1. Introduction

In recent years, the massive growth of communication traffic has prompted the development of next-generation communication technology, such as fifth-generation (5G) systems. Realization of 5G enables large files to be transmitted via the internet through multiple simultaneous connections and low latency communications. Against the background of increasing communication traffic, the future progress of the Internet of Things was predicted\(^1\). In this respect, advances in 5G communication systems are of particular importance. To construct a communication system according to 5G standards, microwave and millimeter wave bands have been used in an attempt to improve the speed of wireless communications; however, this process has been limited by transmission loss of radio waves in the atmosphere\(^2\). Advances in antenna technology are necessary to achieve effective transmission, i.e., the sending and receiving of radio waves without disturbances in communications.

The phased array antenna is promising as a directional antenna, due to its beamforming functionality. Figure 1 shows a schematic diagram of the phased array antenna\(^3,4\).

\[ \phi = d \sin \theta \]

Fig. 1 Structure of phased array antenna.
where \( d \) represents the distance between each antenna element. Based on the operating principle, the phased array antenna has the features of multi-beam radiation and beam scanning in any direction via phase control. Because the transmission loss in the atmosphere can be compensated for by directional control of the synthesized waves, the phased array antenna is suitable for radio-frequency bands and 5G communication applications.

A conventional phase shift method based on changes in the electrical length of the phase shifter has been reported\(^5\). However, arbitrary control of electrical length is difficult with conventional methods, owing to the structure of the phase shifter. In this method, directional control of the synthesized waves becomes discrete. This drawback complicates the application of 5G systems to mobile devices, which are subject to positional changes. In addition, diode switching to control the electrical length induces high dielectric losses for microwaves and millimeter waves. To overcome this issue, phase shifters exploiting the alignment changes of a nematic-phase liquid crystal (LC) have been examined in several studies\(^6\)-\(^8\). The LC molecules have optical anisotropy, and their molecular alignment can be controlled freely to within several kilohertz using a low-frequency alternating current (AC) voltage. Therefore, control of the externally applied AC voltage enables simple and continuous beam steering in any direction. In addition, the phase shifter using an LC has the potential advantage of a smaller size than that of conventional devices. The amount of phase shift (\( \phi \)) is given by equation (2) \(^9\):

\[
\phi = 2\pi df\sqrt{\Delta\varepsilon}/c \tag{2}
\]

where \( f \) is the frequency of the radio wave, \( \Delta\varepsilon \) is the difference in dielectric constant of the LC, and \( c \) is the speed of light in vacuum. This equation states that alignment changes of LC molecules enable continuous changes in phase. However, dielectric losses in LC materials for radio waves strongly depend on the type of LC material. Several studies have investigated the dielectric loss of various LC materials. Of these, a small number of LC molecules have low dielectric losses\(^10\). The optimal structural design of LC molecules for suppression of dielectric loss in radio waves remains unclear. An understanding of the dielectric loss mechanism in LCs is expected to accelerate the development of efficient LC phase shifters.

We have been researching on evaluation of high-frequency dielectric behaviors in LC materials\(^11\). In this paper, we focused on the basic molecular structure of nematic-phase LC molecules and evaluated the relationship between dielectric loss and the fundamental skeleton of LC molecules. The nematic-phase LC material is commonly composed of three skeletons: the core part, the hydrocarbon group (alkyl chain), and the polar group; these three components determine the optical characteristics of the LC and the molecular interactions among LC molecules. We discuss the relationships among these three structural components of the LC and evaluated dielectric loss using the coaxial line method.

### 2. Evaluation method of dielectric loss

The parameter \( \tan\delta \) (loss tangent) is used for quantitative evaluation of dielectric loss, where \( \tan\delta \) is calculated based on the complex permittivity, as follows (\( j \): Fimaginary unit):

\[
\epsilon_{r2} = \epsilon'_{r2} - j\epsilon''_{r2} \tag{3}
\]

where\(^12\):

\[
\tan\delta = \frac{\epsilon''_{r2}}{\epsilon'_{r2}} . \tag{4}
\]

The small value of \( \tan\delta \) corresponds to low dielectric loss. Complex permittivity is calculated based on experimentally obtained S-parameters, as shown in the following equations (5-7) (transmission coefficient \( T \) and reflection coefficient \( \Gamma \)), and assuming a three-layer coaxial line, as shown in Fig. 2\(^13\),\(^14\):

\[
T_{13} = \frac{(1 - \Gamma_{12}^2)e^{-j2\pi f\sqrt{\varepsilon_0\mu_0}\varepsilon_{r2}}}{1 - \Gamma_{12}e^{-j4\pi f\sqrt{\varepsilon_0\mu_0}\varepsilon_{r2}}} \tag{5}
\]

\[
\Gamma = \frac{\Gamma_{12}(1 - e^{-j4\pi f\sqrt{\varepsilon_0\mu_0}\varepsilon_{r2}})}{1 - \Gamma_{12}^2e^{-j4\pi f\sqrt{\varepsilon_0\mu_0}\varepsilon_{r2}}} \tag{6}
\]

\[
\Gamma_{ij} = \frac{Z_j - Z_i}{Z_j + Z_i} = \frac{\sqrt{\mu_j/\varepsilon_j} - \sqrt{\mu_i/\varepsilon_i}}{\sqrt{\mu_j/\varepsilon_j} + \sqrt{\mu_i/\varepsilon_i}} \tag{7}
\]

We used the three-layer coaxial line to prevent liquid leakage of nematic-phase LC materials during S-parameter measurement, as shown in Fig. 2. Based on the above equations, \( \tan\delta \) can be numerically calculated by Newton’s method. The S-parameters were obtained by network analyzer experimentally. In this research, we sealed a nematic LC in the coaxial line with polytetrafluoroethylene (PTFE).

A transverse electromagnetic wave from a vector
network analyzer was incident on the LC for broadband measurements.

Figure 3 shows a cross-sectional schematic diagram of the filled coaxial line with a nematic-phase LC. The two dielectric constants of the LC in the microwave band were obtained using microwave measuring voltage (12 GHz) and a bias voltage (square wave, 1 kHz, 80 Vp-p, Duty ratio: 50%). The bias voltage was switched between the inner and outer conductors of the coaxial line, while controlling the LC alignment. When the coaxial line was in the voltage-off state, the bulk LC molecules assumed a randomly aligned state, as the LC layer was sufficiently thick (1.39 mm). We defined the calculated dielectric constant in the randomly aligned state as $\varepsilon_r(\text{av})$. With a sufficiently high voltage, the long axis of LC molecules was aligned parallel with the electric field between the inner and outer conductors. Here, we defined the calculated dielectric constant under this condition as $\varepsilon_r(||)$. The dielectric constant along the short axis $\varepsilon_r(\perp)$ of the LC molecule can be expressed as follows:

$$\varepsilon_r(\perp) = \frac{3\varepsilon_r(\text{av}) - \varepsilon_r(||)}{2} \quad (8)$$

$\tan\delta(\perp)$ can be calculated using equation (9):

$$\tan\delta(\perp) = \frac{3\varepsilon_r(\text{av}) \cdot \tan\delta(\text{av}) - \varepsilon_r(||) \cdot \tan\delta(||)}{2\varepsilon_r(\perp)} \quad (9)$$

3. Investigation of the optimal molecular structure of the LC

First, we investigated the effect of the chain length of the hydrocarbon group, $\text{C}_n\text{H}_{2n+1}$ [$n = 5, 6$ and $7$], on the dielectric loss using LC materials with a cyano biphenyl group, as these materials exhibit a nematic phase at room temperature. Table 1 summarizes the molecular structures and dielectric loss of the LC materials.

The results showed that $\tan\delta(\perp)$ was not affected by the chain length. This may be because a change in the LC chain length produces only a slight change along the long axis and no changes along the short axis, i.e., a change in the LC chain length does not affect the dielectric loss along the short axis $\tan\delta(\perp)$. In the following discussion, we focus on $\tan\delta(\perp)$ as the dielectric loss.

Next, we investigated the effect of the core structure of the LC molecule on dielectric loss in the two cases described below. The first case is of structural change from cyclohexane to a benzene ring (see the combination of compounds 4 and 1, and of compounds 5 and compound 6 in Table 2). Table 2 summarizes the molecular structure and dielectric loss of the LC materials. The structural changes from a cyclohexanering to a benzene ring reduced dielectric loss. The second case compares compound 6, which has a hydrocarbon spacer ($\text{C}_2\text{H}_4$), with compound 7, which does not. The LC without a hydrocarbon spacer showed lower dielectric loss than that of an LC with a hydrocarbon spacer. On the basis of molecular orbital theory, the structural stability of the benzene ring is higher than that of the cyclohexane ring due to the π-conjugated resonance structure. These results are attributed to the structural rigidity of LC materials. The dielectric loss may be suppressed by a more rigid molecular structure.

| Molecular Structure | No. | $\varepsilon_r$ | $\tan\delta$ |
|--------------------|-----|----------------|--------------|
| $\text{C}_5\text{H}_{11}-\text{CN}$ | 1 | 3.32 | 3.00 | 0.024 | 0.052 |
| $\text{C}_6\text{H}_{13}-\text{CN}$ | 2 | 3.07 | 2.71 | 0.030 | 0.053 |
| $\text{C}_7\text{H}_{15}-\text{CN}$ | 3 | 3.14 | 2.72 | 0.020 | 0.053 |
From the results of Table 2, it was observed that fluorine tends to reduce dielectric loss. Finally, we evaluated the effect of a terminally modified polar group (fluorocarbon system) on dielectric loss. The evaluated dielectric constant and $\tan \delta$ values of fluorinated LC are summarized in Table 3. Compound 8, which has three fluorine atoms, showed a lower dielectric loss than compound 9 having a fluorine atom and compound 10 having no fluorine atom.

The LC molecule, having a fluorine atom as a polar group, experiences the following. First, the fluorocarbon group of the LC has a large polarity due to the electronegativity being strongest for the fluorine atom. For this reason, the C-F bonding energy is relatively high. Second, the rotation of LC molecules is suppressed by steric interactions between adjacent fluorocarbons. Second, fluorinated LCs are less prone to electric field effects, because these LCs have a small polarization. Therefore, we considered that the fluorocarbon group has a role in suppressing molecular vibration loss at radio frequencies.

Based on the above experimental results, a molecular structural design that suppresses molecular vibration, as shown in Fig. 4, is necessary to suppress dielectric loss. By the comparison of core part, we considered that rigid core part of LC suppressed curvature movement of LC molecules as shown in Fig. 4 (a). As the number of benzene rings in core part of LC increase, the LC molecules are more likely to stack. And, vertical translation of molecules is also suppressed, as shown in Fig. 4(b). By the comparison of structure of polar group, we considered that rotation of LC molecule as shown in Fig. 4(c) is suppressed by fluorine.

### Table 2. Dielectric loss of LCs with different core part and polar group structures (12 GHz).

| Molecular Structure | No. | $\varepsilon_r$ || $\tan \delta$ || |
|--------------------|-----|----------------|-----------------|-----|
|                    |     | 3.32           | 3.00            | 0.024 | 0.052 |
| R                  | 4   | 2.53           | 2.44            | 0.027 | 0.070 |
| R                 | 5   | 2.62           | 2.42            | 0.016 | 0.041 |
| R                 | 6   | 2.81           | 2.40            | 0.016 | 0.034 |
| R                 | 7   | 2.67           | 2.35            | 0.016 | 0.028 |

### Table 3. Dielectric loss of an LC having a different number of fluorines as the polar group (12 GHz).

| Molecular Structure | No. | $\varepsilon_r$ || $\tan \delta$ || |
|--------------------|-----|----------------|-----------------|-----|
| R                  | 8   | 3.33           | 2.86            | 0.030 | 0.062 |
| R                  | 9   | 3.43           | 2.61            | 0.029 | 0.062 |
| R                 | 10  | 3.23           | 2.68            | 0.037 | 0.083 |

### 4. Temperature dependence of dielectric loss in the LC

As mentioned above, various structural factors involved in dielectric loss have been indicated. In the case of a solid-state polymer, there are several such factors: rotation of the molecular dipole, crystalline lattice vibration, and the vibration of molecular chains in the amorphous region. These factors are associated with molecular vibration. The strength of molecular vibration is closely related to the environmental temperature.

To clarify the loss factors in a nematic LC, we investigated the temperature dependence of $\tan \delta$. Figure 5 shows the temperature characteristics of $\tan \delta$. The inset equations correspond to a linear approximation of the experimental results.
in compounds 1 and 7. Here we compared compound 1, having a cyano group as the typical LC material, with LC material compound 7, having fluorocarbon, which showed the lowest dielectric loss in this study. The dielectric loss increased with temperature, independent of the LC material. Because dielectric loss is related to temperature, we considered molecular vibration to be the main factor contributing to dielectric loss.

Phase transition points of Compound 1 and 7 are 35 °C and 45.5 °C. In Fig. 4, dielectric losses of LC are not change at phase transition points. In this case, bias voltage was not applied and LC's direction was random. We considered the reason is that directions of LC molecules in nematic LC phase and isotropic phase are random and equal.

5. Conclusions

In summary, we investigated the structural factors of nematic-phase LCs associated with dielectric loss at radio frequencies. As the results, we clarified that dielectric loss is affected by the rigidity of the core part and terminally modified polar groups in the nematic LCs. Also, the environmental temperature of nematic-phase LC materials contributed to a reduction in dielectric loss at radio frequencies. For further reduction of the dielectric loss, control of the molecular vibration of LCs will be important.

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