

# Thermal Conductivity, Heat Capacity, and Thermal Diffusivity of Selected Commercial AlN Substrates<sup>1</sup>

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The thermal transport properties of four commercially available AlN substrates have been investigated using a combination of steady-state and transient techniques. Measurements of thermal conductivity using a guarded longitudinal heat flow apparatus are in good agreement with published room temperature data (in the range  $130\text{--}170 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ ). Laser flash diffusivity measurements combined with heat capacity data yielded anomalously low results. This was determined to be an experimental effect for which a method of correction is presented. Low-temperature measurements of thermal conductivity and heat capacity are used to probe the mechanisms that limit the thermal conductivity in AlN.

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**KEY WORDS:** aluminum nitride; heat capacity; substrate; thermal conductivity; thermal diffusivity.

## 1. INTRODUCTION

Aluminum nitride possesses a superior combination of physical properties which make it an excellent candidate for high-power, high-speed integrated circuit substrates. These properties include a high thermal conductivity, a high electrical resistivity, and a coefficient of thermal expansion similar to that of silicon. Slack [1] calculates the theoretical thermal conductivity of single crystal aluminum nitride to be  $319 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$  at room temperature. This high value is attributed to the simplicity of the crystal structure, its low atomic mass, the existence of strong covalent bonding between the species, and the low anharmonicity within the lattice. Recently a number of polycrystalline aluminum nitride substrates have become com-

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mercially available with quoted thermal conductivities in the range 100 to 250  $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ . The fact that the commercial materials do not achieve the predicted theoretical limit, despite being sintered to full density, is generally attributed to the presence of substitutional oxygen impurities in the nitrogen sublattice which generate aluminum vacancies [2, 3]. The large localized mass differences introduced by the aluminum deficiency lead to considerable anharmonicity within the lattice which impedes phonon transport and causes a dramatic reduction in the thermal conductivity. The objectives of the present study are to address several key issues: (a) What is the most reliable method to determine the thermal conductivity of an aluminum nitride substrate? (b) Are the manufacturers' claims for thermal conductivity realistic based on the available measurement techniques? and (c) What can be learned about the mechanisms governing thermal conductivity in AlN from measurements made at cryogenic temperatures? The thermophysical properties of four different commercial substrates have been investigated using a combination of steady-state and transient heat flow methods, and the results compared and contrasted. Since it is not our intention to compare individual manufacturers, the four materials studied are simply denoted by the letters A through D.

## 2. SAMPLES

The oxygen contents and thermal conductivities quoted by the manufacturers of the four samples investigated are listed in Table I.

## 3. EXPERIMENTAL DETAILS

### 3.1. Thermal Conductivity

The thermal conductivity of the four samples was measured, directly, in the substrate plane, using a microcomputer-controlled guarded

Table I. Sample Description

Sample	Dopant	Oxygen content (mol %)	Manufacturer's <sup>a</sup> thermal conductivity ( $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ )
A	$\text{Y}_2\text{O}_3$		170 (1)
B	$\text{Ca}(\text{NO}_3)_2$	0.441	160 (2)
C	$\text{CaC}_2$	1.650	140 (3)
D	$\text{Y}_2\text{O}_3$		130 (4)

<sup>a</sup> (1) Norton-Toshiba N-170; (2) Shapal; (3) NEC; (4) Norton-Toshiba N-130.

longitudinal heat flow system [4]. Measurements were made in the temperature range 80 to 400 K in a liquid nitrogen cryostat. The samples were cut to 40 mm in length and 6 mm in width. The substrate thicknesses were of the order of 0.5 to 1 mm. Sample heaters were wound directly onto the bar samples to reduce radiative heat losses and the samples were mounted in the cryostat by clamping to the cold sink. Thermocouples were mechanically attached by binding the bead to the samples with a thin copper wire and then coating with a thin layer of Saureisen cement. The thermal conductivity results are presented in Fig. 1.

### 3.2. Heat Capacity

The heat capacity of one sample was determined using the electronic pulse technique [5], in the temperature range 80 to 400 K. This technique was developed in this laboratory to provide a simultaneous determination of thermal diffusivity and heat capacity on insulating materials of low to intermediate thermal conductivity. In principle it is similar to the laser flash technique except that the heat pulse is supplied by depositing a square pulse of electrical energy in a thin graphite resistive heater sprayed onto one side of the sample. Careful impedance matching of the heater to the power supply minimizes reflections and allows integration of the pulse to determine the energy deposited in the sample. By measuring the temperature rise and the mass of the sample, the heat capacity may be calculated. The very high thermal conductivity of the samples in this study precludes a measurement of the thermal diffusivity, as the rise time of the transient is beyond the resolution of the system. The heat capacity data are presented in Fig. 2.

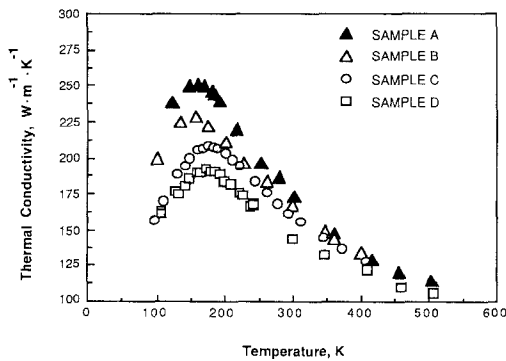


Fig. 1. Temperature dependence of thermal conductivity of AlN substrates.

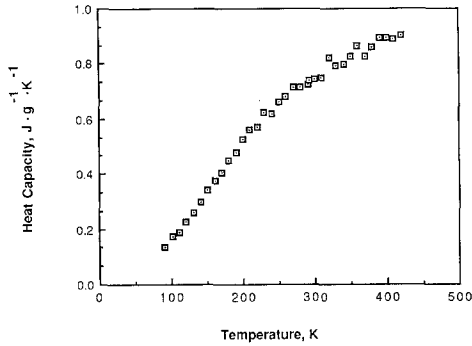


Fig. 2. Temperature dependence of heat capacity of sample A.

### 3.3. Thermal Diffusivity

Measurements of thermal diffusivity normal to the substrate plane were made at room temperature on samples B and C using the laser flash method, first described by Parker et al. [6]. The samples were in the form of 10-mm square slabs, the front faces of which were coated with a thin layer of gold followed by a thin layer of graphite to ensure uniform absorption of the laser beam at the sample surface. The temperature history was measured using an intrinsic thermocouple formed on the sample rear face by pressing the wires into contact with a thin copper strip evaporated onto the sample. Although the finite pulse correction of Taylor and Cape [7] was applied to the measurements, the data appeared to be anomalously low. This effect is believed to be instrumental in origin and a correction for it was developed. This correction is discussed in the next section. The heat capacity and diffusivity data obtained using the transient techniques are compared with the thermal conductivity data obtained using the steady-state longitudinal heat flow method.

### 3.4. Microscopy

Fracture surfaces of each of the samples were photographed in an Amray 1200 C scanning electron microscope. The average grain size of each of the materials was estimated from several micrographs of each sample using the mean linear intercept method with  $G = 1.5L$ .

## 4. RESULTS AND DISCUSSION

### 4.1. Thermal Conductivity

In an electrically insulating material such as AlN, heat conduction is dominated by phonon transport. Thermal resistance may arise from phonon scattering from point defects, grain boundaries, and direct phonon-phonon interactions (Umklapp scattering). Each of these processes limits the phonon mean free path more or less severely. The most severe limitation at room temperature is imposed by Umklapp scattering, and the least by boundary scattering. However, at cryogenic temperatures the effect of lattice defects and grain boundaries becomes more significant. The temperature dependence of thermal conductivity of the four samples measured is shown in Fig. 1. Peaks in thermal conductivity are observed for all four samples at temperatures ranging from 160 to 175 K. The occurrence of the peaks at these relatively high temperatures is characteristic of fine-grained polycrystalline materials [8]. In general the scattering rates of the individual mechanisms depend differently on frequency and temperature. This requires an independent relaxation time for each scattering process. An effective relaxation time  $\tau_e$  can be determined from Eq. (1) [9].

$$\tau_e^{-1} = \tau_b^{-1} + \tau_i^{-1} + \tau_u^{-1} \quad (1)$$

The quantities  $\tau_b$ ,  $\tau_i$ , and  $\tau_u$  are the relaxation times for boundary, impurity, and Umklapp scattering processes, respectively. At temperatures sufficiently above the peak conductivity temperature, the boundary scattering term may be considered a small correction. In the temperature region above 225 K, boundary scattering may be neglected, and the thermal analogue to Matthiessen's rule employed [10]. This was carried out for all four samples by fitting Eq. (2) to the thermal resistance versus temperature data plotted in Fig. 3.

$$k^{-1} = a + bT \quad (2)$$

Information on the amount of static defects in the sample is contained in  $a$ , and  $b$  depends on the details of the crystal structure [11]. The least-squares coefficients  $a$  and  $b$  are listed in Table II. The slopes ( $b$ ) of all four lines are observed to be virtually identical, as expected. However, there is a substantial difference in the defects ( $a$ ) term. This is believed to be related to the different oxygen content of the samples. At present oxygen content data are available only for samples B and C, however, the indication is that the lower the oxygen content, the higher the thermal conductivity.

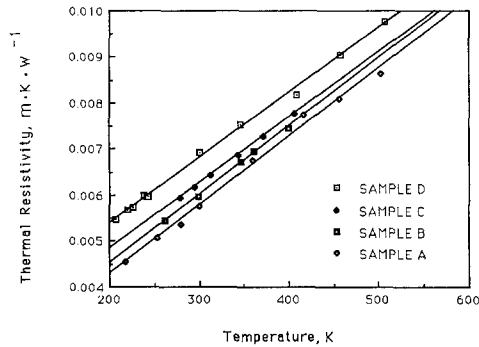


Fig. 3. Linear temperature dependence of the high-temperature thermal resistivity data.

## 4.2. Heat Capacity

The temperature dependence of the heat capacity of AlN is shown in Fig. 2. These data were employed in order to calculate the phonon mean free path (MFP) at the peak conductivity temperature using Eq. (3).

$$k = (CV_s\lambda)/3 \quad (3)$$

In this equation,  $k$  is the thermal conductivity,  $\lambda$  is the mean free path,  $C$  is the heat capacity at constant volume, and  $V_s$  is the speed of sound in the material. A value of  $6.98 \times 10^3 \text{ m} \cdot \text{s}^{-1}$  was used for  $V_s$  [2]. The calculated values of MFP are tabulated in Table II. The mean free path at the peak thermal conductivity temperature is observed to be several orders of magnitude lower than the measured grain size. This is consistent with the notion that lattice defects caused by the oxygen impurities dominate the phonon scattering at this temperature.

Table II. Sample Parameters

Sample	Grain size ( $\mu\text{m}$ )	( $\lambda$ ) MFP (nm)	(a) y intercept ( $\text{m} \cdot \text{K} \cdot \text{W}^{-1}$ )	(b) Slope ( $\text{m} \cdot \text{W}^{-1}$ )
A	5.0	0.85	1.33	1.48
B	4.4	0.83	1.55	1.49
C	5.0	0.63	2.00	1.42
D	7.0	0.60	2.58	1.41

### 4.3. Thermal Diffusivity

The thermal diffusivity data and the heat capacity data were combined to give thermal conductivity values for samples B and C. Thermal conductivity results derived from the raw thermal diffusivity data obtained using the laser flash technique are substantially lower than the manufacturer's quoted values listed in Table II. Both B and C have apparent values of approximately  $38 \text{ W} \cdot \text{m}^{-1} \text{K}^{-1}$ . This is a surprising result. However, Kuramoto and Taniguchi [12] report that for a sample thickness less than 3 mm, measurements of thermal diffusivity on aluminum nitride using the laser flash technique led to the derivation of anomalously low thermal conductivity data. Unfortunately no explanation of this effect is advanced by the authors. Their resolution of the problem was to make thicker samples. However, we have already shown that the maximum MFP, of the AlN samples under investigation, at the peak temperature is considerably less than  $1 \mu\text{m}$ . At room temperature the MFP will be shorter due to the influence of Umklapp scattering. In view of the fact that the grain sizes of the materials range from 4 to  $8 \mu\text{m}$  (which is not untypical for most polycrystalline AlN), it is inconceivable that a sample dimension of 0.5 to 1 mm would in any way influence the intrinsic thermal conductivity of the sample at room temperature. From this standpoint there is no physical justification of a thickness dependent thermal conductivity. This leads us to conclude that the effect is introduced by the technique itself.

The same conclusion was drawn from results obtained on a number of experimental AlN samples measured in this laboratory [13]. To confirm this a series of copper samples was prepared with thicknesses in the range 1 to 8 mm and a definite thickness dependence was established. The results are shown in Fig. 4. The laser flash technique is based on the measurement

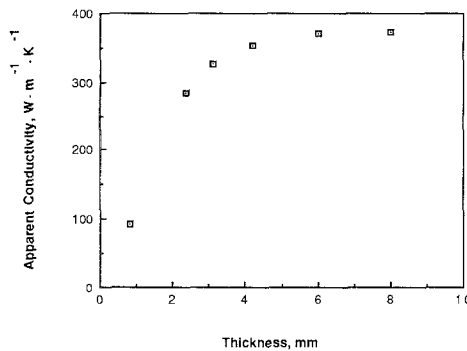


Fig. 4. Effect of sample thickness on thermal conductivity of copper derived from laser flash experiment.

of temperature as a function of time. An AlN sample with a thermal conductivity of  $150 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$  and a thickness of 1 mm would theoretically have a half-rise time of 0.9 ms. This is of the order of the duration of the laser pulse, which in theory is assumed to be instantaneous. However, an adequate theoretical correction for the finite-pulse time effect exists [7] and has been applied to all measurements made on thin samples. The results on the copper samples indicate that this cannot account for the thickness effect that we observe. It is more likely that signal processing by external hardware is responsible. In fact many of the electronic components presently used in the signal amplification and filtering circuitry are subject to finite rise times that are likely to have the effect of retarding the recorded transient response. Plotting the apparent thermal conductivity of the copper samples as a function of the recorded half-rise time (Fig. 5), we observe that the apparent thermal conductivity,  $k$ , appears to be related to an exponential function of time,  $t$ . The following relation was fitted to the data:

$$k = k_0 \{1 - \exp(-t/x)\} \quad (4)$$

where  $k_0$  represents the true conductivity at large  $t$ , and  $x$  is a time constant introduced by the system. A preliminary value of 10 ms has been estimated for the system based on the data shown in Fig. 5. A knowledge of  $x$  permits the calculation of a minimum accurately measurable half-rise time of the order of 30 to 40 ms, below which the system will yield anomalously low thermal property data. This corresponds to the point at which  $k$  saturates in Fig. 5. For samples whose measured half-rise time is less than 400 ms, a correction may be applied to calculate the true thermal conductivity. The correction, Eq. (5), is based on the measured apparent

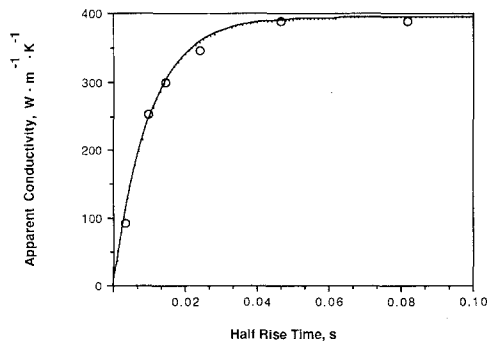


Fig. 5. Apparent thermal conductivity of copper derived from laser flash experiment versus measured half-rise time.



**Table III.** Comparison of Thermal Conductivity ( $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ )  
Results Obtained by Different Techniques

Sample	Manufacturer's data	Longitudinal bar	Laser flash
A	170	173	NA
B	160	166	159
C	140	159	150
D	130	146	NA

thermal conductivity and half-rise time and the time constant of the apparatus.

$$k_0 = k / \{1 - \exp(-t/x)\} \quad (5)$$

Applying Eq. (5), the true thermal conductivities of samples B and C attain values of 159 and 150  $\text{W} \cdot \text{m}^{-1} \cdot \text{K}^{-1}$ , respectively. Both of these results are in good agreement with the data obtained using the longitudinal bar technique and also with the manufacturers' recommended values, presented in Table III for comparison.

## 5. CONCLUSIONS

Comparison of thermal property measurements on four commercially available AlN substrates using steady-state and transient temperature methods permits the following conclusions to be drawn.

(1) The guarded longitudinal heat flow technique is preferable to the laser flash technique for determining the thermal conductivity of thin, high-conductivity ceramic substrates. However, if a system calibration is carried out it appears possible to apply the laser flash technique to such measurements, which would be important if only small amounts of material were available.

(2) Measurements of thermal conductivity at low temperatures can provide information about the mechanisms that limit the thermal conductivity of AlN. Our observations are consistent with the notion that defects occurring due to the presence of oxygen impurities limit thermal conductivity at low temperatures.

(3) The manufacturers' claims for the thermal conductivity of their substrates are in good agreement with the results of our independent measurements.

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