Specialised Hydrocarbon-based Grease for Cryogenic Applications

Discussing the demands of cryogenic applications and research, looking at the behaviour of hydrocarbon-based grease for thermal contact and sealing.

White Paper

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Introduction

Cryogenics typically refers to operation at temperatures below 123K, or -150°C and in many branches of science and medicine it is necessary to perform experiments and run equipment at cryogenic temperatures, down to as low as a few degrees Kelvin. At these temperatures specialist materials are needed in order to provide sealing and thermal conductance, since standard heat sink compounds and sealing formulations may crack or craze, which in turn reduces sealing efficiency and reduces thermal conductivity.

There are many types of grease currently available, such as PFPE based products that quote operating temperatures well below 0°C. However, nearly all these products have a quoted operating temperature range limit of -80°C. Typical heatsink compounds are only quoted as working down to -50°C. The one exception is hydrocarbon based grease (with proprietary additives) which can operate all the way down to -269°C, the temperature at which liquid helium boils.[i]

This paper will discuss the demands of cryogenic applications and research, looking at the behaviour of hydrocarbon based grease for thermal contact and sealing.

Examples of cryogenic applications

There are a number of industries and scientific fields where cryogenic temperatures are utilised. Two examples explored in this paper are research and superconductors.

In university research fields there are many examples of where samples need to be cooled to extremely low temperatures. In these experiments it is necessary to mount a sample onto a cryostat cold finger, in order to achieve very low temperatures. Once the experiment is complete it is necessary to remove the sample again. In this case the compound used for mounting needs to be pliable at room temperature, but harden at lower temperatures. It is also very useful if the compound can be easily removed and cleaned from the sample after the experiment is complete.

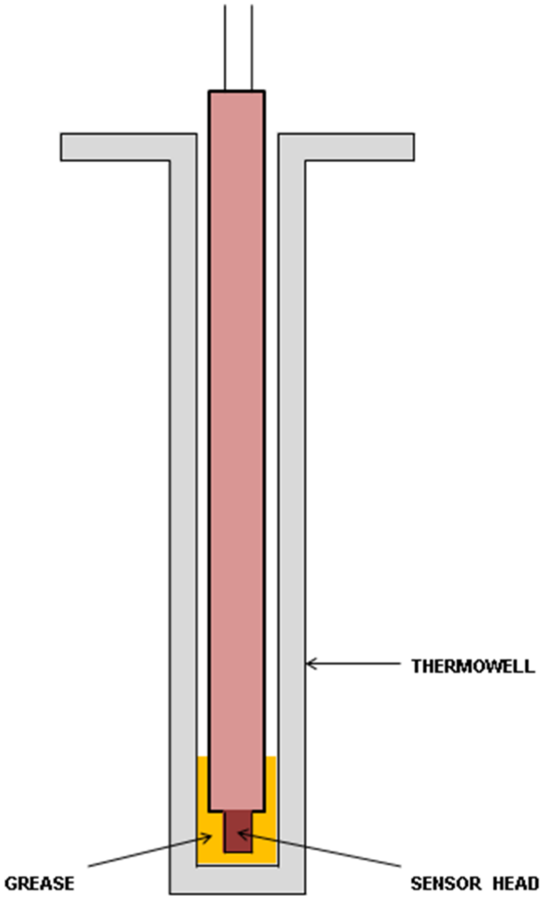
Another key application for cryogenics is within superconductors, where very low temperatures are required to produce the phenomenon of very low electrical resistance. The typical temperature for low temperature superconductors is <30K, while so-called high temperature superconductors operate at temperatures up to 138K. Although considered high temperature this is still well into the cryogenic range and requires the use of materials which will withstand very low temperatures without losing any of their properties.

Superconductors are key to the production of very high strength magnets, used in devices such as Magnetic Resonance Imaging (MRI) scanners and maglev transport systems. More niche applications include particle accelerators and fusion reactors where extremely high power magnet fields are required to provide the necessary containment and acceleration forces. Within these systems there is a need for thermal contact between cooling equipment and the superconducting materials and also for accurate temperature measurement.

Temperature sensors

For temperature sensors it is vital that good thermal contact is maintained to provide accurate measurements. Thermowells are typically used where it is necessary to directly measure the temperature of liquid nitrogen or helium. The thermowell is used to protect the delicate temperature sensors from damage and also allows the placement of the temperature probe into the area of interest, for example into the middle of a pipeline. One challenge of using a thermowell is that good thermal contact needs to be made between the outer wall and the temperature sensor. Hydrocarbon grease is often used to fill thermo-wells in cryogenic equipment, in order to provide this necessary contact between sensor and the medium to be measured. In this type of scenario the grease is ideal as it is easy to apply to the thermo-couple, or insert into the well. The fact that the grease will soften at room temperature means that the sensor head can be easily removed from the thermowell for servicing or replacement if necessary, something that is not possible with an epoxy resin, or similar permanently set thermal interface material. Once the probe is replaced and cooled down the grease hardens once more, providing excellent thermal contact to the outer surface.

Figure 1 - Thermowell application for grease



Cryo-lubrication

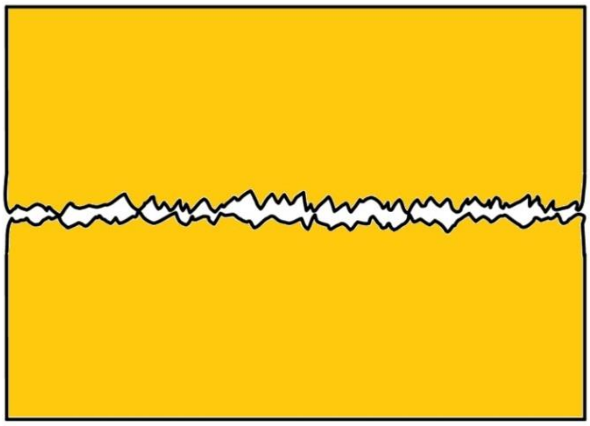
One area where greases are not able to provide a solution is in cryo-lubrication. Regardless of the base oil, used greases will be solid below around 170K, thus proving unsuitable for providing lubrication while in operation. Where grease can be used is as an assembly lubricant, where the compound aids in the construction of a cryogenic system, such as allowing the insertion of a shaft through an O-ring, without damaging the elastomer. In this case grease that can withstand thermal cycling is again useful as it allows the removal and replacement of the shaft for maintenance purposes. However it should be noted that dynamic systems for operating at very low temperatures will require the use of solid lubricant coatings.

Thermal Transfer Performance

Since thermal transfer at cryogenic temperatures is a key area where specially formulated grease can be used, it is useful to look at the comparative performance in more detail. One of the key problems with cryogenics is providing good heat transfer between the cooling equipment and the experimental object. Take for example the mounting of a semiconductor chip on to a cryostat cold finger assembly. In order for the chip to be effectively cooled there is a need to provide excellent thermal contact between the two surfaces.

At cryogenic temperatures thermal transfer is predominantly through direct conduction, since the influences of radiation and convection are negligible. When considering conduction of heat it is necessary to have a good contact between the materials and at the macroscopic level it may seem that two adjoining surfaces are flat, but when looked at on a microscopic level it can be seen that this is not true. The diagram in Figure 2 shows how surface roughness can cause the actual surface area in contact between two materials to be very small. In fact due to microscopic roughness the actual contact area can be as low as 1-2% of the total surface area between two materials. [ii] At room temperature this is less of an issue, since the movement of gas molecules between the two materials assists the heat transfer. However the process is quite inefficient and once the temperature is close to absolute zero the movement of gas molecules slows and thermal transfer is dominated by conduction.

Figure 2 – Surface roughness impact on contact area



The picture in Figure 3 shows the surface of a copper plate at 6000 times magnification, clearly demonstrating the kind of imperfections that can exist, even when the surface appears smooth to the naked eye.

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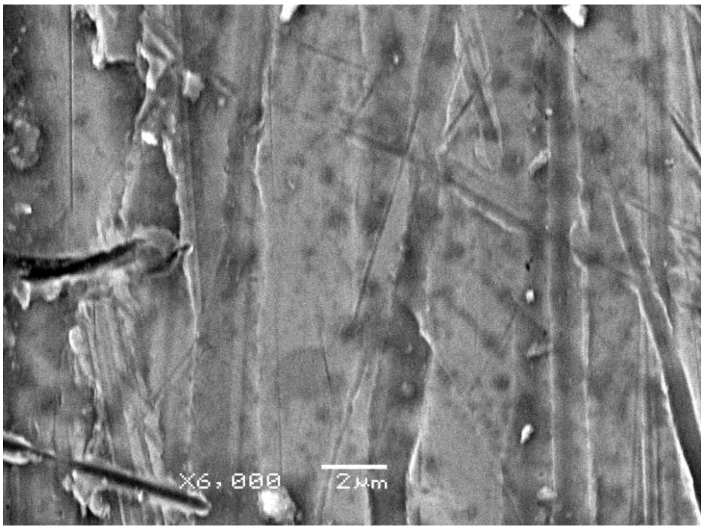
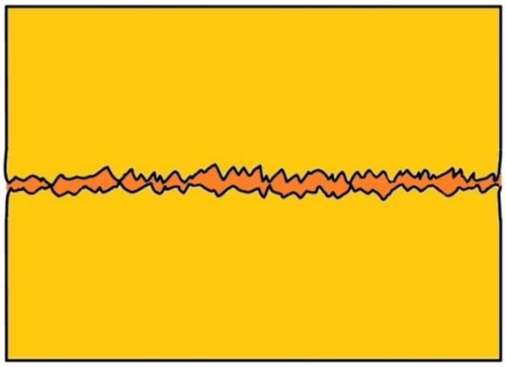
Figure 3 - SEM image of a copper surface



The principle of using a heat transfer grease is straightforward. The grease provides a direct thermally conductive path between the two materials, as shown in Figure 4, which eliminates all the free space and fills in the peaks and valleys.

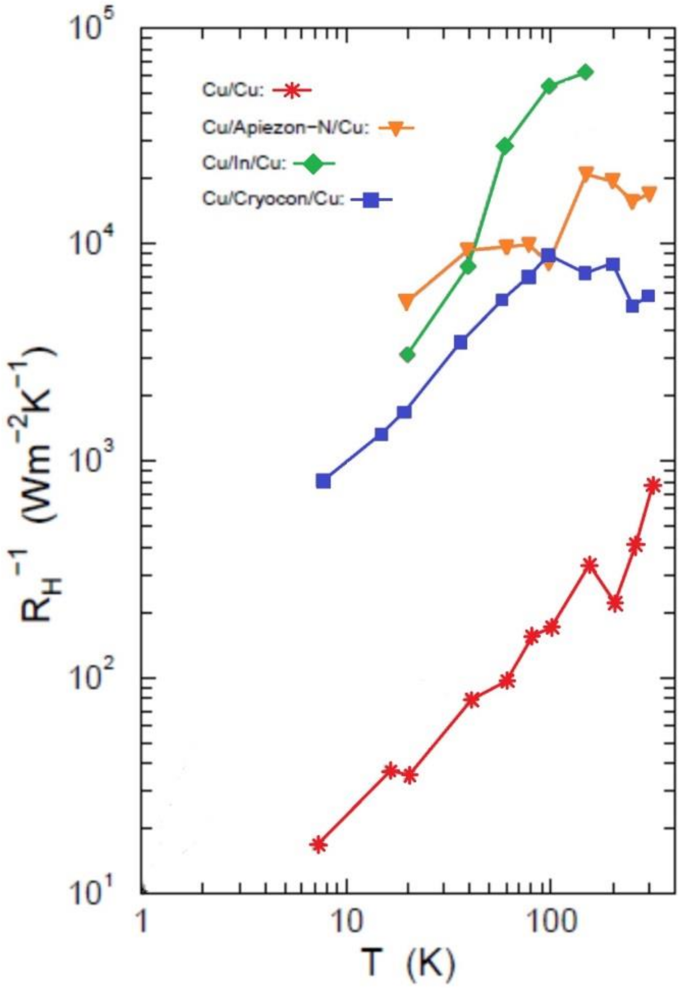
Figure 4 - Application of a thermal grease



There are a vast range of heat-sink compounds which are formulated to provide good thermal conductivity, and some of these contain metal particles to improve the bulk thermal conductivity. The big drawback with these materials for cryogenic use is that the addition of particles increases the space between the contact surfaces and can actually become detrimental to the system's thermal transfer characteristics. Two other well accepted methods for heat transfer under cryogenic conditions are either Indium foil or specially formulated hydrocarbon grease, which has additives to prevent cracking or crazing at very low temperatures.

When comparing the effectiveness of these three solutions it is possible to look at the complete thermal range from room temperature down to liquid helium temperatures, where low temperature superconductors operate. The chart in Figure 5 compares the thermal conductance between copper contacts filled with hydrocarbon grease Apiezon N, a Cu filled grease compound named Cryocon or Indium foil between two copper contacts.[ii]

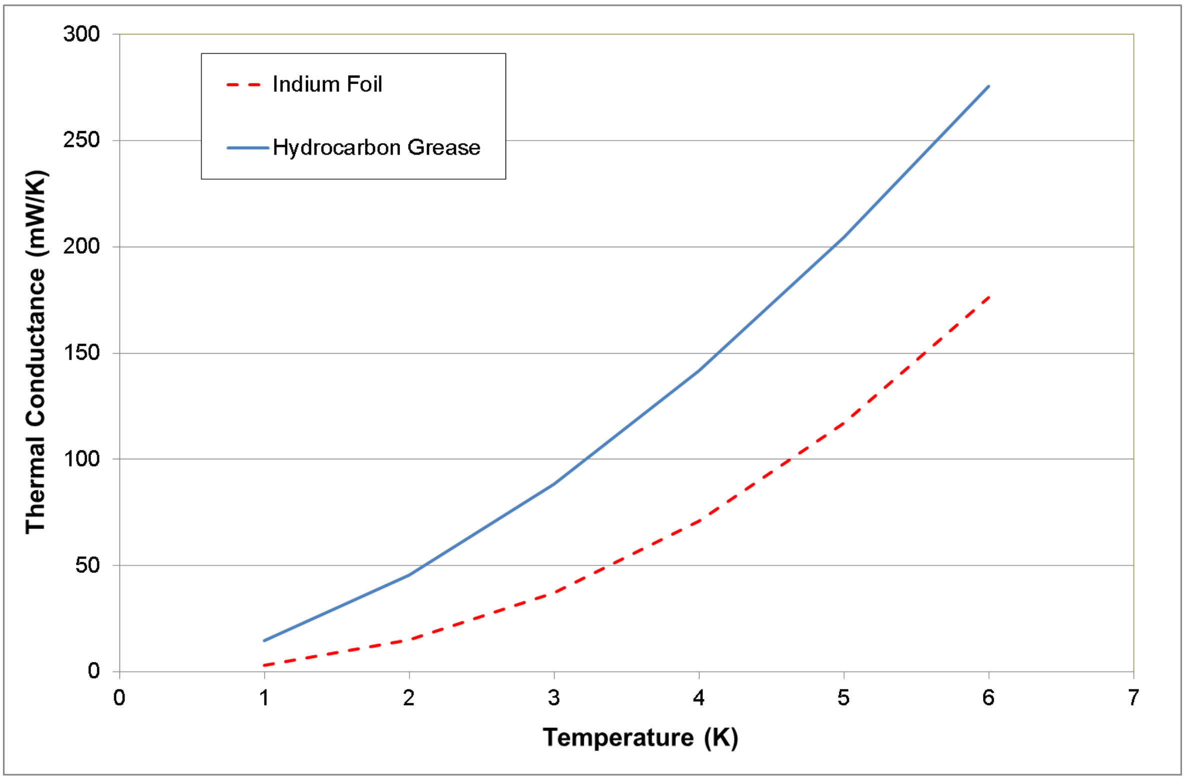
Figure 5 - Thermal conductance of copper contacts with various thermal interfaces



It can be seen that at higher temperatures the indium foil gives the best thermal conductance, but that as the temperature drops below around 40K the Apiezon hydrocarbon grease becomes more effective. The cryocon metal filled grease has a higher thermal conductivity when measured in isolation, but in this type of application the benefit of adding metal particles is reduced, as the distance between the contacts is larger in comparison to the other two solutions.

Taking research into liquid helium temperatures it can be seen that the hydrocarbon grease has much better performance than indium foil when the temperatures drop further to the 1-6K range, as shown in Figure 6[iii]

Figure 6 - Thermal conductance of copper contacts at liquid helium temperatures



These experiments suggest that for cryogenic temperatures hydrocarbon grease offers an excellent thermal contact material, especially for applications which run in the liquid helium range.

A further advantage to using hydrocarbon grease is the ability to easily remove it. This can be especially useful when conducting scientific experiments where a sample needs to be temporarily mounted to a cryostat surface. If indium foil is used then it becomes very difficult to remove the foil once the experiment is complete, whereas hydrocarbon grease can be removed completely by the use of hydrocarbon solvents, such as toluene or d-limonene.

Vapour pressure and vacuum applications

In addition to providing thermal contact hydrocarbon grease can also provide an excellent sealing medium for vacuum systems that are required to operate at cryogenic temperatures. In this type of application key features of the product are the ability to remain crack free down to very low temperatures and to have a low vapour pressure, to prevent outgassing of unwanted material into the vacuum environment.

Figure 7 shows the vapour pressure curve for the same Apiezon grease, showing that the product can be used as a sealing compound, as well as a thermal transfer material.

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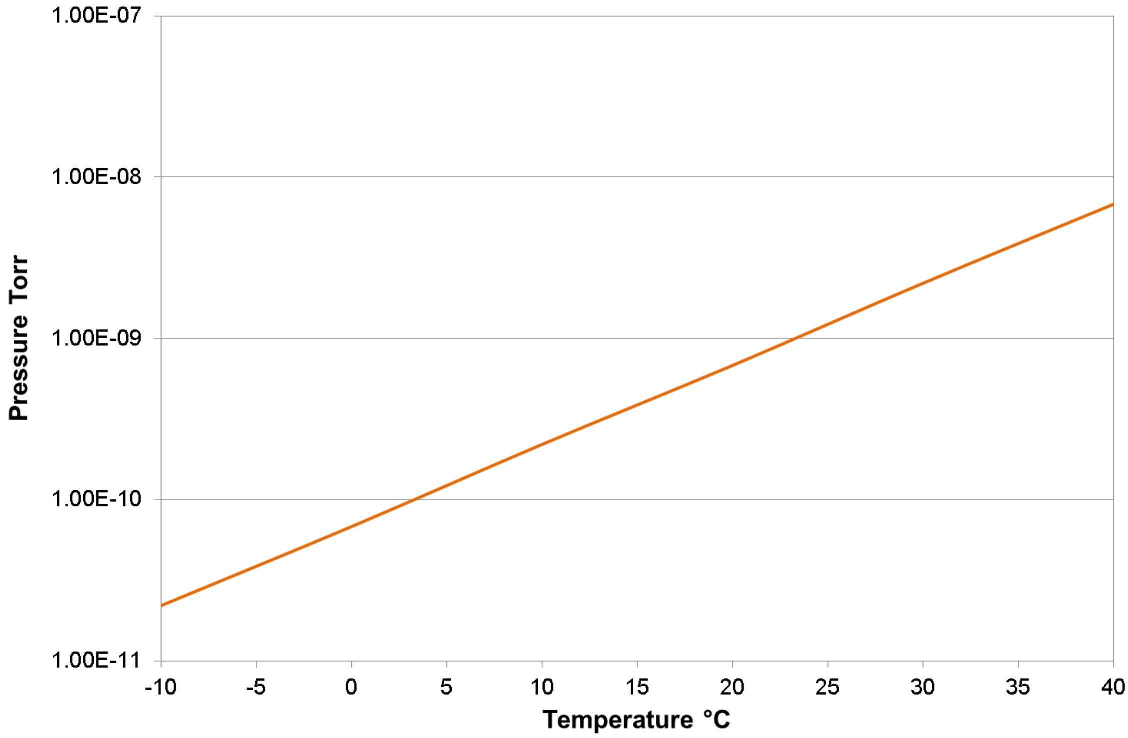
Figure 7 - Vapour pressure of grease



So long as the pressure is kept above the curve for a given temperature no significant outgassing should occur. For example if operating at -10°C the grease will withstand a vacuum pressure of down to 2.2×10^{-11}Torr, which would be considered high vacuum. As the temperature is reduced into the cryogenic range the vapour pressure will decrease, ensuring no outgassing even at ultra-high vacuum.

Conclusions

For cryogenic applications there is often a need to provide good thermal conductivity between two surfaces, down to temperatures as low as a few Kelvin. In order to achieve this a compound is needed which will provide the necessary thermal conductance and survive cycling to extremely low temperatures.

A hydrocarbon grease has been developed and used for many years to provide excellent performance down to liquid helium temperatures. As well as providing good thermal conductivity it can be repeatedly cycled back to room temperature, to allow the removal and replacement of components, such as test devices on cryostat cold fingers. The grease can also be fully removed with the use of common hydrocarbon-based solvents.

This grease can also be utilised as a vacuum sealant, providing very low vapour pressure, which in turn allows use at extremely low pressures into the ultra-high vacuum ranges.

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[i] Apiezon N Grease, Cryogenic High Vacuum Grease, Product Datasheet, November 2012
[ii] Thermal boundary resistance of mechanical contacts between solids at sub-ambient temperatures, E. Gmelin, M. Asen-Palmer, M. Reuther and R. Villar, J. Phys. D: Appl. Phys. 32 (1999) R19-R43
[iii] Thermal conductance of metallic contacts augmented with Indium foil or Apiezon N grease at liquid helium temperatures, J. Salerno, P.Kittel, A.L. Spivak, Cryogenics 1994, Volume 34, Number 8