

Measurements carried out on bulk samples of apiezon N grease in nylon tubes indicate that the grease behaves as a thermal insulator between 4.2 and 1.4 K, and therefore applications utilizing thick grease coatings to provide thermal contact should be avoided. In addition it was observed that the thermal cycling of glass tubes packed with the grease between 300 and 77 K, under

varying conditions of vacuum, can lead to grease column separations which reduce the conductivity further. It is noted that these results may have a bearing on the thermal contact between a heat sink and a small thermometer which has been positioned in a grease hole within the heat sink.

## Thermal conductivity of apiezon N grease at liquid helium temperatures

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Apiezon N grease is a thermal bonding agent which is widely used to provide thermal contact between thermometers, specimens, and different parts of experimental apparatus at cryogenic temperatures.<sup>1-6</sup> There is, however, a lack of low temperature thermal conductivity information on this grease in the literature. The thermal conductivity of several greases has been reported recently<sup>7</sup> and it was decided to extend this study in order to obtain results on apiezon N grease.<sup>8</sup>

### Experimental procedure

The grease was placed in thin-walled nylon tubes with a syringe; care being taken to avoid air bubbles. Gentle warming from a soldering pencil in close proximity was sufficient to reduce the viscosity of the grease enough to rid the grease column of visible air bubbles, unavoidably trapped in the packing process. Further treatment to remove trapped gas, for example, by heating in a vacuum, was not considered advisable since we wished to measure the grease as it is ordinarily supplied and used. The thin nylon walls permitted the packed tube to be inspected for air bubbles with a light source.

Three  $\frac{1}{2}$  W, 10  $\Omega$  Allen Bradley carbon radio resistors were used for thermometers and these were previously cemented into the nylon tube with GE-7031 adhesive. The thermometers divided the grease column into two sections with effective lengths of approximately 2.5 and 5.0 cm, see Figure 1. The use of three thermometers allowed two independent observations of the thermal conductivity to be made and thus presented a check on any heat lost in traversing the length of the grease sample. In addition, discrepancies resulting from incorrect area to length ( $A/L$ ) values, and/or separations or air bubbles in the grease can be more easily noted with

this arrangement, since if one section of the grease column has an  $A/L$  value equal to twice that of the other, for the same heater power and for approximately constant thermal conductivity (that is, for small tempera-

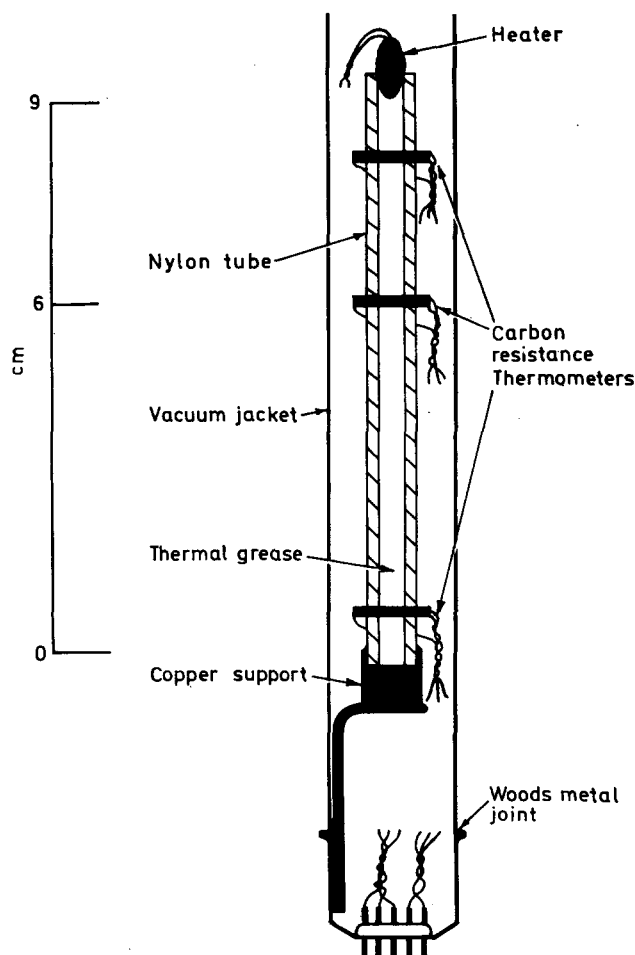


Figure 1. Nylon sample holder for apiezon N grease samples

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ture gradients), the temperature differences between the shorter and longer sections of the grease column should differ by a factor of 2.<sup>9</sup>

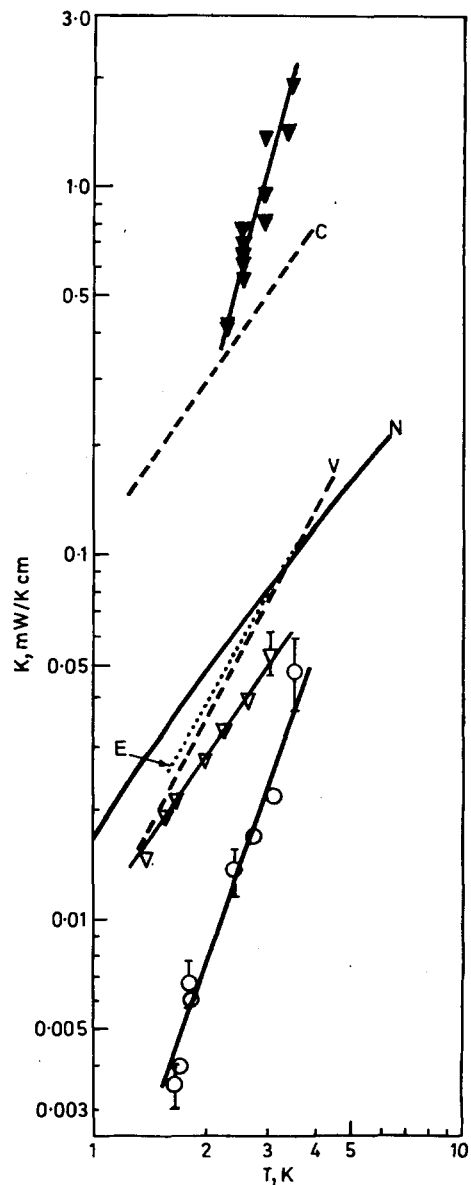
A 2 000  $\Omega$  No. 44 manganin wire heater was non-inductively wound and placed directly into the grease where it was positioned about 1 cm above the top thermometer. To shorten the equilibrium time, a copper form, previously used for the heater winding, was eliminated. The resistances of the heater and the carbon thermometers were measured by a low frequency a.c. resistance bridge. To produce a temperature gradient, a constant current was sent through the heater and a digital voltmeter was used to determine the current by measuring the voltage drop across a standard resistor in series with the heater. Powers of the order of 1  $\mu$ W were used to produce temperature gradients of about 0.01 K/cm. Since the conductivity of apiezon N grease is small, a correction was necessary for the heat conducted by the thin walls of the nylon tube. Additional experimental details have been given elsewhere.<sup>7</sup>

### Apiezon N grease results

The results of experiments on three different apiezon N grease samples are shown as a function of temperature in Figure 2, where they may be compared with conductivity measurements on nylon and other greases. All data shown in the graph has, of course, been corrected for the thermal shorting effect of the nylon tube.<sup>7</sup> The first apiezon N experiment (indicated by open triangles) was carried out with a grease sample having a cross-sectional area of 17.8 mm<sup>2</sup>. In the second experiment (and others which followed) the grease sample was placed within a larger tube with thinner walls. The results of the second experiment, which are indicated by circles in Figure 2, were taken from a sample with a cross-sectional area of 49.5 mm<sup>2</sup>. Typical experimental uncertainties are indicated by the error bars.

For the first two experiments, the method of cooling the grease was similar. The experimental chamber was first evacuated and about 2 000  $\mu$  (266 N/m<sup>2</sup>) of helium gas was introduced at room temperature. The temperature of the chamber was then lowered to 77 K by cooling with helium exchange gas. Next liquid helium was placed around the vacuum jacket as the helium exchange gas pressure within approached about 50  $\mu$  (7 N/m<sup>2</sup>). Calibrations of the carbon resistor thermometers were then carried out against the helium vapour pressure, after which thermal isolation was provided by evacuation.

This procedure was altered for the third experiment, the results of which are shown in Figure 2 by darkened triangles lying about an order of magnitude higher than the thermal conductivity data of the other greases. No pumping was performed at room temperatures. An atmosphere of helium gas was maintained within the vacuum jacket as the grease was cooled to 77 K, after which all but 200  $\mu$  (27 N/m<sup>2</sup>) was pumped out. The vacuum jacket was then cooled to 4.2 K with liquid helium, leaving an exchange gas pressure inside of about 50  $\mu$  (7 N/m<sup>2</sup>) for calibration, and the remaining procedure was identical to experiments 1 and 2. The conductivity of the grease appears to be enhanced if the grease is cooled before it is pumped. We shall comment further on this observation below.



- ▽ 1st apiezon N grease experiment
- 2nd apiezon N grease experiment
- ▼ 3rd apiezon N grease experiment
- N Results for nylon    --- C Results for cry-con
- .....E Results for eccotherm TC-4
- V Results for vacuum grease

Figure 2. A comparison of bulk thermal conductivity measurements on apiezon N grease with those obtained on other greases at liquid helium temperatures

In order to study the effect of cooling under different pressures, glass tubes were packed with the grease in a manner described above. The grease columns had a diameter of 6 mm and a length of 12 cm. Typical results obtained from the first test are shown in Figure 3, which is made up of six separate negatives cut and pieced together. Each picture was taken with the aid of a strong diffused light positioned to pass through the glass tube and grease sample. From Figure 3 the effect of cycling the temperature between 300 and 77 K, as an atmosphere of helium gas is maintained, can be examined. The glass tube containing the grease sample was lowered into a glass jacket into which a gentle flow of helium gas was directed. The entire arrangement was then lowered into a dewar of liquid nitrogen where it was kept for 7–10 min. Although the grease cooled rather quickly in this test, it

must be noted that with the experimental procedure used in the thermal conductivity measurements much more cooling time was allowed.

The tube containing the grease at nitrogen temperatures was extracted from its low temperature environment and immediately photographed. The time required was no longer than 3 s and the result of the first cooling is shown in the first picture on the left in Figure 3. In the second picture, the glass tube and grease sample have returned to ambient temperatures (300 K). The process was repeated twice to produce the next four pictures. The entire test was duplicated and the results were observed to be reproducible. It appears that the effect of thermal cycling is significant in causing a growth of breaks or separations in the grease column. On close inspection, the cracks in the grease appear to be isotropic and numerous. In three instances (out of about six) we observed a complete break or separation in the grease column the first time it was cooled to 77 K. The influence of the walls of the glass tube on grease column separations is believed to be minimal because the grease does not stick to the walls as it cools. One can easily observe this by placing a slug of grease (~1 cm long) in a glass tube (~3 mm i d) and cooling the tube to 77 K. When the grease freezes the entire slug may easily be moved about within the tube by poking it with a stiff rod. It should be noted that the thickness of the grease, shown in the second, fourth, and sixth photographs in Figure 3, is misrepresented owing to an optical reflection effect which makes the thickness of the glass tube walls appear to be part of the thickness of the grease column at room temperature. Therefore in the first, third, and fifth photographs, the contraction of the grease appears exaggerated, because the grease has pulled away from the tube wall and the reflection effect is absent.

It must be emphasized that only the first picture (representing the initial cooling) on the left in Figure 3 corresponds somewhat to the experimental situation in the third apiezon N thermal conductivity experiment, and in addition, the cooling time in the latter is several hours longer. It would appear that the effect of a longer cooling time is to produce fewer cracks in the grease, since fewer strains are present when the entire mass of the grease passes slowly through the glass temperature and experiences a characteristically abrupt change of volume expansion.<sup>10</sup> (We obtained an experimental value of 237 K for the glass temperature of apiezon N grease.) Some experimental evidence for the effects produced by slow and uniform cooling has been obtained from grease samples in glass tubes surrounded by a copper shield, so that the entire mass of the grease might cool uniformly and at a much slower rate than the grease shown in Figure 3. The results for an initial cooling show a smaller number of cracks and a higher degree of localization. For example, whereas in Figure 3 the cracks were large, sometimes producing clear areas extending across the tube, those cracks or pockets in a grease tube, cooled more slowly, are fine and short, seldom having a diameter or length greater than  $\frac{1}{4}$  of the diameter of the grease column. However, complete separations of the grease column still occur with temperature cycling.

In order to examine the effect of pumping on apiezon N grease as it is cooled to 77 K, a glass tube was filled with the grease and placed inside a glass jacket, according to the procedure described above, except that instead of



Figure 3. Effect of cycling the temperature between 300 and 77 K on a glass tube containing apiezon N grease under an atmosphere of helium gas. Beginning on the left, the first, third, and fifth photographs were taken with the tube at 77 K. The second, fourth, and sixth pictures were obtained after the grease temperature returned to 300 K

sending a flow of helium gas into the jacket, it was evacuated and then cooled; the remaining procedure staying the same. With the grease tube at 77 K, pumping was stopped and an atmosphere of helium was placed around the grease sample. The first photograph on the left of Figure 4 was taken immediately after the sample was removed from its nitrogen environment. Many cracks are visible in the grease column. The second photograph reveals a separation of the grease column after the temperature of the grease tube was allowed to return to 300 K. When the procedure was repeated, a huge separation of almost 3 cm was found in the grease column. Photographs 3 and 4 of Figure 4 show this separation with the temperature of the tube, first at 77 K and then at 300 K, respectively. The reproducibility of the qualitative results of Figures 3 and 4 has been observed.

## Discussion

Some of the variables introduced by these results are—the effect of cooling under conditions of different pressures, the contraction of the grease and its effect on the results, the speed of cooling and its effect in the production of cracks, the glass temperature, gas content, the effect of varying the cross-sectional area of the grease column, and possible composition differences in the supply.

For the third apiezon N experiment, the experimental results, which may be roughly approximated by  $K = 0.0135 T^{4.1}$  mW/cm K, show a temperature dependence which is greater than that expected from behaviour characteristic of phonon flow ( $T^3$  temperature dependence).<sup>11</sup> The measured value of the bulk thermal conductivity at 2 K is about  $3 \times 10^{-4}$  W/cm K and it lies within the region of values obtained by Chang and Jones<sup>12</sup> from measurements on various glasses. The value is also approximately three times larger than that

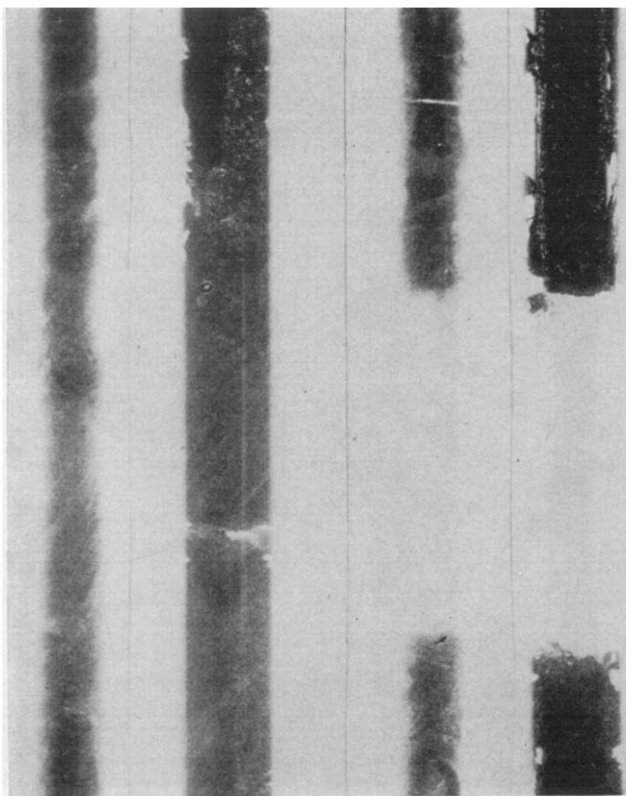


Figure 4. Effect of temperature cycling between 300 and 77 K on a glass tube containing apiezon N grease which is cooled under vacuum. The first and third pictures were taken after the tube had cooled to 77 K under a vacuum. The second and fourth pictures were obtained after the tube was allowed to warm to room temperature

calculated by Challis and Cheeke<sup>13</sup> for the bulk thermal conductivity of epoxy resin (Araldite AY 103). It is interesting to relate the results of the glass grease tube study recorded in Figure 3 to the experimental conductivity shown in Figure 2. As an explanation of why the measured conductivity in experiment 3 (where the sample is kept at 1 atm ( $101 \times 10^3$  N/m<sup>2</sup>) pressure) is larger than that observed in the case of experiments 1 and 2, the following possibility may be offered. When the grease is placed in a vacuum and cooled (as in experiments 1 and 2) the gas bubbles tunnel their way out of the grease, leaving holes or channels behind. Therefore grease which is placed in a vacuum originally, at a temperature above its glass temperature, might be expected to yield a larger thermal resistance because the effective cross-sectional area of the grease column is diminished after cooling.

This explanation may also be used in considering the results of the glass study. Consider the results of Figure 3 where the grease is taken from a 77 K temperature environment and photographed. The cracks which have been produced in the grease in Figure 3 contain helium gas at 77 K and atmospheric pressure. In warming to room temperature this gas will expand and enlarge the cracks. When the sample is cooled again in an atmosphere of helium, the enlarged cracks will accommodate still more gas which contributes to an additional increase in their size on warming. The effect is even more damaging when the grease is cooled in a vacuum. During the first

cooling, holes are produced by the escaping gas which has been trapped in the grease. As soon as the sample is taken out of its environment and photographed, gas enters the cracks and holes. As this comparatively larger volume of gas warms to room temperature, the large separations shown in Figure 4 are produced in the grease column.

It is expected that the area to length ratio of the grease column will be in error due to the cracks as well as the grease contractions. Because of the difficulties involved, no correction to the thermal conductivity was attempted for this source of error. A simple calculation will show that such a correction would have the effect of raising the measured conductivities in the three apiezon N experiments.

In summary, the observations indicate that the experimental procedure is important. In order to achieve a minimum of cracks and/or separations, the grease should be slowly cooled to liquid nitrogen temperature before it is pumped. The cycling of the temperature of a grease column between 300 and 77 K, in the manner performed above, causes a growth of separations or breaks in the grease column. Furthermore, when the pressure is varied, as described above, the separations are much larger. It is interesting to consider the implication of these results for the important application of the thermal contact between a small thermometer and a heat sink. Consider the popular usage in which the thermometer is placed in a hole in the heat sink with grease and left there for the course of many experiments, during which the arrangement is exposed to both temperature cycling and varying conditions of pressure. If the grease is apiezon N, the results reported here indicate that within the hole the grease could be so perforated by cracks, holes, and/or separations that very little would actually be serving to produce a thermal bond with the heat sink. In addition, the poor bulk thermal conductivity of the grease adds suspicion to the quality of such thermal bonds at liquid helium temperatures.

The low temperature thermal conductivity data<sup>7</sup> of several other electrically insulating greases has been included for comparison in Figure 2. They are cry-con<sup>14</sup> (a copper loaded electrically insulating grease), eccotherm TC-4<sup>15</sup> (a specially formulated paste based on silicones and fillers), and silicone high vacuum grease.<sup>16</sup> Cry-con appears to have the most to offer for ordinary bulk applications at liquid helium temperatures, where it has been observed to be highly resistant to cracking. However because of its lower effective viscosity, it has been found to be inferior to apiezon N grease when used in pressed contacts at temperatures below 1 K.<sup>17,18</sup> While the glass tube results in Figure 3, and especially Figure 4, indicate that the measured conductivity of apiezon N grease would decrease under such thermal cycling, the thermal conductivity results of cry-con have been observed to be reproducible after warming to room temperature and cooling again, when using the procedure outlined above for the first and second apiezon N experiment.

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