Improvement of thermal propagation in carbon fiber/thermoplastic composite with hexagonal boron nitride powder

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Thermal propagation and diffusion in the carbon fiber (CF)/thermoplastic (TP) model composites, which was composed of a single CF and thin TP films, were investigated as a basic research of a rapid carbon fiber reinforced thermoplastic (CFRTP) molding by microwave irradiation. In case of the CF/TP model composite with thermally-conductive matrix prepared from polypropylene (PP) and 5 vol.% hexagonal boron nitride (h-BN) powder, the thermal propagation in the matrix was enhanced till 2.3 times as compared to one with PP of the conventional matrix of 0.15 W/(m·K). Its thermal propagation agreed with the one-dimensional heat equation. Moreover, the thermally-conductive matrix led to the suppression of thermal decomposition at the interface between CF and matrix in composites.

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1. Introduction

Among of composite materials, recently, carbon fiber reinforced plastics (CFRP) are attracting attention as light weight structural materials. Therefore, it is important for the transport equipment as automobile et al. to develop the novel method of CFRP manufacturing. Actually, from a viewpoint of mass production, thermoplastics (TP) are expected as a matrix of CFRTP for the automotive field, because the TP resin is able to perform molding easily by heating-melt.

Generally, CFRTP is pressed to fabricate various shapes after heating by an infrared heater. The heating system is complicated and the heating time is long. Therefore, the novel development of short time molding methods for CFRTP manufacturing has been expected at industrial fields. As novel manufacturing of CFRTP, microwave processing has recently been attracting attention because CFRTP is heated easily by microwave irradiation. It is known that carbon fiber (CF) in CFRTP selectively absorbs microwave and then electric charge generated in CF is easily converted into heat, resulting that CFRTP is heated rapidly from an internal body. Therefore, the microwave processing is expected for the rapid manufacturing of CFRTP. However, there are several problems on CFRTP manufacturing using a microwave processing. The uniform heating of CFRTP body using microwave is difficult. Moreover, the interface between CF and TP during microwave irradiation is decomposed by rapid heating of CF because the thermal conductivity of CF and TP is very different. In our previous study published in JCSJ, it was reported that the thermally-conductive TP with h-BN powder caused suppression of thermal degradation of the interface between CF and TP in CFRTP when irradiating microwave. We discussed that the suppression of thermal degradation was affected by the thermal conductivity of matrix in CFRTP. The release of heat accumulated at the interface between CF and TP was predicted. Moreover, in order to achieve a rapid molding technique of CFRTP using a microwave processing, it is important to heat TP in CFRTP homogeneously without thermal degradation. Therefore, it is necessary to study quantitatively the thermal diffusion from CF to TP.

In this paper, we will demonstrate how thermal propagation in CFRTP is quantitatively caused by matrix with different thermally-conductivity. Therefore, the model composite, which a single CF was sandwiched by TP films, was prepared in this work. During microwave irradiation, the thermal diffusion and propagation was observed by a thermo-viewer. We will show how thermal conductivity of matrix is contributed to CFRTP.

2. Experimental procedure

2.1 Materials

All materials were used as received without further purification. Carbon fiber (CF, HTA-12K, 7 μm average diameter) was obtained from TOHO TENAX Co., Ltd. Polypropylene (PP, Noblen Z144, Sumitomo Chemical Co., Ltd.) was used as TP matrix of CFRTP model composite. Hexagonal boron nitride (h-BN, UHP-1, 5 μm average diameter, Showa Denko K. K.) powder was used in this work.

2.2 Preparation procedure of CF/TP model composites

In order to prepare TP film with thermal conductivity, PP pellets and 5 vol.% h-BN powder were mixed at 180°C using an uniaxial kneading equipment (IMC-TAD3, Imoto Machinery Co., LTD.). The rotational speed of screw was adjusted to be 25 rpm. Similarly, PP pellets were kneaded to prepare the film by same screw speed. TP film was adjusted to be 0.25 mm thickness by
heat-pressing the composite, which was kneaded with PP and h-BN at 160°C (AH-10TD, AS ONE Corp.). After a single CF was sandwiched between the prepared TP films, the heat-press was performed at 160°C to be 0.5 mm of the film thickness. Furthermore, the sample was cut to be 15 mm of diameter. The above procedure is represented in Fig. 1.

2.3 Microwave irradiation

The microwave apparatus was used at 2.45 GHz frequency (FDU-201VP-07, Fuji Electronic Industrial Co., Ltd.). Microwave irradiation was carried out by using a single-mode, which can generate a standing wave in an applicator. Figure 1 shows the single-mode microwave applicator. The tuning of apparatus was performed as the top of microwave was hit continuously to the sample. The microwave power was adjusted at 0.1 W in order to suppress the damage of CF.

2.4 Characterization

In order to characterize the thermal diffusion and propagation of composite during microwave irradiation, a thermo-viewer was placed on top of a composite (Fig. 1). The surface texture and cross-section of the composites heated by microwave irradiation were observed using a digital camera (α65, SONY Corp.) and scanning electron microscope (SEM; S4300, HITACHI Ltd.). The disk-shaped TP samples with 3 mm thickness and 50 mm diameter were prepared for the thermal conductivity measurement of matrix by a press molding at 160°C. Its thermal conductivity was measured by a parallel plate method (HC-110, EKO Instruments Co., Ltd.).

3. Results

Figure 2 shows the relationship between temperature and distance from center of samples in (a) PP and (b) 5 vol. % h-BN/PP during microwave irradiation. The temperatures of both PP and h-BN/PP increased 5°C from 26 to 32°C with time. Moreover, its temperatures were almost constant in each samples.

![Fig. 1. Preparation procedure of CF/TP model composites and microwave irradiation system with a single-mode microwave applicator and a thermo-viewer.](image1)

![Fig. 2. Relationship between temperature and distance from center of samples in (a) PP and (b) 5 vol. % h-BN/PP during microwave irradiation.](image2)

![Fig. 3. Thermal images of (a) single CF/PP and (b) single CF/h-BN/PP composite heated by microwave irradiation.](image3)

The thermal conductivity of PP was 0.15 W/(m·K), whereas 5 vol. % h-BN/PP composite was 0.29 W/(m·K). By addition of h-BN powder to PP matrix, the thermal conductivity was enhanced to twice.

Figure 3 shows the thermal images of (a) single CF/PP and (b) single CF/h-BN/PP model composites heated by microwave irradiation, which was obtained by using a thermo-viewer. Matrix in the CF/h-BN/PP became extensively high temperature as compared to CF/PP because the heat of CF heated by microwave irradiation was released from matrix with high thermal conductivity.

Figure 4 shows the surface texture and cross-section of (a, c) CF/PP and (b, d) CF/h-BN/PP composites heated by microwave
irradiation. According to the surface texture image of the CF/PP composite, it was observed that the melting of matrix was caused along with the length direction of CF heated by microwave irradiation [Fig. 4(a)]. On the other hand, the matrix of the CF/h-BN/PP composite was not melted and decomposed by microwave irradiation [Fig. 4(b)]. From the cross-section image of the CF/PP composite, it was observed that the single CF in the composite was lost by microwave irradiation as shown in Fig. 4(c), indicating that CF would be burn out by rapid heating of microwave irradiation. Furthermore, the void with 200μm diameter was observed at the position of a CF in the composite, meaning that the thermal degradation was caused. This phenomenon was consistent with the result reported previous our study.3) On the other hand, in case of the CF/h-BN/PP composite with high thermally-conductive matrix, the thermal degradation of CF and matrix was not caused by microwave irradiation as shown in Figs. 4(b) and 4(d). It is important for microwave irradiation on CFRTP to enhance the thermal conductivity of matrix.

Figure 5 shows the relationship between temperature and distance from CF in case of (a) CF/PP and (b) CF/h-BN/PP composites heated by microwave irradiation. The position of a single CF is represented as 0 mm. The temperature at 0 mm position in microwave-irradiated CF/PP composite was drastically increased till 225°C for 20 s. The temperature at the ±7 mm position departed from the CF was increased from 25 to 37°C by microwave irradiation for 40 s. Moreover, the full width at half maximum (FWHM) of the temperature distribution was 0.9 mm after microwave irradiation of 6 s. On the other hand, in case of CF/h-BN/PP composite, the temperature at 0 mm drastically was increased by microwave irradiation for 6 s and was reached till 160°C for 20 s. The temperature at the position of ±7 mm was increased from 25 to 40°C by microwave irradiation for 40 s and was reached at 50°C by one for 140 s. Moreover, the FWHM of the sample which was microwave-irradiated at 6 s was 1.5 mm, indicating that the thermal propagation property of CF/h-BN/PP composite was 1.7 times excellent as compared to CF/PP one.

4. Discussion

As shown in Fig. 3, in case of the composite with high conductive matrix, the heat of CF irradiated by microwave was diffused into the matrix as compared to conventional resin matrix, meaning that the thermal propagation was enhanced into the matrix of CFRTP. Moreover, the improvement of thermal sticking at the interface between CF and matrix led to suppression of thermal degradation of matrix (Fig. 4). In fact, in case of the CF/PP with low thermally-conductive matrix, the temperature of sample microwave-irradiated for 40 s was reduced from 225 to 180°C, meaning that the degradation was caused as shown in Fig. 4. On the other hand, in case of the CF/h-BN/PP composite with high thermal-conductive matrix, the thermal degradation was not caused even if the irradiation time of 3.5 times was given to the composite. Thus, thermal-conductive matrix prepared from h-BN powder is very effective for suppression of thermal degradation in CFRTP because the heat of CF, which was generated by microwave irradiation, is propagated rapidly to matrix.

In order to discuss the property of thermal propagation in detail, the velocity of thermal propagation was estimated from the temperature as a function of distance from CF when irradiating microwave. The velocity of thermal propagation is represented as a function of distance from CF in Fig. 6. Moreover, the theoretical fitting curve which is estimated from the one-dimensional heat equation15) was added in Fig. 6. The experimental data shows that the velocity on thermal propagation was reduced as the distance of CF. The tendency of behavior was identical with theoretical curve estimated from the one-dimensional heat equation and the both of experimental data and theoretical fitting.
curve were almost similar, meaning that the thermal propagation from CF heated by microwave irradiation was performed to the vertical direction of CF in based on thermal conductive theory.

5. Conclusion

In this paper, we demonstrated the thermal propagation using single CF/PP model composite which was irradiated by microwave. It was found that the thermal propagation on the high thermally-conductive matrix with h-BN was enhanced in composite as compared to the conventional matrix. The behavior of thermal propagation was based on thermal conductive theory. Moreover, in case of the composite with high thermally-conductive matrix, the thermal degradation was not caused when irradiating microwave. The effect of thermal propagation from CF heated must lead to the suppression of thermal degradation.

One important consequence of the present work is that the improvement of thermal conductivity of matrix will become very effective for CFRTP manufacturing utilizing microwave processing from viewpoints of matrix degradation and rapid molding.

Therefore, ceramic powder like h-BN which has high thermal conductive property will become one of important materials for CFRTP manufacturing.

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