

Research Article

Heating Characteristics of Films Prepared with Polymer-Graphite Composites

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Heating films were prepared by using poly(methyl methacrylate) and polybutadiene composites containing graphite. The heating film was prepared by casting the as-made polymer composite on the PET film. Copper electrodes were attached to both ends of the as-prepared film, and the heating characteristics of the film were analyzed while applying the DC voltage. The electrical conductivity and the heating temperature of the heating films depended on the size, the structure, the content, and the dispersion characteristics of the graphite in the composite. The electrical resistance of the heating film was controlled to adjust the heating temperature of the film. The relationship between the physical/chemical structure and the heating characteristics of the composite film was studied by measuring the heating temperature as functions of film thickness and resistance by using an infrared thermal imaging camera. The lower the film resistance, the higher the heating temperature of the film. The surface temperature was uniform throughout the film.

1. Introduction

Heating film is a heating element that generates thermal energy from the film surface, based on the principle of Joule heating (also known as Ohmic heating) [1]. When copper electrodes are attached to both ends of a heating film and a rated voltage is applied on the film, thermal energy is generated over the entire surface of the film. Generally, a heating film radiates thermal energy converted from electric energy. The temperature of a heating film can be easily controlled, and the use of the heating film does not pollute the air, so it has advantages in terms of hygiene and noise [2, 3]. For this reason, the heating films are widely used for home heating systems in Europe and the application areas have been expanded to the industrial fields of dryers, healthcare aids, and building materials as well as housing [4, 5].

In the heating film containing carbon materials as a conductive filler, the electrical resistances caused by the interconnection of a myriad of carbon particles generate the thermal energy when current is applied. The heating films prepared with carbon composites exhibit excellent electrical/thermal conductivity and the weatherability, and the research on the carbon-based heating technology has been actively conducted in the academic and industrial fields [6, 7]. A single-walled carbon nanotube (SWNT) has been used to prepare a transparent heating film, which showed heating characteristics with a transparency of over 90%. The heating films based on silver nanowires have been also reported [8–11]. However, this transparent heating film technology has limitations for industrial application due to expensive SWNT and Ag nanowires. In addition, such heating films have poor mechanical properties such as flexibility and brittleness. On

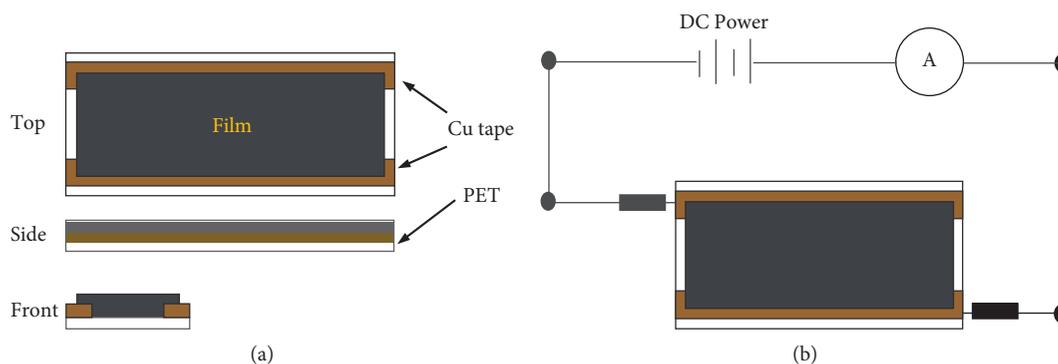


FIGURE 1: Schematics of (a) a heating film and (b) a circuit set for the heating measurement.

the other hand, the polymer composite film containing graphite as a conductive filler is opaque but its mechanical properties can be easily controlled by using various polymeric binders which are widely used in the material industry.

The properties of a heating film are dependent on the kind of polymeric binders and conductive fillers used. As electric current is applied to the heating film, thermal energy is generated on the film surface, in which lots of contact resistances are formed between the conductive particles dispersed in the polymer matrix of the heating film. The thermal energy generated from the heating film is considerably influenced by the polymer binder, the type, and the structure of thermal conductive materials used in the film and the morphology of the as-prepared film [12–16].

Poly(methyl methacrylate) (PMMA) has good weatherability and durability and is suitable for outdoor use [17, 18], and all kinds of processes such as heating, machining, and printing are easily applied to this polymer. Crystalline PMMA is good for heat transfer and is stable at high temperature when used as a binder of a heating film. Polybutadiene (PB) is a rubber material widely used for industrial purposes because it has low density and good weather resistance [19–21]. PB has low glass transition temperature (T_g) ($-20\sim-110^\circ\text{C}$), and it does not crack and is flexible at low temperatures [22]. When PB is used as a polymer binder, the heating films can be used for the pipe applications that require flexibility.

Carbon materials such as graphite, carbon nanotube, carbon fiber, and graphene have high conductivity and excellent mechanical properties and are applied in the fields where lightweight and high-performance composite materials are required. In general, thermally conductive ceramic materials are electrically insulated but graphite is excellent in electrical conductivity because it has π -electrons due to its planar sp^2 carbon-carbon bond structure [23–25]. In addition, graphite has high chemical stability and good workability, so it is easy for precision processing. Graphite is a good candidate as a conductive filler for the heating film because it is good in thermal resistance, thermal shock resistance, and thermal/electrical conductivity [26, 27].

In this study, composite films containing graphite were prepared by using general-purpose polymeric resins of PMMA and PB as binders and their heating characteristics

were analyzed. The temperature elevation of the film surface due to heat generation was analyzed by using an infrared thermal imaging camera while adjusting the graphite amount in composite films, film thickness, and applied voltage.

2. Experimental Section

2.1. Preparation of Composites and Heating Films. To use as binders for the preparation of polymer composites, poly(methyl methacrylate) (PMMA) ($\overline{M}_n = \sim 103000$) and polybutadiene (PB) ($\overline{M}_n = \sim 3,000$) were purchased from LG MMA and Aldrich, respectively. These polymer binders were used without further purification. Toluene (99.9+%, SK chemicals) was used for the solvent. Natural graphite was purchased from Graphite Týn (micronized natural graphite, $<20\ \mu\text{m}$). A PET film (thickness, $125\ \mu\text{m}$) was used as a substrate to which heating films were deposited and a copper tape (width $20\ \text{mm} \times$ thickness $50\ \mu\text{m}$) as the electrode of the heating film.

A composite using PMMA as a binder was prepared as follows: 18 g of PMMA was added to 40 g of toluene, and the mixture was stirred and sufficiently dissolved at 60°C . Then, a certain amount of graphite was added thereto at room temperature and mixed at 500 rpm for 2.5 min, 800 rpm for 5.5 min, and 1000 rpm for 2 min using a paste mixer (PM-500D, SUFLUX) to prepare the final PMMA composite. To prepare a composite using PB as a binder, a given amount of graphite was added to the mixture solution with 20 g of PB and 20 g of toluene and mixed at 500 rpm for 2.5 min and 1000 rpm for 2 min using a paste mixer. The amount of graphite was adjusted to 30, 40, and 50 wt.% for all of the composites regardless of binder species.

The schematics of a heating film is shown in Figure 1. A composite for preparing the heating film was coated on a PET film where a copper tape was attached. The as-coated film thickness was adjusted from 10 to $100\ \mu\text{m}$ using a bar coater. The as-made heating film was dried at room temperature for more than 5 h and further dried at 60°C for overnight.

2.2. Characterization. The thermal stability of the polymer binder used in this study was analyzed by using TGA (TGA N-1000, SCINCO) under an airflow of 40 cc/min, where a

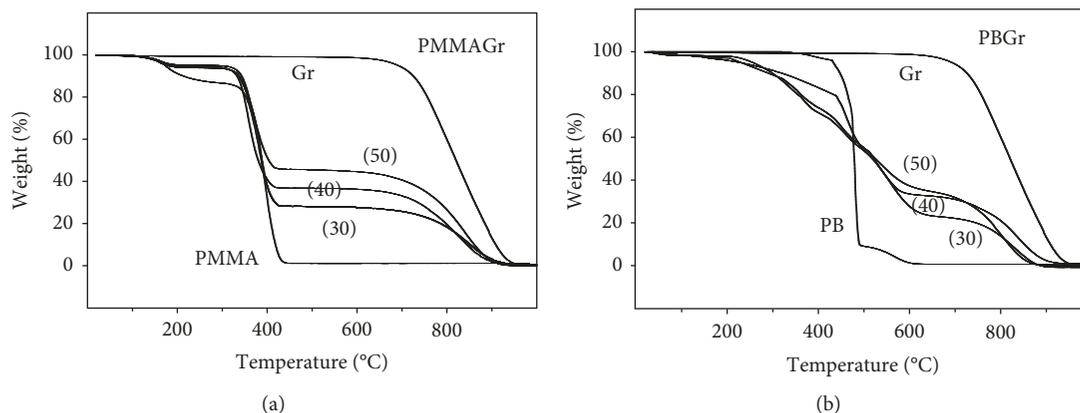


FIGURE 2: TGA thermograms of polymer/graphite composites used in this work. (a) PMMA/graphite composite films (PMMAGr) and (b) PB/graphite composite films (PBGr). The numbers enclosed in parentheses represent the graphite amounts in the composite films. PMMA, PB, and Gr indicate poly(methyl methacrylate), polybutadiene, and graphite, respectively. The measurement was conducted in an airflow of 40 cc/min. The heating rate was 5°C/min.

heating rate was set to 5°C/min. The microstructure of the composite film coated on a substrate was analyzed by using FE-SEM (Quanta 200FEG, operated at 15 kV). Film density was obtained by measuring the volume and mass of the film. The sheet resistance of the as-made heating film was measured by using a Loresta GP Resistivity Meter (MCP-T610, Mitsubishi Chemical Co.) connected to a 4-pin probe. The heating temperature of the film was measured using the setup shown in Figure 1(b). Both Cu electrodes of the heating film were connected to a power supply (GP-4303TP, EZ Digital) to generate thermal energy. A digital multimeter (MAS-830, Mastech) was connected in series to the circuit on the film surface to measure the current during the heating temperature analysis. The heating temperature of the film was analyzed by using an infrared thermal imaging camera (FLIR-C3TM, FLIR Systems), based on the principle of emitting the light of the corresponding wavelength according to the temperature of the subject. Thermal diffusivity (α) of the film deposited with the coating solution was measured by laser flash method (LFA-447, NETZSCH) at 25°C, and the thermal conductivity (λ) was calculated according to the following equation [28, 29].

$$\lambda(T) = \alpha(T) \times C_p(T) \times \rho(T). \quad (1)$$

Here, α (mm²/s), C_p (J/gK), and ρ (g/cm³) are thermal diffusivity, specific heat, and density, respectively. Thermal diffusivity was separately measured in out-of-plane and in-plane directions by using the sample modules for out-of-plane and in-plane analyses, which were provided by NETZSCH.

3. Results and Discussion

TGA was performed to study the thermal stability of poly(methyl methacrylate) (PMMA) and polybutadiene (PB) used as binders of the heating films prepared in this work as shown in Figure 2. The TGA measurement was conducted from room temperature to 800°C at a heating rate of

5°C/min and in an airflow of 40 cc/min. Oxygen in the air atmosphere promotes the oxidative reaction of PMMA, resulting in pyrolysis of the side functional group (-COOCH₃) at ~200°C, followed by decomposition of the main chain at ~350°C. PMMA was completely decomposed at 420°C or more, and PB at 600°C or more, leaving no residue. PB had higher thermal stability than PMMA in the air. This result shows that both PMMA and PB can be used as base resins, considering that the general service condition of the heating film is 100°C or less. Graphite is the material with good thermal stability, and it thermally decomposes at a much higher temperature than general polymeric resin such as PMMA or PB. Graphite began to decompose at a temperature above 600°C in air, which could be clearly distinguished from decomposition of PMMA and PB in the thermograms. The higher the amount of graphite in the composite was, the higher the decomposition curve in the region above 500°C was observed.

Cross-sectional microstructures of the composite films, which were prepared with PMMA (or PB) binder resin and graphite, were compared as shown in Figure 3. It could be seen that graphite particles were randomly arranged, as a whole, irrespective of polymer species. The PMMA composite with a graphite (PMMAGr) film appeared to have a relatively denser morphology than the PB composite with a graphite (PBGr) film. Graphite particles were also more exfoliated in PBGr than PMMAGr. The morphological images support the result that the sheet resistance of the PMMAGr film is relatively lower than that of the PBGr film. It will be further discussed in the following. When using more flexible and amorphous PB resin as a binder, the composite film is expected to have the less dense structure. In fact, the densities of PMMAGr films were higher than those of PBGr films (see Table 1).

Electrical resistance is induced by a large number of interfacial contacts formed between the graphite particles evenly dispersed in the polymer matrix. When voltage is applied to a heating film containing graphite, current flow generates the Joule heat, resulting in the temperature

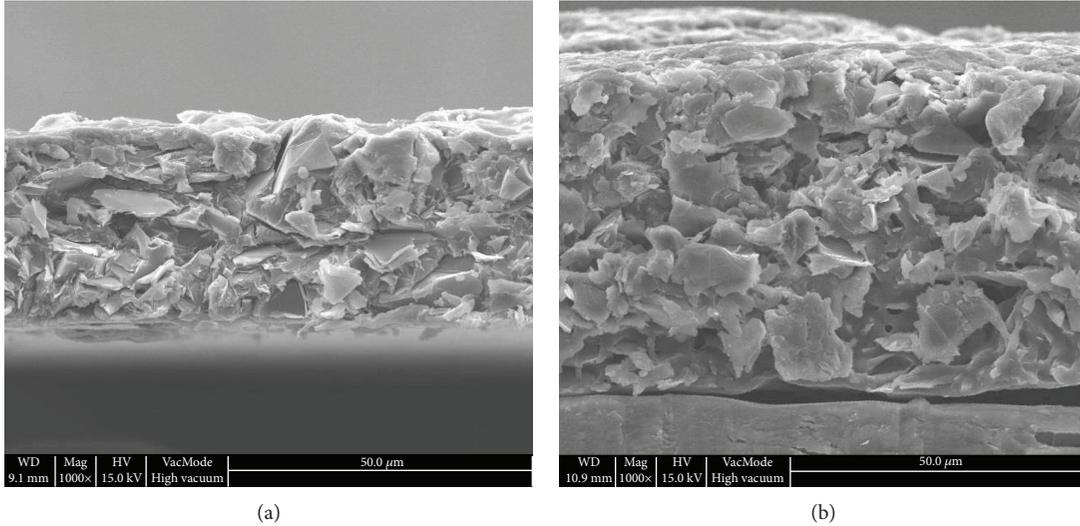


FIGURE 3: Cross-sectional FE-SEM images of (a) PMMAGr and (b) PBGr films containing 40 wt.% of graphite.

TABLE 1: Densities of PMMAGr and PBGr composite films.

Sample	Weight % of graphite in composite		
	30	40	50
PMMAGr	1.24	1.34	1.41
PBGr	1.16	1.12	0.95

elevation of the film [1]. If the film resistance is too low, the film shows the same characteristics as the conductor. On the contrary, if the resistance is too high, it becomes nonconductive. It is difficult to convert electric energy into thermal energy in either case. A suitable level of sheet resistance ($10^1 \sim 10^2 \Omega/\text{sq}$) is required for use as a heating film. Figure 4 shows the effect of the film thickness and the amount of graphite on the sheet resistance of heating films. The 4-pin probe measurement was used to obtain the sheet resistance of the heating film prepared by using the bar coater [30–32]. Regardless of the type of base resin (PMMA or PB), the sheet resistance of the heating films including graphite decreased as the content of graphite increased. That is, the conductivity of the film was correspondingly improved. This indicates that the PMMA composite with a graphite (PMMAGr) film is relatively advantageous in the conduction characteristics than the PB composite with a graphite (PBGr) film, considering the stereoregularity and the polarity effect of the PMMA resin itself [33, 34]. Equation (2) demonstrates that the sheet resistance (R_s) of the film is inversely proportional to the film thickness (t). Therefore, it can be easily understood that the sheet resistance decreases as the film becomes thicker.

$$R_s = \frac{1}{(\sigma_s t)}. \quad (2)$$

Here, σ_s is the in-plane conductivity. When 30 wt.% of graphite was added to both PMMAGr and PBGr, the sheet

resistance of the film was $10^2 \Omega/\text{sq}$ or more. As described above, it was difficult to obtain an adequate level of Joule heating characteristics in the film with high resistance.

Figure 5 shows the sheet resistance and the heating temperature measured on PMMAGr and PBGr films, which were fabricated in the dimension of $100 \mu\text{m} \times 25 \text{ cm} \times 14 \text{ cm}$ and analyzed under DC 25 V. As described above, increasing the amount of the graphite added to the film decreased the sheet resistance of the film (i.e., the conductivity of the film improved) and consequently increased the heating temperature. Under the same condition, the PMMAGr film showed lower resistance and higher heating temperature than the PBGr film. At the same content of graphite, the sheet resistance of the PBGr film was more than 10 times higher. The heating temperature was also higher in the PMMAGr film as the content of graphite increased: PBGr from 30°C to 50°C and PMMAGr from 32°C to 135°C . When 30 wt.% of the graphite was added to both films of PMMAGr and PBGr, the heating temperature was as low as 30°C but the heating temperature above 80°C was measured on the PMMAGr film containing 40 wt.% of graphite. The heating temperature was measured above 130°C on the PMMAGr containing 50 wt.% of graphite. In addition, the heating temperature of the films was stably maintained even under continuous use for over 7 days. The stable heating characteristics and the long-term reliability of the heating film are due to the good thermal stability of the base resin used. As shown in the TGA analysis, PMMA and PB exhibit thermal stability at the temperature below 200°C . Therefore, the heating film might exhibit a thermal stability below 200°C . The saturated temperature shown in the infrared thermal images was uniform within a deviation of 5°C in the whole region, indicating that the graphite particles were evenly distributed in the base resin. Although the film can be fabricated with the graphite content of 50 wt.% or more, it is difficult to keep the mechanical properties of the film such as flexibility and toughness, so that it is not recommended as a composite for heating film.

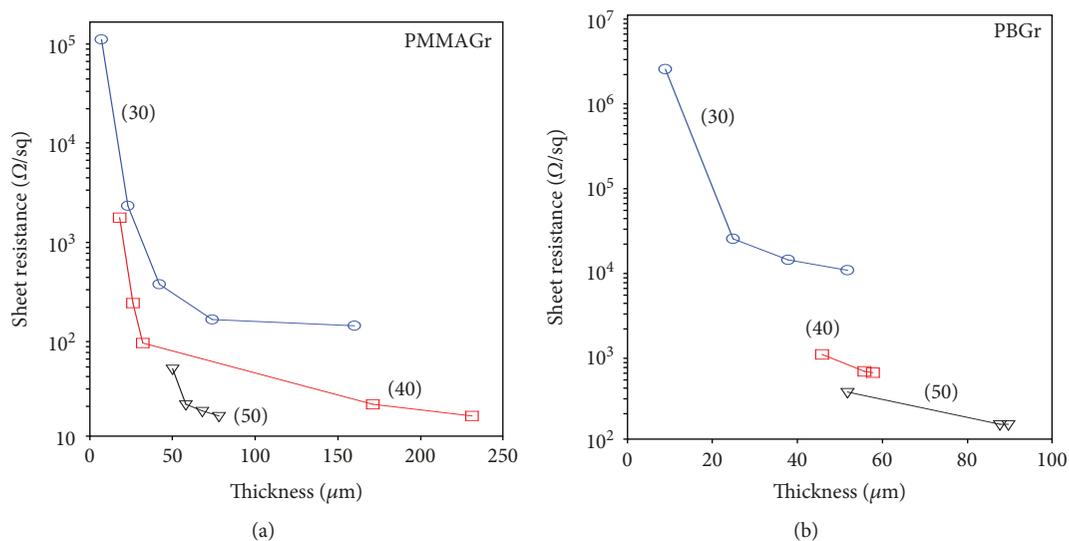


FIGURE 4: Sheet resistance of heating films. (a) PMMAGr and (b) PBGr. The numbers enclosed in parentheses represent the amounts of graphite added to the composite films.

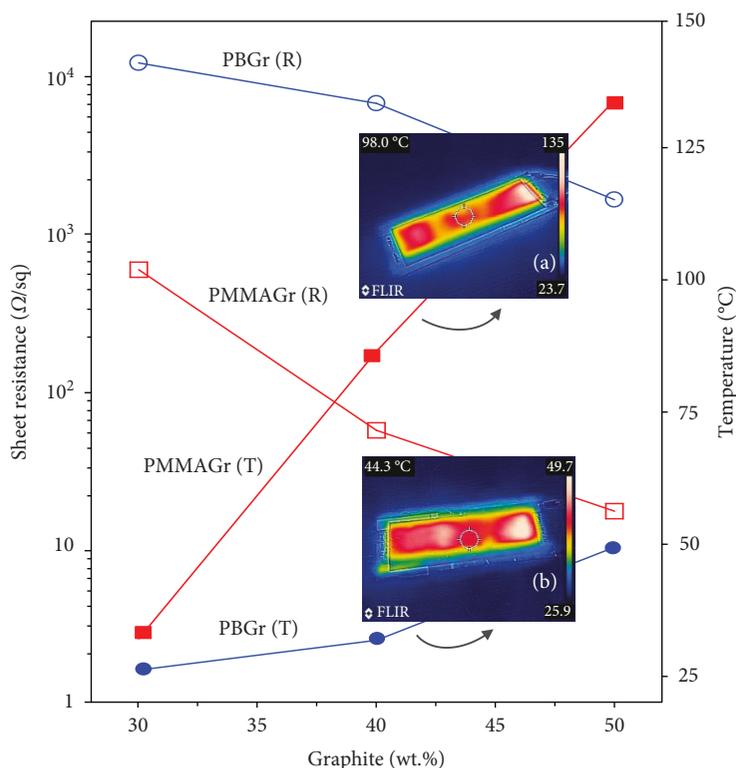


FIGURE 5: Sheet resistance and heating temperature as a function of the amount of graphite added to the heating film. Open symbols (\circ and \square) and filled symbols (\bullet and \blacksquare) represent the sheet resistance and the temperature measured in the film, respectively. The abbreviations in the figure represent as follows: PBGr: polybutadiene/graphite composite film; PMMAGr: poly(methyl methacrylate)/graphite composite film. (R) and (T) after the abbreviations mean the sheet resistance and the temperature, respectively. Infrared thermal images were taken from the heating films while applying DC power of 24 V; (a) PMMAGr and (b) PBGr films including 50 wt.% of graphite. Thickness of both films was about 100 μm .

The thermal conductivity of PMMAGr and PBGr films were analyzed by laser flash method (LFA 447, NETZSCH) according to the ASTM E2585 standard. In-plane thermal

conductivity (λ_{\parallel}) was measured in a wide range from 5 to 20 W/mK, depending on the amount of graphite and the polymer species, and out-of-plane thermal conductivity

TABLE 2: Thermal conductivities of PMMAGr and PBGr composite films.

Thermal conductivity (W/mK)	Sample					
	PMMAGr (wt.%)			PBGr (wt.%)		
	30	40	50	30	40	50
Out-of-plane	0.5	0.7	1.2	0.7	1.2	1.6
In-plane	5.8	13.1	19.9	5.5	6.0	6.4

“wt.%” in parentheses represents the weight percent of graphite in the composite film.

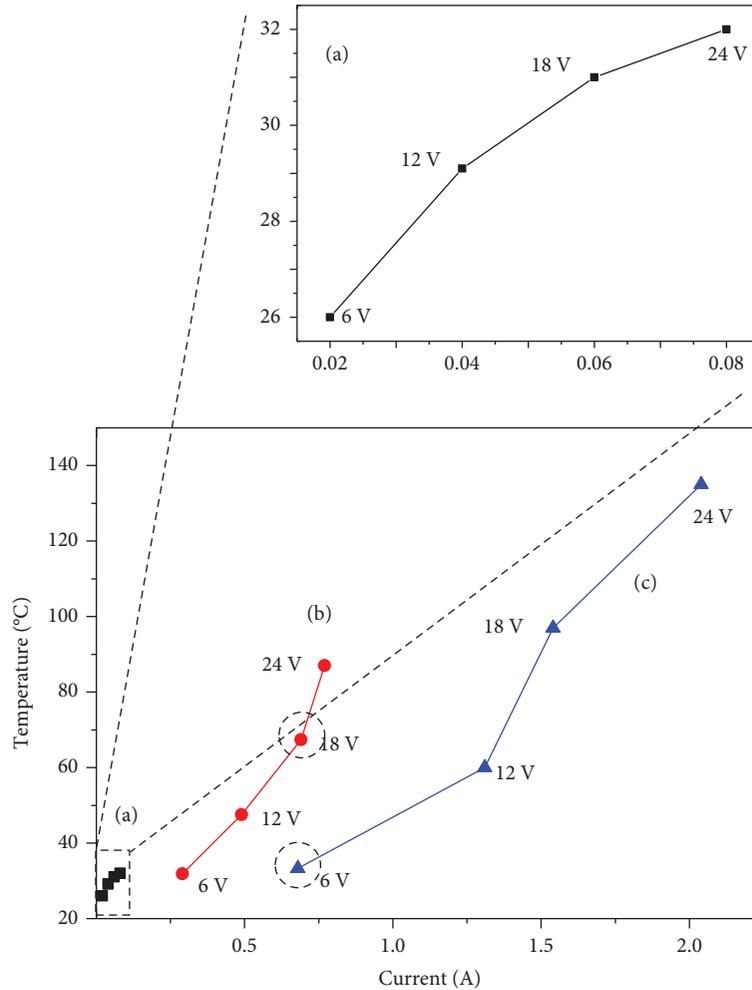


FIGURE 6: Heating temperature measured on the heating film under application of DC power; (a) PMMAGr-30, (b) PMMAGr-40, and (c) PMMAGr-50. The numbers -30, -40, and -50 indicate the graphite amount (wt.%) added to the composite films of $100\ \mu\text{m}$ thickness.

(λ_{\perp}) was about $1\ \text{W/mK}$ regardless of the film species (see Table 2). Considering the characteristic planar structure of graphite, λ_{\perp} of the film containing graphite was considerably lower than its λ_{\parallel} and was not significantly influenced by the amount of the thermal conductive fillers [35, 36]. λ_{\perp} of the PBGr film was relatively higher than that of the PMMAGr film. Thermal conductivity of the film can be explained based on the SEM image shown in Figure 3. Plate-like graphite particles tend to be oriented in the in-plane direction in the film status. This is why the in-plane thermal conductivity, as a whole, is larger than the out-of-plane

thermal conductivity. PMMAGr films with higher densities than PBGr films showed the higher in-plane thermal conductivity. In addition, the graphite particles of Figure 3(b) were randomly oriented in the film but were favorable to be oriented through the film thickness, compared to the graphite in Figure 3(a). Thus, it can be understood that the PBGr film has exhibited a slightly higher λ_{\perp} than the PMMAGr film. The higher the λ_{\parallel} , the higher the heat radiation and temperature rising effect (due to electrical resistance) transferred on the film surface, which is adaptable for use as a heating film.

According to the basic electricity theory, the Joule heating energy (Q) is dependent on the voltage (V), current (I), resistance (R), and time (t) during which the current flows:

$$Q \propto V \times I \times t = \frac{V^2}{R} \times t. \quad (3)$$

The current and the heating temperature measured while applying DC power to each PMMAGr film were summarized in Figure 6. As shown in equation (3), the thermal energy (Q) is proportional to V , I , and t applied to the film at a constant resistance condition. In addition, the thermal energy is proportional to V^2 and inversely proportional to R at a constant current condition. The results could be identified from the dotted circles in Figures 6(b) and 6(c). In the PMMAGr film including 50 wt.% of graphite, the temperature of the film was easily adjusted over a wide range from 32°C to 135°C as the voltage and the current were changed. The heating temperature did not exceed 32°C on the PMMAGr film including 30 wt.% of graphite even when the voltage increased to 24 V. This is because the resistance of the film itself is too high (over $10^2 \Omega/\text{sq}$) to allow current flow even when a voltage is applied.

4. Conclusions

Polymer composites containing graphite were prepared by using poly(methyl methacrylate) and polybutadiene resins with flexibility, durability, and weatherability. As-prepared heating films exhibited stable heating performance under continuous use condition of 200°C or less. The sheet resistance of the film decreased with the increase of the film thickness and the graphite content added to the film. When PMMA was used as a binder, its composite (PMMAGr) film exhibited relatively lower resistance and higher heating characteristics than the film (PBGr) using polybutadiene as a binder. The film requires the sheet resistance of less than $10^2 \Omega/\text{sq}$ to ensure proper heating characteristics. Depending on the type of the base resin and the content of graphite used in the film, the heating temperature could be adjusted up to about 130°C. An electronic controller that can control the input current should be used for more precise temperature control.

Data Availability

The data used to support the findings of this study are included within the article.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

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