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The Paint Prepared Using 2D Materials: An Evaluation of Heat Dissipation and Anticorrosive Performance

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Heat sinks are most widely used in thermal management systems; however, the heat dissipation efficiency is usually limited. Therefore, in order to increase heat dissipation efficiency of the heat sink, the heat-dissipating paint using 2D materials (hexagonal boron nitride (h-BN) and graphene) as thermally conductive additive was designed and evaluated in the present study. The heat dissipation performance of the paint was calculated from temperature difference between the paint-coated and -uncoated specimens mounted on the heat source. The highest heat dissipation performance was obtained when the ratio of h-BN to resin was 1/10 in the paint. In addition, further reduction in the temperature of the test specimen by 6.5 ^{o}C was achieved. The highest heat dissipation performance of the paint prepared using graphene was achieved at a 1/50 ratio of graphene to the resin, and a 6.5 ^{o}C reduction was attained. In addition, graphene exhibited enhanced corrosion resistance property of heat-dissipating paint by inhibiting the growth of the paint blisters.

Keywords: *Paint, h-BN, Graphene, Thermal dissipation, Corrosion resistance*

1. Introduction

As electronic devices with thinner and higher power densities have been developed, thermal management has become one of the critical issues to be addressed [1,2]. This is because the heat accumulated in the electronic device greatly limits the performance of the electronic device and can cause reliability problems [3]. Thus, heat dissipation systems are one of the important goals for the continued development of the electronics industry [1,3]. Heat sinks are the most widely used in thermal management systems, but the heat dissipation efficiency is usually limited [4]. Lots of studies on heat sinks focus on the theoretical analysis, numerical simulation, or optimization of pin parameters such as spacing, height, and orientation of heat sink fins [4]. Although these methods are an effective way to optimize the performance of heat sinks, they are not suitable for small / high power devices and require a new approach to dissipate the heat accumulated in small / high power devices.

Recently, two dimensional (2D) materials such as graphene [5,6], hexagonal boron nitride (h-BN) [7,8], transition metal dichalcogenides (TMDCs) [9,10], and black phosphorous [11,12] have receive particular attention as an additive for heat dissipation materials because of their excellent thermal conductivity. Impressively, the thermal conductivity of graphene is in the range of 4,800 – 5,300 W m^{-1} K^{-1} at room temperature and is much higher than commonly used materials such as copper and aluminum [13]. h-BN has a hexagonal honeycomb lattice structure similar to graphene, but is electrically isolated with a large bandgap of 5.8 eV [1,14]. It has been reported that bulk h-BN has a high thermal conductivity of 360 W m^{-1} K^{-1} at room temperature, and is similar to the thermal conductivity of copper [8]. Thus, many researchers have reported the use of graphene and h-BN as a thermally conductive additive to improve the thermal conductivity of polymer composites [5,7]. The thermal conductive properties of graphene-epoxy nanocomposites as thermal interface materials were evaluated by Shahil and Balandin [15]. According to their report, thermal conductivity of multilayered graphene nanocomposites is reached to 5.1 W m^{-1}

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K^{-1} at 10% of the graphene volume fraction, which corresponds to thermal conductivity enhancement of 2300%. Shtein *et al.* [6] reported that the thermal conductivity enhancement factor (thermal conductivity enhancement factor = thermal conductivity enhancement % / filler volume %) of epoxy nanocomposites prepared using graphene nanoplatelets was achieved above 110 at 15% of the graphene volume fraction. Also, according to a report by Zhu *et al.* [16], the boron nitride nanosheet/cellulose paper exhibited a thermal conductivity of 145.7 W m^{-1} K^{-1}, which was 4000 times better than that of bare cellulose paper. This value was also higher than commercial Al alloy 135. Song *et al.* [17] prepared a nanocomposite film filled with 50 vol% of h-BN and the film reached a thermal conductivity of 30 W m^{-1} K^{-1}. In thermal management systems, the thermal conductivity of thermal interface materials has been improved by many studies, but it does not lead to an improvement in heat release efficiency. Thus, novel approaches are required to effectively release the heat generated in the electronic device to the outside, and heat dissipating paints (or coatings) can be one of the alternatives. In this study, paints were prepared using 2D materials such as graphene and h-BN as thermally conductive additives in order to improve the heat dissipation efficiency of the heat sink, and the heat dissipation performance was evaluated by a comparative measurement method. In addition, the anticorrosive performance of the paint was evaluated by the cyclic salt spray test method.

2. Experimental Methods

2.1 Materials

Graphene (product name: N002-PDR) and h-BN (product name: HW05) are purchased from Angstron Materials and Yeeyoung TTC, respectively, and used as a thermal conductive additive without any other treatment. The graphene used in this study is few-layered graphene having properties

Table 1 Basic properties of thermal conductive additives; reproduced

Materials	Graphene	h-BN
Average lateral size	4 μm	5 μm
Thickness	1 – 2 nm	–
Specific surface area	400 – 800 m^2/g	11 – 13 m^2/g
Tap density	0.005 – 0.01 g/cm^3	0.3 – 0.4 g/cm^3
Carbon	> 95.0%	< 0.3%
Hydrogen	< 2.0%	–
Nitrogen	< 0.05%	–
Oxygen	< 2.5%	< 0.8%
Ash	< 2.5%	–
BN	–	< 99.0%
B_2O_3	–	< 0.5%

* data was obtained from each manufacturers

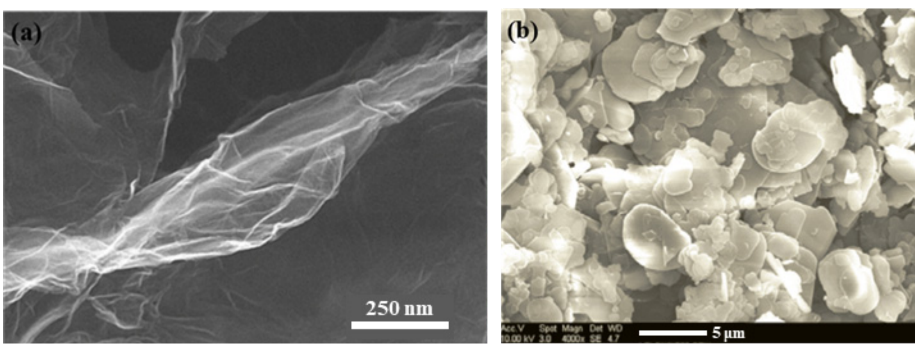


Fig. 1 SEM images of thermal conductive additives; reproduced, (a) graphene (b) h-BN.

such as high surface area and low density as exhibited in Table 1. The basic characteristics of h-BN were also exhibited in Table 1. Fig. 1 showed SEM images of thermal conductive additives and it was obtained from each manufacturers, respectively [18,19]. Silicon-based resin was supplied by Noroo Paint and used as the basic resin in this study. It has unique properties such as high heat resistant and adhesiveness, so it is suitable for use in high temperature application such as kitchen and heating equipment.

2.2 Preparation of Heat-dissipating Paint and Test Specimens

The paint was simply prepared using a high speed dispersion method as follows; Firstly, thermal conductive material and thinner were added into the resin then it was slowly agitated by magnetic stirrer at low rpm for overnight. Thermal conductive materials in resin mixture were dispersed by high rotation dissolver (DISPERMAT® LC75, VWA-Getzmann) at 10,000 rpm for 30 min. As a result, heat-dissipating paints having a good dispersion of the thermally conductive material were obtained. For paints prepared using h-BN, the remaining components except the concentration of the thermally conductive additive were fixed as shown in Table 2. This is to understand the relationship between the concentration of thermally conductive additives and the heat dissipation performance of the paint. The weight ratios of h-BN to resin differed by 10%, 20%, 30% and 40% and denoted B10, B20, B30 and B40, respectively. In the case of paints prepared using graphene, graphene is a nanomaterial having a high surface area as summarized in Table 2, the content of thinner was constantly increased according to the graphene content to maintain a suitable viscosity for the coating. In addition, the paint was represented by G00NS, G05NS, G10NS, G15NS, G20NS and G25NS, respectively, according to the content of graphene. Aluminum (Al) 6061 alloy [20], which is widely used for general purpose, was used to fabricate test specimens for evaluating heat dissipation performance of the paints. Single side of Al alloy plate (50 mm × 150 mm × 2 mm) was coated with heat-dissipating paint using a film applicator (Elcometer 3540) or air brush, and dried overnight in a vacuum oven at 130 ^{o}C.

2.3 Adhesive Strength Test

The adhesive strength test of the paint to the Al alloy plate was carried out in accordance with ISO 2409 [21]. The details are as follows;

1) Clean the coating and dry thoroughly.
2) Cut six lines at a right angle to the horizontal and vertical lines.
3) After attaching the tape, remove it quickly as exhibited in Fig. 2. At this time, the adhesive strength of the tape is defined as 6 to 10 N / 25 mm width.
4) Evaluate the adhesive strength of the paint coating layer by comparing the removed pieces with the grade specified in ISO 2409.

2.4 Measurement of Heat Dissipation Performance

The heat dissipation performance of paints was evaluated by the difference in temperature between paint coated and un-coated test specimens when the heat generated from heat source was transferred to the surroundings through the test specimens as shown in Fig. 3. The evaluation process is described in detail in our previous work [22]. Briefly, in first, all the specimens (paint coated and

Table 2 Composition on ingredients of paint prepared using 2D materials

TCF$^{1)}$	Sample name	TCF/resin	Thinner/resin	Δ thinner / Δ graphene
h-BN	B10	10 %	1	
	B20	20%	1	
	B30	30%	1	
	B40	40%	1	
Graphene	G00NS	n/a	0.5	n/a
	G05NS	0.5%	1	n/a
	G10NS	1.0%	1.2	40
	G15NS	1.5%	1.4	40
	G20NS	2.0%	1.6	40
	G25NS	2.5%	1.8	40

$^{1)}$TCF: thermal conductive filler

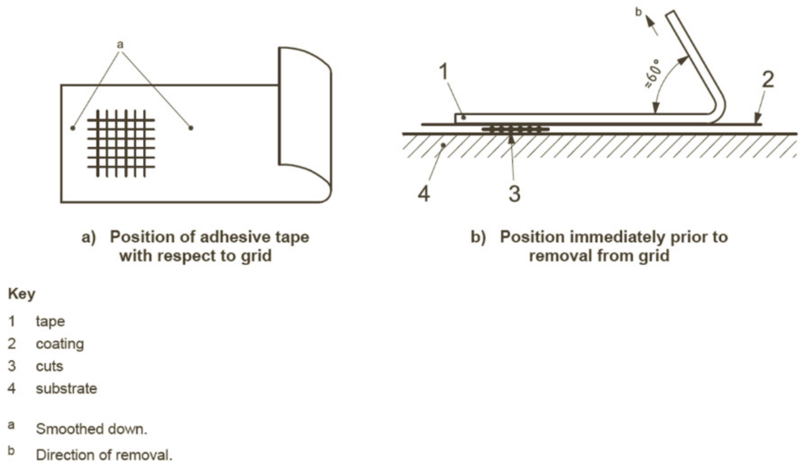


Fig. 2 Positioning of adhesive tape for cross-cut adhesive test, reproduced [21].

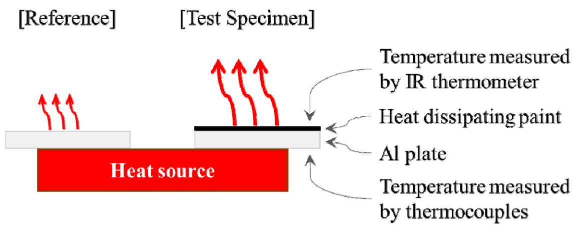


Fig. 3 Schematic diagram for evaluation of heat dissipation performance of the test specimens.

un-coated test specimens) were completely preheated at 30 °C, and then the temperature of the both specimens was observed while the temperature of the heat source was instantaneously increased to 100 °C and maintained at 100 °C for 1 hour. The temperature of test specimens was measured using a thermocouple thermometer and an infrared thermometer. In order to reduce the errors that may occur around the heat source, not only the temperature around the heat source was kept constant, but also the temperature of the test specimen was measured several times to calculate the average and standard deviation.

2.5 Cyclic Salt Spray Test

The cyclic salt spray test is one of the methods to evaluate the corrosion resistance and rust spread of paints. Test specimens for evaluation of corrosion resistance were prepared by coating a heat-dissipating paint on HR steel plate (150 mm × 75 mm × 5 mm) and curing for 3 days under 50% RH. The coating layer of the test specimen was peeled in an X shape (length: 40 mm, cross point angle: 40° ± 5°) according to the method of ASTM D1645 [23] so that the substrate was exposed to the outside. For evaluation of corrosion resistance, a spray solution containing 0.05% sodium chloride and 0.35% ammonium sulfate was used as specified in ASTM G85 [24]. The test was repeated for 28 days, with the spraying and drying process repeated every hour.

3. Results and Discussion

3.1 Observation of Surface Morphology

Fig. 4 and 5 show the surface morphology of test specimens coated with heat-dissipating paint prepared using 2D materials. In the case of h-BN, as shown in the magnified images of Fig. 4, despite being used in a ratio of up to 40% compared to the resin, it could be confirmed that h-BN was well dispersed without any aggregation. As a result, even paints with a high content of h-BN were able to obtain good coating quality. In the heat-dissipating paint prepared using graphene, as exhibited in Fig. 5, it could

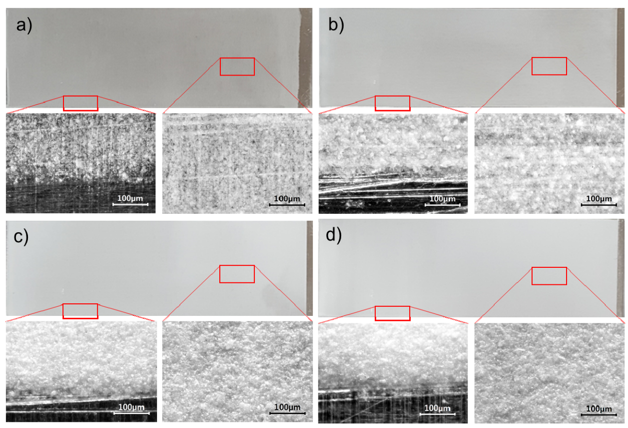


Fig. 4 Surface morphology of test specimens coated with paint prepared using h-BN; reproduced [22], a) B10, b) B20, c) B30, and d) B40. Scale bar is 100 μm.

Fig. 5 Photograph of test specimens coated with paint prepared using graphene. The test specimen size is 5 mm × 15 mm.

be confirmed that the coating was well performed without any defects in all the test specimens.

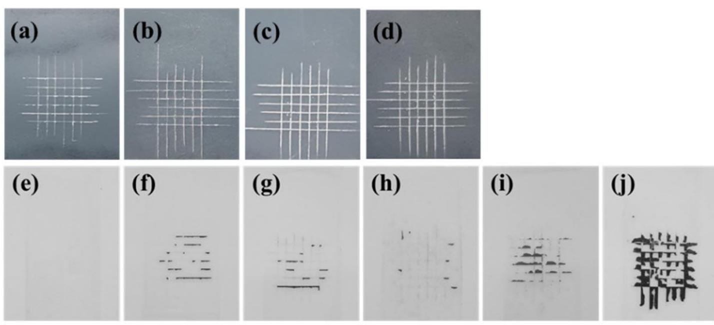
3.2 Adhesive Strength of Paint Prepared using 2D Materials

Adhesive strength is one of the essential requirements of paint and can easily be assessed by the cross-cut adhesion test method. It is a test method for evaluating the resistance to detachment of the paint coating layer from the substrate and is mainly for use in the laboratory but also suitable for field testing [21,25]. Test methods such as number of cuts, cutting interval, adhesive strength of adhesive tape, etc. are described in detail in ISO 2409 or ASTM D3359-09, and briefly summarized below.

1) Number of cut: 6 cuts in each direction of the lattice pattern.
2) Spacing of cut: 1 mm spacing for hard substrate below 60 μm of thickness.
3) Adhesive strength of the tape: between 6 N and 10 N per 25 mm width.

The adhesive strength test on Al substrates of paints prepared using 2D materials was carried out in accordance with ISO 2409 and the results were exhibited in Fig. 6. The results of adhesive strength test by ISO 2409 are all classified into six levels as shown in Fig. 6(2). The first three stages, level 0, level 1, and level 2, are used to indicate the adhesive strength suitable for general use. For paints prepared using h-BN, only small pieces were detached along the cutting pattern. Even though the content of h-BN increased, it was confirmed that the coating layer was well adhered to the Al substrate without deforming. These result corresponds to level 1 of detachment of the surface in the cross-cut area according to the test result classification of ISO 2409. It means that the small pieces of the paint coating layer detached from the Al alloy substrate in the cross-cut area is less than 5%, and the adhesive strength of the paint to the Al alloy substrate is very good. In the case of paints prepared using graphene, when the ratio of graphene to resin was 1.5% or less, it corresponded to level 1 of the ISO 2409 classification, and showed good adhesive strength. However, the adhesive strengths of paints with graphene-to-resin ratios of 2.0% and 2.5% corresponded to levels 2 and 4 of the ISO 2409 classification, respectively. Unfortunately, when the ratio of graphene to resin is 2.5% or more, the adhesive strength of the heat-dissipating paint to the Al alloy substrate is significantly lowered, which is not suitable for use in general applications. Graphene, which is used as a thermally conductive material, is a nanomaterial having a large surface area, so as the ratio of graphene to resin increases,

(1)



(2)

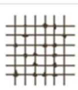
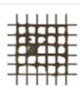
Classification	0	1	2	3	4	5
Detachment	None	< 5%	5% ~ 15%	15% ~ 35%	35% ~ 65%	> 65%
Appearance of surface of cross-cut area	-					-

Fig. 6 (1) Cross-cut adhesion strength test of paint prepared using 2D materials; (a) B10, (b) B20, (c) B30, (d) B40, (e) G00NS, (f) G05NS, (g) G10NS, (h) G15NS, (i) G20NS, and (j) G25NS, (2) Six-step classification of ISO 2409 cross-cut adhesion strength test.

the solid volume and the resin are not balanced. As a result, the adhesive strength of the paint to the substrate is weakened. From the cross-cut adhesion test, it could be easily confirmed that the adhesive strength of the heat-dissipating paint using 2D materials was suitable for use in general applications.

3.3 Evaluation of Heat Dissipation Performance

As a rule, heat transferred from the heat source to the heat sink is mainly released by convection into the surrounding area. If the heat sink is coated with heat-dissipating paint, however, heat is transferred to the outside by radiative emission as well as convection. Therefore, the heat accumulated inside the heat sink is released to outside more quickly, which can further lower the temperature of the electronic devices. The heat dissipation performance of paints prepared using 2D materials was evaluated by the difference in temperature between paint coated and un-coated specimens when heat generated from the heat source was transferred to the surroundings through the substrate, as shown in Fig. 3.

The results of evaluating the heat dissipation performance over time of the paints prepared using h-BN were exhibited in Fig. 7. The temperature of the heat source increases rapidly up to the set temperature, and then remains constant at the set temperature. Therefore, the heat dissipation performance also showed a sharp change as shown in Fig. 7a according to the temperature change of the heat source. Initial heat dissipation performance is able to one of the important factors in the thermal management system because it not only affects the overall heat dissipation performance, but also can protect the device from sudden thermal shocks. Interestingly, our results showed that the initial heat dissipation performance decreases over a period of time. This is because there was a difference in time to reach the highest temperature of each test specimen. In the case of unpainted test specimen, the maximum temperature of the specimen is reached within a short time. On the other hand, for paint-coated test specimen, the heat transferred from the heat source is released to the outside, resulting in longer time to reach the maximum temperature of the specimen. In other words, in the unpainted test specimen, a large amount of heat accumulates in the test specimen within a short time, while in the paint-coated specimen, the heat accumulation rate inside the test specimen is lowered by the effect of heat dissipation. Therefore, the heat dissipating paint can serve to protect the electronic device from thermal shock. The heat dissipation performances of the paints according to the content of h-BN distinguished into the initial section where the temperature of the heat source rapidly increased and the section where the temperature of the heat source was kept constant, and it was compared in detail. And the results were exhibited in Fig. 7b and 7c, respectively. In B10, which the ratio of h-BN to resin is 10%, the initial heat dissipation performance was up to about 8 ℃. However, as the ratio of h-BN to resin increased from 10% to 40%, the initial heat dissipation performance was decreased from 8 ℃ to 6 ℃ (see Fig. 7b). The same tendency was also observed in sections where the temperature of the heat source was kept constant (see Fig. 7c). As shown in the results of Fig. 7, despite the increase in the content of thermally conductive materials in the paint, the reason for the deterioration of the heat dissipation performance may be due to phonon boundary scattering, as reported by Kim [26]. In other words, an increase of the h-BN content in the paint resulted in an increase in the disorder of h-BN, which in turn led to an increase in phonon boundary scattering. As a result, the efficiency of dissipating heat inside the test specimen to the outside was reduced. Interestingly, the heat dissipation performance of all paints gradually increased in the section where the tem-

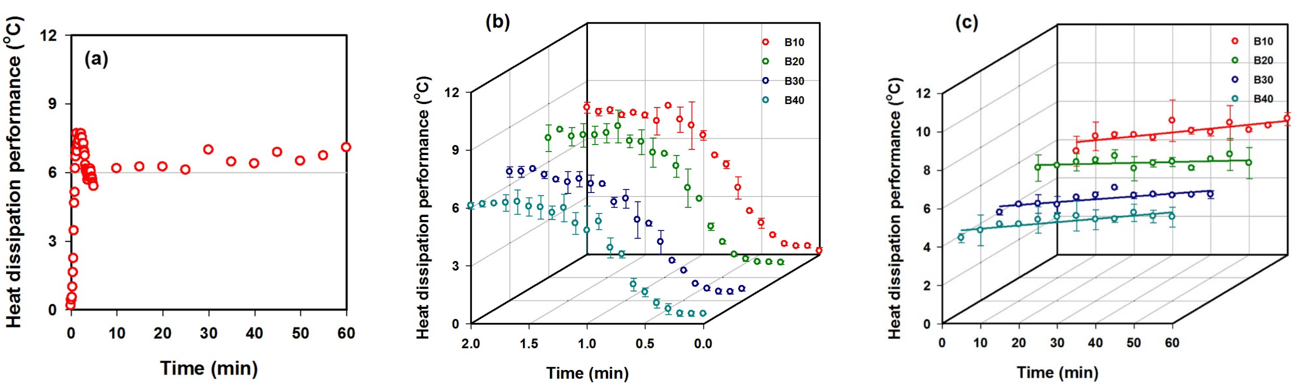


Fig. 7 Heat dissipation performance of paint prepared using h-BN. (a) overall heat dissipation performance, (b) heat dissipation performance at initial stage, (c) heat dissipation performance at continuous stage.

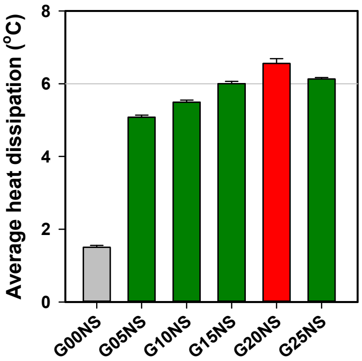


Fig. 8 Average heat dissipation performance of paint prepared using graphene.

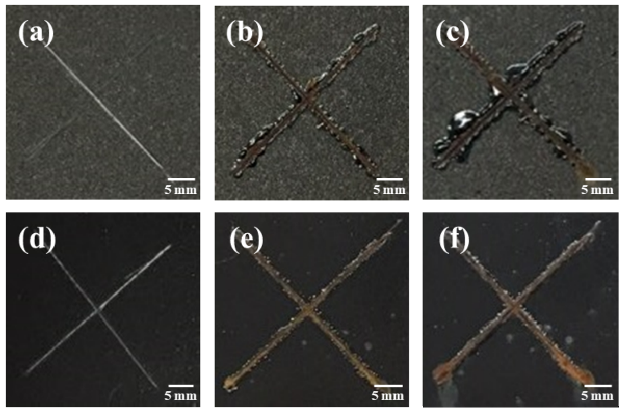


Fig. 9 Anticorrosive performance of paint in salt spray environment, reproduced [31]. (a)-(c) resin-based, (d)-(f) 5% graphene/resin, (a) before test, (b) after 14days, (c) after 28days, (d) before test, (e) after 14days, (f) after 28days.

perature of the heat source is kept constant. This result is believed to be due to the accumulation of heat slowly inside the test specimen, which is not coated with heat-dissipating paint. In summary, B10 showed the best heat dissipation performance, and the average heat dissipation performance was about 6.5 °C.

In paints prepared using graphene, similar heat dissipation performance was observed as described above. In the section where the temperature of the heat source is kept constant, the average heat dissipation performance by graphene content was compared and showed in Fig. 8. As the graphene content increased, the heat dissipation performance of the paint also increased, and the highest heat dissipation performance (about 6.5 °C) was observed when the ratio of graphene to resin was 2%. On the other hand, when the ratio of graphene to resin exceeds 2%, the heat dissipation performance of the paint is reduced because the phonon boundary scattering increases as in the previous study [27].

3.4 Anticorrosive performance of paint prepared using graphene

Heat-dissipating paint should not only have good heat dissipation performance, but also be able to protect the heat sink from corrosion by moisture or salt. It has been reported that graphene has excellent thermal conductivity as well as good corrosion inhibition [28]. According to the report by Cui *et al.* [29], graphene oxide coated with poly dopamine further improved the corrosion resistance of epoxy coatings, and Zhong *et al.* [30] reported on the anticorrosive performance of epoxy coatings using graphene and graphene oxide hybrid materials. In this study, the anticorrosive performance of a heat-dissipating paint having a graphene-to-resin ratio of 0.5% was evaluated by the cyclic salt spray test and the results were exhibited in Fig. 9. In the test specimen coated without graphene, blisters of the paint coating layer were observed within a relatively short time, and the blisters grew up to 5 mm or more after 28 days. On the other hand, for the test specimen coated with the graphene-containing paint, the paint blisters of less than 1 mm were observed at a time similar to the previous case, but it did not grow even after 28 days. As a results, as reported by Prasai [28], graphene used as a thermally conductive material is to working as a corrosion inhibitor to reduce rust spreading and to enhance corrosion resistance of heat-dissipating paints.

4. Conclusions

It was evaluated that the heat dissipation performance of paints prepared using 2D materials such as graphene and h-BN as the thermally conductive additive. The heat-dissipating paint could be easily prepared by the high-speed dispersion method, and it was confirmed that the thermally conductive additives were well dispersed by observing the surface morphology of the paint-coated test specimens. It has been found that the adhesive strength of heat-dissipating paints according to the ISO 2409 test method is suitable for general applications. However, since graphene is a nanomaterial having a large surface area, when the ratio of graphene to resin exceeds 2%, the adhesive strength is sharply lowered, which was not suitable for general use. The best heat dissipation performance was observed when the ratio of h-BN or graphene to resin was 10% and 2%, respectively, and in both cases the temperature of the specimen could be lowered by 6.5 °C compared to the uncoated specimen. On the other hand, if the ratio of the thermal conductive material to resin is increased above a certain level, the heat dissipation performance of the paint is slightly lowered, which may be due to the increase in phonon boundary scattering by the thermal conductive materials. In addition, graphene has enhanced the corrosion resistance of heat-dissipating paint by inhibiting the growth of the paint blisters. As discussed in this study, the paints prepared using 2D materials showed excellent heat dissipation and anticorrosive performances, and are expected to improve heat dissipation efficiency of heat sinks.

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Notes

The authors declare no competing financial interest.

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