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The laser flash method has been used to measure thermal conductivities in a porous metal fiber material at room and elevated temperatures. The anisotropy of heat conduction in this material is reported and a novel technique using a focused laser beam is described to access directly the off-diagonal components of the conductivity tensor. A simulation study shows the anisotropy to be determined by the layered structure of the material. Enhanced conductivities are reported at elevated temperatures and are shown to be due to radiation heat transfer within the porous structure.

I. INTRODUCTION

Porous materials formed from sintered metallic fibers find applications in many fields including filtration and thermal insulation. Our particular interest in these materials is their use as flame holders for surface combustion gas burners.[1] The thermal properties of such materials are not well characterized. Highly porous materials have low thermal conductivities which only vary slightly with the solid component.[2] At high temperatures radiation heat transfer occurs within the pores, increasing the effective thermal conductivity. For example, at temperatures above 1000–1500 K, heat transfer in fibrous ceramic insulation materials is predominantly radiative.[3] If the fibers are arranged in layers then a large anisotropy is expected because conduction within a layer is through bulk fibers, whereas perpendicularly it is inhibited by the high thermal resistance of the contacts between fibers. Tye[4] reports measurements of thermal and electrical conductivities of porous metals and notes anisotropy in samples made from fibers. Kostornov and Galstyan[5] also report thermal conductivities for porous metal fiber materials and show that the highest conductivity is found in the direction perpendicular to the direction of pressing.

The classical methods for measuring the thermal conductivity of solids are based on steady-state heat flow.[2,6] Fairly large samples are required so that one-dimensional approximations may be made and isotropy is assumed. By contrast, the laser flash method used in this work requires only small amounts of sample material, may be used to examine thermal anisotropy, and is readily extended to high temperatures. Parker *et al.*[7] were the first to employ the flash method in which a pulse of thermal energy is supplied to the front surface of a sample and the temperature response of the rear surface is monitored. The thermal diffusivity of the material is calculated from a characteristic time, obtained from the rear face temperature record and the sample thickness. The laser flash method has been used to study heterogeneous dense materials such as fiber reinforced composites[8] and measurements with sponge iron[9] have shown that the technique can also be successfully applied to porous materials.

In this paper we describe laser flash measurements of thermal conductivity for a nonwoven metallic fiber material. The experiments were carried out using a fiber material of a refractory alloy developed for burner applications but the method is applicable to other materials of similar structure.

In Sec. II we describe the material used and show that the layered construction leads to a thermal anisotropy with poor conduction between the layers. In Sec. III we describe the use of the laser flash technique to determine thermal conductivity at room and elevated temperatures. We present a novel method for the direct measurement of off-diagonal conductivity elements. The results are presented and discussed in Sec. IV and compared with a simulation of the conductivity components based on an ordered stacked-fiber model. The radiation enhancement of conductivity observed at elevated temperatures is compared with theory.

II. DESCRIPTION OF MATERIAL

The sintered metal fiber material used, Bekitherm,[10] has been developed and is produced by N.V. Bekaert S.A. The fibers used in this study are of a refractory steel, Fecralloy,[11] with a diameter of 22 μm. The alloy forms a protective alumina coating on the fibers. The structure of the material can be seen in Fig. 1, a scanning electron micrograph of the surface. The fibers are randomly oriented in layers. The material has a porosity of 80% and is produced in sheet form several millimetres thick.

The layered construction of this type of material suggests an ordered model with alternate layers of fibers oriented mutually orthogonally to one another.[12] The fibers must touch longitudinally so that the arrangement, Fig. 2(a), corresponds to a unit cell with longitudinal (z) repeat distance, S_l. Each unit cell has a volume

$$V = S_l S_t^2 = 2 \, dS_t^2 \qquad (1)$$

and contains a solid fiber volume

$$V_s = \pi \, d^2 S_t / 2. \qquad (2)$$

Using the dependence of porosity P on the solid volume

$$P = (V - V_s)/V \qquad (3)$$

one obtains the mean transverse spacing S_t in a layer expressed as a multiple of the fiber diameter:

$$n = S_t/d = \pi/4(1 - P). \qquad (4)$$

For our 80% porous material the mean transverse fiber spacing is about four fiber diameters.

The fiber arrangement shown in Fig. 2(a) would allow normally incident radiation to penetrate straight through the material. A better representation is achieved if each al-
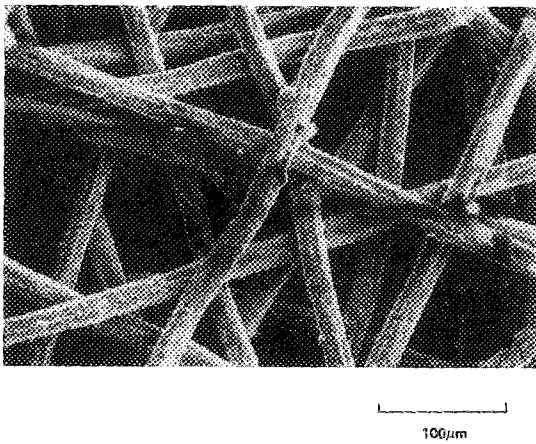
FIG. 1. Scanning electron micrograph of metal fiber material.

ternative, i.e., similarly oriented, layer is translated with respect to one another [Fig. 2(b)]. A natural translation "unit" is the fiber diameter d and such translations leave the configuration invariant. In this arrangement radiation cannot penetrate directly below the $(2n - 1)$th fiber layer, and thus, assuming reasonably high absorptions so that only weak multiple reflections take place, the penetration depth of incident radiation is

$$\delta = (2n - 1)d. \qquad (5)$$

The emissivity of the oxide-coated metal fibers is about 0.6 and within the bounds of the Kirchoff approximation we
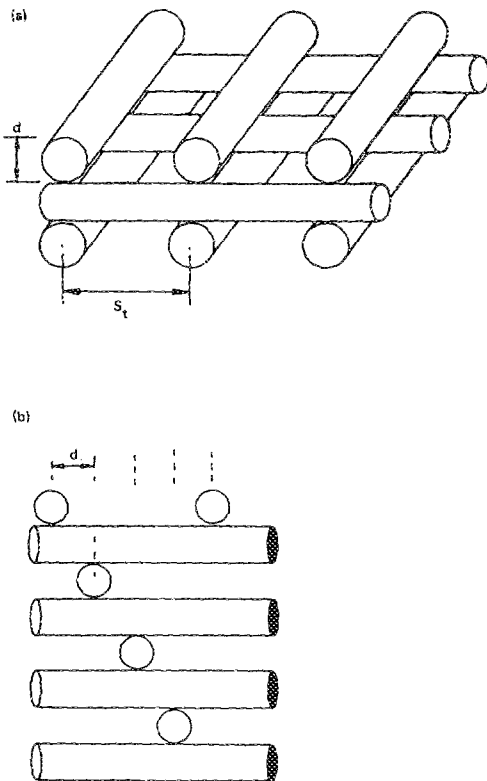

FIG. 2. Models of ordered fiber arrangement. (a) Regular stacked array. (b) Successive layers displaced to simulate random layer distribution.

calculate that only about 2% of incident radiation would penetrate further than δ. The model configuration described above is used in Sec. IV B to simulate the conduction experiments.

Thermal conduction in the fiber material must be anisotropic because the randomly layered construction (see Fig. 1) confers $D_{\infty h}$ symmetry.[13] For conceptual simplicity our model only possesses near D_{4h} symmetry but it will be demonstrated that this is a good approximation. The four elements of the thermal conductivity tensor to be studied (which are not all independent) are λ_{xx}, λ_{zz}, λ_{xy}, and λ_{xz}.

III. MEASUREMENTS

The laser flash technique requires the front surface of the sample to be heated within a time interval that is short compared to the time required for the resulting thermal transient to propagate through the sample. Solution of the heat conduction equation with the appropriate boundary conditions shows the temperature response of the rear surface to be of the form:[7,13]

$$T(t) = T_{\max}\left[1 + 2\sum_{n=1}^{\infty} (-1)^n \exp - \left(\frac{n^2\pi^2}{l^2}\alpha t\right)\right] \qquad (6)$$

where α is the thermal diffusivity and l is the thickness of the sample. Setting $T(t_{1/2}) = \frac{1}{2}T_{\max}$ we obtain:

$$T_{1/2} = 0.138l^2/\alpha = (0.138l^2/\lambda)\rho C_p, \qquad (7)$$

where λ is the thermal conductivity, ρ is the density, and C_p is the heat capacity.

If the laser radiation is able to penetrate the sample to a depth δ then Bretzlaff[14] has shown that Eq. (7) must be modified thus:

$$t_{1/2} = \frac{0.138l^2}{\alpha} - \frac{4l^2}{\pi^2\alpha}\ln\left(1 + \frac{\pi^2\delta^2}{4l^2}\right). \qquad (8)$$

In Sec. II we showed that the penetration depth δ for the material can be approximated by Eq. (5). For the 80% porosity material made from 22-μm-diam fibers we calculate that the shortfall in $t_{1/2}$ caused by penetration of the beam is less than $\frac{1}{2}$%. Thus, the simpler expression (7) may be used to calculate the thermal conductivity of this material from the measured times for the rear surface to reach half of the maximum temperature rise.

The experimental configuration is shown in Fig 3. The heat source was a 1.8-kW CO_2 infrared (IR) laser beam of 1.1 cm diam which was electromechanically chopped to pro-
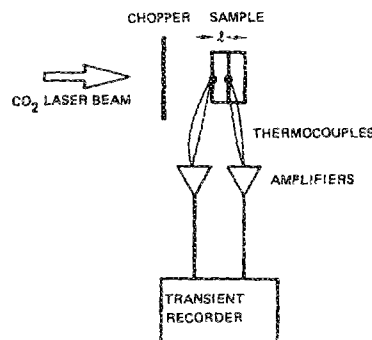

FIG. 3. Schematic of laser flash thermal conductivity experiment.

duce pulses of 30-ms duration. The beam profile was checked for uniformity to ensure that the sample face was reasonably uniformly irradiated.

The sample sizes used were different depending on which component of conductivity was being studied. For λ_{zz}, the component of primary interest to us, the sample was slightly larger than the beam and 2.55 mm thick. The rear face temperature rise (typically 5–10 K) was monitored using a 200-μm bare wire NiCr/NiAl thermocouple sandwiched between two identical pieces of material to achieve a good thermal contact. The signal was amplified 500 times and recorded on a transient recorder. A similar thermocouple placed above the surface of the sample triggered the transient recorder and provided the time origin for the rear face response. For completeness the thermal conductivity of solid Fecralloy was also measured using this technique. In this case the rear face thermocouple was spot welded directly to the metal.

In order to assess the symmetry of the material, in particular the randomness of the fiber arrangement within a layer, it is necessary to measure the off-diagonal components of the conductivity tensor. Irradiation of a complete face of the sample does not permit this because there is no unique path through which conduction takes place and which defines the thermal diffusion length. We have avoided these problems by using a cylindrically focused beam, Fig 4. An 18.6-cm focal length zinc selenide cylindrical lens was used to focus the beam to a line of width 0.03 cm on one side of the sample. When oriented along the k th axis, heat conduction takes place via the element λ_{ij}, where $i \neq j \neq k$. The transverse position of the beam (perpendicular to the beam propagation direction) was obtained by comparing the thermocouple responses on either side of the sample.

Measurements of λ_{zz} at elevated (up to 900 °C) temperatures were obtained by mounting the sample inside an electrically heated tubular furnace. The rear-face thermocouple response was offset by a second circuit junction maintained at the furnace temperature. This enabled good discrimination between the small temperature rise of the rear face and the high temperature environment.

IV. RESULTS AND DISCUSSION

A. Room-temperature measurements

Using a nonfocused beam for face irradiation the conductivities λ_{zz} and λ_{xx} were measured. Figure 5 shows the rear-face thermocouple response for a sample of fiber material 2.55 mm thick, λ_{zz} measurement. The $t_{1/2}$ time is 4.15 s.

The density and heat capacity for the 80% porous fiber material were taken as 1460 kg/m^3 and 460 J/kg K, respectively, based on values for solid Fecralloy of 7300 kg/m^3 and 460 J/kg K taken from the manufacturers literature.

From the experimental data the thermal conductivities λ_{zz} and λ_{xx} were calculated to be 0.13 W/mK and 1.15 W/mK, respectively. These values are the mean of several measurements. We estimate the accuracy to be about $\pm 10\%$. The relative anisotropy $\lambda_{xx}/\lambda_{zz} = 8.8$ shows that the thermal response within a layer is relatively fast compared to that between layers. The focused beam technique was first tested on a sample of solid (isotropic) fecralloy and yielded the same result, $\lambda_s = 9.5$ W/mK, as that obtained by face irradiation using the nonfocused beam method.

A measurement of λ_{xy} in the fiber material gave a value of 1.18 W/mK. The close similarity to the value of λ_{xx} confirms our hypothesis that the actual $D_{\infty h}$ fiber arrangement could be approximated by a D_{4h} model.

The remaining conductivity tensor λ_{zx} is more complex to define as it depends on the longitudinal position of the line heating pulse. By comparing the thermocouple responses as regards the longitudinal position, Fig. 4, it can be shown that

$$t_{1/2}^{ad} - t_{1/2}^{bd} = (0.138/\alpha_{zz})(a^2 - b^2) \tag{9}$$

and

$$\frac{t_{1/2}^{ad} - 0.138d^2/\alpha_{xx}}{t_{1/2}^{bd} - 0.138d^2/\alpha_{xx}} = \frac{a^2}{b^2}. \tag{10}$$

α_{xx} can thus be found and compared with the value obtained above.
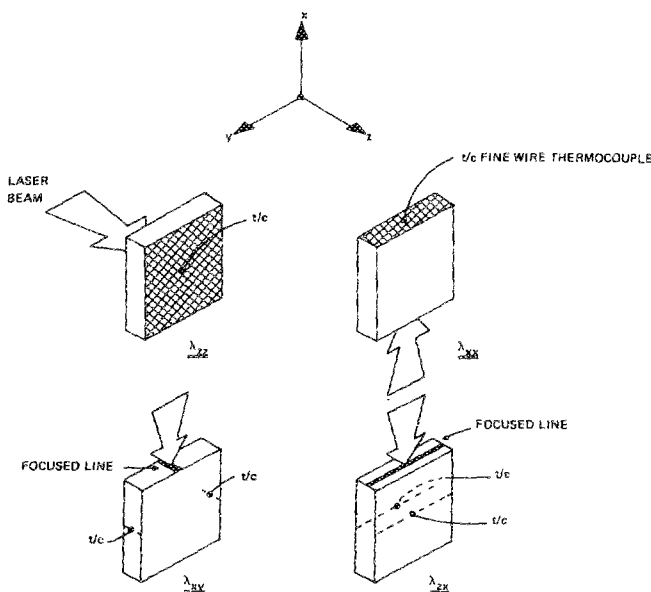
FIG. 4. Laser beam and sample orientations used to access thermal conductivity components λ_{zz}, λ_{xx}, λ_{xy}, λ_{zx}.
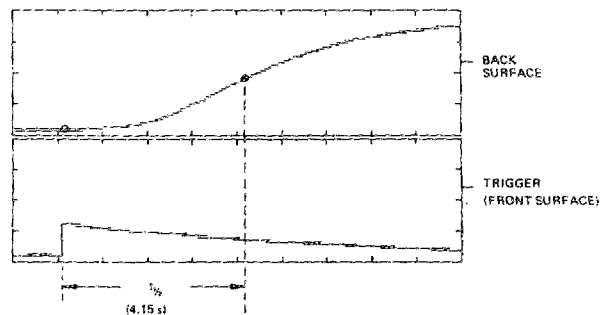

FIG. 5. Temperature rise of rear sample face during measurement of λ_{zz} for metal fiber material, thickness $l = 2.55$ mm.
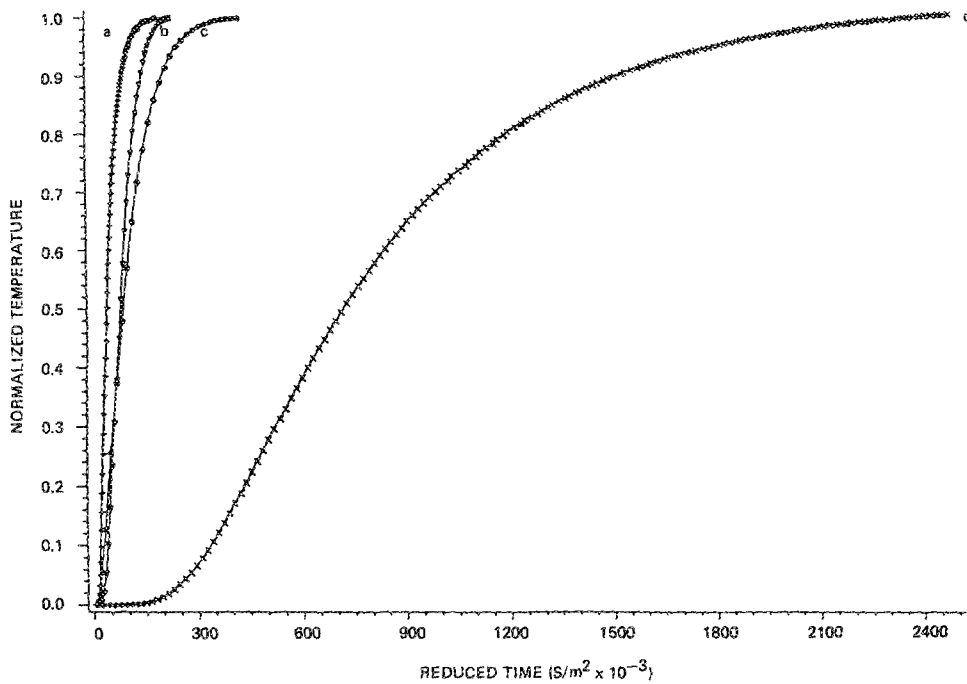
FIG. 6. Thermal diffusion components simulated in an 80% porous material of 22-μm metal fibers and for comparison thermal diffusion in the solid metal; (a) solid metal; (b) xx component fibrous material; (c) xy component fibrous material; (d) zz component fibrous material.

The conductivity λ_{xx} found by this method was 1.09 W/mK, in good agreement with the value of 1.15 W/mK obtained directly by face-to-face heat transfer.

B. Conductivity simulation

The thermal conduction experiments described above have been modeled using the ordered fiber configuration shown in Fig. 2(b) with spatial increments Δr_i equal to the fiber diameter. Application of the thermal diffusion equation to this spatial network yields the following numerical algorithm[15]:

$$T(t + \delta t) = \sum_{i=1}^{6} \mu_i \frac{A_i}{A_l} T_i(t) + \left(1 - \sum_{i=1}^{6} \mu_i \frac{A_i}{A_l}\right) T(t),$$

(11)

where $\mu_i = \alpha \delta t / \Delta r^2$ and the subscript refer to the value at the ith octahedral mesh point with respect to the central point of interest. One of the contributions to the anisotropy arises from the different cross-sectional areas for transverse and longitudinal heat flow. In the transverse direction, $A_i = A_t = \pi d^2 / 4$ for $i = 3$–6, i.e., the four surrounding points transversely located in the same plane. In the longitudinal direction conduction is through the sintered joints, $A_i = A_l = f^2 (\pi d^2 / 4)$ for $i = 1,2$, i.e., the points below and above the point of interest, f is the ratio of the sinter contact diameter to fiber diameter.

Using a value of $f = 0.5$ at every point the model correctly predicts the observed λ_{zz} conductivity. Under the scanning electron microscope we observed that a typical sinter diameter was about $\frac{2}{3}$ of a fiber diameter but it was also clear that not all of the contact points are sintered.

Our computations using Eq. (11), modified for boundary conditions at the sides, edges and corners,[15] were performed for a fiber structure containing 1000 mesh points. Thermal conduction was initiated by a 2-μs heat pulse rais-

ing one face of the material to 100 °C. The temperature of each mesh point is calculated every 2 μs until convergence within 0.1% is obtained. Equation (7) was then used to calculate the thermal conductivity.

By applying varying orientations of linear heat pulses to different faces, temperature profiles were obtained for all of the λ_{ij} elements. These are shown in Fig 6. The y axis represents the normalized rear-face temperature, while for the x axis the time has been divided by the square of the diffusion path length l so that the results are independent of sample thickness. Curve (a) shows the rapid temperature rise for the solid metal. Curve (b) represents transverse heat flow in the fiber material along solid fibers within a layer and characterized by λ_{xx}. For xy flow, curve (c), heat must pass through a sinter contact at the crossover between x and y oriented fibers. There is only a slight slowing down for xy compared to xx flow because only one sinter contact need be used in any particular diffusion path.

For longitudinal zz flow, curve (d), the temperature rise is slowest because of the high thermal resistance between layers. The ordering of the thermal conductivities is, thus,

$$\lambda_{zz} \ll \lambda_{xy} \sim \lambda_{xx} < \lambda_s.$$

Their calculated values are shown in Table I together

TABLE I. Measured and simulated flash conductivities in 80% porous 22-μm-diameter sintered metal fiber material.

	Observed (W/mK)	Calculated (W/mK)
λ_s	9.5	$\cdots$
λ_{xx}	1.15	1.10
λ_{xy}	1.18	1.22
λ_{zz}	0.13	0.14

1974   J. Appl. Phys., Vol. 63, No. 6, 15 March 1988

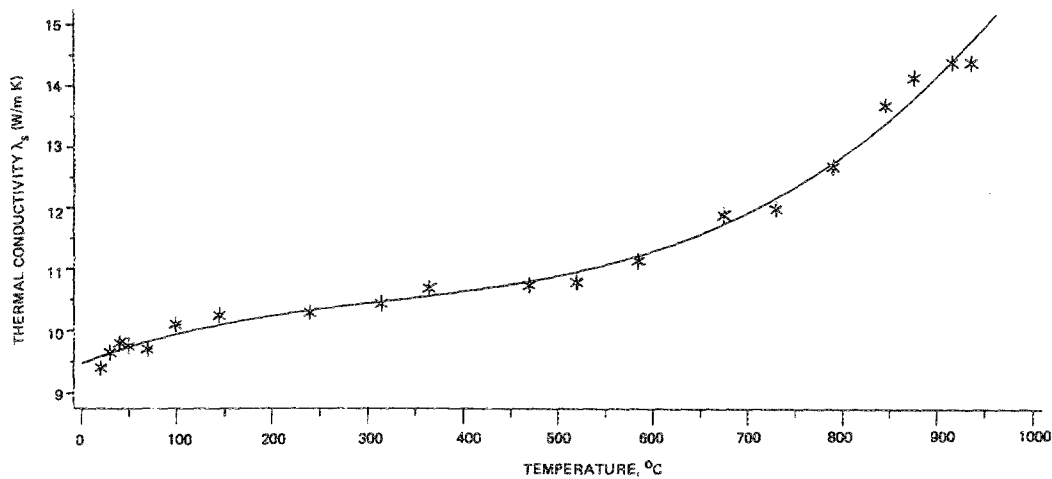M. Golombok and L. C. Shirvill   1974

FIG. 7. Thermal conductivity of solid Fecralloy (λ_s) as a function of temperature.

with the equivalent experimentally determined values. The agreement is satisfactory. This simulation can also be applied to the calculation of thermal conductivities at different porosities and with different diameter fibers.

C. Elevated temperature measurements

At elevated temperatures radiation heat transfer within the pores of the fiber material is of increasing importance and thus the thermal conductivity is expected to increase with temperature. There will also be an increasing contribution from increased electron carrier mobility within the metal fibers themselves. A necessary preliminary was therefore to characterize thermal conductivity as a function of temperature for the solid material. The results, (Fig. 7) show a 50% increase in thermal conductivity of the solid between room temperature and 900 °C.

Figure 8 shows the results of λ_{zz} measurements for the metal fiber material between room temperature and 900 °C. The thermal conductivity increases from a value of 0.13

W/mK already quoted at room temperature to 0.28 W/mK at 900 °C.

It is reasonable to assume that the conductivity behavior measured for the solid will apply to the individual fibers. The conductivity in the fiber material which is due purely to true conduction is determined by the ratio of fibrous material conductivity λ_{zz} to solid conductivity λ_s at a reference temperature where there is negligible radiant contribution to conduction in the fiber material. We take the ratio at room temperature $\lambda_{zz} / \lambda_s = 1/73$ as constant to describe the temperature variation of nonradiant conduction in the fiber material.

Cabannes[3] has shown that the radiation component of conductivity in porous materials λ_{rad} is of the form

$$\lambda_{rad} = BT^3, \tag{12}$$

where B is a constant depending on the material properties. The total conductivity is then given by

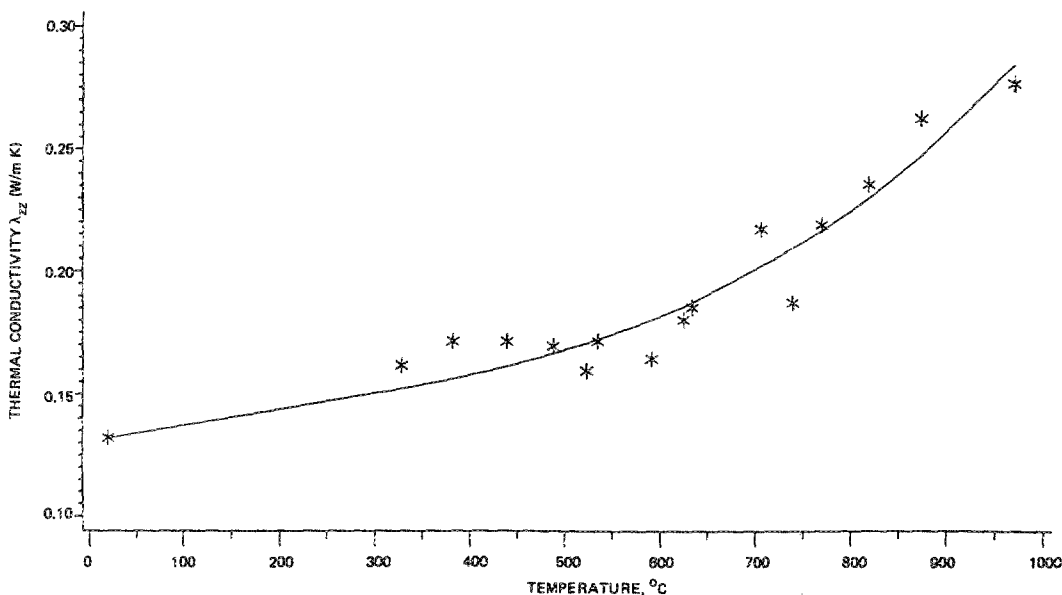$$\lambda(T) = \tfrac{1}{73}\lambda_s(T) + BT^n. \tag{13}$$

FIG. 8. Thermal conductivity (λ_{zz}) through 80% porous metal fiber material made from 22-μm Fecralloy fibers as a function of temperature. (*) Experimental data; (–) $\lambda(T) = \tfrac{1}{73}\lambda_s(T) + 2.5\times10^{-11}T^{2.99}$.

Using the data for the solid material given in Fig. 7 a least-squares fit of the experimental data, (Fig. 8) yields $n = 2.99$, in good agreement with Eq. (12). The value of $B = 2.5 \times 10^{-11}$ is comparable to that found for ceramic fiber insulation materials.[3]

V. CONCLUSIONS

(1) The laser flash method has been successfully used to determine the anisotropic thermal conductivities of a porous metal fiber material. The conductivity in a direction normal to the lay of the fibers, λ_{zz}, was found to be 8.8 times smaller than within the layers, λ_{xx}, and 73 times smaller than the conductivity of the solid metal.

(2) By focusing the laser beam with a cylindrical lens the off-diagonal components of the thermal conductivity tensor, λ_{xy} and λ_{zx}, can be directly accessed.

(3) Numerical simulations of heat conduction using an ordered model representation of the material gave results that are in good agreement with the experimental data.

(4) Measurements at elevated temperatures show that the thermal conductivity of the fiber material is greatly enhanced by radiation heat transfer through the pores. Comparison with values for the solid metal, obtained by the same method, show a T^3 dependence for the radiant component consistent with theory.

[1]L. C. Shirvill, in *Proceedings of the 1986 International Gas Research Conference* (Government Institutes, Inc., Rockville, MD, 1987), p. 837.
[2]R. P. Tye, *Thermal Conductivity* (Academic, London, 1969).
[3]F. Cabannes, Rev. Int. Hautes Temper. Refract. **17**, 120 (1980).
[4]R. P. Tye, ASME Report No. 73-HT-47 (1973).
[5]A. G. Kostornov and L.G. Galstyan, Porosh. Metall. **3**, 88 (1984).
[6]ASTM C177-85 Standard test method.
[7]W. J. Parker, R. J. Jenkins, C. P. Butler, and G. L. Abbot, J. Appl. Phys. **32**, 1679 (1961).
[8]R. E. Taylor, J. Jortner, and H. Groot, Carbon **23**, 215 (1985).
[9]H. W. Gudenau, H.A. Friedricks, and P. K. Rademacher, Arch. Eisenhuttenwes **52**, 261 (1981).
[10]Bekitherm is a trademark of N.V. Bekaert S.A, Zwevegem, Belgium.
[11]Fecralloy is a trademark of UKAEA, Didcot, England.
[12]H. M. Strong, F. P. Bundy, and H. P. Bovenkerk, J. Appl. Phys. **31**, 39 (1960).
[13]H. S. Carslaw and J. C. Jaeger, *Conduction of Heat in Solids* (Oxford University Press, Oxford, 1959).
[14]R. S. Bretzlaff, J. Appl. Phys. **58**, 2816 (1985).
[15]F. M. White, *Heat Transfer* (Addison-Wesley, Reading, MA, 1984).