Properties of graphite blocks consisting of ordered graphite flakes with bimodal particle size distribution

Moon H. Lee1,2 | Byung C. Kim1 | Ho Y. Kim3 | Euigyung Jeong2 | Sooyeon Jeong4 | Kwang S. Park1 | Joong T. Han4 | Jong S. Woo1

1. Advanced Center of Engineering, Morgan Advanced Materials, Dae-gu, South Korea
2. Department of Textile System Engineering, Kyungpook National University, Dae-gu, South Korea
3. Changwon Industry Promotion Agency, Changwon, South Korea
4. Nano Hybrid Technology Research Center, Korea Electrotechnology Research Institute, Changwon, South Korea

Correspondence
Jong S. Woo, Advanced Center of Engineering, Morgan Advanced Materials, 23, Dalseong2cha 4-ro, Guji-myeon, Dalseong-gun, Dae-gu 43013, South Korea.
Email: justinjongsok.woo@morganplc.com

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Abstract
Petroleum pitch-based graphite blocks (GBs) attract considerable attention owing to their low cost, high yield, and possible application as heat sink components. However, no studies have systematically investigated the particle size distribution of GBs, their structures, and the relationship between the structures and thermal properties resulting from graphitization. Hence, herein, we present a novel method for fabricating petroleum pitch-based GBs using natural graphite flakes (GFs) of sizes ranging between 50 and 500 μm and a bimodal particle size distribution. Furthermore, the effects of the mean GF particle size, pitch binder content, and microstructure of the GBs with low GF contents on compression molding with elevated hot-pressing temperatures after graphitization are investigated. To investigate the effect of packing density with a GF size of 500 μm, we mix a small-sized GF (50 μm) and low pitch binder content by kneading. In particular, specimens in the in-plane direction with a bimodal particle size distribution of GFs and 10 wt% pitch binder yield optimum density and thermal conductivity values of 1.74 g m⁻³ and 384 W m⁻¹ K⁻¹, respectively, after high-temperature graphitization at 2600°C. These findings provide insights for developing high-performance GBs for use in heat sink components with high thermal conductivity.

KEYWORDS
compression molding, graphite blocks, graphite flake, graphitization, petroleum pitch

1 INTRODUCTION
Numerous studies have reported that carbon particle formation of arrayed structures in graphite blocks (GBs) enhances thermal and mechanical properties, thus making GBs applicable as heat sink components in electronic devices and automobiles.1,2 The rapid development of technology has necessitated smaller, lightweight, and highly efficient electronic devices. This miniaturization of devices requires heat sink parts with high thermal conductivity and excellent mechanical properties.3-5 Carbon-based materials such as graphite,6 graphene,7 carbon nanotubes (CNTs),8 and carbon fibers9 are ideal fillers for GBs owing to their exceptional electrical and thermal conductivity, which overcome the limitations of other binder materials. However, several of these materials exhibit certain limitations such as the relatively high price of
high thermally conductive carbon fibers and CNTs and the nonuniform distribution of graphite layers in the GBs. The aforementioned disadvantages hinder the commercialization of these materials.

Natural graphite flakes (GFs) have extremely high electrical and thermal conductivities along the direction parallel to the graphite layers and are conventionally utilized as refractory materials and additive fillers for composites owing to their low cost, lightweight, and high thermal conductivity.\textsuperscript{10-15} GBs are traditionally fabricated from binder coal tar or petroleum pitch, and coke filler is added via cold isostatic pressing through kneading or extrusion, followed by several impregnation processes, pyrolysis, and high-temperature graphitization (>2500°C). GBs have been utilized as electrode materials for the metallurgical industry, earths for blast furnaces, electric brushes and high-speed train pantograph slides, and heat exchangers and sinks for high-power electronic devices. These practical applications, which are associated with high-temperature performance, effective thermal management, low coefficients of thermal expansion (CTE), and high-friction properties, have attracted continuous attention.\textsuperscript{16,17}

In particular, petroleum pitch-based GBs have attracted significant attention owing to their high yield and low cost. Consequently, high thermally conductive GBs have been widely employed as heat sink materials. Petroleum-based pitch binders are generally classified as pyrolysis fuel oil (PFO), fluid catalytic cracking decant oil, vacuum residue, and deasphalted oil, depending on the manufacturing process. As a byproduct of the naphtha cracking process, PFO slurry oil has a low concentration of heterogeneous elements such as sulfur, nitrogen, and metals.\textsuperscript{18-21} However, the thermal conductivity of common GBs produced using pitch binder and coke powders is as low as 70–150 Wm\textsuperscript{−1} K\textsuperscript{−1}, indicating considerable potential for the development of GBs with higher thermal conductivity and improved mechanical properties.\textsuperscript{12-23}

Numerous factors determine the thermal and mechanical properties of GBs, including the type of pitch binder, the carbon particle size, and the method of formation. Yuan et al. reported GBs fabricated from natural GFs and mesophase pitch via a hot-pressing procedure using the pitch binder. When the pitch content was 14 wt%, the thermal conductivity and flexural strength were 522 Wm\textsuperscript{−1} K\textsuperscript{−1} and 7.7 MPa, respectively.\textsuperscript{24} Gao et al. fabricated natural GFs (495 μm) and spinning pitch using a hot-pressing technique (500°C, 10 MPa). Maximum thermal conductivity (485 Wm\textsuperscript{−1} K\textsuperscript{−1}) was achieved with 16 wt% pitch content in the direction perpendicular to the hot-pressing direction of graphitization.\textsuperscript{25} Zhao et al. prepared short-fiber-reinforced GBs by mixing natural GFs (size: 270 μm), mesophase pitch, and short carbon fibers, followed by hot-pressing to improve the mechanical properties. As a result, the flexural and compressive strengths of the GBs improved to 39.6 and 65.5 MPa, respectively.\textsuperscript{17} However, the arrangement of the pitch binder and various-sized GFs with bimodal size distribution of GBs, evaluation of the structure, and relationship between the structure and thermal properties resulting from graphitization have not been systematically investigated.

Therefore, in this study, we aimed to improve the thermal conductivity of GBs using compression molding with kneading, utilizing various-sized GFs and various concentrations of pitch binders. Furthermore, we investigated the effect of the bimodal particle size distribution of GFs and hot-pressing temperatures on the properties and microstructure of the graphitized GBs. In addition, correlations between the effect of the microstructure of the graphitized GBs and the network pathways between the pitch binder and the small-sized GFs were also studied.

## EXPERIMENTAL SECTION/METHODS

### 2.1 Materials

PFO, manufactured using a mild thermal reaction pilot-scale unit (900°C, heating rate: 10°C min\textsuperscript{−1}, Korea Research Institute of Chemical Technology), was used to fabricate the pitch binder (softening point: 156.5°C, carbon yield: ∼38.3 wt%), as shown in Table S1. Natural GFs with average particle diameters of 50 μm (#3557, Asbury Carbons, USA) and 500 μm (#3763, Asbury Carbons, USA) were purchased and used without further purification. The corresponding scanning electron microscopy (SEM) images and GF particle size distributions are shown in Figures S1 and S2(A), respectively. Table 1 shows the elemental analysis of the petroleum-based pitch binders. The petroleum-based pitch binders were synthesized from PFO because it has fewer impurities, such as sulfur, nitrogen, and metal elements.

| C     | H     | N     | S     | O     | C/H ratio (%) |
|-------|-------|-------|-------|-------|---------------|
| 91.66 | 5.92  | –     | –     | 2.42  | 19.54         |

**Table 1** Elemental analysis of petroleum-based pitch binders
The X-ray diffraction (XRD) pattern of the pure GF powders revealed that the $L_a$ and $L_c$ values were similar based on the graphite particle size, as shown in Figure S2(B) and Table S2. $L_c$ and $L_a$ were calculated using the following equations:

$$L_c = \frac{0.89 \lambda}{(\beta_{002} \cos \theta_{002})}, \quad (1)$$

$$L_a = \frac{1.84 \lambda}{(\beta_{100} \cos \theta_{100})}, \quad (2)$$

where $\lambda$ is the wavelength of the X-ray, and $\beta$ and $\theta$ are the full width at half maximum and Bragg angles of the diffraction peak, respectively.

## 2.2 Fabrication of GBs with pitch and natural GFs by compression molding

A comparatively homogeneous distribution of the raw materials was achieved by micronizing the pitch binder using the pitch lump in a household mixer. Uniformly mixed pitch/GF mixtures with different pitch contents (5, 10, 15, and 20 wt%) and varying GF sizes (50 and 500 $\mu$m) and contents (10, 15, 20, and 25 wt%) were prepared using a kneader (10 rpm, 1 h, 180°C, PN-1, Shokai Co., Japan) in Figure S3. Then, the mixture powders were crushed using an electrical pulverizer (5000 rpm, 30 s, Tube Mill 100 control, IKA, Germany) and fabricated with different GFs and pitch binder weight ratios, into compacted green blocks (dimensions: 60 × 60 × 7 mm) using compression molding under 30 MPa (1 h, HT-10 T, ILSHIN Autoclave, Korea). The hot-pressing temperature was increased from 25 to 400°C. The schematic of the basic process of the GBs is shown in Scheme 1. Carbonization treatment was performed at a heating rate of 4°C min$^{-1}$ to ∼1400°C to inhibit the rapid release of volatiles and thereby reduce the delamination or cracking of the unidirectional laminates. The carbonized blocks were subsequently graphitized under an argon atmosphere, inside a furnace that was heated to 2600°C at a heating rate of 5°C min$^{-1}$.

## 2.3 Characterization

Elemental analysis of the pitch binder was performed to confirm the C, H, N, and S elemental composition and C/H ratio using an organic elemental analyzer (EA, Flash EA 2000, Thermo Scientific, USA). The particle size distribution in pristine natural GF was measured via laser scanning with a particle size analyzer (Matersizer 2000, Malvern, UK). The XRD (PW 3830, Philips, NL) patterns, with the scanning angles ranging from 10° to 60°, were obtained using an X-ray diffractometer with Cu Kα radiation ($\lambda = 1.5418$ Å) to determine the primary structure and parameters of the GF crystal: $d_{002}$ and $L_c$, where $d_{002}$ is the graphite interlayer distance, and $L_c$ is the average crystallite diameter. The density and porosity of the GBs were determined by measuring the dried ($W_1$), saturated ($W_2$), and suspended ($W_3$) weights using Archimedes’ method.

**Scheme 1** Illustration of pitch/graphite flake powders mixing by kneading and the formation of graphite blocks using compression molding
Density \((\text{g cm}^{-1}) = \frac{W_1}{W_2 - W_3}\)  

Porosity \((\%) = \frac{W_2 - W_1}{W_2 - W_3}\)

The fracture and polished surfaces of the GBs were observed using field-emission SEM (FE-SEM; S-4800, Hitachi, Japan) and optical microscopy (EPIPHOT 200, Nikon, Japan), respectively. The thermal conductivity of the samples (size: 10 × 10 mm, thickness: 2 mm) was evaluated using a thermal conductivity meter (Netzsch, LFA 457, Germany) through a laser flash method. The 3-point flexural strength of the specimens was measured perpendicular to the compression direction using a universal testing machine (UTM; DEC-M200KC, Dawha Testing Machine, Korea). Each specimen was cut into 50 × 4 mm with a thickness of 3 mm according to the international standard ASTM 790. The average flexural strength was calculated based on the results of five tests. The CTE was measured by thermomechanical analysis (TAg Instruments Q400, USA) within the temperature range of 25–1000°C in an argon atmosphere at a heating rate of 10°C min\(^{-1}\) using a cubic specimen with dimensions of 5 × 5 × 5 mm. The CTE values of the specimen were measured in directions perpendicular (in-plane direction) and parallel (through-plane direction) to the applied pressure.

3 | RESULTS AND DISCUSSION

3.1 | Preparation of GBs with various GF sizes by compression molding

We propose a novel approach that uses compression molding through a kneading process to prepare different pitch binder contents and GBs consisting of large-sized GFs that are isolated small-sized GFs after graphitization. As shown in Scheme 1, the kneading process was conducted at 180°C, which is higher than the softening temperature of pitch (156.5°C). To overcome dispersion, a high-energy kneading process was used to prepare the pitch binder and GF mixtures prior to compression molding. This method is cost-effective and widely used in mixture powders to produce common GBs. In particular, the shear force can wrap GFs with molten pitch through the kneading process, while the compressive force can orient them perpendicular to the compression direction.

Prior to investigating the GBs containing a bimodal particle size distribution of various GF sizes, we investigated the GF sizes based on the pitch binder contents. It is essential to control the content of the pitch binder to achieve compact GBs with relatively high density and low porosity after high-temperature heat treatment (graphitization). Table 2 shows the density and porosity of the GBs as functions of the pitch binder content with various-sized GFs. The density was inversely proportional to pitch content, but the porosity was proportional to the pitch content. Over the range of GF sizes and pitch binder contents studied, the density of the GBs decreased as pitch content increased and particle size decreased. The effects of GF size on the density and porosity of the GBs can be explained from two perspectives. First, the GF was more isolated in the GBs under an optimum pitch binder content, thereby improving density and porosity. Second, poor dispersion favored the agglomeration of GF sizes based on the pitch binder content; thus, small-sized GFs with large specific surface areas decreased the density and porosity compared with large-sized GF particles. Moreover, the addition of pitch can only increase the content of the binder phase in the resulting GBs, which improves the mechanical strength of the GBs. However, both cases with different GF particle sizes exhibited swelling of GBs at a pitch binder content of 15 wt% after graphitization.

Compression molding is an important factor impacting the microstructure and performance of the GBs; the samples prepared using different pitch contents at 2600°C were investigated. The fracture surfaces of the GBs with different GF contents and particle sizes were observed by SEM, as shown in Figure 1. In this study, the micropores in the GBs were generated from two sources: the original voids among the GFs and the pores in the pitch residue formed by pitch decomposition at high temperatures. The former and latter decreased and increased with pitch additions, respectively. When the two factors work together, porosity tends to fluctuate, which explains the sudden reduction in open porosity depending on the pitch content and process parameter. The SEM images of the cross-sections parallel to the compression direction reveal that most GFs with large-sized particles are stacked parallel to each other to form a highly oriented structure, as shown in Figure 1(D–F), suggesting that the pitch binder fills the interspaces of the GFs and forms a conducting link between the GFs. However, analysis of the small-sized GFs revealed that several voids occurred due to the aggregated pitch
TABLE 2  Properties of graphite blocks according to the pitch binder contents with various-sized natural graphite flakes

| GF sizes (μm) | Pitch binder content (wt%) | Density (g cm\(^{-3}\)) | Carbonization (°C) | Graphitization (°C) |
|---------------|---------------------------|-------------------------|--------------------|--------------------|
|               | After compression molding | Density (g cm\(^{-3}\)) | Porosity (%)       | Density (g cm\(^{-3}\)) | Porosity (%) |
| 50            | 5                         | 2.016                   | 1.689              | 19.87              | 1.575       | 20.91       |
|               | 10                        | 1.939                   | 1.664              | 21.88              | 1.486       | 24.28       |
|               | 15                        | 1.901                   | 1.584              | 24.39              | 1.397       | 29.67       |
|               | 20                        | 1.873                   | 1.445              | 29.85              | 1.188       | 36.89       |
| 500           | 5                         | 2.100                   | 1.807              | 13.89              | 1.754       | 15.40       |
|               | 10                        | 1.989                   | 1.759              | 17.42              | 1.652       | 19.54       |
|               | 15                        | 1.948                   | 1.689              | 19.38              | 1.487       | 23.52       |
|               | 20                        | 1.882                   | 1.547              | 24.90              | 1.207       | 32.87       |

FIGURE 1  Typical microstructure after polished surface graphite blocks with graphite flake sizes of (A–C) 50 μm and (D–F) 500 μm at GF compositions of: (i) 5 wt%, (ii) 10 wt%, and (iii) 15 wt% via scanning electron microscopy (Inset: schematic of various-sized GFs with the pitch binder after compression molding)

binder after graphitization, as shown in Figure 1(A–C). As the pitch binder content increased, additional molten pitch entered the voids between the GFs under pressure in the compression molding, thus increasing the distance between the nonoriented crystalline and major planes of the GFs.

3.2  | Effect of the bimodal particle size distribution of GFs

The main objective of this study is to investigate the effect of the mean size of the GFs, according to the pitch binder content and mixed small-sized GF contents, on the density and thermal conductivity of GBs after high-temperature heat treatment. The effectiveness of incorporating different GF sizes into the GBs depends on their dispersion and contents.
Moreover, the dispersion of GFs is important because isolated small-sized GFs functioning as secondary fillers connect the highly conducting large-sized GFs in GBs.

First, we systemically identified the process parameters of the GBs with mixed small-sized GFs using raised compression molding. The hot-forming process was performed at temperatures ranging from 25 to 400°C. The density of the GBs ranged from 1.72 to 1.85 g cm\(^{-3}\) after graphitization, as shown in Figure S4(A). The fracture surfaces of the GBs hot-pressed at different temperatures were observed by SEM and optical microscopy, as shown in Figures S4(B–D) and S5. Upon increasing the molding temperature, the GFs of the filler and pitch binder became closer to each other, leading to denser and more compact GBs. The density and properties of the GBs could be improved using a high compression molding temperature, but the equipment parameter was limited.

Theoretically, two particle packing models occur, that is, loosening and wall effects, with different particle sizes, by bimodal particle size distribution systems.\(^{26-28}\) The loosening effect occurs when the coarse spherical graphite is dominant, and the fine spherical graphite is not sufficiently small to fit into the voids along with the coarse spherical graphite without distorting the skeleton of the coarse particles. By contrast, the wall effect occurs when the fine spherical graphite is dominant and regularly packed. This results in the formation of additional voids in the vicinity of the coarse spherical graphite surfaces. The packing density of the GF mixture powders is assumed to have binary size distribution, platelet shapes that are perfectly circular, equal density and aspect ratio of the spherical graphite, and noninteraction between the spherical graphite. The volume fractions of the fine and coarse spherical graphite are \(\phi_f\) and \(\phi_c\), respectively. Moreover, \(\phi_v\) is the volume fraction of void in the graphite mixture. The packing densities of the fine and spherical graphite are \(\alpha_f\) and \(\alpha_c\), respectively. The packing density (\(\rho\)) of the mixture is expressed as follows:

\[
\rho = \phi_f + \phi_c + \phi_v.
\]

(5)

When the mixture is rich in coarse spherical graphite, the fine spherical graphite is located in the voids of the coarse spherical graphite. Hence, the remaining space that can be occupied by the fine spherical graphite is \(1 - \phi_c\), which was considered by the loosening effect. Therefore, the packing density with rich-coarse spherical graphite (\(\rho_c\)) is obtained from the following equation\(^{29,30}\):

\[
\rho_c = \alpha_c + f \cdot \phi_f,
\]

(6)

where \(f\) is related to the loosening effect. Parameter \(f\) is equivalent to 1 when \(r_f \ll r_c\) and to 0 when \(r_f = r_c\). When the fine spherical graphite is rich in the mixture, the coarse spherical graphite surrounds it. The fine spherical graphite particles and the packing density are given by

\[
\rho_f = \alpha_f + \phi_c \cdot (1 - \alpha_f) \ g.
\]

(7)

where \(g\) is related to the wall effect and is determined by \(g = 1 - r_f/r_c\).

First, we calculated the packing density of the GF mixtures with particle sizes ranging between 50 and 500 \(\mu\)m, resulting in the highest packing fraction (0.95) at small-sized GFs and 22 wt% GF. In terms of the GBs, if the space without GFs is completely filled by pitch binders, the bulk density is calculated by adding the packing fraction of the theoretical density of the natural GF (2.24 g cm\(^{-3}\)) and pitch binder (1.05 g cm\(^{-3}\)).\(^{31}\) The results of the bulk density of bimodal particle size distribution with the pitch binder as a function of various small-sized GF contents are shown in Figure 2. The experimental and theoretical density curves increase and decrease before and suddenly after the transition point, respectively. Compared with the predicted theoretical density, the bulk density of small-sized GFs was lower owing to the presence of more voids from the randomly oriented GFs. The bulk density was improved using a bimodal particle size distribution of coarse and fine GFs, and 20 wt% small-sized GF yielded the highest bulk density (2.043 g cm\(^{-3}\)). This is ascribed to the excellent packing ability of the mixture of coarse and fine GFs.

Table 3 lists the physical properties of GBs with various pitch contents and a constant GF ratio (i.e., 50:500 \(\mu\)m = 80:20) fabricated using the same mixing and compression molding. The density of the GBs generally decreases with the content of the pitch binder, both before and after graphitization, and the porosity increases with the pitch binder content. This is because the lower molecular weight of the pitch binder decomposes with the heat treatment resulting in the generation of gaseous species that produce porous structures within GBs. Small-sized GFs (50 \(\mu\)m) in the mixture are expected to improve the packing density and act as connecting pathways between the large-sized GFs (500 \(\mu\)m) of the GBs, thereby increasing the density and decreasing the porosity compared with the intrinsic small- and large-sized GFs.
To determine the mechanism underlying the aforementioned improvement, the effects of the network pathways and bimodal particle size distribution of GFs on the thermal and mechanical properties were systematically analyzed. The thermal conductivity of GBs strongly correlates with the orientation and dispersion of GFs. We analyzed the thermal conductivity and flexural strength as a function of the pitch binder content of the GBs after heat treatment, as shown in Figure 3. The optimum pitch binder content strikes a balance between thermal conductivity and flexural strength. In particular, the apparent density and thermal conductivity in the in-plane (through-plane) direction with a bimodal particle size distribution of GBs with 10 wt% of pitch binder content achieved optimum values, that is, a thermal conductivity of 384 Wm$^{-1}$ K$^{-1}$ (21 Wm$^{-1}$ K$^{-1}$) and flexural strength of 7 MPa after high-temperature graphitization at 2600$^\circ$C. Moreover, we measured the CTE of GBs in two directions (in-plane and through-plane) according to the pitch binder contents as shown in Figure S7. Owing to the high orientation of GFs, the in-plane CTE values decreased by 12 times compared with those in the through-plane direction. The CTE values are close (nearly equal) in all directions in isotropic GBs, whereas in this case, the GFs have anisotropic CTE values.

To better understand the effect of the pitch binder content on the properties of the GBs, the fracture morphology on polished surfaces of the samples was investigated (Figures 4 and S6). The morphology, particularly the dispersion of particles, may provide insights into the physical properties of the GBs. Figure 4(A) shows that some voids (∼30 μm) occurred at the pitch binder (5 wt%) after graphitization because it could not wrap as a binder between large-sized and small-sized GFs. However, at a pitch binder content of 15 wt%, several large voids (∼70 μm) occurred due to the aggregated pitch binder after graphitization, as shown in Figure 4(B). As the pitch binder content increased, additional molten pitch entered the voids between the small-sized and large-sized GFs under the compression molding pressure, which

3.3 Physical properties and microstructure of bimodal particle size distribution in GBs

![FIGURE 2](A) Schematic of bimodal particle size distribution with various-sized graphite flakes. (B) Density of graphite blocks as a function of the small-sized graphite flake contents

![TABLE 3](Properties of graphite blocks with various pitch binder contents after graphitization)

| Graphite contents (wt%) | After compression molding | Graphitization (°C) |
|-------------------------|---------------------------|---------------------|
|                         | Density (g cm$^{-3}$)     | Density (g cm$^{-3}$) | Porosity (%) |
| 50 μm 500 μm             |                           |                     |
| 20 80                    | 2.133                     | 1.874               | 12.84        |
| 10                       | 2.050                     | 1.743               | 17.00        |
| 15                       | 1.8445                    | 1.571               | 23.37        |

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**FIGURE 3** Thermal conductivity and flexural strength of the graphitized blocks as functions of the pitch binder content (i.e., 50:500 μm = 80:20)

**FIGURE 4** Scanning electron microscopy images of the polished fracture surfaces of graphite blocks based on the pitch binder contents: (A) 5 wt%, (B) 15 wt%, (C) 10 wt%, and (D) high magnification of (C)

results in the nonoriented crystalline planes being away from the major planes of the GFs. Smooth surfaces and no microcracks or large holes are observed in the plane perpendicular to the compression direction, indicating that the GFs were well-wrapped and connected by the pitch binder. In particular, the small-sized GF particles would positively impact the interparticle conduction, enhancing the overall thermal conductivity and flexural strength (Figure 4(C,D)).

## 4 | CONCLUSION

We investigated GBs according to the pitch binder content and mean size of GFs through kneading, compression molding, and high-temperature heat treatment. In particular, the GBs were compared with investigate the correlations between the
effect of pitch binder contents and bimodal particle size distribution based on small-sized GFs and the microstructure of the GBs on the density and physical properties. Graphitized GBs achieved optimum thermal conductivity (384 Wm\(^{-1}\) K\(^{-1}\)) and flexural strength (7 MPa) with small-sized GF and pitch binder contents of 20 and 10 wt\%, and carbonization and graphitization temperatures of \(\sim 1400\) and \(\sim 2600\)°C, respectively, and compression molding at elevated temperatures. The proposed method is commercially attractive because it is cost-effective and can be used to fabricate GBs that are applicable as heat sink components with high thermal conductivity.

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ORCID
Jong S. Woo [https://orcid.org/0000-0002-5867-1881](https://orcid.org/0000-0002-5867-1881)

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