Aerogel-Based Insulation Materials for Cryogenic Applications

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Abstract. Many different aerogel-based materials are now being used in thermal insulation systems for cryogenic applications. These materials include flexible composite blankets, bulk-fill particles, and polymer composites in both evacuated and non-evacuated environments. In ambient environments, aerogels provide superior thermal performance compared to conventional polymeric foam and cellular glass insulations while offering unique advantages in avoiding problems with weathering, moisture, and mechanical damage. Aerogels are also used as spacer materials in multilayer insulation systems. These layered systems provide combined structural-thermal capability for cryogenic systems in either vacuum-jacketed or externally-applied insulation designs. Test data (effective thermal conductivity) include a wide range of both commercial and experimental aerogel materials. Testing was performed using laboratory cryostats and standard methods including full range vacuum (from ambient pressure to high vacuum) and boundary temperatures 293 K and 78 K. Examples of aerogel-based insulation systems are given for both evacuated and non-evacuated applications.

1. Introduction

Thermal insulation systems for cryogenic applications span a wide range of requirements and call for demanding levels of performance. Aerogels and their composites continue to be developed to take advantage of highly tailorable processing techniques that enable specialized end products and unique thermo-physical properties attributed to their nano-porous internal networks. Following breakthroughs in the area of solution-gelation process chemistry in the 1980s to 1990s, a number of aerogel materials have become commercially available in three categories: flexible composite blankets, bulk-fill particles, and layered composite systems. However, cryogenic insulation systems are technically complex and must be designed to meet a host of requirements including (just to name a few) mechanical, thermal, chemical compatibility, size/weight, and environmental or vacuum exposure. Added to these list of requirements is the need for cost effectiveness in design as performance must always justify the cost. Although it is true that an insulation system is better than another insulation system only in the comparative sense, understanding the real-world performance of the installed system is the key piece of information that trumps all others.

Aerogel-based materials used in cryogenic thermal insulation systems are economically justified in many instances. But like any other insulation, none is a universal fit for all situations. Testing has included cryostat thermal performance using boiloff calorimetry in environments from ambient pressure to high vacuum. Examples of field applications include liquefied natural gas (111 K), liquid oxygen (90 K), liquid nitrogen (77 K), cryo-compressed hydrogen (~40 K), and liquid hydrogen (20 K).

2. Materials and Insulation Systems

A total of six different aerogel materials were tested for cryogenic-vacuum thermal performance. The aerogel bulk-fill material was manufactured by Cabot Corp. and all aerogel blanket materials were manufactured by Aspen Aerogels, Inc. The aerogel blanket materials, with the exception of the melamine type material (G2-113) are all composite materials that include a fiber reinforcement material in the production process. In addition, two systems are layered composite insulation (LCI) systems of radiation shielding layers in combination with aerogel composite blanket materials. System G1-191 is a layered composite of pairs of ultra-low density (ULD) aerogel blanket and double-aluminized Mylar film. System A193 is a layered composite of pairs of aerogel composite paper (0.7 mm thick) and double-aluminized Mylar film. The aerogel materials/systems tested and their basic physical properties are summarized in Table 1.

Additional insulation materials and systems have been similarly tested for comparison and reference [1]. The descriptions of these additional test specimens are given in Table 2. These materials include "vacuum only" and a closed-cell rigid polyisocyanurate foam [2] on one end of the spectrum of thermal performance followed by the glass bubbles system [3] and a baseline of multilayer insulation (MLI) on the other end [4].

Table 1. Physical properties of aerogel-based test specimens.

Cryostat	Test Series	Test Specimen	No. of Layers	Total Thickness* (mm)	Density* (kg/m^3)
C100	A108	Bulk-fill aerogel beads	1	25	80
C100	A111	Pyrogel® aerogel blanket (black)	6	18	125
C100	A194	Cyrogel® aerogel blanket	2	20	130
C500	G2-109	Spaceloft® Subsea (grey)	4	20	152
C500	G1-190	ULD^ aerogel blanket white	8	23	55
C500	G2-113	ULD^ melamine flexible aerogel grey	8	21	65
C500	G1-191	ULD^ Aerogel MLI layered composite	8	23	52
C100	A193	Aerogel MLI layered composite (0.7-mm aerogel paper)	7	5	91

*As tested.
^Ultra-Low Density (ULD).

Table 2. Physical properties of additional insulation test specimens for comparison.

Cryostat	Test Series	Test Specimen	No. of Layers	Total Thickness* (mm)	Density* (kg/m^3)
C100	A114	Vacuum Only (black surfaces)	1	25	n/a
C500	G1-157	SOFI Foam BX-265	1	25	42
C100	A102	Glass Bubbles K1	1	25	65
C100	Total of 26	Kaganer Line (MLI Baseline); average of 26 different MLI test specimens	From 10 to 80	~22 typical	~50 typical

*As tested.

Three main limitations on the use of MLI systems are summarized as follows: 1) high vacuum is required for operation (and in the first place, it is not possible to vacuum-jacket all hardware), 2) not all hardware can be suitably wrapped or properly covered, and 3) localized compression will ruin the thermal performance. MLI cannot withstand mechanical loading. Compared to the no load (<0.007 kPa) condition for six different MLI systems tested with an average heat flux of 0.6 W/m^2, a mere 0.7 kPa (0.1 psi) will cause a 15x increase in heat flux. A modest 7-kPa load will cause an approximate 40x increase while a 70-kPa load will cause a more than 100x increase [5].

For a given cryogenic application, how should one choose among MLI, bulk-fill, foams, aerogels, or layered composites of aerogel blankets or some combination of materials? The choice depends on four main factors: 1) heat load requirement; 2) physical design of system; 3) installation build process; and 4) operational and maintenance requirements.

In ambient pressure applications, an alternative to closed-cell foam is the layered composite extreme (LCX) system as previously reported [6]. The LCX system is an MLI system but for open-air environments. Its various combinations of aerogel blanket and compressible barrier layers, provide unique performance benefits where complex shapes, weathering, moisture, and mechanical damage are problematic. This breathable (non-sealed) type insulation system has been proven at 20 K on operational LH2 systems based on the characteristic of the aerogel as a hydrophobic, nano-porous, and amorphous (non-cellular) composite that does not cryopump, beyond initial cooldown, even as low as 4 K [7]. Select aerogel blanket materials such as the Pyrogel® product line of Aspen Aerogels provide high temperature capability to 923 K (1200 °F) where fire protection might be needed for cryofuel systems.

3. Experimental Method and Apparatus

The experimental work used two cryostat test instruments for absolute thermal performance measurement in the full range of vacuum-pressure environments. Both are steady-state boiloff calorimeters using liquid nitrogen as the cryogen. The Cryostat-100 has a vertical cylindrical cold mass assembly that is 1-m tall by 167-mm diameter while the Cryostat-500 has a horizontal flat plate cold mass assembly of 204-mm diameter [8-9]. The cold and warm boundary temperatures for all tests are 78 K and 293 K, respectively, unless otherwise noted. The large temperature difference (ΔT) is representative of the majority of cryogenic applications where the objective is to isolate the cold from the ambient (hot) environment). The effective thermal conductivity (k_e in mW/m-K) and heat flux (q in W/m^2) are calculated for the standard ΔT of 215 K. The test methodology follows the guidance of ASTM C1774, Annex A1 and Annex A3, respectively [10]. In addition, the standard ASTM C740 is followed as applicable for MLI systems [11]. Simplified schematics for both cryostats are given in Figure 1.

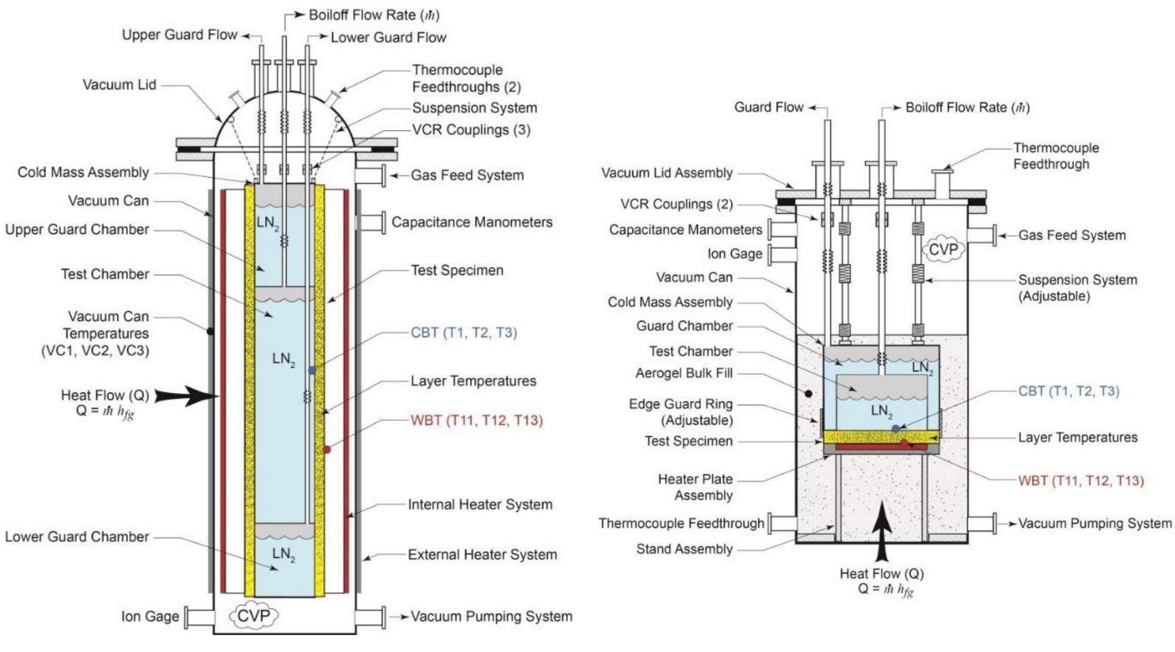

Figure 1. Simplified schematics for insulation test instruments Cryostat-100 (left) and Cryostat-500 (right).

For high vacuum tests, each test specimen was heated and evacuated according to standard laboratory procedures. The typical process includes heating to approximately 323 K in conjunction with evacuation and gaseous nitrogen purge cycles (a minimum of three times) followed by at least 48 hours of continuous vacuum pumping.

Temperature sensors (Type E, 30 gage thermocouples) are placed within the specimen at specific intervals through the thickness from the cold side to the warm side. Because the thermal performance is measured under steady-state conditions, the heat flow rate through the thickness of the test specimen in constant for all layers and at all points through its thickness. With the additional temperature sensors, intermediate or interlayer thermal conductivity values (λ) can be calculated and reported with the mean temperature (T_m) for each layer. Four interlayer temperature sensors were used. Therefore, a total of 14 λ points can be calculated in addition to the k_e for the full ΔT. In this way, a single test can yield a host of data points for determining the temperature dependence of heat transmission through a test specimen operating between a large temperature difference.

4. Cryogenic-Vacuum Test Results

The cryostat test results for all test series are presented in Figure 2. Included for comparison are the additional materials/systems as discussed. For all tests, the boundary temperatures are approximately 293 K and 78 K and the residual gas in nitrogen.

4.1. Comparisons among, aerogels, layered aerogels, and baseline materials

For each test specimen, the total thickness, number of layers, and bulk density are given in the legend. In higher vacuum, the higher thermal conductivity systems are foam (G1-157) and "vacuum only" (A114) while the lowest thermal conductivity systems are the MLI baseline (Kaganer line) and layered composites of aerogel/MLI. The six aerogel materials (without any radiation shield layers) are generally in the middle at CVP below 10 millitorr, but all are superior at CVP above 1000 millitorr. The data for glass bubbles (A102), dominated by gas conduction heat transfer, clearly shows the free molecular to continuum transition at around 50 millitorr. In the soft vacuum region up to ambient pressure (760,000 millitorr), all aerogel materials are obviously superior. The two layered composites (A193 and G1-191) are examples of combining the advantages of two different material systems (aerogel and MLI) for specific applications that may require tolerance for vacuum degradation and/or mechanical loading.

4.2. Effective thermal conductivity of aerogel materials

The results for the six aerogel materials are repeated in Figure 3 for detailed examination. In high vacuum, the ULD aerogel (G1-190) and Spaceloft Subsea Gray (G2-109) gave the lowest k_e. Through the moderate vacuum (from 50 to 1,000 millitorr) and soft vacuum (from 1,000 to 50,000 millitorr) ranges is where some significant differences appear according to the characteristics of the aerogel on the nanoscopic level, the fiber matrix (if any) and physical arrangement on the microscopic level, and the interstitial spaces (if any) on the macroscopic level. Even with this simplistic analytical view, the differences can at least be somewhat resolved by considering the four modes of heat transfer (solid conduction, radiation, gas conduction, and convection) all in play for this region of CVP.

Additional test data points (not shown) were taken in all cases to verify the unique shapes of the data for each aerogel material. For example, the aerogel beads (A108) specimen has more free interstitial space for gas conduction and convection to occur while the ULD aerogel (G1-190) specimen has the finest pore size and hence lowest gaseous conduction. For ambient pressure conditions, the two ULD aerogels show the highest k_e while the twice heavier Cryogel (A194) is superior. Complete data sets for the Cryogel material, including different gas environments, is previously reported [12].

Considering the relative proportions of the individual modes of heat transfer is the starting place for the detailed design and analysis of any cryogenic insulation system whether it is for high vacuum, moderate/degraded vacuum, soft vacuum, or ambient pressure.

4.3. Layer temperature profiles and lambda calculation

Plots of the layer temperature distributions for a select test specimen (ULD Aerogel – G1-190) are given in Figure 4. Other material-only (no radiation shield layers) data are similar. The key distinction is the progression of inflection of the curves from high vacuum to soft vacuum where the steepest inflection always occurs at the first layer (coldest layer) and under higher vacuum conditions.

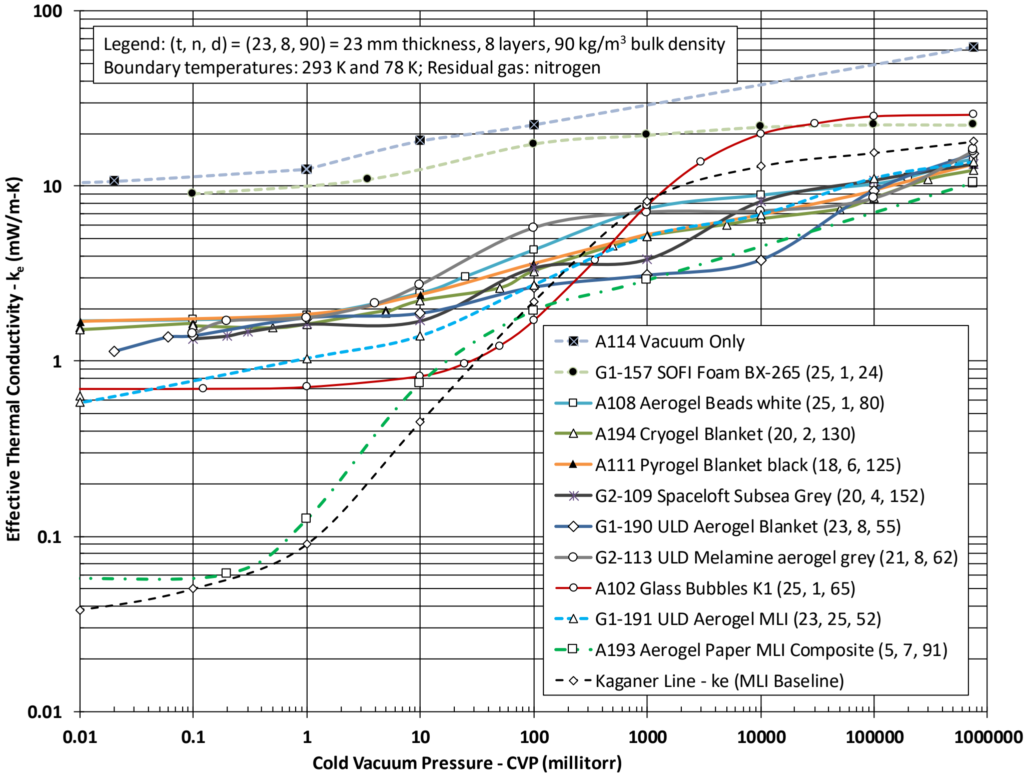

Figure 2. Variation of effective thermal conductivity with cold vacuum pressure for aerogel materials in comparison with a variety of other cryogenic insulation systems (boundary temperatures are 293 K and 78 K; residual gas is nitrogen).

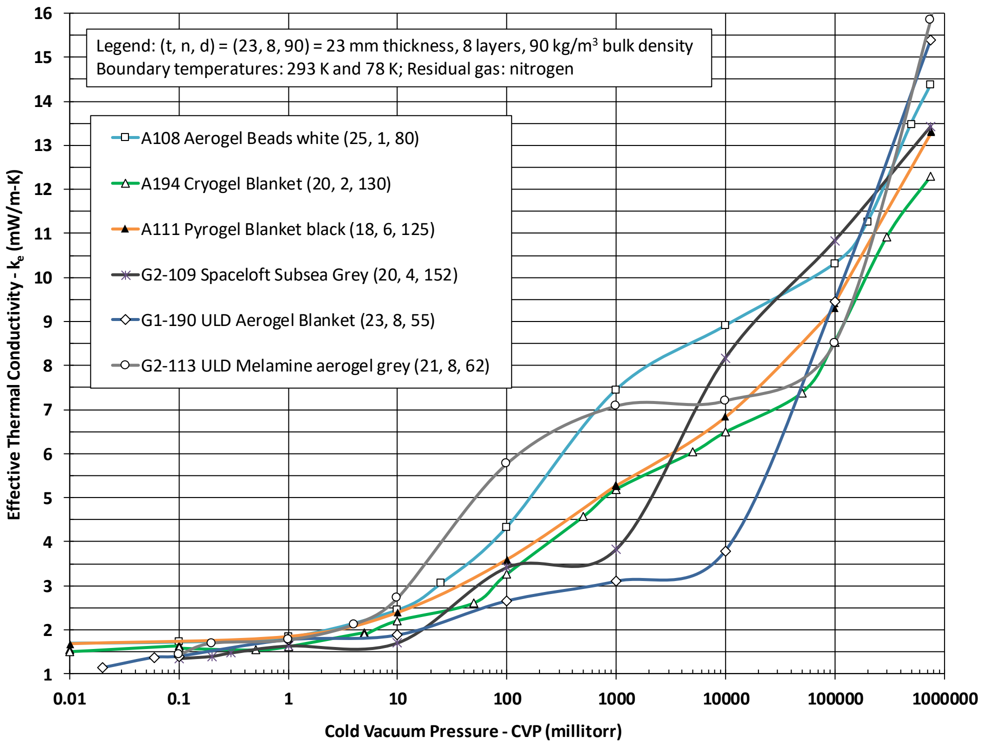

Figure 3. Variation of effective thermal conductivity with cold vacuum pressure for aerogel materials (boundary temperatures are 293 K and 78 K; residual gas is nitrogen).

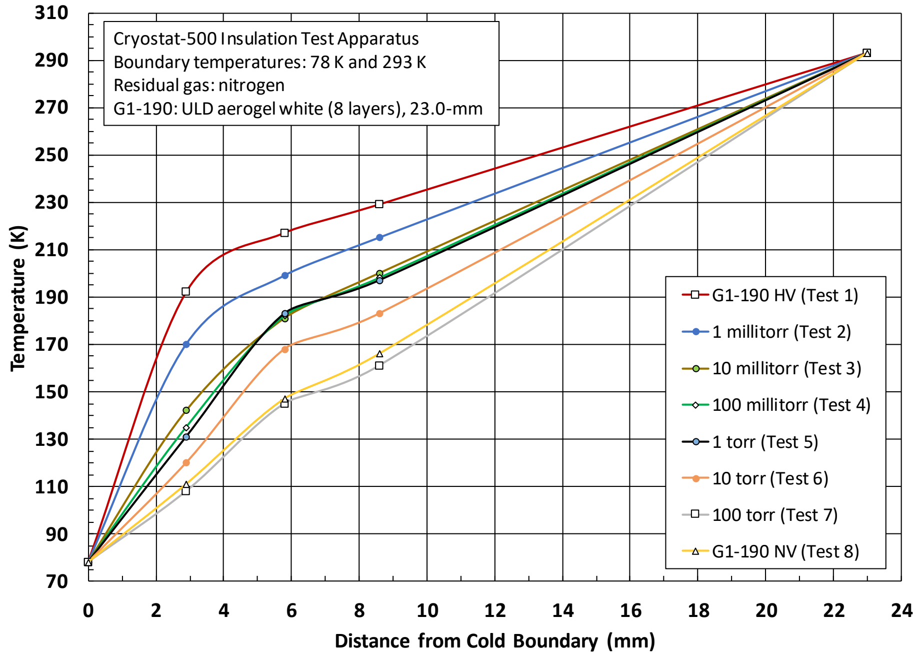

Figure 4. Layer temperature profiles for G1-190 ULD aerogel white for all cold vacuum pressures.

5. Thermal Analysis and Discussion

Here we will examine two areas of detail with respect to layered composite insulation (LCI) systems. In this case, we refer to alternating layers of aerogel blanket material and radiation shielding films. The arrangement of aerogel blanket materials and MLI systems, in different combinations has been previously reported [13].

The given cryostat data are for the laboratory standard temperatures of 293 K (warm boundary) and 78 K (cold boundary). Using standard temperatures facilitates repeatable testing and is helpful for direct comparison among different materials/systems. However, most applications may pertain to conditions above the standard 293 K warm boundary temperature and/or with cold boundary temperatures above (111 K for LNG) or below (20 K for LH2) the standard 78 K. Therefore, tools for analyzing the boundary temperature extremes and the internal (through-the-thickness) temperature distributions are important to arrive at the proper engineering of a given end-use application.

5.1. Lambda calculations for temperature dependence

The intermediate or interlayer temperature sensors used in the cryostat testing provide additional data to determine the temperature dependence of thermal conductivity within the two prescribed boundary temperatures. The use of three intermediate temperature sensors creates four layers, numbered from one to four, from the cold side. Given below are the basic nomenclature and equations:

$$Q = k_e * A_e * \Delta T/\Delta x \quad \text{(Fourier equation)} \quad (1)$$

$$q = Q/A_e \quad \text{(constant)} \quad (2)$$

$$q = q_1 = q_2 = q_3 = q_4 = \lambda_4 * \Delta T_4/\Delta x_4 \quad \text{(and so on)} \quad (3)$$

$$T_m = (T_{colder} + T_{warmer})/2 \;\; \text{or} \;\; T_{m4} = (T_{c4} + T_{w4})/2 \quad \text{(and so on)} \quad (4)$$

The heat flux (q) is constant for a steady-state heat flow rate and therefore the heat flux through each layer of the flat disk test specimen is constant. Because the ΔT and Δx for each layer is the known, the thermal conductivity (λ) for each layer can be readily calculated and reported for the corresponding mean temperature (T_m). Plotted in Figure 5 are the resulting calculations of the intermediate thermal conductivity values (λ) as a function of the mean temperature (T_m).

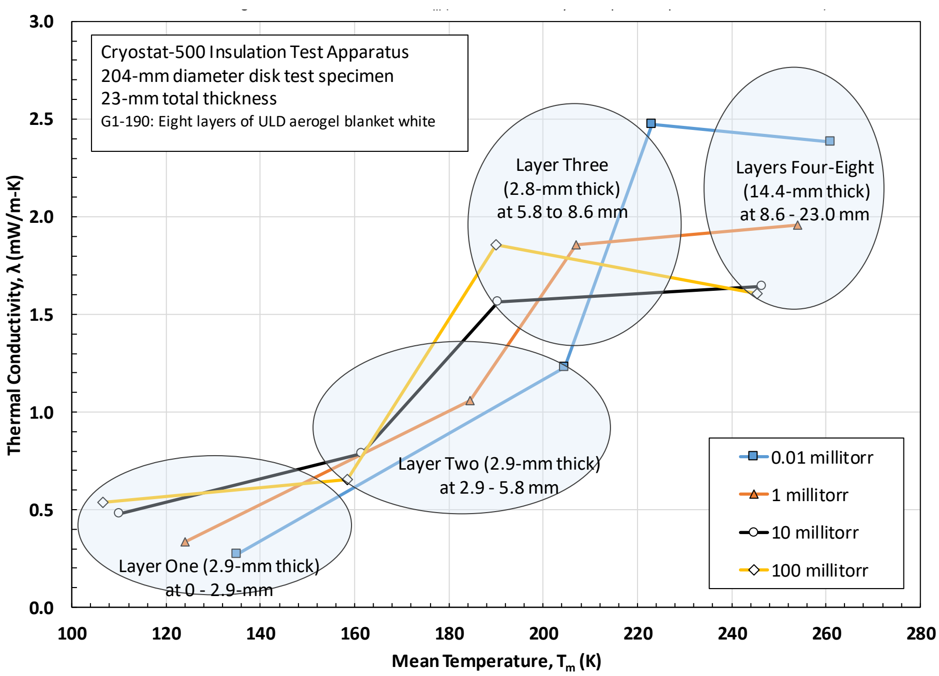

Figure 5. Intermediate thermal conductivities calculated for G1-190 ULD aerogel white: variation of lambda with mean temperature for four different cold vacuum pressures.

5.2. Thermal performance estimates for warmer/colder boundary temperatures

Aerogel-based MLI systems (layered composites) like the A193 and G1-191 systems given in Figure 2 provide options for cryogenic insulation where the factors of compression loading, installation, vacuum degradation, and operational efficiency come into play. However, needed for any approach is an estimate of the system thermal performance for specific boundary temperatures. A library of test data for MLI under high vacuum ($<10^{-5}$ torr) provides a preliminary way of estimating the increases in heat transmission for warm boundary temperatures up to 350 K [4]. Given in Table 8 are the increases in heat flux (q), on a percentage basis, for an average of 12 different MLI systems from a baseline WBT of 293 K. Conversely, data from Flynn provide the reductions in heat flux for an MLI system with decreasing cold boundary temperatures compared to a 78 K baseline [14].

From baseline heat flux (q_{base}) test data at the standard boundary temperatures of 293 K and 78 K, a first-order estimation of the thermal performance for a specific layered system design is calculated using a warm boundary temperature factor (b_w) and a cold boundary temperature factor (b_c) as follows:

$$q_{design} = b_c * b_w * q_{base} \qquad (5)$$

For example, the heat flux estimate for a system operating at boundary temperatures of 325 K / 78 K is approximately the same thermal performance as the baseline of 293 K and 78 K (q_{design}=1.32*0.79*q_{base} = 1.04*q_{base}. Even though the theoretical heat flux is proportional to the ΔT (and T^4 for the radiation portion), the more important and influential factor is the materials' heat transmission characteristics that occur at the progressively lower temperatures combined with the likely improvement of the level of vacuum. The vacuum level inside the materials is always the dominate factor, but testing under real-world conditions is required to determine its effect.

Table 8. Increase of heat flux for increasing WBT (for MLI system with constant CBT = 78 K) [4].

WBT (K)	ΔT	% increase, ΔT	% increase, q	factor b_w
293	215	baseline	baseline	1.00
305	227	6	14	1.14
325	247	15	32	1.32
350	272	27	46	1.46

Table 9. Increase of heat flux for decreasing CBT (for MLI system with constant WBT = 300 K) [14].

CBT (K)	ΔT	% increase, ΔT	% increase, q	factor b_c
76	224	baseline	baseline	1.00
40	260	16	14*	0.86
20	280	25	21	0.79
4	296	32	33	0.67

*Interpolated value

6. Conclusion

Cryogenic-vacuum thermal performance of aerogel-based insulation systems is provided for a number of different systems for different applications. Examples of field applications show the unique thermo-economic performance advantages of insulation systems when combined the proper understanding of laboratory data, design approach, and reality of installation on complex hardware. Aerogels include blanket type, bulk-fill type, and layered composites including radiation shield layers. Each has its own advantages and weaknesses with the key being understanding of the installed (rendered) system in comparison with alternative system designs.

Future aerogel materials under development will likely lead to further advances that may enable entirely new cryogenic applications and multi-functional systems. However, a number of aerogel materials are widely commercially available today, proven for cryogenic use in both vacuum and non-vacuum environments at temperatures from approximately 4 K to 400 K, and higher.

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