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Study on the Preparation of a High-Efficiency Carbon Fiber Dissipating Coating

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Abstract: The working temperature of electronic components directly determines their service life and stability. In order to ensure normal operation of electronic components, cooling the coating is one of the best ways to solve the problem. Based on an acrylic amino-resin system, a dissipating coating was prepared with carbon fiber (CF) as the main thermal conductive filler. The influence of the CF content on the thermal conductivity was determined by the single factor method. The surface structure was observed by scanning electron microscopy (SEM). The results show: With the increase of the CF mass fraction, both the heat dispersion and heat conduction coefficient of the coating tend to increase at first and then decrease, and the heat dissipation effect is optimum when the CF mass fraction is 12.3 wt %. At this point, the coating shows an excellent comprehensive performance, such as 1st level adhesion, H grade hardness, and thermal conductivity of 1.61 W/m·K. Furthermore, this paper explored the radiating mechanism of coating in which CF produces a coating which forms a heat “channel” for rapid heat conduction. When the optimal value is exceeded, the cooling effect is reduced because of the accumulation and the anisotropy of CF.

Keywords: dissipating coating; carbon fiber; heat channel; radiating mechanism

1. Introduction

At present, electronic components are developing at a high frequency, as are both density and miniaturization, thus the heat flow density per unit area is increasing rapidly, and the internal temperature can rise sharply if the heat cannot be dissipated in time. The working temperature of electronic components directly determines their service life and stability [1], so the performance and life of the components will be decreased significantly if the temperature exceeds the normal operating temperature range. Research has shown that more than half of equipment failure is caused by high temperature [2]. Traditional cooling methods, such as forced air cooling, have been unable to meet the requirements of modern electronic equipment [3]. Considering security and performance, electronic components should not be directly exposed to the air, and need to be coated to improve the performance of the electronic components. The application of coating technology not only solves the problem of heat dissipation of electronic components but also contributes to new heat coating technology development.

Li Minghui [4] prepared a kind of polyamide based thermal conductive and insulation composite material with AlN, Al_2O_3, and BN respectively as thermal conductive fillers, and it turned out that the distribution of the thermal conductive fillers in the polymer was uniform, and the thermal conductivity of the composites increased gradually with increased filler content, favorable for heat transfer. Srikar, R. [5] provided a kind of nano fiber coating by using electrospun polymeric materials on heat transfer surfaces, and the study found that the material had good wettability after coating with nano materials; the water droplets could penetrate more easily into the interior of the material, thus having the effect of evaporation and heat dissipation. Yang Qinghao [6] offered a kind of polyurethane coating with alumina particles and silica particles as a mixed thermal conductive filler, with a thermal conductivity of 2.2 W/m·K and adhesion of the 2nd level. Lei Dingfeng [7] afforded an epoxy resin thermal conductive plastic by adding Al_2O_3 by treating with a silane coupling agent KH-560 in the E-20 type epoxy resin, and found that its thermal conductivity was 1.33 W/m·K and its adhesion was of the 0 level. Zhang Xueping [8] used an epoxy resin system and added spherical alumina and other additives to obtain a thermosetting thermal insulation coating, The upshot was that its thermal conductivity was 1.1 W/m·K with a breakdown strength of not less than 35 MV/m.

Coating technology cannot only solve the problem of heat dissipation, but also prevents the corrosion of components and the danger of static electricity. However, from the above study, it was found that most heat coatings are of traditional solvent based paint, and produce large amounts of toxic and harmful waste gas and waste water in the manufacturing process. At the same time, the prepared coatings have poor mechanical properties, high production costs, low temperature resistance, and a bad insulation performance.

To solve the above problems, a kind of dissipating coating was prepared by using the high thermal conductivity and high radiation of CF as the main filler and acrylic resin as the matrix. The single factor method was used to determine how the the mass fraction of CF in the coating affected heat conduction. The morphology of the coating was observed by a scanning electron microscope (SEM), and the heat dissipation mechanism of the coating was investigated by the results of the thermal conductivity measurements and the cooling effect. Finally the application of the coating performance was tested.

2. Materials and Methods

2.1. Materials

According to the principle of heat transfer, it is important to focus on the increase of the radiation and conduction of the coating, so the choice of raw materials must have high thermal conductivity and high radiation. CF is a type of material with high heat conductivity while the thermal conductivity in the fiber direction can be higher than copper, up to 700 W/m·K [9]. Moreover, CF has good mechanical properties and very good radiation ability. In this study, high strength and high modulus CF filaments were selected, the surface of which was treated with a silane coupling agent. Then, the fibers were ground and dried to obtain powdered short CF. In this way, it not only retained the excellent performance of CF, but also had an increased specific surface area, so that it was easy to have evenly dispersed resin lubrication. In this experiment, the specific parameters of CF are shown in Table 1.

Table 1. The parameters of carbon fiber (CF).

Parameter	CF
Tensile strength (MPa)	>3500
Density (g/cm^3)	1.75
Filament diameter (μm)	7
Carbon content (wt %)	>95
Fineness (mesh)	300
Aspect ratio	3:1–4:1
Thermal conductivity of fiber direction (W/m·K)	120

Nanometer aluminum nitride (AlN) is a kind of inorganic ceramic, and has excellent thermal conductivity. The thermal conductivity of a single crystal of AlN can be as high as 320 W/m·K at room temperature. At the same time, AlN has advantages of good insulation, a small thermal expansion coefficient, and good mechanical properties, as well as being a good weather resistant and high-temperature resistant filler. When AlN was added into the matrix, due to its small particle size and high conductivity, the CF can be linked with the "island-island" distribution [10,11]. Sericite powder has excellent infrared radiation ability and steady chemical properties, so that it can reduce the damage of light and heat on the film and increase the acid and alkali resistance of the coating [12] when added into the coating. Silicon dioxide has a strong absorption in the spectrum at wavelengths longer than 8 μm, hence it can form a good layer of radiation cooling with sericite powder [13].

When acrylic polyurethane resin is used as the main film-forming material, the filling can be well dispersed in the matrix, so that the coating has good mechanical properties and extensive construction performance [14]. The thermal conductivity and radiation of the coating can be improved by using CF of high thermal conductivity and high radiation.

Based on the anisotropy of the thermal conductivity of CF, its dosage is the biggest effect factor on coating thermal conductivity. Therefore, this paper mainly aimed to study the influence of the heat transfer performance of the coating with different contents of CF.

In this paper, in order to meet the required mechanical properties, corrosion resistance and environmental protection of the coating, detailed information of the raw materials and contents is shown in Table 2.

Table 2. The detailed information of the raw materials and content.

Material	Specifications	Manufacturer	Mass (g)
Silicone-acrylic resin	Solid containing 50 wt %, silicon containing 40 wt %	Sihui Xinda Chemical Industry Co. Ltd., Guangzhou, China	36
AlN	Modified AlN powder	Qingzhou Zhengda Chemical Co. Ltd., Qingzhou, Shandong, China	8
Sericite powder	–	Chuzhou Gerui Mining Co. Ltd., Chuzhou, Anhui, China	13
Silicon dioxide	200 meshes	Shijiazhuang Gold and Nano Chemical Co. Ltd., Guangzhou, China	1.5
Amino resin	Solid containing 60%	Nanjing Witten Chemical Technology Co. Ltd., Nanjing, China	11
Cosolvent (butyl acetate, two acetone alcohol, xylene)	Analysis of pure	Zhongshan Yongsheng Chemical Co. Ltd., Guangzhou, China	20–35
Additives (BYK-ATU, BYK-355)	–	BYK(Shanghai) Co. Ltd., Shanghai, China	0.5–2
CF (300 mesh)	Average length: 45 μm	Zibo Xinnuo lubricants Co. Ltd., Zibo, Shandong, China	Variable (gradient)

2.2. Preparation of Coatings

First, the silicone modified acrylic resin and dispersant were added into a 200 mL reaction tank. Next, were added the CF, aluminum nitride, sericite powder, and anti-settling agent. When the mixture was evenly dispersed, it was diluted 2–3 times with the solvent of the corresponding mass fraction. Afterward, the pulverized zirconium beads of weight 1.5 times the total weight were successively added to the tank and dispersed at 1200 rpm for 1 h, and the amino resin was added and redispersed at 1000 rpm. Finally, the 200 mesh filter was used to remove impurities to obtain the product when the fineness was lower than 50 μm. The preparation method of the target coating is shown in Figure 1.

The target coating was diluted by a diluent, and this diluent was prepared by using butyl acetate, xylene, and two acetone alcohol as mentioned above in the ratio of 1:1:1. When the viscosity was 13–16 Pa·s, the coating was sprayed onto the surface of the treated substrate with an air gun. Then, after waiting until the coating had been flattened, the substrate was placed in an oven at 150 °C. After 30 min of treatment, the cured film was taken out to obtain the test sample.

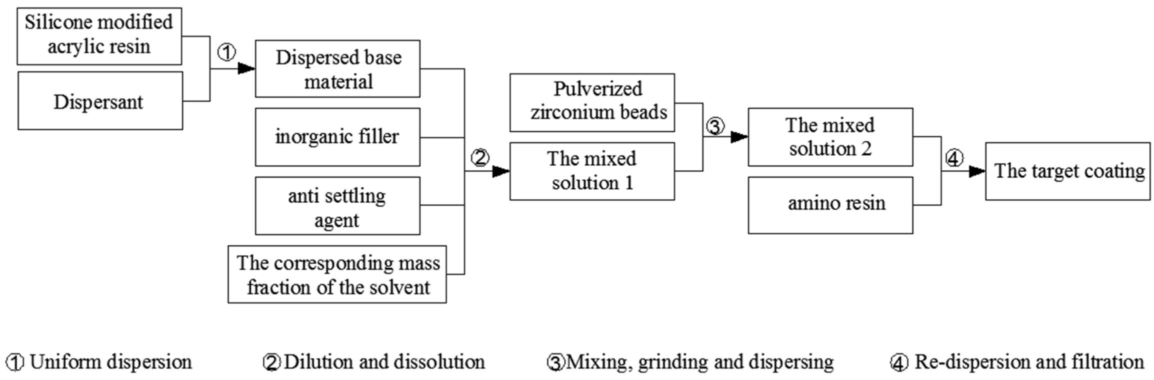


Figure 1. The preparation method of the target coating.

2.3. Performance Testing

2.3.1. Measurement of Thermal Conductivity of the Coating

The coating was made into a circular sample with a diameter of 6.5 cm and a thickness of about 3.5 mm. The thermal conductivity of the coating was tested by using a thermal conductivity analyzer of Hot Disk TPS2500 in 7854 probe with 5 s heating time and the 0.3 W heating power [15], Sweden hot disk Ltd., Uppsala, Sweden.

2.3.2. Measurement of Heat Dissipation of the Coating

The specimen with the cooling coating and the blank specimen were placed in a 180 °C constant temperature oven for 1 h, Shanghai Yaoshi instrument equipment factory, China, then it was removed. The cooling curve was measured by a furnace temperature tracker (type SMT-7-128-500-K) in the case of natural convection, Beijing Savemation Tech Co., Ltd., Beijing, China.

2.3.3. Coating Surface Structure Observation

The prepared coating samples were coated with an aluminum plate of 1×1 cm^2. To give higher resolution images, a thin layer of gold was coated on the sample by a sputter coater.

The surface structure of the coating was observed by a SEM3700 scanning electron microscope (SEM) under a voltage of 5 kV and a magnification of 500, 1000, and 3000, Germany visitec Ltd., Heidelberg, Germany.

2.3.4. Measurement of Mechanical Properties of Coating

- Hardness: First, the 5 um thickness sample coating was fixed on the horizontal test platform. Then, in conditions in which the lead core was not broken, in degrees, a series of pencils, with hardness of 5 B to 5 H, were pushed 1 cm along the coating at about 45°. Each hard lead core was pushed 5 times. If two or more of the 5 lines were scratched, the pencil mark of the previous hardness was designated as the hardness of the coating.
- Impact resistance: The coated steel plates were placed on the impactor. Then, a heavy cone (1 kg weight) was dropped from a certain height to the center of the coating on the model, and each high impact was tested 3–5 times. If there was not an obvious crack or spalling under the 4 times magnification, the height was further increased until a crack or spalling occurred, and the final height was designated as the impact strength of the coating.
- Temperature resistance: The thermal stability of the coating was analyzed by placing a sample of 10 mg in the sample chamber of the STA449C thermo-gravimetric analysis (TGA), Taiwan National Taiwan University Co., Ltd., Dong Guan, China. The conditions of the test were such that under a nitrogen atmosphere, the heating rate was 10 °C/min, the air velocity 20 mL/min,

and the scanning temperature was increased from 100 °C to 700 °C. In the resulting TG curve, the highest temperature resistance of the coating was the temperature at which the weight began to decrease.

- Adhesion: First, the coating was spread on the aluminum plate. Then, A F107 paint film scriber was used to draw a cross grid shape in the model, with an incision until the base material. A hairbrush was used to brush 5 times along the diagonal direction of the lattice cross, after which adhesive tape was used close to the incision and pulled off quickly. The above test was repeated several times, and the level of adhesion of the coating was determined in accordance with the ISO adhesion grade standard under microscope observation.

3. Results and Discussion

3.1. Effect of CF Mass Fraction on the Thermal Conductivity of Coatings

The relationship between the thermal conductivity and the mass fraction of the CF of the coating is shown in Figure 2.

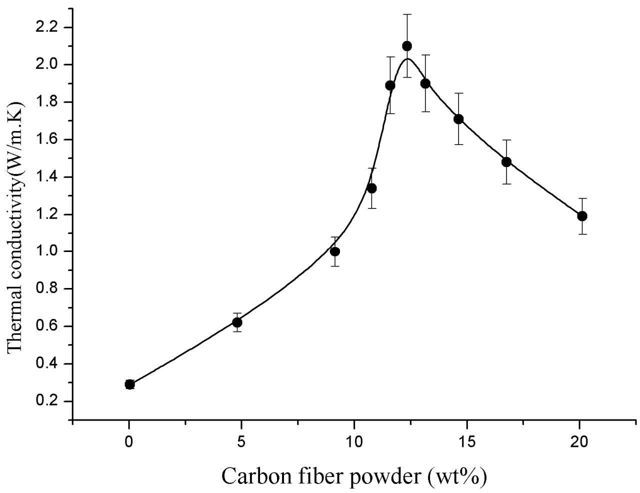


Figure 2. The relationship between the thermal conductivity and CF mass fraction of coating.

From Figure 2, with the increase of CF mass fraction, the coating thermal conductivity shows a tendency in which the curve increases rapidly at first and then reduces slowly. The layer has a maximum heat transfer coefficient of 2.1 W/m·K when the CF mass fraction is 12.3 wt %. Analyzing the cause, at the beginning, the CF is dispersed in the matrix, and the thermal conductivity of the substrate plays an important role, so its thermal conductivity is lower. When the mass fraction of CF is less than 12.3 wt %, with the increase of the CF mass fraction, a bridge forms between the CF. Then, the amount of dominant influence the coefficient of thermal conductivity in the sample increases continuously, and a thermal conduction network is developed, so that the thermal conductivity increases. When the CF mass fraction is more than 12.3 wt %, with the increase of CF mass fraction, the excess CF causes destruction of the network, which leads to the failure of the thermal conductivity network, so that the thermal conductivity is reduced, at this time its thermal conductivity is completely dominated by the CF content.

3.2. Effect of CF Mass Fraction on the Cooling Range of the Coating

Figure 3 shows the relationship between the amount of CF and the cooling temperature of the coating. As seen in Figure 3, the cooling range of the specimen shows a tendency in which the curve begins to increase rapidly and then decreases slowly as the mass fraction of CF increases. When the mass fraction of CF is 12.3 wt %, the coating has a maximum cooling range. According to the analysis of Figure 2, within the scope of this study, the cooling curve of the coating is consistent with the thermal conductivity curve, and the thermal conductivity of the coating is optimum at the maximum temperature.

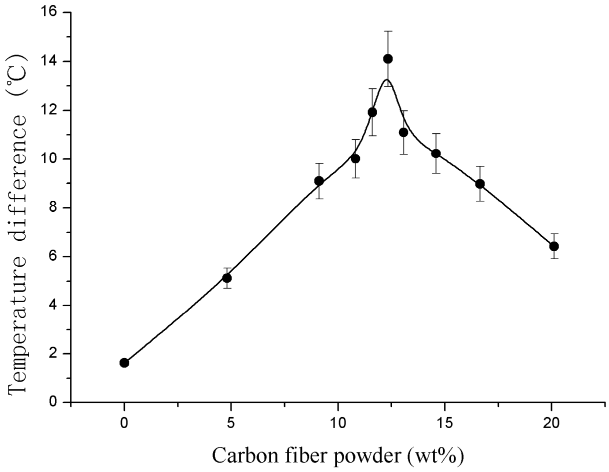


Figure 3. The relationship between the coating CF mass fraction and temperature.

3.3. Analysis of Coating Morphology and Mechanism Analysis

Traditional resin is a poor conductor of heat, and its heat conduction coefficient is lower than 0.5 W/m·K at 25 °C. For instance, epoxy resin is only 0.2 W/m·K [15]. The thermal conductivity of filled polymer materials is mainly determined by the thermal conductivity of the filler and the polymer matrix [16], the filler content and the arrangement of the filler in the polymer matrix [17]. According to Agari-Y's theory of the network structure of heat conduction [18], when the filler content is too low, the filler is dispersed in the matrix, and the thermal conductivity of the substrate plays an important role. With the increase of filler content, a bridge forms between filler particles, and it gradually forms a heat conductive network that penetrates through the substrate, thus increasing the thermal conductivity. When the filler content is too high, it introduces a parallel and vertical heat conduction mechanism, and then the explanation is as follows: The thermal conductivity of the composite is the highest when the thermal conduction channel is parallel to the substrate in the direction of heat conduction, and the thermal conductivity is the lowest when the thermal conduction channel is vertical to the substrate in the direction of heat conduction. As can be seen from Figure 2, the thermal conductivity increases with the increase of the CF mass fraction, which obeys the theory of the network structure of heat conduction.

Figure 4 shows the surface morphology structure of the coating in the SEM observation, and Figure 5 shows the schematic diagram of the specific effect of coating cooling. This diagram is based on Agari-Y's theory of the network structure of heat conduction, Figures 3 and 4. From Figure 4, with the increase of mass fraction of CF, the coating structure gradually proceeds from smooth to rough. From Figure 4a to Figure 4c, the amount of CF is very small, thus the filler is distributed inside the resin in "Island-Island" form, which means the heat conductive body is still the resin, resulting in low thermal conductivity of the coating. From Figure 4d to Figure 4f, the thermal conductivity of the coating increases rapidly. With continuous overlap between fillers, a structure is formed inside the coating which is similar to the "channel" (Figure 5a). This structure is good for the rapid passage of heat, so that the cooling temperature of the coating decreases rapidly. This channel is most obvious at 12.3 wt %. Then, from Figure 4g to Figure 4i, with the increase of CF content, the phenomenon of CF piling up gradually emerges, and a larger gap is formed (as shown by the circles in Figure 4). The anisotropy of thermal conductivity of CF leads to a barrier between the heat conduction "channels" (Figure 5b), thus the cooling temperature range of the coating decreases.

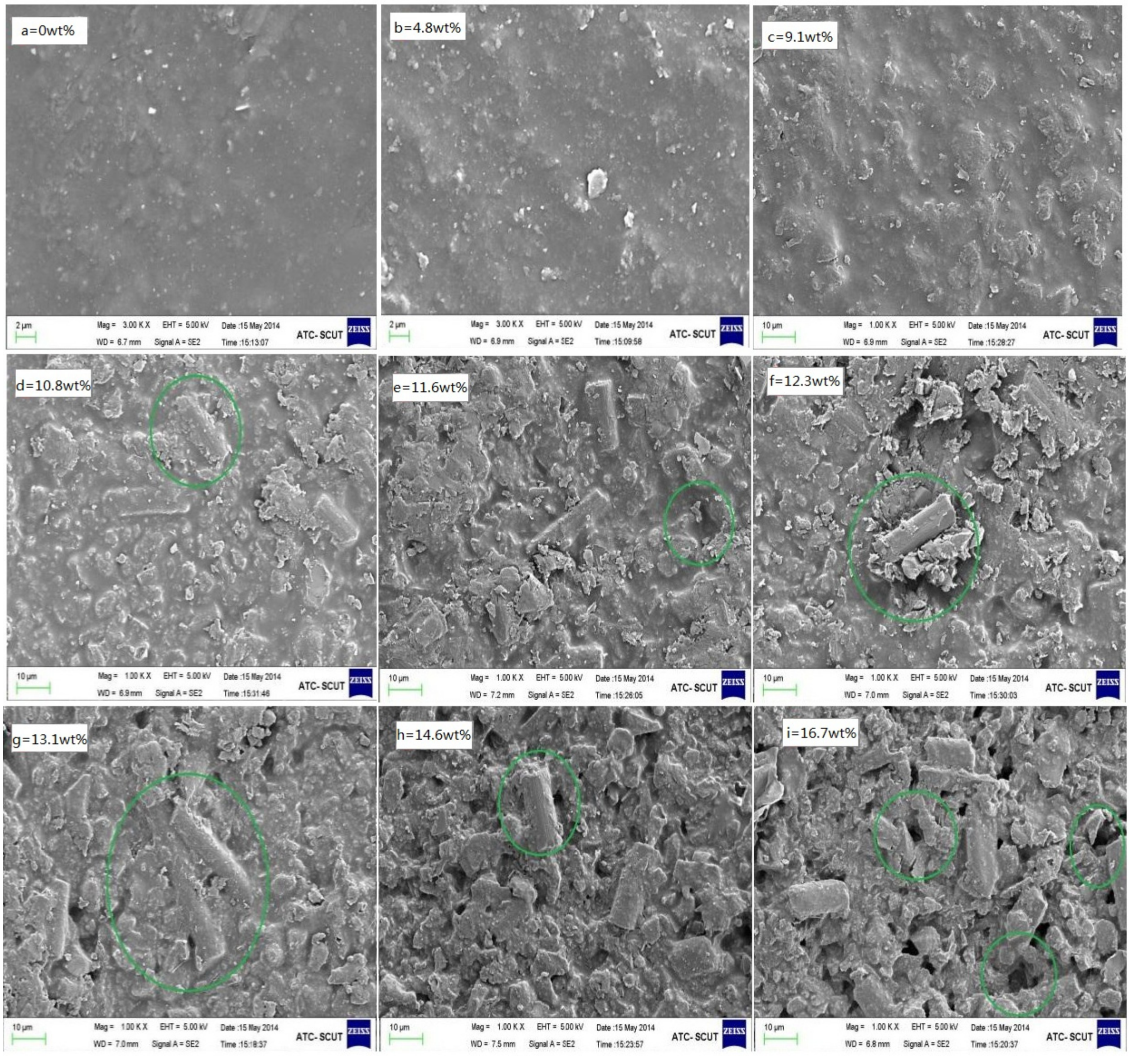
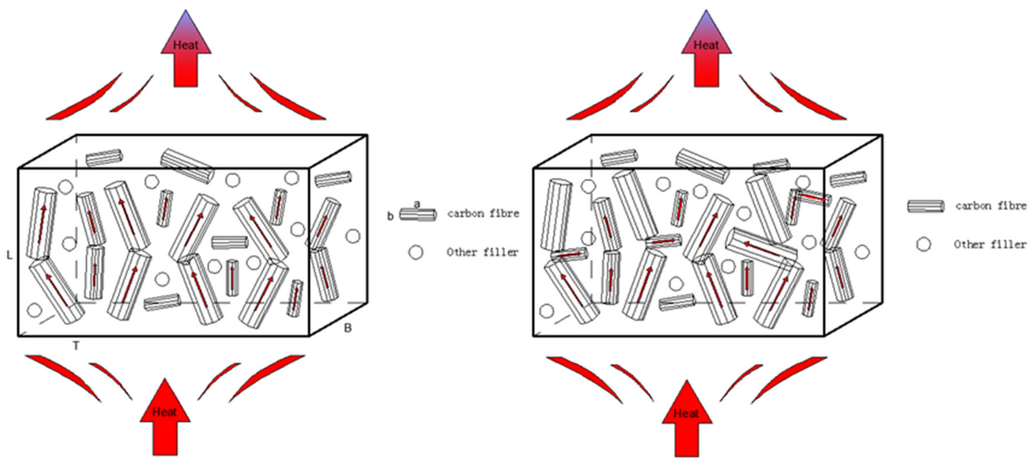


Figure 4. Scanning electron microscopy (SEM) micrographs of different mass percentages of CF.



(**a**) (**b**)

Figure 5. Schematic diagram of coating and effect of heat dissipation, (**a**) the mass fraction of CF is at critical value (red arrows represent the direction of heat transfer); (**b**) Excess CF in which *L* is the film thickness, *T* is the length, *B* is the width, CF parameters: Length *a*, diameter *b* (m).

The theoretical thermal conductivity of the coating [18] can be expressed as:

$$\lambda = K \times \mu \times \pi(b/2)/(T \times B) \tag{1}$$

$$A = \phi LTB/\left[\pi \times a \times (b/2)^2\right] \tag{2}$$

In which λ is the thermal conductivity (W/m·K), is the filler ratio of the thermal conduction, K is the axial thermal conductivity of the filler (W/m·K), A is the number of fillers, ϕ is the filler volume fraction.

According to the dimensionless equation, the theoretical thermal conductivity of the coating canbe derived by the filler material with the aspect ratio a/b and the axial thermal conductivity of K. The single filler volume can be obtained by the length and diameter of the filler, and the number of fillers can be determined by the volume fraction of the filler. Because there is only a good connection between the upper and lower surface of the coating, the filler can play a role in thermal conductivity, thus μ depends on the CF's content and arrangement. According to the bottom area of the single filler, the total bottom area with thermal conductivity can be derived. Then the proportion of the filler that acts as a heat conductor in the total area can be obtained. The theoretical thermal conductivity of the coating is this proportion multiplied by the effective thermal conductivity of the length of filler.

In this experiment, the main filler is CF. With the coating of the CF mass fraction of 11.6 wt % taken as an example, the theoretical thermal conductivity was calculated as follows: The calculated ϕ value of CF is 12.9%. The μ value is about 10% measured by SEM. The experimental result [18] shows that the coating has good thermal conductivity only when the thickness of the film is similar to the length of the CF, so this makes L approximately equal to a. The proportion of CF that acts as a heat conductor in the total area is calculated as 1.6%. The axial thermal conductivity of CF is 120 W/m·K, so the theoretical thermal conductivity of the coating is 1.76 W/m·K. The experimental results show that the average thermal conductivity of the coating is 1.89 W/m·K.

Figure 6 compares the theoretical value with the actual test results of the coating thermal conductivity. From Figure 6, when the CF content is too low or too high, the relative error of theory and practice is relatively large, thus the theoretical formula is not applicable. This paper analyzed the reasons for the difference and came to two possible conclusions: (1) With low CF content, the filler is not enough, and cannot constitute a "channel". In high CF content, the accumulation effect of filler appears, so that the CF is randomly dispersed in the coating and gradually decreases its orientation. A larger gap forms inside the coating, and hinders the formation of heat conduction channels. (2) The arrangement and proportion of the CF in the theoretical calculation are not properly set up.

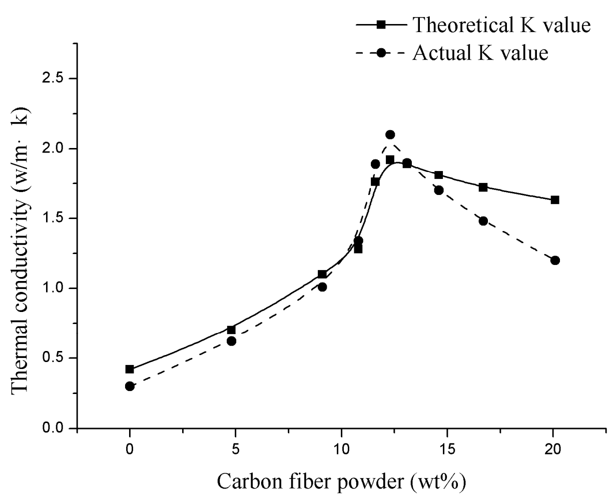


Figure 6. Comparison between theoretical value and actual test results of coating thermal conductivity.

4. Application Performance Testing

4.1. Coating Performance Index

The performance of the CF mass fraction of 12.3 wt % coating was measured. The results are listed in Table 3.

Table 3. The performance test results of the coating.

Test	Result
Hardness Impact resistance (cm)	H 50
Adhesion	Level 1
Temperature resistance	paint not taken off at 200 °C × 200 h

In the current research of heat transfer coating, the cooling effect of the coating is often more of a concern, and the mechanical properties of the coating are ignored; e.g., poor adhesion (less than 2), low hardness (less than B), and impact resistance. Although the thermal conductivity of the existing coating is relatively high (about 1.5), the coating falls off easily, and is not resistant to scrape and has a very small application value. It can be seen from the data in Table 3 that the coating in this paper has good mechanical and surface properties.

4.2. Application

A sample of heat sink was obtained by polishing, degreasing, and rusting treatment and then, the sample was sprayed with the target coating solution. The target coating solution was a kind of coating which was diluted to the appropriate viscosity by the diluent. After the coating was flattened, the sample was baked in an oven at 130 °C for 30 min.

In this experiment, the coating with the CF content of 12.3 wt % was selected as the experimental material, and the GY6-150 motorcycle cylinder (the motorcycle cylinder is Jingjiu gasoline engine cylinder by Yongkang Jingjiu power machinery Co., Ltd., Yongkang, China.) was used as the experimental specimen, as shown in Figure 7. The black one was sprayed with the coating of the heat dissipation, and the white one was sprayed with the coating without filler (as a control specimen). The test point of the cooling range is shown in Figure 7, where a and b are the two edges of the specimen, c is the center of the specimen. The cooling range of the coating was the temperature change of the test point a after 15 min (the difference between the test points b and c is not significant). Since the cooling curve of the blank specimen was not affected by the CF content, it could be used as a standard of measurement, and the difference caused by ambient temperature was eliminated.

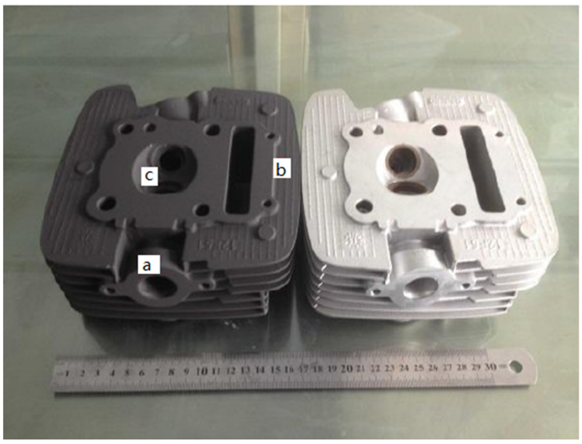


Figure 7. Experimental test cylinder.

Figure 8 is the comparison of the cooling curves with or without filler. The coated motorcycle cylinder shows faster cooling rate than the blank motorcycle cylinder. When they were cooled after 15 min, the temperature difference reached 15 °C, and this indicated that the coating of this paper had a significant cooling effect.

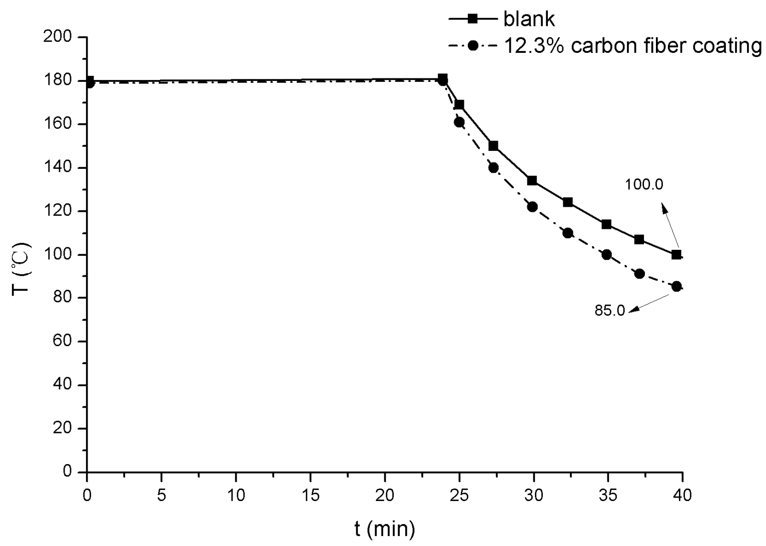


Figure 8. Comparison of cooling curves with or without filler.

5. Conclusions

According to the results of this study, the conclusions obtained are as follows:

- Single factor experiments indicate: With the increase of CF content, the thermal conductivity and cooling temperature range of the coating tend to increase rapidly and then decrease slowly. When the mass fraction of CF is 12.3 wt %, the coating has the best cooling effect.
- The SEM results of the coating show: With the increase of CF content, a "channel" is formed inside the coating. Due to the continuous overlap between the fillers, the "channel" forms a type of rapid thermal conduction. When the CF content is excessive, the simple random accumulation reduces the orientation of the fibers, and leads to the "channel" being weakened.
- When the CF mass fraction is 12.3 wt %, the thermal conductivity is the highest, and the coating has good weather resistance, high temperature resistance, and good insulation which meet the performance requirements of the coating application.

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Author Contributions: Jing Li and Yeming Liu conceived and designed the experiments; Yeming Liu performed the experiments; Yeming Liu and Xue Li analyzed the data; Xue Li, Chunlei Fan, Huan Yao and Xuyang Chen wrote the paper.

Conflicts of Interest: The authors declare no conflict of interest.

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