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CARBON FIBER THERMAL CONDUCTIVITY MEASUREMENT AND ANALYSIS

by

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CARBON FIBER THERMAL CONDUCTIVITY MEASUREMENT AND ANALYSIS

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Abstract

The paper describes the working principle of the laser flash method and the results of measuring the axial thermal conductivity of carbon fibers using the flash method. The relation between heat transfer and carbon fiber microstructure is discussed and the directional effect of the axial thermal conductivity of carbon fibers is suggested. The relation between the thermal conductivity of carbon fibers and that of carbon fiber reinforced composites is discussed and analyzed along with the problem of the anisotropy of carbon fiber reinforced composites.

Subject terms: carbon fiber, carbon fiber reinforced composite, thermal conductivity, and measuring techniques.

I. Background

With resin, carbon, metal, glass and ceramic, carbon fibers can form composite materials. The performance of these materials is closely related with the performance of the carbon fiber itself.

Heat transfer can be expressed in terms of thermal diffusivity (α) and thermal conductivity (λ); these two are related as follows:

$$\lambda=a\cdot\rho\cdot C_p \qquad (1)$$

In the equation, ρ is density and C_p is specific thermal capacity.

In carbon fiber thermal conductivity data given in certain reference papers, most cases are derived from the measured thermal conductivity of composite materials. However, since the thermal conductivity of composite materials is related not only to the ratio of base material substrate, but also closely to the existence of gas holes, fiber orientation and composition technique. Therefore, errors in the derived data are quite substantial.

Direct measurement of carbon fiber thermal conductivity is quite difficult; these difficulties have not been reported in China, and there are few such reports abroad. R. E. Taylor of TPRL in Purdue University (United States) conducted measurements with the direct electric conduction method and the laser pulse method [1]; however, there are widely different (more than 50 percent) measurement results reported by the different authors of these papers.

Recently, the laser pulse measurement method was employed by the author and his colleagues in analyzing the axial direction thermal conductivity problem of carbon fibers; this article will describe the measurement methods, discuss the relation between heat transfer and microstructure, as well as the relationship of heat transfer as between fiber composite material and fiber.

II. Laser Pulse Method of Measuring Axial Direction Thermal Conductivity of Carbon Fibers

The laser pulse method [2] [3] was proposed by W. J. Parker in 1961. Under the operating principle of the method, if there is a small circular sheet in an isothermal adiabatic environment, when the front face of the sheet is irradiated by a laser pulse, the temperature variation in the specimen can be expressed with the following formula:

$$T(x,t)=\frac{1}{L}\int_0^L T(x,0)\,dx+\frac{2}{L}\sum_{n=1}^{\infty}\exp\left(\frac{-n^2\pi^2}{L^2}at\right)\cos\frac{n\pi x}{L}\int_0^L T(x,0)\cos\frac{n\pi x}{L}\,dx \quad (2)$$

The rule is shown in Fig. 1.

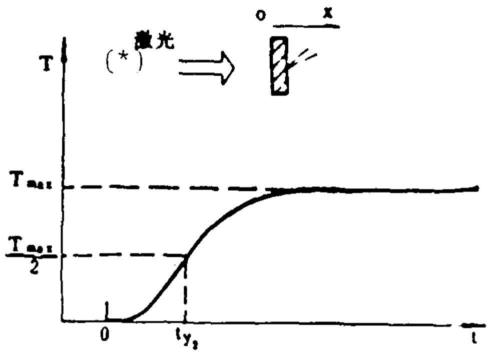

Fig. 1. Temperature rise curve at back of test specimen

Key: (*) Laser.

When the energy of a laser pulse is absorbed by a test specimen in its very thin surface layer, formula (2) can be simplified into:

$$\frac{T(L,t)}{T(L,\max)}=1+2\sum_{n=1}^{\infty}(-1)^n\exp\left(\frac{-n^2\pi^2 a}{L^2}t\right) \quad (3)$$

In the formula, L is the thickness of test specimen; t is time; T is temperature, and T(L, t) and T(L, max) are the rear face temperature and the highest rear face temperature of the test specimen.

When one half is made on T(L, t)/T(L, max), formula (3) is approximated as $\alpha = 0.139L^2/t_{1/2}$ (4)

In the equation, $t_{\frac{1}{2}}$ and $\frac{T(L,t)}{T(L,\max)}$ are the corresponding time of 1/2.

This method is highly regarded owing to the small test specimen, rapid measurement speed and high precision. However, whether this method is suitable to carbon fibers is a problem calling for serious discussion. In particular, appropriate methods should be adopted for preparation of the test specimen and measurement of rear face temperature.

As to test specimen preparation, the laser pulse method does not involve the cross sectional area of the test specimen during calculation; the primary necessity is absence of light leakage. The author and his colleagues applied the method by using a heat contraction tube to solidify multibundle fibers to first fabricate fiber rod. After a small ring made of solid adiabatic material was wrapped around the fiber rod, the rod was then cut into sheets. The feasibility of this specimen preparation method was experimentally confirmed by using fine copper filament as the test specimen.

As to the measurement of the rear face temperature, success was attained with the method of attaching a low-inertia thermocouple cemented sheet.

To prove the feasibility and reliability of the laser pulse method for measuring axial direction fiber thermal conductivity, measurements were made on electrical engineering grade, pure fine copper filament and T300 carbon fibers; the results are shown as follows:

Table 1. Determination Value of Axial Direction Thermal Diffusivity for T300 Carbon Fiber

第n次 (*)	a m²/s	第n次 (*)	a m²/s	第n次 (*)	a m²/s
1	0.0405×10^{-4}	2	0.0370×10^{-4}	3	0.0372×10^{-4}
4	0.0430×10^{-4}	5	0.0407×10^{-4}	6	0.0388×10^{-4}
7	0.0430×10^{-4}	8	0.0420×10^{-4}	9	0.0384×10^{-4}
10	0.0397×10^{-4}	11	0.0402×10^{-4}	12	0.0315×10^{-4}
13	0.0375×10^{-4}	14	0.0391×10^{-4}	15	0.0366×10^{-4}
16	0.0430×10^{-4}	17	0.0315×10^{-4}	18	0.0400×10^{-4}
19	0.0389×10^{-4}	20	0.0366×10^{-4}	21	0.0369×10^{-4}
22	0.0390×10^{-4}	23	0.0430×10^{-4}	24	0.0397×10^{-4}
25	0.0366×10^{-4}	26	0.0361×10^{-4}		
	$\bar{a}\pm s=(0.0382\pm0.0032)\times10^{-4}$, $\Delta\%=8.3$				

Key: (*) n-th time.

The determination value of thermal conductivity at room temperature for electrical engineering grade, pure copper filament is 232.37 W/(m·K); the literature value is 240.74 W/(m·k); the deviation value is 3.6 percent.

The result of measuring electrical engineering grade, pure copper filament proves the feasibility and reliability of the method. The determination result of T 300 carbon fiber reveals the reproducibility and repeatability of the measurement results. Thus, it is feasible in using laser pulse method to measure the axial direction thermal conductivity of carbon fiber. The outstanding advantage of this method is its avoidance of difficult problems in the conventional measurement methods of thermal conductivity that require precisely measuring the cross sectional area and temperature difference of test specimen.

III. Measurement Results of Carbon Fiber Axial Direction Thermal Conductivity and Carbon Fiber Microstructure

The above mentioned method was used in measurements of thermal diffusivities and thermal conductivities of several conventional types of carbon fibers; the results are listed in Table II.

From the following table, there is a considerable difference of thermal conductivities of carbon fiber for different filaments and different graphitization degrees; this phenomenon is closely related to carbon fiber microstructure.

Table II. Axial Direction Thermal Diffusivities and Thermal Conductivities of Carbon Fibers

(1) 纤 维 名 称	(2) 热扩散率 m^2/s	(3) 热导率 $W/(m\cdot K)$
(4) 石墨纤维M40	0.114×10^{-4}	15.83
碳纤维T300 (5)	0.0375×10^{-4}	4.90
(6) 国产聚丙烯腈碳纤维	0.0355×10^{-4}	4.44
美国粘胶基碳纤维CCA-3 (7)	0.0153×10^{-4}	1.65
(8) 国产粘胶基碳纤维	0.0127×10^{-4}	1.30
国产棉质粘胶基碳纤维 (9)	0.0068×10^{-4}	0.749
(10) 国产木质粘胶基碳纤维	0.0084×10^{-4}	0.967

Key: (1) Name of fiber; (2) Thermal diffusivity; (3) Thermal conductivity; (4) Graphite fiber; (5) Carbon fiber; (6) Polyacrylonitrile carbon fiber made in China; (7) Viscose base carbon fiber made in the United States; (8) Viscose base carbon fiber made in China; (9) Cotton viscose base carbon fiber made in China; (10) Woody viscose base carbon fiber made in China.

As a material of structural element, carbon fiber is a microcrystal composed of randomly laid carbon layers; carbon fiber is formed by carbonizing organic fibers. During thermal decomposition of organic fibers, noncarbon atoms are expelled;

the remaining carbon atoms in the adjacent molecular chains are rearranged into hexagonal sheet layer. Due to thermal traction and elongation, the chain shaped molecules will turn along the axial fiber direction to take on the orientation property of the fiber. During carbonization, the carbon content in the fiber gradually increases; the microstructure changes from random type to ordered type. With higher carbonization temperature, microcrystals grow steadily; the interlayer distance D002 gradually decreases. Variations in fine structure and composition state determine the difference in thermal conductivity of various carbon fibers. Table III lists the microstructure parameters* and heat transfer performance data of several types of carbon fiber.

Table III. Microstructure Parameters and Heat Transfer Property of Carbon Fibers

纤维名称 (1)	d002Å	Lc Å	La Å	n	V 10^4Å	α度 (2)	y= h/β	P %	a m²/s	λ W/(m·K)
M40	3.433	31.0	82.8	11	168	12.9	99.6	93	0.114×10^{-4}	15.83
PANT300	3.535	14.0	42.6	6	2.0	17.68	16.8	72	0.0375×10^{-4}	4.90
(3) 国产PAN碳纤维	3.526	13.9	34.0	6	1.3	18.70	12.1	75	0.0355×10^{-4}	4.44
(4) 粘胶基碳纤维	3.781	12.0	38.0	4	1.36	(5) 散乱	4.0	0	0.0127×10^{-4}	1.30

Key: (1) Name of fiber; (2) Alpha degree; (3) PAN carbon fiber made in China; (4) Viscose base carbon fiber; (5) Randomly scattered.

Heat transfer is based on the collision of microparticles and energy transfer inside an object; essentially, heat is conducted by the interaction and collision of phonons. Based on the theory of solid state physics, the thermal conductivity

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Footnote *: The microstructure parameters were measured by Comrade Wang Minrong of Beijing Institute of Materials Science.

phonon-based heat transfer can be calculated by using the following equation:

$$\lambda(T) = \frac{1}{3} C_v(T) \cdot \bar{v} \cdot l \qquad (5)$$

In the equation, C_v is the thermal capacity of phonon per unit volume; $\bar{v}$ is the average phonon velocity; and $\bar{l}$ is the average free path of a phonon.

Generally, variations in C_v and v are not large; however, $\bar{l}$ is an easily varied parameter; therefore, thermal conductivity is mainly determined by $\bar{l}$. The value of $\bar{l}$ is mainly determined by two scattering processes: one is the scattering caused by collisions among phonons, and the other is scattering at the crystal boundaries between phonons and crystal. Defects and collision of impurities cause scattering. Therefore the thermal conductivity of carbon fiber is not only a function of temperature, but is also closely related to the kinds of original filaments and the carbonization techniques. Or, in other words, the thermal conductivity of carbon fiber is closely related to microstructure, defects, gas holes and existing impurities (as factors) of carbon fibers.

As to the structural property of carbon fiber, its thermal conductivity exhibits apparent anisotropic and orientational preference. The thermal conductivity of carbon fiber is tens of times greater in the axial direction than in the diametral direction; however, axial direction thermal conductivity is closely related to the orientation degree of fiber microstructure. Table IV lists experimental data to prove this conclusion.

Table IV. Orientation Effect of Axial Direction Thermal Conductivity of Carbon Fiber

纤维名称 (1)	M40	T300	(2) 国产PAN	(3) 美国CCA-3	(4) 国产粘胶
(5) 取向角	12.90	17.68	18.70	—	乱 (6)
有序度 (7)	99.6	16.3	12.1	—	4.0
(8) 热导率	15.83	4.90	4.44	1.65	1.30

Key: (1) Name of fiber; (2) PAN made in China; (3) CCA-3 made in the United States; (4) Viscose made in China; (5) Orientation angle; (6) Randomly scattered; (7) Degree of orderliness; (8) Thermal conductivity.

IV. Relationship Between Heat Transfer Property of Carbon Fiber Composite Material and Thermal Conductivity of Carbon Fiber

Carbon fiber composite material is a multiphase system, where heat transfer is more complicated to analyze than the heat transfer mechanism of homogeneous materials; the heat transfer property is closely related to thermal conductivity. Citing an example of three kinds of composite materials (carbon/carbon, carbon/epoxy and carbon/phenol formaldehyde), research was conducted on the relation among them; the experimental data are listed in Figs. 2 through 5.

Fig. 2 shows thermal diffusivities and thermal conductivities of the three-dimensionally woven carbon/carbon materials. From the figure, we see that there are different thermal conductivities along the Z direction, x-y direction and 45 degree direction

$$a_z > a_{x-y} > a_{45^\circ}$$
$$\lambda_z > \lambda_{x-y} > \lambda_{45^\circ}$$

This phenomenon shows a wide difference in heat transfer properties of carbon fiber and deposited carbon in carbon/carbon composite material. Moreover, in the three-dimensionally woven materials the number of Z-direction carbon fiber bundles is the highest; the 45-degree direction is closely related to this weaving pattern with an unoccupied direction in the spatial distribution of carbon fiber.

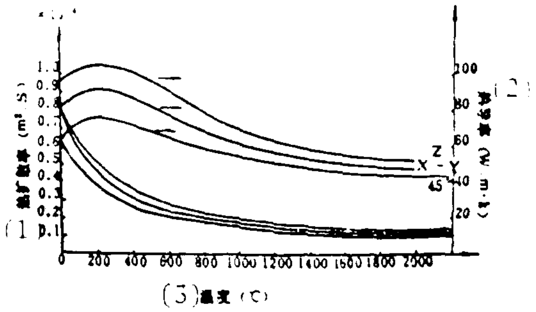

Fig. 2. Thermal diffusivities and thermal conductivities of carbon/carbon composite material

Key: (1) Thermal diffusivity; (2) Thermal conductivity; (3) Temperature.

Fig. 3 shows the thermal diffusivities of composite material along the x and y directions (parallel to fiber direction), the z direction (perpendicular to fiber layer) and 45 degree direction; the composite material is composed of resin in the orthogonal or single direction laying pattern of different kinds of carbon fiber. Curves 1, 2, 3 and 7 show, respectively, the thermal diffusivities of graphite fiber M40 and epoxy resin composite materials along the y, x, 45 degree and z directions. Since the test specimen is orthogonal laid of nonwoven cloth, the thermal diffusivities along the x and y directions are much higher than the thermal diffusivity along the z direction. However, diffusivities along the x and y directions are relatively close in value; diffusivity along 45 degree direction is considerably smaller than that along the x and y directions. Curves 4, 8 and

10 show, respectively, the thermal diffusivities of composite material with polyacrylonitrile carbon fiber and 648 epoxy formaldehyde resin along the x, y and z directions.

Since the test specimen is nonwoven cloth laid in a single direction, the thermal diffusivity parallel to the x direction of the fiber is much greater than in the y and z directions. Curves 11, 12 and 13 are the thermal diffusivities in the x, y and z directions, respectively, of composite material of rayon-based carbon fiber and formaldehyde resin.

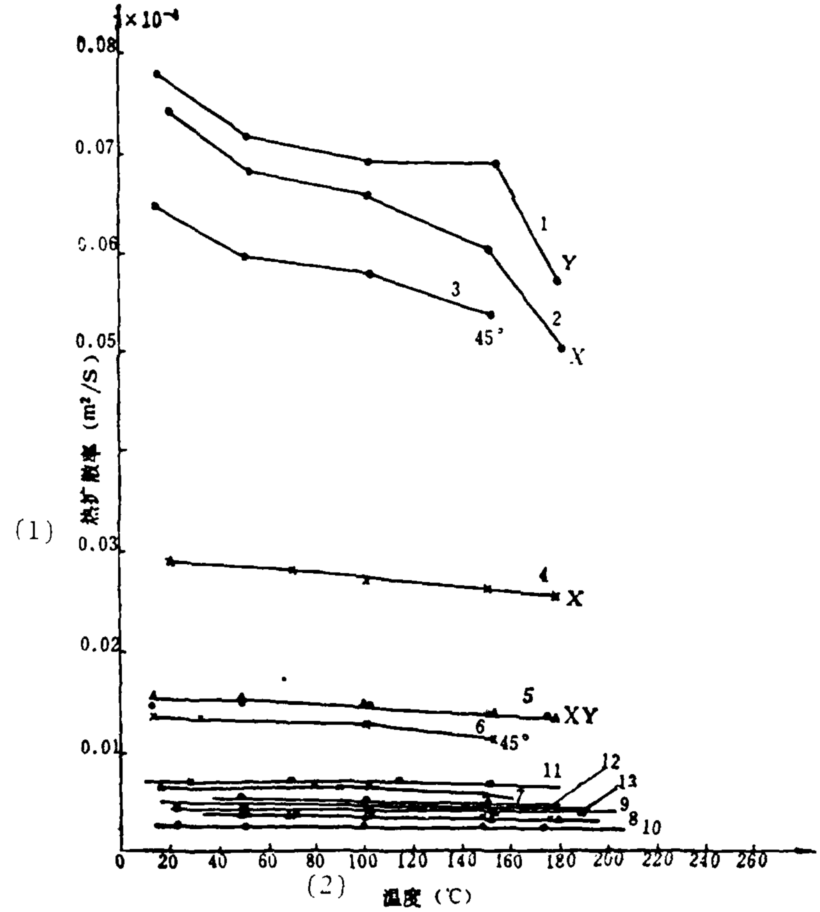

Fig. 3. Relation between thermal diffusivity and temperature of carbon fiber composite material.

Key: (1) Thermal diffusivity; (2) Temperature.

Fig. 4 shows the relation between the heat transfer property and the fiber laying direction of polyacrylonitrile carbon fiber and epoxy resin composite material. The figure shows the thermal diffusivity of heat flow and carbon fiber at the included angles of 0, 30, 45, 60 and 90 degrees. Fig. 5 shows the relation curves of thermal diffusivity and included angle θ.

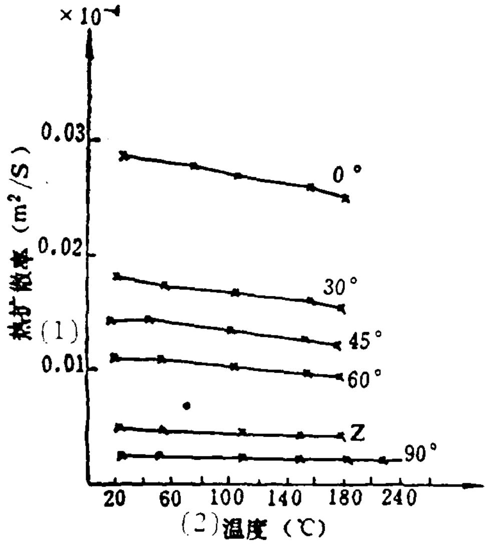

Fig. 4. Relation between thermal diffusivity and carbon fiber orientation

Key: (1) Thermal diffusivity; (2) Temperature.

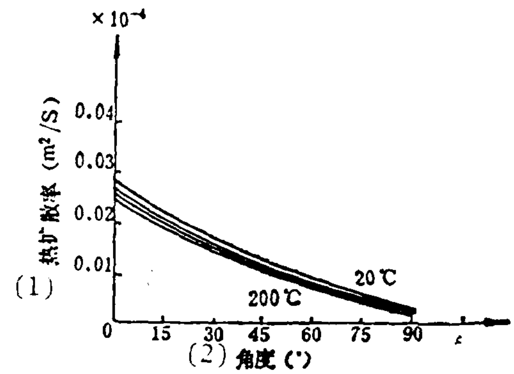

Fig. 5. Relation between thermal diffusivity on the one hand, and the included angle between heat direction and fiber axis, on the other

Key: (1) Thermal diffusivity; (2) Angle.

From the figures, the heat transfer property of carbon fiber composite material exhibits apparent anisotropy; this anisotropy can be expressed with $\lambda_{\parallel}/\lambda_{\perp}$ and $a_{\parallel}/a_{\perp}$ values. $\lambda_{\parallel}$, $a_{\parallel}$, $\lambda_{\perp}$ and $a_{\perp}$ are, respectively, the thermal conductivity and thermal diffusivity parallel (or perpendicular) to the fiber direction. Therefore, the thermal conductivity of carbon fiber composite material is not only a function of temperature, but also a function of spatial distribution; particular attention should be given when designing components with intricate shapes.

The anisotropy of heat transfer in carbon fiber composite material is closely related to the thermal conductivity of the fiber as such. Citing an example of a composite material using M40 graphite fiber, polyacrolonitrile carbon fiber and viscose base carbon fiber with resin, its $\alpha_{\parallel}/\alpha_{\perp}$ are, respectively, 10.85, 4.29 and 1.26. Moreover, the interdependent relation between the heat transfer properties of composite material and fiber in different directions is dissimilar; the relation is especially apparent for the case parallel to fiber direction. Citing an example of the three above mentioned materials: $\alpha_{\parallel(A)}:\alpha_{\parallel(B)}:\alpha_{\parallel(C)}=10.93:2.14:1$, $\alpha_{\perp(A)}:\alpha_{\perp(B)}:\alpha_{\perp(C)}=1.27:0.85:1$, $\alpha_{F(A)}:\alpha_{F(B)}:\alpha_{F(C)}=12.15:3.75:1$. In the equations, A, B and C represent, respectively, M40, polyacrylonitrile and viscose base carbon fiber; α_F represents the axial direction thermal diffusivity of the fiber as such. It is apparent that variation of $\alpha_{\parallel}$ is quite large, parallel to the fiber direction; moreover, the ratio is very close to that of the α_F ratio. This phenomenon can be explained by the Kingery theory [4]; the structure of carbon fiber resin composite material is quite similar to that of the phase distribution model of Kingery's parallel mode. The total heat transfer parallel to the layer direction corresponds to the heat transfer $\lambda_{\parallel}=V_1\lambda_1+V_2\lambda_2$ of the parallel circuit of two kinds of layers. The total heat transfer perpendicular to the layer direction corresponds to heat transfer $\lambda_{\perp}=V_1/\lambda_1+V_2/\lambda_2$ of the series circuit of two layers. In the equation, $\bar{V}$, $\bar{V}$, λ_1, and λ_2 are, respectively, the volume percentage content and thermal conductivity of two kinds of layer surface. When $\lambda_1 \gg \lambda_2$, $\lambda_{\parallel}$ is mainly related to λ_1; however, $\lambda_{\perp}$ is mainly determined by λ_2. This situation should receive close attention in designing materials.

V. Conclusions

The laser flash method is a feasible method of measuring the axial direction thermal conductivity of carbon fiber; the anisotropy exhibited by carbon fiber heat transfer has an apparent orientational preference; all the above mentioned factors are closely related to fiber microstructure. The heat transfer property of carbon fiber composite material is also anisotropic; this anisotropy can be expressed with $\alpha_{\parallel}/\alpha_{\perp}$ and $\lambda_{\parallel}/\lambda_{\perp}$. The value is closely related to the heat transfer property of carbon fiber itself and orientation of the fiber.

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