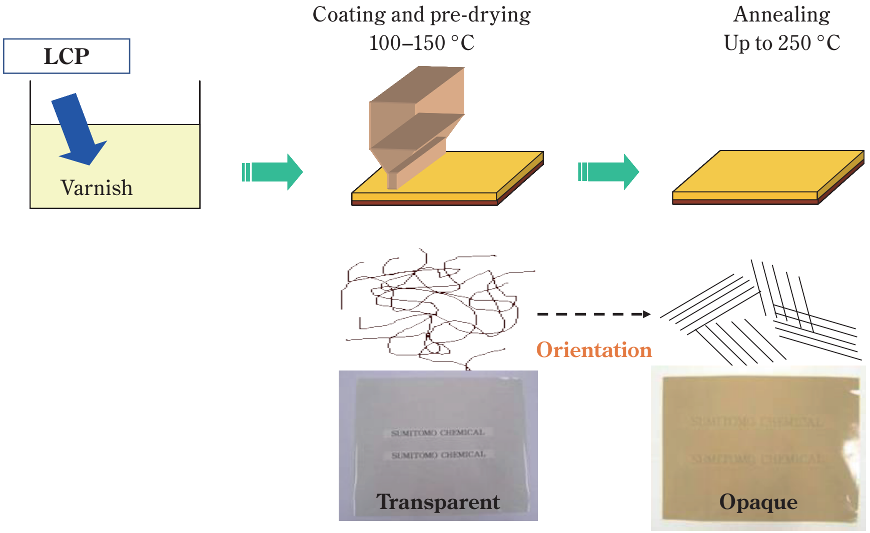


Fig. 7 Production process of film using soluble LCP

The soluble LCP developed by our company has stable dielectric properties up to the high-frequency range and properties such as high heat resistance, easy anisotropy control, and excellent conductor adhesion. We have already reported an application of this technology to high-frequency circuit boards in three-layer films using a solvent-cast polyimide film[8].

In the present report, we focus on the high functionality of the soluble LCPs that our company is currently developing. In particular, we aimed to improve the dielectric properties of the soluble LCP by adopting a new approach that utilizes the properties that soluble LCPs can handle in a solution state. Specifically, we attempted to apply a new LCP micropowder with a low dielectric dissipation factor to a solution of LCP that can be dissolved in the general-purpose solvent developed independently by our company. The performance evaluation results of the hybrid material that we obtained through this process at high frequencies are detailed below.

Fig. 8 shows the dielectric dissipation factor of the LCP used in the micropowder at 10 GHz.

Orientation polarization is a primary factor that impacts the dielectric dissipation factor in the GHz band. In this LCP, a structure that fixes the polarized part in the molecule and suppresses molecular motion was adopted in the molecular design. We successfully reduced the dielectric dissipation factor to 0.001, compared to 0.004 for general LCP, as a result of our approach to reducing the dielectric dispassion factor.

Moreover, we adopted a method called jet mill grinding to turn the LCP into micropowder. Jet mill grinding is a technology that uses the energy of gas jetted at high speed to reduce the size of materials. It enables the grinding of materials to a uniform and fine particle size, and allows the efficient production of micropowder (average particle size 10–20 μm) even from LCP, which is generally difficult to grind (**Fig. 9**)[9].

Table 2 lists the results of comparing the dielectric properties of hybrid LCP and modified polyimide to

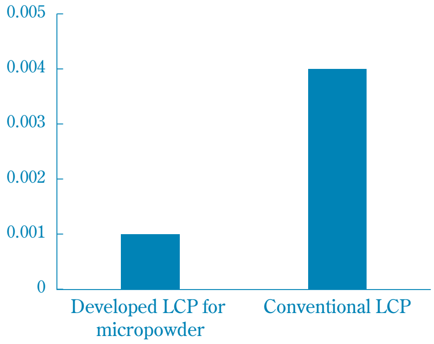


Fig. 8 Dielectric dissipation factor of developed LCP for micropowder at 10 GHz

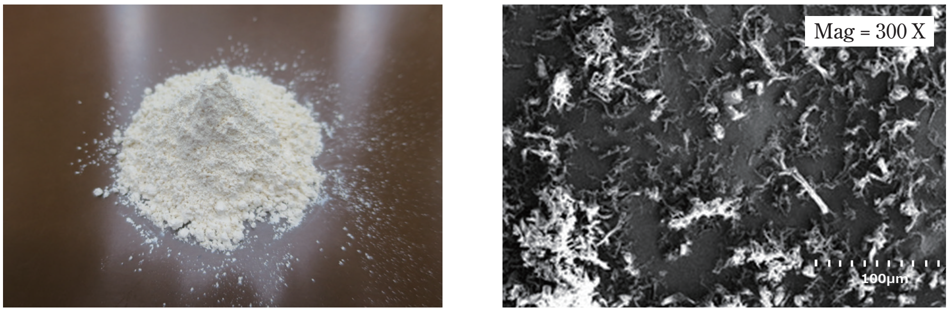


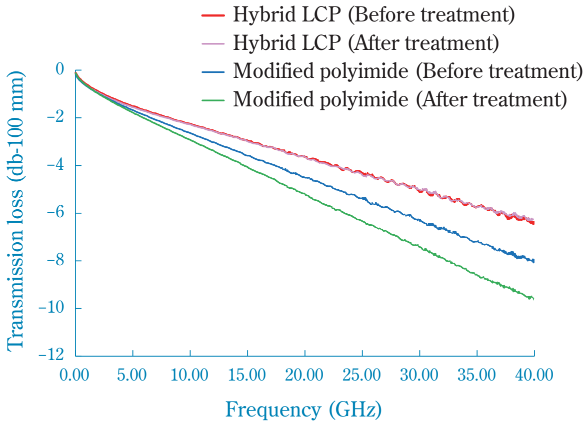
Fig. 9 Appearance (left side) and scanning electron microscope images of micropowder (right side)

Table 2 Comparison of environmental condition dependence of dielectric properties between hybrid LCP and modified polyimide at 10 GHz (cylindrical cavity resonator method)

	Pre-conditions	Hybrid LCP (Soluble LCP and developed LCP micropowder)	Modified polyimide
Relative permittivity (*Dk*)	5 h at 120 °C	3.5	3.6
	72 h at 25 °C and 50%RH	3.5	3.7
	48 h at 23 °C in water	3.5	3.7
Dielectric dissipation factor (*Df*)	5 h at 120 °C	0.0016	0.0009
	72 h at 25 °C and 50%RH	0.0017	0.0032
	48 h at 23 °C in water	0.0021	0.0064

which micropowder has been applied. Modified polyimide exhibits excellent properties in an absolutely dry state. However, the dielectric dissipation factor of modified polyimide was found to greatly increase under the standard environmental conditions of 25 °C and 50% relative humidity and water immersion conditions. This is believed to be caused by the polar groups derived from the imide skeleton in the polyimide that absorbs water by forming hydrogen bonds with water molecules. Meanwhile, hybrid LCP has a low water absorption rate, dielectric properties that are almost independent of environmental conditions, and good dielectric properties even under high humidity conditions.

Circuit boards were fabricated using double-sided copper-clad laminates made of modified polyimide and hybrid LCP, and the transmission loss was simultaneously compared and evaluated. **Fig. 10** shows the transmission loss of each circuit board material in the frequency range of 1–40 GHz. The hybrid LCP that uses micropowder exhibited significantly lower transmission loss than modified polyimide substrate materials. Measurements at 40 GHz exhibited transmission loss that was approximately 2.5% lower than that of modified polyimide materials. Furthermore, we investigated the transmission loss after immersion in water at 23 °C for 48 h. The modified polyimide was impacted by water absorption, with the transmission loss increasing remarkably. Meanwhile, the hybrid LCP hardly exhibited any change and maintained stable transmission characteristics.



Measurement equipment: Network analyzer E8363B (Keysight Technologies, Inc.)
Measured frequency range: 1–40 GHz
Measurement condition: C-24/23/50
Water absorption treatment condition: D-23/48
Insulator thickness: 50 μm
Copper foil thickness: 12 μm
Pattern length: 100 mm
Mode: Differential mode

Fig. 10 Comparison of transmission loss between hybrid LCP and modified polyimide

This result indicates that the hybrid LCP exhibits excellent resistance to humidity fluctuations, supporting its applicability as a highly reliable circuit board material.

This technology is anticipated to enhance the performance of next-generation high-speed communication boards and significantly contribute to the expansion of high-speed communication applications.

Injection molding grade for next-generation high-speed communication

Injection molding applications other than the above-mentioned film applications require low-dielectric constant materials for parts used in next-generation high-speed communication to suppress the attenuation of electrical signals, and applications to high-speed communication connectors are being particularly considered. LCP has excellent heat resistance, mechanical strength, electrical properties, and dimensional accuracy apart from high fluidity, enabling the use of injection molding to mold fine products. Moreover, LCP has low water absorption, enabling it to maintain stable dielectric properties even in changing environments. This is because water has a very high dielectric constant of approximately 80 and a dielectric dissipation factor of approximately 0.3, which results in significant changes in the dielectric constant and dielectric dissipation factor if the resin absorbs even a small amount of water. In this respect, LCP is also advantageous as a material for high-speed communication connectors.

Our company launched and sold SUMIKASUPER E6205L and SUMIKASUPER SR1205L as low-dielectric constant grades that utilize these LCP characteristics (**Table 3**).

In particular, SR1205L achieves improved dielectric properties by applying the uniquely developed LCP resin of our company, which has a low dielectric dissipation factor and smaller changes in dielectric constant and dielectric dissipation factor with respect to frequency and temperature compared to our standard grade E6807LHF (**Fig. 11**).

Furthermore, only low-dielectric constant materials have attracted attention to date, but as products become smaller and circuits become denser, impedance

Table 3 Dielectric properties of SR1205L and E6205L at 1.0 and 10 GHz

		Low relative permittivity (*Dk*) & low dielectric dissipation factor (*Df*)	Low relative permittivity (*Dk*)	Standard
		SR1205L	E6205L	E6808LHF Z
Relative permittivity (*Dk*)	1.0 GHz*1	2.8	2.9	3.6
	10 GHz*2	2.7	2.8	3.7
Dielectric dissipation factor (*Df*)	1.0 GHz*1	0.002	0.006	0.004
	10 GHz*2	0.003	0.005	0.005

*1: Measurement along *Z*-axis by the capacitance method using an impedance analyzer.
*2: Measurement along TD direction by cylindrical empty cavity resonator method which is compliant with IEC 62810.

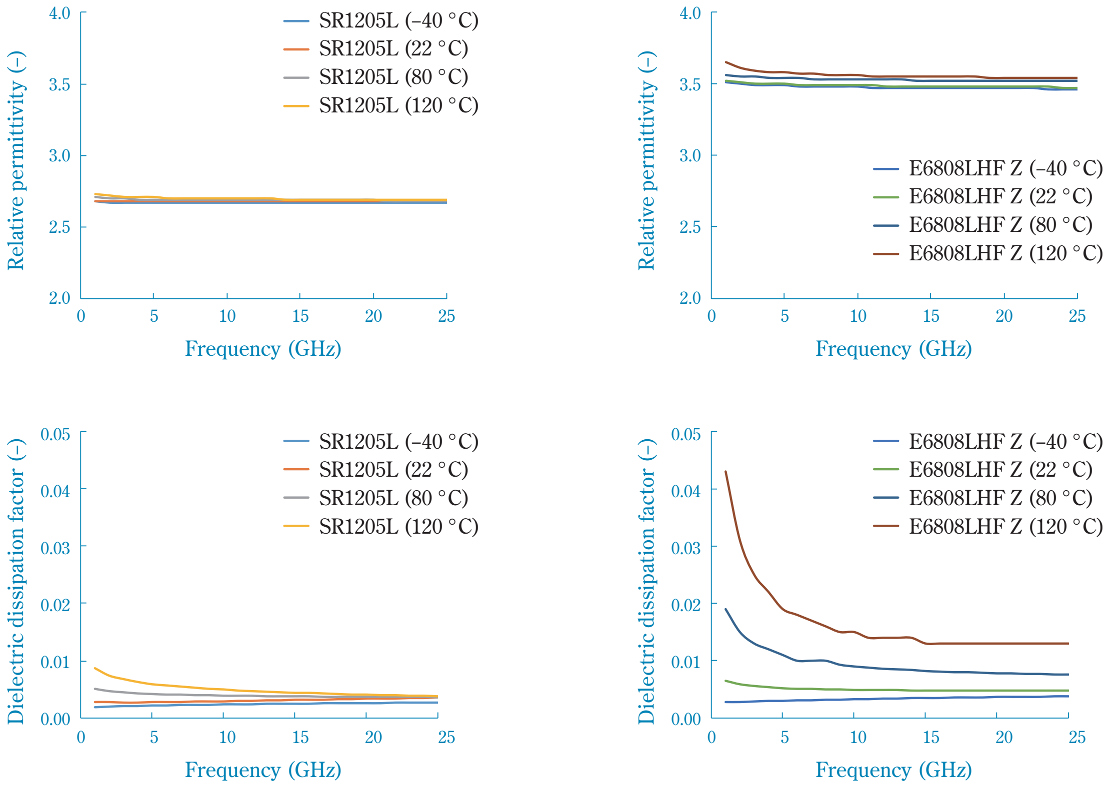


Fig. 11 Temperature and frequency dependence of relative permittivity and dielectric dissipation factor measured by triplate-line resonator

matching between contacts and insulators (insulating bodies) is also expected to become crucial in the future. Previous designs achieved impedance matching by adjusting the effective dielectric constant by making adjustments in the insulator structure, but smaller products with denser circuits can make it more difficult to secure space within the insulator for impedance matching10). This is thought to lead to more opportunities to select resin materials with appropriate impedance for the resin material to be used as an insulator, and materials with various dielectric constant values will be required for adjusting impedance. Therefore, our company is currently developing the SUMIKASUPER SZ6920 series, which includes low-dielectric constant materials and resin materials with a wide range of dielectric constants (**Table 4**). Additionally, we are also developing grades with a wide range of dielectric constants in the SUMIKASUPER SR1920 series, which uses an LCP with a low dielectric dissipation factor (**Table 5**).

We believe that the use of these grades will increase the freedom of product design and enable the optimization of transmission characteristics by simply changing the insulator material with the same product shape, and we are proposing these options to customers.

Table 4 Dielectric properties of the SZ6920 series (grades with controlled relative permittivity) at 1.0 and 10 GHz

		Relative permittivity (*Dk*) controlled				Low relative permittivity (*Df*)	Standard
		SZ6920				E6205L	E6808LHF Z
		RP25	RP30	RP40	RP50		
Relative permittivity (*Dk*)	1.0 GHz*1	2.5	3.0	3.8	4.9	2.9	3.6
	10 GHz*2	2.4	3.0	3.9	5.1	2.8	3.7
Dielectric dissipation factor (*Df*)	1.0 GHz*1	0.003	0.004	0.005	0.005	0.006	0.004
	10 GHz*2	0.004	0.004	0.003	0.003	0.005	0.005

*1: Measurement along *Z*-axis by the capacitance method using an impedance analyzer.
*2: Measurement along TD direction by cylindrical cavity resonator method which is compliant with IEC 62810.

Table 5 Dielectric properties of the SR1920 series (low dielectric dissipation factor grades with controlled relative permittivity) at 1.0 and 10 GHz

		Relative permittivity (*Dk*) controlled & low dielectric dissipation factor (*Df*)				Low relative permittivity (*Dk*) & low dielectric dissipation factor (*Df*)	Standard
		SR1920				SR1205L	E6808LHF Z
		RP25	RP30	RP40	RP50		
Relative permittivity (*Dk*)	1.0 GHz*1	2.5	3.0	3.8	4.8	2.8	3.6
	10 GHz*2	2.4	3.0	3.9	5.1	2.7	3.7
Dielectric dissipation factor (*Df*)	1.0 GHz*1	0.002	0.002	0.001	0.001	0.002	0.004
	10 GHz*2	0.003	0.002	0.001	0.001	0.003	0.005

*1: Measurement along *Z*-axis by the capacitance method using an impedance analyzer.
*2: Measurement along TD direction by cylindrical cavity resonator method which is compliant with IEC 62810.

Summary

The high-orientation properties of LCP result in the material being easily oriented in one direction, and it is generally believed that manufacturing a biaxially stretched film using this material is extremely difficult. However, LCP that is soluble in solvents by controlling the molecular structure has been proposed, and we have sought a solution to the above problem by establishing an extrusion molding technology that combines solvent casting-based film formation technology and precision film formation technology with precision control technology of the microstructure. Furthermore, for injection molding applications, we are developing materials that contribute to greater freedom in circuit design and improved transmission characteristics by offering a variety of resin materials with a wide range of dielectric constants while utilizing the high fluidity and low water absorption properties of LCP.

Our efforts are aimed at overcoming the difficulty of processing high performing LCPs by combining a variety of technologies, and we intend to continue development so that it will become a material that can be widely used in commercial applications.

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