Fabrication and Simulation of a Layered Ultrahigh Thermally Conductive Material of Lamellar Stacking Graphene and Polydopamine on an Aluminum Substrate

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ABSTRACT: As technology continues to develop, electronic devices are becoming ever more integrated. The high level of integration results in a higher volume of calculations and higher heat generation. Metal materials have always been good conductors of heat and are commonly used in thermally conductive devices. However, the thermal conductivity of metallic materials decreases at elevated temperatures. Therefore, it is reasonable to develop new composite materials as thermal conductivity materials. In the experiments, a novel composite material with a sandwich structure has been designed. The material uses metallic aluminum (Al) as a substrate. Then, the metallic aluminum was soaked in a polydopamine (PDA) solution. Graphene (G) on the surface of the material was then enriched using an electrophoretic method. The material was removed and annealed to form the G-PDA-Al composite. According to the measurement, the thermal conductivity of the material is 492 W·m⁻¹·K⁻¹, which means ultrahigh thermal conductivity. Elongation experiments were carried out, and they increased the strength of the material by 12.4%. The formation of the material was then analyzed. The construction of the material was then carefully examined. The surface morphology, elemental composition, and structures were investigated by using scanning electron microscopy equipped by a scanning electron microscope, X-ray diffraction, infrared spectroscopy, and X-ray photoelectron spectroscopy. Differences from ordinary thermal materials were obtained based on calculations. A flexible thermal conductor was fabricated by using this material. The device can reduce the spontaneous combustion of ternary lithium batteries.

1. INTRODUCTION

Metals have always been good conductors of heat. The use of electrons in metals allows for the rapid transfer of heat. As the technology of batteries and circuits gradually advances, the heat dissipation materials previously used in thermal management are no longer adequate.1 Excessive temperatures can desolder electronic components, reduce the battery activity, and even cause thermal runaway problems.2−4 New thermally conductive materials need to be developed to meet the needs of thermal management.5

In recent years, new thermally conductive materials have become a hot topic of research. Most of these high thermal conductivity and ultrahigh thermal conductivity materials are based on composite materials. When the thermal conductivity of the material exceeds 400 W·m⁻¹·K⁻¹, it is called ultrahigh thermal conductivity material. In particular, they are mixed constructions of nine metal and nonmetallic substances as the thermal conductivity of metals decreases with increasing temperature.6−8 At the same time, the thermal conductivity of alloys is lower than that of their constituent metals. This is mainly because the transfer of electrons achieves heat conduction in metals, and an increase in temperature reduces the rate of electron movement. Alloys also contain other metals that form a different lattice, affecting electron transfer.9 This is why nonmetals have been the subject of research. The nonmetallic materials with high thermal conductivity are mainly those of the element carbon. Among these are graphite, graphene (G), diamond, and carbon nanotubes, all of which have ultrahigh thermal conductivity.10−12 Among these materials, graphene has an advantage over graphite, diamond, and carbon nanotube (CNT) due to its shape and better coupling properties with the substrate. It has an advantage over graphite, diamond, and CNT. Graphene conducts heat utilizing phonons. ZA modes are dominant in graphene heat transport.13 According to the study, the thermal conductivity of graphene is influenced by the number of layers, shape, etc. The number of layers of graphene can be determined by Raman mapping. This has also become a standard test tool for graphene-based composites. Also frequently used is the

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application of graphene as a thermally conductive material. Whether as a filler or surface material, the medium that acts as a heat transfer agent in graphene is phonons rather than electrons. This also provides the basis for the construction of future composites. Elshina et al. have used aluminum (Al), alumina, and graphene to obtain a highly thermally conductive material by adding aluminum and graphene particles to it using a ceramic material composed of alumina. The problem is that graphene cannot form a thermally conductive network, and the increase in thermal conductivity is limited. Goyal and Balandin propose to combine metal particles with graphene particles to create a highly thermally conductive material made up of nanoparticles. Due to the composition, the probability of graphene contact can only increase by increasing the amount of graphene, which lacks a fixed index as a production method. Based on previous methodological studies, the construction of thermally conductive graphene networks on metal surfaces by some technique is a feasible preparation method. However, Wejrzanowski et al.’s research has shown that the natural connection between the metal and graphene reduces the overall thermal conductivity of the material, with a gap of two orders of magnitude between the thermal conductivity of its interstitial voids and the performance of the material. Therefore, the search for a material that can combine graphene with metal and reduce its holes is the focus of the research.

Unlike previous preparation methods, electrophoresis is used. Electrophoresis means that the charged particles move in the opposite direction of the electrode under a direct current field, and the electrophoretic method allows the target to be enriched near the electrode. There are four main stages, namely, electrolysis, electrophoretic movement, electrodeposition, and electroosmosis stages. In previous studies, electrophoresis has been used extensively in applying paints, resulting in rich, homogeneous, and flat coatings. Mu et al. used electrochemical methods to construct anticorrosion coatings on the surface of metallic aluminum. The electrochemical construction method is fast and allows the thickness of the layer to be controlled to ensure a uniform coating.

In this paper, a method for the construction of an ultrahigh conducting thermal material is presented. Using Al as a substrate, it is immersed in a polydopamine (PDA) solution. After some time, it is removed and electrophoresed through an electrochemical workstation for a while. The electrophoresis solution is a graphene oxide (GO) solution. Finally, the sample is removed and annealed to obtain a thermally conductive composite material. After the material is obtained, the material has ultrahigh thermal conductivity with a thermal conductivity of 492 W·m⁻¹·K⁻¹. The surface graphene is tightly bonded with an adhesion force of 6.18 mN, which is of use. Using this material, a flexible thermally conductive device has been constructed to address the problem of both heat and volume change in the operating environment of tertiary lithium batteries. This flexible device has a significant improvement in thermal conductivity compared to existing flexible thermal conductivity devices.

2. RESULTS AND DISCUSSION

2.1. Surface Performance Analysis. The thermal conductivity of each sample was first investigated. According
to the data in Figure 1a, a significant increase in thermal conductivity was found to have occurred for sample 6. The thermal conductivity of sample 6 was found to be ultrahigh conducting. To verify the performance of the material, a modulus of elasticity test was carried out. The tensile strength of sample 1 is shown in Figure 1b. The tensile strength of sample 6 is shown in Figure 1c. According to the test, the tensile strength of sample 6 increased by 12.4% relative to sample 1. This proves that the material still has high power.

Figure 2. (a) Schematic diagram of material composition. (b) Low-magnification SEM image of sample 4. (c) Low-magnification SEM image of sample 6. (d) High-magnification SEM image of sample 6. (e) Energy-dispersive X-ray spectroscopy (EDS) plot of sample 5. (f) EDS plot of sample 6.

Figure 3. (a) X-ray diffraction (XRD) spectra of samples 5 and 6. (b) Fourier transform infrared spectroscopy (FTIR) of samples 5, 6, and 7. (c) Raman spectra of samples 5 and 6. (d) X-ray photoelectron spectroscopy (XPS) of sample 6. (e) XPS of Al 2p. (f) XPS of C 1s.
previous laboratory materials, so adhesion experiments were carried out on it.\textsuperscript{19} The purpose of the adhesion test was to ensure that the graphene would not come off quickly, and the results of the adhesion test are shown in Figure 1d.

2.2. Composition Analysis. According to the scanning electron microscopy (SEM) analysis, the formation mechanism should be described in Figure 2a. First, a polydopamine film should have formed on the surface of the aluminum sheet; then, the polydopamine would have adsorbed graphene oxide, which was annealed to form a reduced graphene film. A low-magnification electron microscopy image of sample 4 is shown in Figure 2b, with the polydopamine in a uniform cover of flakes. A low-magnification electron microscopy image of sample 6 is shown in Figure 2c, where graphene is distributed in a folded paper-like pattern. Figure 2d shows a high-magnification electron microscopy image of sample 6, where the graphene film can be seen under the high-magnification electron microscopy image. This is supposed to be the reason for the high thermal conductivity of G-PDA-Al. However, it is not enough to perform these analyses; the next step is to characterize the composition.

Figure 2e,f shows the EDS plots of samples 5 and 6, where the elemental distribution of the models can be seen. C, Al, and O dominate the basic allotment of the samples. There is a significant decrease in the oxygen content in (e) compared to (f), which indicates that graphene oxide has been reduced by annealing to form graphene and, therefore, the oxygen content has decreased. This analysis corroborates the analysis above, but the relationship between polydopamine and graphene is not clear enough to be analyzed next.\textsuperscript{20}

2.3. Chemical Bond Energy Analysis. Figure 3a shows the XRD drawings of samples 5 and 6. According to the XRD figure, the characteristic peaks of aluminum appear at 44.7°, 65.1°, and 78.2° (JCPDS Standard Card 04-0787), indicating that no other substances are generated from aluminum as a substrate. At the same time, the characteristic peaks of graphene oxide (GO) appeared at 18° and 20°. After annealing, the distinct peaks of sample 6 decreased significantly, indicating that GO decreased. At the same time, compared with sample 5, the characteristic peaks of sample 6 at 27° and 42° were significantly more robust, among which the distinct rise at 42° was apparent, which was the characteristic peak of graphene (JCPDS Standard Card 65-612), indicating that the amount of graphene increased. After XRD, it can be concluded that graphene oxide is reduced to form graphene after annealing, which adheres to the surface of aluminum.\textsuperscript{21}

Figure 3b shows the FTIR spectra of samples 5, 6, and 7. The broad peaks around 3500 cm\(^{-1}\) are the O—H and N—H stretching vibrational peaks. The absorption peaks labeled with “•” are the O—C=O stretching vibration peaks, the absorption peaks marked with “◊” are the superimposed peaks of the C= C stretching vibration of the benzene ring and the N—H bending vibration, the absorption peak labeled with “◊” is the deformation vibration peak of C—O—H, the absorption peaks labeled with “*” are the telescoping vibration peaks of C—OH, and the absorption peaks labeled with “Δ” are the C—O stretching vibration peaks.\textsuperscript{22} The O—C=O stretching vibration peak of sample 5 was more substantial than that of sample 6, indicating that the annealing process at higher temperatures caused the sample to lose some of its O—C=O functional groups on the surface. The absorption peak of the selection was shifted to the right after annealing, indicating that the degree of conjugation was increased by annealing. After annealing, the C—O—H deformation vibration peak disappeared, and the C—OH stretching vibration peak appeared and was weaker at 300 °C than at 200 °C. The stretching vibration peak of C—O indicates that some of the oxygen-containing functional groups were removed during the annealing process. Graphene oxide was reduced to graphene, and the higher the temperature, the more oxygen-containing functional groups were lost.

Figure 3c shows the Raman spectra of samples 5 and 6. The absorption peaks at Stoke shifts of 1349, 1597, and 2700 cm\(^{-1}\) are the carbon material’s D, G, and 2D peaks. The D peak is related to the intrinsic defects and disorders in the material itself, and the G peak is connected to the vibrations of the C sp\(^2\) atoms in the face. The ratio of I\(_D\)/I\(_G\) is commonly used to evaluate the defect density of graphite materials.\textsuperscript{23} The I\(_D\)/I\(_G\) ratio of sample 4 was 0.81, and that of sample 5 was 0.74. The I\(_D\)/I\(_G\) ratio of sample 5 was smaller than that of sample 4, indicating that some of the oxygen-containing functional groups were removed during the annealing process while reducing the defect density of the sample, and GO was reduced to graphene. The ratios of I\(_D\)/I\(_G\) for samples 4 and 5 were 0.20 and 0.28, respectively, indicating that the graphene attached to the surface of the samples was a bilayer or multilayer.\textsuperscript{24}

In Figure 3, the XPS full spectrum of sample 6 is shown. According to the spectrum, elements C, N, and O appear in it. Because of the processing accuracy of XPS, only a tiny amount of aluminum appears in the graph (Table 1). Elements C, N, and O indicate the presence of graphene and dopamine. To further observe the bond energy bonding, the study of slit fitting was carried out. According to Figure 3d,f, it can be seen that C mainly exists in the form of the C—C—C bond, which is also the main bond energy distribution of polydopamine and graphene. In particular, in Figure 3f, peak fitting was carried out for C 1s, and the characteristic peak of C was explained. Further analysis of the C bond can be carried out as follows according to peak fitting: This distributes π-cross bonds, which are the standard bond energies of polydopamine and graphene. The polydopamine connection to graphene is also in the form of a C—O bond. According to the peaking data in Figure 3e, it is found that it mainly exists in the form of an Al—O bond, indicating that the connection mode of polydopamine and aluminum is connected primarily in the form of an oxygen bond.\textsuperscript{25}

2.4. Formation Mechanism. The mechanism of formation of the material is shown in the diagram, which can be divided into three parts.

In the first part, from a macroscopic point of view, the reaction is shown in Figure 4a, where the aluminum sheet is soaked to form a spherical polydopamine on the surface, which is then electrochemically attracted so that polydopamine can adsorb to graphene oxide. This results in the formation of a graphene shell on the surface. The second part starts with a

| Table 1. Percentage of Elements in Sample 6 |
|------------------------------------------|
| C 1s | N 1s | O 1s | Al 2p |
| sample 6 | 67.5 | 4.3 | 21.5 | 3.2 |
microscopic chemical reaction, the reaction flow of which is shown in Figure 4b. First, the dopamine molecules undergo agglomeration under alkaline conditions to form 5,6-dihydraxyindote. Then, the agglomeration reaction forms PDA, which connects graphene to the aluminum sheet via an oxygen bond. This acts as a bridge. In Figure 4c, the model of dopamine is shown by Gaussian calculations, where the bond angle distribution and the individual atomic distribution can be observed. This accurately represents the chemical process and describes the polymerization of dopamine in terms of the bond angle distribution and the nuclear distribution.

2.5. Model Calculation. Once the material composition was precise, the thermal conductivity of G-CVD-Al was calculated for the same $V_v$ to differentiate it from the previous material.

According to model a (Figure 5), the thermal ability to let heat flow can be calculated according to the limits/guidelines of lamellar things made up of different items in clearly connected or related studies. The volume fraction of various things or materials ($V_v$) can be received/begotten by eq 1.

$$V_v = \frac{h_{Al-G} + h_G}{h_{sum}} \%$$

where $h_{Al-G}$ is the thickness of the Al-G interface, $h_G$ is the thickness of the G layer, and $h_{sum}$ is the thickness of the composite material. The $h_{sum}$ can be obtained by eq 2.

$$h_{sum} = h_G + h_{Al} + 2h_{Al-G} [\mu m]$$

where $h_{Al}$ is the thickness of the Al layer.

The thermal conductivity parallel to the elongated Al layer ($\epsilon_{\parallel}$) can be obtained by eq 3.

$$\epsilon_{\parallel} = \frac{h_{sum}}{h_{Al} \epsilon_{Al} + h_G \epsilon_G + 2h_{Al-G} \epsilon_{Al-G}} \text{[W.m}^{-1}.\text{K}^{-1}]$$

(3)

where $\epsilon_{Al}$, $\epsilon_{G}$, and $\epsilon_{Al-G}$ are the thermal conductivities of the aluminum layer, G layer, and Al-G interface, respectively.

The thermal conductivity across the Al layer ($\epsilon_{\perp}$) can be obtained from eq 4.

$$\epsilon_{\perp} = \frac{h_{Al} \epsilon_{Al} + h_G \epsilon_{G,\perp} + 2h_{Al-G} \epsilon_{Al-G}}{h_{sum}} \text{[W.m}^{-1}.\text{K}^{-1}]$$

(4)

Combined with previous studies, the parameters of G-CVD-Al are shown in Table 2.

### Table 2. Parameters of the Lamellar Composite Fabricated within These Studies

| $\theta$ | $\epsilon_{Al}$ | $\epsilon_{Al-G}$ | $\epsilon_{G,\parallel}$ | $\epsilon_{G,\perp}$ | $N_G$ | $h_{Al}$ |
|---------|-----------------|-------------------|---------------------|-------------------|------|--------|
| model a | 198             | 50–350            | 0.03–0.3            | 1000              | 0.67 | 2      | 0.3    |

$N_G$ is the number of layers.

The thermal conductivity enhancement (TCE) can be obtained by eq 5:

$$\text{TCE} = \epsilon - \epsilon_{Al} \text{[W.m}^{-1}.\text{K}^{-1}]$$

(5)

The microscopic TCE parallel to the elongated Al layer (TCE$_\parallel$) and microscopic TCE across the Al layer (TCE$_\perp$) can be obtained by eqs 6 and 7, respectively:

$$\text{TCE}_{\parallel} = \epsilon_{\parallel} - \epsilon_{Al} \text{[W.m}^{-1}.\text{K}^{-1}]$$

(6)

$$\text{TCE}_{\perp} = \epsilon_{\perp} - \epsilon_{Al} \text{[W.m}^{-1}.\text{K}^{-1}]$$

(7)
According to the model, the overall thermal conductivity of aluminum is not significantly improved if only G is used to form the film, and the longitudinal improvement of G-CVD-Al is only 0.05 W·m⁻¹·K⁻¹; thus, the TCE of G-CVD-Al is 32 W·m⁻¹·K⁻¹ (Table 3). The adhesion force significantly decreases Al-G for G-PDA-Al and conventional G-CVD-Al, which is the main reason for the improved thermal conductivity. Briefly, $h_{\text{Al-G}}$ decreases, leading to phonons’ rapid and barrier-free propagation between spaces, resulting in a substantial increase in thermal conductivity. This result is almost identical to the data obtained from a conventional G thermal conductivity film.27

The graphene surface composition of the two materials, G-CVD-Al and G-PDA-Al, can be seen in Figure 6. In conventional G-CVD-Al composites, the surface is enriched with graphene, but the arrangement of graphene is haphazard, and the composition cannot form a lamellar structure. Unlike PDA, which is used to be a robust surface coating, G-PDA-Al is used as an attractive structure, and PDA not only fixes graphene on the Al surface but also allows graphene to form a layered design on the Al surface. In turn, the layered structure graphene creates a thermally conductive network on the composite surface, which is responsible for the ultrahigh thermal conductivity of the composite.

In the study, a ultrahigh conducting thermal network was built using very little graphene content. The higher the thermal conductivity, the better the performance.28,29 Compared with almost the same materials, creating a thermal network is more reasonable. The thermal conductivity of the material has reached the ultrahigh conducting thermal performance. At the same time, graphene is tightly bound to aluminum.

Several studies similar to this study were selected and compared regarding the graphene content and thermal conductivity. These studies are enumerated in Figure 7. Also marked in the figure is the range of superthermal conductivity. The direction of the minimum graphene fraction is also indicated. Based on the comparison, this study uses the lowest percentage of graphene among the studies that use graphene to construct superconducting materials while also achieving superconductivity. According to the survey, graphene can present different thermal conductivity methods for single-layer and multilayer graphene due to other construction methods. Therefore, suitable preparation methods can substantially improve the thermal conductivity of graphene and thus the overall thermal conductivity of the material.35

### Table 3. Calculated Values of the Lamellar Composite Fabricated within These Studies

| material | $h$ | $V_r$ | $\epsilon_\parallel$ | $\epsilon_\perp$ | TCE$_\perp$ | TCE$_\parallel$ |
|----------|-----|-------|----------------------|------------------|-------------|--------------|
| Al       | 0   | 198   | 0.23                 |                  |             |              |
| G/CVD@Al | 100 | 0.1%  | 230                  | 0.24             | 0.01        | 32           |
| G/PDA@Al | 100 | 0.1%  | 492                  | 0.29             | 0.06        | 294          |

| Figure 6. Cause diagram of thermal conductivity difference between G-CVD-Al and G-PDA-Al. |

| Figure 7. Comparison of the thermal conductivity of similar materials.23,30–34 |

2.6. Application of Flexible Thermally Conductive Devices. Once the G-PDA-Al composite was obtained, a lot of exploration was done to understand how it could be used. A
A flexible thermally conductive device has been designed whose surface is composed of G-PDA-Al, which is then connected to a thermal insulation sponge via PET. The composition structure is shown in Figure 8.

The characteristic of this thermally conductive device is that it deforms when subjected to pressure and returns to its original shape when the pressure is removed. The details are shown in Figure 8b, c. The characteristics of this device can be applied to the thermal management of ternary lithium batteries. Ternary lithium batteries are now a widely used power supply device. They are used in a large number of applications in electric vehicles and electric flying machines. During the operation of a ternary lithium battery, there is a change from chemical energy to electrical energy. There is also heat and volume expansion taking place. Conventional thermally conductive devices cannot solve this problem and can only widen the spacing between the cells and use thermally conductive silicone filling. But flexible thermally conductive devices can solve this problem very well. In contrast to the original flexible thermally conductive device with a thermal conductivity of 212 W m⁻¹ K⁻¹, the flexible thermally conductive device has a thermal conductivity of 492 W m⁻¹ K⁻¹. Compared to the original machine, the new flexible thermally conductive device is a significant improvement.

2.7. Experiments with G-PDA-Al in Thermal Management. Based on the ultrahigh thermal conductivity of the G-PDA-Al material, a finned radiator from a home computer was selected. The radiator was composed of aluminum, and the radiator was cleaned using deionized water and ethanol. The finned heat sink of the G-PDA-Al material was then prepared. The frequency of the central processing unit (CPU) was then fixed, and an infrared camera observed the finned heat sink. The details are shown in Figure 9.

Figure 9 shows the use of G-PDA-Al material in thermal management. After locking the CPU frequency, the heat generated by the CPU is fixed. Figure 9b shows an infrared image of the original aluminum heat sink in an operating environment. In Figure 9c, the infrared imaging in G-PDA-Al is demonstrated. According to the comparison, the temperature drop between the two is about 8 °C. This shows that in thermal management, the use of superthermally conductive materials can effectively reduce the temperature of critical electronic components.

2.8. Mechanistic Analysis. Based on the analysis above, the formation process of G-PDA-Al is explained. PDA has super adhesion and can form a film on the aluminum surface quickly under alkaline conditions, which contains a large number of hydroxyl and amino functional groups. GO was attached by electrophoresis. Then, after annealing, polydopamine reduced GO and interacted with graphene (oxygen bonding and π–π conjugation), which significantly reduced the interfacial thermal resistance. Because of the addition of polydopamine, graphene arranges like fallen leaves under pressure, forming a good thermal network. Thus, the thermal conductivity of the composite is improved.

3. CONCLUSIONS

In summary, a graphene/polydopamine composite material on an aluminum substrate (G-PDA-Al) with ultrahigh thermal conductivity was obtained. The optimal preparation conditions were determined by orthogonal experiments. Then, the thermal conductivity of the composite was obtained as 492
W·m⁻¹·K⁻¹ based on the experimental results. The thermal conductivity was enhanced by 107.59% compared to that of the aluminum substrate. Graphene is bonded to the aluminum substrate tightly. The surface morphology, chemical composition, and thermal conductivity of the well-prepared G-PDA-Al composite material were evaluated. The thickness of the thin graphene film was controlled by the electrophoretic method. Compared with immersion, the electrophoresis method is faster, and the film formation is fuller and more uniform. The formation mechanism of G-PDA-Al was that graphene oxide was adsorbed to the surface of the aluminum sheet through the use of PDA before being reduced to reduced graphene oxide during annealing. Due to the PDA bonds with both the aluminum substrate and graphene, the thermal conductivity of the composite was affected. The increase in the thermal conductivity of the G-PDA-Al material, unlike the rise in the thermal conductivity of the conventional G-CVD-Al material, was analyzed through calculations. The calculation results showed that PDA controlled the growth direction of G and G lamellar stacking layer-by-layer on the aluminum substrate. Based on a flexible heat conduction device structure, the material was used to make a flexible thermally conductive device with a thermal conductivity of 492 W·m⁻¹·K⁻¹. It was better than the existing flexible thermally conductive devices. The thermally conductive devices were prepared to meet the practical application.

4. EXPERIMENTAL SECTION

4.1. Materials. Aluminum sheets were obtained from Beijing Aerospace Corporation. Graphite was obtained from Qingdao Hua Tai Lubricant Sealing S&T Co. (Qingdao, China). Dopamine hydrochloride was obtained from J&K Scientific Ltd. Other raw materials were purchased as described in the Supporting Information.

4.2. Preparation of GO. GO was synthesized from graphite powder with an improved Hummer’s method. A homogeneous GO aqueous suspension (0.5 mg/mL) was achieved by dispersing GO in water with ultrasonication for 3 h.38,39

4.3. Preparation of G-PDA-Al. Aluminum sheets with a size of 10 mm × 10 mm × 0.1 mm were ultrasonically washed with ethanol, acetone, and deionized water for 5 min to remove organic contaminants on the surface. Dopamine hydrochloride was dissolved in deionized water to prepare the dopamine solution (20 mg/mL), and then the pH was adjusted to approximately 8.5 using a 2 mol/L NaOH solution. Subsequently, the cleaned aluminum sheet was set perpendicularly in the above-prepared dopamine solution for 1 h at room temperature.

A three-electrode system was established using the PDA-Al sheet as the negative electrode, followed by the glycolic electrode as the positive electrode and the platinum electrode (EPD) as the counter electrode. A constant voltage of 5 V was set. The sample was then removed and annealed. The electrophoresis solution was 40 mg/mL GO solution. By electrophoresis, GO can be deposited on the surface of PDA-Al to form G-PDA-Al composites. The preparation process flow chart of G-PDA-Al is shown in Figure 10.

The optimal reaction conditions were obtained according to the orthogonal experiment. See the orthogonal experimental data in the Supporting Information for more information on the materials design based on the results of the orthogonal experiments. To determine the optimum preparation conditions, the thermal conductivity of the samples under different
preparation conditions, such as the immersion time in the dopamine aqueous solution and GO aqueous suspension and the annealing temperature and annealing time, were investigated; the preparation conditions are shown in Table 4.

4.4. Characterization. The volume of the object was determined by the Archimedes drainage method. The weight was weighed using a balance. The thermal diffusivity ($\alpha$) of the sample was measured using a laser flash thermal analyzer (LFA447, NanoFlash, Germany) at room temperature. The temperature distribution at $x = L$ ($x$ is the distance from the front of the specimen) was obtained when a laser pulse of heat was uniformly absorbed by a surrounding adiabatic, homogeneous, opaque, circular specimen of thickness $L$. A one-dimensional heat flow was generated on the back of the specimen. The time was taken for the temperature at the back of the sample to reach half its maximum value under laser pulse irradiation. Therefore, the thermal diffusivity $\alpha$ can be calculated by measuring $t$ and $L$.

The specific heat (CP) in this paper was measured by a physical property measurement system (PPMS-9T, Quantum Design, USA) at room temperature. The density ($\rho$) was calculated according to eq 8:

$$\rho = m/V$$  \hspace{1cm} (8)

where $m$ is the mass of the sample, and $V$ is the volume of the sample.

The thermal conductivity ($K$) is based on eq 9:

$$K = \alpha \times CP \times \rho$$  \hspace{1cm} (9)

where $\alpha$ is the thermal diffusivity of the sample, and CP is the specific heat of the sample.

The electrochemical workstation was the CHI-760E electrochemical workstation.

The adhesion force test was measured by using a nanoscratch meter (Ti980, Bruker, Germany).

The surface morphology and chemical composition were characterized using scanning electron microscopy (SEM; NovaNano 430) with energy-dispersive X-ray spectroscopy (EDS). The crystal structure was investigated by X-ray powder diffraction (XRD, D8 Advance, Bruker, Germany) with a Cu Kα radiation source in a continuous scanning mode (40 kV, 40 mA, and $\lambda = 0.15418$ nm) and a scanning rate of $2^\circ$ min$^{-1}$. The surface chemical composition of the sample was analyzed by X-ray photoelectron spectroscopy (XPS, model PHI 5300, Physical Electronics, USA) using 250 W Mg Kα ($\lambda = 0.9891$ nm) X-ray as the excitation source in the constant analyzer energy mode with C 1s at 284.8 eV as the reference. The infrared spectra of the samples were obtained from attenuated total reflectance-Fourier transform infrared (ATR-FTIR) spectroscopy ( Nicolet 6700) equipped with a liquid nitrogen-cooled MCT detector. Raman spectroscopy was conducted using a micro-Raman system (LabRAM HR Evolution, France) equipped with a 532 nm Ar-ion incident laser. The vibrational Raman Stokes spectra were recorded from 200 to 3500 cm$^{-1}$.

**ASSOCIATED CONTENT**

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.1c05957.

Scheme of factors and levels of orthogonal experiment (Table S1); orthogonal experimental table, four factors, and three levels (Table S2); scheme of factors and levels of orthogonal experiment (Table S3); further experiment of orthogonal experiment (Table S4); sample thermal conductivity data of orthogonal experiment (Figure S1); thermal conductivity distribution of supplementary experiment (Figure S2) (PDF)

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**Notes**

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