Sanyo Chemical

It is a translation based on the content of corporate PR magazine
"Sanyo Chemical News" issued in New Year in 2023.

Thermally Conductive Polyurethane Gap Filler Achieving Both High Thermal Conductivity and Low Viscosity

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In recent years, the transition to the 5th generation mobile communication system, "5G", has rapidly advanced to sustain an increasing global communication volume caused by the wide-spread diffusion of smartphones, cloud services, and internet of things (IoT), as well as the adoption of automated operations in several fields. The amount of heat generated by electronic devices has increased as these have become more sophisticated, highly functional, and efficient, owing to an improved processing speed. However, the cooling conditions have become more demanding owing to the further miniaturization, high-density implementation, and reduction in thermally conductive area and routes of modern electronic devices.

To prevent malfunctions and damages, and improve quality and reliability of such devices, improved thermal measures for electronic devices have become a necessary requirement. To achieve this, the improvement of the performance of thermal interface materials (TIMs), which are used between electronic parts (heat-generating bodies) and thermally conductive parts (heat sink), has been increasingly studied.

In this article, we present the thermally conductive gap filler products, which are a type of TIMs, developed by our company.

Types of TIMs and the expanding market scale[1)]

TIMs include thermally conductive gap fillers, which are liquid at the time of application and then solidify, thermally conductive greases, which remain in a liquid state, and solid sheet-shaped thermally conductive sheets. **Table 1** shows a performance comparison of these different materials, indicating the most appropriate based on the conditions of the destination use, such as the amount of heat generated, distance between the heat generation and thermally conductive part, shape constraints, and reworking, adhesion, and workability requirements. Thermally conductive gap fillers are more advantageous compared to thermally conductive sheets because of the easier automated application, absence of waste materials, improved adhesion and followability, and low contact thermal resistance. Moreover, compared to thermally conductive greases, gap fillers present excellent pump-out, bleed-out, and vibration resistance. At present, TIMs have the largest market size for thermally conductive sheets, followed by gap fillers and greases. However, the demand for thermally conductive gap fillers is expected to grow considerably in the future owing to the increasing request for TIMs with automated manufacturing process for on-board electronic devices, including electric vehicle applications.

Contact thermal resistance: an important factor for thermally conductive in TIMs

Thermally conductive sheets are traditionally considered as the most advantageous TIM materials for their thermal conductivity. Compared to sheets, thermally conductive gap fillers and greases tend to have lower thermal conductivities because improving the filling rate while ensuring fluidity is a challenging task.

An advantage of thermally conductive gap fillers and greases is that they are in liquid form at the time of application, which allows them to adhere to the interface, thereby reducing the contact thermal resistance at the interface. Therefore, a high thermally conductive effect is expected. **Figure 1** illustrates an example of contact thermal

Table 1 Performance comparison among various TIMs

	Thermal conductivity	Contact thermal resistance	Automatic coating	Uniform coating performance	Film thinning	Pump-out resistance	Occurrence of part displacement	Reworkability
Gap fille	△	○	○	×	×	○	○	×
Grease	△	○	○	×	○	×	×	○
Sheet	○	×	×	○	×	○	○	○

resistance of the substrate interface using a thermally conductive gap filler. The seemingly smooth surfaces of the substrate and metal present micro-irregularities. These create contact points of microscopic size that cause contact thermal resistance owing to the presence of air at the contact interface. The contact thermal resistance is affected by how easily the gap filler occupies such surface irregularities when injected between the heating element and heat sink. Considering the factors contributing to the thermally conductive characteristics of TIMs, the contact thermal resistance is as important as the heat conductivity of the material.

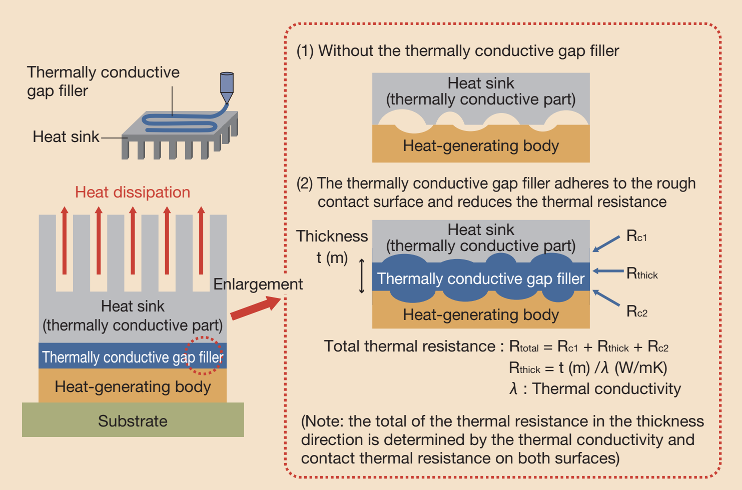


Figure 1 Contact thermal resistance of a thermally conductive gap filler


Before curing: Paste
After curing: Curing possible in desired form

Photo 1 Appearance of the urethane-based thermally conductive gap fillers developed by our company.

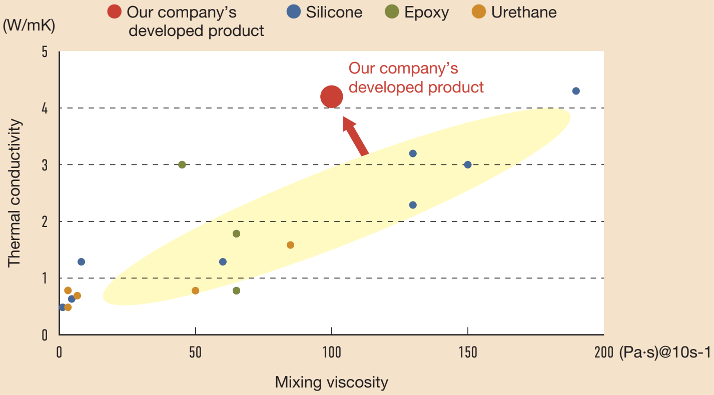


Figure 2 Correlation between viscosity and thermal conductivity of urethane-based thermally conductive gas filler

Characteristics of our urethane-based thermally conductive gap fillers

To provide a better heat management solution, our company designed and developed materials that could address conventional issues and achieve both thermal conductivity and fluidity, while maintaining the advantages of existing thermally conductive gap fillers. The developed products are urethane-based thermally conductive gap fillers obtained by mixing two solutions: a polyol solution containing a thermally conductive filler and a polyisocyanate solution. As shown in **Photo 1**, the result is a paste-like liquid that can be molded into an arbitrary shape at room temperature.

When the viscosity of the thermally conductive gap filler increases, the fluidity deteriorates. Since increasing the amount of thermally-conductive filler increases the thermal conductivity, achieving both high thermal conductivity and fluidity (low viscosity) represents a challenging aspect. Moreover, an increase in viscosity affects the ability of the filler to follow the unevenness of the heat-generating body or sink surface, resulting in a high contact thermal resistance. In this study, using a proprietary interface control technology, our company successfully produced a new filler that shows increased wettability at a high concentration of thermally conductive particles in the resin, while maintaining low viscosity and high fluidity. **Figure 2** shows the correlation between the thermal conductivity and viscosity of common gap-filler products. As described above, the mixing viscosity of the liquid tends to increase as the thermal conductivity increases. The urethane-based thermally

conductive fillers developed by our company have a better balance between thermal conductivity and viscosity than conventional commercial products, according to our research. Therefore, our developed products show good gap filler applicability and can accommodate automation of TIM mounting, as well as contribute to improvement of production efficiency. In addition, our materials can adsorb part of the stress caused by the deflection of the electronic parts and heat sink following compression.
In addition, as our products are urethane-based and non-silicone, there is no concern about defective continuity due to volatilization of low-molecular-weight siloxane components, which is a problem with silicone-based TIM used in electronic devices. Lastly, our products can be used for various applications, including electronic devices and automotive applications, because they do not easily peel off under vibration owing to the flexibility of urethane and the insulating-type filler used to produce them.

Dispersion mechanism of thermally conductive filler in our products

The key to achieving both high thermal conductivity and low viscosity was ensuring the optimal dispersion of the high-concentrated thermally conductive fillers using our company’s interface control technology. As shown in the schematic in **Figure 3**, without a dispersant the thermally conductive filler aggregates forming fine air bubbles among the particles. When our company’s proprietary dispersant is added, it adsorbs onto the thermally conductive filler surface improving the wettability of the dispersion medium. This process releases the air effectively, thus, enabling the production of uniform-paste materials that do not form aggregates, even when a large amount of thermally conductive filler is mixed in the dispersion medium.

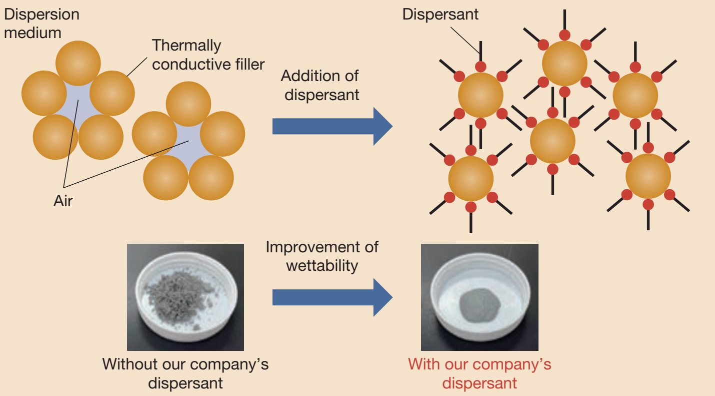


Figure 3 Dispersion effect of thermally conductive filler by dispersant

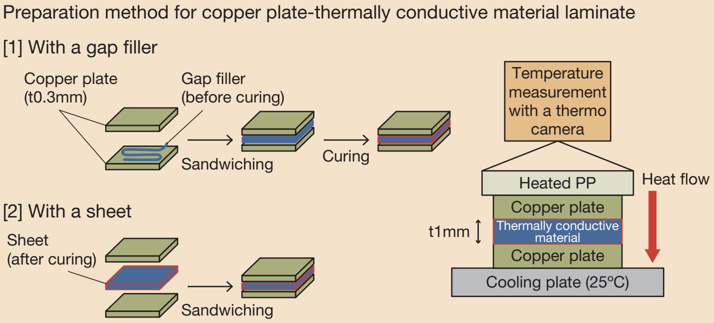


Figure 4 Copper plate-thermally conductive material laminate for thermal conductivity evaluation

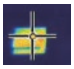
	Without thermally conductive material (Air layer)	With our company’s polyurethane gap filler (4.3W/mK)	Thermally conductive silicone sheet from another company (4.5W/mK)
Immediately after starting	 75.2°C	 74.0°C	 74.7°C
After 60 seconds	 48.3°C	 33.3°C	 40.1°C

Figure 5 Thermally conductive effect comparison

Thermally conductive characteristics of our products

We verified the thermally conductive effect of the products developed by our company. As shown in **Figure 4**, we observed the changes in the temperature of a polypropylene (PP) plate surface with a thermo camera after sandwiching each one of the thermally conductive materials between two copper plates and placing a heated PP plate on top. As evidenced by the results shown in **Figure 5**, our products can release heat more efficiently. In fact, they present a faster temperature drop rate for the heated PP plate than that of the sample without a thermally conductive material (i.e., including spacers only), or with a silicone thermally conductive sheet from another company with the same thermal conductivity. The enhanced thermally conductive effect of the developed products, compared to that of the thermally conductive sheet with a similar heat conductivity, results from their ability to follow the microscopic irregularities of the substrate surface owing to their initial liquid form, thus, reducing the contact heat resistance after molding.

Although the performances of our materials in practical applications need further exploration, in this study, we demonstrated that the developed products have excellent thermally conductive characteristics and the ability to meet the needs for automated manufacturing process.

Lineup of our developed products

Table 2 shows the lineup of the urethane-based thermally conductive gap filler products currently under development at our company.

In addition to the general product line with high thermal conductivity, a product line adhering to the nonpolar PP substrate, which is generally a difficult task, is also under development, always utilizing the urethane design technology developed by our company.

Although silicone-based gap filler products generally have low adhesiveness to the substrate, the ability to control or add adhesiveness to various types of substrates is the strength of our company’s urethane design technology. Lastly, our lineup includes a product line that shows limited liquid dripping or pump-out by obtaining high thixotropy using our rheology control technology.

Future developments

As electronic devices, such as automotive devices, power supply/energy devices, and communication modules, are becoming smaller, more sophisticated, and more functional, the required performance of the thermal measures in these devices is expected to increase in the near future, considering also the development of IoT. Our company will continue to develop high-quality products that meet the ever-changing market requests using our interface control and urethane design technology developed over several years. At the same time, we aim at testing practical applications of our thermally conductive gap fillers as heat management solutions to respond to future technological requirements.

Literature

1) “Current situation of heat control/thermally conductive material market and development of new applications in 2019,” FUJI KEIZAI Co., Ltd.

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In Japan
Sales & Marketing Dept. of Polyurethane Division
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In U.S.A
Sanyo Chemical America Incorporated
https://sanyochemicalamerica.com/

Table 2 **Lineup of urethane-based thermally conductive gap fillers in development**

Product name	RCG-020	RCG-022	RCG-028	RCG-029	Evaluation method
Outline	Standard product	High thermal conductivity Low viscosity product	Light weight Low wear PP adhesion product	Light weight Low wear High thixotropy product	–
Type (mixing ratio is in volume ratio)	2-liquid curing (Liquid A : Liquid B = 1 : 1)	2-liquid curing (Liquid A : Liquid B = 1 : 1)	2-liquid curing (Liquid A : Liquid B = 4 : 1)	2-liquid curing (Liquid A : Liquid B = 1 : 1)	–
Heat conduction (W/mK)	4.0	4.3	2.8	3.1	JIS R1611 (laser flash method)
Asker C hardness	94	94	90	90	JIS K7312
Paste viscosity (Pa·s)	Liquid A: 90 Liquid B: 50 @0.1/s, 40°C	Liquid A: 80 Liquid B: 20 @0.1/s, 40°C	When mixing A and B 136 @1/s, 40°C	Liquid A: 150 Liquid B: 150 @1/s, 25°C	ASTM D2556 (MCR 92 manufactured by Anton Paar)
Density (g/cm^3)	2.8	3.0	2.1	2.1	JIS K7112 (water replacement method)
Adhesiveness (25°C)	To glass epoxy*1 : ○ To PPS*2 : ○ To PP*3 : × To PET*4 : ○ To ED steel plate*5 : ○	To glass epoxy: ○ To PPS: ○ To PP: × To PET: ○ To ED steel plate: ○	To PET: ○ To ED steel plate: ○ To PP: ○	To glass epoxy: ○ To PPS: ○ To PP: × To PET: ○ To ED steel plate: ○	JIS K6850 Tensile shearing adhesion strength is 0.1 MPa or more⇒○

*1: L6504C1 manufactured by Nikkan Industries Co., Ltd. *2: General-purpose product. *3: Standard test plate manufactured by Nippon Testpanel Co., Ltd. *4: L#100S10 manufactured by Toray Industries, Inc. *5: Standard test plate from SPCC to SD. (Reported values are representative values and not specification values. This lineup lists the products under development and is subject to change without notice.)