Heat capacity of Apiezon N grease from 1 to 50 K

A. J. Bevolo

Thermal contact between various components of a low temperature calorimeter can be improved by the application of a thin layer of vacuum grease. For specific heat measurements the need for a precise value of the specific heat C of the grease is usually avoided by including the grease in the addenda. Recently we measured the specific heat of 2H $NbSe_2$ from 1 to 55 K.[1] Because of its layered structure 2H $NbSe_2$ can only be grown in thin plates of the order of 0.1 g. To obtain a sufficiently large sample many such plates had to be placed in the calorimeter at once. Thermal contact between the individual pieces of the sample was accomplished by using a small amount of Apiezon N grease. To correct for the effect of the grease required accurate values of its specific heat. To our knowledge the only data available was that of Bunting et al [2] which covered the range from 80 to 324 K. Although the heat capacity of a closely related material, Apiezon T grease, is known [3] from 1 to 350 K, we could not be certain that its specific heat was identical to that of Apiezon N. This note reports the specific heat of Apiezon N from 1 to 50 K.

The specific heat of a 1.026 g sample of Apiezon N grease was measured by the heat pulse method using a calorimeter with an isothermal shield.[4] Because of the low thermal diffusivity of the sample equilibrium times ranged from a fraction of a second near 1 K to 20 minutes at the highest temperatures. The precision of the measurements ranged from 0.5% at low temperatures to about 1.5% at the highest temperatures. Because of the long equilibrium times at high temperatures it is difficult to estimate the accuracy of our data. We minimized the extrapolation errors by taking large ΔT pulses (from 5 to 10%) at temperatures above 20 K. As the heat capacity is nearly linear with T in this temperature range the curvature correction is very small. We estimate that the accuracy is of the order of the precision.

Fig.1 shows the data for Apiezon N plotted as C/T versus T^2 for $1 < T^2 < 5$ K. In addition several points are shown for Apiezon T obtained from equation 1 of reference 3 using 1 cal = 4.184 J. Although neither set of data lie along a straight line, it is clear from the graph that when the data is extrapolated to 0 K Apiezon T has a linear term in its heat capacity while Apiezon N does not. Within a few percent the difference between the heat capacity for these two greases below 2 K can be accounted for by this linear term, so that Apiezon T has a specific heat that is 20% higher than Apiezon N at 1 K. Even taking into account this linear contribution there are subtle differences in the heat capacity of these two materials. For instance, the Apiezon N data for $1 < T < 4$ K can be fitted within the experimental precision by

$$C = 21.6\,T^3 + 2.55\,T^4\ \mu\mathrm{J g^{-1}\,K^{-1}} \tag{1a}$$

or

$$C = 5.16\,T^3 + 0.612\,T^4\ \mu\mathrm{cal\,g^{-1}\,K^{-1}} \tag{1b}$$

The author is with the Ames Laboratory – USAEC and Department of Physics, Iowa State University, Ames, Iowa 50010, USA.
Received 24 June 1974.

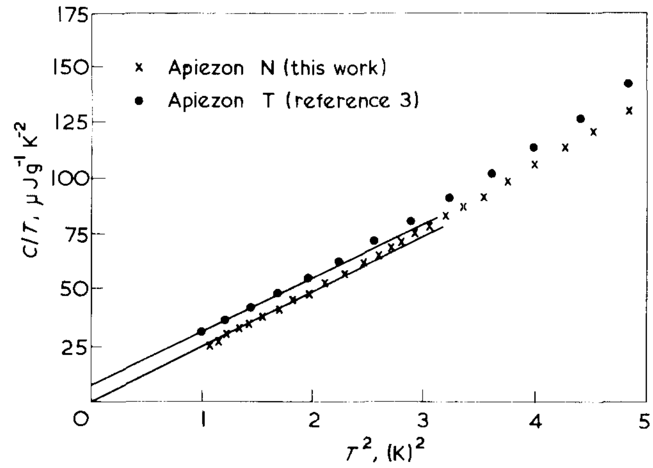

Fig.1 Specific heat of Apiezon N and Apiezon T plotted as C/T versus T^2 showing the difference between the two greases. The straight line extrapolations to 0 K show the presence and absence of an intercept (and therefore of a linear term in T) for Apiezon T and N respectively

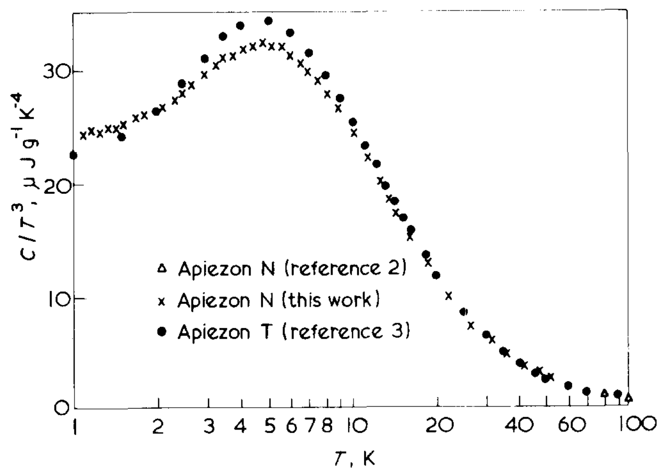

Fig.2 Specific heat of Apiezon N and Apiezon T plotted as C/T^3 versus T on a semilogarithmic plot from 1 to 50 K. For Apiezon T the linear term (8.5 T mJ g^{-1} K^{-1}) has been subtracted before plotting the data from reference 3

despite attempts to use a four-term series in odd powers of T of the type found adequate for Apiezon T by Bunting et al.[3] It is of interest to note that the coefficient of the T^3 term for both greases differs by only 2% (5.16 for Apiezon N and 5.065 for Apiezon T). A convenient representation of the C of these two substances is shown in Fig.2, where C/T^3 is plotted versus T in a semi-logarithmic plot. The Apiezon T grease as shown has had the linear term subtracted before being plotted. Both show the same general behaviour exhibited by most materials when plotted this way. The broad peak is due to the deviation of the heat capacity predicted by the Debye model at low temperatures, that is $C_D = \beta T^3$. When plotted as C/T^3 versus T, C_D gives a straight line with zero slope. According to Katz[5] the temperature at which the peak occurs T_m is

Table 1. Specific heat of Apiezon N(C_N) and Apiezon T(C_T)

T, K	C_N, mJ g^{-1} K^{-1}	C_T^*, mJ g^{-1} K^{-1}	T, K	C_N, mJ g^{-1} K^{-1}	C_T^*, mJ g^{-1} K^{-1}
1	0.024	0.0311	13	42.4	44.1
2	0.212	0.228	14	49.0	50.8
3	0.793	0.866	15	56.0	57.7
4	2.025	2.218	16	63.4	64.9
5	4.05	4.33	18	78.1	79.6
6	6.70	7.24	20	93.0	94.8
7	10.2	10.88	25	133	133.9
8	14.4	15.23	30	175	173.4
9	19.1	20.12	35	215	212.7
10	24.4	25.56	40	253	250.9
11	30.2	31.4	45	290	288.0
12	36.2	37.6	50	326	324.0

* Data taken from Table 1, reference 3 using 1 cal = 4.184 J

related to the frequency ν_{max} at which the phonon density of states reaches its first maximum, while the height of the heat capacity peak is correlated with the number of phonon modes in the density of states peak. If the phonon density of states can be represented by a Debye term proportional to ν^2 and an Einstein peak at ν_{max}, then $T_m = h\nu_{max}/5\,k$. For both of the greases T_m = 4.8 K which gives ν_{max} of approximately 0.5 THz. The somewhat higher peak for the Apiezon T grease indicates a larger number of these low frequency phonons occur for this grease when compared to Apiezon N. For comparison we have listed the specific heat of both greases from 1 to 50 K in Table 1. For $80 \leq T \leq 300$ K the heat capacity of the two greases are compared in Fig.2 of reference 6, which shows good agreement except between 200 and 300 K where both materials exhibit anomalous behaviour.

The presence of a linear term in the heat capacity of the Apiezon T and its absence in Apiezon N are not understood. Being good insulators [7] there cannot be any electronic contribution to their heat capacity. Both greases are presumably amorphous solids at these temperatures, which are well below their respective glass transition temperatures.[6] It is known [8] that amorphous solids have a linear term in their heat capacity at low temperatures. Anderson et al [9] has proposed that there exist in all amorphous solids atomic configurations that can exist in two discrete energy states which, over the whole sample, have a distribution of energy gaps. The exact nature of these entities is not yet clear. According to previous discussion [2,3] the only difference in the composition of the two greases is that Apiezon T contains a hydrated aluminium soap whereas the Apiezon N does not. It is possible that the soap so modifies the short-range order of the grease so that it introduces states of the type postulated by Anderson. In any case the fact remains that because of the linear term in its heat capacity Apiezon T has a higher heat capacity at low temperatures than Apiezon N. It is therefore recommended that Apiezon N be used in specific heat cryostats operating below 4 K.

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