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# Newly Developed Innovative LCP Resins Applied into LED Usages

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Recently Sumitomo Chemical Co., Ltd. has developed two types of innovative liquid-crystalline polymer resins (abbreviated to LCP) for use in the area of high-power LED applications. One is a novel catalyst-polymerized LCP and the other one is our original soluble LCP. Catalyst-polymerized LCP has been applied to high-power LED package housing materials by making the best use of the resin whiteness and heat resistance. Some commercial compound grades mainly composed of catalyst-polymerized resin have already been accepted by customers in LED applications such as TVs, lighting and pocket projectors.

On the other hand, by succeeding in the solubilization of LCP and the development of cast films, Sumitomo Chemical has made applications in substrates incorporated into components which use films with a high level loading of inorganic filler compared to extrusion and tubular film processes. By using original techniques, we have developed super high thermal conductive circuit boards for use in high-power LED applications such as BLU and lighting to extend LED lifetimes. In the present report we introduce the latest situation.

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## Introduction

It has been quite a while since we began debating how we could contribute to the sustainable growth of humankind with global environmental perspectives after the era during which we mainly pursued convenience in daily living. In Green and Sustainable Chemistry (GSC), which can be considered the symbol of this trend, the roadmap to sustainable society is examined from the perspective of future challenges under the four major themes of energy, resources, environment and lifestyle. The significant reduction of CO<sub>2</sub> discharge goal set forth by the Japanese government (the 2020 discharge goal has been set as a 25% reduction from the 1990 discharge goal) is also a clear indicator of the future prospects for electronics and packaging materials.<sup>1)</sup> One of the areas of which sustainable growth can be expected is LED (Light Emitting Diode) devices. An LED is a diode created by binding the p- and n-type semiconductors. The GaN-based blue LED was developed in 1993 and the white LED, which is suitable for general lighting fixtures, emerged in 1996. Hence, the

scope of LED application has expanded. Special features of LEDs include longevity, low power consumption, low UV ray and no infrared emission. By the time LEDs meet the eyes of consumers, they have to go through various processing steps such as LED chip processing (the radiation source), LED chip packaging and LED package modularization. After going through these steps, the LED modules are finally manufactured into lighting fixtures. Sumitomo Chemical has already contributed to the provision of the materials required for each of the above steps in many different forms, including high-purity alumina and high-purity organic metal compound MO (TMG (trimethylgallium) and TMI (trimethylindium)) used as substrate materials, as well as process materials such as polymethyl methacrylic acid (PMMA) resin for light-guiding plates.<sup>2)</sup>

The range of application has expanded from the initial application for lighting décor and switches through LCD backlight units (BLUs) to the general-purpose lighting fixtures and vehicle interior lighting. From this point forward, LEDs with high output, or so-called high-power LEDs, will be the indispensable development-factor tech-

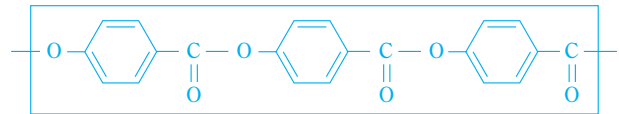
nology for BLUs with greater brightness, general-purpose lighting fixtures and vehicle interior lighting. Therefore, the requirements of the materials used in such applications will differ from those required for low-power LEDs, thus demanding technological innovation in materials as well. More specifically, the significant improvement in the luminous efficiency and flux of light will enhance the large volume current, thereby serving to dramatically increase the contribution of peripheral materials to the LED's lifetime. In fact, it is urgent that we improve both the heat resistance of resins used for package housing and the heat-conductivity of packaging materials.

Currently, Sumitomo Chemical is proactively tackling such challenges through its molecular design for LCPs (Liquid Crystal Polymers), as the polymerization and compound technologies by which successful commercialization has been achieved since 1979. Specifically, the company has begun applying its proprietary LCP-related technologies, which have been released one after another since 2002, and the new technologies (including resin whitening using the newly developed catalyst process and solubilization of LCPs) to LED package housings and ultra-heat dissipating substrates for LED modules. This paper will introduce the related factor technologies and some of the applications achieved through the use of such technologies.

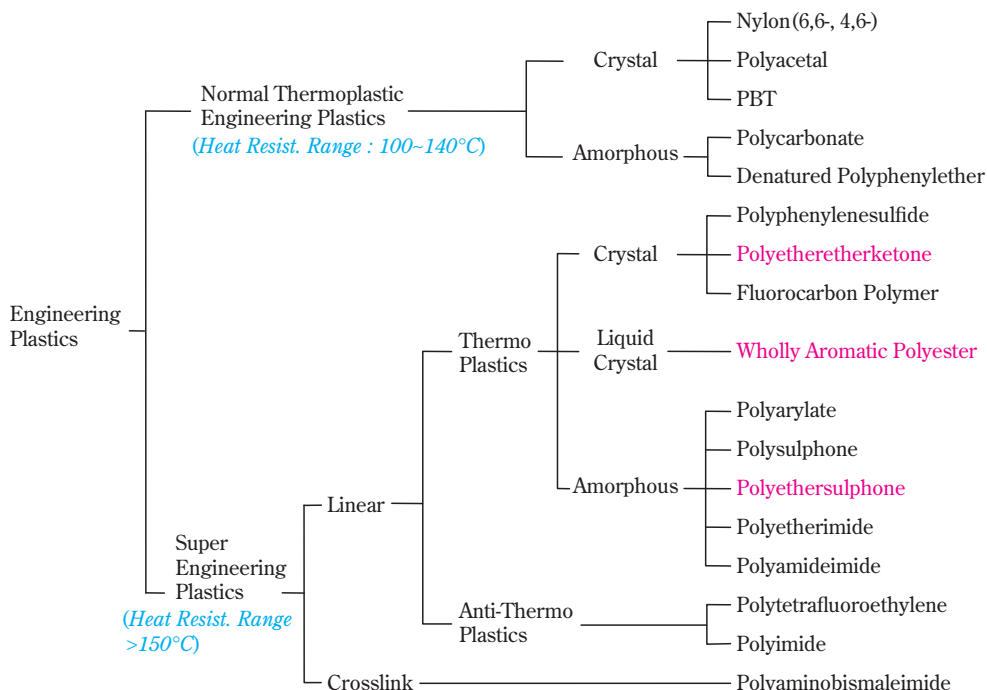
## Molecular Design of Liquid Crystal Polymer (LCP) and Its Polymerization Technology

### 1. Molecular Design of LCP

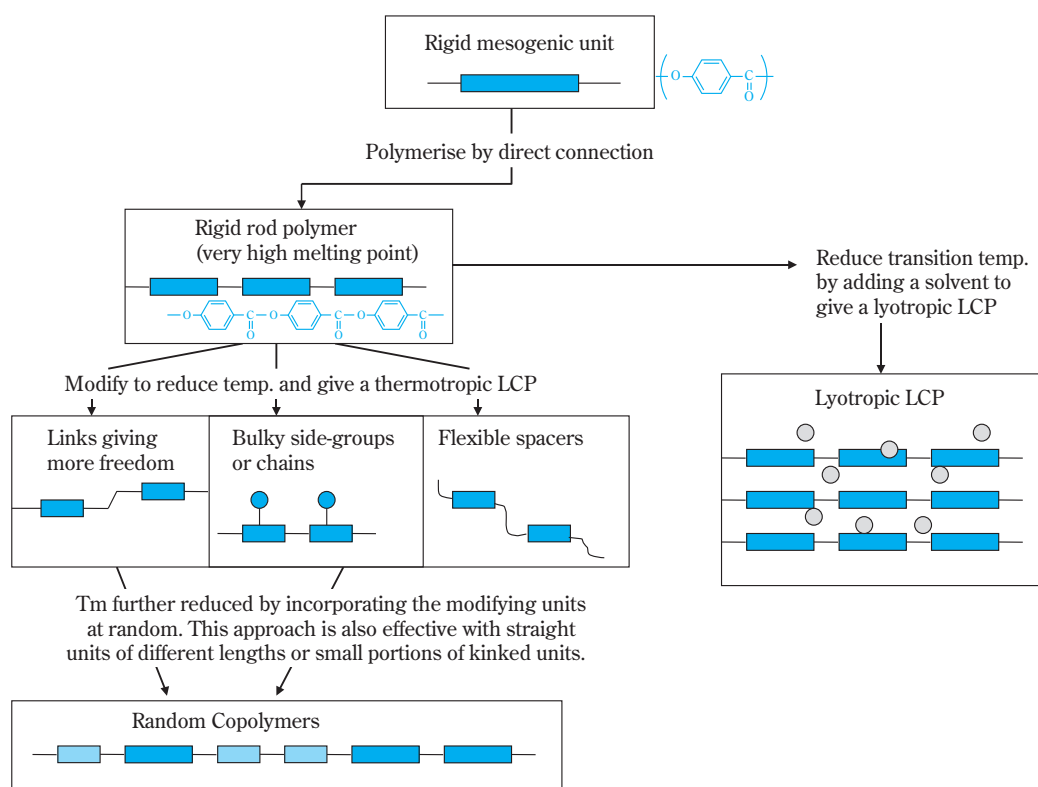
As shown in Fig. 1, while most of the resins are named after their chemical structures (such as polyethylene), the name of liquid crystal polymer alone indicates its thermodynamic status as a polymer in the liquid crystalline state. Its chemical structure comprises fully aromatic co-polyesters, which in turn contain the liquid crystal formation trigger (mesogen) having POB (parahydroxy benzoic acid) as the main component in its principal chain, as shown in Fig. 2. In order to indicate the liquid crystal state, it is necessary that molecules have either a long and slender stick-like shape or a flat plate-like shape, and that they have the appropriate intermolecular force in order to maintain the parallel orientation of molecules. When the melting point of the polymer is higher than the thermal decomposition temperature, melt processing cannot be performed. Because the melting point of POB homopolymer can be



**Fig. 2** Chemical structure of liquid crystalline state generator



**Fig. 1** Standard classification of engineering plastics



**Fig. 3** Schematic description of conventional design of liquid-crystalline polymer

extremely high, exceeding 500°C, the following methods are undertaken in order to lower the melting point so that the melt processing can be performed: partially introducing a soft aliphatic chain such as ethylene glycol; copolymerizing irregular components such as biphenol; copolymerizing a small amount of a component with low symmetry; and introducing substituents into the aromatic compounds (Fig. 3).<sup>3)</sup>

LCP products with various structures are currently on the market in addition to the LCP products classified by the heat resistance (deflection temperature under load) of type I to III, as determined by a U.S. survey company (Table 1). The LCP market is expected to reach 40,000

tons in 2010, mainly for the application in electronic components obtained through high-precision molding such as connectors and relays. It is predicted that as the trend of miniaturization intensifies in the future, the LCP market will, for a while, continue to expand at the annual growth rate of more than 10% due to its outstanding heat resistance, which allows it to be compatible with the lead-free soldering process, as well as its excellent environmental suitability (i.e., being halogen-free).<sup>4)</sup>

## 2. LCP Polymerization Method

Melt polymerization is generally used in the manufacture of LCPs, which can be obtained by undertaking the

**Table 1** Classical classification of standard commercial LCP grade

Type	Chemical Struct.	HDT*	Commercial Grade
Ekonal (Type I)		300°C ~	SumikaSuper® Xydar®
Vectra (Type II)		240 ~ 280°C	Vectra®
X7G (Type III)		~ 210°C	Rodrun® Novaccurate®

\* HDT: Heat Deformation Temperature

following procedures: Acetylation is performed on a hydroxyl group—which is represented by the above POB—such as aromatic hydroxy carboxylic acid, aromatic dicarboxylic acid and aromatic diol using acetic anhydride. A hydroxyl group is then heated to near 300°C, causing the polycondensation reaction with diacetic acid and thereby producing LCPs (Fig. 4). In some cases the LCP manufactured through the melt polymerization, as described above, is used as the molding material without any further processing. However, in other occasions LCPs are used after the following processes: LCPs are extracted via melt polymerization using an oligomer with a low degree of polymerization, and the degree of polymerization is then increased through solid-phase polymerization. The LCPs obtained after these processes demonstrate various properties, including high strength/high modulus, heat resistance, dimensional stability, low flash, flame resistance, damping and gas barrier property. Due to rigid molecular chain, these LCPs show high strength/high modulus without needing to be enhanced with glass fibers. They even have the nickname “self-enhanced plastic.” The level of the specific modulus of a highly oriented, thin molded article is even equivalent to that of metals such as aluminum.<sup>5)</sup> This fact, together with high heat resistance, suggests that it is the ideal material for the substitute of metals, as materials are increasingly becoming lighter for the purpose of reducing the fuel consumption of automobiles, etc.

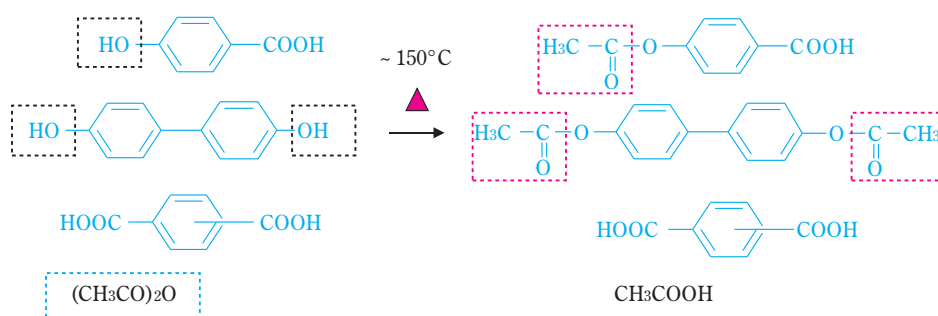
## LCP Polymerization Technology Using Catalysts

As shown in Fig. 4, the LCP polymerization process consists of two processes: the acetylation of a phenolic hydroxyl group and the diacetic acid polycondensation reaction between the acetoxy group and carboxylic acid. However, because the polycondensation occurs at the high temperature of 300°C, it was known that the procedure did not usually employ an organic catalyst, and therefore the reaction was caused without any catalyst or only by using an organic metal such as potassium acetate or sodium acetate.

Although some organic metals (including potassium acetate) have activity toward the polycondensation reaction, previously no catalyst usable for both acetylation and polycondensation reactions, which was also usable with one-pot polymerization, had been found. Responding to this situation, in October 2002 Sumitomo Chemical discovered the innovative basic organic catalyst that had activity toward both acetylation and diacetic acid polycondensation reactions.<sup>6)</sup>

Compared to the conventional LCP manufacturing method which doesn't use a catalyst, the new manufacturing method has increased the speed of acetylation and polycondensation significantly, thus greatly improving the productivity and allowing us to achieve high-quality LCP with the following outstanding properties: narrower molecular weight distribution due to the

### Acetylation Reaction



### Acidolysis Reaction Melt or Solid Phase

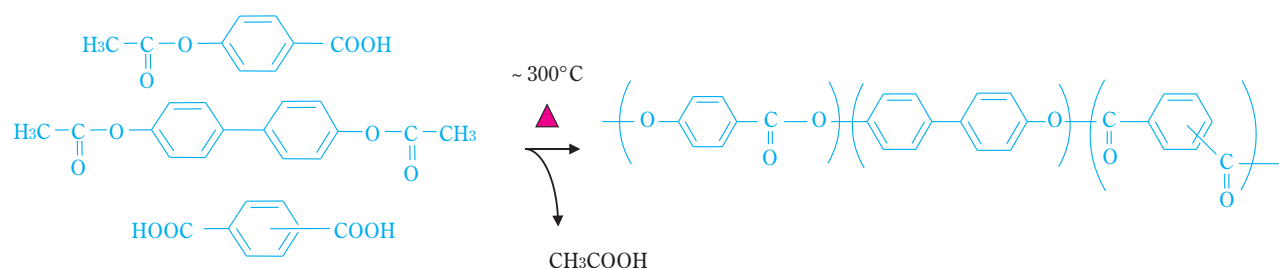
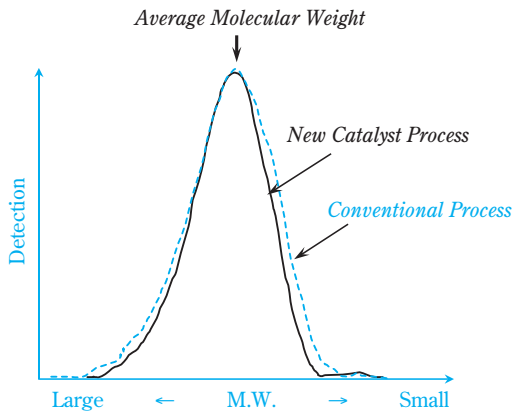
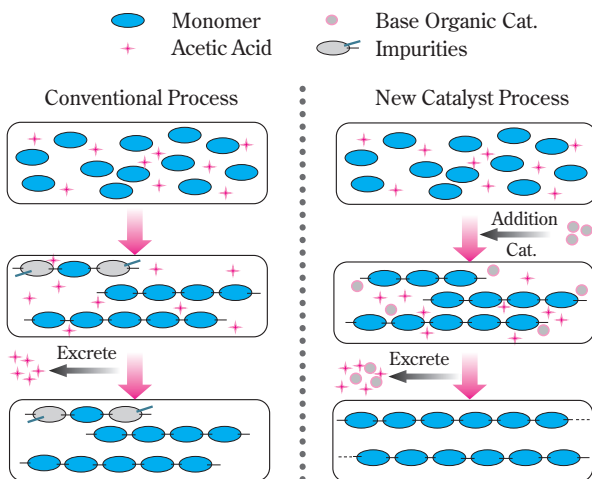


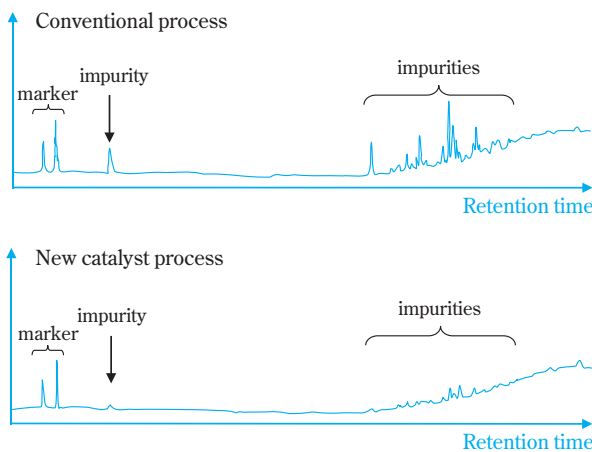
Fig. 4 Standard LCP polymerization process



**Fig. 5** Comparison of molecular weight distribution between conventional and new catalyst process



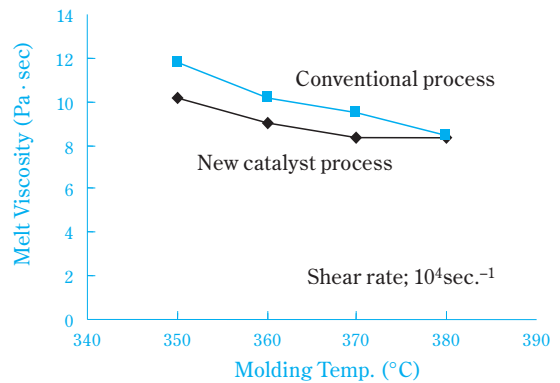
**Fig. 6** Schematic description of comparison between conventional and new catalyst process



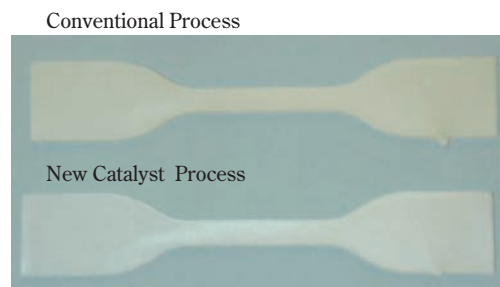
**Fig. 7** Specialized chromatography analysis for detection of impurities in LCP

improved reaction selectivity (Fig. 5); fewer side reactions and impurities (Fig. 6); and less gas evolution in the molded articles (Fig. 7).

Because this catalyst was excreted outside the system of reaction together with the byproduct of acetic acid during the temperature rising process, it was not retained in the polymer. Therefore, in the early stages we began applying the LCP obtained through this new method to major application areas, such as connectors, by making the most of its outstanding fluidity (Fig. 8) and the blister resistance shown during the soldering process.<sup>7), 8)</sup> At the same time it was discovered that the LCP color tone could be improved by controlling the side reaction of coloring during the acetylation and polycondensation reactions (Fig. 9), whereupon the examination of its application for LEDs was initiated.



**Fig. 8** Comparison of melt viscosity between conventional and new catalyst process (E6006L)



**Fig. 9** Picture of neat-resin molded dumbbells of conventional and new catalyst process

### White Grade LCP and Its Application for LED

#### 1. White Grade LCP and Its Application to LED

Although under the current circumstances, LCPs are the second leading packaging material following heat-resistant nylon, in its history the use of LCPs for LED

cases was significant because it served as a springboard when making the decision for LCP market launch.<sup>9)</sup> Although the LCP market has been taken over by nylon due to its whiteness in resins as well as its cost-effectiveness, LCPs are most popularly used for high-temperature packaging methods such as eutectic gold-tin soldering, which is undertaken as a lead-free measure and a solution for the environmental problem. The expectation regarding LCPs is currently growing along with improvement in the brightness and output power of LEDs, because they demonstrate less discoloration due to heat as compared to nylon. Under such circumstances the type-I LCPs such as Sumitomo Chemical's SUMIKASUPER<sup>®</sup> LCP (which can be clearly differentiated from 6T or 9T nylon in terms of heat resistance) are garnering attention (Table 1).

Conventionally, discoloration of the resin caused by the side reaction was occasionally seen when using LCP due to the fact that the polymerization process was performed at a temperature exceeding 300°C. For this reason, in the LED application whiteness was enhanced using titanium oxide or other similar chemical compounds. However, the problem was that it had less initial reflectivity than nylon did. In order to solve the problem, Sumitomo Chemical has adopted a polymerization method that uses the aforementioned basic organic catalyst, thus reducing the side reactions, whitening resins (Fig. 9), and significantly improving the reflectivity and productivity of the molded article.

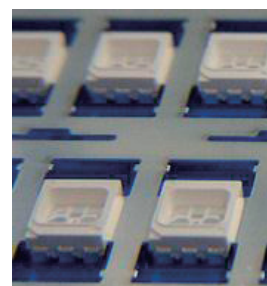
Sumitomo Chemical, in order to take back the LED package housing market which was once taken over by nylon, is proactively expanding LCP sales armored by resins with high reflectivity for LED package housing through the use of LCPs (as obtained through the new catalyst process) for its base, its compound technology and the productivity that can accommodate the rapidly emerging LED market (neat resin: 9,200 ton/year). Also, making the most of its outstanding whiteness, heat resistance and fluidity, which can be achieved through

the new catalyst process, Sumitomo Chemical enhances the application of special-grade LCPs not only for the area of packaging but also for the area of connectors used in lighting fixture to connect one board to another, such as inverters, which require high reflectivity.

## 2. Compound Grades for LED Package

In February of last year, Sumitomo Chemical developed and launched the following new LCP grades (Table 2) as the high-brightness grades of SUMIKA SUPER<sup>®</sup>: the ultra-high heat-resistant standard grade SZ4709, with reflectivity of 82% at a wavelength of 460nm; and the highly heat-resistant white grade SZ6709, with reflectivity of 88%. These new grades are used not only by domestic LED manufacturers but also by those in overseas countries such as Taiwan and Korea for use in pocket projectors, liquid crystal TVs and high-power LEDs (Fig. 10 shows one example). Both grades are SMT (surface mount technology) compatible and have heat resistance that can be used for eutectic gold-tin soldering as a solution for the environmental problem.

Moreover, because whitening is achievable with a minimal amount of titanium oxide, it has outstanding Izod impact strength and tensile strength, thus enhancing the strength of LED packages and reducing cracks during assembly.



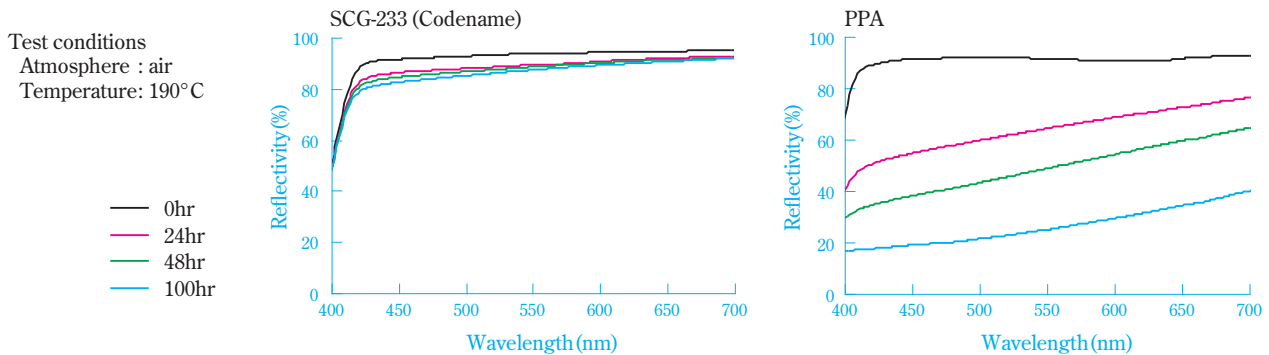
**Fig. 10** Picture of LED packages molded by commercial grade

**Table 2** Sumitomo's commercial and R&D LED grades

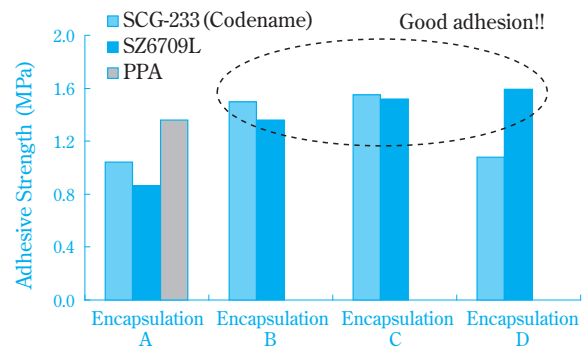
Grade		Reflectivity @460nm	Heat performance	Applications
SZ4709	For AuSn soldering and SMT use	82%	Highest	Pocket projector Lighting
SZ6709L	For AuSn soldering and SMT use	88%	Higher	TV Lighting
SCG-233 (Codename)	For SMT use	92%	High	Under evaluation at domestic and overseas LED manufactures

**Table 3** Summary of physical properties for LED package housing grade including R&D grade

			For AuSn solder use	For SMT use	For SMT use	Others	
			SZ4709	SZ6709L	SCG-233 (Codename)	LCP-D	
	Test method	Unit					
Reflectivity	640nm	JIS K7105-1981	%	91	92	95	93
	520nm			87	90	93	91
	460nm			82	88	92	89
Specific gravity	ASTM D792	-	1.89	1.89	1.99	2.19	
Mold shrinkage	MD	Sumitomo Chemical Method	%	0.15	0.17	0.32	0.23
	TD			1.05	0.80	0.68	0.63
Izod impact strength	ASTM D256	J/m	400	310	220	110	
Tensile strength	ASTM D638	MPa	120	115	100	85	
Elongation at break	ASTM D638	%	6.0	5.0	3.5	2.8	
Flexural strength	ASTM D790	MPa	140	140	133	110	
Flexural modulus	ASTM D790	GPa	12.5	11.0	10.3	13.1	
DTUL	1.82MPa	ASTM D648	°C	305	265	234	235
	0.45MPa			332	290	273	-
Soldering resistance	Sumitomo Chemical Method	°C	320	300	265	240	

**Fig. 11** Reflectivity after thermal aging of SCG-233 (Codename) compared with PPA

Furthermore, Sumitomo Chemical has already succeeded in the development of the highly white grade with reflectivity of 92% SCG-233 (development code), which can accommodate SMT (Table 3).<sup>11)</sup> This grade does not show a significant deterioration in reflectivity after heating, but its greatest feature is that the deterioration of whiteness due to long-term thermal aging is extremely minimal even in an atmosphere having a temperature of 190°C, such that the reflectivity decreased by only seven points 100 hours after at 460nm (Fig. 11). Furthermore, it decreased by only two to three points 48 hours after the Xe lamp irradiation. As a result of measurement performed in compliance with ASTM D4541, it showed the characteristics that the neither adhesion property nor mechanical strength had any problem in actual use (Fig. 12). The new grade SCG-233 (development code) has been introduced, and sample work has already been conducted targeting PC monitors, car navigation monitors and liquid crystal TVs. It has thus received high recognition.

**Fig. 12** Adhesive strength to encapsulation resin compared with PPA

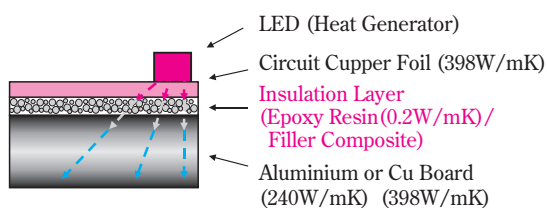
### Roles of Heat Dissipating Materials in LED

The heat source of LED is local because the radiative zone is small, thus causing the radiation density to be extremely high. Therefore, if the radiation design is inadequate it will affect its lifetime as well as the luminous efficiency. The higher the output of LED is, the

more important the radiation design will be. As a mounting material also, the heat discharge characteristic is an important prescribed property during the LED packaging and module processes.<sup>1)</sup>

The heat radiation of the LED is a mechanism by which to conduct heat through conduction, and it is a structure to eventually radiate the heat produced by an LED chip to the atmosphere via various types of materials. Metal-based heat-dissipating materials conduct heat extremely well in terms of thermal conductivity. However, an LED cannot be mounted merely with a metal-based conductive material alone. Therefore, insulating materials with high thermal conductivity are extremely important for LED packages and modules.

Heat-dissipating substrates, to which LED packages and chips have been mounted, consist of copper or aluminum metals in the form of wiring layers or heat-conductive layers. While copper and aluminum have high thermal conductivity of 398W/mK and 240W/mK, respectively, epoxy resin used for an insulating layer of the general-purpose substrate has extremely low heat-conductivity of 0.2W/mK, and it was a bottleneck during the heat-transfer process (Fig. 13). Therefore, for a high-power LED, ceramic insulating materials having heat conductivity of 20W/mK (such as alumina) are used as the substrate. However, it has been a problem that using alumina for LED packages can be extremely costly. Although its thermal conductivity is high, the heat resistance required for a substrate can be enormous due to the extreme thickness of the substrate (600 $\mu$ m), thus making the thermal conductivity of alumina insufficient.



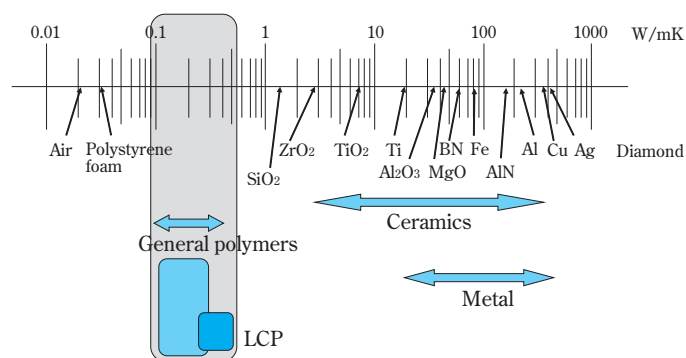
**Fig. 13** Constitution of metal-base heat dissipation circuit board

## LCP Orientation and Improving Thermal Conductivity of LCP

### 1. LCP Orientation and Improving Thermal Conductivity of LCP

Because the composite material of filler and plastic having thermal conductivity are used for thermally con-

ductive materials, it seems that fillers play a major role in thermal conduction. However, the heat resistance of a plastic material used for the matrix of such a composite material is large, and therefore the effect of the thermal conduction of plastic material toward the composite material is predominant over that of fillers. For that reason the improvement of the thermal conductivity of matrix materials has been vigorously attempted.<sup>12)</sup> It is generally known that because the resins used for the matrix do not possess free electrons, unlike metals having free electrons that are beneficial for thermal conduction, their thermal conduction is controlled by phonons. The level of thermal conduction is smaller in phonon conduction than that in electron conduction, which utilizes free electrons. Particularly, because resins have many amorphous regions that are coherent due to the Van der Waals force, the phonons responsible for thermal conduction are susceptible to dispersion, thus causing the thermal conductivity to be smaller than that of other materials (Fig. 14).



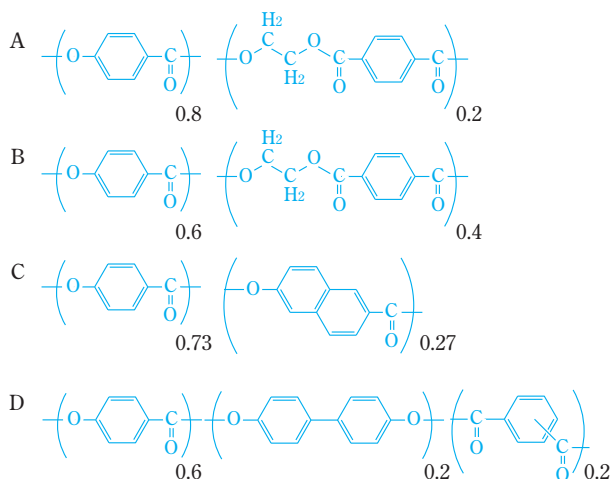
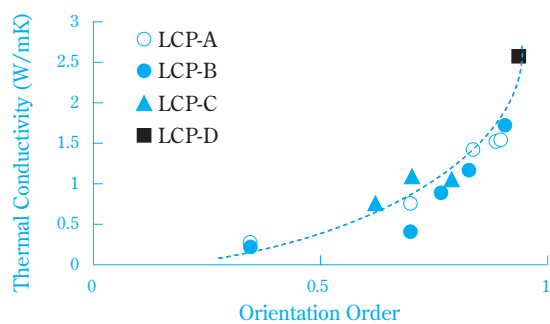
**Fig. 14** Thermal conductivity of general polymers and LCP

It can be assumed that if the resin structure can be controlled at the nano level in order to reduce phonon dispersion, the thermal conductivity of the resin itself can be improved. The control of LCP orientation is now being studied as a means to achieve that goal. As shown in Table 4, because LCP has a high-crystalline, high-order structure in which molecules are micro-periodically aligned, and because no interface exists due to the lack of a phase that separates the amorphous and crystalline structures, it shows that the thermal conductivity is twice as great as that of regular resins even in the state in which the molecules are not macroscopically aligned. It has already been reported, as a result of the NEDO project, that by controlling the orientation of this resin through the magnetic field orientation the thermal



**Table 4** List of various plastics thermal conductivities

Resin	Thermal Conductivity (W/mK at 27°C)	Resin	Thermal Conductivity (W/mK at 27°C)
Polyethylene	0.33–0.52	LCP	0.3–0.4
Polypropylene	0.12	(Polydomain)	
Polystyrene	0.08–0.14	Soluble LCP	0.4–1.2
PVC	0.13–0.19	(Polydomain)	
ABS	0.2–0.33	PI	0.2
PMMA	0.17–0.25	Epoxy Resin	0.17–0.21
POM	0.23	LC Epoxy Resin	
PBT	0.18–0.29	(Polydomain)	0.28–0.43
PET	0.15	PC	0.19
PA	0.25	Polyurethane	0.31
PPS	0.22	Phenol Resin	0.13–0.25
PPO	0.19	PTFE	0.35–0.42


**Fig. 15** Relationship between orientation order of various LCP and thermal conductivity

conductivity can be improved to more than 2.5W/mK, even with the resin alone (Fig. 15, Tables 5 and 6).<sup>13</sup> In fact, when conducting injection molding the flow direction of the molecules (MD) demonstrates thermal conductivity five times greater than that of the traverse direction (TD) (Fig. 16). Therefore, this resin is extremely useful as a matrix resin for high-thermal-conductive LCP materials.

**Table 5** Thermal conductivities of various LCP applied 0T or 10 T into Z (thickness)-direction

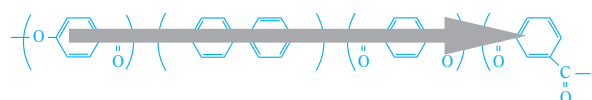
Applied magnetic field to Z direction	LCP			
	A	B	C	D
Thermal conductivity (W/mK)	0.26	0.28	0.41	0.20
	1.71	1.51	1.09	2.56
	(0.91)	(0.90)	(0.79)	(0.94)

( ) : Orientation order

**Table 6** Comparison of thermal conductivities between LCP and liquid crystal epoxy resin

	At 10T magnetic field applied	
	Thermal conductivity (W/mK)	
	Thermotropic LCP	LC Epoxy Resin
In plane (X-direction)	0.21	0.19
In plane (Y-direction)	0.24	0.32
Thickness (Z-direction)	1.55	0.69

Highly thermal conductivity of LCP in oriented direction

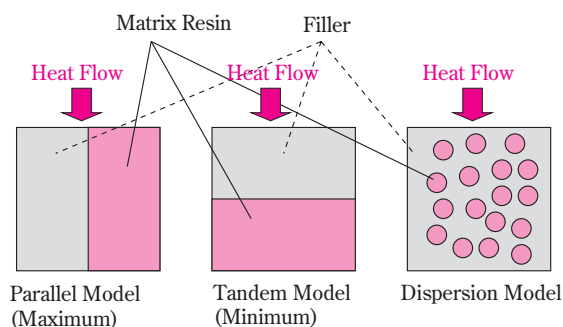


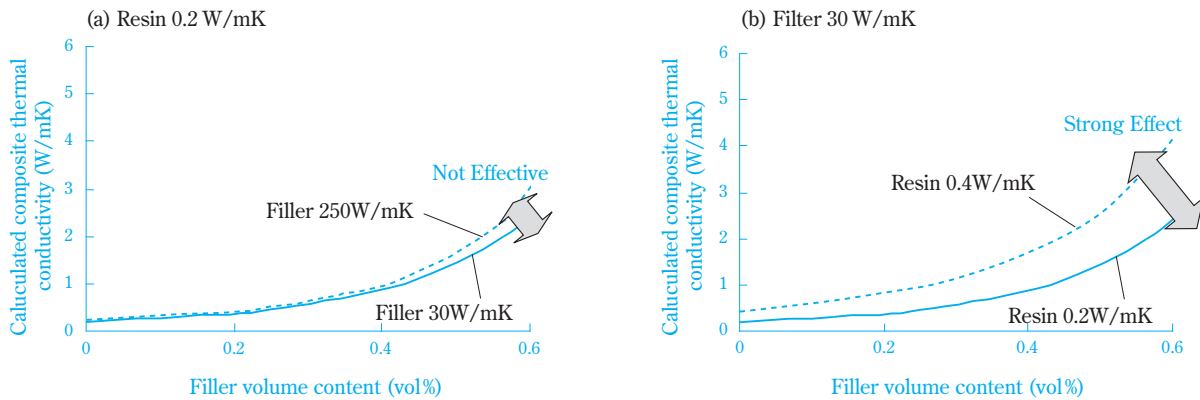
	TD	MD
Degree of orientation / %	50	85
Thermal Conductivity / W/mK	0.3	1.6

**Fig. 16** Highly thermal conductivity of LCP in oriented direction

## 2. LCP Composite Using Fillers that Have High Thermal Conductivity

The thermal conductivity of resin/filler composite can be classified into parallel, tandem and disperse models (Fig. 17).<sup>14</sup> All models must be densely filled with high-

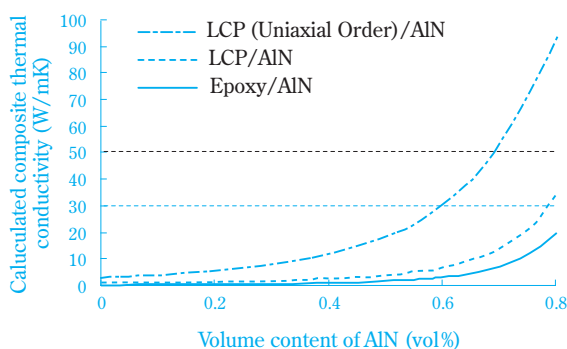

**Fig. 17** Various theoretical thermal conductive models for resin/filler composite



**Fig. 18** Relationship between filler content and thermal conductivity estimated using dispersion theory ((a) in case of fixed resin thermal conductivity (b) in case of fixed filler thermal conductivity)

thermal-conductive fillers in order to improve the thermal conductivity of the composite. **Figs. 18** (a) and (b) show the relationship between the filler volume content and the thermal conductivity of the filler obtained using Kanari's equation<sup>15)</sup> on the dispersion model. Although inside the composite heat is conducted mainly through the fillers, which have high thermal conductivity, the interface between one filler and another is separated by resin, which serves as a matrix. For that reason the thermal conduction of matrix resin becomes rate determination in the thermal conduction channel, and it will therefore be more effective to improve the thermal conductivity of the matrix resin than that of the filler.

**Fig. 19** indicates the calculation result when using epoxy (0.2W/mK) and LCP (0.4W/mK) as matrix resins and aluminum nitride (250W/mK) as filler. The figure shows the fact that the thermal conductivity varies significantly, even with the same volume content of aluminum nitride filler. While the thermal conductivity of LCP can exceed 30W/mK, this cannot occur in epoxy



**Fig. 19** Comparison between LCP and Epoxy resin estimated by dispersion theory (Thermal conductivity of Aluminum Nitride (AlN) assumed to be 250 W/mK)

resin. Furthermore, as described above, the thermal conductivity of LCP can approach 3W/mK due to the orientational control, and the theoretical potential of the composite can exceed 50W/mK when it is filled with high-conductivity fillers such as aluminum nitride. In regard to actual application of LCPs, because the same thermal conductivity (heat radiation property) can be achieved with less volume content of fillers, the use of LCPs with high thermal conductivity as matrix materials will expand the scope of application to areas where the special characteristics of resins (adhesive property, viscosity, flexibility and strength) can be effectively utilized.

## LCP Solubilization Technology and Heat Dissipating Substrates

### 1. Solubilization of LCPs

Conventionally, when processing LCPs into films the characteristic of resins—high susceptibility to orientation in a unidirection while melting—made the process quite a challenge.<sup>16)</sup> In order to solve this problem, as shown in **Fig. 2** and **Fig. 3**, Sumitomo Chemical modified the conventional molecular design of LCPs, which were by intrinsically insoluble, by taking advantage of the properties of thermotropic aromatic polyester to not orient, so that the LCPs would become soluble in a general-purpose solvent. Through this new molecular design Sumitomo Chemical has successfully obtained a film that shows a macroscopically isotropic property by removing the solvent through evaporation. The achievement was reported in this 'SUMITOMO KAGAKU in 2005.<sup>17)</sup>

The film thus obtained was transparent under the “as cast” state, in which the solvent was removed. Although it became opaque after performing heat-treatment and showed a high-order crystalline structure in which mol-

ecules were microscopically and periodically aligned, it was also confirmed that the film thus obtained was macroscopically isotropic, based on measurement using the microwave molecular orientation meter. Consequently, the film's electrical properties and dimensions of substrates and sheets became more stable. The film also showed adhesive property with high-performance semiconductors, flexibility and high resistance to dielectric breakdown, thereby expanding the scope of LCP application to the use as a heat-dissipating material (which the conventional technology could not achieve), insulating material for circuit boards and pre-pregs.<sup>18)–33)</sup>

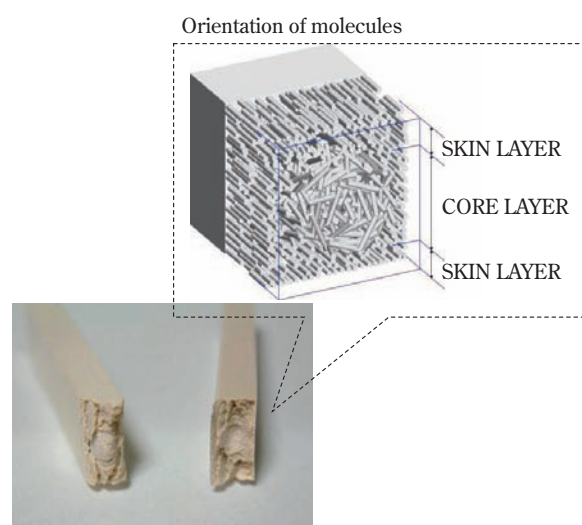
## 2. Heat-Dissipating Substrates Using Soluble LCP for Insulating Layer

As shown in Table 4, LCP shows better thermal conductivity compared to epoxy and polyimide resins. However, it was more difficult to improve the thermal conductivity of conventional LCP than that of epoxy and polyimide resins (both of which can easily be made into a composite with high-thermal-conductive fillers) because LCP did not dissolve in a solvent at all (Table 7). Moreover, conventional LCP had shortcomings such as the lamella structure shown in Fig. 20, which was created during the melt processing significantly prevented thermal conduction in the depth-wise direction.

Using this proprietarily developed soluble LCP, Sumitomo Chemical began developing printed circuit board materials having high thermal conductivity through the combined use of high-thermal-conductive fillers. The company aims to develop printed circuit board and aluminum substrates that demonstrate high thermal conductivity. For example, the physical properties of the printed circuit board materials obtained through the use

**Table 7** Thermal conductivities of various materials and their filler composite supplier performance

	Soluble LCP	Melt-process LCP Film	Epoxy Resin
Possible Filler Loading Volume	~ 80vol%	~ 50vol%	~ 80vol%
Resin Thermal Conductivity (W/mK)	0.4–1.2	0.3–0.4	0.2
Supplier	Sumitomo Chemical Co.	Several	Several



**Fig. 20** Cross section view of fracture of molded article

of glass pre-pregs having high thermal conductivity and those of the aluminum substrate containing soluble LCP in its insulating layer are shown in Tables 8 and 9, respectively.

Compared to the conventional substrates such as FR-4, the printed circuit board obtained through the use

**Table 8** Physical properties of printed circuit board using soluble LCP compared with FR4

Item	Method	Unit	FR4	Soluble LCP Glass Cross (1078-EGC)	
				Non Filler	Including Alumina
Thermal conductivity	Thermal resist method	W/mK	0.5	0.7	2.6
CTE	TMA (50-100°C)/TD-direction	ppm/°C	22.4	18.1	11.6
Tensile strength modulus	Autograph/MD-direction	MPa	224	242	126
		GPa	8.4	11.7	15.0
Moisture absorption	23°C/24Hr Dipping	wt%	0.57	0.29	0.33
Peeling strength	90° Direction/TD-direction (Cu Foil: 3EC-VLP)	N/cm	15.1	11.2	8.7
Solder resis.	260°C × 30sec Dipping	Blister/ Deformation/ Coloration	O.K.	O.K.	O.K.

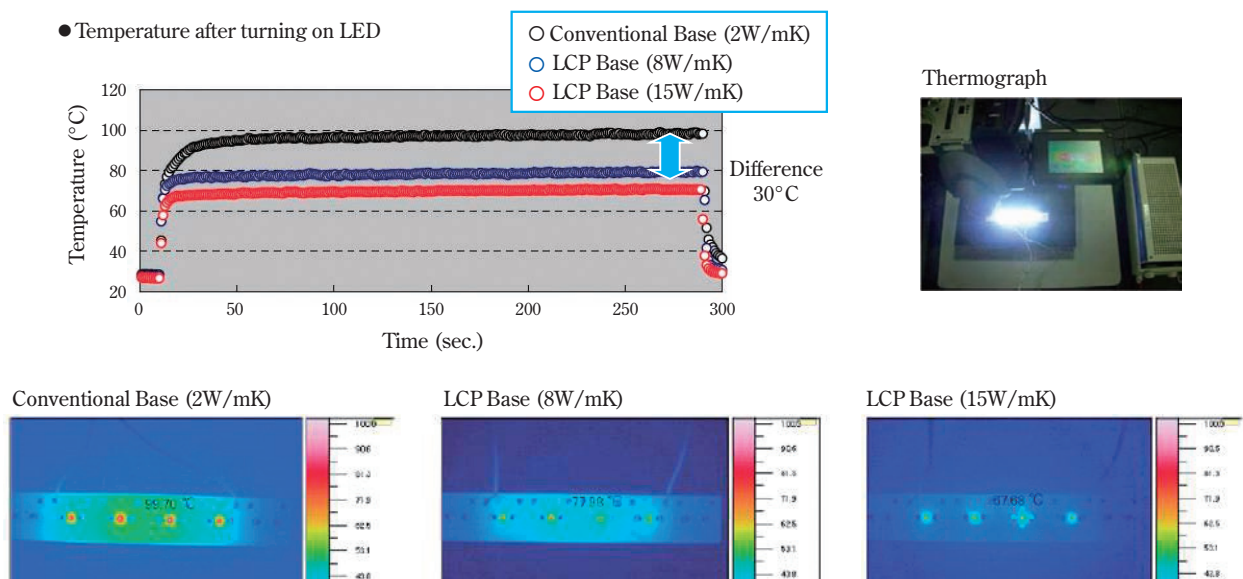
**Table 9** Physical properties of aluminum base circuit board using soluble LCP compared with conventional board

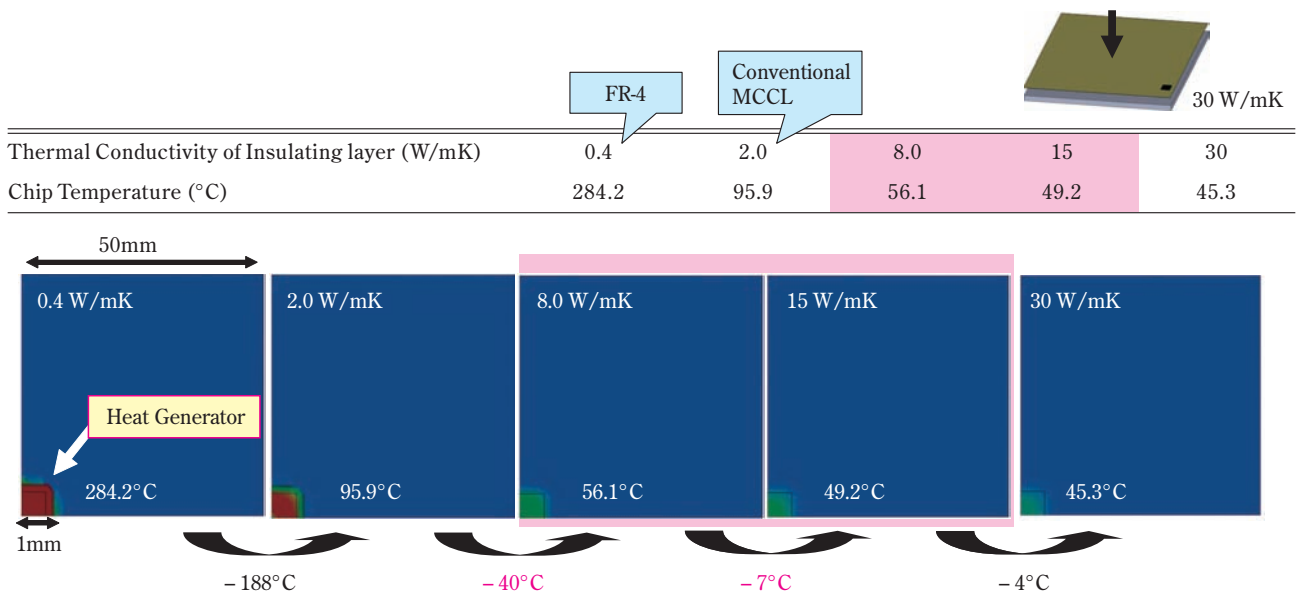
Item	Test Method	Condition	Unit	R&D grade		Conventional Base
				8W/mK	15W/mK	2W/mK
Cu	–	–	μm	70	70	70
Insulation Layer	–	–	μm	100	100	80
Al	–	–	mm	2	2	2
Peel strength	JIS C6481	No treatment	N/cm	10	9	20
		PCT24Hr		10	8	–
Dielectric breakdown	JIS C2110	No treatment	kV(AC)	10	10	5–8
		PCT24Hr		10	10	–
Thermal resistivity	Sumitomo's original	No treatment	°C/W	0.19	0.16	0.5–0.6
		PCT24Hr		0.2	0.16	–
Thermal conductivity	Thermal resistivity	No treatment	W/mK	8	15	1.6
Solder resistance	JIS C6481	No treatment		O.K.	O.K.	O.K.
	300°C × 4min.	Boil Water 1Hr		O.K.	O.K.	O.K.
Volume resistivity	JIS C6481	No treatment	Ω · cm	3.00E+16	3.00E+16	2.00E+13
Dielectric constant	1MHz	No treatment		4	4.1	4.5
loss		No treatment		0.01	0.01	0.02
Moisture absorption	JIS C6481	E-24/50 → D-24/23	wt%	0.4	0.4	0.5
Surface resistivity	121°C/97%	Before	MΩ	3.00E+07	3.00E+07	–
	96Hr/DC100V	After		2.00E+07	2.00E+07	–
Solder crack resistance	Sumitomo's original	-40 ↔ 125°C 15min.	Cycles	1000 <	1000 <	–
CTE	TMA	-40~150°C	ppm/°C	18	15	18
Tg	DMA	-40°C	°C	210	210	130

of glass pre-pregs shows not only high thermal conductivity but also the following outstanding properties that the soluble LCP has: excellent heat resistance (repair solder heat resistance), low thermal expansibility and the adhesive property toward conductors. Regarding aluminum substrates, the productization of the ultra-high-thermal-conductive substrate, which can even surpass the thermal conductivity of ceramic substrates such as alumina, has been determined through

joint development with the leading metal-base substrate manufacturer.

Based on the results of simulation test and experiments, it has been proved that the temperature rise of the electronic circuits of LED and power devices can be controlled (Fig. 21 and 22), and LCP substrates have begun garnering attention in the market as substrates that demonstrate greater heat dissipation than that of FR-4 and conventional metal-base substrates.

**Fig. 21** Heat transfer experiment of developed LCP aluminum base circuit board



**Fig. 22** Theoretical CAE thermal simulation of aluminum base LCP circuit board

## Conclusion

As described above, we have introduced the two major application areas of Sumitomo Chemical's LCPs: LED package housings and lighting-fixture components, specifically focusing on the following areas; (1) LED package housings to which the new resin whitening technology (catalyst process) is applied and (2) heat-dissipating substrates for LED modules that utilize the soluble LCP.

Armored with the unique resin manufacturing method (whitening through the catalyst process) and high production capacity (10,000-ton level in neat resins), Sumitomo Chemical is working to further expand the scope of LCP application to the following areas by making the most of the original LCP properties such as formability, dimensional stability, high heat resistance, low evolved gas volumes and high thermal conductivity, displays such as the light source (side view type) for cellular phone backlights (applied to SCG-233 (development code)); general-purpose lighting fixtures using high luminous LEDs (applied to SCG-233 (development code) and SZ6709); color rendering lighting fixtures and displays, such as light sources for liquid crystal backlights (applied to SCG-233 (development code), SZ6709 and SZ4709); and high-output light sources such as vehicle headlights (applied to SZ4709). Furthermore, we will continue to conduct research and development on the improvement of reflectivity (94% was achieved at the laboratory level) and light resistance, in order to achieve a clear differentiation from nylon resins.

Regarding the application of our proprietary soluble LCP to substrates, we have also successfully developed the ultra-high-thermal-conductive substrate, which can even surpass the thermal conductivity of ceramic substrates such as alumina, thus finally determining our goal for the development of a material that can serve as a business foundation.

We are planning to have all the LCP models function as a driving force for the growth of Sumitomo Chemical's business in SEP (Super Engineering Plastic).

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