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UC-33, Propulsion Systems
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UNION CARBIDE CORPORATION

LINDE DIVISION

FINAL REPORT OF RESEARCH AND DEVELOPMENT

IN A THERMAL INSULATION STUDY

Phase I

March 28, 1966 - September 27, 1967

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A B S T R A C T

The objective of this development program was to evaluate, develop where necessary and demonstrate thermal insulation systems for use in radioisotope power systems. The insulation systems investigated were multilayer type where highly reflective radiation shields separated by low thermal-conductivity spacers operate in a vacuum.

Radiation shield material selections of aluminum, copper and nickel based on previous work were verified and an operating temperature-design life trade off study based on loss of shield by vaporization was performed. As a result, the maximum operating temperature for each of the above foils of given thickness and design life has been determined. Tantalum foil had been considered for use at temperatures above the use temperature of nickel but was rejected due to observed incompatibility with silica fiber spacers. The fibrous spacers evaluated for use were glass fiber paper, aluminum flake opacified glass fiber paper, Refrasil quartz fiber paper, copper flake opacified Refrasil quartz fiber paper and Astroquartz cloth.

The thermal conductivity of six different spacer-shield combinations was determined as a function of temperature. The measured thermal conductivities ranged between $0.15-3.5 \times 10^{-3}$ BTU/hr.ft. $^{\circ}$ R over the temperature range $T_{\text{hot face}}=400^{\circ}\text{F}$ to 1800°F and $T_{\text{cold face}}=70^{\circ}\text{F}$. Three of the combinations were tested for 2000 hours with no thermal performance degradation. A fourth, tantalum foil-Astroquartz cloth, exhibited degradation due to the formation of high emissivity compounds of tantalum.

An analytical study was made to determine the thermal effects of air exposure during ground hold conditions on multilayer insulations at operating temperatures and in vacuum. All three radiation shield materials, aluminum, copper, and nickel are capable of some air exposure before deterioration of thermal conductivity in vacuum occurs. An analytical method was developed to predict the thermal conductivity of insulation in vacuum after air exposure. Some experimental verification was achieved using a copper-refrasil system.

An analytical study was made to assess the effect of various edge conditions on the thermal performance of multilayer insulation panels. The effects of panel width, gap width or angle between panels, panel thickness and exposed edges were investigated. The thermal conductivity of a panel ten inches wide, 0.2 inches thick with a gap of 0.050 inch would be about 1.5 times that of a panel without a gap. A preliminary assessment of the affect of filling the gap with a medium performance insulation material shows that considerable improvement that can be attained.

The processing required to render the radiation shield foils and spacers physically and chemically stable at operating temperature in vacuum was determined.

Low offgassing rates, below 0.1 micron liters per hour per square foot were measured for all materials at their respective operating temperatures, except Astroquartz Cloth and Refrasil quartz fiber paper which exhibited this rate up to 1700°F. At 1800°F these materials exhibited offgassing rates of 1-3 micron liters per hour per square foot. Offgas compositions were determined by mass spectrometer gas analysis and found to be predominantly hydrogen.

A preliminary evaluation of the problems associated with the design and fabrication of high temperature vacuum enclosures was made. An Inconel-600 test specimen was fabricated and tested at 1800°F in a one side vacuum, one side air environment. Leakage developed after three hundred hours with cause of failure attributed to oxidation penetration accelerated by the high stress level. A second specimen, Hastelloy-X, successfully completed 10,000 hours of testing at 1400°F with a vacuum on one side and air on the other.

Material compatibility experiments to determine long-term compatibility of insulation materials with themselves and with vacuum enclosure materials in vacuum and at operating temperature were made. Tantalum foil and silica fibers were found to be incompatible.

The resistance of five processed insulation combinations to vibration levels typical of service requirements was evaluated with no physical deterioration of any of the combinations observed by visual inspections.

Physical and chemical tests were performed on the spacers in the "as-received" and "after processed conditions" to determine their suitability for insulation application. All candidates possessed sufficient strength after processing for insulation application.

Based on the results of this program the feasibility of using the multifoil type of insulations on radioisotope power systems has been demonstrated.

1.0 SUMMARY

The insulation systems studied under this contract were the multi-foil type in which reflective radiation shields are separated by low thermal conductivity spacers.

For a composite insulation system operating between ambient and 1800°F with a life of 1 to 5 years the following radiation shield materials; aluminum, copper and nickel were recommended. Tantalum foil, selected as an alternate for nickel was rejected because of incompatibility with silica fiber spacers. Glass fiber paper, aluminum flake opacified glass fiber paper, Refrasil quartz fiber paper, copper flake opacified Refrasil quartz fiber paper and Astroquartz cloth were the spacer materials evaluated.

Thermal conductivity as a function of temperature was determined for selected combinations of the above foils and spacers and the low thermal conductivities anticipated were experimentally verified. During 2000 hour thermal stability tests of selected insulation combinations the only combination displaying thermal degradation was tantalum foil-Astroquartz Cloth. This was attributed to the formation of high emissivity compounds of tantalum.

An analytical study to assess the effects of insulation panel width, gap width between panels and panel thickness on panel thermal performance was made. The thermal conductivity of a panel 10 inches wide, 0.2 inches thick with a gap of 0.050 inches is about 1.5 times greater than the thermal conductivity of the same panel without a gap. This deterioration is considerably reduced by introduction of a low conductivity filler in the gap.

An analytical study to determine the thermal effects of air exposure during ground hold conditions on insulations at operating temperature and in vacuum showed that all three foil materials, aluminum, copper, and nickel, can be exposed to some extent before significant degradation occurs. An analytical method was developed to predict the effect of air exposure on the subsequent vacuum performance.

Insulation processing requirements were established for the foils and spacers to render the insulation physically and chemically stable at operating temperature and under vacuum.

Offgassing rates and compositions were determined for the insulation materials and getter requirements to maintain low pressure in sealed systems established.

A preliminary evaluation of the problems associated with the design and fabrication of high temperature vacuum enclosures was made. One enclosure specimen, Hastelloy-X, successfully completed 10,000 hour of testing at 1400°F. with an external vacuum and internal air pressure.

A first order assessment of the physical resistance of processed insulations to vibrational loads, typical of service requirements, was obtained. No physical deterioration of any of the combinations was observed following the limited testing.

Significant progress has been made toward the objectives of evaluation and demonstration of using the multi-foil type of insulations on radio-isotope powered heat sources and thermal-electric conversion devices.

2.0 INTRODUCTION

Multi-foil insulations can provide an extremely efficient solution to problems of thermal insulation where total insulation weight and volume are important. Their low thermal conductivity makes them attractive for terrestrial, deep-sea and space applications. They are especially suited for space applications where system weight is of paramount importance.

A useful thermal insulation must impede heat transfer by solid conduction, gas conduction, and radiation. It must also possess chemical, physical, and mechanical stability. However, both the mode of heat transfer and the properties of insulation materials are dependent on temperature. For example, radiant heat transfer becomes increasingly important and the emissivity, vapor pressure, and reactivity of materials rise as temperature goes up. No single combination, therefore, presents the optimum thermal barrier in more than a limited temperature range. Thus, each material under consideration must be thoroughly evaluated for the conditions under which the system is intended to be used.

The principal parts of a multi-foil insulation system are the vacuum envelope, mechanical support members, reflective foils, low conductance spacers and getters for vacuum maintenance. The insulation itself consists of reflective foils and fibrous spacers which are usually located in a vacuum envelope.

Oriented, highly reflective foils are used to minimize the flow of radiant energy. These foils are constructed from aluminum in moderate temperature regions of the insulation system; and copper, nickel, and other high melting point metals in high-temperature regions. The shields are as thin as is mechanically practical, to permit minimum weight and flexible contour fitting of the finished insulation.

Fibrous spacers of glass paper at moderate temperature levels, or quartz paper at higher temperature levels, separate the foils. These papers are constructed of small fibers to increase the number of interface resistance points and thus increase the effective length of conduction paths through the fibers. In addition, small fibers produce small voids to minimize residual gaseous conduction.

Linde has also developed a glass fiber paper which contains a uniform dispersion of minute metallic flakes. This material is called opacified paper insulation. It has the unique characteristic of a low thermal conductivity in both the normal and lateral direction. It can therefore be applied over difficult geometries several layers at a time without an adverse effect on performance. Opacified paper contains aluminum flakes for moderate temperature levels and copper, nickel or other flakes for higher temperatures.

For any one application, a composite of the above shields and separators may be employed at different operating temperature levels in order to gain optimum performance at a minimum of cost.

This study contract was performed in order to further advance the state of the art, to evaluate, and demonstrate the applicability of multi-layer

insulations for engineering application to radioisotope powered heat sources and thermo-electric conversion devices. In order to meet the above objective the following properties, characteristics and system components were studied. Insulation material selections were made and their operating temperature limits defined. Thermal conductivity measurements of selected combinations of foils and spacers were made. Processing and offgassing experiments were made to determine the long term vacuum problems which might exist and to determine gettering requirements for sealed systems. For non-sealed systems, such as aerospace applications, the effects of air exposure (ground hold) on the subsequent performance in vacuum (space) were studied analytically and experimentally. In order to assist practical application an analytical study on the thermal effect of gaps and exposed edges of insulation segments was performed. These results will be experimentally verified in Phase II of this contract. The physical integrity of these material combinations was evaluated by observing their resistance to vibrations typical of many applications and by the experience gained in the repetitive preparation, use and application of these materials to the various test devices during the course of property measurements. Material compatibility tests and observations were performed in conjunction with other tests to determine if any long term problems might occur. Finally, an investigation into the requirements of the vacuum enclosure for sealed systems was made, materials of construction specified and physical testing begun.

The work on this contract was performed under the direction of the United States Atomic Energy Commission, Isotopic Power Branch, Division of Space Nuclear Systems. The AEC technical monitor was Mr. D. L. Krenz, Sandia Corporation and the Linde Project Manager was Mr. W. D. DeWitt. Other Linde personnel contributing to this program were Messrs, N. C. Gibbon, R. L. Reid and D. J. Webster.

Additional assistance was rendered by Mr. C. M. Heath, and Messrs. L. D. Potts and D. Suckow.

3.0 TECHNICAL REPORT

3.1 Metal Foil Materials Study

Foil vaporization rate within an insulation system is an important factor in selecting materials for insulation system radiation shields, especially for systems requiring a life of one to five years. Although foil life can be increased with an increase in foil thickness, there is, of course, a practical thickness limit posed by fabricability requirements. In addition, increased thickness would result in greater system weight. Candidate materials initially selected for study were aluminum, copper, and nickel foils with the possible use of refractories or precious metals at high temperatures, 1600 - 1800°F.

This work first involved a literature survey to determine physical properties, notably vapor pressure, of candidate foil materials. Knowledge of these properties was then used in a design study in which parameters such as mission life, foil thickness and operating temperature were investigated.

The vaporization rate of a metallic foil can be calculated by assuming free vaporization of the foil in a vacuum, but this relationship does not account for the retardation of foil vaporization by the insulation system environment itself. The effect of the close proximity of spacers and other foils on vaporization rate must be considered.

3.1.1 Foil Vaporization in an Insulation System

When the vaporization rate of a foil within an insulation system is analyzed, two types of vapor flow in the insulation system can be considered. The vapor may flow:

1. Perpendicular to the foils through and into the spacers from the hotter to the cooler foils, and
2. Parallel to the foils to areas in the insulation having a lower partial pressure of the metallic vapor.

The vapor flow perpendicular to the foils is a complicated process composed of several mechanisms and phases. In order to calculate for the net loss of the hottest foil, we can consider two models. One of these models yields a vaporization rate that is believed to be high and the other model gives a rate which is believed to be low. Since the answers from the two models are different by only about 40°F. for a five year maximum operating temperature, the solution is suitably bracketed.

For the first model, consider the two hottest foils of the insulation section. If we neglect entirely the effect of the spacer, the net vaporization rate will be the difference in the free vaporization rates of the two foils.* That is, let w_1 be the vaporization rate of foil 1 at T_1 if this foil

* Assuming material leaving one foil is deposited on the other in such a manner that it will identically restore the structure in the form prior to vaporization

were isolated in an infinite vacuum. Also, let w_2 be the vaporization rate of foil 2 at T_2 under the same conditions. Then the net vaporization rate, m_I , is

$$m_I = w_1 - w_2 \quad (1)$$

For nickel, the free vaporization rate is given by:

$$\log w = 9.40 - 0.5 \log T - \frac{20960}{T} \quad (2)$$

where

w = vaporization rate, $\text{gm/cm}^2 \text{ sec.}$
 T = temperature, $^{\circ}\text{K.}$

The equation for copper is:

$$\log w = 8.63 - 0.5 \log T - \frac{16980}{T} \quad (3)$$

(Aluminum need not be considered in this and following development since its vapor pressure and vaporization rate are very low at all temperatures below its melting point, 1220°F). Equation (1) with (2) and (3) thus give one model for the vaporization analysis. (Designated Model I.) Maximum temperatures for five year life as a function of $(T_1 - T_2)$ are given in Figures 1 and 2 for 0.5 mil copper and nickel foils.

For any desired life, the total thickness evaporated is given by:

$$Q = \frac{m_I \theta}{(2.54 \times 10^{-3}) \rho} \quad (4)$$

where

Q = thickness, mil
 m_I = vaporization rate calculated from (1) and (2) or (3)
 θ = time, sec.
 ρ = density of metal foil, gm/cm^3

Solving (4) for the foil life for any thickness and temperatures, we obtain:

$$\theta = \frac{(2.54 \times 10^{-3}) \rho Q}{m_I} \quad (5)$$

Another model can be considered that does include some effects of the spacer. When the system reaches operating temperature, the initial rate of vaporization will be high since the fibers will tend to adsorb a thin layer of atoms at a high, but difficult to calculate, rate. When the adsorption energy has been exhausted, the fibers will then act as minute foils and experience a net gain of metal atoms since foil 1 is hotter than any of the fibers and thus will supply atoms faster than atoms can be vaporized from the lower temperature fibers. However, in this model, (Model II), this effect will be neglected and the fibers will be assumed inactive after the initial adsorption

energy is exhausted.

Since the adsorption rate is unknown, this process will be assumed to occur in zero time. In order to put on upper bound on the thickness of the adsorption layer on the glass fiber, we can first draw on a crude analogy of the adsorption of water vapor on glass. Brunauer reports that glass generally adsorbs about 6 or 7 monolayers of water but sometimes as high as 40 monolayers. However, to be conservative we will assume that 350 monolayers of metal (3.4 micro-inches) will be adsorbed by the glass fibers (assumes 2.5 angstroms per layer). It then remains to calculate the actual surface area of the fiber spacer to determine the thickness of metal corresponding to the 3.4 micro-inch coating on each fiber.

After the metal thickness to coat the fibers has been sacrificed, the vapor flow will be assumed to occur as dictated by the temperature differences between two foils and the conductance of the spacer. That is, once the fibers have been coated, they are assumed to be inactive with all vaporization and condensation occurring at the foils.

In this analysis Refrasil quartz fiber paper was assumed to be the spacer.

If we consider the fibers as cylinder, the ratios of the actual fiber surface area to sheet surface area is given by:

$$S = \frac{4\alpha}{d_f \rho_G} \quad (6)$$

where

$$\begin{aligned} \rho_G &= \text{density of solid quartz, gm/cm}^3 \\ d_f &= \text{fiber diameter, cm.} \\ \alpha &= \text{sheet weight, gm/cm}^2 \end{aligned}$$

For quartz paper, we have:

$$\begin{aligned} \rho_G &= 2.60 \text{ gm/cm}^3 \\ d_f &= 0.000127 \text{ cm.} \\ \alpha &= 2.56 \times 10^{-3} \text{ gm/cm}^2 \end{aligned}$$

Substituting in (6), we find

$$S = 29.2$$

Therefore, even though the porosity of the paper is well over 90%, the actual internal surface area is 29.2 times the measured surface area. Consequently, the thickness of metal lost from the radiation shield required to coat the fibers to a thickness of 3.4 micro-inches is approximately 0.1 mil. Even though it would take a considerable amount of time for this vapor deposition to occur; to be conservative, this process will be assumed to take place instantaneously.

As stated earlier, after the adsorption layer forms, the net flow of metal from the hottest foil will be assumed controlled by the vapor pressures of the first and second foils and the hydrodynamic conductance of the spacer. We will assume that the external side of the number one foil does not evaporate (e.g., it might be wrapped tightly on an impervious surface). The point of interest then is the maximum temperature of operation whereby the hottest 0.5 mil thick copper or nickel foil will disappear. Actually, all interior foils experience a net weight gain during the time required for vaporization of the first foil. Therefore, at the time the first foil has completely vaporized, the interior foils probably have not been affected significantly by the vaporization process. The maximum use temperatures generated by this model (and also Model I) are only strictly valid for the hottest foil of a subsection and are conservative for all other foils of the subsection.

In order to calculate the perpendicular flow rate through the metal coated fiber spacer, the temperatures (and hence the vapor pressures) of adjacent foils at the hot end must be known. The temperature difference between foils at the hot end is generally small due to the non-linear nature of the temperature profile. The temperatures of the first and second foils for an actual system could easily be calculated after the insulation material composition and boundary temperatures of that system have been determined. The temperature difference between two foils is not expected to exceed 10°F for most systems.

After the temperatures are determined, the corresponding vapor pressures can be calculated by the general relation in (7) taken from Dushman.

$$\log P = A - \frac{B}{T} \quad (7)$$

where P is in microns and T in °K. For nickel, A = 12.75 and B = 20,960. For copper, A = 11.96 and B = 16900. For aluminum, A = 11.79 and B = 15,940. The mass flow rate through the insulation can then be calculated from (8)

$$m_{II} = \frac{S (P_H - P_2) M}{122.8 D R T_H} \quad (8)$$

where:

- S = conductance of spacer, CFM-ft./ft.²
- D = distance between adjacent foils, ft.
- P_H = vapor pressure at hottest foil, mm Hg.
- P₂ = vapor pressure at second foil, mm Hg.
- M = molecular weight of metal, gm/g mole
- T_H = hot side temperature, °R
- R = universal gas constant, 554.9 mm.Hg.ft.³/lb. mole °R
- m_{II} = mass flow rate, gm/cm.² - sec.

Equations (7) and (8) thus give a means of calculating the mass flow rate through the spacer assuming that the fibers are coated with metal so

that adsorption has ceased.

As an example of the previous calculations, consider copper at 1400°F. at a layer density of 100 layers/inch. For a 9°F. temperature difference ($T_2 = 1391^\circ\text{F.}$), from (7), we find $P_H = 3.37 \times 10^{-8}$ and $P_2 = 2.804 \times 10^{-8}$. From best available Linde in-house data, the conductance of glass spacer in both the perpendicular and longitudinal direction is approximately 10.0 CFM ft./ft.². With $D = 7.92 \times 10^{-4}$ ft., substituting in equation (8) yields a mass flow rate of 3.60×10^{-11} gm/cm² sec. The maximum use temperatures generated by this model (Model II) are given in Figures 1 and 2.

The flow of vapor parallel to the foils can be calculated in a similar manner. The mass flow rate per square centimeter of foil area is given by (9)

$$m_L = \frac{F P_H D M}{122.8 L^2 R T_H} \quad (9)$$

where:

- F = longitudinal conductance, CFM-ft./ft.²
- L = length of path to region of low metal vapor pressure, ft.
- m_L = longitudinal mass flow rate, gm/cm² - sec.

In (9), the vapor pressure in the "surroundings", L distance from the point of interest, was taken as zero to obtain the maximum rate.² In the case of the copper at 1400°F., using a value of $F = 10.0$ CFM-ft./ft.² and a path length of one foot, (9) gives an m_L of 1.34×10^{-16} gm/cm² sec. which is a factor of 3×10^5 less than the rate through the spacer. If we let $m_L = m_{II}$, we can solve for the path length where the two flows would be equal. With $m_L = m_{II} = 3.60 \times 10^{-11}$, L is calculated to be 0.0232 inches. Therefore, except within 0.0232 inch of the edge of an insulation panel or material discontinuity, the vaporization will be controlled by the perpendicular flow through the spacer and the vaporization rate will be the rate calculated by the perpendicular flow rate.

With the vaporization rate for Model II given by (8) and with the fact that about 0.1 mil of foil will be required to coat the fibers, the total thickness of foil evaporated at any time can be obtained from (10).

$$Q = 0.1 + \frac{m_{II} \theta}{(2.54 \times 10^{-3}) \rho} \quad (10)$$

where:

- m_{II} = vaporization rate calculated from (8), gm/cm² sec.

Conversely, Q can be taken as the foil thickness and θ , the foil life, can be calculated from (11)

$$\theta = \frac{(2.54 \times 10^{-3})}{m_{II}} (Q - 0.1) \quad (11)$$

In examining the validity of Models I and II, we can make the following observations:

1. Model I gives too high a vaporization rate since the fiber spacer will develop a coating that will be at a higher temperature than foil 2 and thus contribute a higher back pressure than using the temperature of foil 2. Also the rate is too high since the blockage by the spacer has been neglected.
2. Model II gives too low a vaporization rate since the continual buildup of metal on the fiber has been neglected. This buildup provides effective "sinks" for metal atoms which would give a higher rate than the calculated from (6).

Thus, to be conservative, it is recommended that the Model I curves on Figures 1 and 2 be used to determine the maximum operating temperature of an insulation system.

Note further that all of the previous development has been important based upon certain assumptions. The equation is really only valid for the first foil of a system where the first foil is in intimate contact with the hot vessel being insulated. That is, vaporization only occurs from one side of the foil. It should also be noted that all foils except the first actually have a net gain of metal until the first foil has vaporized. Therefore, the foil life calculated for a given foil thickness is only for the first foil so that the thermal performance would probably be unaffected until some later time.

Over a long period of time, condensation of the vaporized metal on spacer fibers could result in higher solid conduction. Quantitative data in this respect is not yet available. It is believed, however, that for 5 - 10 year periods, these effects will be both small and concentrated to the hottest layers making their effect on overall performance small.

3.1.1.1 Foil Life

For use in multi-layer insulation systems foil thickness on the order of .25 - .50 mil have been found to be practical from the standpoint of handleability and have still provided a reasonably high layer density, 100 - 140 layers per inch. These thin foils also result in low insulation system weight since foil weight constitutes the major portion of insulation system weight. This is an important consideration when considering space applications where system weight is of paramount importance.

From Figure 1 using the conservative Model I, we see that the upper temperature limit of 0.5 mil copper foil for a five year life could therefore vary between about 1390°F. - 1470°F. based on the bracketing conditions

TEMPERATURE LIMIT FOR
0.5 MIL COPPER VS. ADJACENT
FOIL TEMPERATURE DIFFERENCE

FIGURE 1

FIGURE 2

of an adjacent foil temperature difference of $2^\circ - 10^\circ$.

The temperature difference between adjacent foils in an actual insulation system depends on the insulation thickness. Systems having small thicknesses of insulation have steep temperature gradients and hence relatively large temperature differences between adjacent foils.

Similar calculations for 0.5 mil nickel foil resulted in Figure 2. Figure 2 shows that 0.5 mil nickel could be used at 1800°F . for a 1 - 5 year design life based on the assumption that a 3°F . temperature difference existed between adjacent foils at the hot boundary. As mentioned earlier, calculations on aluminum revealed that from the standpoint of vaporization a 0.25 mil foil can be used at all temperatures below its melting point (1220°F .) for a five year life. Experience has shown that aluminum foil tends to soften when used above 1000°F . The spacer material used with aluminum foil, glass fiber paper, also tends to soften and melt when subjected to temperatures of approximately $900 - 1000^\circ\text{F}$. An upper operational temperature of 900°F . is, therefore, recommended for aluminum.

The following radiation shield foils and temperature ranges are recommended for an insulation system with a design life of one to five years:

Aluminum foil	- 0.00025 inch thick	$100^\circ\text{F} - 900^\circ\text{F}$
Copper foil	- 0.0005 inch thick	$900^\circ\text{F} - (1390^\circ - 1470^\circ\text{F})$
Nickel foil	- 0.0005 inch thick	$(1390^\circ - 1470^\circ\text{F}) - (1730^\circ - 1830^\circ\text{F})$

3.1.2 Free Vaporization of Foils in Vacuum

Based on the Langmuir⁴⁵ expression an equation was derived to calculate the foil life that would be expected without the vaporization retarding effect of the insulation system.

A derivation of the foil life equation is as follows:

If	L_s	- foil life in seconds
	M	- foil material molecular weight
	P	- foil vapor pressure at T_k in mm Hg
	T_k	- foil temperature in degrees Kelvin
	t_c	- foil thickness in centimeters
	W	- vaporization rate per unit area for the foil material in grams/cm ² sec.
	ρ	- foil density in grams/cm ³
	w	- foil weight per unit area in grams/cm ²

We can see that:

$$L_s = \frac{w}{2 * W} \quad (12)$$

* Assumes that vaporization occurs from both sides of the foil.

However,

$$\omega = t_c \rho \quad (13)$$

and from the Langmuir equation,

$$W = .0585P \sqrt{\frac{M}{T_k}} \quad (14)$$

Thus it follows that

$$L_s = \frac{t_c \rho}{2} \frac{1}{.0585P} \sqrt{\frac{T_k}{M}} \quad (15)$$

and changing units for L, T and t,

$$\therefore L_d = 1.87 \times 10^{-4} \frac{t_i \rho}{P} \sqrt{\frac{T_r}{M}} \quad (16)$$

where:

L_d = foil life in days
 T_r = foil temperature in degrees Rankine
 t_i = foil thickness in inches

Based on equation (16) for 0.5 mil foil and a five year life, upper temperature limits of 1270°F. and 1640°F. were established for copper and nickel foils, respectively. It is now believed that these temperature limitations set by the foil life insulation system study (Section 3.1.1.1) are more realistic.

Chronologically the temperature limitations from this "free" vaporization study were completed first and used as guides for establishing the operating conditions of the thermal conductivity and offgassing tasks (Section 3.3.1.5 and 3.4.2). Based on these conservative temperature limitations a study was initiated to select a "backup" material for nickel in the temperature regime of 1640 - 1800°F.

In order to select a backup for nickel foil at temperatures above 1640°F., a survey of the better known and most extensively used refractory and precious metals was made. The survey as presented in Table I consisted of the tabulation of available physical properties in order to select a material that could be readily procured so as not to substantially delay the project schedule.

Of the precious metals, gold and silver show extremely low emissivities but are excluded from consideration because of their low melting points.

TABLE I
HIGH TEMPERATURE METALS

Candidate	Melting Point, C°	Total Emissivity at 1800°F.	Foil Availability	Vapor Pressure	
				F°	mm Hg
Refractory Metals:					
Tungsten	3370	0.150	5.0 mil (sheet)	2240	10 ⁻¹¹
Tantalum	2850	0.160	0.3 to 4.0 mil thick 12 in. wide, coil length	3020	10 ⁻¹¹
Molybdenum	2620	0.166	0.3 to 4.0 mil thick 12 in. wide, coil length	2440	10 ⁻¹¹
Columbium	2415	0.150	0.3 to 4.0 mil thick 12 in. wide, coil length	2720	10 ⁻¹¹
Precious Metals:					
Platinum	1755	0.140	0.1 to 1.0 mil thick 12 in. wide, 20 in. long	1950	10 ⁻¹¹
Palladium	1555	0.120	0.1 to 1.0 mil thick 12 in. wide, 20 in. long	1815	10 ⁻⁶
Rhodium	1955	0.05	1.5 mil thick, 8 in. x 16 in.	3560	10 ⁻³
Ruthenium	1950		No		
Iridium	2350		1.5 mil, 8 in. x 16 in.		
Osmium	2700		No		
Gold	1060	0.030	Yes		
Silver	960	0.050	Yes		

Ruthenium and osmium are not available as foils. Platinum, palladium, iridium and rhodium are available as foils but not in continuous coil lengths. Also, the vapor pressure of palladium is relatively high, 10^{-6} mm Hg at 1815°F. , indicating a slightly higher vaporization rate than nickel and, therefore, not acceptable.

Tungsten is not available as a thin foil.

Columbium, tantalum, and molybdenum are available as foils and they possess similar vapor pressures, emissivities and melting points.

Cost of 0.5 mil thick, 12 inch wide foils are as follows:

Tantalum	-	\$70/lb.
Columbium		\$200/lb.
Molybdenum	-	\$250/lb.

From a mechanical viewpoint, the ease of working of these metals is, in order: tantalum, columbium and molybdenum. Tantalum and columbium can be worked at room temperature while molybdenum requires heating. Molybdenum oxidizes rapidly above 1300°F.

Tantalum and columbium possess similar characteristics with the major difference being in density and nuclear cross-section. Density of tantalum is almost twice that of columbium, and the nuclear cross-section of tantalum is nearly 20 times that of columbium.

Based on its vapor pressure, emissivity, cost, ease of working and availability, tantalum was selected as a backup radiation shield foil for nickel. Tantalum was later rejected because of incompatibility with Astro-quartz Cloth spacer and trace contaminants in the vacuum space, Section 3.5.1.3.1.

As Figure 2 shows, a backup is probably not needed for nickel since nickel is good to at least 1730°F. and one or two extra foils can be added to allow for vaporization. (Or possibly one mil foil could be used for the first few layers.)

3.2 Insulation Spacer Selection

The scope of this task was to survey potential insulation spacer materials within the following types and select promising candidates for further investigation.

Three basic types of spacers were considered:

- (1) glass or quartz fiber paper
- (2) woven quartz cloth
- (3) metal flake opacified glass or quartz fiber paper.
This type of spacer consists of a fibrous paper into which metal flakes, on the order of -325 mesh, have been introduced during fabrication.

Vendor contacts were initiated and a survey of the literature yielded appropriate data for a variety of spacer material candidates. The survey included both papers and fabrics produced from silica fibers.

Some selected physical properties for the various products are presented in Table II. These are: thicknesses, temperature limits, breaking strength, specific gravities; and, when available, heat capacities, thermal conductivities, and areal shrinkage. Breaking strengths are given for two directions of the fabrics (warp and fill) and two directions of the papers (machine and cross).

The chemical composition (on a binder-free basis) of the various products is presented in Table III. The primary component is SiO_2 , and the chief impurities are metal oxides. Various binders are used by manufacturers and generally range from one to three percent of the compositions. Dexter and Sons, Inc. uses Refrasil fiber by H. I. Thompson Co. for its Refrasil paper, as well as the copper opacified quartz fiber paper. However, the aluminum opacified glass papers are made with a glass fiber and aluminum flake.

The fabrics differ from each other in the purity of the silica used for the yarn. For example, J. P. Stevens and Co. produces a high purity silica yarn (Astroquartz) in addition to a lower purity product (Astrosil), and this seems similar to the H. I. Thompson Refrasil fabric. The H. I. Thompson Irish Refrasil is made from a yarn containing one to three percent Cr_2O_3 and a minimum of 96 percent SiO_2 .

Most of the fabrics and papers have an organic binder that may be removed or stabilized by heat treating in air. Vendor recommended temperatures vary from 500 to 1000°F.

Sneed and Brasted⁷¹ report that silica (SiO_2) has at least eleven (and possibly as many as fifteen) distinct crystalline and vitreous forms. Three of these forms (the α quartz, α tridymite, and α cristobalite) are stable at room temperatures; but at elevated temperatures undergo transitions to β forms which are normally metastable at room temperatures.

TABLE II

Physical Properties of Candidate Materials

Company and Trade Name	Thick- ness (mils)	Temp. Limits (°F.)			Breaking Strength		C _p Btu lb. °F.	k Btu-in. (hr. ft ² °F.)	Specific Gravity	Areal Shrinkage After Exposure at 1800°F. (%)
		Manufacturer's Recommendation	Strain Point	Melt Point	Warp (lb./in.)	Fill (lb./in.)				
<u>Dexter and Sons, Inc.</u>										
Refrasil Paper	5				.308*	.132**			.190	
1.1 g/ft ² Alum. Opac. Glass Paper	18				.660*	.154**			.270	
2.5 g/ft ² Alum. Opac. Glass Paper	35				1.101*	.440**			.290	
5.5 g/ft ² Copper Opac. Quartz Paper	15									
106 Glass Fiber Paper	2.8				.442*	.165**			.190	
<u>J. P. Stevens and Co., Inc.</u>										
Astroquartz 503	5.3		1950	3000			.165		.831	
594	8.5		1950	3000					.37	
581	12		1950	3000	185	170			.901	
527	8		1950	3000	140	110			1.010	
533	7		1950	3000	125	125			.961	
570	27		1950	3000	500	320			1.015	
593	10		1950	3000	150	145			.964	
Astrosil 64	15			3000	28	25			.889	5
82	13			3000	28	25			1.057	5
84	26			3000	55	50			.948	5
88	52			3000	100	75				5
75	42			3000	75	50				5
<u>H. I. Thompson Co.</u>										
Refrasil UCC 100-46	15			3000	20	15			.895	
100-28	13			3000	33	30			1.033	
100-38	18			3000	35	30			.960	
Irish Refrasil C1554-28	14	2600						.65-600°F.	.896	
C1554-48	28	2600			30	20		1.10-1800°F.	1.049	
Refrasil Open Weave SS1452	17	2000			20	15			.679	8

* Manufacturer's Machine Direction

** Manufacturer's Cross Direction

[illegible]

The crystalline form of silica which is present in the yarn seems to be an important factor influencing the breaking strength of the fabric. However, the only manufacturer to give information on the crystalline structure of its fiber is J. P. Stevens. For their product, "Astroquartz", they say that at:

1900°F. - Devitrification into the crystalline form known as cristobalite takes place. This stiffens the material but seems to have no other effect on the physical properties.

2370°F. - A melting into a pasty form occurs.

Also the melting point is given as 3000°F. and vaporization is said to occur at 3600°F.

Some manufacturers list strain points and operating limits, and these would correspond to the Astroquartz transitions at 1900°F. and 2370°F., respectively. The crystalline phase transitions seem to be responsible for the brittleness observed in fabrics that have been heat treated at high temperatures. However, the loss of the binder following heat treatment may also contribute significantly to the brittleness.

All of the products, with the exception of H. I. Thompson's "Irish Refrasil", seem to have similar temperature characteristics. J. P. Stevens and Co. has supplied additional temperature data for the breaking strength of their Astroquartz No. 581 and Astrosil No. 82. This is presented in Table IV. The phase transition for Astroquartz seems to be responsible for the zero breaking strength reported at 2000°F. Also the sharp decrease in the Astroquartz breaking strength between 400°F. and 800°F. may possibly indicate yet another phase.

Astrosil exhibits a somewhat different behavior, yet the breaking strength does decrease sharply, though still maintaining some strength, near 2000°F.

Astroquartz fabric would be an ideal spacer material since it can be produced as thin as 5.3 mils. Although suffering a loss of tensile strength after high temperature exposure, it still retains sufficient strength after processing to be fabricable, Section 3.5.1.

Breaking strength properties of the type presented for Astroquartz and Astrosil are not available for H. I. Thompson products; but, since their products appear similar to the Astrosil, they can probably be expected to behave as Astrosil does. "Irish" Refrasil is somewhat unique in that it is rated for use up to a temperature of 2600°F. It has the additional advantage of having a lower thermal conductivity than that reported for Refrasil.

After consideration of the data in Tables II, III, and IV, the following vendors and materials were selected as the most promising:

H. I. Thompson Co.

Refrasil C100-28

Irish Refrasil C1554-28

TABLE IV

EFFECTS OF TEMPERATURE ON BREAKING STRENGTH*

<u>Conditions</u>	<u>BREAKING STRENGTH, LB/IN.</u>			
	<u>Astroquartz No. 581</u>		<u>Astrosil No. 82</u>	
	<u>Warp</u>	<u>Fill</u>	<u>Warp</u>	<u>Fill</u>
Original fabric (room temperature)	200	254	120	72
After 24 hr @ 400°F	230	276	124	64
After 24 hr @ 800°F	64	66	97	65
After 24 hr @ 1200°F	42	51	110	72
After 24 hr @ 1600°F	25	33	73	53
After 24 hr @ 2000°F	0	0	28	6

 *Data supplied by J. P. Stevens Co.

J. P. Stevens & Co., Inc.

Astroquartz 503
Astrosil 82

Dexter and Sons, Inc.

Refrasil Paper
No. 106 Glass Fiber Paper
1.1 gm/ft.² alum. opac. glass
fiber paper
2.2 gm/ft.² alum. opac. glass
fiber paper
5.5 gm/ft.² copper opacified
quartz fiber paper

Samples of these spacers were obtained and submitted, in the "as received" condition, to the analytical laboratory to be analyzed for a residual carbon content.

These residual carbon contents in Table V, were used as criterion to evaluate the effectiveness of the spacer processing techniques developed in the processing task, Section 3.4.1, in the removal of carbonaceous materials.

TABLE VPRELIMINARY SPACER CANDIDATESRESIDUAL CARBON CONTENT (AS RECEIVED CONDITION)

<u>Spacer Material</u>	<u>% Carbon</u>
Astroquartz Cloth No. 503	0.24 ± .01
Astrosil Cloth No. 82	0.11 ± .01
Refrasil Cloth No. C100-28	0.15 ± .01
Irish Refrasil Cloth No. C1554-28	0.12 ± .01
Dexter Refrasil Paper	2.2 ± .01
Dexter 106 Glass Paper	0.1 ± .1
Dexter 1.1 gm/ft. ² Al Opacified Glass Paper	1.0 ± .1
Dexter 2.2 gm/ft. ² Al Opacified Glass Paper	1.1 ± .1
Dexter 5.5 gm/ft. ² Cu Opacified Refrasil Paper	0.84 ± .1

Final selection of the candidate spacers was based on:

- (1) ability of the candidate to undergo thermochemical processing for removal of carbonaceous binders, Section 3.4.1.
- (2) sufficient strength and physical stability for insulation system fabrication after processing, Section 3.5.1.
- (3) fabricability of processed spacers, on thermal conductivity test apparatus, Section 3.3.1.

- (4) acceptable offgassing characteristics after processing, Section 3.4.2.
- (5) resistance of the processed spacers to a typical vibrational environment, Section 3.5.2.

Another characteristic of each spacer that must be considered is its thickness. Minimum thickness is desirable for the spacers in order to provide as high a layer density of radiation shields as possible.

The candidate spacers selected were:

- (1) Astroquartz Cloth No. 503
- (2) Refrasil Quartz Fiber Paper
- (3) Glass Fiber Paper
- (4) Aluminum flake opacified glass fiber paper, 1.1 g/ft.²
- (5) Copper flake opacified Refrasil Quartz fiber paper

Astrosil Cloth, Refrasil Cloth, Irish Refrasil Cloth and Dexter 2.2 g/ft.² aluminum opacified glass paper were satisfactory from strength and processing viewpoints but are thicker than the selected candidates.

3.3 Insulation Thermal Performance

3.3.1 Thermal Conductivity in Vacuum

The aims of the thermal conductivity tests were to determine the thermal performance of the insulation materials in an installed configuration as a function of temperature and to determine any tendency of the installed materials to undergo thermal deterioration as a result of long-term exposure to temperature and vacuum.

3.3.1.1 Multiple Layer Insulation Thermal Analysis

A multiple layer insulating system is comprised of three essential elements; (1) highly reflective shields, (2) low conductance fillers to separate the shields and (3) vacuum. In such an insulation, heat is transported by gaseous conduction, solid (or filler) conduction, and radiation. Although the three modes are, strictly speaking, not separable, it was shown by Wang⁷⁸ that for the systems considered in this study, treating the total heat transported as a sum of three independent contributions is a reasonable approximation. A thermal conductivity expression of the form derived by Wang has been used to obtain an empirical correlation of the experimental data. Wang's derivation is reproduced here for convenience.

$$q_o = q_f + q_g + q_r \quad (17)$$

where:

- q_o = overall heat transfer rate, Btu/hr.
- q_f = filler solid conduction rate, Btu/hr.
- q_g = gaseous conduction rate, Btu/hr.
- q_r = radiation heat transfer rate, Btu/hr.

Assuming the insulating space to be composed of a multiplicity of reflective shields separated by low conductance fillers, then the temperature change ΔT across any two neighboring shields is small compared to the absolute temperature, T . Let us proceed to consider these three contributing modes of heat transfer individually between two neighboring shields.

The heat transfer rate of the filler due to solid conduction may be defined as:

$$q_f = k_f A \frac{\Delta T}{\Delta X} \quad (18)$$

where:

- k_f = equivalent filler thermal conductivity at T , Btu/hr-ft-°R
- A = heat transfer area, ft.²
- ΔT = temperature difference, °R
- ΔX = distance between radiation shields, ft.

k_f , it should be noted, represents the effective conductivity of a multitude of fibers or particles and, although its magnitude cannot be directly calculated from the solid conductivity of the filler material alone, it could be assumed that k_f is proportional to the conductivity of the parent filler material, thus

$$k_f = c k_{pf} \quad (19)$$

where k_{pf} is the conductivity of the parent filler material and, since the thermal conductivity of the latter may usually be expressed as a simple power function of temperature, it follows then

$$k_f = a T^d \quad (20)$$

where:

a = an empirical constant
 d = an exponent

The exponent, d , is that which is obtained for the parent filler material and lies usually within the limits $.25 < d < 1$. As a first approximation $d = 1$ has been used.

The constant, a , must be sought empirically since for a given filler material, a , will vary with bulk density, fiber or particle size and shape, etc.

Substituting in equation (18):

$$q_f = a T A \frac{\Delta T}{\Delta X} \quad (21)$$

In insulating spaces filled with a fine particle or fibrous material, gas convection may be neglected. Gaseous conduction is, according to kinetic theory, a function of temperature only, provided that the distance between the intervening particles or fibers within which gaseous conduction is operative is larger, by an order of magnitude or more, than the mean free path of the gas molecules at the prevailing pressure and temperature. The interstitial space in a matrix of fine powders or fibers is usually so small that the above stipulation is not adhered to even at moderate vacuums; in such cases the heat transferred due to gaseous conduction may be, approximated by the following equation, due to Kistler:³⁹

$$q_g \approx k_g A \left(\frac{L}{L+1} \right) \frac{\Delta T}{\Delta X} \quad (22)$$

where k_g is the "classical" thermal conductivity of the free gas, Btu/hr-ft-°R.

L is the average distance which the gas molecule travels, before it collides with a solid (filler) structural member, microns.

l is the mean free path of the gas at the prevailing pressure and temperature, microns.

k_g as a first approximation may be expressed as a linear function of temperature, so that

$$q_g = k_g A \frac{T}{530} \left(\frac{L}{L+1} \right) \frac{\Delta T}{\Delta X} \quad (23)$$

where k_g is the conductivity of the free gas at 530°R, Btu/hr-ft-°R.

Substituting for l the expression:

$$l = \frac{l_o T}{530P} \quad (24)$$

where:

p is the absolute pressure in, say, microns of mercury.

l_o is the mean free path of the gas at l_o Hg pressure and 530°R, in microns.

Then substituting (24) in (23) we obtain:

$$q_g = \frac{k_g T}{530} \frac{Lp}{Lp + \frac{l_o T}{530}} \frac{A \Delta T}{\Delta X} \quad (25)$$

Furthermore, at moderate temperatures, low pressures and for fillers having interstitial spaces between structural members in the order of 100 μ or less,

$$Lp \ll \frac{l_o T}{530}, \text{ so that approximately}$$

$$q_g = \frac{k_g Lp}{l_o} \frac{A \Delta T}{\Delta X} \quad (26)$$

The rate of heat transfer due to radiation between two reflective surfaces for low values of emissivity is approximately:

$$q_r = \frac{\delta e A}{2} [(T + \Delta T)^4 - T^4] \quad (27)$$

where:

δ = Stefan-Boltzmann constant, Btu/hr-ft²-°R⁴
 e = emissivity of the reflective surfaces

The temperature difference between two adjacent radiation shields, ΔT , is typically very small compared to the source temperature, T . If $T \gg \Delta T$,

then expanding equation (27):

$$q_r \approx 2\delta\epsilon AT^3 \Delta T \quad (28)$$

Substituting equations (21), (26), and (28) in (17):

$$q_o = A \frac{\Delta T}{\Delta X} \left[aT + \frac{k-Lp}{l_o} + 2\delta\epsilon\Delta XT^3 \right] \quad (29)$$

Overall heat transfer rate, q_o , may also be defined in terms of an overall thermal conductivity, k_o .

$$q_o = k_o A \frac{\Delta T}{\Delta X} \quad (30)$$

and from (29)

$$k_o = aT + \frac{k-Lp}{l_o} + 2\delta\epsilon\Delta XT^3 \quad (31)$$

The total number of insulation compartments bounded by radiation shields may be defined as N , then defining n as the number of compartments per unit thickness, ΔX :

$$N = n \Delta X \quad (32)$$

For $N = 1$

$$n = \frac{1}{\Delta X} \quad (33)$$

Substituting in equation (31), an equation for the point conductivity of multiple layer insulation is obtained:

$$k_o = aT + \frac{k-Lp}{l_o} + \frac{2\delta\epsilon T^3}{n} \quad (34)$$

The overall thermal conductivity is a point value and in differential form:

$$q_o = k_o A \frac{dT}{dX} \quad (35)$$

Rearranging:

$$\frac{q_o}{A} \int_0^x dx = \int_{T_1}^{T_2} k_o dt \quad (36)$$

where the limits o and x and T_1 and T_2 mark the two extremities of the insulated space.

Integrating and substituting (34):

$$\frac{q_o}{A} \Delta X = \frac{a}{2} (T_1^2 - T_2^2) + \frac{k_g L_p}{l_o} (T_1 - T_2) + \frac{\delta \epsilon}{2n} (T_1^4 - T_2^4) \quad (37)$$

Defining an apparent, mean thermal conductivity, k_a , such that:

$$q_o = \frac{k_a A (T_1 - T_2)}{X} \quad (38)$$

and

$$\frac{q_o X}{A} = k_a (T_1 - T_2) \quad (39)$$

where

X = total insulation thickness

Then equations (39) and (37) are equal.

Solving equation (39) for k_a :

$$k_a = \frac{a}{2} \left(\frac{T_1^2 - T_2^2}{T_1 - T_2} \right) + \left(\frac{k_g L_p}{l_o} \right) + \frac{\delta \epsilon}{2n} \left(\frac{T_1^4 - T_2^4}{T_1 - T_2} \right) \quad (40)$$

The data obtained from the thermal conductivity testing, $k_a = f(T_1, T_2)$, was used in equation (40) to obtain knowledge of the constants, (a) and $(\frac{\delta \epsilon}{2n})$, or since ϵ is the only unknown in this term, the effective value of ϵ can be determined.

Equation (40) then can be used to predict thermal performance of an insulation between any boundary temperature within a given temperature range once the constants a and $\frac{\delta \epsilon}{2n}$ have been determined.

At the lower pressures, (< 0.1 micron mercury), at which the data were collected, the effect of gaseous conduction is negligible and

$$\frac{k_g L_p}{l_o} = 0$$

Letting $b = \frac{2\sigma\epsilon}{n}$, equation (40) may then be written as

$$k_a = \frac{a}{2} \left(\frac{T_1^2 - T_2^2}{T_1 - T_2} \right) + \frac{b}{4} \left(\frac{T_1^4 - T_2^4}{T_1 - T_2} \right) \quad (41)$$

Equation (41) is of the form:

$$y = mx + b \quad (42)$$

where:

y = dependent variable
m = slope
x = independent variable
b = intercept

Rearranging equation (41):

$$k_a \left(\frac{T_1^2 - T_2^2}{T_1 - T_2} \right) = \frac{a}{2} + \frac{b}{4} \left(\frac{T_1^4 - T_2^4}{T_1 - T_2} \right) \quad (43)$$

It should be recognized that many assumptions have led to the development of (41) that make the equation approximate. However, since the constants are determined from experimental data, the equation is useful in that a value from conductivity as predicted by the data can be regenerated from the equation much the same way as if an ordinary polynomial had been used. (41) is probably somewhat better than a polynomial for purposes of extrapolation or interpolation.

Plotting the experimentally determined values,

$$k_a \left(\frac{T_1^2 - T_2^2}{T_1 - T_2} \right) \text{ vs. } \left(\frac{T_1^4 - T_2^4}{T_1 - T_2} \right)$$

a straight line results, the slope of which is $b/4$ and the intercept is $a/2$. The constants, $a/2$ and $b/4$, can be grossly interpreted as solid conduction and radiation term coefficients for a given combination of insulation materials. As such, these coefficients for different multilayer insulations can be compared and an evaluation of the insulation made.

Table VI presents the pertinent data from the cylindrical tests and the $a/2$ and $b/4$ constants for (41).

Table 6

Summary of Thermal Conductivity Tests

Foil and Specification	Spacer and Specification	Layers	Layers/inch	Gas Pressure (μ Hg)	T _H (°F)	T _C (°F)	k $\left(\frac{\text{BTU } 10^{-3}}{\text{hr-ft-}^\circ\text{F}}\right)$	a/2 $\left(\frac{\text{BTU } 10^{-6}}{\text{hr-ft-}^\circ\text{R}^2}\right)$	b/4 $\left(\frac{\text{BTU } 10^{-12}}{\text{hr-ft-}^\circ\text{R}^4}\right)$	ϵ
Al Type AA-1145-0 Cold rolled, 1/4 mil thick unprocessed	No. 106 glass fiber paper, 3 mil thick, 1.7 gms/ft ² processed by air oxidation	60	120	.05 - .1	482	109.4	.174			
					779	143.6	.289			
					878	161.6	.381*	.053	.052	.086
Al Type AA-1145-0 Cold rolled, 1/4 mil thick unprocessed	Aluminum flake opacified glass fiber paper processed by air oxidation, 3 mil thick, 1.1 gm/ft ²	75	150		399.2	89.6	.085			
					734	104	.206			
					842	113	.270	.01	.066	.137
Cu-electrodeposited, 1/2 mil thick, processed by Linde Foil Cleaning Process	Refrasil quartz fiber paper, processed by air oxidation, 5 mil thick, 2.4 gms/ft ²	50	83.3	.045 - .1	720	158	.310			
					982	203	.440			
					1243	258	.630	.08	.053	.062
Cu-electrodeposited, 1/2 mil thick, processed by Linde Foil Cleaning Process	Copper flake opacified Refrasil quartz fiber paper 10 mil thick, 5.5 gms/ft ²	48	70.4	.07 - .1	635	185	.70			
					977	275	1.03			
					1265	347	1.33	.31	.061	.060
**Ni-type 200 rolled 0.62 mil thick, unprocessed	Astroquartz cloth, processed by air oxidation method, 5 mil thick, 10.4 gms/ft ²	60	120	.1 - .28	503	130	.66			
					850	212	1.32			
					1268	329	2.29			
					1625	450†	3.00			
					1947	550†	4.53	.31	.15	0.25
Ni-type 200 rolled 0.62 mil thick unprocessed	Refrasil quartz fiber paper, processed by air oxidation, 5 mil thick, 2.4 gms/ft ²	55	96.1		815	176	.66			
					1238	275	1.42			
					1616	338	2.48⊗	-.03	.18	0.24
Ta-99.9% pure 0.3 mil. thick, unprocessed	Astroquartz cloth, processed by air oxidation method, 5 mil thick, 10.4 gm/ft ²	42	140	.09-.10	617	136.4	.466			
					1005	212	.932			
					1351.5	285.8	1.853	.03	.159	.306

* Believed 13% high due to electrical leakage.

** Data for this test is believed about 25% high due to thermal shorting through the head.

† Extrapolated from preceding data due to cold side thermocouple failure.

⊗ Believed 10% high due to damage incurred in replacing defected hot side thermocouples.

3.3.1.2 Thermal Conductivity Test Apparatus

Since alternate layer insulations using metallic foils are strongly non-isotropic, thermal conductivity measurement of these materials should be performed in such a way that each metallic foil within the insulation composite is essentially an isotherm. Failure to maintain this condition will result in lateral heat flux through the insulation, causing significant errors in measurement. In order to measure the thermal conductivity of these materials and to simultaneously provide for realistic fabricability determinations and simulation of application conditions, the test apparatus as described by Figure 3 was designed and built.

On this device, the insulation material is installed around the central tube, shown schematically in Figure 4. This installation can be considered to be a fabricability determination and, if the candidate materials do not lend themselves to this installation, thermal conductivity study is hardly warranted. After installation of the insulation around the central tube, the unit is placed within an outer vacuum-tight shell, Figure 5, and connected to a suitable vacuum system. Measurement of the thermal conductivity of the installed insulation materials is determined by measuring the steady state power required to maintain the inner (hot) boundary of the insulation at a constant temperature with the outer insulation boundary at a lower constant temperature. Details of the test apparatus and instrumentation are presented in Appendix A. Figure 6 is a circuit schematic showing temperature and power measurement instrumentation. Figure 7 is a photograph showing the thermal conductivity test apparatus and instrumentation during actual testing. Figures 10 through 13 show various stages in the wrapping of a typical tester.

3.3.1.2.1 Heater Design and Fabrication

A detailed discussion of the heater selection activities is presented in Appendix A.

The heater selected for use in the thermal conductivity tester was the resistance wire type.

The heater consisted of a hollow alumina tube (re-crystallized pure alumina - 99% Al_2O_3) fitted with threaded grooves for supporting the resistance heating wire. Figure 8 shows from left to right: the alumina heater core wound with tungsten wire and end clips, the alumina heater sleeve to guard against electrical shorting, and the insulation tube. Assembly consisted of inserting the wire wound ceramic core into the ceramic sleeve, securing the end clips and inserting the ceramic assembly into the insulation tube.

For testing temperatures up to 900°F., a nickel-chromium alloy (Trophet A) heater wire was used. For testing temperatures from 900°F. to 1800°F. tungsten or a platinum - 10% rhodium alloy wire was used.

During high temperature thermal conductivity testing, an appreciable amount of the total heat input may be lost through the heater leads and thermocouple wires if they have not been carefully designed. An estimate can be obtained for these losses if it is assumed that the entire available temp-

FIGURE 3

INSULATION TEST SCHEMATIC

FIGURE 4

Figure 5: Thermal Conductivity Tester
Outer Vacuum Shell

THERMAL CONDUCTIVITY TEST
INSTRUMENTATION AND POWER SUPPLY CIRCUIT SCHEMATIC

FIGURE 6

Figure 7 Thermal Conductivity Apparatus and Instrumentation Setup

erature drop occurred over the insulation thickness. The heat loss by thermal leakage is then calculated from the conduction losses and, in the case of heater lead wires, the heat generated in the wire itself. The basis for the heat loss calculation is presented in Appendix A.

3.3.1.3 Test Procedure

After installing the insulation on the test core-heater assembly and obtaining dimensional measurements, the insulation assembly is placed within the vacuum chamber and evacuated. During this operation the vacuum chamber is leak checked by means of a helium mass spectrometer leak detector. Power input to the heater is carefully initiated and the hot face temperature level is increased to the first data point. The temperature controller is in use at this time to insure that the desired data point temperature is not exceeded. When steady-state conditions are established, constant temperature at constant power input, readings of hot and cold face temperature, power input and vacuum level are made. This procedure is repeated at increased temperatures resulting in thermal performance measurements at new sets of boundary temperatures (hot face and cold face temperatures).

3.3.1.4 Data Reduction

The data reduction procedure for determination of the insulation thermal conductivity involves:

1. calculation of an energy balance around the entire test system: $Q_{\text{insulation}} = Q_{\text{total power input}} - Q_{\text{losses}}$
2. solution of the basic Fourier heat transfer equation

Details of the data reduction derivation are presented in Appendix A.

The apparent thermal conductivity of the insulation is calculated from:

$$k_a = \frac{VI (3.414) - Q_{\text{losses}}}{S (T_1 - T_2)} \quad (44)$$

where:

k	= thermal conductivity, Btu/hr.ft.°R.
V^a	= voltage
I	= amperage
Q_{losses}	= instrumentation and heat lead thermal loss, Btu/hr.
S	= insulation shape factor, area/thickness, ft.
T_1	= hot face temperature, °R.
T_2	= cold face temperature, °R.

Figure 8 Coil Heater

3.3.1.5 Thermal Conductivity Tests In Vacuum

In Figure 9 the apparent thermal conductivity of each insulation combination tested is presented as a function of hot boundary temperature while the cold boundary is maintained at ambient, 70°F. During actual testing, cold boundary temperature did vary somewhat with hot boundary temperature. The curves of Figure 9 were constructed from actual measured data corrected to a constant low boundary temperature of 70°F in order to compare the tested combinations.

Inspection of Figure 9 shows that the anticipated low thermal conductivity of the multilayer insulation systems, 0.15 to 3.5×10^{-3} Btu/hr.ft.R° over the temperature range $T_{\text{hot}} = 400^{\circ}\text{F}$ - 1800°F at $T_{\text{cold}} = 70^{\circ}\text{F}$ was verified.

Aluminum foil with 106 glass fiber paper exhibited the lowest thermal conductivity at hot side temperatures up to the recommended maximum temperature of 900°F. Copper shield systems exhibited the next lowest conductivity over their recommended temperature range and should be used from 900°F to approximately 1500°F. For approximately 1500°F to 1800°F nickel foil is recommended.

Both Refrasil quartz fiber paper and Astroquartz cloth are recommended for usage between 900°F and 1800°F. Refrasil paper appears to display a lower thermal conductivity when installed in an insulation combination but is physically more fragile than Astroquartz Cloth. Both materials are recommended however, the selection dictated by the particular application or usage involved.

THERMAL CONDUCTIVITY OF THE INSULATION COMBINATIONS

FIGURE 9

Comparison of the tantalum-Astroquartz and nickel-Astroquartz combinations shows that the thermal conductivity of the tantalum-Astroquartz combination was lower although the same spacer was used in both tests and the installed layer density of the tantalum Astroquartz was greater. A high layer density insulation combination would be expected to have a high solid conduction component of heat transfer that would result in a greater thermal conductivity particularly at lower temperatures, less than 1000°F. Examination of the nickel-Astroquartz after testing disclosed thermal shorting, occasional contacting of alternate layers of nickel foil, had occurred during installation resulting in the higher thermal conductivity. Although tantalum foil displayed incompatibility during long-term, 2000-hour, testing and is not recommended as a radiation shield material, it is believed that the short life thermal conductivity-temperature relationship of tantalum-Astroquartz has been established and more significantly, the solid conductivity characteristics of the Astroquartz cloth spacer measured.

The thermal conductivity of the copper foil - copper opacified Refrasil quartz paper combination is seen to be higher than that of the copper foil-Refrasil quartz fiber paper combination. The layer density, 70.4 layers per inch, of the copper foil-copper opacified Refrasil quartz paper combination was about the highest that may be practically achieved because of the thickness of the copper flake opacified Refrasil quartz fiber paper spacer, nominal 0.010 inches, while the layer density of the copper foil-Refrasil paper combination, 83.3 layers per inch, does not represent a maximum for this combination. It is believed that the higher thermal conductivity observed for the copper foil-copper opacified Refrasil paper combination is the result of the higher spacer density due to the compression, and hence an increased solid conductivity component of heat transfer.

During Phase II work the thermal conductivity VS temperature relationships of various insulation combinations including nickel-Astroquartz and copper-copper opacified Refrasil paper will be examined with a flat plate thermal conductivity tester. This tester will allow investigation of the layer density VS thermal conductivity relationship of the insulation combinations in order to establish the optimum layer density. With the cylindrical type tester described in this report thermal performance data were obtained at only one layer density.

3.3.1.5.1 Aluminum Foil-Glass Fiber Paper

The radiation shield material used in this test was type AA-1145-0, rolled aluminum foil; 1/4 mil thick and unprocessed (section

3.4.1.1). The spacer was No. 106 glass fiber paper, processed by the air oxidation method (section 3.4.1.2), 3 mil nominal thickness and with a nominal weight of 1.7 grams/square foot.

Sixty layers of this insulation were applied to the cylindrical test portion to a thickness of 1/2 inch (120 layers/inch) as shown in Figure 10. The insulated test core and heater assembly were placed in a holding fixture to complete the end application, Figure 11. The insulation was then folded around the ends as shown in Figure 12, a view of the top head. The electrically insulated heater and thermocouple wires can be seen passing through the insulation.

Figure 13 shows the insulated cylinder prior to initiation of thermal testing.

Results of thermal conductivity testing of this combination are presented in Table VII. The pressure level during these tests was maintained by dynamic pumping and ranged from 0.05 to 0.1 micron of mercury pressure.

While point 3 (Table VII) was being established, small fluctuations were noted in the hot side temperature profile on the temperature controller. Electrical checks of the apparatus showed that apparently an electrical leak had developed in the insulated heater lead wires as they passed through the insulation. The electrical short was transmitted to the thermocouples in contact with the insulation foils and caused fluctuations in the output signal. The power to the heater was momentarily interrupted while the thermocouple readings were taken to establish hot and cold face temperatures. It is believed that the thermal conductivity calculated for this point is approximately 13 per cent higher than the actual value because of power loss due to the electrical short. At the conclusion of the test, while a mass spectrometer analysis of the vacuum space was being conducted, the vacuum line from the tester to the mass spectrometer was accidentally broken and the insulated tester suddenly exposed to air.

Visual examination of the insulation combination after disassembly showed the aluminum foil radiation shields to be bright and shiny throughout the combination. The first 12 layers, starting at the hot side, of glass fiber paper appeared slightly yellow-brown in color but still soft and handleable. It was suspected that this color change might indicate metallic contamination, but an X-ray diffraction analysis disclosed no crystalline material to be present. The rest of the glass fiber paper layers appeared unchanged from the installed condition.

Employing the methods discussed in Section 3.3.1.1, the experimental data from this test is plotted in Figure 14 in the form of:

$$k_a \left(\frac{T_1 - T_2}{T_1^2 - T_2^2} \right) \text{ vs } \frac{T_1^4 - T_2^4}{T_1^2 - T_2^2}$$

Figure 10 Aluminum Foil - Glass Fiber Paper Combination,
Application to Test Apparatus Cylindrical Section

Figure 11. Aluminum Foil - Glass Fiber Paper Combination in Fixture
Before End Insulation Application

Figure 12. Aluminum Foil - Glass Fiber Paper Combination with End Insulation Application

Figure 13. Aluminum Foil - Glass Fiber Paper Combination
Insulated Tester Before Thermal Testing

TABLE VII

THERMAL CONDUCTIVITY OF ALUMINUM FOIL-GLASS FIBER PAPER
INSULATION COMBINATION

<u>Data Point</u> <u>No.</u>	<u>Cold Side</u> <u>Temperature, °F</u>	<u>Hot Side</u> <u>Temperature, °F</u>	<u>Thermal Conductivity</u> <u>Btu/hr-ft-°F x 10⁻³</u>
1	109.4	482.0	0.174
2	143.6	779.0	0.289
3*	161.6	878.0	0.381

*This value is estimated to be approximately 13 per cent higher than the actual thermal conductivity because of power losses, due to an electrical short, that occurred while the data point was being established.

APPARENT THERMAL CONDUCTIVITY
CORRELATION-ALUMINUM FOIL-
GLASS FIBER PAPER

Figure 14

The intercept of the line, $a/2$ (the solid conductivity constant) = $.053 \times 10^{-6} \frac{\text{Btu}}{\text{hr-ft-}^\circ\text{R}^2}$ and the slope, $b/4$ (the radiation constant) = $.52 \times 10^{-13} \frac{\text{Btu}}{\text{hr-ft-}^\circ\text{R}^4}$.

Substituting these constants in Equation (36), we obtain an expression that permits the calculation of thermal conductivity for this composite at any set of boundary conditions from approximately 900°F to 100°F.

$$k_a = .053 \times 10^{-6} \left(\frac{T_1^2 - T_2^2}{T_1 - T_2} \right) + .52 \times 10^{-13} \left(\frac{T_1^4 - T_2^4}{T_1 - T_2} \right) \quad (45)$$

An effective emissivity for the aluminum foil radiation shields may be calculated from:

$$\begin{aligned} b/4 &= \epsilon/2n = .52 \times 10^{-13} \\ &= .0086 \end{aligned}$$

In comparison, handbook data indicates emissivities of 0.01 at ambient and 0.03 at 1000°F for bare aluminum not in contact with spacer material.

3.3.1.5.2 Aluminum Foil - Aluminum Flake Opacified Glass Fiber Paper

The radiation shield material used in this test was type AA-1145-0 aluminum foil; 1/4 mil thick and unprocessed (Section 3.4.1.1). The spacer was aluminum flake opacified glass fiber paper processed by the air oxidation method (Section 3.4.1.2), nominal thickness 3 mils and weighing 1.1 grams per square foot, nominal.

Seventy-five layers of this combination were applied to the cylindrical portion of the thermal conductivity tester in a thickness of 1/2 inch, 150 layers/inch.

Results of thermal conductivity testing of this combination are presented in Table VIII. Employing the methods discussed in Section 3.3.1.1, the experimental data from this test is plotted in Figure 15 in the form of:

$$k_a \left(\frac{T_1^2 - T_2^2}{T_1 - T_2} \right) \quad \text{vs.} \quad \left(\frac{T_1^4 - T_2^4}{T_1^2 - T_2^2} \right)$$

The intercept of the line, $a/2$ (the solid conductivity constant) = $-.0162 \times 10^{-6} \text{ Btu/hr-ft-}^\circ\text{R}^2$. and the slope, $b/4$ (the radiation constant) = $.76 \times 10^{-13} \text{ Btu/hr-ft-}^\circ\text{R}^4$.

Substituting these constants in Equation (36), we obtain an expression that permits the calculation of thermal conductivity for this composite at any set of boundary conditions from approximately 850°F to 100°F.

$$k_a = -.0162 \times 10^{-6} \left(\frac{T_1^2 - T_2^2}{T_1 - T_2} \right) + .76 \times 10^{-13} \left(\frac{T_1^4 - T_2^4}{T_1 - T_2} \right)$$

An effective emissivity for the aluminum foil radiation shields may be calculated from:

$$b/4 = \frac{1}{2n} = .76 \times 10^{-13}$$

$$= .0133$$

3.3.1.5.3 Copper Foil-Refrasil Quartz Fiber Paper

The radiation shield material used in this test was electro-deposited copper foil; 1/2 mil thick and processed by the Linde Foil Cleaning Process (Section 3.4.1.1). The spacer used was Refrasil Quartz Fiber Paper, processed by the air oxidation process (Section 3.4.1.2), 5 mil nominal thickness and with a nominal weight of 2.4 grams per square foot.

Fifty layers of this combination were applied to the cylindrical portion of the tester to a thickness of 0.6 inch, resulting in a layer density of 83.3 layers/inch. Refrasil quartz fiber paper was first processed by the oxidation process and then used as a separator during processing of the copper foil. This method allowed application of the spacer-foil combination from a single holding shaft. A higher layer density could not be achieved because of slippage of the combination on the transfer shaft when tension was applied during wrapping. Results of the thermal conductivity testing are presented in Table X.

TABLE VIII

THERMAL CONDUCTIVITY OF ALUMINUM FOIL-ALUMINUM FLAKE OPACIFIED GLASS FIBER PAPER

<u>Data Point Number</u>	<u>Temperature, F°</u>		<u>Thermal Conductivity BTU/hr/ft/°R x 10⁻³</u>
	<u>Cold Side</u>	<u>Hot Side</u>	
1	89.6	399.2	0.085
2	104.0	734	0.206
3	113.0	842	0.270

Figure 15.

APPARENT THERMAL CONDUCTIVITY CORRELATION -
ALUMINUM FOIL - ALUMINUM FLAKE
OPACIFIED GLASS FIBER PAPER

TABLE IX

MASS SPECTROMETER ANALYSES OF TEST SYSTEMS

<u>Insulation System</u>	<u>Temp. (°F.)</u>	<u>H₂ (%)</u>	<u>CO (%)</u>	<u>CO₂ (%)</u>	<u>O₂ (%)</u>	<u>N₂ (%)</u>	<u>Hydrocarbons (%)</u>
Copper foil-Refrasil paper	1260	67	18	2	0	11	Trace
Copper foil-copper opacified paper	1650	78	15	2	0	3	Trace
Nickel foil-Astroquartz cloth	1600	69	20	1	0	2	8
Nickel foil-Refrasil paper	1630	3	66	1	0	27	2
Tantalum foil-Astroquartz cloth	1500	20	71	1	2	5	Trace

The vacuum level during these tests was maintained by dynamic pumping and ranged from 0.045 to 0.10 microns of mercury pressure. This insulation combination was subjected to 2065 hours of testing at a hot face temperature of approximately 1260°F with no change in thermal performance. During these long-term tests, the power required to maintain the insulation hot and cold boundary temperatures at a constant level was the measure of the thermal stability of the insulation system.

From Figure 16 the slope of the line, $\frac{b}{4} = .52 \times 10^{-13}$ Btu/hr-ft°R⁴ and the intercept $\frac{a}{2} = 0.095 \times 10^{-6}$ Btu/hr-ft°R². The constant $\frac{a}{2}$, the solid conductivity coefficient, and $\frac{b}{4}$, the radiation coefficient, substituted in the general thermal conductivity equation define the temperature dependence of the insulation system.

$$k_a = .095 \times 10^{-6} \left(\frac{T_1^2 - T_2^2}{T_1 - T_2} \right) + .52 \times 10^{-13} \left(\frac{T_1^4 - T_2^4}{T_1 - T_2} \right) \quad (46)$$

Equation (46) permits the calculation of the thermal conductivity of this insulation combination between any set of boundary conditions from approximately 100°F to 1300°F.

The effective emissivity of the copper foil radiation shields may be obtained from the relationship:

$$\frac{b}{4} = \frac{\sigma \epsilon}{2n} = .52 \times 10^{-13}$$

where σ = Stephan-Boltzmann constant
 ϵ = effective emissivity of the radiation shields
 n = number of compartments bounded by the radiation shields per unit thickness of insulated space

Solving for the emissivity: $\epsilon = .059$

This effective emissivity represents the emissivity of the copper foils in this test configuration (copper foils in contact with Re-frasil quartz fiber paper spacer) over the boundary temperatures of the test.

Handbook values indicate a fairly constant value of 0.035 from ambient to 1500°F for bare copper not in contact with spacer materials.

TABLE XTHERMAL CONDUCTIVITY OF COPPER FOIL-REFRASIL QUARTZ FIBER
PAPER INSULATION COMBINATION

<u>Data Point No.</u>	<u>Cold Side</u> <u>Temperature, °F</u>	<u>Hot Side</u> <u>Temperature, F°</u>	<u>Thermal Conductivity</u> <u>BTU/hr ft F° x 10⁻³</u>
1	158.0	720.0	0.324
2	203.0	982.4	0.455
3	257.8	1243.0	0.641

APPARENT THERMAL CONDUCTIVITY CORRELATION
COPPER FOIL-REFRASIL QUARTZ FIBER PAPER

Figure 16

A mass spectrometer analysis of the vacuum chamber conducted while the insulation tester hot face was operating at approximately 1260°F indicated the following compositions:

Hydrogen	67.0%
Carbon monoxide	18.0%
Carbon dioxide	1.8%
Nitrogen	10.7%
Hydrocarbons	Trace

This is in reasonable agreement with offgas compositions determined for copper foil and Refrasil paper during the offgassing experiments (Section 3.4.2).

The test apparatus was next revised for air exposure experiments by replacement of the original tungsten wire heater with a nickel-chromium alloy heater capable of air exposure. These experiments are discussed in Section 3.3.3.

3.3.1.5.4 Copper Foil-Copper Flake Opacified Refrasil Quartz Fiber Paper

The radiation shield material used in this test was electro-deposited copper foil; 1/2 mil thick and processed by the Linde Foil Cleaning Process (Section 3.4.1.1). The spacer was copper flake opacified Refrasil quartz fiber paper processed by the Linde Insulation Cleaning Process (Section 3.4.1.2); of 10 mil nominal thickness and with a nominal weight of 5.5 grams per square foot.

Forty-eight layers of this combination were applied to the cylindrical portion of a thermal conductivity tester in a thickness of 0.54 inches resulting in a layer density of 70.4 layers/inch. This layer density represents about the highest that may be practically achieved because of the thickness of the copper flake opacified Refrasil quartz fiber paper spacer, nominal 0.010 inches. The nominal thicknesses of spacers such as Astroquartz cloth and Refrasil quartz fiber paper is approximately 0.005 inches.

The results of thermal conductivity testing of this combination are presented in Table XI.

TABLE XI

THERMAL CONDUCTIVITY OF COPPER FOIL-COPPER FLAKE OPACIFIED REFRASIL QUARTZ FIBER PAPER INSULATION COMBINATION

<u>Data Point No.</u>	<u>Cold Side Temperature, °F</u>	<u>Hot Side Temperature, °F</u>	<u>Thermal Conductivity Btu/hr.ft.°F x 10⁻³</u>
1	185	635	0.725
2	275	977	1.05
3	347	1265	1.36

The pressure level during these tests was maintained by dynamic pumping and ranged from 0.07 to 0.10 micron of mercury. This combination was subjected to 2155 hours of testing at approximately 1270°F hot side temperature with no apparent change in insulation thermal performance.

The solid conductivity coefficient, $a/2$, and the radiation coefficient, $b/4$, were obtained from Figure 17.

Substituting in the general thermal conductivity equation:

$$k_a = .323 \times 10^{-6} \left(\frac{T_1^2 - T_2^2}{T_1 - T_2} \right) + .83 \times 10^{-13} \left(\frac{T_1^4 - T_2^4}{T_1 - T_2} \right) \quad (42) \quad (47)$$

Calculating an effective emissivity of the copper foil radiation shields from the relationship:

$$\frac{b}{4} = \frac{\sigma \epsilon}{2n} = .83 \times 10^{-13}$$

where:

- σ = Stefan-Boltzmann constant
- ϵ = effective emissivity
- n = number of compartments bounded by the radiation shields per unit thickness of insulation space

Solving for the emissivity, $\epsilon = 0.081$

Comparing the data for this test with that of the copper foil-Refrasil quartz fiber paper combination (Section 3.3.1.5.3).

	Copper=Copper Opacified Refrasil Paper			Copper=Refrasil Paper		
$T_{hot}, ^\circ F$	635	977	1265	720	982.4	1243
$k, \text{Btu/hr.ft.}^\circ R \times 10^{-3}$	0.725	1.05	1.36	0.324	0.455	0.641
$a/2, \text{Btu/hr.ft.}^\circ R^2 \times 10^{-6}$		0.323			0.095	
$b/4, \text{Btu/hr.ft.}^\circ R^4 \times 10^{-13}$		.830			0.520	
$\epsilon,$		0.081			0.059	
$N, \text{layers/inch}$		70.4			83.3	

Analyzing the results of the tests, it can be seen that the effective emissivity and radiation coefficients are fairly comparable, bearing in mind that these values represent emissivities that are influenced by the spacer materials that they contact. The solid conductivity coefficient of the copper-copper opacified Refrasil paper combination is about 3 times that of the copper-Refrasil paper combination and the

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FIGURE 17

apparent thermal conductivity is roughly two times greater over the temperature range investigated. The layer density, 70.4 layers per inch, of the copper-copper Refrasil paper combination is about the highest that may be practically achieved because of the thickness of the copper flake opacified Refrasil quartz fiber paper spacer, nominal 0.010 inches, while the layer density of the copper-Refrasil paper combination, 83.3 layers per inch, does not represent a maximum. It is believed that the higher thermal conductivity observed for the copper-copper Refrasil paper combination is the result of the higher layer density application which resulted in a greater spacer contact, due to the compression, and hence an increased solid conductivity component of heat transfer. In order to evaluate the optimum layer density to which the copper-copper opacified Refrasil paper combination should be applied for minimum thermal conductivity, the layer density-thermal conductivity relationship will have to be established. This would involve a thermal conductivity evaluation at a minimum of two more layer densities.

A mass spectrometer analysis of the vacuum chamber conducted while the insulation tester hot face was operating at approximately 1650°F indicated the following composition:

Hydrogen	78.1%
Carbon monoxide	15.3%
Nitrogen	2.4%
Carbon dioxide	2.0%
Hydrocarbons	Trace

This is in reasonable agreement with offgassing compositions determined for copper foil and copper opacified Refrasil paper in the offgassing tests. (Section 3.4.2).

3.3.1.5.4.1 Copper Foil - Copper Flake Opacified Refrasil Quartz Fiber Paper - Thermal Conductivity in Inert Gas

At the conclusion of the 2000 hour thermal performance test the vacuum of the test apparatus was broken with Argon gas to one atmospheric pressure and thermal performance testing in this gas initiated.

At pressure levels on the order of one atmosphere the mean-free path of the molecules is very small with the molecules experiencing many collisions, so that heat transport within the insulation system is primarily by gaseous conduction. It is, therefore, expected that the inert gas thermal conductivity data of this test can be extrapolated to other foil-paper combinations because the thermal effects of gaseous conduction should be the same for composites having spacers with the same interstitial fiber spacing.

The results of the testing are presented in Table XII.

TABLE XIITHERMAL CONDUCTIVITY OF COPPER FOIL-COPPER FLAKEOPACIFIED REFRASIL QUARTZ FIBER PAPER IN ARGON GAS AT ONE ATM. PRESSURE

<u>Data Point No.</u>	<u>Cold Side Temperature, °F</u>	<u>Hot Side Temperature, °F</u>	<u>Thermal Conductivity Btu/hr.ft.°F x 10⁻³</u>
1	140	203	15.65
2	257	491	19.85
3	366.8	770	22.03
4	494.6	1094	24.86

These measured thermal conductivity values range from 22 to 35.5% greater than the thermal conductivity of Argon gas itself over these temperature ranges. This difference observed is attributed to the solid conductivity and radiation transport of the multi foil system as well as gas convection.

3.3.1.5.5 Nickel Foil-Astroquartz Cloth

The radiation shield material used in this test was type 200, rolled nickel foil; 0.62 mil thick and unprocessed (Section 3.4.1.1). The spacer was Astroquartz cloth; processed by the air oxidation method (Section 3.4.1.2), 5 mil nominal thickness and with a nominal weight of 10.4 grams per square foot.

Sixty layers of this combination were applied to the cylindrical tester to a thickness of 1/2 inch (layer density of 120 layers per inch). Figure 18 shows the insulated tester with insulated heater and thermocouple wires protruding from it before initiation of thermal performance testing. Results of thermal conductivity testing of this combination are presented in Table XIII. Pressure level during these tests was maintained by dynamic pumping and ranged from 0.28 to 0.10 microns of mercury pressure.

Comparison of the data from this test with that of the tantalum foil - Astroquartz cloth combination (3.3.1.5.7) disclosed that the solid conductivity coefficient, $a/2$, of the tantalum-astroquartz was about 10 times lower than that of the nickel-Astroquartz. Examination of the nickel-Astroquartz combination after testing disclosed the presence of thermal shorting, contact of alternate nickel foil layers, which would thermally appear as an increase in solid conductivity. Thus it is believed that the data of Table XII for nickel-Astroquartz is high by about 25%. This comparison is discussed in detail in Section 3.3.1.5.7. The cold side temperatures of points 4 and 5 are extrapolated from the preceding temperature data because the cold side thermocouple became inoperable during data point 4. A run was made at 1947°F

Figure 18. Nickel Foil - Astroquartz Cloth Insulated Tester

TABLE XIIITHERMAL CONDUCTIVITY OF NICKEL FOIL-ASTROQUARTZCLOTH INSULATION COMPOSITE

<u>Data Point Number</u>	<u>Cold Side Temperature °F</u>	<u>Hot Side Temperature °F</u>	<u>Thermal Conductivity Btu/hr-ft-R° x 10⁻³</u>
1	130	503	0.68
2	212	850	1.35
3	329	1268	2.32
4	450*	1625	3.01
5	550*	1947	4.54

*Extrapolated from preceding temperature data.

in order to note the thermal performance of this combination at this temperature and also to check the reliability of the apparatus, particularly the tungsten coil heater. The solid conductivity coefficient $a/2$ and the radiation coefficient $b/4$ are obtained from Figure 19. The data were fitted with polynomials using a least squares technique. The intercept of this line is: $a/2 = .31 \times 10^{-6}$ Btu/hr.ft. $^{\circ}R^2$ and the slope $b/4 = 1.47 \times 10^{-13}$ Btu/hr.-ft. $^{\circ}R^4$. Substituting these constants, we obtain an equation defining the temperature dependence of this insulation system:

$$k_a = .31 \times 10^{-6} \left(\frac{T_1^2 - T_2^2}{T_1 - T_2} \right) + 1.47 \times 10^{-13} \left(\frac{T_1^4 - T_2^4}{T_1 - T_2} \right) \quad (48)$$

This equation permits the calculation of thermal conductivity between any set of boundary temperatures from approximately 2000°F to 100°F.

The effective emissivity of the nickel radiation shields may be obtained from the relationship:

$$b = \frac{2\sigma\epsilon}{n}$$

$$\frac{b}{4} = \frac{\sigma\epsilon}{2n} = 1.47 \times 10^{-13}$$

Solving for the emissivity:

$$\epsilon = .24$$

The effective emissivity represents the emissivity of the nickel foils in this test configuration (nickel foils in contact with Astroquartz cloth spacer) over the boundary temperatures of the test.

Handbooks indicate values ranging from 0.06 at ambient to 0.18 at 2000°F. Handbook values are determined for bare materials which are not in contact with spacer materials (Astroquartz cloth).

A mass spectrometer analysis of the vacuum chamber while the insulation tester hot face was approximately 1600°F indicated the following composition:

Hydrogen	67.0%
Carbon monoxide	20.0%
Water vapor.....	3.3
Carbon dioxide	0.5
Nitrogen	1.0
Hydrocarbons	8.1
Argon	0.1

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CORRELATION-NICKEL FOIL-
ASTROQUARTZ CLOTH

Figure 19

This analysis is similar to the offgas analysis observed for Astroquartz cloth, Section 3.4.2, showing the gas to be composed primarily of hydrogen, carbon monoxide and hydrocarbons. It is believed that most of the water vapor was picked up from the vacuum transfer line between the vacuum chamber and the mass spectrometer.

3.3.1.5.6 Nickel Foil-Refrasil Quartz Fiber Paper

The radiation shield material used in this test was type 200, rolled nickel foil; 0.62 mils thick and unprocessed (Section 3.4.1.1). The spacer material was Refrasil quartz fiber paper; processed by the air oxidation process (Section 3.4.1.2), 5 mil nominal thickness with a nominal weight of 2.37 grams per square foot.

Fifty-five layers of the insulation were applied to the cylindrical portion of a thermal conductivity tester to a thickness of 0.572 inch (96.1 layers/inch). Test results are presented in Table XIV.

After data point 2 had been established and the temperature level was being increased to obtain data point 3, fluctuations were observed in both hot face thermocouples. Electrical checks with an ohmmeter disclosed that both thermocouples were broken. Since the replacement of the original hot face thermocouples would involve time consuming disassembly and subsequent reinsulation of the top head insulation, it was decided to insert two new hot face thermocouples into the insulation wrap without disassembly. The technique used, involved boring two holes through the top head of the insulation at such a location that the holes just cleared the outside of the stainless steel tester core and thus contacted the hot innermost layer of the insulation. The holes were produced with a 1/8" O.D. stainless steel rod, the end of which was sharpened to a fine point. The hot face thermocouples were encased in 1/8" O.D. ceramic insulation sleeves and then fitted into the holes. After installation of the new thermocouples, testing was resumed and the third data point was established. The third data point is about 10% higher than that which would be obtained from an extrapolation of the two preceding points obtained before installation of the new thermocouples. This method of repair apparently resulted in much less thermal performance degradation than occurred when the insulation was disassembled (tantalum foil-Astroquartz cloth test, Section 3.3.1.5.7, copper foil-Refrasil paper, air exposure tests, Section 3.3.3).

This combination was subjected to a 2017 hour thermal performance test at approximately 1630°F hot side temperature with no change in insulation thermal performance.

The solid conductivity coefficient, $a/2$, and the radiation coefficient, $b/4$, were obtained from Figure 20.

where: $a/2$ is the intercept and $b/4$ is the slope.

TABLE XIV

THERMAL CONDUCTIVITY OF NICKEL FOIL-REFRASIL QUARTZ
FIBER PAPER INSULATION COMBINATION

<u>Data</u> <u>Point No.</u>	<u>Cold Side</u> <u>Temperature, °F</u>	<u>Hot Side</u> <u>Temperature, °F</u>	<u>Thermal Conductivity</u> <u>Btu/hr-ft-°F x 10⁻³</u>
1	176	815	0.73
2	275	1238	1.23
3*	338	1616	2.11

*This value is estimated to be approximately 10% higher than the actual thermal conductivity because it was obtained after the insulation system had been modified by the addition of two new hot face thermocouples. This was necessitated because of failure of the original thermocouples.

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QUARTZ FIBER PAPER

$$\frac{k_B (T_1 - T_2)}{T_1^2 - T_2^2} \times \text{BTU/HR FT } ^\circ\text{R}^2$$

$$\frac{T_1^4 - T_2^4}{T_1^2 - T_2^2} \times 10^7, ^\circ\text{R}^2$$

FIGURE 20

Substituting in the general thermal conductivity equation:

$$k_a = 0.20 \times 10^{-6} \left(\frac{T_1^2 - T_2^2}{T_1 - T_2} \right) + 1.23 \times 10^{-13} \left(\frac{T_1^4 - T_2^4}{T_1 - T_2} \right) \quad (49)$$

An effective emissivity of the nickel foil radiation shields obtained from the relationship:

$$\frac{b}{4} = \frac{\sigma \epsilon}{2n} = 1.23 \times 10^{-13}$$

where:

σ = Stefan-Boltzmann constant

ϵ = effective emissivity

n = number of compartments bounded by the radiation shields per unit thickness of insulated space

and solving for the emissivity, $\epsilon = 0.163$

This is in reasonable agreement with handbook data which indicates values ranging from 0.06 at ambient to 0.18 at 2000°F. Handbook values, however, are determined for bare materials which are not in contact with spacer materials (Refrasil paper).

A mass spectrometer analysis conducted at a hot face temperature of approximately 1630°F indicated the following composition:

Hydrogen	3.13%
Carbon monoxide	66.38%
Carbon dioxide	1.27%
Nitrogen	27.34%
Hydrocarbons	1.88%

This is in reasonable agreement with offgassing compositions determined for nickel and Refrasil paper in the offgassing tests (Section 3.4.2).

3.3.1.5.7 Tantalum Foil-Astroquartz Cloth

The radiation shield material used in this test was rolled, 99.9% pure tantalum foil; 0.3 mil thick and unprocessed (Section 3.4.1.1). The spacer material was Astroquartz Cloth; processed by the air oxidation method (Section 3.4.1.2), 5 mil nominal thickness with a nominal weight of 10.4 grams per square foot.

Forty-two layers of this combination were applied to the cylindrical portion of a thermal conductivity tester in a thickness of 0.3 inches resulting in a layer density of 140 layers/inch.

Results of the thermal conductivity testing are presented in Table XV. The pressure level during these tests was maintained by

TABLE XVTHERMAL CONDUCTIVITY OF TANTALUM FOIL-ASTROQUARTZ CLOTH
INSULATION COMBINATION

<u>Data Point No.</u>	<u>Cold Side Temperature, F°</u>	<u>Hot Side Temperature, F°</u>	<u>Thermal Conductivity, BTU/hr ft F° x 10⁻³</u>
1	136.4	617.0	0.466
2	212.0	1005.0	0.932
3	285.8	1351.5	1.853

dynamic pumping and ranged from 0.09 to 0.10 microns of mercury pressure.

In Figure 21 the experimental thermal conductivity data is plotted and $\frac{a}{2}$ was found to equal 0.03×10^{-6} Btu/hr.ft. $^{\circ}$ R 2 and $\frac{b}{4}$ equalled 1.59×10^{-13} Btu/hr.ft. $^{\circ}$ R 4 .

These constants are substituted in the general thermal conductivity equation to define the temperature dependence of the insulation system:

$$k_a = .03 \times 10^{-6} \left(\frac{T_1^2 - T_2^2}{T_1 - T_2} \right) + 1.59 \times 10^{-13} \left(\frac{T_1^4 - T_2^4}{T_1 - T_2} \right) \quad (50)$$

This equation permits the calculation of thermal conductivity between any set of boundary temperatures from approximately 100 $^{\circ}$ F to 1800 $^{\circ}$ F.

An effective emissivity for the tantalum foil radiation shields is calculated from;

$$\frac{b}{4} = \frac{\sigma \epsilon}{2n} = 1.59 \times 10^{-13}$$

Solving for the emissivity:

$$\epsilon = .306$$

The effective emissivity value appears high when compared with handbook values of tantalum which range from 0.035 at ambient to 0.05 at 2500 $^{\circ}$ F. Handbook values however are determined for bare materials which are not contacted by other materials. In the case of the tantalum foil-Astroquartz cloth combination, the tantalum foil was tightly wrapped (140 layers/inch) in an insulation system and the foil emissivity may be influenced by the presence of the Astroquartz spacer.

These data are next compared with the nickel foil-Astroquartz cloth insulation combination (Section 3.3.1.5.5) because the spacer Astroquartz cloth was the same in both cases.

	Effective Emissivity, ϵ	$\frac{b}{4}$, BTU/hr ft $^{\circ}$ R 4	$\frac{a}{2}$, BTU/hr ft $^{\circ}$ R 2	Layer Density Layers/inch
Nickel foil-Astroquartz cloth	0.240	1.47×10^{-13}	0.31×10^{-6}	120
Tantalum foil-Astroquartz cloth	0.306	1.59×10^{-13}	0.03×10^{-6}	140

While the effective emissivities and radiative conductivity coefficients, $\frac{b}{4}$, were roughly the same, the solid conductivity coefficient $\frac{a}{2}$, of the tantalum-Astroquartz cloth composite was almost 10 times

Figure 21

lower than that of the nickel-Astroquartz. This seemed contradictory because the tantalum-Astroquartz was applied at a higher layer density than the nickel-Astroquartz. The insulation system that is applied at a higher layer density would be expected to have a higher solid conductivity component because of an increase in contact area of the spacer fibers due to compression. The raw data and thermal conductivity calculations for both insulation systems were rechecked as well as the instrumentation used on the tests. No errors were found. Next the nickel-Astroquartz cloth insulation system, which had previously been removed from a test core and stored, was examined. Evidence of thermal shorting, contact of alternate nickel foil layers, through breaks in the Astroquartz spacer was observed in the "knuckle" area where the cylindrical portion was folded over the ends of the test core. The effects of these thermal shorts would appear as an apparent increase in the solid conductivity component of heat transfer. This effect would result in a higher overall thermal conductivity for this insulation system than is actually the case.

Thus it appears that the thermal conductivity data of the nickel foil-Astroquartz cloth combination, Section 3.3.1.5.5, is about 25% higher than would be expected. It is recommended that the nickel foil-Astroquartz cloth test be repeated to obtain reliable data.

The 2000 hour thermal performance test was initiated and after 517 hours of testing, with no thermal performance degradation, a heater failure occurred. The top head of the insulated tester was disassembled to provide access to the heater. Examination of the platinum-rhodium heater wire showed that the wire had failed in two places. Cause of the failure was attributed to contaminants, manganese, silicon, aluminum, copper, and chromium, which reacted with the platinum-rhodium to form brittle bimetallic alloys. It is believed that the source of the contaminants was the thermal conductivity tester on which the insulation is wrapped. The tester is constructed from Type 310 stainless steel and is within the vacuum space with the platinum-rhodium heater. The wire vendor recommended a nickel tester as being more suitable from a compatibility viewpoint. Instead of fabricating a new nickel core, a tungsten heater was substituted for the platinum-rhodium heater and the existing stainless steel core and its installed insulation reused.

After replacement of the heater, the calorimeter heat leak increased by a factor of approximately 2.4 as a result of damage to the insulation incurred during heater repairs. When thermal performance testing was resumed a constant hot face temperature could not be maintained at constant power input. Hot face temperature fluctuated between 1590°F and 1463°F at power inputs which should have yielded hot side temperatures in excess of 1800°F.

Electrical checks showed that an electrical short had occurred allowing some current to be shunted through the insulation to the thermocouples. It was decided to continue the test rather than attempt another

disassembly of the insulation for heater and/or thermocouple repairs which would probably further degrade insulation performance due to the handling involved. Pressure level of the tester at this time was approximately 0.15 micron of mercury and leak detection with a helium mass spectrometer leak detector disclosed a small leak in a thermocouple vacuum feedthrough. After repair of the leak the pressure level decreased to approximately 0.05 micron of mercury. Hot face temperature continued to decrease, however.

A mass spectrometer gas analysis conducted after approximately 2000 hours of testing, disclosed the following vacuum environment compositions:

Hydrogen	19.8%
Nitrogen	5.3
Carbon monoxide	71.4
Carbon dioxide	0.9
Oxygen	1.5
Hydrocarbons	Trace

This analysis is similar to those taken for Astroquartz cloth during the offgassing tests with the exception of the 1.5% oxygen. Leak detection with a helium mass spectrometer disclosed the presence of a second leak around a thermocouple feedthrough. The leak was repaired and the test terminated after 2300 hours of operation.

Examination of the insulation showed that from the outer or cold face of the insulation to about 1/2 the distance through the wrap both the tantalum foil and Astroquartz cloth were quite flexible. The middle zone and inner hot face foils and spacer, however, were very brittle. The foils were badly discolored. Samples of the tantalum foil were taken from various portions of the insulation for laboratory analysis. The analysis, discussed in detail in Section 3.5.1.3.1, showed that the tantalum had reacted to form tantalum oxide, tantalum silicide and other compounds such as iron tantalum. It is believed that the formation of these high emissivity compounds resulted in the observed thermal performance degradation. Tantalum is not recommended as a radiation shield because of its incompatibility with the Astroquartz cloth spacer.

3.3.2 Insulation Panels - Thermal Effects of Joints and Exposed Edges

Insulation can be installed on multi-sided configurations in such a manner that each radiation shield essentially envelopes the item, leaving no gaps or radiation windows in the shield. This procedure has been widely used on applications requiring the absolute minimum in heat leak but its employment is frequently time consuming and expensive. In the case of complicated configurations it may be impossible. Insulation applications in these cases can be made by the use of insulation panels which are butted or otherwise joined together. On some applications the use of these insulation panels result in configurations where the panel edges are exposed to the environment. The analytical study described in this section was performed to investigate the effect of panel joints and exposed edges on panel thermal conductivity and to document methods for predicting the thermal performance of such panels.

A computer program for evaluating the effect on thermal performance of various panel geometries and segment edge conditions was developed. Edge conditions handled by the program include heat transfer between edges of adjacent panels, across either a rectangular or wedge-shaped gap, by solid conduction (filled gap) or radiation (unfilled gap), and radiation from a panel edge to an infinite wall. The geometric parameters investigated for a rectangular gap were: panel width, gap width, and panel thickness. For the wedge shaped gap, the parameters were: panel width, wedge angle, and panel thickness. For the exposed edge problem, panel width, panel thickness, and ambient temperature were investigated. The effect of these variables is presented in the form of parametric curves. Under severe conditions, the effective thermal conductivity with a gap can be as much as ten times the conductivity without a gap. Under more typical conditions, the thermal conductivity with a gap would be about 1.5 times the conductivity without a gap. With a material of medium thermal conductivity in the gap, losses were decreased to the extent that the effective conductivity of typical panels was about 10% higher than the conductivity without a gap.

3.3.2.1 Computer Program

The purpose of the computer program was to determine the effects of joints and exposed edges upon the overall thermal performance of insulation panels. Edge conditions handled by the program include (See Figure 22).

- a.) Radiation or solid conduction across a rectangular gap joint.
- b.) Radiation or solid conduction across an angular gap joint (warm edges in contact).
- c.) Radiation from an exposed edge to an infinite wall at a specified temperature.

The mathematical model upon which the computer program is based assumes that the panel consists of filler sheets (glass, quartz, etc.) alternated with highly reflective foils in a layered structure. The panel dimension in the direction parallel to both the interpanel joint and the insulation layers is large enough so that a two-dimensional thermal analysis, in the (Y - Y) plane normal to this direction, will suffice. The X direction is chosen to be parallel to the layers, the Y direction normal to the layers. Schematic drawings of an insulation panel under the influence of different edge conditions are shown in Figure 22.

The development of the computer program is presented in Appendix B.

3.3.2.2 Parametric Study

Table B-I presented in Appendix B lists input parameters and four of the five output heat fluxes for 103 cases analyzed to investigate the effects of varying edge conditions and panel dimensions.

An energy balance written around an insulation panel would consist of:

$$Q_w = Q_l + Q_e \quad (51)$$

where:

Q_w	=	flux from warm side of panel
Q_l	=	flux from panel edge
Q_e	=	flux from cold side of panel
Q_{wg}	=	flux from warm side of gap
Q_{cg}	=	flux from cold side of gap

SCHEMATIC OF INSULATION PANEL FOR DIFFERENT EDGE CONDITIONS

FIGURE 22

The flux incident to the cold side of the gap, Q_{cg} ,

$$Q_{cg} = Q_l + Q_{wg} \quad (52)$$

Combining equations (51) and (52), the total heat balance around the panel and gap is given by:

$$Q_w + Q_{wg} = Q_{cg} + Q_e \quad (53)$$

The effective conductivity of the panels containing rectangular or wedge shaped gaps or radiating to an infinite wall was determined by the following equations:

For the rectangular gap:

$$K_e = \frac{2[(Q_{wg}) + (Q_w)] h}{(W + d) \Delta t} \quad (54)$$

For the wedge gap and exposed edge:

$$K_e = \frac{(Q_w) h}{W \Delta t} \quad (55)$$

where:

h = panel thickness, ft.
W = panel width, ft.
d = gap width, ft.
 Δt = °R.

In the wedge gap cases, W refers to panel width at the warm side. In Table B-I, Appendix B, the uneffected conductivity (K_o) was determined by cases 20-22 which had zero gap width. The effected conductivity and the ratio of the effected conductivity to the uneffected conductivity for all cases also appear in the Table.

The ratio of effected to uneffected conductivity for unfilled gaps has been plotted in Figures 23-30 and Figures B1-B14, Appendix B-I. Panel width

EFFECT OF PANEL WIDTH ON THERMAL CONDUCTIVITY
FOR A GAP WIDTH OF .02 INCH.

 K_e/K_o

FIGURE 23

EFFECT OF PANEL WIDTH ON THERMAL CONDUCTIVITY
FOR A GAP WIDTH OF .05 INCH.

FIGURE 24

×

LT 80

FIGURE 25

EFFECT OF PANEL WIDTH ON THERMAL CONDUCTIVITY
FOR AN ANGLE OF 10 DEGREES

- = 0.1 inch panel thickness
□ = 0.2 inch panel thickness
X = 0.4 inch panel thickness

K_e/K_o

FIGURE 26

EFFECT OF PANEL WIDTH ON THERMAL CONDUCTIVITY
FOR AN ANGLE OF 30 DEGREES

EFFECT OF PANEL WIDTH ON THERMAL CONDUCTIVITY
FOR AN ANGLE OF 60 DEGREES

 K_e/K_o

FIGURE 28

EFFECT OF PANEL WIDTH ON THERMAL CONDUCTIVITY
FOR AN ANGLE OF 90 DEGREES

 K_e/K_o

- = 0.1 inch thickness
- = 0.2 inch thickness
- × = 0.4 inch thickness
- ⊗ = 0.4 inch thickness
filled gap

FIGURE 29

EFFECT OF PANEL WIDTH ON THERMAL CONDUCTIVITY
FOR A PANEL WITH EDGES RADIATING TO SPACE

- ⊙ = 0.1 inch panel thickness
□ = 0.2 inch panel thickness
× = 0.4 inch panel thickness

K_e/K_o

FIGURE 30

is probably the most important parameter and, therefore, Figures 23-30 would probably be the most useful for design. The data is also cross plotted against gap width, panel thickness, and angle in Appendix B, Figures B1-B14. As can be seen from the graphs, the least effect on the conductivity is obtained with long, thin panels with a small gap or angle. Conversely, the worst effect is obtained with short, thick panels having a large gap or angle.

The addition of a filler in the gaps greatly improved even the worst of the cases investigated. The worst gap cases appear on Figures 25 and 29. Also plotted on these two graphs are the results for the thickest panels with gaps filled. The base conductivity is at most affected by 40% and with panels 10 inches or more wide, the effect is 10% or less.

An investigation was made to determine the effect on panel thermal conductivity of exposed panel edges viewing an infinite wall at several levels. Virtually no effect was seen as the wall temperature varied from 0°R. to 400°R. (e.g. See Table B-I, Appendix B, Cases 23, 26, 29; Cases 24, 27, 30; and Cases 25, 28, and 31.) This is due to the fourth power temperature dependence of the radiation which overwhelms the relatively low wall temperatures. Figure 30 shows the ratio of conductivities for various panel widths and thicknesses for this exposed edge case.

The temperature profiles that appear in Figures B-15 - B-19 (Appendix B) are useful for understanding the methods of heat transfer within the insulation as effected by the gap or edge. Figure B-15 shows that even at the center of 25 inch wide panels at a 90° angle, there is a considerable distortion of the temperature profile. On the other hand, Figure B-16 shows that little distortion occurs in 25 inch wide panels separated by a rectangular gap. Figure B-17 gives a comparison of a wedge and a rectangular gap. Little distortion occurs with a rectangular gap and considerable distortion occurs with the wedge gap. Added loss in the rectangular gap case are primarily due to radiation straight through the gap while the wedge gap losses are due to lateral transfer through the panels and radiation from the edges. This mechanism is illustrated more clearly in Figures B-18 and B-19. Figure B-18 shows that large lateral gradients exist throughout the thickness of panel so that heat is continually flowing towards the edge. Figure B-19 shows that much smaller gradients exist in the rectangular gap problem (note the broken and expanded scale on the plot). Also note that heat is actually being fed into the panel from the gap near the cold side of the panel. This effect can also be seen by comparing the magnitudes of Q_e for the two cases (61 and 91) in Table B-I, Appendix B.

From the results of this study, the following conclusions can be drawn:

1. Gaps and edges will not have an overwhelming influence on panel thermal performance if thin, wide panels are used with small gaps or angles. For example, a panel 50 inches wide, 0.1 inch thick with a 0.05 inch gap displayed an effected to uneffected thermal conductivity ratio of only 1.098.

2. The greatest degradation of thermal conductivity occurs with short, thick panels having a large gap or angle between panels.

3. The addition of a modest filler to gaps can greatly reduce the effected thermal conductivity of an insulation panel.

3.3.3 Air Exposure of Vacuum Insulations

Although high temperature Super Insulation requires a vacuum environment for optimum thermal performance, in space applications the vacuum is automatically provided and, therefore, a vacuum-tight enclosure is not needed. However, many of these applications involve radioisotope power systems that must be fueled on the ground and a considerable hold time elapses during which the insulation is hot before it is exposed to the low pressures of space. Since some of the metallic foils in insulation systems have relatively poor oxidation resistance, oxide will form on the foils. Depending on the amount of oxide on the foils, the reflectivity and, thus, the thermal performance may or may not be affected. If affected, however, it may be possible to remove the oxide and thus negate any adverse effects. It was the object of this study to investigate the oxidation rate relations, the decomposition, vaporization, and reduction possibilities, and the actual effect of an oxide on the surface of a foil. Consequently, this study was divided into four parts:

- 1). Determination of the ground hold temperatures that would be expected in typical insulation systems.
- 2). Investigation of the temperature-time-thickness of oxide parameters of the oxidation process.
- 3). Study of the possibilities of reduction, vaporization, or decomposition of the oxide.
- 4). Literature search for experimental data, and the development of analytical methods to predict the effect of oxide on the reflectivity of metallic foils.

3.3.3.1 Air Exposure Temperature Profile

In order to derive the equations governing the insulation system temperature profile under air exposure, we let Q_T = total heat produced by the generator, Btu/hr.; Q_S = heat passing through the thermoelectric elements and through supports in vacuum; Q_I = heat passing through the insulation in vacuum. If we assume that in vacuum ϕ part of the heat passes through the insulation, we can write:

$$\frac{Q_S}{Q_T} = 1 - \phi ; \frac{Q_I}{Q_T} = \phi$$

Also in vacuum, if T_1 , T_2 , T_3 , and T_4 are the boundary and interface temperatures in a three component system, (See Figure 31) we can write for purely one-dimensional conditions:

$$Q_I = \frac{A k_A (T_1 - T_2)}{X_A} = \phi Q_T \quad (56)$$

where:

- A_A = average area of insulation, ft.²
 k_A = thermal conductivity of first insulation section, Btu/hr.-ft.-°F.
 X_A = thickness of first insulation section, ft.

but

$$X_A = x_A \ell \quad (57)$$

where:

- ℓ = total insulation thickness, ft.
 x_A = first insulation section fraction of total thickness

Based on equations (56) and (57) an expression describing the thermal performance of an insulation system during air exposure may be derived:

$$\frac{\phi x_A^a}{2k_A (T_1 - T_2)} (T_H^2 - T_L^2) + \left[\frac{\phi x_A^b}{k_A (T_1 - T_2)} + \frac{(1 - \phi)}{(T_1 - T_4)} \right] (T_H - T_L) = 1 \quad (58)$$

a and b are constants describing the temperature dependence of the thermal conductivity of air. All quantities in equation 58 are known except T_H and T_L . One more equation or boundary condition must be known. If the outside area of the insulated system is large, or the generator output is small, or the outside is forced cooled during air exposure, then the outside temperature is equal to the ambient temperature, T_A ; therefore:

$$T_L = T_A \quad (59)$$

and equation 58 can be solved for T_H . If, however, Q_T is large, or the outside boundary is small, T_L will be greater than T_A . This condition is described by:

$$\frac{a}{2} T^2 + b T = \frac{a}{2} T_H^2 + b T_H - \frac{x}{1} \left[\frac{a}{2} (T_H^2 - T_L^2) + b (T_H - T_L) \right] \quad (60)$$

With T_H and T_L known from the previous analysis, equation 60 gives an equation in terms of T and $\left(\frac{x}{1}\right)$.

Details of these derivations are presented in Appendix C.

Two typical insulation systems were investigated. One system was a composite of nickel-astroquartz cloth (Section A), copper-Refrasil quartz

AIR EXPOSURE NOMENCLATURE

paper (Section B), and aluminum-glass paper (Section C.)

With boundary temperatures $T_1 = 1800^\circ\text{F.}$, $T_2 = 1270^\circ\text{F.}$, $T_3 = 900^\circ\text{F.}$, $T_4 = 700^\circ\text{F.}$, the fractions of each type were determined to be $x_A = 0.88$, $x_B = 0.11$, and $x_C = 0.01$. The corresponding thermal conductivities between the prescribed temperatures were $k_A = 5.99 \times 10^{-3}$ Btu/hr.ft. $^\circ\text{F.}$, $k_B = 1.07 \times 10^{-3}$ Btu/hr.ft. $^\circ\text{F.}$, and $k_C = 0.175 \times 10^{-3}$ Btu/hr.ft. $^\circ\text{F.}$ In order to determine ϕ , fraction of heat passing through the insulation under vacuum conditions, the exact insulation geometry and heat flux fractions chargeable to the insulation, supports, and thermoelectric elements must be known. Since detailed calculations of this sort are beyond the scope of this study, an assumption was made, based on known radioisotope power system designs, that 5% of the total heat flux passed through the insulation during vacuum operation ($\phi = 0.05$). With this 1800 - 700 $^\circ\text{F.}$ system, two types of outside boundary conditions were investigated. First, the outside boundary was assumed to be at ambient temperature, $T_A = 70^\circ\text{F.}$ Equation 58 and C-41 were then used to calculate the temperature profile. The second condition was for a 7000 watt generator (total isotope output) with 10 square feet outside area. This is believed to be an extreme but possible application. The outside surface of the generator system was assumed to be essentially black so that $\epsilon = 1$. T_L was obtained by heat balance (see Appendix C). Equation 52 was used to obtain T_H , and equation 54 to give the profile. The vacuum profile and the two air exposure profiles appear in Figure 32.

The second insulation system to be investigated consisted entirely of aluminum and glass paper with $T_1 = 800^\circ\text{F.}$ and $T_2 = 200^\circ\text{F.}$ so that $x_A = 1.0$. The conductivity between these temperatures was determined to be 0.122×10^{-3} Btu/hr.ft. $^\circ\text{F.}$ Again ϕ was assumed to be 0.05. Two types of boundary conditions were also investigated for this case. The first condition was an ambient boundary as before while the second was for a 700 watt generator with 5 square feet outside area. The second condition also represents an extreme but possible condition. The vacuum and air exposure profiles for this 800 - 200 $^\circ\text{F.}$ system are shown in Figure 33.

3.3.3.2 Oxidation Rate Relations

Based on the operating temperatures to be expected during air exposure, a study was initiated to investigate the oxidation rate vs. air exposure time characteristics of the metal foils used in insulation systems. Nickel, copper, and aluminum were the metals of primary interest. Platinum and gold were also investigated as possible back-up materials.

The oxidation rate of nickel was obtained from Kubaschewski⁴². The parabolic rate constant is given as:

$$400 - 850^\circ\text{C} \quad (752 - 1562^\circ\text{F})$$

$$K_p = 8 \times 10^{-4} e^{(-41,200/RT)}$$

Figure 32

HOT AND COLD BOUNDARY TEMPERATURE, °F

Figure 33

900 - 1050°C (1652-1922°F)

$$K_p = 1.2 \times 10^{-2} e^{(-68,300/RT)}$$

where T is in °K and K_p is in $g^2 cm^{-4} sec^{-1}$ and R is the universal gas constant. The K_p was changed from weight gain units into oxide thickness units by assuming that the oxide layer had the density of the pure oxide. The conversion constants calculated were:

NiO	159.5 $\frac{\mu g/cm^2}{\mu Oxide}$
CuO	178.3 $\frac{\mu g/cm^2}{\mu Oxide}$
Al ₂ O ₃	174.2 $\frac{\mu g/cm^2}{\mu Oxide}$

Figure 34 presents the square of the nickel oxide thickness as a function of the exposure time.

Tylecote⁷³ compiled data and presented a plot of the parabolic rate constant k as a function of temperature for copper. We found that this data could be closely predicted by the following equation in the range 100 - 980°F.

$$\log k = -3933/T - 2.2$$

where T is in °K and k is in $g^2 cm^{-4} hr^{-1}$. This equation was used to generate Figure 35.

Smeltzer⁷⁰ gave plots of the oxidation rate constant for aluminum. Calculations were made directly from these graphs to generate Figure 36 for the oxidation of aluminum. As Figure 36 shows, although a thin oxide layer forms quickly on the aluminum, the layer is impervious so that further oxidation is controlled by the diffusion of oxygen through the aluminum oxide. The rate of oxidation decreases sharply.

Two oxidation resistant metals that could be used if severe oxidation conditions are expected are platinum and gold. Even at 1700°F, (which as Figure 32 shows is much higher than any expected air exposure temperature), platinum is extremely oxidation resistant. An oxide is formed at this temperature but immediately vaporizes such that the platinum loses weight at the rate of 0.24 mg/(dec)² day. During a 30 day exposure at this temperature, the weight loss is equivalent to 0.00232 mil which is only about 1% of the total foil thickness. Other metals in the platinum group, e.g. rhodium, have similar properties.

Oxide Thickness as a Function of Air Exposure
Time for Nickel Foil

Figure 34

Oxide Thickness as a Function of Air Exposure
Time for Copper Foil

Figure 35

Oxide Thickness as a Function of Air Exposure
Time for Aluminum

Figure 36

Gold does not oxidize in air⁷⁴. The factor that determines its maximum operating temperature is its vaporization rate. Calculations based on data from Dushman¹¹ show that for a five year life with one mil foil, the maximum temperature of operation is 1500°F. This temperature is based upon the maximum possible vaporization rate under these conditions. Actually, the rate is less due to a dampening effect of the surrounding insulation. Gold, therefore, probably can be used at temperatures greater than 1500°F.

3.3.3.3 Restoration of Foil Surfaces

The three ways of removing the oxide and thus returning the foil surface to its original metallic condition are vaporization, reduction, and decomposition. Each of the three metals of importance (copper, nickel, aluminum) will be discussed in regard to the three possibilities for restoration of the surface. It should be noted that data for some of these mechanisms are non-existent or available only for conditions different than those encountered in the insulation system.

Copper - Copper oxide could possibly be reduced to pure copper by the offgassing products of the insulation which is predominantly hydrogen. Reduction rate data at low pressures could not be obtained. However, the rate can be inferred from differential thermal analysis data of the oxide. Figure 37, taken from Keely³⁷, shows a DTA curve for copper oxide being reduced by hydrogen at a heating rate of 5 - 6°C/Min. From this curve we can infer that above 340°C. (644°F.), the reduction of copper oxide by hydrogen proceeds rapidly. However, this reference only tells us that the reaction proceeds rapidly when a liberal supply of reducing gas is present. In the space environment, the hydrogen is supplied at the insulation offgassing rate. If G_S is the hydrogen offgassing rate in micron liters per hour per foot², then $G_S / (760,000)(22.4)$ g mole/hr.ft.² of hydrogen is produced. For monoxides, the following reaction occurs.

Therefore, $G_S / (760,000)(22.4)$ g mole/hr.ft.² of oxide is consumed. Converting this into an oxide thickness, we find that the rate of reduction is

$$\phi = 6.32 \times 10^{-7} \frac{G_S M_O}{\rho_O} \quad (61)$$

where:

$$\begin{aligned} \phi &= \text{rate of reduction (}\mu\text{Oxide/hr.)} \\ M_O &= \text{gram molecular weight of the oxide} \\ \rho_O &= \text{density of the oxide, (gm/cm}^3\text{)} \end{aligned}$$

Using a G_S of 1.0 μ l/hr.ft.² and the molecular weight and density of Cu_2O , we find that $\phi = 1.51 \times 10^{-5}$ μ /hr. Thus, the rate will be controlled by the rate of offgassing. From Figure 32 we find that the maximum exposure temperature

HEATING CURVE

COOLING CURVE

STOP POINTS

REDUCTION OF COPPER OXIDE

HEATING RATE = 5-6° C/MIN

for copper is 650°F. If we assume a maximum ground hold time of 30 days, Figure 35 shows that about 10 microns of oxide will have formed. With a $\dot{O}$ of 1.51×10^{-5} μ /hr., we can then calculate that approximately 75.6 years would be required to reduce the oxide. However, if the cold boundary is at 70°F. during air exposure, Figure 32 shows that the exposure temperature will be only 200°F. If the ground hold time is assumed to be one day, from Figure 35 only 0.013 microns of oxide would be formed. This oxide layer would be removed by reduction in 35 days. Therefore, reduction might be of some use in removing the oxide. Of course, the relatively thin oxide layer, 0.013 microns, might not have an effect on the reflectivity and in this event the hydrogen reduction possibilities might allow the exposure time to be lengthened.

Another possibility for the removal of copper oxide is decomposition. At 1270°F., we can calculate from Dushman¹¹ that at 0.0316 torr of oxygen, cupric oxide (CuO) will transform to cuprous oxide (Cu_2O). This transition would probably not be particularly beneficial but at 10^{-8} torr of oxygen, cuprous oxide will transform to pure copper. The main source of oxygen in applications involving space vehicles, is the oxygen in the atmosphere. Data from Koelle⁴¹ was plotted in Figure 38 to determine the partial pressure of atmospheric gases as a function of altitude. As can be seen, the oxygen pressure is below 10^{-8} torr at approximately 150 miles. Therefore, if the mission height is greater than 150 miles, the oxide should decompose. However, the rate of this decomposition is important. In studying the reduction of the oxide by hydrogen, we came to the conclusion that the reduction should proceed rapidly if the temperature was above a critical temperature (644°F.) and if a plentiful supply of hydrogen was present. Since decomposition is another form of reduction, it seems probable that if the oxygen pressure is below a critical value (10^{-8} torr at 1270°F.), then the decomposition should proceed rapidly. Vapor pressure data for copper oxide could not be obtained but the stability of the oxide suggests that its vapor pressure is probably much lower than for the parent metal.

Nickel - Many of the same comments that applied to the reduction of copper oxide also apply to the reduction of nickel oxide. The DTA curve of nickel oxide being reduced by hydrogen taken from Keely and Maynor³⁷ appears in Figure 39. Note that above about 320°C. (608°F.) the reaction is essentially complete. The reduction is therefore rapid if the temperature is above 608°F. and if a plentiful supply of hydrogen is available. However, as with copper, the rate is governed by the offgassing rate of the insulation. Taking an offgassing rate, $G_s = 1.0$ μ l/hr.ft.² and the molecular weight and density of NiO, equation 56 gives a reduction rate of 6.34×10^{-6} μ Oxide/hr. From Figure 32, we find that the maximum exposure temperature for nickel is 1300°F. A 30 day exposure at 1300°F. gives 6.63 microns of oxide (Figure 34). With a rate of 6.34×10^{-6} μ oxide/hr., the reduction time is 119 years. A one-day exposure at 900°F. gives 0.046 microns which requires 10 months. Reduction therefore seems of little value for nickel.

From Dushman¹¹, we find that NiO will decompose at 1640°F. if the oxygen pressure is below 10^{-9} torr. From Figure 38 we find that decomposition will occur for missions greater than 250 miles in altitude. As with copper, it is expected that decomposition would proceed rapidly.

ATMOSPHERIC COMPOSITION AS A FUNCTION OF HEIGHT

FIGURE 38

REDUCTION OF NICKEL OXIDE

Figure 40, taken from Wong⁷⁹, shows that at 900°C. (1652°F.), NiO evaporates at a rate of 3×10^{-4} A/sec. (3×10^{-8} g/sec.). For the 6.63 microns of oxide (30 days at 1300°F.), the time for vaporization would therefore be seven years. For the 0.046 microns (1 day at 900°F.), the time is 18 days. Therefore, vaporization shows some promise for thin oxide layers.

Aluminum - Reduction, decomposition, or vaporization data was not found for aluminum. This is probably because aluminum shows such good oxidation resistance that there has been no incentive to obtain such data. The effect of the thin oxide layer formed will be investigated later in the report.

To summarize this section, it appears that the best hope for restoration of the metallic surface lies in the decomposition of the oxides of nickel and copper at high altitudes. At lower altitudes, the oxide may be removed by reduction from the offgassing or from vaporization if the air exposure conditions have not been too severe. It now remains to determine how much oxide can be tolerated before the thermal performance is affected.

3.3.3.4 Effect of Oxide on Reflectivity

There has been very little experimental data generated on the effect of an oxide on the reflectivity of a metallic surface at elevated temperatures. Some data was obtained at or near room temperature^{24, 25, 27} but it is difficult to generalize this data to higher temperatures because at these elevated temperatures, the emission of the oxide due to its temperature becomes important. Some qualitative data^{77, 20} is available for surfaces with "light oxide" or "heavy oxide". The only quantitative experimental data located was by Shelton and Akers⁶⁸ and Oubre⁶¹ for nickel oxide on nickel. A considerable number of theoretical studies*, have been made concerning the effect of a coating on the radiation properties of a surface. However, most of these studies were directed toward solar absorptance and none derived the solution for the problem of interest. It was therefore necessary to develop a correlation to predict the effect of oxide on the reflectivity of a metallic surface at elevated temperature. The basis of the correlation is summarized as follows:

Experimental data was found for the effect of oxide on reflectivity for nickel. From fundamental differential equations for radiation in absorbing and scattering media, an equation was developed to predict the effect of oxide on reflectivity from knowledge of the absorption and scattering coefficients of the media. Next, an equation was developed to relate the ratio of the absorption to the scattering coefficient to the emissivity of the pure oxide. These equations were then used to extract the absorption and scattering coefficients as a function of temperature from experimental data for oxidized nickel. Using the scattering coefficient of nickel oxide as being representative of the scattering coefficient of aluminum and copper oxide, absorption coefficients were calculated for aluminum and copper oxide. Using these calculated coefficients, the effect of oxide on reflectivity for copper and aluminum at several temperatures was calculated. This correlation is presented in

* References 4, 15, 16, 17, 21, 23, 29, 36, 40, 43, 48, 50, 51, 52, 64, 75, 76

detail in Appendix C.

The reflectivities of nickel oxide, copper oxide and aluminum oxide as a function of oxide thickness are shown in Figures 41 through 43, respectively.

3.3.3.5 Thermal Conductivity of an Oxidized Insulation System

Section 3.3.3.4 gave a method for predicting the reflectivities of foils in an insulation system that has been exposed to air under specified conditions. This section shows how to use this data to calculate the thermal conductivity in vacuum of a Super Insulation system that has been exposed to air.

The thermal conductivity of Super Insulation at any temperature is given by the following equation:

$$k = aT + bT^3 \quad (62)$$

where:

k = thermal conductivity, Btu/hr-ft.°R.
 T = temperature, °R.

The constants a and b are determined from the intercept ($\frac{a}{2}$) and slope ($\frac{b}{4}$) of a plot of $k_a (T_1 - T_2) / (T_1^2 - T_2^2)$ vs. $(T_1^4 - T_2^4) / (T_1^2 - T_2^2)$. k_a is the mean conductivity between temperatures T_1 and T_2 (Section 3.3.1.1). In terms of fundamental parameters, b can be written as:

$$b = \frac{2\sigma\epsilon}{n} \quad (63)$$

where:

σ = Stefan-Boltzmann constant
 ϵ = foil emissivity
 n = layer density

After an insulation system has been exposed to air, the emissivity of each foil is different due to the presence of different amounts of oxide. Thus, a plot of $k_a (T_1 - T_2) / (T_1^2 - T_2^2)$ vs. $(T_1^4 - T_2^4) / (T_1^2 - T_2^2)$ will not yield a straight line. However, the system can still be defined mathematically if the emissivities and temperatures of the foils are known.

Under known exposure conditions, the amount of oxide on each foil can be determined from the graphs presented in the oxidation section. In order to determine the reflectivities of the foils, the temperature profile during vacuum operation must be known. This profile is, of course, changed from the unexposed case due to the reduced reflectivities of the foils. An iterative approach could be used to circumvent this problem. For example, the profile could just be assumed equal to that of an unexposed system, the reflectivities could be obtained by the methods of Section 3.3.3.4, the new

MAXIMUM RATE OF EVAPORATION PLOTTED AGAINST TEMPERATURE

REFLECTIVITY OF OXIDIZED NICKEL AS A FUNCTION OF OXIDE THICKNESS

FIGURE 41

REFLECTIVITY OF OXIDIZED COPPER AS A FUNCTION OF OXIDE THICKNESS

FIGURE 42

FIGURE 43

profile could then be calculated, and the process repeated until the calculated profile was identical to the assumed profile. However, exploratory calculations revealed that the profile for an exposed system was similar to that of an unexposed system so that the reflectivities could be calculated from the unexposed profile with little error involved. Therefore, the reflectivities can be obtained from the unexposed temperature profiles.

An empirical equation can be used to describe the emissivity (obtained from reflectivity) - Temperature relationship existing in a given exposed insulation system operating in vacuum. Denote this function by $\epsilon(T)$.

If we let

$$B = \frac{2\sigma}{n} \quad (64)$$

then 63 can be written as:

$$b = B\epsilon(T) \quad (65)$$

Substituting 65 into 62, we obtain:

$$k = aT + B\epsilon(T) T^3 \quad (66)$$

We can also write

$$q = ka \frac{dT}{dx} \quad (67)$$

Integrating 61, we obtain:

$$q \int_{T_1}^{T_2} \frac{dx}{k} = \int_{T_1}^{T_2} \frac{dT}{aT + B\epsilon(T) T^3}$$

$$q = \frac{A}{L} \int_{T_1}^{T_2} [aT + B\epsilon(T) T^3] dT$$

$$q = \frac{A}{L} \frac{a}{2} [(T_1^2 - T_2^2) + B \int_{T_1}^{T_2} \epsilon(T) T^3 dT] \quad (68)$$

where:

L = thickness of insulation

Another expression for q is:

$$\dot{q} = k_{ae} A (T_1 - T_2) / L \quad (69)$$

where:

k_{ae} = mean thermal conductivity after air exposure.

Equating 68 and 69 and solving for k_{ae} , we obtain:

$$k_{ae} = \frac{a}{2} \frac{(T_1^2 - T_2^2)}{(T_1 - T_2)} + \frac{B}{(T_1 - T_2)} \int_{T_2}^{T_1} \epsilon(T) T^3 dT \quad (70)$$

If, for example, the function $\epsilon(T)$ is a polynomial of degree n so that

$$\epsilon(T) = \sum_{i=0}^n C_i T^i \quad (71)$$

then 70 becomes

$$k_{ae} = \frac{a}{2} \frac{(T_1^2 - T_2^2)}{(T_1 - T_2)} + \frac{B}{(T_1 - T_2)} \int_{T_2}^{T_1} \sum_{i=0}^n C_i T^{i+3} dT \quad (72)$$

$$k_{ae} = \frac{a}{2} \frac{(T_1^2 - T_2^2)}{(T_1 - T_2)} + \frac{B}{(T_1 - T_2)} \sum_{i=0}^n \frac{C_i}{i+4} (T_1^{i+4} - T_2^{i+4}) \quad (73)$$

We can derive an equation for the temperature profile in vacuum of the air exposed system by integrating to a point x from the hot side, similar to the derivation of the air exposed profile in Section 3.3.3.1. For a polynomial of degree n , the equation for the profile is:

$$\frac{a}{2} T^2 + B \sum_{i=0}^n \frac{C_i}{i+4} T^{i+4} = \frac{a}{2} T_1^2 + B \sum_{i=0}^n \frac{C_i}{i+4} T_1^{i+4}$$

$$- \frac{x}{1} \left[\frac{a}{2} (T_1^2 - T_2^2) + B \sum_{i=0}^n \frac{C_i}{i+4} (T_1^{i+4} - T_2^{i+4}) \right] \quad (74)$$

We can check the k_{ae} calculated by (73) with the k measured on the copper foil-quartz paper combination after air exposure. Table XIV gives the exposure times, hot and cold face temperatures, and conductivities for each of the four exposures. The exposures temperatures within the insulation were calculated by assuming a linear profile during air exposure. The oxide thickness after the i th exposure, was obtained from Figure 35. The temperatures in vacuum after each air exposure were taken from the profile for the unexposed case given in Figure 44. The reflectivity was obtained from Figure 42. A plot of the emissivity-temperature relationship for the oxidized system operating in vacuum is given in Figure 45. A least squares curve was fit to this data on the IBM 360 computer. k_{ae} was calculated from (73) for each exposure. The k_a (unexposed conductivity) is also tabulated.

The temperature profile within the oxidized tester was calculated by equation (74) and is given in Figure 44. As assumed earlier, the oxidized profile is similar to the unoxidized profile.

If we assume that the foils are turned into black bodies ($\epsilon=1$) by the oxidation process, we can obtain an upper limit for the conductivity of an oxidized system. This assumption is conservative because the emissivity approaches that of the pure oxide which is less than 1. Between the temperature limits 1755°F. and 392°F., the black body conductivity, k_B , was calculated to be 11.35×10^{-3} Btu/hr.ft.°F. Thus, even a completely oxidized system has a reasonably low conductivity. The ratio $k_{ae} - k_a / k_B - k_a$ gives an indication of the relative degree of oxidation of the system. For the oxidized tester, this ratio is 0.272.

To summarize; in order to determine the effect of air exposure on the thermal conductivity of an insulation system, the following procedure should be used.

- 1). Determine the insulation temperature profile during air exposure using the methods presented in Section 3.3.3.1.
- 2). With the temperatures obtained, use Figures 34, 35 and 36 to determine the thickness of oxide that will be present on the foils after the expected exposure time. This should be repeated for several locations within the insulation system thermal gradient.
- 3). Depending on the altitude of the mission and the time specified to reach maximum thermal efficiency, use the information in Section 3.3.3.3 to predict whether or not the oxide will dissociate, vaporize, or be reduced.
- 4). If the conditions of the mission are such that the oxide will not be removed within the necessary time, use Figures 41, 42, and 43 to determine the emissivity - temperature relationship that will exist in the oxidized system during vacuum operation. Describe this relationship mathematically by fitting a curve to the emissivity - temperature data.

TABLE XVI

EXPERIMENTAL AND CALCULATED DATA
ON AIR-EXPOSED COPPER FOIL

		<u>Exposure No. 1</u>		<u>Exposure No. 2</u>		<u>Exposure No. 3</u>		<u>Exposure No. 4</u>		<u>Total Oxide</u>	<u>In Vacuum After Exposure No. 4</u>		
		<u>Temp</u>	<u>Oxide Thickness</u>	<u>Temp</u>	<u>Oxide Thickness</u>	<u>Temp</u>	<u>Oxide Thickness</u>	<u>Temp</u>	<u>Oxide Thickness</u>		<u>Temp</u>	<u>Reflec- tivity</u>	<u>Emis- sivity</u>
<u>x/l</u>		<u>T₁</u>	<u>D₁</u>	<u>T₂</u>	<u>D₂</u>	<u>T₃</u>	<u>D₃</u>	<u>T₄</u>	<u>D₄</u>	<u>D</u>			
		<u>(°F)</u>	<u>(Micron)</u>	<u>(°F)</u>	<u>(Micron)</u>	<u>(°F)</u>	<u>(Micron)</u>	<u>(°F)</u>	<u>(Micron)</u>	<u>(Micron)</u>	<u>(°F)</u>		
Hot Face	0	223	0.02	342	0.1643	380	0.2098	371	0.3082	0.7023	1295	0.54	0.46
	0.2	207	0.0164	335	0.1095	362	0.1581	367	0.2345	0.5185	1190	0.56	0.44
	0.4	192	0.0114	304	0.0678	329	0.1049	334	0.1549	0.3390	1070	0.60	0.40
	0.6	178	0.0089	272	0.0469	295	0.0608	300	0.0894	0.2060	920	0.66	0.34
	0.8	155	0.0077	240	0.029	262	0.0424	266	0.0603	0.1399	720	0.67	0.33
Cold Face	1.0	149	0.0071	207	0.0148	234	0.0245	234	0.0361	0.0825	392	0.58	0.42

T_i and D_i are the temperature and oxide thickness formed during the i th exposure. $D = \sum_{i=1}^4 D_i$.
 T is the operating temperature in vacuum.

First, second and third exposures were for 24 hours each. Exposure No. 4 was for 48 hours.

VACUUM TEMPERATURE PROFILE - COPPER FOIL - REFRASIL QUARTZ

FIGURE 44

EMISSION VS. TEMPERATURE PARABOLIC TUNE FIT -
COPPER FOIL - REFRASIL QUARTZ FIBER PAPER

FIGURE 45

5). Use equation 67 to calculate the conductivity of the oxidized insulation system.

3.3.3.6 Copper Foil-Refrasil Quartz Fiber Paper Combination Air Exposure Experiments

The objective of this work was to experimentally evaluate the effects of air exposure during typical ground hold conditions on the thermal performance of a copper foil - Refrasil paper insulation combination. After air exposure, the effect of oxidation on the copper foils was determined by re-evaluating the thermal conductivity in vacuum. Guidelines for selecting the operating conditions, temperature, and exposure times for the experiments were determined from Sections 3.3.3.4 and 3.3.3.5.

This test apparatus was revised for air exposure experiments by replacement of the existing tungsten heater with a nickel-chromium alloy heater capable of air exposure (Section 3.3.1.5.3). This was accomplished by disassembling the top head of the insulated tester, removing the tungsten heater, and replacing it with a nickel-chromium alloy heater.

A new base point thermal conductivity was established for this combination in vacuum, at a hot face temperature of 1292°F., and a cold face temperature of 297°F. A thermal conductivity of 1.18×10^{-3} Btu/hr-ft-°R. was obtained. This value is nearly 70% higher than the thermal conductivity obtained from the original test data (Section 3.3.1.5.3). This increased thermal conductivity is attributed to damage of the insulation that occurred during revision of the heater. This new base conductivity was used as a reference point to determine the effects of air exposure on vacuum thermal conductivity.

Testing was conducted by cooling the insulation combination in vacuum to the desired air exposure temperature and then exposing to air for the specified time period. At the conclusion of the air exposure period, the tester was evacuated to less than 0.1 micron mercury and the thermal conductivity re-evaluated at the base point conditions.

Air exposure times, temperature, and thermal conductivity of the insulation in air, together with the thermal conductivity measured in vacuum, after each exposure, are presented in Table XVI.

Four separate air exposure experiments were conducted, with the last exposure of 48 hours duration. The thermal conductivity of the insulation in air was measured and found to average about 79% greater than the thermal conductivity of air itself, the difference attributed to solid conductivity and radiation transport of the insulation. This copper foil-Refrasil quartz paper wrap was applied at a relatively low layer density while the copper foil-copper opacified Refrasil quartz paper wrap (Section 3.3.1.5.4) was applied at about its maximum layer density. The greater deviation of the thermal conductivity of the copper foil-Refrasil paper wrap from the thermal conductivity of air, 79%, may also be due to gaseous convection within the insulation itself. The thermal conductivity in vacuum was seen to increase

after each air exposure, indicating that an oxide layer was being formed on the foils with subsequent increase in foil emissivity. The vacuum thermal conductivity increased by a factor of about 2 over the base vacuum conductivity as a result of the exposure experiments. It is significant to note that if the thermal conductivity of a copper foil-Refrasil quartz fiber paper insulation system initially operating between 1292 and 297°F. is increased by a factor of 2 due to air exposure, it is still about 10% lower than the thermal conductivity of the next most efficient insulation system capable of operation in this temperature range, nickel foil-Refrasil paper.

Copper foil-Refrasil Paper
unexposed

1292°F

$$\begin{aligned} \text{(Section 3.3.1.5.3)} \quad k/_{297^\circ\text{F}} &= 0.095 \times 10^{-6} \left(\frac{T_1^2 - T_2^2}{T_1 - T_2} \right) + \\ &+ 0.52 \times 10^{-13} \left(\frac{T_1^4 - T_2^4}{T_1 - T_2} \right) = 0.712 \times 10^{-3} \end{aligned} \quad (75)$$

Copper foil-Refrasil Paper
exposed

$$k = (2.1) (0.712 \times 10^{-3}) = 1.47 \times 10^{-3} \quad (76)$$

Nickel foil-Refrasil Paper
unexposed
(Section 3.3.1.5.6)

$$\begin{aligned} k/ &= 0.2 \times 10^{-6} \frac{1292^\circ\text{F}}{\left(\frac{T_1^2 - T_2^2}{T_1 - T_2} \right) +} \\ &+ 1.23 \times 10^{-13} \left(\frac{T_1^4 - T_2^4}{T_1 - T_2} \right) = 1.62 \times 10^{-3} \end{aligned} \quad (77)$$

TABLE XVIIEFFECT OF AIR EXPOSURE ON A COPPER FOILREFRASIL QUARTZ FIBER PAPER INSULATION COMBINATION

<u>Test Condition</u>	<u>Hot Face Temperature, °F</u>	<u>Cold Face Temperature °F</u>	<u>Thermal Conductivity Btu/hr.ft.°R x 10⁻³</u>
Base Point in Vacuum	1292	297	1.18
Air Exposure No. 1-24 hrs.	223	149	31.20
In Vacuum After Air Exposure No. 1	1272	302	1.23
Air Exposure No. 2-24 hrs.	342	207	34.80
In Vacuum After Air Exposure No. 2	1287	356	1.77
Air Exposure No. 3-24 hrs.	380	234	-
In Vacuum After Air Exposure No. 3	1280	374	2.14
Air Exposure No. 4-48 hrs.	371	233.6	39.7
In Vacuum After Air Exposure No. 4	1295	392	2.46

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3.3.3.7 Nickel Foil-Refrasil Quartz Fiber Paper Combination Air Exposure

The objective of this work is to experimentally evaluate the effects of air exposure, during typical ground hold conditions, on the thermal performance of a nickel foil - Refrasil quartz fiber paper insulation combination (Ni-Refrasil). After air exposure, the effect of air exposure (oxidation) on the nickel foils is determined by re-evaluating the thermal conductivity in vacuum.

Air exposure times, temperatures, and thermal conductivity of the nickel foil - Refrasil quartz fiber paper insulation in air, together with the thermal conductivity measured in vacuum, after each exposure are presented in Table XVIII, Columns 1-3. Also included in Table XVIII for comparative purposes are the previous data from air exposure testing of the copper foil - Refrasil quartz fiber paper insulation.

The thermal conductivity of the nickel foil - Refrasil quartz fiber paper combination in air was measured and found to average about 43% greater than the thermal conductivity of air. This increase is attributed to the thermal conductivity of the insulation itself and also to gaseous convection within the insulation layers.

From Table XVIII the thermal conductivity in vacuum can be seen to increase after each air exposure, Column 3, indicating that an oxide layer was being formed on the foils with subsequent increase in foil emissivity. The measured thermal conductivities in vacuum after the various air exposures were corrected to the boundary temperatures of the base point (Column 4, Table XVIII) and then ratioed to the base point conductivity (Column 5, Table XVIII).

Next, the experimentally measured vacuum thermal conductivities after air exposure were compared with thermal conductivities that were predicted analytically from the correlation developed in Section (3.3.3.5). This comparison is presented in Columns 6 and 7, Table XVIII.

The following is a brief discussion of how this analytical method of predicting thermal conductivities in vacuum after air exposure was applied to the experimental data from these air exposure tests.

The theoretical correlation expresses thermal conductivity after air exposure is:

$$k_{ae} = \frac{a}{2} \left(\frac{T_1^2 - T_2^2}{T_1 - T_2} \right) + \frac{2\sigma}{n(T_1 - T_2)} \int_{T_2}^{T_1} \epsilon(T) T^3 dT \quad (78)$$

where

a = solid conductivity coefficient
 σ = Stefan-Boltzmann constant

Table XVIII

THERMAL EFFECTS OF AIR EXPOSURE

	HOT FACE TEMPERATURE, °F	COLD FACE TEMPERATURE, °F	THERMAL CONDUCTIVITY k_a BTU/HR.FT.°R X 10 ⁻³	THERMAL CONDUCTIVITY CORRECTED TO BASE POINT TEMPERATURES k_{ae} BTU/HR.FT.°R X 10 ⁻³	$\left(\frac{k_{ae}}{k_a}\right)$ MEAS = K	$\left(\frac{k_{ae}}{k_a}\right)$ CALC = K ¹	$\frac{K^1 - K}{K}$
<u>NICKEL FOIL- REFRASIL QUARTZ FIBER PAPER</u>							
BASE POINT IN VACUUM,	1600	253	2.61	2.61			
AIR EXPOSURE NO. 1-70 HRS.	900	380	35.27				
IN VACUUM AFTER AIR EXPOSURE NO. 1	1610	261	3.02	2.98	1.141	1.027	-0.100
AIR EXPOSURE NO. 2-143 HRS.	887	385	36.45				
IN VACUUM AFTER AIR EXPOSURE NO. 2	1589	275	3.42	3.414	1.308	1.058	-0.191
AIR EXPOSURE NO. 3-363 HRS.	1064	453	40.03				
IN VACUUM AFTER AIR EXPOSURE NO. 3	1599	289	3.97	3.892	1.491	1.238	-0.170
<u>COPPER FOIL- REFRASIL QUARTZ FIBER PAPER</u>							
BASE POINT IN VACUUM,	1292	297	1.18	1.18			
AIR EXPOSURE NO. 1-24 HRS.	223	149	31.20				
IN VACUUM AFTER AIR EXPOSURE NO. 1	1272	302	1.23	1.247	1.061	1.129	0.064
AIR EXPOSURE NO. 2-24 HRS.	342	207	34.80				
IN VACUUM AFTER AIR EXPOSURE NO. 2	1287	356	1.77	1.712	1.457	1.704	0.170
AIR EXPOSURE NO. 3-24 HRS.	380	234					
IN VACUUM AFTER AIR EXPOSURE NO. 3	1280	374	2.14	2.056	1.750	2.500	0.429
AIR EXPOSURE NO. 4-48 HRS.	371	233.6	39.7				
IN VACUUM AFTER AIR EXPOSURE NO. 4	1295	392	2.46	2.296	1.954	3.330	0.704

n = layer density
 T_1 = hot side temperature
 T_2 = cold side temperature

The solid conductivity coefficient, $a/2$, is obtained from the empirical correlation. Solid conductivity, k_{sc} is believed to be higher than that given by:

$$a/2 \left(\frac{T_1^2 - T_2^2}{T_1 - T_2} \right)$$

and that the difference between the two is charged to the radiative conductivity. This difference between actual solid conductivity and the value used in equation (7) may help to explain the lack of agreement between the predicted and experimentally measured thermal conductivities for the copper foil - Refrasil paper combination shown in Table XVIII. Another possibility for this disagreement, a restricted rate of diffusion or oxygen supply, is also discussed later in this section.

The foil emissivity function, $\epsilon(T)$, is a polynomial of degree, n , so that:

$$\epsilon(T) = \sum_{i=0}^n C_i T^i \quad (79)$$

The foil emissivity after air exposure is a function of oxide thickness, absorption coefficient, scattering coefficient, and base metal reflectivity.

Consequently, the emissivity during vacuum operation of an oxidized system is a function of both temperature and X/L , where X/L is the dimensionless position in the wrap relative to a total thickness L . Therefore, in order to determine the emissivity relationship for a particular insulation wrap, the temperatures both during air exposure and after air exposure must be determined at several X/L positions in the wrap. The temperature during air exposure is not difficult to obtain since the profile can be closely approximated knowing the conductivity of air as a function of temperature. For the temperature in vacuum after air exposure, first assume a temperature profile, calculate the emissivities at several X/L positions, and then fit these emissivities and temperatures with a polynomial, equation (79).

Note that ϵ is actually a function of T and T_A (temperature during air exposure) so that this relationship is only valid for a particular set of air exposure and vacuum conditions.

The vacuum thermal conductivity after air exposure can then be determined from equation (78). From equation (78) an expression for the insulation temperature profile in vacuum after air exposure can be derived by integrating to a point x from the hot side. For a polynomial of degree n , the equation for the profile is:

$$\frac{a}{2} T^2 + B \sum_{i=0}^n \frac{C_i}{i+4} T^{i+4} = \frac{a}{2} T_1^2 + B \sum_{i=0}^n \frac{C_i}{i+4} T_1^{i+4} - \frac{x}{\ell} \left[\frac{a}{2} (T_1^2 - T_2^2) + B \sum_{i=0}^n \frac{C_i}{i+4} (T_1^{i+4} - T_2^{i+4}) \right] \quad (80)$$

where

$$B = \frac{2\sigma}{n}$$

From equation (80) the vacuum temperature profile can be recalculated.

This procedure is repeated until the calculated profile becomes equal to the assumed profile. Calculations have shown, however, that even under extreme air exposure conditions where significant increases in the thermal conductivity occur, the temperature profile is still reasonably close to the profile before air exposure.

These equations governing air exposure have been programmed on the IBM 360 computer. The emissivity - temperature relationship for copper foil - Refrasil quartz fiber paper and nickel foil - Refrasil quartz fiber paper during vacuum operation after an air exposure are given in Figures 46 and 47 respectively. The calculated conductivity deterioration ratio is given in Table XVIII, Columns 6 and 7. The calculated ratio versus the measured ratio is plotted in Figure 48. The percent deviation is plotted against the measured deterioration ratio in Figure 49.

Figure 49 shows that the analytical equation predicts the air exposure effect within 20% for nickel but shows a difference of up to 80% for these particular air exposure conditions for copper. This 20% deviation for nickel is within the range of uncertainties that were expected, due to the complexities and assumptions involved in the analysis. The 80% deviation for copper points to additional factors that

COPPER FOIL - REFRAASIL QUARTZ FIBER PAPER

Figure 46

NICKEL FOIL - REFRASIL QUARTZ FIBER PAPER

FIGURE 47

COMPARISON OF CALCULATED
WITH MEASURED THERMAL
CONDUCTIVITIES AFTER AIR EXPOSURE

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○ NICKEL FOIL - REFRASIL QUARTZ FIBER PAPER

× COPPER FOIL - REFRASIL QUARTZ FIBER PAPER

$\left(\frac{k_{ae}}{k_o}\right)$ CALC. $\uparrow$

$\left(\frac{k_{ae}}{k_o}\right)$ MEAS. $\rightarrow$

FIGURE 48

DEVIATION OF THEORETICAL THERMAL CONDUCTIVITY
FROM EXPERIMENTAL THERMAL CONDUCTIVITY

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○ NICKEL FOIL - REFRASIL QUARTZ FIBER PAPER

× COPPER FOIL - REFRASIL QUARTZ FIBER PAPER

Figure 49

might have been present in this exposure.

Upon re-examination of the air exposure conditions of the copper foil - Refrasil quartz fiber paper combination an aspect was uncovered that might have caused the copper to oxidize less than expected and hence cause the calculated performance to be pessimistic.

The experimental procedure for air exposure was to open the tester vacuum system valve to the atmosphere for the desired length of time while adjusting the heater power input to obtain the desired hot side temperature. Consider the volume above an element of foil surface at some interior position in the wrap. This volume is in effect a dead end volume when the air enters the tester for an air exposure. One fifth of the air entering this volume (the oxygen) reacts to form oxide. This one fifth volume is replaced by air which contains one fifth oxygen. The total amount of oxygen thus far reacted is $1/5 + (1/5)(1/5)$. The process continues until, in the limit, pure nitrogen is present above the foil element. The total fraction of the volume above an element of foil surface that would contribute oxygen would be:

$$\sum_{n=1}^{\infty} \left(\frac{1}{5}\right)^n = \frac{1}{5} + \left(\frac{1}{5}\right)^2 + \left(\frac{1}{5}\right)^3 + \dots = \frac{1}{4} \quad (81)$$

Therefore, only one fourth of the volume above the foil element would contribute oxygen in the initial stage of oxidation. Approximate calculations show that this amount of oxygen leads to the formation of only 0.01 microns which decreases the reflectivity only a negligible amount.

The second stage of oxidation involves the extraction of oxygen that diffuses from the external air through the nitrogen shroud. This is, of course, a very slow and time dependent process. Note from Table XVIII that the times for copper exposure were much shorter than for nickel exposure. Since the nickel data agrees approximately with the calculated effect, it is reasonable to assume that the diffusion rate was equal to or greater than the oxidation rate. If we make the assumption that the oxidation rate equalled the diffusion rate in the nickel test, we can back calculate the conductance parameters for the nickel wrap from the equation

$$Q = D\Delta P \left(\frac{A}{\Delta x}\right) \quad (82)$$

where

- Q = flow rate calculated from the oxidation rate (cm^3/sec)
- D = diffusion coefficient of oxygen into nitrogen at the desired temperature
- ΔP = partial pressure difference, atm.

A = area normal to diffusion, cm^2
 Δx = path length for diffusion cm

From the literature, a diffusion constant at 650°F of $0.74 \text{ cm}^2/\text{sec.}$ was obtained. A partial pressure difference of 0.2 atm. was used and $(A/\Delta x)$ was calculated as $0.21 \text{ cm}^2/\text{cm}$. Substituting this $(A/\Delta x)$ into (11) and using the total time for all the copper air exposures, the total amount of oxygen that diffused into the copper wrap was calculated. The amount of oxide formed from this oxygen and the air exposed conductivity were then calculated. The ratio of exposed to non-exposed conductivity was then calculated to be 2.5. The actual measured ratio was 1.95 giving a percent deviation of 28%. Considering that it was necessary to make several assumptions in these last series of calculations. This agreement with the measured results is quite good.

The main point that should be emphasized in using the theoretical calculations, is that a sufficient supply of air is assumed; that is, the rate of diffusion or oxygen supply must be greater than the oxidation rate at the temperature of interest. Since the diffusion rate for the copper exposures was apparently less than the oxidation rate, the results from these copper exposures could not be expected to agree with the theoretical calculations. In actual practice, if short exposure times are expected, it should be realized that the calculated effect may be conservative, that is, the actual effect may be less severe.

3.3.3.8 Air Exposure Correlation

It has been the intent in this air exposure investigation to develop a theoretical method for predicting the effects of air exposure upon thermal performance of Super Insulation systems and to verify experimentally this theoretical method. Considering the assumptions the analytical method and the possibilities of errors in the experiments, a first order analytical prediction of deterioration seems possible.

Table XIX summarizes all the experimental data and the corresponding theoretical calculations. The calculated values were obtained using the methods presented in Section 3.3.3.7. The new curves for emissivity versus temperature at a particular position in the wrap are given in Figures 50 and 51. Figures 52 and 53 compare the deterioration ratios and amounts of deteriorations for calculated and measured points. Note that the calculated values are now closer to the experimental data, particularly for the copper foil-Refrasil quartz fiber paper exposure.

These latest calculational results were obtained after the input data to the theoretical method were improved. One of the important equations that led to the determination of the absorption and scattering constants for the oxides is the following: (Appendix C)

$$\frac{a}{s} = \frac{\epsilon_{\infty}}{2(1-\epsilon)} \quad (83)$$

where a and s are the absorption and scattering constants of the oxide layer and ϵ is the emissivity of an infinitely thick oxide layer. As an approximation, the (as a function of temperature) was taken as the emissivity of pure oxide as reported by Gubareff, et al³. However, new data by Wade has been located which more closely fulfills the requirements of the theory. It gives the emissivity vs. temperature relationship for copper oxidized at 1400°f for 40 minutes.

Consulting Figure 35, we find that approximately 10 microns of oxide will have formed on the copper. Figure 42 shows that this thickness of coating is essentially "infinite" as far as Equation 83 is concerned. Since this data is available, it should be more appropriate for use in 83 than the emissivity of pure oxide since the sample of pure oxide may not have been prepared in the same way that an "infinite" coating will form on the metal surface. A different crystal structure or a different degree of order in the two forms of oxide may lead to substantially different emissivity-temperature relationships.

Wade's data for copper oxide can be predicted by the following equation:

$$\begin{aligned} &= 0.495 \text{ for } T \text{ } 1060^{\circ}\text{R} \\ &= .798 - .454 \times 10^{-3}T + .139 \times 10^{-9}T^3 \quad (84) \\ &\text{for } T \text{ } 1060^{\circ}\text{R} \end{aligned}$$

This relationship is substantially different from the previous data used for copper oxide since the previous data predicted a decreasing emissivity with temperature while this new data predicts an increasing emissivity with temperature (which is usually the case for this type of materials).

Sully, et al⁵ has given the emissivity-temperature relationship for a nickel oxide coating that was essentially infinite in thickness. This data can be predicted by:

$$\begin{aligned} &= .00013T + .36 \text{ for } T \text{ } 1200^{\circ}\text{R} \\ &= .6366 - .3049 \times 10^{-3}T + .1763 \times 10^{-6}T^2 \text{ for } T \text{ } 1200^{\circ}\text{R} \end{aligned}$$

This relationship is not substantially different than the nickel oxide data previously used.

The agreement between calculated and experimental deterioration due to air exposure is considered to be quite good in light of the many assumptions in the analysis (e.g. oxide layer have same density as pure oxide, scattering coefficient of copper oxide is the same as nickel oxide). Also, much of the data needed to use the theoretical results may be somewhat questionable or hard to interpret as demonstrated above by the influence of the emissivity data for copper oxide. In addition, errors may be present in the air exposures of the copper foil-Refrasil paper and nickel foil-Refrasil paper testers. The point of most concern is whether or not the foils were exposed to a plentiful supply of oxygen.

While Figures 52 and 53 show substantial disagreements between calculated and measured deterioration, the possibility of disagreement is understandable in light of some of the assumptions and the crude nature of the experiments. Specifically, the following points might be in error in the analytical method.

(1) Use of the equation (41) to separate solid conduction and radiation. This also forces use of the "effective" emissivity as the base emissivity in the calculations.

(2) Assumption that the scattering coefficient of copper oxide is the same as that of nickel oxide.

(3) Assumption of oxide density equal to that of the pure oxide.

(4) Use of literature data for emissivity of heavily oxidized foils, oxidation rates, and emissivity vs. oxide weight data for nickel; any of which may be questionable.

In addition, the following factors may have led to errors in the measurement of the oxidized conductivity of the cylindrical testers.

Tablx XIX

THERMAL EFFECTS OF AIR EXPOSURE

Nickel Foil - Refrasil Quartz Fiber Paper	Hot Face Temperature, °F.	Cold Face Temperature, °F.	Thermal Conductivity k_a Btu/hr.ft.°R x 10 ⁻³	Thermal Conductivity Corrected to Base Point Temperatures		$\left(\frac{k_{ae}}{k_a}\right)$ Meas	$\left(\frac{k_{ae}}{k_a}\right)$ Calc	$\left(\frac{k_{ae}}{k_a}\right)$ Meas	$\left(\frac{k_{ae}}{k_a}\right)$ Calc
				k_{ae}					
Base Point in Vacuum	1600	253	2.61	$k_a = 2.51$					
Air Exposure No. 1 - 70 hr.	900	380	35.27						
In Vacuum After Air Exposure No. 1	1610	261	3.02	2.38		1.141	1.037	368	0966
Air Exposure No. 2 - 143 hr.	887	385	36.45						
In Vacuum After Air Exposure No. 2	1589	275	3.42	3.414		1.308	1.077	.804	.201
Air Exposure No. 3 - 363 hr.	1064	453	40.03						
In Vacuum After Air Exposure No. 3	1599	239	3.97	3.892		1.491	1.293	1.282	.765
Air Exposure No. 4 - 425 hr.	1225	504	41.7						
In Vacuum After Air Exposure No. 4	1586	286	3.95	3.93		1.504	1.451	1.315	1.177
Copper Foil - Refrasil Quartz Fiber Paper									
Base Point in Vacuum	1292	297	1.18	$k_a = 1.18$					
Air Exposure No. 1 - 24 hr.	223	149	31.20						
In Vacuum After Air Exposure No. 1	1272	302	1.23	1.247		1.061	1.081	.0720	.0956
Air Exposure No. 2 - 24 hr.	342	207	34.80						
In Vacuum After Air Exposure No. 2	1287	356	1.77	1.712		1.457	1.491	.539	.579
Air Exposure No. 3 - 24 hr.	380	234							
In Vacuum After Air Exposure No. 3	1280	374	2.14	2.056		1.750	2.32	.885	1.558
Air Exposure No. 4 - 48 hr.	371	233.6	39.7						
In Vacuum After Air Exposure No. 4	1295	392	2.46	2.296		1.954	2.588	1.126	1.874

FIGURE 50
Improved EMISSIVITY TEMPERATURE RELATIONSHIP AFTER AIR EXPOSURE
NICKEL FOIL - REFRAASIL QUARTZ FIBER PAPER

FIGURE 51
Improved EMISSIVITY - TEMPERATURE RELATIONSHIP
AFTER AIR EXPOSURE
COPPER FOIL-REFRASIL QUARTZ FIBER PAPER

FIGURE 52

A Better COMPARISON OF CALCULATED WITH MEASURED THERMAL CONDUCTIVITIES
AFTER AIR EXPOSURE

○ NICKEL FOIL - REFRASIL QUARTZ FIBER PAPER

× COPPER FOIL - REFRASIL QUARTZ FIBER PAPER

(5) Insufficient oxygen contact causing oxygen starvation.

(6) General inaccuracies in the cylindrical measurements which might have resulted in up to 40% error in the measurements.

The effect of each of the above factors on the results is difficult to assess due to the complexities of the equations. Point (5) was discussed in the last quarterly report and could have resulted in high predicted values particularly for copper. The measurement errors consist of instrument and heat loss calculation inaccuracies. One obvious example of possible experimental errors can be noted by comparing the third and fourth nickel foil-Refrasil paper exposure points. The fourth point was exposed 160°F higher than the third point and for 60 hours longer and yet the effect on the conductivity was barely detectable, whereas the third point showed a rather substantial change from the second point. Since most of the foils in the tester are only oxidized slightly, the conductivity should not be leveling out by any reasonable theory. Even though a thorough analysis of the effects listed above has not yet been made, it is felt that the disagreement is more due to experimental deficiencies than defects in the analytical calculations.

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3.4 Vacuum Maintenance

3.4.1 Insulation Processing

3.4.1.1 Radiation Shields Processing

Since thermal performance of a multilayer insulation system is dependent on low emissivity of the radiation shield foils, thermochemical processing of the candidate foils was investigated.

Foil samples evaluated included:

- .30 mil tantalum foil (Wah Chang Corp.)
- .25 mil rolled aluminum foil (Alcoa)
- .5 mil electrodeposited copper foil (Anaconda American Brass Co.)
- .5 mil rolled copper foil (Hamilton Watch Co.)
- 1.0 mil rolled nickel foil (Somers Brass Co.)
- .6 mil rolled nickel foil (Somers Brass Co.)

The Aluminum and nickel foils are in a relatively clean "as received" condition and do not require much cleaning. The two copper foils had heavy discolorations, and the cleaning procedures were concentrated on the task of removing these. Figure 54 is a photograph, 200 magnification of "as received" copper foil.

A literature search yielded several possible cleaning procedures among which were

- Solvent cleaning
- Acid pickling
- Chemical brightening
- Linde foil cleaning process
- Alkaline cleaning
- Emulsion cleaning
- Vapor degreasing
- Electrolytic brightening
- Ultrasonic techniques

These procedures, with the exception of the Linde foil cleaning process, are discussed in Reference 1.

Loose surface dust and oils can be removed with a solvent, emulsion, vapor degreasing, or an ultrasonic treatment. Since the foils are relatively clean in terms of surface dirt, more complicated cleaning procedures such as alkaline and emulsion cleaning (which require soaking and rinsing) and vapor degreasing and ultrasonic techniques (which require special equipment) are unnecessary.

Figure 54. As-Received Copper Foil, 200X Magnification

Figure 55. Processed Copper Foil, 200X Magnification

The solvent wipe was tested on the two copper foils using:

Acetone
Toluene
Benzene
Chloroethane
Methyl ethyl ketone
SD-1 (alcohol)
Kerosene

However, no visible change was noted. This would seem to indicate that the discolorations were due to oxide formations, and a vendor contact with Anaconda American Brass Company confirmed this for their foil.

An acid pickling solution (which is essentially similar to a chemical brightening) was employed on the Anaconda foil and worked well. The solution consisted of:

25% by volume HNO_3
60% by volume H_2SO_4
0.2% by volume HCl
14.8% by volume H_2O

Here, however, the procedure involves soaking, rinsing and drying; and would become complicated for large rolls of foil. The electrolytic brightening was ruled out because the use of specialized equipment is involved.

The Linde foil cleaning process was evaluated for the following materials and conditions:

<u>Foil</u>	<u>Temperature, °F</u>	<u>Time, * hours</u>
Aluminum	800	4 - 6
Copper	1400-1600	4 - 6
Nickel	1800	4 - 6

Although the Linde foil cleaning process was effective in removing copper oxide films at 1400°F, traces of the advanced oxidation present on the Anaconda foil remained in the form of a rough surface. This could be removed and a smooth surface formed by processing at 1600°F. The experiment at 1600°F yielded an attractive foil that was very workable and quite bright on both sides. Figure 55 is a photograph, 200 magnification, showing a sample of copper foil after application of the Linde foil cleaning process at 1600°F. This would be in contrast to a heavily oxidized, relatively dull foil in the "as received" condition.

Tantalum foil is cleaned by ultrasonic solvent immersion techniques by the vendor and is free from surface dirt and discolorations. Therefore, techniques such as alkali cleaning or emulsion cleaning are not required. The Linde foil cleaning process was employed and, after

*Exact processing time is dependent upon initial foil condition.

processing at 1800°F for six hours, it was found that the tantalum foil had become very brittle and discolored. In order to gain insight into this problem, a laboratory X-ray diffraction analysis was performed on the processed foil. X-ray diffraction analysis disclosed that the following compounds of tantalum were present: Ta_2O_5 approximately 40 per cent, TaN approximately 10 per cent, and the balance unidentifiable.

Since not every compound of tantalum has been identified by the ASTM for standards, some of the compounds formed during the processing are reported as unidentifiable. It is theorized that the unidentifiable compounds could be a partial oxidation product, a different valence state or a mixed molecule such as $TaOCl$, or $TaNCl$. The mixed molecules could result from reaction of the tantalum with trace gaseous impurities, a characteristic of refractory metals.

Based on these results, it appears that the Linde foil cleaning process is not suited for cleaning tantalum foil.

Samples of tantalum foil were next exposed to vacuum to establish preliminary justification for continued evaluation.

Samples of tantalum foil were exposed to vacuum, 10^{-4} to 10^{-5} mmHg at 1800°F for periods up to 150 hours. In one experiment the tantalum foil was wrapped loosely with unprocessed Refrasil quartz fiber paper and exposed to a vacuum of 10^{-4} to 10^{-5} mmHg at 1800°F for approximately 72 hours. The tantalum foil was somewhat darker, probably from offgassing constituents from the unprocessed Refrasil quartz paper. The foil was slightly more brittle to the touch after the experiment but still was very handleable. In the second experiment the tantalum foil was exposed to 10^{-4} to 10^{-5} mmHg at 1800°F for approximately 150 hours during the vacuum enclosure background and specimen tests (Section 3.4.3). The tantalum foil was slightly more brittle to the touch after exposure but was still bright and handleable.

From these preliminary experiments, simulating operational environments, it appeared that tantalum foil justified further study. However material compatibility studies later disqualified tantalum as a candidate radiation shield material, Section 3.5.1.3.1.

3.4.1.1.1 Analysis of Radiation Shield Processing Techniques

Normal reflectance measurements were obtained for various foils, both before and after cleaning. These measurements were made in two ways -- one using visible light and the other infrared. The cleaning procedure used was that of the Linde foil cleaning process except for one measurement in the visible light region made on copper foil cleaned by the acid pickling technique.

The measurements in the visible light region were made utilizing an optical microscope, and the results of these tests are presented in Table XX. The values in the table represent reflectivities rela-

TABLE XX.RELATIVE FOIL REFLECTIVITIES

<u>Sample</u>		<u>As-received</u>	<u>After Linde Foil Cleaning Process</u>	<u>After Acid Pickling</u>
1/4 mil Al	Dull Side	0.37	0.28	--
	Bright Side	0.42	0.81	--
1/2 mil Al	Dull Side	0.34	--	--
	Bright Side	0.44	--	--
1/2 mil Cu*	Dull Side	0.10	0.7	0.47
	Bright Side	0.22	0.7	0.60
1/2 mil Cu**	--	0.4	0.96	--
1 mil Ni	--	0.34	0.6	--

*Produced by Anaconda American Brass Company

**Produced by Hamilton Watch Company

tive to a polished aluminum standard -- the relative reflectivity of the standard being unity. The values are not necessarily linear with respect to reflectance but do give an indication of the quality of the cleaning process. The closer the relative reflectivity approaches to unity, the brighter the surface.

Copper foil showed a marked improvement in reflectivity after cleaning but little improvement could be noted for nickel and aluminum. The dull side of the aluminum foil actually showed a decrease after cleaning.

The reflectance measurements in the infrared region of the light spectrum were made with a infrared spectrophotometer. When an incident radiation (infrared) is imposed on a surface, a portion is absorbed, a portion is transmitted, a portion is scattered and a portion is reflected. The spectrophotometer measures only the reflected portion of the incident infrared energy. The wave length of the radiation was varied according to the service temperature of the foil. By assuming the foil to be a black body, the wave lengths, λ of the radiation emitted from the foil at the given temperatures are:

For aluminum	830 > T > 150°F	4.0 < λ < 8.0
For copper	1630 > T > 830°F	2.5 < λ < 4.0
For nickel	2140 > T > 830°F	2.0 < λ < 4.0
For Tantalum	2140 > T > 830°F	2.0 < λ < 4.0

Each of the above candidate foils was examined, over the indicated wave lengths, against one of three standards depending on the reflectivity of the sample. During testing the standard was first tested in air to obtain a base point and the sample was then tested against the standard. A reflective index is obtained by multiplying the percent reflectivity of the standard with that of the sample. The reflective index is not related to the emissivity of the foils and should not be used as a measure of emissivity. Because different standards were used with the different foils the reflective index cannot be used to compare one foil against another. The only purpose of these measurements was to compare the reflective index of the foils before and after processing to determine if an improvement was achieved. Figures 56, 57, and 58 show the as received and after processed reflective indexes for aluminum, copper and nickel respectively. Considerable scatter of data is observed in the figures with the uncertainties indicated by the shaded areas. Since the spectrophotometer measures the spectral reflection characteristics by infrared energy source the surface condition of the foil, such as wrinkles or roughness, influences the reflective index. For example if the infra red beam is impinged on a flat section of foil a great portion will be reflected. However, if the beam is impinged on a wrinkled area a large portion will be scattered so that a lesser amount will be reflected. This makes it difficult to compare the reflectance index of two foils having different surface characteristics and no definite conclusions can be drawn.

FIGURE 56

NICKEL FOIL REFLECTIVITY INDEX AS A FUNCTION OF TEMPERATURE

FIGURE 58

Aluminum and copper foils showed some improvement in reflectivity after processing but no change was observed in the nickel foil.

When processing larger quantities, approximately 100 linear ft., of nickel, aluminum and copper foils was attempted, severe wrinkling of nickel and aluminum foils was encountered.

Investigation of the previous small-scale nickel foil processing experiments revealed that the in-house nickel foil, 1 mil thick, that was used in these small-scale experiments had been annealed during rolling by the vendor at 1600°F. The 0.62 mil nickel foil procured for this contract has been annealed at 1020°F. When processing was attempted at 1200 to 1800°F with the 0.62 mil nickel foil, wrinkling of the foil occurred apparently due to the relieving of stresses incurred during rolling. An acceptable technique which produced a relatively wrinkle-free foil involved rolling the foil on the metal core with previously processed Refrasil quartz fiber paper spacer with processing temperature limited to 900°F.

Wrinkling of the aluminum foil during processing was attributed to the handling procedures used rather than a processing temperature dependence.

Wrinkling is undesirable due to increased difficulties in installation.

Copper foil was successfully processed by rolling alternate layers of foil and previously processed quartz paper spacer material under tension on a metal core, thus maintaining support of the foil during processing. Processing temperature was 1600°F.

Conclusions

The processing of nickel and aluminum foils is not recommended for the following reasons.

- 1). lack of definite improvement in reflectivity.
- 2). the offgassing rate, Section 3.4.2, of nickel and aluminum is not lowered by processing.
- 3). wrinkling of nickel and aluminum was experienced during large scale processing.

Processing of copper foil by the Linde Foil Cleaning Process @ 1600°F for 4 - 6 hours is recommended.

3.4.1.2 Spacer Processing

Thermochemical processing of multi layer insulation spacers is required in order to stabilize the materials for high temperature service

and also to reduce the offgassing rate of the spacers to an acceptable level.

The problem of cleaning a spacer material of its organic contamination can be solved by oxidation of the organic in an oxidizing stream at a high temperature. Experimentally this involves placing the spacer in a furnace under a controlled temperature and passing a fixed oxygen concentration gas stream through it.

Theoretically, it is desired to describe the condition of the spacer in terms of known variables so that optimum process conditions may be selected. The theory may be checked experimentally by monitoring the carbon dioxide concentration of the oxidizing stream as it leaves the furnace. The monitoring was done with a Beckman infrared carbon dioxide analyzer, and a correction for the response time of the instrument was made. Details of this correction are given in Appendix D.

Two processing procedures were evaluated: the air oxidation procedure and the Linde Insulation Cleaning Process. The apparatus employed in the air oxidation process is shown schematically in Figure 59. Photographs of the apparatus are shown in Figure 60 and 61. The air oxidation procedure consists of passing oxidizer gas, 20% oxygen, 80% nitrogen through the furnace tube containing the spacer for 12-14 hours at the recommended processing temperature.

Monitoring of the reaction was achieved by continuous analysis of the carbon dioxide concentration of the exiting gas stream with a Beckman Infrared Carbon dioxide analyzer. Final reaction completeness was obtained by comparing the residual carbon content of the spacers "as received", with the residual carbon content after processing. It was noted that at higher temperatures of reaction, or processing, lower residual carbon contents were obtained. The effectiveness of the cleaning procedure increased with temperature but the effects of long, greater than 12-14 hours, cleaning times and lower, less than 20%, oxygen concentrations seemed to be negligible.

Table XXI shows the residual carbon contents of the candidate spacers for various conditions.

Inspection of the table shows that the oxidizer process appears superior to the Linde process in reducing carbon content of the candidate spacers. Residual carbon content was the criterion selected as the measure of reaction completeness.

Residual carbon content of glass fiber paper does not appear to be reduced by either the oxidation process or Linde process. This is also indicated by the results of the weight loss tests, Section 3.5.1, which shows no loss in weight for glass fiber paper after processing. Since no binder is used in the manufacture of this paper, it is suspected that the residual carbon reported may be contained in a trace compound

FIGURE 59

Figure 60 Insulation Processing Furnaces

Figure 61 Insulation Processing Furnaces Control Panel

TABLE XXI

RESIDUAL CARBON CONTENTS

Material	As Received Carbon Content-%	AFTER AIR OXIDATION						AFTER LINDE PROCESS		
		800°F	900°F	1200°F	1280°F	1500°F	1800°F	800°F	1280°F	1800°F
Non-Opacified Spacers										
Astroquartz Cloth	0.24		0.031	0.019		0.014	0.011			0.049
Astrosil Cloth	0.11						0.041			0.056
Refrasil Quartz Fiber Paper	1.40		0.52	0.55		0.59	0.24			0.08
Glass Fiber Paper	0.24	0.25						0.24		
Opacified Spacers										
Aluminum Flake Opacified Glass Fiber Paper	1.00	0.46						0.71		
Copper Flake Opacified Refrasil Quartz Fiber Paper	0.84				0.16				0.29	

which does not decompose at the 800-1000°F operational temperature of the glass paper.

After processing by the air oxidation method, 20% oxygen, 80% nitrogen, at 1280°F the copper flake opacified Refrasil quartz fiber paper, turned light blue in color and displayed a weight gain of 4.8%, Section 3.5.1. It was believed that the color change and weight gain occurred because of oxidation of the copper flake dispersed throughout the spacer. This is not considered a desirable condition as the emissivity of oxidized copper is higher than that of the pure metal and could result in poor thermal performance of the spacer. Processing by the Linde Insulation Cleaning Process resulted in the color taking on a slightly gray cast but still remaining pink. This processed spacer displayed a weight loss of 6%, which indicates removal of organic impurities without oxidation of the copper flakes.

An X-ray diffraction analysis was made to determine the copper oxide and copper content of the spacer after processing by both the oxidation process and the Linde process.

The X-ray diffraction analysis disclosed the following:

As-Received..... entirely elemental copper

After Linde Insulation Process.. entirely elemental copper

After Oxidation Process..... entirely cupric oxide

Since no oxide was formed due to processing by the Linde Process, and satisfactory offgassing rates were obtained (Section 3.4.2) the Linde Process is recommended.

3.4.1.2.1 Spacer Processing Recommendations

The basic processing procedures established in Section 3.4.1.2 were modified based on information generated in the Physical Stability (Section 3.5.1) and Offgassing (Section 3.4.2) Tasks.

Because of high offgassing rates determined for copper flake opacified Refrasil quartz fiber paper, Refrasil quartz fiber paper and Astroquartz cloth (Section 3.4.2 - Offgassing), the processing techniques employed for these candidates were re-examined.

It was believed that one method of reducing the offgassing rate would involve processing at a higher temperature than the temperature at which the offgassing rate was determined. The processing temperature of the copper flake opacified Refrasil quartz fiber paper was then increased from 1270°F to 1400°F. The processing technique used was that of the Linde Insulation Cleaning Process. Following this 1400°F processing an offgassing test was performed on the spacer at its maximum

recommended operational temperature, 1270°F, and a very low offgassing rate was observed (Section 3.4.2).

Physical stability tests (Section 3.5.1) showed that physical strength of Refrasil quartz fiber paper and Astroquartz cloth decreased rapidly if the processing temperature was increased to 1700°F. A modified air oxidation processing technique was next explored to reduce the offgassing rate of these candidates rather than employing an increased processing temperature. This technique consisted of first passing oxidizer gas, 20% oxygen, 80% nitrogen, through the furnace tube containing the spacer for 12-14 hours at processing temperature. Next the spacer was subjected to 3 pressure-vacuum cycles with oxidizer gas in order to insure adequate oxygen penetration and removal of gases formed by the oxidation reaction. These cycles consisted of first pressurizing the furnace tube with oxidizer gas at 10 psig for about 20 minutes and then evacuating to about 3 mm mercury for about 20 minutes. Offgassing tests were conducted on the Refrasil paper and Astroquartz cloth at 1700°F and 1800°F. Both materials exhibited high offgassing rates at 1800°F after 34 hours of pumping at 1900°F (Section 3.4.2 - Offgassing). It therefore appears that use of the modified air oxidation procedure did not reduce the offgassing rates of these candidates below the rate achieved with the original air oxidation processing technique.

The following processing cycles are recommended for the candidate spacers:

<u>Material</u>	<u>Processing Temperature, °F</u>	<u>Time, Hours</u>	<u>Procedure</u>
Refrasil Quartz Fiber Paper	1700°F	15	Air Oxidation (20% O ₂ -80% N ₂)
Astroquartz Cloth	1700°F	15	Air Oxidation (20% O ₂ -80% N ₂)
Copper Flake Opacified Refrasil Quartz Fiber Paper	1400°F	15	Linde Insulation Cleaning Process
Aluminum Flake Opacified Glass Fiber Paper	900°F	15	Air Oxidation (20% O ₂ -80% N ₂)
Glass Fiber Paper	900°F	15	Air Oxidation (20% O ₂ -80% N ₂)
Copper Foil	1600°F	15	Linde Foil Cleaning Process

3.4.2. Offgassing

A high vacuum level on the order of 0.1 to 0.5 microns of mercury should be maintained within a high temperature super insulation system for its optimum thermal performance. This vacuum exists in space applications as part of the normal surroundings; however, with sealed systems, the system itself must be capable of sustaining its high vacuum level. If the insulation system is leak tight and, further, if all materials of the system are completely devoid of gas, a low pressure would be maintained indefinitely. But, such is not the case, since materials composing the system such as metal foils and fibrous spacers contain dissolved and adsorbed gases which are liberated slowly in vacuum.

It is for this reason that knowledge of offgassing characteristics of insulation materials in a vacuum environment is important. Normally, offgassing from materials, even following thermal-chemical processing, is high immediately after exposure to the vacuum environment. In these early stages of the degassing period it is believed that desorption of gases takes place near the material surface. Thereafter, the outgassing process is mainly diffusion from the interior of the material and some sublimation. Thus the outgassing rates should continue to decrease.

Removal of these gases is accomplished by chemical getters which are placed within the system. These getters chemically react with the gases to form non-gaseous products. Offgassing rates should not be too high, otherwise, an impractical amount of getter will be needed to remove the gases. In order to ensure a low offgassing rate, an insulation system usually undergoes a bakeout cycle before it is sealed off. During the bakeout cycle, the system is connected to a vacuum pump to continually remove gases and, at the same time, the system is heated above the system operating temperature. Total gas removed during the bakeout period and subsequent outgassing rate is highly dependent on the temperature during bakeout and also on the duration of the outgassing period; the quantity of gas removed increasing and the terminal outgassing rate decreasing with temperature and duration.

Offgassing rates were determined for: aluminum foil, tantalum foil, nickel foil, copper foil, glass fiber paper, aluminum flake opacified glass fiber paper, copper flake opacified Refrasil quartz fiber paper, Astroquartz cloth, and Refrasil quartz fiber paper. It will be seen later that the thermo-chemical processing and the bakeout cycle do not always help lower outgassing rates.

In the subsequent sections, test apparatus, test procedures, and offgas constituent analysis are described. Offgassing data, their characteristics, and an example of getter requirement calculation are presented.

3.4.2.1 Test Apparatus

The experimental equipment included two 4 inch I.D. vacuum tube furnaces with volumes of 14.25 liters each and with temperature limits of 1900°F. One vacuum tube was constructed of Type 304 stainless steel and the other of Inconel-600 superalloy. Diffusion pumps in conjunction with liquid nitrogen cold traps were used to produce the vacuum environment. Philips cold cathode and Pirani thermocouple gauges were used with strip

chart recorders to provide a continuous readout of the pressures in the chambers. Figure 62 is a photograph of the test apparatus. The apparatus from left to right includes: the mass spectrometer, Type 304 stainless steel offgassing furnace and liquid nitrogen cold trap. Analysis of the offgas constituents is conducted by means of a Model 210 Aero-Vac mass spectrometer gas analyzer. Since nitrogen and carbon monoxide have the same mass, it is difficult to distinguish between the two with a gas analyzer (mass spectrometer). With the aid of secondary nitrogen mass peaks, the maximum amount of nitrogen and the minimum amount of carbon monoxide can be determined.

3.4.2.2 Test Procedure

The initial step in the offgassing test procedure involved calibration of the vacuum chamber in accordance with the bakeout cycle given below.

1. Pumpdown at $(T + 100)^{\circ}\text{F}$. for 17 hours, where T is the insulation system operating temperature.
2. Cool to $T^{\circ}\text{F}$ while pumping for 1/2 hour.
3. Pressure rise for 8 hours at $T^{\circ}\text{F}$

This cycle was repeated twice, and pressure-time data were recorded during both the pumpdown and pressure-rise periods. A typical bakeout cycle is presented graphically in Figure 63. The gas constituent analysis was conducted at the end of each cycle with a mass spectrometer gas analyzer. These tests establish the vacuum chamber characteristics.

Following calibration, the chamber was cooled to 140°F and returned to atmospheric pressure by injecting dry argon gas. A temperature of 140°F . was selected as the lower temperature rather than ambient in order to minimize contamination from atmospheric moisture when the chamber was open. The spacer or shield material to be tested was then placed in the chamber and the same bakeout cycle was repeated.

3.4.2.3 Materials Tested

The materials tested and the corresponding temperatures are listed below:

Radiation Shield Foils

Aluminum Foil	-	900°F. 100°F.
Tantalum Foil	-	1800°F.
Copper Foil	-	1270°F.
Nickel Foil	-	1640°F.

Spacers

Glass Fiber Paper	-	900°F. 100°F.
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Figure 62. Offgassing Test Apparatus

FIGURE 63

Aluminum Flake Opacified Glass Fiber Paper	- 900°F.
Astroquartz Cloth	- 1800°F. 1700°F.
Refrasil Quartz Fiber Paper	- 1800°F. 1700°F. 1400°F.
Copper Flake Opacified Refrasil Quartz Fiber Paper	- 1270°F.

For each of the above tests, at least one empty chamber recalibration test was made. For most of the above tests, two empty chamber runs were made, both before and after the sample test. A list of all tests performed is given in Appendix E.

3.4.2.4 Experimental Data

Essentially all pertinent data of the sample tests are presented in Tables XXII and XXIII. For every material, each sample test is presented including the processing, bakeout, and offgassing temperatures. The offgassing rate expressed in micron-liters per square foot of material/hour is generally given at 0.3 micron of mercury pressure for each pressure rise except for materials with a very high offgassing rate. For the materials with a high offgassing rate, the pressure rose too rapidly through the 0.3 micron range for accurate calculations to be made. The rate was then determined at 2.0 microns. .3 micron was chosen, because at elevated temperatures (400-1800°F.) super insulation exhibits an excellent thermal performance at this vacuum range.

The offgassing data from Tables XVIII and XIX has been plotted in Appendix E. The data was smoothed whenever an obvious inconsistency existed in a manner that would be conservative. That is, the curve was moved upward to the higher offgassing rate when two points seemed inconsistent. Actually, only a few points needed this adjustment. The code given with each curve can be interpreted as follows:

ST	P 900	OG 900	(0.3)
Sample Test Run Number	Processing Temp. (°F.) (UP if un- processed)	Offgassing Temp. (°F.)	Pressure for Offgassing Measurements

Offgassing rates are calculated from the pressure rise curves (Figures 66 and 67 typical) by subtracting the slopes of the empty chamber and chamber plus sample pressure rise curves and the results multiplied by the system volume and divided by the sample area to obtain the offgassing rate. Mathematically this method is expressed as:

$$G_s = \frac{V}{A} \left[\left(\frac{dP'}{dt} \right)_{P_1} - \left(\frac{dP}{dt} \right)_{P_1} \right] \quad (85)$$

where: S = Offgassing rate, micron-liters per hr-sq.ft.

V = System volume, liters

A = Sample area, length x width, sq.ft.

$\frac{dP}{dt}$ = Instantaneous slope of empty system pumpdown curve, microns/hr.

$\frac{dP'}{dt}$ = Instantaneous slope of system with sample pumpdown curve, microns/hr.

P_1 = Pressure at which both dP/dt and dP'/dt are measured.

Equation (85) is derived from the separate mass accountability equations

$$\frac{VdP'}{dt} = -G_s - G_c \quad (86)$$

For Chamber and Sample

$$\frac{VdP}{dt} = G_c \quad (87)$$

For Empty Chamber

G_s = Sample offgassing rates

G_c = Chamber offgassing rate

The simplified equation (85) for G_s is obtained following the assumption that at any given pressure P_1 the chamber outgassing rate is identical regardless of whether the test chamber is full or empty. Although this assumption implies a concession, the assumption is believed adequate. In most cases the offgas analysis for the fuel and empty chambers was similar - if the constituent analysis were identical the assumption would be correct. For the cases where the sample rate G_s was very large, the subtraction is inconsequential.

To illustrate how the rate results are obtained, consider the pressure rise curves in Figures 66 and 67. The symbols ST refer to Sample Test No. and RC designates Recalibration of Chamber. Shown are the data points from the tantalum foil test and the corresponding empty chamber test. The curves shown were fit by a least squares program for the IBM-360 computer which determined the coefficients of the following equation.

$$\log p = a_0 + a_1 \log t + a_2 (\log t)^2 \quad (88)$$

where,

P = pressure, microns

a_0, a_1, a_2 = coefficients

t = time, minutes

Several more complicated equations were tried that produced smaller standard deviations, but these did not yield curves with constantly decreasing first derivatives, a physical constraint imposed by the offgassing phenomenon. The computer program developed also gives a tabulation of the first derivative as a function of pressure. To obtain the values of offgassing rate shown in Table XXII, the derivative at 0.3 micron pressure was taken from the computer printouts for the sample and empty chamber rises. The resulting offgassing rate (G_s) was then divided by the one-side area of the sample to give an offgassing rate per square foot.

Figures 68 and 69 illustrate a phenomenon that occurred frequently in the series of offgassing tests. As the Figures show, the pressure rise curves for the empty chamber fell considerably above the sample curves, indicating that negative offgassing or adsorption was taking place. The materials that exhibit an apparent adsorption rate have apparently become adsorption activated by the overtemperature bakeout before the offgassing test. This behavior occurred with many materials as indicated in Tables XXII and XXIII by the negative rates and was substantiated by repeat runs.

3.4.2.5 Offgas Constituent Analysis

The final constituent analyses presented in Tables XXII and XXIII must be viewed in the face of the several analytical concessions which were required in order to be able to present the results in that form. Specifically:

(a) It is assumed that the mol fraction (x_i) of each constituent as determined at the end of the test is constant throughout the pressure-rise period. This assumption is reasonable as long as desorption of gases reaches completion during the pumpdown portion of the bakeout cycle and the outgassing process is mainly by diffusion during the pressure-rise period. The assumption is rigorously true only for single component offgas. Ideally, the mol fraction should be made time dependent by analyzing offgas constituent throughout the pressure-rise period; practically, however, such a measurement will be too time consuming. A compromise can be made in future work if more accurate data than those available are required, by performing the offgas constituent analysis at several pressures during the pressure-rise period as shown in Figure 69.

(b) It was found that a significant amount of water and argon were present in some analyses. Although it is possible for one of the constituent of the fibrous sample outgas to be water, water shows up also in the foil tests. It is suspected that the water detected reflects permeation into the sampling line through elastomer O-rings during the gas sampling period. Argon is present because all transfer lines and the offgassing chamber are purged with argon gas before bringing back the chamber to the atmospheric pressure.

In view of this reasoning the raw constituent analysis was adjusted prior to further calculation by subtracting the water and argon

percentages and then readjusting the remaining fractions to total 100%. Adjustment to the remaining percentages was weighted according to the magnitude of each remaining fraction. This method should give reasonably accurate results since empty and full chamber analysis were nearly the same except when a material with very high offgassing rate was tested. In the case of a sample with high offgassing rates, the sample gas overwhelms the chamber gas. Thus what is being measured is substantially all sample gas with the exception of water and argon; these were subsequently subtracted.

(c) Since nitrogen and carbon monoxide have the same mass, it is difficult to distinguish between the two with a mass spectrometer gas analyzer. It was possible only to determine maximum amount of nitrogen and minimum amount of carbon monoxide with the aid of secondary nitrogen mass peaks.

Following the above limitations, particularly (a) and (b) the simple relation

$$\bar{x}_{is} = \frac{\bar{x}_{i(s+c)} G_{(s+c)} - \bar{x}_{ic} G_c}{G_{s+c} - G_c} \quad (89)$$

where

G_{s+c} = outgassing rate of sample plus chamber

G_c = offgassing rate of chamber alone

$G_{s+c} - G_c = G_s$ = sample offgassing rate

$\bar{x}_{is}$ = mol fraction of gas species (i) from sample (s) assumed constant throughout pressure rise period.

is used to determine the mol fraction of each species (i) assignable to the sample ($\bar{x}_{is}$).

The limitations of this simple analysis can perhaps be clarified, and the requirements for a more rigorous analysis set forth by presenting the mathematical development and associated simplifications necessary to derive the above expression.

Let the total quantity of gas accumulated in the chamber, whose volume is V, after a period of time (t) be:

$$PV = V \int_0^t P_g d\tau \quad (90)$$

where, P = pressure in chamber

$\dot{P}$ = instantaneous pressure gradient in chamber, $dP/d\tau$

Let the gas mixture consist of (n) species such that we can write,

$$P = \sum_{i=1}^n P_i, \quad \text{and also} \quad \dot{P} = \sum_{i=1}^n \dot{P}_i$$

Let there be three distinct sources for the accumulated gas which are:
1. The test sample (s), 2. The test chamber (c), and 3. The test system lines during sampling only (l). Then, equation 85 may be rewritten as:

$$\begin{aligned} PV &= P_{s+c+l} V \\ &= (P_s + P_{c+l}) V \\ &= \sum_{i=1}^n (P_{is} + P_{i(c+l)}) V \\ &= V \sum_{i=1}^n \int_0^t (\dot{P}_{is} + \dot{P}_{i(c+l)}) d\tau \end{aligned} \quad (91)$$

Further, if we let (x_i) be the time dependent mol fraction associated with the time dependent partial pressure (P_i) we can write:

$$P_{i(s+c+l)} = x_{i(s+c+l)} P_{s+c+l}$$

$$P_{i(c+l)} = x_{i(c+l)} P_{c+l}$$

$$P_{is} = x_{is} P_s$$

and, by differentiating the above with respect to time we get:

$$\dot{P}_{i(s+c+l)} = \dot{x}_{i(s+c+l)} P_{s+c+l} + x_{i(s+c+l)} \dot{P}_{s+c+l}$$

$$\dot{P}_{i(c+l)} = \dot{x}_{i(c+l)} P_{c+l} + x_{i(c+l)} \dot{P}_{c+l}$$

$$\dot{P}_{is} = \dot{x}_{is} P_s + x_{is} \dot{P}_s$$

Substituting the above into equation 91, noting that PV can also be expressed as

$$\sum_{i=1}^n \int_0^t P_{i(s+c+1)} V d\tau,$$

we obtain the following:

$$V \int_0^t \left[\sum_{i=1}^n \left(x_{i(s+c+1)} P_{i(s+c+1)} + x_{i(s+c+1)} P_{i(s+c+1)} \right) \right] d\tau = V \int_0^t \left[\sum_{i=1}^n \left(x_{i(s+c+1)} P_{i(s+c+1)} + x_{i(s+c+1)} P_{i(s+c+1)} \right) \right] d\tau \quad (92)$$

We are interested in two essential results:

1. A meaningful outgassing rate for the sample - G_s
2. The sample outgassing species and their distribution - x_{is} .

Integration of equation (92) coupled with a sufficient amount of experimental data can yield at least a valid time history of (G_s) and (x_{is}) for the time period of the experiment (ie., (t)), provided there is no interaction between the sample outgassing and the chamber outgassing.

From an experiment of reasonable duration, it may then be possible to infer a meaningful value of G_s and (x_{is}).

The experimental data required would be:

1. A history of ($P_{(s+c+1)}$) vs. (τ) for the time period (τ).
2. A history of ($P_{(c+1)}$) vs. (τ) for the same time period.
3. A history of $x_{i(s+c+1)}$ vs. (τ) for the same time period.
4. A history of $x_{i(c+1)}$ vs. (τ) for the same time period.

The required data is shown graphically in Figure 69.

However, as noted previously, we do not have these required experimental data. In fact, only one gas analysis was made at the conclusion of the pressure-rise period ($t = 8$ hours); thus, any changes in con-

centration of the species (i) with time (i.e., Figure 69) are not available. Any attempt to use Equation (92) must then be compromised by the only available assumption:

$$x_i(t) = x_i = \text{constant}$$

or,

$$\dot{x}_i = 0$$

This assumption may be reasonable provided the outgas process is diffusion from the interior of the materials and every element of the system is contributing positively to the gas sample.

The assumption is most valid for materials which have undergone a high temperature bakeout so that desorption is negligible compared to diffusion.

As stated previously, the assumption is only rigorously correct for those cases where the gas analysis only showed a single component. Assuming $x_i = \text{constant}$ and neglecting the contribution of lines as previously suggested, Equation (92) can be written as:

$$\bar{V}\bar{X}_{i(s+c)} \int_0^t \dot{P}_{s+c} d\tau = \bar{V}\bar{X}_{is} \int_0^t \dot{P}_s d\tau + \bar{V}\bar{X}_{ic} \int_0^t \dot{P}_c d\tau \quad (93)$$

Rewriting Equation 93 by replacing (t) by (t + dt) we have:

$$\bar{V}\bar{X}_{i(s+c)} \int_0^{t+dt} \dot{P}_{s+c} d\tau = \bar{V}\bar{X}_{is} \int_0^{t+dt} \dot{P}_s d\tau + \bar{V}\bar{X}_{ic} \int_0^{t+dt} \dot{P}_c d\tau \quad (94)$$

Subtracting Equation 93 from Equation 83:

$$\bar{V}\bar{X}_{i(s+c)} \int_t^{t+dt} \dot{P}_{s+c} d\tau = \bar{V}\bar{X}_{is} \int_t^{t+dt} \dot{P}_s d\tau + \bar{V}\bar{X}_{ic} \int_t^{t+dt} \dot{P}_c d\tau$$

or,

$$\bar{X}_{i(s+c)} \dot{P}_{s+c} V = \bar{X}_{is} \dot{P}_s V + \bar{X}_{ic} \dot{P}_c V \quad (95)$$

By rearranging Equation 95, we finally obtain the simplified form at time (t):

$$\bar{X}_{is} = \frac{\bar{X}_{i(s+c)} G_{s+c} - \bar{X}_{ic} G_c}{G_{s+c} - G_c} \quad (96)$$

where

$$\begin{aligned} G_{s+c} &= \dot{P}_{s+c} V; \text{ total offgassing rate} \\ G_c &= \dot{P}_c V; \text{ chamber offgassing rate} \\ G_{s+c} - G_c &= G_s = \dot{P}_s V; \text{ sample offgassing rate} \end{aligned}$$

Although the outgassing mechanism is assumed mainly by diffusion in arriving at Equation 91, no mention was made concerning its validity. For this task, two typical materials, metal foil and spacer paper, were chosen and their pressure rise curves were plotted against time. Figure 61 gives the pressure-time curve of tantalum foil. Although the figure shows only 6 hours of the pressure rise, it is clear that during a typical pressure-rise period of 8 hours the chamber pressure increases almost linearly after less than one hour of the start of the pressure-rise period, i.e., the error encountered will not be great by assuming the outgassing mode being diffusion throughout the period. Figure 72 gives the pressure-time curve of glass fiber paper. Although the curve is not as linear as that for tantalum foil, it is still very near linear especially after two hours of the start of the pressure-rise period; and the offgassing mode can be considered to be diffusion throughout the pressure-rise period without sacrificing accuracy significantly. Thus, it seems that it is reasonable to assume the offgassing mode to be diffusion.

OUTGASSING MODE FOR SECOND
PRESSURE RISE OF TANTALUM
(ST9 UP OG1800)

OUTGASSING MODE FOR SECOND
PRESSURE RISE OF GLASS FIBER PAPER
(ST11 P1000 OG900)

3.4.2.6 Offgassing Rates

Essentially all pertinent data of the sample tests are presented in Tables XXII and XXIII. For every material, each sample test is presented including the processing, bakeout, and offgassing temperatures. The offgassing rate expressed in micron-liters per sq. ft. of material/hour is generally given at 0.3 micron of mercury pressure for each pressure rise except for materials with a very high offgassing rate or a very low offgassing rate. For the materials with a high offgassing rate, the pressure rose too rapidly through the 0.3 micron range for accurate calculations to be made. The rate was then determined at 2.0 microns. For materials with a very low offgassing rate the pressure level did not increase to 0.3 micron during the eight-hour pressure rise and the rate was determined at 0.1 micron. Since offgassing rate decreases with increasing pressure, one can be assured that the rate will be below the tabulated value if the pressure is above the stated pressure.

For design purposes, the data is presented as plots of offgassing rate as a function of hours of pumping time at the overtemperature. This provides the designer with a means of estimating the required bakeout period to obtain a designated offgassing rate. The pressure at which the offgassing rate is measured is a parameter in this type of plot. Since 0.3 micron is a suitable pressure to obtain good thermal efficiency for Super Insulation at elevated temperature (400 - 1800°F), this pressure is a reasonable choice as a common parameter for these graphs.

To illustrate how the rate results are obtained, consider the pressure rise curves in Figures 66 and 67. The symbols ST refer to Sample Test No. and RC designates Recalibration of Chamber. Shown are the data points from the tantalum foil test and the corresponding empty chamber test. The curves shown were fit by a least squares program for the IBM-360 computer which determined the coefficients of the following equation.

Table XXII

METALLIC FOIL OFFGAS COMPOSITION

Material	Processing Temp. (°F)	Over-Temp. (°F)	Offgassing Temp. (°F)	Gaseous Component						Offgassing Rate at Pressure (P)		Pressure, Microns, Hg	Pressure Rise No.	Hours at Over-Temp.	Sample Run No.	Empty Chamber Run No.
				H ₂ (%)	CO ₂ (%)	H ₂ O (%)	Hydro-Carbons (%)	N ₂ -CO (%)	Max. N ₂ (%)	Min. CO (%)	Rate, Micron-Liters Hr ft ²					
Aluminum Foil	900	1000	900	99.52	--	--	.34	.13	--	--	.102	(0.3)	1	17	7	11
	900	1000	900	97.94	--	--	1.41	.64	--	--	.146	(0.3)	2	34	7	11
	900	1000	900	99.20	--	--	.53	.26	--	--	.08	(0.3)	3	42	7	Estimated
	900	1000	900	89.26	.16	--	1.36	9.2	--	--	.02	(0.3)	4	57	7	Estimated
Aluminum Foil	UP*	1000	900	99.36	--	--	.50	.14	--	--	.00926	(0.3)	1	17	8	11
	UP	1000	900	99.42	--	--	.42	.15	--	--	.0486	(0.3)	2	34	8	11
Aluminum Foil	UP	200	100	40.95	--	54.26	--	4.79	--	--	-.081 -.043	(0.3)	1	17	22	24 26
	UP	200	100	57.47	--	35.13	.17	7.22	--	--	-.012 -.0041	(0.3)	2	34	22	24 26
Tantalum Foil	UP	1900	1800	99.64	--	--	.17	.18	--	--	.0397	(0.3)	1	17	9	13
	UP	1900	1800	--	--	--	.17	.18	--	--	.0090	(0.3)	2	34	9	13
Copper Foil	1550	1370	1270	96.60	--	--	1.54	1.85	--	--	-.0777 -.136	(0.3)	1	17	17	18 19
	1550	1370	1270	95.44	.04	--	1.92	2.58	--	--	-.00588 -.0285	(0.3)	2	34	17	18 19
Nickel Foil	1640	1740	1640	80.03	.09	--	1.59	18.27	2.37	15.90	-.0433 -.00791	(0.3)	1	17	12	15 17
	1640	1740	1640	79.47	.14	--	1.06	19.31	1.18	18.10	-.00352 -.000358	(0.3)	2	34	12	15 17
Nickel Foil	UP	1740	1640	98.63	--	--	.09	1.26	--	--	.000348	(0.3)	1	17	14	17
	UP	1740	1640	97.94	--	--	.16	1.88	--	--	-.000324	(0.3)	2	34	14	17

UP* - Unprocessed

Table XXIII

PROCESSED SPACER OFFGAS COMPOSITION

MATERIAL	PROCESSING TEMP. (°F)	OVER- TEMP. (°F)	OFFGASSING TEMP. (°F)	GASEOUS COMPOSITION						OFFGASSING RATE AT PRESSURE (P)		PRESSURE, MICRONS, HG	PRESSURE RISE NO.	HOURS AT OVER-TEMP.	SAMPLE RUN NO.	EMPTY CHAMBE RUN NO.
				H ₂ (%)	CO ₂ (%)	H ₂ O (%)	HYDRO- CARBONS (%)	N ₂ -CO (%)	MAX. N ₂ (%)	MIN. CO (%)	RATE, MICRON-LITERS HR FT ²					
GLASS FIBER PAPER	1000	1000	900	85.53	---	--	1.36	13.09	4.88	8.21	-.0706	(0.3)	1	17	11	14
	1000	1000	900	78.02	.63	--	.63	20.70	.50	20.19	-.0327 -.00827 -.0082	(0.3)	2	34	11	16 14 16
GLASS FIBER PAPER	1000	200	100	13.50	--	78.05	--	8.45	3.15	5.30	-.325	(0.3)	1	17	21	24
	1000	200	100	50.22	--	42.56	--	7.22	0	7.22	-.111 -.143 -.0632	(0.3)	2	34	11	26 24 26
ALUMINUM FLAKE OPACIFIED GLASS FIBER PAPER	1000	1000	900	99.00	.01	--	.35	.63			-.321	(0.3)	1	17	13	14
	1000	1000	900	98.19	.03	--	.63	1.14			-.149 -.139 -.0754	(0.3)	2	34	11	16 14 16
COPPER FLAKE OPACIFIED REFRASIL QUARTZ FIBER PAPER	1200	1370	1270	17.55	4.55	--	6.39	71.48	3.09	68.39	6.63	(0.3)	1	17	19	19
	1200	1370	1270	27.68	4.53	--	5.16	62.61	4.65	57.96	5.19 3.03 2.80	(0.3)	2	34	11	21 19 21
	1400	1370	1270	8.73	15.44	--	8.88	66.92	2.12	64.79	.241 -.240	(0.3)	1	17	25	30 31
	1400	1370	1270	67.56	---	--	2.12	27.84	1.85	25.98	-.00239 -.0351	(0.3)	2	34	11	30 31
ASTROQUARTZ CLOTH	1700	1900	1800	27.87	.31	--	4.50	67.31	3.64	63.66	.577 .538	(0.3)	1	17	16	13 20
	1700	1900	1800	43.68	---	--	3.65	52.65	2.97	49.68	-.000403 -.295	(0.3)	2	34	16	13 20
	1700	1900	1800	16.34	.36	--	3.65	79.64	2.31	77.31	3.89 3.67	(0.3)	1	17	24	28 32
	1700	1900	1800								3.10 3.48	(0.3)	2	34	11	28 32
	1700	1900	1800	15.75	.32	--	6.35	77.57	3.11	74.45	.0017	(0.3)	3	105	11	ESTIMATED
	1700	1800	1700	58.57	.37	--	3.41	37.63	7.00	30.63	-.169	(0.3)	1	17	28	33
	1700	1800	1700	47.18	.38	--	2.90	49.52	12.78	36.73	-.00133	(0.3)	2	34	28	33
	1700	1900	1800	59.25	.01	--	.48	40.23	27.18	13.04	4.00	(2.0)	1	17	29	34
	1700	1900	1800	25.74	---	--	.15	74.23	53.59	20.62	2.848	(2.0)	2	34	29	34
	1700	1900	1800	5.88	.10	--	5.52	88.48	4.28	84.19	4.58	(0.3)	1	17	18	20
REFRASIL QUARTZ FIBER PAPER	1700	1900	1800	5.51	--	--	6.07	88.40	4.58	83.81	.941	(0.3)	2	34	18	20
	1700	1900	1800	1.93	.07	--	6.08	91.90	4.88	87.02	.55	(0.3)	3	86	18	ESTIMATED
	1650	1550	1550	26.99	2.40	--	7.42	63.17	2.31	60.84		(0.3)	A	17	20	22
	1650	1550	1400	88.39	.12	--	2.17	9.30	2.15	7.14	69.7 2.07 2.07	(0.3)	1	17	20	25 22 25
	1650	1550	1400	67.78	.40	--	5.60	26.19	4.86	21.34	.000163 .000163	(0.3)	2	34	20	22 25
	1650	1550	1550	21.77	5.53	--	8.00	64.63	2.50	62.11		(0.3)	B	51	20	22 25
	1650	1550	1400	88.99	.07	--	2.19	8.74	1.72	7.00	63.7 .0737 .0737 .912	(0.3)	3	51	20	22 25 27
	1650	1550	1550								4.05 -.320	(0.4)	A	17	23	29 27
	1650	1550	1400	93.86	.23	--	.87	5.02	2.03	2.98	-.391	(0.3)	1	17	23	27 29
	1650	1550	1400	91.32	.24	--	1.21	7.21	2.51	4.69	-1.035 -.0110	(0.3)	2	34	23	27 29
	1700	1800	1700	64.88	.40	--	2.86	31.85	3.12	28.70	-.114	(0.3)	1	17	26	33
	1700	1800	1700	50.32	.33	--	2.59	46.69	6.69	39.99	-.0002	(0.3)	2	34	26	33
	1700	1900	1800	31.68	.43	--	1.80	66.07	27.52	38.55	3.49	(2.0)	1	17	30	34
	1700	1900	1800	1.19	.01	--	.13	98.66	64.87	33.80	1.18	(2.0)	2	34	30	34

Figure 66

SECOND PRESSURE RISE FOR TANTALUM
FOIL AND CORRESPONDING EMPTY
CHAMBER TEST

X ST 9 Tantalum
O RC 13 Empty chamber

Figure 67

$$\log p = a_0 + a_1 \log t + a_2 (\log t)^2 \quad (97)$$

where,

P = pressure, microns
 a_0, a_1, a_2 = coefficients
 t = time, minutes

Several more complicated equations were tried that produced smaller standard deviations, but these did not yield curves with constantly decreasing first derivatives, a physical constraint imposed by the offgassing phenomenon. The computer program developed also gives a tabulation of the first derivative as a function of pressure. To obtain the values of offgassing rate shown in Table XXII, the derivative at 0.3 micron pressure was taken from the computer printouts for the sample and empty chamber rises and inserted in Equation (90). The resulting answer for G_s was then divided by the one-side area of the sample to give an offgassing rate per square foot.

Figures 68 and 69 illustrate a phenomenon that occurred frequently in the series of offgassing tests. As the Figures show, the pressure rise curves for the empty chamber fell considerably above the sample curves, indicating that negative offgassing or adsorption was taking place. The materials that exhibit an apparent adsorption rate have apparently become adsorption activated by the overtemperature bake-out before the offgassing test. This behavior occurred with many materials as indicated in Tables XXII and XXIII by the negative rates and was substantiated by repeat runs.

The offgassing data from Tables XXII and XXIII has been plotted in Appendix E. The data was smoothed whenever an obvious inconsistency existed in a manner that would be conservative. That is, the curve was moved upward to the higher offgassing rate when two points seemed inconsistent. Actually, only a few points needed this adjustment. The code given with each curve can be interpreted as follows:

ST	P 900	OG 900	(0.3)
Sample test Run Number	Processing Temp. (°F) (UP if unprocessed)	Offgassing Temp. (°F)	Pressure for Offgassing Measurements

As mentioned previously, if the pressure is higher than the indicated pressure, the offgassing rate will be less than the indicated rate. For a typical system, getter calculations show that rates above 0.1 micron-liter/ft² hr. require sizable amount of getter so that

Figure 68

Figure 69

a high offgasser might be defined as a material whose rate is above 0.1 after 34 hours of pumping at the overtemperature. A short discussion of the offgassing characteristics of each material follows. Offgassing curves for each material are presented in Appendix E.

3.4.2.7 Offgassing Characteristics of Material

A brief discussion of the offgassing characteristics of each material will be presented.

Aluminum foil - At 900°F, processed aluminum is acceptable but unprocessed aluminum is considerably better. Processing does not seem to reduce the offgassing rate. At 100°F, aluminum appeared as an adsorber.

Tantalum foil - Acceptable offgassing rate for unprocessed tantalum at 1800°F.

Copper foil - Adsorption at 1270°F by processed copper.

Nickel foil - Both unprocessed and processed nickel exhibited very low rates. Again it seems unnecessary to process the foil since further reduction in offgassing rate is not required.

Glass fiber paper - Adsorber at both 900°F and 100°F.

Aluminum flake opacified glass fiber paper - Adsorber at 900°F.

Copper flake opacified Refrasil quartz fiber paper - Very low rate if processed at 1400°F. These two curves indicate the dependence of offgassing rate on processing temperature.

Astroquartz cloth - Three tests were made under identical conditions of processing at 1700°F, bakeout at 1900°F and offgassing at 1800°F. The cloth always showed a high offgassing rate during the first pressure rise but sometimes showed a high rate and sometimes a low rate at the second pressure rise. This indicates that these conditions are near a critical point between low and high rates so that small random variations in test conditions markedly affect the rate. To be conservative, the curve showing the highest rate should be used. Small scale processing tests showed that the processing temperature cannot be raised above 1700°F without a phase transition occurring that significantly decreases the strength of the cloth. The cloth has a very low offgassing rate at 1700°F when processed at 1700°F and baked out at 1800°F.

Refrasil quartz fiber paper - Two tests were made with processing at 1700°F, bakeout at 1900°F and offgassing at 1800°F. Both tests showed a high rate. A low offgassing rate at a temperature of 1400°F was obtained for Refrasil paper which was processed at 1650°F and baked out at 1500°F. An adsorption rate was obtained at a temperature of 1700°F when processing was conducted at 1700°F and bakeout at 1800°F. As with the Astroquartz cloth, small scale processing tests showed that increasing the processing temperature above 1700°F resulted in a sharp decrease in strength of the Refrasil paper. The offgassing rate measured at the overtemperature (1550°F)

10

is seen to be considerably higher than the rate measured at 1400°F.

The rates presented in Appendix E are summarized in Figure 70. Where disagreement existed, the highest (most conservative) rate curve was used. Materials displaying an adsorption rate were assigned zero off-gassing rates. Curves for processed aluminum and nickel foil are not shown since processing is no longer recommended for these foils. Tantalum is not shown because it is not recommended.

Table XXIV summarizes the composition data from Tables XXII and XXIII using the composition of the second pressure rise since this is the composition that would be exposed to the getter for systems with a 34-hour bakeout period. (The compositions are usually almost the same for both rises).

Table XXIV should be used in conjunction with Figure 70 to determine getter requirements.

OFFGASSING RATE MICRON LITERS/FT² HR

Figure 70

TABLE XXIV

INSULATION MATERIALS OFFGAS COMPOSITION FOR GETTER DESIGN

<u>Material</u>	<u>Processing Temp. (°F)</u>	<u>Over- Temp. (°F)</u>	<u>Offgassing Temp. (°F)</u>	<u>Gaseous Composition</u>				
				<u>H₂ (%)</u>	<u>CO₂ (%)</u>	<u>H₂O (%)</u>	<u>Hydro- Carbons (%)</u>	<u>CO (%)</u>
Aluminum Foil	UP	1000	900	99.42	--	--	.42	.15
Tantalum Foil	UP	1900	1800	99.64	--	--	.17	.18
Nickel Foil	UP	1740	1640	97.94	--	--	.16	1.88
Copper Opacified Quartz Paper	1400	1370	1270	67.56	--	--	2.12	27.84
Quartz Cloth	1700	1900	1800	25.74	--	--	.15	74.23
Quartz Cloth	1700	1800	1700	47.18	.38	--	2.90	49.52
Quartz Paper	1700	1900	1800	1.19	.01	--	.13	98.66
Quartz Paper	1700	1800	1700	50.32	.38	--	2.59	46.69
Quartz Paper	1650	1550	1400	88.99	.07	--	2.19	8.74

3.4.2.8 Sample Getter Calculation

To illustrate getter requirement calculations, a hypothetical system with a design life of 5 years operating between 70°F and 1200°F will be examined. Insulation materials that might be required for the system are as follows:

<u>Temperature (°F)</u>	<u>Foil</u>	<u>Spacer</u>
900-1200	Cu	Quartz paper
70- 900	Al	Glass fiber paper

Capacitys of Linde getter are approximately:

<u>Gas</u>	<u>C, Capacity (ul/gm)</u>
H ₂	100,000
CO	8,000
CO ₂	5,000
H ₂ O	10,000
Hydrocarbon	23,000

From Figure 70 after a 34-hour bakeout period, the following offgassing rates would be observed.

<u>Material</u>	<u>Temp. (°F)</u>	<u>Offgassing Rate (ul/ft²hr)</u>
Cu	1270	0
Al	900	0.045
Glass paper	900	0
Quartz paper	1700	0.0018

Since offgassing rate increases with increasing temperature, we are conservative when using the offgassing rate at the maximum temperature of the material as the offgassing rate of the entire insulation thickness.

Assuming that 100 ft² of aluminum foil is present, getter requirements are calculated as follows: In 5 years, 100 ft² of aluminum would offgas:

$$(.045 \text{ ul/hr.ft}^2) (100 \text{ ft}^2) (5 \text{ yrs.}) \left(\frac{8765 \text{ hrs.}}{\text{yr.}}\right) = 197,000 \text{ ul}$$

From Table XXIV, the amount of each gas would be:

$$\begin{aligned} \text{H}_2, (.9942) (197,000) &= 195,900 \text{ ul} \\ \text{CO}, (.0015) (197,000) &= 296 \text{ ul} \end{aligned}$$

Hydrocarbon, (.0042) (197,000) = 826 μ l

Using the capacities given, the getter requirements for aluminum would be:

$$\frac{195,000 \mu\text{l}}{100,000 \mu\text{l/gm}} + \frac{296 \mu\text{l}}{8,000 \mu\text{l/gm}} + \frac{826 \mu\text{l}}{23,000 \mu\text{l/gm}} = 2.03 \text{ gm}$$

In a similar matter, the getter requirements for the other materials can be calculated. For a typical system, getter calculations show that rates above 0.1 micron - liter/ft²-hr. require sizable amount of getter, so a high offgasser might be defined as a material whose rate is above 0.1 after 34 hours of pumping at the overtemperature.

3.4.3 Vacuum Enclosure

The objective of the vacuum enclosure investigation was to select materials of construction for a typical vacuum enclosure environment and conduct small scale tests to develop design confidence.

The initial environmental requirements for the vacuum enclosure dictated that the inside wall of a vacuum enclosure withstand 1800°F with air on one side and vacuum on the other. These requirements were reviewed with the AEC midway through the investigation and based on radioisotope thermoelectric generator requirements in the near future, a temperature requirement of 1400°F was defined.

The initial efforts of this work, Section 3.4.3.1 Material Selection, 3.4.3.2 - Super Alloy Problem Areas and Material Selections, and 3.4.3.3 - Inconel 600 Enclosure Specimen Testing, were directed toward the 1800°F temperature level, while Section 3.4.3.5 - Hastelloy X Enclosure Specimen Testing deals with the 1400°F temperature objective.

3.4.3.1 Material Selection Study

Because of the high temperature conditions (1000-1800°F) in which the vacuum enclosure must operate, with one wall exposed to vacuum and the other exposed to air, the superalloys were deemed the most suitable materials of construction. The possibility of coated refractories was investigated but in general the coating of refractory metals for long term service in an oxidizing environment has not yet been completely successful.

A preliminary list of candidate materials and their compositions is shown in Table XXV⁷, the Candidate Materials table. These preliminary candidates were determined to be the super alloys most suitable for the service requirements, 1800°F operation in vacuum and in air, by literature search, vendor contact and discussions with in-house metallurgical consultants.

Based on the enclosure requirements, the criteria list of Table XXVI was generated which was used as a guide in selecting the candidate vacuum enclosure materials. Each of the properties listed in Table XXVI are discussed in detail in Appendix F, together with supporting data.

3.4.3.2 Super Alloy Problem Areas and Candidate Material Selection

The material selection study, Section 3.4.3.1, disclosed three main problem areas:

TABLE XXV
LIST OF CANDIDATE MATERIALS

Alloy Name	COMPOSITION, WEIGHT PER CENT													Vendor
	C	Mn	Si	Cr	Co	Mo	W	Cb + Ta	Ti	Al	Fe	Ni	Other	
Hastelloy-X	.05/ .15	1.00*	1.00*	20.5/ 23.0	.5/ 2.50	2/10	.020/ 1.00				17/20	Bal.		Haynes-Stellite
Haynes-25	.05/ .15	1/2	1.00	19/21	Bal.	----	14/16				3.00*	9/11		Haynes-Stellite
Inconel-600	.04	.20	.20	15.8							7.20	76	.0075 .10Cu	International Nickel 184
Inconel-625	.10*	.50*	.50*	20.0/ 23.0	1.00*	8.0/ 10.0		3.15/ 4.15	.40*	.40*	5.00*	Bal.		International Nickel
Rene-41	.09	----	----	19.0	11	10			3.10	1.50	----	Bal.		General Electric

*Maximum

TABLE XXVICRITERIA FOR COMPARISON OF CANDIDATE ENCLOSURE MATERIALS

Availability
Compatibility
Diffusion rate of air
Elongation
Fatigue strength
Machineability
Melting practice
Notch sensitivity
Oxidation resistance
Phase change embrittlement resistance
Strength, creep
Strength, rupture
Thermal conductivity
Vaporization rate
Weldability

- 1). The vaporization of alloy constituents at high temperature in a vacuum environment.
- 2). Intergranular oxide penetration after long exposure time, 1-5 years at elevated temperature in air.
- 3). Possible diffusion of air components through super alloys.

A detailed discussion of these problem areas is presented in Appendix F.

After an extensive literature search and consultation with recognized authorities in this field the following conclusions were drawn.

1). The superalloys which appeared to best meet the vacuum enclosure requirements were those with high oxidation resistance, such as Inconel-625, Inconel-600, and Hastelloy-X. These were the materials selected for experimental evaluation.

2). Testing appeared to be the best way of determining the suitability of the candidate materials.

3). Precipitation hardenable superalloys should be avoided because of changes in physical properties at elevated temperature.

3.4.3.3 Test Apparatus and Specimen Design

The test apparatus is shown schematically in Figure 71. Basically, the apparatus consists of an electrically heated tube furnace housing a vacuum chamber tube. Test specimens fabricated from the candidate enclosure materials are inserted into the vacuum chamber tube for testing. Testing consisted of maintaining the specimen at elevated temperature, exposing to the external surface to vacuum and pressurizing internally with air. In this way the specimen can be stressed to a predetermined level based on test enclosure geometry and air pressure. Samples of insulation materials, arranged so that they were at a temperature level corresponding to their operational limits, were placed in the vacuum chamber tube so that:

- 1). The test specimen is exposed to a vacuum environment typical of actual service conditions and
- 2). Material compatibility evaluations could be made on the insulation material and enclosure specimen.

A detailed test procedure and data analysis procedure is presented in Appendix F.

VACUUM ENCLOSURE - MATERIAL TESTER

Figure 71. Vacuum Enclosure Material Tester

3.4.3.4 Inconel-600 Enclosure Specimen Fabrication and Testing

Test enclosure specimen No. 1, Inconel-600, consisted of a 3-1/2 inch O.D. tube, 12 inches in length. The tube was fabricated from cold rolled, deep drawn, pickled and annealed 0.018 inch thick sheet stock. The test enclosure specimen included three welds, a longitudinal seam weld, and an edge weld to each end plate. A tungsten inert gas weld was utilized at each weld for the entire assembly. The specimen was leak tested after fabrication with a helium mass spectrometer leak detector sensitive to 10^{-10} cc air STP/sec.

The specimen and insulation compatibility test samples were attached to the holding fixture as shown in Figure 72, extreme right. Compatibility test samples of insulation materials were placed in the furnace tube vacuum space so that they were at a temperature level within their operational limits. The specimen itself was also wrapped with approximately five layers of Astroquartz cloth and tantalum foil for further material compatibility test purposes. Close-up photographs of the insulation materials and enclosure test specimen are shown in Figures 73 and 74 respectively. The test specimen and insulation samples, Figure 72 extreme right, were inserted into the vacuum chamber tube, Figure 72 extreme left, and testing initiated.

Test conditions consisted of maintaining the specimen at 1800°F with an external vacuum and one-atmosphere of air pressure internally. One atmosphere of air pressure internally yielded a hoop stress of approximately 1300 psi in the test specimen cylindrical portion. This stress is 300 psi less than the 2000 hour rupture stress and 150 psi greater than the 10,000 hour rupture stress of 1150 psi. A helium mass spectrometer was utilized to leak test the test specimen after it was brought up to a temperature of 1800°F. After 115 hours of operation at 1800°F a vacuum pump blank-off test yielded a pressure of 8×10^{-5} torr. There was no measurable pressure rise during a four-hour blank-off test. A mass spectrometer analysis of the gas indicated 65.3% hydrogen, 22.6% nitrogen, 11.8% water and trace quantities of hydrocarbons. It is believed that the water vapor present is picked up from the vacuum transfer lines from the vacuum tube to the mass spectrometer. A helium mass spectrometer leak test after 140 hours indicated a leak on the order of 1×10^{-7} cc air STP/sec. A helium mass spectrometer leak test after 260 hours of operation indicated leakage on the order of 6×10^{-7} cc air STP/sec. After 300 hours of operation a leak test indicated complete loss of vacuum integrity and the test was terminated.

3.4.3.4.1 Inconel-600 Enclosure Specimen Test Analysis

The enclosure specimen and insulation compatibility test samples after removal from the vacuum chamber are shown in Figure 75. Figure 76 is a close-up photograph of the Inconel-600 specimen wrapped with Astroquartz cloth insulation and tantalum foil. After removal of the tantalum and Astroquartz cloth, Figure 77, it was noted that the metal beneath the insulation appeared to have undergone less vaporization

Figure 72. Vacuum Enclosure Test Components

Figure 73. Insulation-Materials Compatibility Test Assembly

Figure 74. Inconel-600 Test Specimen Before Testing

Figure 75. Inconel-600 Test Specimen and Insulation Materials
Compatibility Samples After Testing

Figure 76. Inconel-600 Test Specimen After Testing with Astroquartz Cloth

Figure 77. Inconel-600 Test Specimen After Testing with Astroquartz Cloth Removed

than the exposed portion. This could be due to a condensation of vaporized metallic components under the insulation which did not occur on the exposed portion. Analysis of metallic deposits collected on the Astroquartz cloth disclosed high concentrations of chromium and manganese substantiating this theory. The exposed portion of the specimen was bright and shiny contrasting with the dull appearance of the surface beneath the insulation.

The specimen surface was examined under an optical microscope, exact leakage areas were determined by helium mass spectrometer leak detection and specimen microstructures made of parent material and weld areas. Details of these metallurgical examinations are presented in Appendix F.

Failure Analysis

It is believed that the cause of failure was due to subsurface oxidation penetration greatly accelerated by the stress level, 1300 psi. The presence of subsurface oxidation is indicated in Figures F-10 and F-12 of Appendix F by the dark void areas in the specimen cross-section. Depth of penetration appears high for the test duration, 300 hours, when compared to similar previous experiments. Brunhouse and Titus⁸ reported a maximum internal oxidation penetration of about 1.3 mils for Inconel at 1750°F over 1000 hours in 300 psi air.

The increased depths of subsurface oxide penetration and hence loss of vacuum integrity are likely due to a combination of these effects:

1. Effect of Stress on Diffusion⁵³

Stress can increase subsurface oxidation because increased diffusion rates occur within the stress areas, thereby increasing the frequency with which metals and oxygen react.

2. Effect of Stress on Metal Deformation⁵³

Under the influence of stress the rate of metal deformation can exceed the rate of growth of oxides. Protective oxides normally cover a metal with a self-healing layer which is in compression. However, if the metal beneath the oxide layer is elongated at such a rate that the oxide formed is not sufficient to keep up with the rate of elongation, fresh metal will continually be exposed as oxidation proceeds. Lund and Wagner⁵³ observed that this stress level roughly corresponded to the stress required to produce 1% elongation in 100 hours.

The stress level chosen for the specimen tests was reconsidered from the viewpoint of a practical design stress criterion that would typically be chosen for an actual application. For a long term, 5 year application the design stress in most cases would be based on a

creep stress to produce a maximum allowable total elongation. It was therefore decided that in future testing of enclosure specimens the stress level would be based on a creep stress rather than a rupture stress.

3.4.3.5 Hastelloy-X Enclosure Specimen Fabrication and Testing

Nominal dimensions of the Hastelloy-X test specimen were 3-1/4" O.D. x 12" long with a wall thickness of 0.012". The sheet stock from which the tube was fabricated was cold rolled and annealed (2,150°F). The specimen included three welds, a longitudinal seam weld and an edge weld to each end plate. Tungsten inert gas welding was utilized for the entire assembly.

Test conditions consisted of maintaining the specimen at 1400°F with an external vacuum and an internal air pressure of 37.2 psia. The internal air pressure of 37.2 psia was required to obtain a 5000 psi hoop stress in the specimen.

Previously, it was planned that this specimen would be stressed to the 50,000 hour 1% creep stress. However, examination of vendor literature and discussions with personnel at the Stellite Division of Union Carbide Corporation disclosed that available creep stress data may not apply for thin sections, 0.010 through 0.020 inches and that some reduction in the published creep strength data might occur. Because of the lack of available creep stress data for thin sheets over long time periods, it was decided to select a shorter time basis for the stress level. The stress chosen was one half of the published stress value for a 1% total elongation in 2000 hours at 1400°F. This stress is 5000 psi and was maintained within ± 100 psi during the testing.

A mass spectrometer analysis of the constituents present in the vacuum space external to the specimen was determined at regular intervals throughout the 2000 hour test period. An analysis at the initiation of the test indicated the following major components: 58% hydrogen, 21% water, 18% carbon monoxide, and trace quantities of nitrogen and carbon dioxide.

Samples, approximately 5 square feet in area, of each of the following insulation materials were included in the vacuum chamber tube: aluminum foil, copper foil, nickel foil, glass fiber paper, Refrasil quartz fiber paper, and Astroquartz cloth. These materials were placed in the vacuum chamber tube in such a manner that they were at temperature levels within their operational limits. The insulation materials provide a vacuum environment typical of actual service conditions for the test specimen during the test and will be examined at the conclusion of the test for evidence of incompatibility.

In order to investigate compatibility of the Hastelloy-X test specimen with typical radioisotope power system enclosure materials, test coupons of Hastelloy-X have been placed in direct contact

with coupons of beryllium and Hastelloy-C (power system enclosure materials) on the air side of the test enclosure test specimen.

3.4.3.5.1 Hastelloy-X Enclosure Specimen Test Analysis (2000 Hours)

Testing of the enclosure upto 2000 hours was completed in Phase I. Further testing upto 10,000 hours was conducted in Phase II. Results of this entire test effort, however, are discussed in these following sections to provide unity.

3.4.3.5.2 Hastelloy-X Enclosure Specimen Test Analysis (10,000 hours)

Analysis of the Hastelloy-X vacuum enclosure specimen, exposed to simultaneous air and vacuum environments at 1400°F for 10,000 hours continuous testing, was completed in Phase II. Effects of this high temperature environment on mechanical properties and overall materials stability and compatibility are summarized.

The 10,000 hour long-term test run was interrupted at intervals of 2,000, 4,000 and 6,000 hours in order to: (1) check gas leak rate through the specimen, (2) analyze residual gas in vacuum space, (3) measure circumferential creep and (4) observe any vacuum-side physical changes in base metal surface or weld areas. In all cases, these data were compared to initial, time-zero values.

After completing the 10,000-hour cycle, a final set of the above data were taken. In addition, an overall evaluation of the exposed Hastelloy-X specimen was made, including tensile strength testing, compatibility sample evaluation, and photomicrograph studies of cross-sections of selected base metal and weld area samples.

3.4.3.5.2.1 Vacuum Integrity and Materials Stability

The stability of the Hastelloy-X specimen in a vacuum environment is indicated by the results summarized in Table XXVII. The specimen remained virtually leak-tight throughout the test period, with the measured rate (helium mass spectrometer testing) less than 1×10^{-8} STD cc air/second. The consistency of the residual gas analysis of the vacuum space gave further indication of specimen tightness at 1400°F.

A carbide coating on the exterior surface of the Hastelloy-X enclosure specimen is clearly shown on Figure 84. Observations at interim periods established that the coating formed in the period between the 6,000 and 10,000-hour inspections. Preliminary analysis of the coating indicated predominately chromium carbide. The chromium, an alloy constituent of the Hastelloy-X, could have been available at the vacuum surface by virtue of vaporization of this constituent at 1400°F. Speculation on the presence of carbon and/or chromium carbide is as follows:

(1) Migration from the Inconel furnace tube (100-200°F higher than the sample) and deposition on the 1400°F Hastelloy-X surface.

(2) Presence of hydrocarbons, due to back-streaming* of pump oil into the furnace chamber, which could have reacted with the free chromium.

The fact that the coating formed between 6,000 and 10,000 hours intervals would support the second possibility. In any event, the carbide coating appears to have had no significant effect on the behavior of the Hastelloy-X specimen.

* The cold-trap on the pumping system, which had been operational up to 6000+ hours, was inadvertently allowed to warm to room temperature.

Hastelloy-X Specimen and Insulation
Material Compatibility Test Samples
after 2000 hours of testing.

Figure 78

Hastelloy-X Specimen
after 2000 hours of
testing.

HASTELLOY-X SPECIMEN SURFACE, AS RECEIVED
(18X - mag.)

HASTELLOY-X SPECIMEN, VACUUM SURFACE AFTER 2000 HOURS TESTING
(18X - MAG.)

HASTELLOY-X SPECIMEN, VACUUM SIDE LONGITUDINAL WELD AFTER 2000 HOURS
TESTING
(18X - MAG.)

HASTELLOY-X SPECIMEN, VACUUM SIDE EDGE WELD AFTER 2000 HOURS TESTING
(18X - MAG.)

Photographs (18 power magnification) of the base metal and weld surfaces are given in Figures 85 and 86. Vacuum surfaces are pictured at 0, 4,000, and 10,000-hour exposure times. The oxidized surface (air side) is shown after 10,000-hour exposure. In general, no obvious detrimental effects in either the weld area or base metal could be detected at any of the exposure conditions. The carbide coating can be noted on the 10,000-hour, vacuum-side photographs of base metal and weld.

The 10,000-hour inspection of the Hastelloy-X/Hastelloy-C and Hastelloy-X/Beryllium compatibility samples, which had been air exposed at 1400°F, revealed the following:

(1) The Hastelloy-X/Hastelloy-C sample indicated no interaction or individual deterioration at the interface.

(2) No interaction between the Hastelloy-X and beryllium. The beryllium, however, had severely oxidized.

In the 10,000-hour, 1400°F exposure, essentially all the beryllium had oxidized (BeO) and turned to powder. This occurrence is supported by outside data on oxidation of beryllium. This reference reports that "the thin oxide that forms on beryllium on exposure to air at ambient temperature is highly protective up to about 1100°F. Above 1400°F, the oxide film becomes unprotective and, in time, the metal can turn to powder". Apparently, under the pressurized air environment (37.2 psia), the critical temperature was exceeded at the indicated 1400°F level.

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Figure 84 - Hastelloy-X Vacuum Enclosure Specimen After
10,000-Hour, 1400°F Exposure

FIGURE 85 - SURFACE PHOTOGRAPHS (18X)-HASTELLOY-X BASE METAL

AS- RECEIVED - VACUUM SIDE

4,000 HOURS, 1400°F EXPOSURE
VACUUM SIDE

10,000 HOURS, 1400°F EXPOSURE
VACUUM SIDE

10,000 HOURS, 1400°F EXPOSURE
AIR SIDE

LONGITUDINAL WELD - NO EXPOSURE

LONGITUDINAL WELD AFTER 4000 HOURS, 1400°F
VACUUM SIDE

LONGITUDINAL WELD AFTER 10,000 HOURS, 1400°F
VACUUM SIDE

LONGITUDINAL WELD AFTER 10,000 HOURS, 1400°F
AIR SIDE

TABLE XXVII

HASTELLOY-X ENCLOSURE - VACUUM INTEGRITY

Testing Time	Leak Rate* cc air(STP)/sec.	Vacuum Space* Gas Analysis	Observations
Initial (0 time)	$< 1 \times 10^{-8}$	58% - H ₂ 21% - H ₂ O 18% - CO Trace - N ₂ & CO ₂	Hastelloy-X specimen clean; in as-received condition.
2000 hours.	$< 1 \times 10^{-8}$	78% - H ₂ 13% - CO 7.5% - N ₂ 1.2% - CH _x [Air diffusion through specimen not indicated]	Vacuum side surface of Hastelloy-X specimen slightly tarnished; possibly a light oxide coat developed during slightly higher vacuum pressure periods in early pumpdown stages. Surface photographs (18X) show no visible change in base metal or weld surfaces. Vaporization does not appear to be significant.
4000 hours	$< 1 \times 10^{-8}$	49% - H ₂ 24% - CO + N ₂ 14% - CH _x 13% - CO ₂ [No air diffusion indicated]	Observations at 2000 hours apply.
6000 hours	$< 1 \times 10^{-8}$	-----	Observations at 2000 hours apply.
10,000 hours	$< 1 \times 10^{-8}$	81% - H ₂ 9% - N ₂ + CO 7.4% - CH _x 2.6% - CO ₂ [No air diffusion indicated]	The Hastelloy-X tube specimen was covered entirely by a light coating, loosely held in the central (1400°F) portion of the tube and tightly bonded in the end regions (see Figure 2.5.2). Analysis indicated a carbide coating, most likely chromium carbide. In areas where coating was removed, the Hastelloy-X surface appeared normal.

* Leak test and gas analysis
performed with specimen at 1400°F

3.4.3.2.2 Mechanical Properties

Tensile strength, yield strength, and total specimen creep were measured for the as-received Hastelloy-X specimen and the 10,000-hour, 1400°F exposed specimen. The results were compared and referenced to manufacturer's published data showing effects of 1400°F aging on strength of Hastelloy-X sheet.

Total Specimen Creep

Total elongation of the Hastelloy-X specimen, measured in the circumferential direction (loaded under equivalent 5000 psi hoop stress), was obtained at intervals throughout the long-term test. Results are as follows:

Test Time hrs	Temperature °F	Stress psi	Total Elongation %
0	1400	5000	0
2000	1400	5000	0.31
4000	1400	5000	0.62
6000	1400	5000	0.62
10,000	1400	5000	0.79

As indicated, total specimen elongation was less than 1% for the 10,000-hour exposure.

Manufacturer's data giving stress required for 1% total elongation in Hastelloy-X sheet for various temperatures and times, are given in Figure 87. The 1400°F line has been estimated and included. In designing the total creep test, it was assumed**that Hastelloy-X in thin section (.010-.020 in.) would exhibit reduced creep strength in comparison to the published data on thicker sheet. As a result, the test stress level was chosen as one-half the corresponding published value for a particular temperature, time condition. The selected "target" condition is indicated on Figure 87; ie., for 1% total elongation in 2000 hours at 1400°F, a stress level of 5000 psi was selected.

The actual results indicate that the one-half strength assumption for the .012 in. Hastelloy-X sheet was conservative. In fact, the preliminary indication is that the published data may be quite suitable for predicting creep behavior of thin Hastelloy-X sheet.

* "Target" condition

** Assumption based on a study of the published data and discussions with manufacturer's technical personnel.

Figure 87 - Average One Percent Total Elongation - Hastelloy-X Sheet

Comparison of published and actual data, in this case, may be somewhat imprecise since the actual cylindrical specimen was subjected to two-dimensional creep stress (longitudinal as well as circumferential). However, the longitudinal stress level (estimated at 2500 psi) is sufficiently low so that its effect at 1400°F is assumed small. No change in the longitudinal dimension was observed after 10,000 hours.

Tensile and Yield Strength

Ambient temperature strengths of Hastelloy-X base metal and weld zone samples were determined. Table XXVIII gives data on ultimate and yield strength and percent elongation for as-received and 10,000-hour, 1400°F aged materials. The samples were prepared such that the tensile loading was applied in the circumferential direction, and perpendicular to the longitudinal weld.

Based on the results of Table XXVII the following observations are made:

- (1) A noticeable deterioration in ultimate tensile strength was brought about by the 1400°F exposure. Reduction of strength was particularly significant in the weld areas.
- (2) Marked increase in brittleness (loss of elongation) in both the base metal and weld samples.
- (3) The reduction of properties has been attributed to general precipitation of carbides, which has been described as characteristic of Hastelloy-X in 1400°F exposure.
- (4) Properties deterioration due to grain boundary attack at the vacuum and oxidized surfaces is considered minor.
- (5) Substantial deterioration in properties at the weld areas is attributed to porosity of the weld "heat affected zone" (HAZ). This porosity, which occurred under 1400°F exposure, did not extend to the weld nugget. All 1400°F weld samples failed in the HAZ.

Further discussion on causes of properties deterioration for the 1400°F samples will be presented in Section 3.4.3.5.2.3, in conjunction with the photomicrographs.

Published data on room temperature strength and elongation of 1400°F aged Hastelloy-X are presented in Figures 85 and 86. Data are extrapolated from 1000 hours to 10,000 hours (as indicated by the dashed line). Measured strength data on as-received and 1400°F base metal and weld samples are included for comparison.

The strength and elongation data for Hastelloy-X base material are in good agreement (~10%) with the extrapolated literature data. This implies that the Hastelloy-X, in thin (.012 in.) section, behaves in characteristic fashion when exposed to long-term 1400°F aging. The surface effects due to the combination vacuum and oxidizing environments do not significantly alter this behavior. This appears to lend further credence to the utility of literature data in estimating the effects of these rather specialized high temperature environments on Hastelloy-X mechanical properties.

TABLE XXVIII
ROOM TEMPERATURE STRENGTH OF
HASTELLOY-X VACUUM ENCLOSURE MATERIAL

	As-Received Material				10,000-hr, 1400°F Aged Material			
	Base Metal		Weld Zone		Base Metal		Weld Zone	
	A	B	A _w	B _w	A	B	A _w	B _w
Ultimate Strength (KSI)	118.8	118.8	109.8	115.5	109.0	103.9	85.2	55.7*
Yield Strength .2% Offset (KSI)	55.0	48.3	57.3	52.4	59.8	56.5	65.5	—*
Elongation (%)	42.	38.	24.	33.	8.	7.	3.	2.
Fracture Location	B	B	<u>HAZ</u> W.I.	B.	B	B	<u>HAZ</u> W.I.	<u>HAZ</u> W.I.

W.I. -- weld interface

B -- base material

HAZ -- heat-affected zone

* Fracture below yield

Strength data for the as-received weld sections were also in good agreement with the literature data. Elongation values were substantially lower, possibly due to embrittlement of the HAZ during welding. The very low 10,000-hour ultimate strengths, in comparison with the extrapolated literature values, again point up the significant effect of the severe porosity observed in the HAZ.

3.4.3.4.2.3 Hastelloy-X Photomicrographs

Photomicrographs (200X magnification) of typical Hastelloy-X base metal sections, at time zero and after 10,000 hours, 1400°F exposure, are presented in Figure 90. Samples A_e and B_e are identical to A and B, the former having been etched to show grain structure.

The vaporization of alloy constituents and/or impurities in the grain boundaries is indicated at the vacuum-side surface. Some mild porosity is also apparent in the Hastelloy-X cross-section, close to the vacuum surface. This porosity is quite random and does not appear to have a significant effect on strength properties. Surface penetration due to oxidation at the "air-side" surface is minimal. The chemically etched photomicrographs clearly illustrate the general carbide precipitation at 1400°F, which results in loss of strength and ductility as discussed in Section 3.4.3.5.2.2.

Photomicrographs of the weld "heat-affected zone" (HAZ) after 10,000 hours, 1400°F exposure are shown in Figure 91. The acute porosity, which appears to be responsible for the sharp reduction in weld zone strength, is clearly illustrated. The following observations are made:

- (1) In opposition to what was seen on the base metal photographs, the porosity in the HAZ is concentrated nearer the oxidized side of the Hastelloy-X sample.
- (2) Surface oxidation effects are more pronounced than on the base metal samples.
- (3) The weld nugget is not affected.

Cross-sections of typical original (before 1400°F exposure) weld areas, fabricated with materials and techniques identical to those used in making the welds shown in Figure 91, were also studied under 100X magnification. No evidence of porosity in the heat affected zone was noted on these original welds (Figure 92). This implies, at least, that the porosity in some way results from the 1400°F, long-term exposure and is not caused by the original welding operation.

The porosity problem and its adverse effect on strength are well illustrated. Its cause is not readily explained. It is felt that in-depth metallurgical investigation is required to gain the necessary insight into this phenomenon. This type of study was beyond the scope of our Phase II contractual efforts.

Figure 88 - Room Temperature Strength of Aged Hastelloy-X

Figure 89 - Room Temperature Tensile Elongation of Aged Hastelloy-X

FIGURE 90 - PHOTOMICROGRAPHS (200X) OF HASTELLOY-X BASE METAL CROSS-SECTION

VACUUM
SIDE

AIR SIDE

A. AS-RECEIVED

B. 10,000-HOUR, 1400°F EXPOSURE

VACUUM
SIDE

AIR SIDE

A_e

AS-RECEIVED

B_e

10,000-HOUR, 1400°F EXPOSURE

FIGURE 891 - PHOTOMICROGRAPHS - HASTELLOY-X
WELD AREA CROSS-SECTION

VACUUM SIDE

WELD NUGGET

AIR SIDE

(200X) WELD HEAT-AFFECTED ZONE

VACUUM SIDE

WELD NUGGET

AIR SIDE

(100X; ETCHED) WELD HEAT-AFFECTED ZONE

Figure 92 - Photomicrograph of Unexposed Hastelloy-X
Weld Area

Weld Nugget

(100X; Etched) Weld Zone
No High Temperature Exposure

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3.5 Insulation Environmental Evaluations

3.5.1 Stability and Compatibility

The purpose of this work was to investigate any changes that take place in the insulation materials after processing or after long term, 2000 hour, exposure to operating temperatures and vacuum. Included are physical changes, such as reduction of tensile strength, that occur in spacer materials after processing as well as compatibility observations made on insulations after long term temperature - vacuum exposure. Initial testing involved tensile strength tests, shrinkage and weight loss tests on candidate spacer materials before and after processing. Tensile strength tests were also conducted on selected candidates after 2000 hour thermal conductivity tests. The compatibility of insulation materials with themselves (thermal conductivity test samples) and with candidate vacuum enclosure material was determined after 2000 hour exposure to operating temperature and vacuum.

3.5.1.1 Tensile Tests

Tensile tests were performed in triplicate on the candidate spacers in the "as received" condition and after processing. The tests were conducted with a Dillon tensile tester, Figure 93. The results are summarized in Table XXIX. Inspection of the table shows that the strength of Astroquartz cloth drops off rapidly above 1500°F, but it still retains more strength than the paper-type spacers. Refrasil cloth, Irish Refrasil and Astrosil cloth perform well from a strength viewpoint but are not as attractive as Astroquartz cloth because of their thickness, nominal 14 mils. The strength of the paper-type spacers is approximately four to seven times less than their original strength after processing with the exception of glass fiber paper which loses about 30% of its strength.

Astroquartz Cloth

Tensile strength measurements were made on samples of Astroquartz cloth taken from the nickel foil-Astroquartz cloth combination thermal conductivity tests, Section 3.3.1.5.5. The tensile strength tests were performed in triplicate using the Dillon tester. The nickel foil-Astroquartz cloth combination was not a 2000-hour test specimen, but had undergone approximately 600 hours of testing during which time the hot face temperature was varied from 503 to 1947°F.

Spacer samples were taken from three different temperature "zones" of the insulation system:

- 1). Cold face zone: fifth layer counting from the outside or cold boundary.
- 2). Intermediate zone: 30th layer counting from the outside cold face boundary.

Figure 93 Dillon Tensile Tester

TABLE XXIXSPACER TENSILE TESTS

<u>Material</u>	<u>As Received</u>	<u>BREAKING STRENGTH (LB/IN.-WARP DIRECTION)</u>				
		<u>After Oxidizer Gas Processing</u>				
		<u>800°F</u>	<u>1200°F</u>	<u>1280°F</u>	<u>1500°F</u>	<u>1800°F</u>
Refrasil Cloth No. C10028	76.5					54.3
Irish Refrasil No. C1554-28	56.0					42.0
Astroquartz No. 503	66.0		43.5		24.5	3.9
Astrosil No. 82	80.0					48.2
Refrasil Quartz Fiber Paper	0.28					0.063
Aluminum Flake Opacified Glass Fiber Paper	0.55	0.084				
Copper Flake Opacified Refrasil Quartz Fiber Paper	0.22			0.03		
Glass Fiber Paper	0.105	0.069				

- 3). Hot face zone: 55th layer counting from the cold face boundary.

Approximate zone temperatures and exposure times are shown below:

<u>Exposure Time, hr.</u>	<u>Approximate Temperature, °F</u>		
	<u>Hot Zone</u>	<u>Intermediate Zone</u>	<u>Cold Zone</u>
72	500	400	130
72	850	700	212
120	880	750	220
120	1270	1150	329
48	1420	1280	370
72	1630	1500	450
96	1947	1700	550

A decrease in strength with exposure to increased temperature was noted during examination and preparation of the tensile strength test samples. This was verified by the test results presented in Table XXX.

TABLE XXX

ASTROQUARTZ CLOTH TENSILE TEST (600 Hour Temperature Exposure)

<u>Insulation Spacer Location</u>	<u>Breaking Strength, lb/in Warp Direction</u>
<u>Cold Face Insulation Zone:</u> 5th insulation layer from cold face - approximate temperature exposure - 130°F - 550°F	18.30
<u>Middle Insulation Zone:</u> 30th insulation layer from cold face - approximate temperature exposure - 450°F - 1700°F	4.50
<u>Hot Face Insulation Zone:</u> 55th insulation layer from cold face - approximate temperature exposure - 500°F - 1947°F	1.36

Although tensile strength decreased after high temperature exposure all samples were still handleable.

Tensile strength measurements were made on samples of Astroquartz cloth taken from the tantalum foil-Astroquartz cloth 2,000 hour thermal conductivity test, Section 3.3.1.5.7.

Astroquartz cloth samples were taken from two different temperature zones of the insulation system:

- 1). Cold Face Zone: third spacer layer counting from the outside or cold face boundary.
- 2). Intermediate Zone: 21st spacer layer counting from the outside or cold face boundary.

Tensile test measurements were attempted on the Astroquartz cloth samples in the hot zone - 21st through 42nd layers, but the material was so brittle that the tensile test specimen could not be fabricated. The results of the tensile strength measurements together with approximate exposure temperatures and times are presented in Table XXXI. A decrease in the tensile strength of the Astroquartz cloth with exposure to the higher temperatures can again be seen.

Tensile strength measurements were also made on samples of Astroquartz cloth taken from the Hastelloy-X vacuum enclosure-materials compatibility test at the conclusion of 2000 hours of testing at a temperature level of 1400°F (Section 3.4.3.5). Tensile strength was determined to be 10.5 lb./in. with the material displaying good handling characteristics.

Refrasil Quartz Fiber Paper

Tensile strength measurements were made on samples of Refrasil paper taken from the Hastelloy-X vacuum enclosure-materials compatibility task at the conclusion of 2000 hours of testing at 1400°F (Section 3.4.3.5). Tensile strength was found to be .029 lbs./in. As received tensile strength of Refrasil paper was 0.28 lb/in. and after short term exposure at 1800°F tensile strength was 0.063 lbs/inch (Table XXIX).

Tensile strength measurements were also made on samples of Refrasil paper taken from the copper foil-Refrasil quartz fiber paper 2000 hour thermal conductivity test, Section 3.3.1.5.3. This combination was also subjected to approximately 1000 hours of air exposure testing, Section 3.3.3.6. Refrasil paper samples were taken from three different temperature zones of the insulation systems:

- 1). Cold face zone - fourth spacer layer counting from the outside or cold face boundary.
- 2). Intermediate zone - twentieth spacer layer counting from the outside or cold face boundary.

TABLE XXXI

ASTROQUARTZ CLOTH TENSILE TEST

<u>Insulation Spacer Location</u>	<u>Approximate Exposure</u>		<u>Breaking Strength, lb./in. Warp Direction</u>
	<u>Time, hours</u>	<u>Temperature, °F</u>	
<u>Cold Face Insulation Zone</u>			17.5
3rd insulation layer from Cold face	500	385	
	600	320-535	
	1200	660	
<u>Middle Insulation Zone</u>			3.75
21st insulation layer from Cold face	500	1600	
	600	850-1500	
	1200	1350-1420	

- 3). Hot Zone - forty-sixth layer counting from the cold face boundary.

The results of the tensile strength measurements together with approximate exposure times and temperatures are presented in Table XXXI. Tensile strength appeared to be relatively insensitive to temperature exposure and did not show a decrease at higher temperature as did Astroquartz Cloth.

Copper Flake Opacified Refrasil Quartz Fiber Paper

Tensile strength measurements were made on samples of copper opacified Refrasil paper taken from the 2000 hour copper foil - copper flake opacified Refrasil quartz fiber paper thermal conductivity test, Section 3.3.1.5.4. This combination was also subjected to approximately 400 hours of argon inert gas exposure thermal testing, Section 3.3.1.5.4.1. Samples were taken from three different temperature zones:

- 1). Cold face zone - third spacer layer counting from the outside or cold face boundary.
- 2). Intermediate zone - twenty fourth spacer layer counting from the outside or cold face boundary.
- 3). Hot face zone - forty-sixth spacer layer counting from the outside or cold face boundary.

The results of the tests together with approximate exposure temperatures and times are presented in Table XXXIII. Tensile strength appears to decrease slightly after long term exposure to temperatures in excess of approximately 400-500°F and then shows little change with increased temperature.

3.5.1.2 Shrinkage and Weight Loss Tests

Shrinkage and weight loss experiments were conducted in triplicate on the candidate spacers.

The results of these tests are presented in Table XXVIII. It can be seen that no change in area was observed for Astrosil, Refrasil and Irish Refrasil. A rather large shrinkage was observed for Refrasil quartz fiber paper, and this can be attributed to the large amounts of binder used for the production of this paper. Astroquartz, however, underwent an area increase, and this is probably due to a physical change, such as a phase change, in the structure of the fibers, see Section 3.2, Spacer Selection.

A weight gain was noted for copper flake opacified Refrasil quartz fiber paper processed by the air oxidation method due to oxidation of the copper flakes. A weight loss, however, was noted when the Linde Insulation Cleaning Process was used, indicating removal of the binder with no oxidation of the copper flake, see Section 3.4.1., processing.

TABLE XXXII

REFRASIL QUARTZ FIBER PAPER TENSILE TEST

<u>Insulation Spacer Location</u>	<u>Approximate Exposure</u>		<u>Breaking Strength lb./inch, Warp Direction</u>
	<u>Time, hours</u>	<u>Temperature, F°</u>	
Cold face zone	2000	258	0.022
	150	200	
	850	350	
Intermediate zone	2000	1000	0.021
	150	300	
	850	1150	
Hot face zone	2000	1245	0.021
	150	350	
	850	1290

TABLE XXXIII

COPPER FLAKE OPACIFIED REFRASIL QUARTZ FIBER PAPER TENSILE TEST

<u>Insulation Spacer Location</u>	<u>Time, hours</u>	<u>Temperature, F°</u>	<u>Breaking Strength lbs/inch Warp Direction</u>
Cold face zone	2000	347	0.073
	100	140	
	100	257	
	100	367	
	100	495	
Intermediate zone	2000	1100	0.053
	100	175	
	100	400	
	100	650	
	100	900	
Hot face zone	2000	1265	0.055
	100	200	
	100	500	
	100	770	
	100	1094	

TABLE XXXIV

SHRINKAGE AND WEIGHT LOSS AFTER AIR OXIDATION

<u>Material</u>	<u>Areal Shrinkage, %</u>	<u>Weight Loss, %</u>	<u>Processing Tempera- ture, °F</u>
Astroquartz No. 503	-9.4	2.1	1800
Astrosil No. 82	0	4.5	1800
Refrasil No. C100-28	0	3.8	1800
Irish Refrasil No. C1554-28	0	5.4	1800
Refrasil Quartz Fiber Paper	7.3	11.4	1800
Glass Fiber Paper	0.4	0	800
Aluminum Flake Opacified Glass Fiber Paper	2.6	2.4	800
Copper Flake Opacified Refrasil Quartz Fiber Paper	1.3	-4.8 (gain) 6.0*	1280

*Processed by Linde Insulation Cleaning Process

No weight loss and an extremely small areal shrinkage were noted in the case of the glass fiber paper since no binder is used in the manufacture of this spacer. This is substantiated by the apparent constant residual carbon content before and after processing, Table XXI, Section 3.4.1.

3.5.1.3 Material Compatibility

Material compatibility observations and chemical analyses were made on samples of the candidate insulation materials taken from the thermal conductivity and vacuum enclosure tasks. The thermal conductivity tests provided not only exposure to vacuum and temperature but also to various temperature levels due to the thermal gradient existing in the insulation from the hot to cold face boundary. The effect of this gradient on tensile strength was seen in Section 3.5.1.1. During the vacuum enclosure task samples of insulation materials were exposed to candidate enclosure materials, Inconel 600 (Section 3.4.3.4) and Hastelloy X (Section 3.4.3.5) as well as long term vacuum and temperatures.

3.5.1.3.1 Insulation Materials

Glass Fiber Paper

Visual examination of this material after the aluminum foil-glass paper combination thermal conductivity test, Section 3.3.1, showed that about the first twelve layers from the hot boundary were slightly yellow brown in color but still soft and handleable. An X-ray diffraction analysis of the discolored layers was made to investigate possible metallic contamination but no crystalline material was found. No change in thermal performance of the insulation combination was observed over the test period, approximately 700 hours. Temperature exposures were: cold side; 109°F to 162°F, hot side; 482°F to 878°F.

Inspection of glass fiber paper samples at the conclusion of the 300 hour Inconel-600 Vacuum enclosure specimen test and at the 2000 hour point of the Hastelloy-X vacuum enclosure specimen test disclosed no evidence of glass fiber paper incompatibility. The samples exposed to temperatures of 800-900°F during these tests were soft and handleable. Laboratory analysis conducted after testing disclosed that no metallic compounds were present.

Astroquartz Cloth

After the short term, approximately 600 hours, nickel foil-Astroquartz cloth thermal conductivity test, Section 3.3.1.5.5, an examination of the material showed loss of strength in the hot face zones but no evidence of incompatibility. Temperature exposures ranged from 130°F to 550°F on the cold boundary and from 503°F to 1947°F on the hot

boundary. No changes in the thermal performance of the insulation system were noted over the test period.

Similar examination of the tantalum-Astroquartz thermal conductivity test disclosed that the Astroquartz had become very brittle in the hot zone and was compacted with the tantalum foil for about the first seven layers from the hot boundary. The rest of the layers displayed reduction of strength but were handleable. Temperature exposure ranged from 136°F to 662°F on the cold side and 617°F to 1800°F on the hot side during thermal conductivity testing.

Inspection of the Astroquartz cloth samples at the conclusion of the 300 hour Inconel 600 vacuum enclosure specimen and 2000 hour point of the Hastelloy-X specimen disclosed no evidence of incompatibility. The samples were exposed to temperatures of 1800°F during the Inconel-600 testing and 1400°F during the Hastelloy-X testing. A laboratory analysis of the Astroquartz spacer from the Hastelloy-X vacuum enclosure test and tantalum-Astroquartz thermal conductivity test was conducted and no metallic compounds were found.

Refrasil Quartz Fiber Paper

Examination of this material from the 2000 hour copper foil-Refrasil paper thermal conductivity test disclosed no evidence of incompatibility. Laboratory analysis was conducted on this spacer and no metallic compounds were detected. Temperature exposure during testing ranged from 158°F to 258°F on the cold face to 720°F to 1260°F on the hot face.

Copper Flake Opacified Refrasil Quartz Fiber Paper

Examination of this material from the copper foil-copper opacified Refrasil paper thermal conductivity test disclosed no evidence of incompatibility. A laboratory analysis was conducted and no metallic compounds were found.* Temperature exposures during the 2,000 hour test were 185°F to 347°F on the cold side and 635°F to 1265°F on the hot side.

Aluminum Foil

Visual examination of this foil after the aluminum foil-glass fiber paper thermal conductivity test, Section 3.3.1, disclosed that the layers were bright and shiny throughout the insulation wrap.

Examination of aluminum foil after the 300 hour Inconel-600 and 2000 hour Hastelloy-X vacuum enclosure tests showed no visual evidence of incompatibility as the foils retained a shiny appearance.

* With the exception of elemental copper which was used as the opacifier.

Copper Foil

Examination of the copper foils from the copper foil-Refrasil quartz fiber paper insulation combination that was subjected to air exposure at the conclusion of its 2000 hour performance test disclosed variations in foil color that were attributed to oxidation. The foils were a yellow-orange color at the cold boundary and changed progressively to a dull pink at the hot boundary. A visual loss of brightness was observed from the orange yellow to dull pink.

Examination of copper foil at the conclusion of the 2000 hour copper foil-copper opacified Refrasil paper thermal conductivity tests showed that the foils were bright and shiny with no visual evidence of incompatibility.

Copper foil samples were also bright and shiny after the 300 hour Inconel-600 and 2000 hour Hastelloy-X vacuum enclosure specimen tests.

Nickel Foil

Inspection was made of nickel foils from the 600 hour nickel foil-Astroquartz cloth thermal conductivity test, the 300 hour Inconel-600 vacuum enclosure test and the 2000 hour Hastelloy-X vacuum enclosure test. The foils were bright and shiny with no evidence of incompatibility. The nickel foils of the 2000 hour nickel-Refrasil paper thermal conductivity test will be examined at the conclusion of that test.

Tantalum Foil

Short term, approximately 120 hour, exposure of tantalum foil to vacuum and elevated temperature disclosed no undesirable effects (Section 3.4.1) and examination of tantalum foil samples at the conclusion of the 300 hour, 1800°F Inconel-600 vacuum enclosure test disclosed no evidence of incompatibility. The foils were slightly more brittle to the touch than in the "as received" conditions but still bright and shiny.

However, after the 2000 hour tantalum foil-Astroquartz cloth thermal conductivity test, examination of the insulated tester after removal from the vacuum enclosure showed that the outer, cold face tantalum layer had a blue-gray appearance. From the second foil to about the 23rd layer, (cold zone - counting from the outside or cold face) the surface of the foils was quite bright and the foils themselves were flexible. Temperature exposure of the insulation at this point, 23rd layer, ranged from approximately 1650°F to 1375°F during the testing. From the 23rd to 35th layers (intermediate zone - counting from the outer or cold side), the tantalum foil and Astroquartz cloth were very brittle and the 25th to 42nd layers, hot zone, were extremely wrinkled and compacted together. Samples of the tantalum foil from the various temperature zones of the insulation wrap were submitted for X-ray diffraction analysis.

The blue-gray outer layer was analyzed and found to contain a small fraction of tantalum metal, <10%, with the balance being unidentifiable.

Since not every compound of tantalum has been identified by the ASTM for standards, some of the compounds formed are reported as unidentifiable. It is theorized that the unidentifiable compounds could be a partial oxidation product, a different valance state or a mixed molecule such as $TaOCl$, $TaNCl$. The mixed molecules could have resulted from reaction of the tantalum with trace impurities present in the vacuum space.

A foil sample taken from the middle zone, 21st layer counting from the cold face boundary, was analyzed to be mostly tantalum silicide, Ta_2Si , with a small quantity of tantalum metal and an unidentifiable crystalline material, <10%. This indicates that the tantalum foil had reacted with silicon present in the quartz cloth spacer during the thermal performance testing.

Foil samples taken from the hot zone, 25th-42nd layers counting from the cold face boundary indicated that iron tantalum, Fe_7Ta_3 , was the major component with small quantities of tantalum silicide, Ta_2Si , and tantalum pentoxide, Ta_2O_5 . An emission spectrum disclosed that trace quantities of iron, aluminum, chromium and nickel were also present.

These analyses indicate that tantalum foil reacted with silicon from the Astroquartz spacer and contaminants such as iron, aluminum, nickel and chromium originating from the thermal conductivity tester itself. The thermal conductivity tester was fabricated from Type 310 stainless steel and is in communication with the insulation within the vacuum space. The tantalum oxide was probably formed due to air leakage into the tester, during testing, Section 3.3.1.5.7. The foil brittleness, darkening of color and deterioration of thermal performance over the 2000 hour test period, Section 3.3.1.5.7., is thus attributed to the formation of the previously described tantalum compounds which resulted in an increased emissivity of the foil. Because tantalum foil reacted with the Astroquartz spacer and trace contaminants present in the vacuum space, with a subsequent degradation of thermal performance, it is not recommended as a candidate radiation shield material.

3.5.1.3.2 Vacuum Enclosure-Insulation Materials Compatibility

Inconel-600 Specimen

This enclosure specimen was subjected to 300 hours of testing at 1800°F in a vacuum environment containing samples of the following insulation materials, each placed within the vacuum chamber so that they were at temperature levels within their operational limits: aluminum foil, copper foil, nickel foil, tantalum foil, glass fiber paper, Refrasil quartz fiber paper and Astroquartz cloth. No evidence of incompatibility of the insulation materials or the Inconel-600 specimen

were observed.

Hastelloy-X Specimen

Samples of each of the following insulation materials were included in the vacuum chamber tube: aluminum foil, copper foil, nickel foil, glass fiber paper, Refrasil quartz fiber paper, and Astroquartz cloth. The materials were placed in the vacuum chamber tube in such a manner that they were at temperature levels within their operational limits.

In order to investigate compatibility of the Hastelloy-X specimen with typical radioisotope power system enclosure materials, test coupons of Hastelloy-X were placed in direct contact with coupons of beryllium and Hastelloy-C (power system enclosure materials) on the air side of the test enclosure specimen.

After 2000 hours of testing in vacuum at 1400°F the specimen and insulation samples were examined. No evidence of incompatibility of the insulation materials or Hastelloy-X specimen was observed.

3.5.2 Preliminary Insulation Dynamic Tests

The objective of this work was to ascertain the apparent resistance of the processed candidate insulation materials to vibration levels typical of service requirements. Testing involved placing multiple layer insulation combinations within a rigid, square, holding fixture, mounting the fixture on an electrodynamic shaker table and subjecting the apparatus to acceleration loadings at ambient temperature. No visual damage to any of the combinations tested was observed.

Each of the following insulation combinations has been subjected to the acceleration levels of Figure 94, (based on Curve C, Figure 514-3, MIL-STD-810A, June 23, 1966):

- 1). Aluminum foil - glass fiber paper
- 2). Aluminum foil - aluminum flake opacified glass fiber paper
- 3). Copper foil - Refrasil quartz fiber paper
- 4). Nickel foil - Astroquartz cloth
- 5). Copper flake opacified Refrasil quartz fiber paper

VIBRATION PROFILE FOR PRELIMINARY DYNAMIC TEST TASK

FIGURE 94

Table XXXV summarizes physical properties of the insulation evaluated.

Before vibrational testing the insulation combinations were processed as described in Section 3.4.1 with the exception of the aluminum foil. Based on previous experience with both processed and as-received aluminum foil, it was felt that processing would not alter the foil resistance to vibrational loads; and, in order to expedite testing, unprocessed aluminum foil was used. Sample sizes were 5 in. x 5 in. x 3/8 in. in height.

Previous insulation application experience has shown that the minimum pressure at which Linde Super Insulations can be applied is approximately 0.05 psi. At this condition the insulation layers have maximum freedom of movement and hence maximum opportunity for vibrational damage. Thus, it was decided to dynamically test all the insulation samples at a layer density equivalent to a 0.05 psi loading. In addition, two of the insulation combinations, copper foil - Refrasil quartz fiber paper and nickel foil-Astroquartz cloth, were tested at a layer density equivalent to 0.1 psi loading in order to determine vibrational resistance at an increased load. Curves for compressive load vs. insulation layer density of each insulation combination were plotted based on data obtained with the load density tester in the range between 0.01 and 0.1 psi. The resulting curves to obtain the layer density vs. load data and the load density tester used are shown in Appendix G. The number of layers included in the test fixture compartments for each insulation to be tested was based on the layer density curves shown in Appendix G for the desired test loading. Table XXXV presents a summary of the insulations tested and the layer densities at which they were tested.

Test Apparatus and Test Procedure

The vibrational test fixture used included three compartments, each to contain 3/8-inch thickness of insulation.

Three samples of each composite being studied were tested simultaneously, one perpendicular, one parallel and one at a 45 degree angle to the acceleration load.

Figure 95 is a schematic of the vibrational test fixture while Figure 96 shows the vibration test fixture with a sample insulation combination placed in the 45 degree angle compartment.

Vibration testing was accomplished on a 1200-pound Sinusoidal machine manufactured by Uholtz-Dicke and shown in Figure 97. It was anticipated that the acceleration loading profile of Figure 75 could be obtained by automatic programming of the vibrational machine throughout the desired frequency range. However, the vibrational fixture displayed three resonance frequencies at approximately 600, 1200 and 1800 cycles per second. During a programmed run the resulting acceleration loading became so high at these frequencies that the automatic overload

TABLE XXXV
INSULATION COMPONENT DATA

<u>Foils</u>	<u>Thick- ness</u>	<u>Type</u>	<u>Method of Production</u>	<u>Vendor</u>
Aluminum	.00025"	AA-1145-0	Rolled	Alcoa
Copper	.0005"	---	Electro Deposited	Circuit Foil
Nickel	.00062"	200	Rolled	Somers Brass
<u>Spacers</u>	<u>Thick- ness</u>	<u>Type</u>	<u>Weight</u>	<u>Vendor</u>
Glass Fiber Paper	.003"	223	1.7 gm/ft ²	C.H.Dexter
Refrasil Quartz Fiber Paper	.005"	X-666	2.37 gm/ft ²	C.H.Dexter
Astroquartz Cloth	.005"	503	10.4 gm/ft ²	J.P.Stevens
<u>Opacified Paper</u>	<u>Thick- ness</u>	<u>Type</u>	<u>Weight</u>	<u>Vendor</u>
Aluminum Flake Opaci- fied Glass Fiber Paper	.003"	---	1.1 gm/ft ²	C.H.Dexter
Copper Flake Opacified Refrasil Quartz Fiber Paper	.010"	---	5.5 gm/ft ²	C.H.Dexter

TABLE XXXVI
INSULATION COMBINATIONS TESTED

	Test Layer Density (Based on Load Density Tester Loading)	
	<u>0.05 psi</u>	<u>0.1 psi</u>
Aluminum Foil-Glass Fiber Paper	122 layers/in.	-----
Aluminum Foil - Aluminum Flake Opacified Glass Fiber Paper	158 layers/in.	-----
Copper Foil - Refrasil Quartz Fiber Paper	83 layers/in.	89 layers/in.
Nickel Foil - Astroquartz Cloth	115 layers/in.	123 layers/in.
Copper Flake Opacified Refrasil Quartz Fiber Paper	62 layers/in.	-----

INSULATION
CLAMPING
DEVICE
(CUT-AWAY)

INSULATION
SAMPLE

VIBRATOR
SUPPORT

CLAMPING SURFACES

INSULATION VIBRATOR FIXTURE

Figure 96. Vibration Test Fixture with Insulation Composite

Figure 97. Vibrational Test Apparatus

protection device shut the tester down. Thus in order to accomplish the test it was necessary to manually control the acceleration input at these frequencies. An accelerometer was bolted rigidly to the fixture and the accelerometer readings observed on an oscilloscope so that machine input could be controlled where necessary to avoid overloads at the resonance frequencies. In all cases, however, insulation combinations were subjected to loadings equivalent to or greater than those of Figure 75.

Test Results

No visual damage to any of the components listed was observed.

In Appendix G photographs show samples of each of the insulations before and after vibrational testing. Because there were no detrimental effects to any of the insulations at any of the three angles of loading, only one insulation sample for each material is shown in each of the before and after photographs.

4.0 CONCLUSIONS AND RECOMMENDATIONS

4.1 CONCLUSIONS

The objectives of this insulation study program have been substantially met. Specific results of this program are:

4.1.1 The very low thermal conductivity of the vacuum foil type of insulations has been demonstrated. Aluminum, copper and nickel systems have demonstrated stability up to 900°, 1400° and 1600°F., respectively, for periods up to 2000 hours.

4.1.2 All insulation materials except the tantalum and silica combination were shown to be compatible. Because of the tantalum-silica incompatibility, tantalum is not recommended for thermal radiation shields in these systems.

4.1.3 The effect of air exposure (as would occur on ground fueled space systems) for typical foil materials on their subsequent thermal performance in vacuum is analytically shown to be minor for moderate exposure times and temperatures. For copper, the most sensitive to oxidation, in a subsequent vacuum environment on a deep space mission (above 150 miles) the oxide formed during ground hold should decompose, possibly restoring the original performance. A correlation has been developed which can be used to predict the thermal conductivity of insulation systems in vacuum after air exposure. Some experimental evaluations of the correlations has been performed.

4.1.4 By suitable processing, the insulation materials can be made sufficiently stable for operation in vacuum at high temperature.

4.1.5 The outgassing rates of suitable insulation materials have been measured and their gettering requirements defined. All materials required for use in sealed systems at operating temperatures up to 1700°F. have acceptably low outgassing characteristics.

4.1.6 Physical strength determinations were made for all insulation materials before and after processing. Based on these physical strength determinations, dynamic tests, and actual application to test apparatus, it is believed the materials will be fabricable to most thermoelectric converter requirements.

4.1.7 The fabrication of vacuum enclosures for 1400°F. operation in air for long-term life has been demonstrated. A Hastelloy-X specimen was tested in this environment for 10,000 hours with no leakage.

4.1.8. The effect of gaps between panels of vacuum-foil insulation was shown analytically to be of controllable effect so that greater adaptability in the application of this type of insulation system can be realized.

4.2 RECOMMENDATIONS

In the course of the development work of this contract further investigation of various areas became apparent. In order to more completely develop multilayer insulations for engineering application to radioisotope power systems, the following recommendations are offered:

4.2.1

Thermal conductivity values for the individual insulation combinations have thus far been obtained at one value of layer density. In order to thermally optimize the multilayer insulation system, the optimum foil spacing or layers per inch density for each temperature range must be selected. It is recommended that thermal conductivity, as a function of layer density, be investigated for the candidate insulations. With this kind of information it will be possible to better define the relationship between radiation and solid conduction in these systems as affected by layer density. Because of the number of experiments involved in this type of study, a flat plate type thermal conductivity tester is recommended.

4.2.2

Since the opacified paper spacers contain metallic flakes of extremely small dimensions, it is recommended that the long-term stability of these opacified papers, without metallic radiation shield foils, be investigated by long-term, 2000 to 10,000 hour, stability tests.

4.2.3

It is recommended that the longitudinal thermal conductivity of opacified paper spacers be experimentally determined so that their effect on insulation system thermal performance can be evaluated. This is uniquely significant because of the promising application of these materials in areas of discontinuity of the multi foil insulations such as joints between segments and around penetrations.

4.2.4

Experiments should be conducted to evaluate the air exposure characteristics of insulations containing aluminum and nickel foils. These experiments will supplement the experiment performed on copper in Phase I and is necessary to verify the analytical prediction of the behavior of these two additional materials.

4.2.5

In order to verify the thermal performance of a composite insulation system, an experimental thermal conductivity determination of such a system should be made. To demonstrate the suitability of this

composite insulation system for long-term usage, a 10,000 hour thermal stability test should be performed.

4.2.6

An analytical study of the thermal effects of joints between insulation panels and exposed edges of insulation panels has been made. It is recommended that this work be experimentally verified and methods developed to minimize the effect of these joints.

4.2.7

The analytical joints study in Item 4.2.6 assumed that joint depths equal the entire insulation thickness. For applications requiring greater total insulation thickness, it may be thermally advantageous to stagger joint locations. It is recommended that this concept be analytically and experimentally studied.

4.2.8

To reduce heat losses associated with penetration of the insulation by components such as thermoelectric elements or structural supports, an analytical and experimental investigation should be conducted to determine the best way to insulate around these components.

4.2.9

To assure that multilayer insulations are adaptable for application to radioisotope power sources, improved procedures must be developed for applying the insulation to geometric shapes typical of actual isotope power generators. Primary emphasis should be placed on studying the application of these insulations to double curvature surfaces. The present procedure of installing one layer at a time is exceedingly slow and expensive and typically results in a non-uniform insulation thickness.

4.2.10

Methods for attachment of multilayer insulation panels should be investigated to develop techniques for holding the insulation in place with acceptable and predictable thermal performance during exposure to typical dynamic loads. This is especially needed for those applications (typically aero-space) where physically restraining boundaries to contain the insulation are not provided.

4.2.11

A preliminary visual evaluation of the physical resistance of processed insulation materials to dynamic environments has already been carried out. It is recommended that this be expanded to include evaluation of the thermal performance of processed insulations before and after exposure to dynamic loads.

4.2.12

To compliment the development of ultra-low conductivity insulation materials for use on radioisotope powered systems it is recommended that a study be initiated to survey, develop and evaluate structural materials and structural design configurations for application in the same range of temperatures and other environment requirements as specified for the insulations. Development of thermally-structurally efficient supports for integration with the vacuum-foil insulations will be necessary to provide for the most effective use of the vacuum-foil insulation and to minimize the total unproductive heat loss from the system.

4.2.13

Because of the increasing emphasis upon the non-sealed, space applications for vacuum-foil insulations it will be necessary to determine the time requirements to evacuate these insulations so that the time to achieve vacuum-level, thermal performance will be known. Since the time required is governed by offgassing as well as flow conductance considerations, direct measurement for various flow distances should be performed. This will enable accurate calculations to be made for all practical configurations.

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6.0 APPENDICES

APPENDIX A

THERMAL CONDUCTIVITY TESTS IN VACUUM

TEST APPARATUS AND INSTRUMENTATION

The insulation tester consisted of a hollow stainless steel cylinder, Type 310, 3 inches in diameter by 18 inches long, equipped with suitable mounts for installing internally the resistance heater elements. The outside diameter of the tester was the insulation hot face. The heating element consisted of a resistance wire wound on a ceramic tube. The heater leads were electrically insulated as they pass through the insulation.

The vacuum test chamber consisted of a stainless steel container, 6.5 in. I. D. x 24 in. high, with an O-ring flange access port for installing the insulated test core. Evacuation port and vacuum feed-throughs were provided in the flange head.

Temperature measurement was achieved by three Chromel-Alumel thermocouples fabricated from 28 gauge wire. Two thermocouples were mounted at the hot surface (outer surface of the 3-inch diameter test core) and one on the cold surface (outer layer of insulation). Thermocouple wires were electrically insulated where they pass through the insulation.

Thermocouple output was monitored on a multi-point temperature recorder, ambient temperature compensated.

A voltmeter and ammeter monitored voltage drop across the core heater and current input to the heater, respectively. These measurements were used to evaluate power consumption. The power input to the core heater was regulated by a variac. A temperature controller, was used to control power input while thermal equilibrium was being established.

A Phillips cold cathode gauge was used to monitor dynamic vacuum pressure.

Heater Design and Fabrication

The following requirements were established in a heater that must be capable of operating in high vacuum, 10^{-4} to 10^{-5} torr, for periods of 2000 hours at temperatures up to 1800°F.

- 1). Low vapor pressure at 1800°F to minimize vaporation rate when subjected to hard vacuum for 2000 hours.
- 2). Relatively high electrical resistance.
- 3). Low conductivity to minimize extraneous heat leak out of the insulation system.
- 4). Ductile enough to permit installation and normal handling without breaking.

An investigation of various types of heaters for use with the thermal conductivity tester showed that two types of heaters appeared suitable for this application. One heater employed a coiled heating wire element while the other used a graphite tape heating element. Fabrication difficulties were encountered with the graphite tape heater and since the aim of this program was to thermally evaluate insulations rather than to develop and evaluate heater designs further work on the graphite heater was terminated.

For the temperature range of 100°F. to 900°F., when testing aluminum foil-glass fiber paper and aluminum foil-aluminum opacified glass paper combinations, Trophet A, a resistance wire composed of 80% nickel - 20% chromium alloy was selected. The extremely low thermal conductivity of this alloy, 6.4 BTU/hr.ft.°R as compared with 97.5 BTU/hr.ft.°R. for tungsten, makes it extremely attractive as extraneous heater wire heat leak to the insulated tester is considerably lessened. Discussions with vendors indicated that this heater element will operate successfully in vacuum in the temperature range of 1000°F. for at least 2000 hours.

Among several metals that were considered for the 900°F. to 1800°F. temperature range, tungsten was found to be the most suitable. Nickel and chromium base alloys were not strong candidates because of their relatively high vaporization rates in vacuum at elevated temperatures. Vendor contact confirmed this. Based on the low vaporization rate of tungsten in vacuum at elevated temperature and its property to remain ductile at 2000°F., this wire was recommended. Platinum-rhodium alloy was also selected for use as a heater element based on its low vapor pressure - high melting point properties. However, a platinum-rhodium heater installed on a tantalum-Astroquartz cloth combination failed during testing due to the formation of brittle bimetallic alloys from contaminants originating from the Type 310 stainless steel insulation tester. Details of the failure are given in Section 3.3.1.5.7.

Data Reduction

The data reduction procedure for determination of insulation thermal conductivity involved the solution of the basic Fourier heat conduction equation.

Writing an energy balance equation for the insulation test system:

$$Q_{\text{insulation}} = Q_{\text{total}} - Q_{\text{losses}} \quad (\text{A-1})$$

where: Q_{total} = Power input to the heater

$Q_{\text{insulation}}$ = Heat transferred from the heater through the insulation

Q_{losses} = Heat transferred from the heater through heater leads, thermocouple leads, and their insulation plus power losses through the heater leads

$Q_{\text{insulation}}$ may be expressed in the form of:

$$Q_{\text{insulation}} = S k_a (T_1 - T_2), \text{ BTU/hr.} \quad (\text{A-2})$$

where: S = Shape factor, area/thickness, ft.
 k_a = Apparent thermal conductivity, Btu/hr-ft-°R
 T_1 = Hot face temperature, °F
 T_2 = Cold face temperature, °F

The shape factor, S , is obtained from geometrical measurements and is the sum of the cylindrical portion shape factor and end sections shape factor.

$$S = S_{\text{cylinder}} + S_{\text{ends}}, \text{ ft.} \quad (\text{A-3})$$

$$S_{\text{cylindrical}} = \frac{2\pi L}{\ln \frac{d_2}{d_1}} \quad (\text{A-4})$$

where: L = Length of test core + 1/2 thickness of the installed insulation at each end of the core

d_2 = Outside diameter of the insulation as installed on the tester core

d_1 = Outside diameter of the test core which equals inside diameter of the insulation

S_{ends} of the tester is calculated from:

$$S_{\text{ends}} \approx \frac{2\pi r_1 r_2}{t} \quad (\text{A-5})$$

where: r_1 = Inside radius of the insulation cylinder

r_2 = Outside radius of the insulation cylinder

t = Thickness of the end insulation

Total power delivered to the tester may be expressed as:

$$Q_{\text{total}} = VI(3.414), \text{ Btu/hr.} \quad (\text{A-6})$$

where: V = Voltage across the heater leads, volts

I = Current flowing through the heater leads, amps

Q_{losses} may be expressed as a summation of individual components:

$$Q_{\text{losses}} = Q_{\text{heater leads}} + Q_{\text{heater lead insulation}} + Q_{\text{thermocouples}} + Q_{\text{thermocouple insulation}} + Q_{\text{power losses}} \quad (\text{A-7})$$

Heater leads, thermocouple leads and their insulation losses may be estimated by assuming the entire temperature drop occurs through the thickness of the insulation; that is

$$Q_i = \frac{k_i A_i (T_1 - T_2)}{t} \quad (\text{A-8})$$

where: Q_i = Lead loss for heater leads, thermocouples or their insulation

k_i = Mean thermal conductivity of each component between T_1 and T_2

t = Thickness of lead

This approximation was later checked using a penetration computer program developed in Phase II and found to be accurate within 50%. Since the testers were generally designed such that the losses were a small part of the total, this approximation is not thought to influence the k by more than 5%.

Power losses due to resistance of the heater wires outside of the heater core may be calculated by:

$$Q_{\text{power}} = I^2 R \quad (\text{3.414}) \quad (\text{A-9})$$

where: Q_{power} = Power losses, BTU/hr.

I = Current, amps

R = Wire resistance, corrected for temperature, ohms

Substituting equations A-9 and A-13 in A-8, we obtain:

$$Sk_a (T_1 - T_2) = VI (3.414) - Q_{\text{losses}} \quad (\text{A-10})$$

The apparent thermal conductivity of the insulation may then be calculated from:

$$k_a = \frac{VI (3.414) - Q_{\text{losses}}}{S (T_1 - T_2)} \quad (\text{A-11})$$

APPENDIX B

THERMAL EFFECTS OF INSULATION PANEL JOINTS AND EXPOSED EDGES

COMPUTER PROGRAM DEVELOPMENT - THERMAL EFFECTS OF JOINTS AND EDGES ON INSULATION PANELS

In this analysis it was assumed that the unfilled gaps are bounded by reflective foils, of the same material as the foils within the insulation panel. These foils act as blackbody radiators when viewed from the panel edge. However, when both a warm-side and cold-side gap foil are present (rectangular gap), each foil appears gray to the other.

The temperature T inside the panel is found by solving the steady state Fourier heat conduction equation,

$$\frac{\partial}{\partial X} (k_X \frac{\partial T}{\partial X}) + \frac{\partial}{\partial Y} (k_Y \frac{\partial T}{\partial Y}) = 0 \quad (B-1)$$

subject to the following boundary conditions:

- (a) $T(X, 0) = T_W$ (prescribed warm-side temperature)
- (b) $T(X, h) = T_C$ (prescribed cold-side temperature)
- (c) $\frac{\partial T}{\partial X} (\frac{W}{2}, Y) = 0$ (adiabatic plane through midsection of panel)
- (d) $k_X \frac{\partial T}{\partial X} (0, Y) = Q_L$ (heat-loss mechanism at panel edge)

If the joint contains a solid filler, the temperature within the gap will also be determined by (B-1). However, now the thermal conductivities k_X and k_Y will be different. Also, if the gap is angular, the first term on the left hand side of (B-1) becomes:

$$\frac{1}{Y^2} \frac{\partial}{\partial \theta} (k_X \frac{\partial T}{\partial \theta})$$

where θ is the angular distance from the edge of the panel, and Y becomes the radial distance from the warm-face contact line of the two panels. The boundary conditions for a rectangular gap are

- (a') $T(-X, 0) = T_W$ (same as (a))
- (b') $T(-X, h) = T_C$ (same as (b))
- (c') $\frac{\partial T}{\partial X} (-\frac{d}{2}, Y) = 0$ (adiabatic plane through midsection of gap)
- (d') $k_X \frac{\partial T}{\partial X} (0, Y) = Q_L$ (heat gained or lost at edge of panel equals heat lost or gained at edge of gap)

and for an angular gap are:

$$\begin{aligned}
 (a'') \quad \frac{\partial T}{\partial Y} (\theta, 0) &= 0 && \text{(no radial heat conduction at gap apex)} \\
 (b'') \quad T (\theta, h) &= T_C && \text{(similar to (b) and (b'))} \\
 (c'') \quad \frac{\partial T}{\partial \theta} \left(\frac{\Phi}{2}, Y \right) &= 0 && \text{(similar to (c) and (c'))} \\
 (d'') \quad (k_X/Y) \frac{\partial T}{\partial \theta} (0, Y) &= Q_L && \text{(similar to (d'))}
 \end{aligned}$$

Although the edge heat loss Q_L is conductive for a filled gap,* it is radiative for an empty gap. If the panel is divided into $(N + 1)$ increments in the Y direction, the edge heat loss at the j th level is given by

$$\begin{aligned}
 (Q_L)_j &= (h_W)_j (T_j - T_W) + (h_C)_j (T_j - T_C) \\
 &+ \sum_{i=1}^N (Q_e)_{i,j}
 \end{aligned} \tag{B-2}$$

$(h_W)_j$, the heat transfer coefficient for radiation between the j th edge increment and the warm face of the gap, is given by

$$(h_W)_j = \sigma \cdot dY \cdot (f_W)_j \cdot (T_j^2 + T_W^2) \cdot (T_j + T_W) \tag{B-3}$$

$(h_C)_j$, the edge-increment-to-gap-cold-face radiative heat transfer coefficient, is given by

$$(h_C)_j = \sigma \cdot dY \cdot (f_C)_j \cdot (T_j^2 + T_C^2) \cdot (T_j + T_W) \tag{B-4}$$

The radiative heat transfer between the j th panel increment and the i th opposite-panel increment (on other side of the gap) is given by

$$(Q_e)_{i,j} = \sigma \cdot dY \cdot (f_e)_{i,j} \cdot (T_j^4 - T_i^4) \tag{B-5}$$

In the above equations,

$$dY = h / (N + 1)$$

$$\sigma = \text{Stefan-Boltzmann Constant} = 1.73 \times 10^{-9} \text{ BTU-hr}^{-1}\text{-ft}^{-2}\text{-}^\circ\text{R}^{-4}$$

$$(f_W)_j = j \text{ th edge increment-gap warm face view factor}$$

* Semitransparent - changed in Phase II.

$(f_c)_j$ = j th edge increment-gap cold face view factor

$(f_e)_{i,j}$ = j th edge increment-i th opposite edge increment view factor

T_j = edge temperature at boundary between j th and (j + 1) th increment

Since $(f_w)_j = 0$ for an angular gap, there is no edge-to-gap-warm-face heat exchange in this case. As indicated in the above equations, we have assumed all surfaces to be black.

Because of the strong nonlinearity imposed by the radiative boundary conditions and temperature dependence of the thermal conductivities, analytical solution of (B-1) is impractical. Instead, we set up a finite difference mesh within the panel and, for filled gap, within the gap (Equation B-6). Finite difference approximations to (B-1) and the appropriate boundary conditions are then made at all mesh points, resulting in $M \times N$ linear equations in like number of unknown temperatures (M and N are the numbers of mesh points in the X (and Y directions, respectively). The equation at the mesh point located at the i th X or θ position and j th Y position assumes the general form:

$$\begin{aligned} & a \cdot T_{i+1,j} + b \cdot T_{i-1,j} + c \cdot T_{i,j+1} + d \cdot T_{i,j-1} \\ & - (a + b + c + d + e + e' + f + f') \cdot T_{i,j} \\ & = (e + e') \cdot T_w + (f + f') \cdot T_c + g \end{aligned} \quad (B-6)$$

Some of the coefficients a , b , c , d , e , e' , f , f' , and g are generally zero. The quantities $a - b$ and $c - d - e - f$ depend upon, respectively, the thermal conductivities k_x and k_y in the panel and/or the filled gap. The quantities e' and f' depend upon the heat transfer coefficients for radiation from the panel edge to the warm and cold faces of the gap, whereas g depends upon the edge-to-edge radiative heat transfer coefficients.

The $M \times N$ linear equations can be written in matrix form as

$$A \cdot T = B \quad (B-7)$$

where A is an $(M \times N)$ th order square coefficient matrix, T is an $(M \times N)$ element vector of unknown temperatures $T_{i,j}$, and B is an $(M \times N)$ element constant vector.

The temperature at each mesh point is governed only by the temperatures of nearest neighbor mesh points in both the X and Y directions. Thus, if the difference equations are arranged so that the coefficients for the equation written for mesh point (i,j) occupy the k th rows of

A and B, and the unknown temperature $T_{i,j}$ occupies the k th row of T, where $K = i + M \cdot (j-1)$, the coefficient matrix becomes block-tri-diagonal, of block order N and block size M. In other words, A subdivides into N^2 submatrices, each of which is square and of order M. Furthermore, all these submatrices except those lying along and immediately to the left or right of the main diagonal of A are null, i.e., all (M^2) elements are zeros. Further description of systems of linear equations possessing this structure and the technique used to solve them is given in the literature.*

The Fortran computer program consists of 19 subprograms - a main program, 14 subroutines, and 4 functions. Program calculations proceed sequentially as follows. Once the data is read in, the program calculates X and Y increment parameters and radiative view factors. Next, it calculates a starting temperature profile which is one-dimensional (since the lateral thermal conductivity k_x is assumed infinite). This starting profile is then used to evaluate insulation and filler thermal conductivities and edge-loss radiative heat transfer coefficients. Next, the program sets up and solves the $M \times N$ set of simultaneous linear equations, uses the resulting two-dimensional temperature profile to recompute the temperature-dependent parameters listed above, and repeats the cycle until convergence, which occurs when the weighted sum of the per cent changes in the heat fluxes falls below a certain tolerance. The program prints out temperatures and heat fluxes at the end of every cycle.

The computer program was used to run 103 cases to investigate the effects of varying edge conditions and panel dimensions. For all cases, the warm-side and cold-side temperatures were prescribed at 1500°R and 500°R , respectively. The insulation consisted of glass filler sheets alternated with 0.15 mil thick aluminum foils. The gap filler was aluminum opacified paper. Properties of these substances are summarized below:

Thermal Conductivity of Glass Filler

$$k_g(T) = (1.06 \times 10^{-7}) \cdot T \quad (\text{B-8})$$

Thermal Conductivity of Aluminum

$$k_m(T) = 244.2 - .05 \cdot T \quad ; \quad T \leq 1200^\circ\text{R} \quad (\text{B-9})$$

$$k_m(T) = 214.2 - .025T \quad ; \quad T > 1200^\circ\text{R} \quad (\text{B-10})$$

* Niendorf, L. R. and Nies, G. E., "Investigation of a Lightweight Self-evacuating Prefabricated Multi-layer Insulation system for Cryogenic Space Propulsion Stages", Final Report NASA Contract No. NAS3-6289, July, 1966.

Emissivity of Aluminum

$$\epsilon(T) = (.0001) T - .029 \quad \text{for } T > 520^\circ\text{R} \quad (\text{B-11})$$

Thermal Conductivity of Aluminum Opacified Paper

$$\text{*Parallel to layers: } k_1 = (2.76 \times 10^{-4}) T - .0545 \quad (\text{B-12})$$

$$\text{*Normal to layers: } k_n = (1.79 \times 10^{-7}) T + (6.4 \times 10^{-13}) T^3 \quad (\text{B-13})$$

In the above equations, temperatures and thermal conductivities are expressed in degrees Rankine and $\text{BTU-hr}^{-1} \cdot \text{ft}^{-1} \cdot ^\circ\text{R}^{-1}$, respectively.

Table B-I lists input parameters and four of the five output heat fluxes for all 103 cases. Of particular importance is the considerable reduction in the joint heat leak that occurs when opacified papers are placed in the joint gap - compare for example filled gap cases 1 - 3 with unfilled gap cases 4 - 6.

* These thermal conductivity data (parallel and normal to layers) was extrapolated from the best available inhouse cryogenic data.

TABLE B-I - RESULTS FOR PROBLEMS RUN WITH PROGRAM

CASE NO.	1	2	3	4	5	6	7	8	9	10	11
Gap Angle (°)	0	0	0	0	0	0	0	0	0	0	0
Panel Thickness (in)	.1	.2	.4	.1	.2	.4	.1	.1	.4	.4	.1
Panel Width (in)	10	10	10	10	10	10	10	100	10	100	50
Gap Width (in)	.5	.5	.5	.5	.5	.5	.05	.5	.05	.5	.05
Gap Filler ?	✓	✓	✓	0	0	0	0	0	0	0	0
Exposed Edge?	0	0	0	0	0	0	0	0	0	0	0
Temp. of Infinite Wall (°R)	-	-	-	-	-	-	-	-	-	-	-
Q (BTU-hr ⁻¹ -ft ⁻¹)	20.46	10.25	5.130	22.89	14.35	9.69	21.05	200.3	5.758	55.83	99.68
Q^w (BTU-hr ⁻¹ -ft ⁻¹)	19.84	9.89	4.934	22.36	13.77	8.80	20.68	199.1	5.478	54.32	99.17
Q_e (BTU-hr ⁻¹ -ft ⁻¹)	.62	.36	.196	.53	.58	.89	.36	1.2	.280	1.51	.51
Q_l (BTU-hr ⁻¹ -ft ⁻¹)	1.59	.78	.385	22.71	33.45	45.81	4.61	22.4	.264	44.61	4.50
K^{wg} (BTU-hr ⁻¹ -ft ⁻¹ °R ⁻¹)x10 ⁻³	.420	.420	.420	.8686	1.821	4.229	.5106	.4432	.6683	.7995	.4163
K_e/K_o	1.108	1.108	1.108	2.291	4.802	11.15	1.347	1.169	1.763	2.108	1.098

CASE NO.	12	13	14	15	16	17	18	19	20	21	22
Gap Angle (°)	0	10	30	60	10	30	60	30	0	0	0
Panel Thickness (in)	.1	.1	.1	.1	.4	.4	.4	.1	.1	.2	.4
Panel Width (in)	25	10	10	10	10	10	10	10	10	10	10
Gap Width (in)	.05	-	-	-	-	-	-	-	0	0	0
Gap Filler?	0	0	0	0	0	0	0	✓	0	0	0
Exposed Edge?	0	0	0	0	0	0	0	0	0	0	0
Temp. of Infinite Wall (°R)	-	-	-	-	-	-	-	-	-	-	-
Q (BTU-hr ⁻¹ -ft ⁻¹)	50.56	20.37	23.02	27.06	7.38	13.33	19.03	19.75	18.95	9.48	4.74
Q^w (BTU-hr ⁻¹ -ft ⁻¹)	50.11	19.07	17.50	15.84	3.53	2.08	1.16	19.51	18.95	9.48	4.74
Q_e (BTU-hr ⁻¹ -ft ⁻¹)	.45	1.29	5.42	11.22	3.85	11.24	17.88	.24	-	-	-
Q_l (BTU-hr ⁻¹ -ft ⁻¹)	4.56	-	-	-	-	-	-	-	-	-	-
K^{wg} (BTU-hr ⁻¹ -ft ⁻¹ °R ⁻¹)x10 ⁻³	.4401	.4074	.4588	.5220	.5904	1.056	1.458	.395	.3792	.3792	.3792
K_e/K_o	1.161	1.074	1.210	1.377	1.557	2.785	3.845	1.042	1.000	1.000	1.000

✓ - Yes
○ - No

Q : Flux from warm side of panel
 Q^w : Flux into cold side of panel
 Q_e : Flux from panel edge
 Q_l : Flux from warm side of gap

All Fluxes are for a half-panel (and half-gap), since temperature profile is symmetric about panel and gap midplanes.

B-C

TABLE B-1 - RESULTS FOR PROBLEMS RUN WITH PROGRAM

Cont.

CASE NO.	23	24	25	26	27	28	29	30	31	32	33
Gap Angle (°)	0	0	0	0	0	0	0	0	0	0	0
Panel Thickness (in)	.1	.2	.4	.1	.2	.4	.1	.2	.4	.1	.2
Panel Width (in)	10	10	10	10	10	10	10	10	10	2	2
Gap Width (in)	-	-	-	-	-	-	-	-	-	.05	.05
Gap Filler?	0	0	0	0	0	0	0	0	0	0	0
Exposed Edge?	✓	✓	✓	✓	✓	✓	✓	✓	✓	0	0
Temp. of Infinite Wall (°R)	0	0	0	200	200	200	400	400	400	-	-
Q (BTU-hr ⁻¹ -ft ⁻¹)	33.73	29.14	25.13	33.76	29.05	25.16	33.68	28.80	24.86	5.206	2.893
Q^w (BTU-hr ⁻¹ -ft ⁻¹)	12.61	1.94	-.34	12.62	1.95	-.33	12.72	2.10	-.15	4.866	2.593
Q_e (BTU-hr ⁻¹ -ft ⁻¹)	21.12	27.20	25.47	21.14	27.10	25.48	20.96	26.70	25.01	.340	.300
Q_l (BTU-hr ⁻¹ -ft ⁻¹)	-	-	-	-	-	-	-	-	-	4.682	3.956
K^{wg} (BTU-hr ⁻¹ -ft ⁻¹ -°R ⁻¹) $\times 10^{-3}$	.6746	1.166	2.010	.6752	1.162	2.013	.6736	1.152	1.989	.9647	1.336
K_e/K_o	1.779	3.075	5.301	1.781	3.064	5.309	1.776	3.038	5.245	2.544	3.523

CASE NO.	34	35	36	37	38	39	40	41	42	43	44
Gap Angle (°)	30	60	90	30	60	90	0	0	0	0	0
Panel Thickness (in)	.4	.4	.4	.4	.4	.4	.1	.2	.4	.4	.1
Panel Width (in)	10	10	10	2	2	2	2	2	2	2	2
Gap Width (in)	-	-	-	-	-	-	.02	.02	.02	.05	.1
Gap Filler?	✓	✓	✓	✓	✓	✓	0	0	0	0	0
Exposed Edge?	0	0	0	0	0	0	0	0	0	0	0
Temp. of Infinite Wall (°R)	-	-	-	-	-	-	-	-	-	-	-
Q (BTU-hr ⁻¹ -ft ⁻¹)	5.045	5.177	5.310	1.090	1.214	1.337	4.441	2.304	1.170	1.490	5.915
Q^w (BTU-hr ⁻¹ -ft ⁻¹)	4.807	4.728	4.663	.924	.884	.866	4.269	2.170	1.111	1.297	5.448
Q_e (BTU-hr ⁻¹ -ft ⁻¹)	0.237	.448	.647	.166	.331	.477	.172	.134	.059	.192	.468
Q_l (BTU-hr ⁻¹ -ft ⁻¹)	-	-	-	-	-	-	1.319	.886	.522	3.839	9.323
K^{wg} (BTU-hr ⁻¹ -ft ⁻¹ -°R ⁻¹) $\times 10^{-3}$	.4036	.4142	.4248	.436	.4856	.5348	.5703	.6317	.6701	1.689	1.451
K_e/K_o	1.064	1.092	1.120	1.150	1.281	1.410	1.504	1.666	1.767	4.454	3.826

✓ - Yes
○ - No

Q : Flux from warm side of panel
 Q^w : Flux into cold side of panel
 Q_e : Flux from panel edge
 Q_l : Flux from warm side of gap
 Q^{wg}

All Fluxes are for a half-panel (and half-gap), since temperature profile is symmetric about panel and gap midplanes.

TABLE B-I - RESULTS FOR PROBLEMS RUN WITH PROGRAM

Cont.

CASE NO.	45	46	47	48	49	50	51	52	53	54	55
Gap Angle (°)	0	0	0	0	0	0	0	0	0	0	0
Panel Thickness (in)	.2	.4	.1	.2	.4	.2	.1	.2	.4	.1	.2
Panel Width (in)	2	2	10	10	10	10	10	10	10	25	25
Gap Width (in)	.1	.1	.02	.02	.02	.05	.1	.1	.1	.02	.02
Gap Filler?	0	0	0	0	0	0	0	0	0	0	0
Exposed Edge?	0	0	0	0	0	0	0	0	0	0	0
Temp. of Infinite Wall (°R)	-	-	-	-	-	-	-	-	-	-	-
Q_w (BTU-hr ⁻¹ -ft ⁻¹)	3.585	1.899	20.17	10.20	5.155	11.03	21.83	12.05	6.631	49.62	24.92
Q_e (BTU-hr ⁻¹ -ft ⁻¹)	3.112	1.560	19.99	10.06	5.058	10.70	21.38	11.60	6.188	49.40	24.77
Q_l (BTU-hr ⁻¹ -ft ⁻¹)	.473	.339	.13	.14	.097	.33	.46	.45	.443	.22	.15
Q_{wg} (BTU-hr ⁻¹ -ft ⁻¹)	9.891	8.478	1.30	.85	.496	3.78	9.25	9.58	8.012	1.28	.85
K_e^{wg} (BTU-hr ⁻¹ -ft ⁻¹ -°R ⁻¹) $\times 10^{-3}$	2.567	3.953	.4285	.4411	.4512	.5895	.6154	.8566	1.16	.4069	.4120
K_e/K_o	6.770	10.42	1.130	1.163	1.190	1.555	1.623	2.259	3.059	1.073	1.086

CASE NO.	56	57	58	59	60	61	62	63	64	65	66
Gap Angle (°)	0	0	0	0	0	0	10	10	10	30	30
Panel Thickness (in)	.4	.2	.4	.1	.2	.4	.1	.2	.4	.1	.2
Panel Width (in)	25	25	25	25	25	25	2	2	2	2	2
Gap Width (in)	.02	.05	.05	.1	.1	.1	-	-	-	-	-
Gap Filler ?	0	0	0	0	0	0	0	0	0	0	0
Exposed Edge ?	0	0	0	0	0	0	0	0	0	0	0
Temp. of Infinite Wall (°R)	-	-	-	-	-	-	-	-	-	-	-
Q_w (BTU-hr ⁻¹ -ft ⁻¹)	12.52	25.81	13.19	51.42	26.91	14.20	4.647	3.176	2.620	6.943	6.024
Q_e (BTU-hr ⁻¹ -ft ⁻¹)	12.42	25.43	12.89	50.81	26.37	13.73	3.452	1.338	.436	2.548	.775
Q_l (BTU-hr ⁻¹ -ft ⁻¹)	.10	.39	.30	.61	.55	.47	1.195	1.838	2.184	4.394	5.248
Q_{wg} (BTU-hr ⁻¹ -ft ⁻¹)	.49	3.74	2.59	9.18	9.52	7.89	-	-	-	-	-
K_e^{wg} (BTU-hr ⁻¹ -ft ⁻¹ -°R ⁻¹) $\times 10^{-3}$	.4160	.4719	.5040	.4829	.5806	.7041	.4647	.6352	1.048	.6943	1.205
K_e/K_o	1.097	1.244	1.329	1.273	1.531	1.857	1.225	1.675	2.764	1.831	1.178

✓ - Yes
○ - No

Q_w : Flux from warm side of panel)
 Q_e : Flux into cold side of panel)
 Q_l : Flux from panel edge)
 Q_{wg} : Flux from warm side of gap)

All Fluxes are for a half-panel (and half-gap), since temperature profile is symmetric about panel and gap midplanes.

TABLE B-I - RESULTS FOR PROBLEMS RUN WITH PROGRAM

Cont.

CASE NO.	67	68	69	70	71	72	73	74	75	76	77
Gap Angle (°)	30	60	60	60	90	90	90	10	30	60	90
Panel Thickness (in)	.4	.1	.2	.4	.1	.2	.4	.2	.2	.2	.1
Panel Width (in)	2	2	2	2	2	2	2	10	10	10	10
Gap Width (in)	-	-	-	-	-	-	-	-	-	-	-
Gap Filler ?	0	0	0	0	0	0	0	0	0	0	0
Exposed Edge ?	0	0	0	0	0	0	0	0	0	0	0
Temp. of Infinite Wall (°R)	-	-	-	-	-	-	-	-	-	-	-
Q (BTU-hr ⁻¹ -ft ⁻¹)	5.312	9.796	8.742	7.075	11.77	10.20	7.744	11.24	15.77	21.31	30.03
Q^w (BTU-hr ⁻¹ -ft ⁻¹)	.225	1.686	.418	.106	1.15	.22	.049	8.84	6.86	4.92	14.51
Q_e (BTU-hr ⁻¹ -ft ⁻¹)	5.088	8.109	8.324	6.969	10.62	9.98	7.695	2.40	8.90	16.38	15.51
Q_l (BTU-hr ⁻¹ -ft ⁻¹)	-	-	-	-	-	-	-	-	-	-	-
K_{e}^{wg} (BTU-hr ⁻¹ -ft ⁻¹ -°R ⁻¹) × 10 ⁻³	2.125	.9796	1.748	2.83	1.177	2.041	3.098	.4496	.6308	.8524	.6006
K_e/K_o	5.604	2.583	4.610	7.463	3.104	5.382	8.170	1.186	1.664	2.248	1.584

CASE NO.	78	79	80	81	82	83	84	85	86	87	88
Gap Angle (°)	90	90	10	10	10	30	30	30	60	60	60
Panel Thickness (in)	.2	.4	.1	.2	.4	.1	.2	.4	.1	.2	.4
Panel Width (in)	10	10	25	25	25	25	25	25	25	25	25
Gap Width (in)	-	-	-	-	-	-	-	-	-	-	-
Gap Filler ?	0	0	0	0	0	0	0	0	0	0	0
Exposed Edge ?	0	0	0	0	0	0	0	0	0	0	0
Temp. of Infinite Wall (°R)	-	-	-	-	-	-	-	-	-	-	-
Q (BTU-hr ⁻¹ -ft ⁻¹)	24.89	22.02	49.80	25.97	14.93	52.47	30.84	22.51	56.83	37.14	30.33
Q^w (BTU-hr ⁻¹ -ft ⁻¹)	3.68	.64	48.48	23.47	10.46	46.95	21.15	7.58	45.24	18.57	5.08
Q_e (BTU-hr ⁻¹ -ft ⁻¹)	21.21	21.38	1.32	2.49	4.47	5.52	9.69	14.93	11.59	18.57	25.25
Q_l (BTU-hr ⁻¹ -ft ⁻¹)	-	-	-	-	-	-	-	-	-	-	-
K_{e}^{wg} (BTU-hr ⁻¹ -ft ⁻¹ -°R ⁻¹) × 10 ⁻³	.9956	1.762	.396	.4155	.4778	.4198	.4934	.7203	.4546	.5942	.9706
K_e/K_o	2.626	4.647	1.044	1.096	1.260	1.107	1.301	1.900	1.199	1.567	2.560

✓ = Yes
○ = No

Q : Flux from warm side of panel
 Q^w : Flux into cold side of panel
 Q_e : Flux from panel edge
 Q_{wg} : Flux from warm side of gap

All Fluxes are for a half-panel (and half-gap), since temperature profile is symmetric about panel and gap mid-planes.

-R-3-

TABLE E-1 - RESULTS FOR PROBLEMS RUN WITH PROGRAM

Cont.

CASE NO.	89	90	91	92	93	94	95	96	97	98	99
Gap Angle (°)	90	90	90	0	0	0	0	0	0	0	10
Panel Thickness (in)	.1	.2	.4	.1	.2	.4	.1	.2	.4	.4	.1
Panel Width (in)	25	25	25	2	2	2	25	25	25	2	2
Gap Width (in)	-	-	-	-	-	-	-	-	-	.1	-
Gap Filler ?	0	0	0	0	0	0	0	0	0	✓	✓
Exposed Edge ?	0	0	0	✓	✓	✓	✓	✓	✓	✓	✓
Temp. of Infinite Wall (°R)	-	-	-	0	0	0	0	0	0	0	0
Q_w (BTU-hr ⁻¹ -ft ⁻¹)	60.15	41.42	34.85	13.96	11.76	8.229	64.23	46.64	40.14	1.010	3.963
Q_e (BTU-hr ⁻¹ -ft ⁻¹)	43.86	16.79	3.62	.38	-.18	-.179	41.76	14.16	1.49	.998	3.889
Q_l (BTU-hr ⁻¹ -ft ⁻¹)	16.30	24.63	31.23	13.58	11.94	8.409	22.47	32.48	38.65	.011	.074
Q_{wg} (BTU-hr ⁻¹ -ft ⁻¹)	-	-	-	-	-	-	-	-	-	.075	-
K_e^{wg} (BTU-hr ⁻¹ -ft ⁻¹ -°R ⁻¹)x10 ⁻³	.4812	.6627	1.115	1.396	2.352	3.292	.5138	.7462	1.284	.5348	.3963
K_e^e/K_o	1.269	1.748	2.940	3.681	6.203	8.681	1.355	1.968	3.386	1.410	1.045

CASE NO.	100	101	102	103
Gap Angle (°)	0	90	0	0
Panel Thickness (in)	.1	.4	.4	.4
Panel Width (in)	2	25	25	10
Gap Width (in)	.02	-	.1	.1
Gap Filler ?	✓	✓	✓	✓
Exposed Edge ?	0	0	0	0
Temp. of Infinite Wall (°R)	-	-	-	-
Q_w (BTU-hr ⁻¹ -ft ⁻¹)	3.959	12.67	12.30	4.954
Q_e (BTU-hr ⁻¹ -ft ⁻¹)	3.920	11.99	12.26	4.902
Q_l (BTU-hr ⁻¹ -ft ⁻¹)	.039	.67	.043	.052
Q_{wg} (BTU-hr ⁻¹ -ft ⁻¹)	.060	-	.075	.075
K_e^{wg} (BTU-hr ⁻¹ -ft ⁻¹ -°R ⁻¹)x10 ⁻³	.3979	.4054	.3944	.3983
K_e^e/K_o	1.049	1.069	1.040	1.050

✓ - Yes
0 - No

Q_w : Flux from warm side of panel
 Q_e : Flux into cold side of panel
 Q_l : Flux from panel edge
 Q_{wg} : Flux from warm side of gap

All Fluxes are for a half-panel (and half-gap), since temperature profile is symmetric about panel and gap mid-planes.

PARAMETRIC CURVES - PANEL THERMAL PERFORMANCE AS A FUNCTION OF PANEL
THICKNESS, WIDTH AND GAP WIDTH.

FIGURE B-1

EFFECT OF PANEL THICKNESS ON THERMAL CONDUCTIVITY
FOR A GAP WIDTH OF .05 INCH.

K_e/K_o

- = 2 inch panel width
- = 10 inch panel width
- × = 25 inch panel width

PANEL THICKNESS, INCHES

FIGURE B-2

EFFECT OF PANEL THICKNESS ON THERMAL
CONDUCTIVITY FOR A GAP OF 0.1 INCH.

K_e/K_o

FIGURE B-3

EFFECT OF GAP WIDTH ON THERMAL CONDUCTIVITY
FOR A PANEL THICKNESS OF 0.10 INCH.

k_e/k_o

- = 2 inch panel width
- = 10 inch panel width
- × = 25 inch panel width

FIGURE B-4

EFFECT OF GAP WIDTH ON THERMAL CONDUCTIVITY
FOR A PANEL THICKNESS OF 0.40 INCH.

K_e/K_o

- = 2 inch panel width
- = 10 inch panel width
- × = 25 inch panel width

FIGURE B-6

EFFECT OF PANEL THICKNESS ON THERMAL CONDUCTIVITY
FOR AN ANGLE OF 10 DEGREES.

FIGURE B-7

EFFECT OF PANEL THICKNESS ON THERMAL CONDUCTIVITY
FOR AN ANGLE OF 30 DEGREES.

FIGURE B-8

EFFECT OF PANEL THICKNESS ON THERMAL CONDUCTIVITY
FOR AN ANGLE OF 60 DEGREES.

K_e/K_o

FIGURE B-9

EFFECT OF PANEL THICKNESS ON THERMAL CONDUCTIVITY
FOR AN ANGLE OF 90 DEGREES

K_e/K_o

- = 2 inch panel width
- = 10 inch panel width
- × = 25 inch panel width

FIGURE B-10

FIGURE B-11

FIGURE B-12

FIGURE B-13

FIGURE B-14

PANEL TEMPERATURE PROFILES NORMAL TO PANEL LAYERS, CASE 91

FIGURE B-15

PANEL TEMPERATURE PROFILES NORMAL TO PANEL LAYERS, CASE 60

- UNAFFECTED PROFILE
- × CASE AT $\frac{x}{w} = 0$
60 (CENTER OF PANEL)
- CASE AT $\frac{x}{w} = 1.0$
60 (EDGE)

$W = 25 \text{ IN. (} \approx 12.5 \text{)}$

$d = 0.1 \text{ IN.}$

$h = 0.4 \text{ IN.}$

$$\frac{K_e}{K_o} = 1,531$$

FIGURE B-16

PANEL TEMPERATURE PROFILE NORMAL TO PANEL LAYERS, CASES 46 AND 73

FIGURE B-17

LONGITUDINAL PANEL TEMPERATURE PROFILES, CASE 91

CASE 91

$W = 25$

ANGLE = 90°

$h = .4$

$\frac{K_e}{K_o} = 2.91$

FIGURE B-18

LONGITUDINAL PANEL TEMPERATURE PROFILES CASE 61

FIGURE B-19

APPENDIX C

I. REFLECTIVITY OF OXIDIZED METAL FOILS

II. AIR EXPOSURE TEMPERATURE PROFILES

I. REFLECTIVITY OF OXIDIZED METAL FOILS

Consider the oxidized surface shown schematically in Figure C-1. We divide the total radiant flux into two parts²¹.

I = flux in the positive x direction

J = flux in the negative x direction

When the radiant flux I passes through an infinitesimal layer dx of the oxide, a fraction $a I dx$ of the flux is absorbed and a fraction $s I dx$ is lost by scattering in the negative x direction. (a and s are thus defined as the absorption and scattering coefficients with units of reciprocal length.) Also, a quantity of flux $s J dx$ is added by scattering from the flux J . In addition, due to the temperature of the layer, the flux $a E_0 dx$ is contributed in the direction of negative x , where E_0 designates the black-body radiation at the temperature of interest. We therefore obtain:

$$\frac{dI}{dx} = - (a+s) I + sJ + a E_0 \quad (C-1)$$

In the same manner, we can write for the flux, J ,

$$\frac{dJ}{dx} = (a+s) J - sI - a E_0 \quad (C-2)$$

Sideways scattering has not been considered in (C-1) and (C-2). It is assumed that any radiation lost in the sideward direction is compensated for by scattering from adjacent areas. This implies that the thickness of the coating is small compared with the dimensions of the area under question. (C-1) and (C-2) also assume completely diffuse radiation and isotropic scattering in the forward and backward directions. The solution of these equations is given by Hamaker²¹ as:

$$I = A (1 - \beta_0) e^{\sigma_0 x} + B (1 + \beta_0) e^{-\sigma_0 x} + E_0 \quad (C-3)$$

$$J = A (1 + \beta_0) e^{\sigma_0 x} + B (1 - \beta_0) e^{-\sigma_0 x} + E_0 \quad (C-4)$$

OXIDIZED METAL SURFACE WITH OXIDE THICKNESS D

FIGURE C-1

where:

$$\beta_o = \sqrt{\frac{a}{a+2s}} = \sqrt{\frac{a/s}{a/s+2}} \quad (C-5)$$

$$\sigma_o = \sqrt{a(a+2s)} \quad (C-6)$$

A and B are constants to be determined by the boundary conditions. The boundary conditions for the case of interest were not applied by Hamaker. These are:

$$I = I_o \text{ and } x = 0 \quad (C-7)$$

$$J = rI + (1-r)E_o \text{ at } x = D \quad (C-8)$$

where r is the reflectivity of the parent metal. Boundary conditions (C-8) can more conveniently be put in the form:

$$(1+\rho)J - (1-\rho)J = 2\rho E_o \text{ at } x = D \quad (C-9)$$

$$\text{where: } \rho = \frac{1-r}{1+r} \quad (C-10)$$

The constants A and B for (C-3) and (C-4) subject of (C-7) and (C-9) are:

$$A = \frac{e^{-\sigma_o D} (\beta_o - \rho) (I_o - E_o)}{e^{\sigma_o D} (1 + \beta_o) (\rho + \beta_o) + e^{-\sigma_o D} (\beta_o - \rho) (1 - \beta_o)} \quad (C-11)$$

$$B = \frac{e^{\sigma_o D} (I_o - E_o) (\rho + \beta_o)}{e^{\sigma_o D} (1 + \beta_o) (\rho + \beta_o) - e^{-\sigma_o D} (\rho - \beta_o) (1 - \beta_o)} \quad (C-12)$$

Substituting (C-11) and (C-12) into (C-3) and (C-4) gives the solution for I and J. The solution for J is:

$$J = \frac{e^{-\sigma_o D} e^{\sigma_o x} (1+\beta_o)(\beta_o-\rho)(I_o-E_o) + e^{\sigma_o D} e^{-\sigma_o x} (1-\beta_o)(\rho+\beta_o)(I_o-E_o)}{e^{\sigma_o D} (1+\beta_o)(\rho+\beta_o) + e^{-\sigma_o D} (\beta_o-\rho)(1-\beta_o)} + E_o \quad (C-13)$$

Evaluating (C-13) at $x = 0$, we have for J_o ,

$$J_o = \frac{I_o [e^{\sigma_o D} (1-\beta_o)(\rho+\beta_o) + e^{-\sigma_o D} (1+\beta_o)(\beta_o-\rho)] + 2 E_o \beta_o [(\rho-\beta_o) e^{-\sigma_o D} + (\rho+\beta_o) e^{\sigma_o D}]}{e^{\sigma_o D} (1+\beta_o)(\rho+\beta_o) + e^{-\sigma_o D} (\beta_o-\rho)(1-\beta_o)} \quad (C-14)$$

But we also know that the flux J_o can be written as:

$$J_o = R I_o + \epsilon E_o \quad (C-15)$$

where R is the effective reflectivity of the oxidized surface and ϵ is the effective emissivity of the surface. Therefore, comparing (C-14) and (C-15) we can write:

$$R = \frac{e^{\sigma_o D} (1-\beta_o)(\rho+\beta_o) + e^{-\sigma_o D} (1+\beta_o)(\beta_o-\rho)}{e^{\sigma_o D} (1+\beta_o)(\rho+\beta_o) + e^{-\sigma_o D} (1-\beta_o)(\beta_o-\rho)} \quad (C-16)$$

$$\epsilon = \frac{2 \beta_o [(\rho-\beta_o) e^{-\sigma_o D} + (\rho+\beta_o) e^{\sigma_o D}]}{e^{\sigma_o D} (1+\beta_o)(\rho+\beta_o) + e^{-\sigma_o D} (1-\beta_o)(\beta_o-\rho)} \quad (C-17)$$

(C-16) and (C-17) are thus the equations for the reflectivity and emissivity of the oxidized surface as a function of a , s , r , and D . One other relation that can be used to check (C-16) and (C-17) is:

$$R + \epsilon = 1 \quad (C-18)$$

A check of (C-16) and (C-17) reveals that they meet this requirement.

One other useful relationship can be derived from (C-17). If we let D approach infinity, i.e., pure oxide, we find

$$\epsilon_{\infty} = \frac{2 \beta_o}{1 + \beta_o} \quad (C-19)$$

Substituting (C-5) into (C-19) and solving for a/s , we find

$$\frac{a}{s} = \frac{\epsilon_{\infty}^2}{2 (1 - \epsilon_{\infty})} \quad (C-20)$$

Therefore, we can relate the ratio of the absorption coefficient to the scattering coefficient directly to the emissivity of the pure oxide which can be obtained from the literature. Therefore, if either a or s is known, we can completely predict the effect of the oxide layer. A plot of equation (C-20) appears in Figure C-2.

Unfortunately, no a or s data could be found in the literature for the oxides of interest (nickel, copper, aluminum). The only a , s data found was for zirconium dioxide⁴¹. However, we have developed a method to back calculate a and s from measured data of the emissivity of oxidized metal surfaces. Using this method, the absorption and scattering coefficients as a function of temperature has been calculated from Oubre's data⁶¹ on oxidized polycrystalline nickel.

Oubre's data has been reproduced in Figures C-3 and C-4. Oubre reported his data in terms of weight gain. This was converted to an oxide thickness in microns by assuming that the layer had the density of the pure oxide. The information needed for calculating a and s from the experimental data is:

1. a plot of the emissivity of the metal as a function of temperature,
2. a plot of the emissivity of the oxide as a function of temperature,
3. a plot of emissivity versus temperature for various oxide thicknesses (such as Figure C-4),
4. Equations (C-16) and (C-20) together with the defining relations.

For nickel, the emissivity of the metal as a function of temperature appears in Figure C-3. Figure C-5 gives the emissivity of nickel oxide as a function of temperature.²⁰ As an example, consider 500°C (932°F) as the temperature of interest. From Figure C-3, nickel has an emissivity at 500°C of 0.09 ($r = 0.91$). From Figure C-5, $\epsilon_{\infty} = 0.505$. From equation (C-20), $a/s = 0.258$. From (C-5) and (C-20), $\beta_o = 0.336$ and $\rho = 0.0471$. Using equation (C-16) and various values of $\sigma_o D$, the 500°C curve of Figure C-6 was generated. From Figure C-3, for oxide thicknesses

FIGURE C-2

FIGURE C-3

EMISSION VS. TEMPERATURE-EFFECT OF OXIDE THICKNESS FOR POLYCRYSTALLINE NICKEL

REFLECTIVITY OF OXIDIZED NICKEL AS A FUNCTION OF OXIDE THICKNESS

FIGURE C-4

C-9

EMISSION OF NICKEL OXIDE

FIGURE C-5

of 0.162μ , 0.508μ , 0.757μ the emissivities are 0.16, 0.284, 0.328, respectively. With these emissivities, the corresponding values of $\sigma_o D$ can be obtained from Figure C-6. With D known, three values of $\sigma_o D$ can be obtained as 7726 cm^{-1} , 7680 cm^{-1} , and 6866 cm^{-1} . From (C-5) we can write:

$$s = \frac{\sigma_o}{\sqrt{a/s (a/s + 2)}} \quad (\text{C-21})$$

We thus calculate values of s of 10180 cm^{-1} , 10120 cm^{-1} , and 9063 cm^{-1} . Since the thickness of the oxide layer is much greater than the thickness of a monolayer of the oxide, the scattering coefficient should not be a function of oxide thickness. Therefore, the preceding values must be different due to experimental error and should be averaged to give a value of $s = 9781\text{ cm}^{-1}$. Since a/s is known, a can be calculated as 2490 cm^{-1} . In a similar manner, s and a were calculated at other temperatures and plotted as a function of temperature in Figures C-7 and C-8. Thus, a and s have been calculated as a function of temperature and equation (C-16) could be used to calculate the reflectivity of the oxidized nickel surface at other temperatures and oxide thicknesses.

In order to predict the reflectivity of oxidized metal surfaces, other than nickel, we first examine the nature of the scattering and absorption coefficients. Absorption is a fundamental property of the absorbing media depending on lattice structure, vibrational energy levels of the atoms and other basic properties. Some materials show strong absorption bands at some wave lengths and not at others. The absorbing characteristics are also manifested in the emissivity of the materials since emissivity equals absorptivity by Kirchoff's Laws. The absorption coefficient is therefore probably unique for each oxide and the coefficient of one oxide cannot be related to that of another oxide. The scattering coefficient, however, is primarily determined by physical characteristics of the oxide; that is, the number and size of voids or scattering centers in the oxide. Since all of these oxides are formed under identical conditions, it seems probable that the scattering coefficient - temperature relation for the oxides of aluminum and copper might be nearly the same as for nickel.

With the scattering coefficients of copper and aluminum assumed equal to that of nickel, the absorption coefficients can be obtained from a plot of the emissivity of the oxides as a function of temperature and equation (C-20) to determine a/s . The emissivities of the oxides as a function of temperature are given in Figure (C-9). The absorption coefficients obtained with (C-20) are given in Figure C-8. With the absorption and scattering coefficients known, the reflectivity of the oxidized surface can be obtained by equation (C-16). The reflectivity of unoxidized copper¹⁹ was taken as a constant of 0.97 in the range of $900-1300^\circ\text{F}$ and the reflectivity of aluminum¹⁹ as 0.99 in the range 300 to 900°F . The reflectivities of the oxidized metals as a function of oxide thickness are given in Figures C-10 and C-11 as calculated by this procedure.

C-11

EMISSION VS. $\epsilon_0 D$

FIGURE C-6

CALCULATED SCATTERING COEFF. FOR N₁₀

FIGURE C-7

CALCULATED ABSORPTION COEFF. AS A FUNCTION OF TEMPERATURE

FIGURE C-8

EMISSION OF COPPER AND ALUMINUM OXIDES AS A FUNCTION OF TEMPERATURE

FIGURE C-9

REFLECTIVITY OF OXIDIZED COPPER AS A FUNCTION OF OXIDE THICKNESS

FIGURE C-10

REFLECTIVITY OF OXIDIZED ALUMINUM

- ⊗ 900° F
- × 600° F
- ⊙ 300° F

FIGURE C-11

II. AIR EXPOSURE TEMPERATURE PROFILE

In order to derive the equations governing the insulation system under air exposure, we let Q_T = total heat produced by the generator, Btu/hr.

Q_S = heat passing through the thermoelectric elements and through supports in vacuum.

Q_I = heat passing through the insulation in vacuum.

If we assume that in vacuum ϕ part of the heat passes through the insulation, we can write:

$$\frac{Q_S}{Q_T} = 1 - \phi ; \quad \frac{Q_I}{Q_T} = \phi$$

Also in vacuum, if T_1 , T_2 , T_3 , and T_4 are the boundary and interface temperatures in a three component system, assuming a one-dimensional heat flow (See Figure C-12) we can write:

$$Q_I = \frac{A_A k_A (T_1 - T_2)}{X_A} = \phi Q_T \quad (C-22)$$

where:

A_A = average area of insulation, ft.²

k_A = thermal conductivity of first insulation section, Btu/hr.-ft.-°F

X_A = thickness of first insulation section, ft.

$$\text{but } X_A = x_A \ell \quad (C-23)$$

where:

ℓ = total insulation thickness, ft.

x_A = first insulation section fraction of total thickness

(C-23) into (C-22) gives upon rearrangement:

AIR EXPOSURE NOMENCLATURE

FIGURE C-12

$$\frac{A_A}{L} = \frac{\phi Q_T x_A}{k_A (T_1 - T_2)} \quad (C-24)$$

Similarly in vacuum, we can write for the supports,

$$Q_S = \frac{k_S A_S}{L} (T_1 - T_4) = (1 - \phi) Q_T \quad (C-25)$$

where:

A_S = total cross sectional area of supports and elements, ft.²

k_S = thermal conductivity of supports and elements, Btu/hr. ft.°F.

L = characteristic length of supports and elements, Ft.

Rearranging (C-25), we obtain

$$\frac{k_S A_S}{L} = \frac{(1 - \phi) Q_T}{(T_1 - T_4)} \quad (C-26)$$

Thermal conductivity data for air from Kreith⁴⁴ was plotted as a function of temperature and was found to fit the following equation:

$$k_{\text{air}} = aT + b \quad (C-27)$$

where:

$$\begin{aligned} a &= 1.895 \times 10^{-5} & 0^\circ\text{F} \leq T \leq 700^\circ\text{F} \\ b &= 4.583 \times 10^{-3} \end{aligned} \quad (C-28)$$

$$\begin{aligned} a &= 1.535 \times 10^{-5} & 700^\circ\text{F} \leq T \leq 2000^\circ\text{F} \\ b &= 9.439 \times 10^{-3} \end{aligned} \quad (C-29)$$

k is in Btu/hr.ft.°R and T is in °R.

During air exposure we can write

$$Q_I' = k_{\text{air}} A_A \frac{dT}{dx} \quad (C-30)$$

where:

$$Q_I' = \text{heat passing through the insulation during air exposure, Btu/hr.}$$

Substituting (C-2) into (C-3) and integrating we obtain:

$$Q_I' \int_0^L dx = -A_A \int_{T_H}^{T_L} (aT + b) dT$$

$$Q_I' = \frac{A_A}{L} \left[\frac{a}{2} (T_H^2 - T_L^2) + b (T_H - T_L) \right] \quad (C-31)$$

where:

T_H = hot side temperature during air exposure, °R.

T_L = cold side temperature during air exposure, °R.

Substituting (C-31) into (C-3) and rearranging, we obtain:

$$\frac{Q_I'}{Q_T} = \frac{\phi x_A}{k_A (T_1 - T_2)} \left[\frac{a}{2} (T_H^2 - T_L^2) + b (T_H - T_L) \right] \quad (C-32)$$

Similarly, for the supports and elements under air exposure, we can write:

$$Q_S' = k_S A_s \frac{dT}{dx} \quad (C-33)$$

Where: Q_S' = heat passing through the supports and elements during air exposure, Btu/hr.

Integrating, substituting (C-33) and rearranging, and assuming K_S to be constant, we obtain:

$$\frac{Q_S'}{Q_T} = \frac{(1 - \phi)}{(T_1 - T_4)} (T_H - T_L) \quad (C-34)$$

By a heat balance during air exposure, we must satisfy:

$$\frac{Q_I'}{Q_T} + \frac{Q_S'}{Q_T} = 1 \quad (C-35)$$

Substituting (C-32) and (C-34) into (C-35), we have:

$$\frac{\phi x_A^a}{2k_A (T_1 - T_2)} (T_H^2 - T_L^2) + \left[\frac{\phi x_A^b}{k_A (T_1 - T_2)} + \frac{(1 - \phi)}{(T_1 - T_2)} \right] (T_H - T_L) = 1 \quad (C-36)$$

All quantities in equation (C-36) are known except T_H and T_L . One more equation or boundary conditions must be known. If the outside area of the insulated system is large, or the generator output is small, or the outside is forced cooled during air exposure, then the outside temperature is equal to the ambient temperature, T_A ; therefore:

$$T_L = T_A \quad (C-37)$$

and equation (C-36) can be solved for T_H . If, however, Q_T is large, or the outside boundary is small, T_L will be greater than T_A . At the outer boundary, we can write the following equation:

$$Q_T = hA_o (T_L - T_A) + A_o \epsilon \sigma (T_L^4 - T_A^4) \quad (C-38)$$

where:

- h = convection coefficient at outer boundary, Btu/hr.ft.²
- A_o = outside area, ft.
- ϵ = emissivity of outside surface
- σ = Stefan-Boltzman constant, 1713×10^{-12} Btu/hr.ft.²°R⁴

We notice that if the outer boundary is known to be at ambient temperature during air exposure, then (C-36) gives the hot side temperature without knowledge of Q_T and A_o . Only ϕ and the composition and temperatures of the insulation in vacuum need be known. However, when T_L is known to be greater than T_A , then the power and outside area of the generator must be known.

In order to derive an equation for the temperature profile, we integrate (C-30) to a point x within the insulation.

$$Q_I' \int_0^x dx = -A_A \int_{T_H}^T k_{air} dT$$

$$Q_I' = \frac{A_A}{x} \left[\frac{a}{2} (T_H^2 - T^2) + b (T_H - T) \right] \quad (C-39)$$

We can also write:

$$Q_I' = \int_x^{\ell} dx = -A_A \int_T^{T_L} k_{air} dT \quad (C-40)$$

$$Q_I' = \frac{A_A}{\ell - x} \left[\frac{a}{2} (T^2 - T_L^2) + b (T - T_L) \right]$$

Equating (C-39) and (C-40) and rearranging, we obtain:

$$\frac{a}{2} T^2 + b T = \frac{a}{2} T_H^2 + b T_H - \frac{x}{\ell} \left[\frac{a}{2} (T_H^2 - T_L^2) + b (T_H - T_L) \right] \quad (C-41)$$

With T_H and T_L known from the previous analysis, equation (C-41) gives an equation in terms of T and $\left(\frac{x}{\ell}\right)$.

APPENDIX D

SPACER PROCESSING ANALYSIS

A flat sheet of spacer material of length, L^* , and width, w , may be taken as a theoretical model. This is to be placed in a tube furnace so that the flow path of the oxidizing stream will coincide with the direction of the length of the spacer. The distance coordinate, y , will also be in this direction.

A CO_2 material balance may be written for a differential volume, $(S\Delta y)$, that flows past the spacer from $y = 0$ to $y = L$:

$$(S\Delta y) \frac{dc}{dt} = 2w\Delta y N_c$$

where: S = cross-sectional area of the furnace tube
 Δy = differential length of the spacer
 c = CO_2 concentration in the differential volume
 t = time
 N_c = flux of CO_2 from one side of the spacer

Rearranging:

$$\frac{dc}{dt} = \left(\frac{2w}{S}\right) N_c \quad (D-1)$$

The flux N_c will be a function of both time and distance, y , from the start of the spacer. However, it should be noted that $N_c = 0$ at any point such that:

$$t = \frac{y}{v}$$

where v = velocity of the oxidizing stream; that is, the reaction does not start until the oxidizing stream reaches the point, y . By introducing a new time variable:

$$\tau = t - \frac{y}{v}$$

N_c will remain constant for a fixed τ . Equation (D-1) may then be integrated at constant τ with the boundary conditions:

$$\begin{aligned} y &= 0 \\ t &= \tau \\ c &= 0 \end{aligned}$$

to yield:

$$\begin{aligned} c &= \left(\frac{2w}{S}\right) N_c (t - \tau) \\ c &= \left(\frac{2wN_c}{S}\right) \frac{y}{v} \end{aligned}$$

*A table of nomenclature is presented at the end of the appendix.

Since the concentration leaving the furnace is of interest and since this concentration would be that of the differential volume at $y = L$:

$$c \Big|_{y=L} = \left(\frac{2wL}{Sv} \right) N_c \quad (D-2)$$

N_c is now a function of τ alone and may be approximated by a power law.

$$\begin{aligned} N_c &= k C_{O_2}^a C_B^b \\ &= N_c^0 \left(\frac{C_B}{C_B^0} \right)^b \end{aligned} \quad (D-3)$$

where: k = rate constant
 C_{O_2} = oxygen concentration of oxidizing stream
 C_B = the areal concentration of carbon in the binder
 C_B^0 = the areal concentration of carbon in the binder at $\tau = 0$
 N_c^0 = the flux of CO_2 at $\tau = 0$
 a, b = exponents

Since there is a one to one correspondence between the moles of CO_2 produced and the moles of carbon oxidized in the binder, the rate equation for the dissipation of carbon in the binder would then be the negative of the CO_2 flux.

$$\frac{dC_B}{d\tau} = -k C_{O_2}^a C_B^b$$

Integrating with the initial condition

at $\tau = 0$

$$C_B = C_B^0$$

yields for $b = 1$

$$\frac{C_B}{C_B^0} = e^{-k C_{O_2}^a \tau} \quad (D-4)$$

and for $b \neq 1$

$$\frac{C_B}{C_B^0} = \left\{ 1 - (1-b) \left[\frac{kC_{O_2}^a}{C_B^0(1-b)} \right] \tau \right\}^{\frac{1}{1-b}}$$

$$= \left[1 - \frac{\tau}{\beta} \right]^{\frac{1}{1-b}} \quad (D-5)$$

where β is the total reaction time.

Experiments were run to simulate the model, and the data indicate that the reaction time is infinite. Equation (D-4) would then be the applicable one. Combining this with equations (D-3) and (D-2):

$$C_{CO_2} = \left(\frac{2N_c^0 L w}{S v} \right) e^{-kC_{O_2}^a \tau} \quad (D-6)$$

Here the reaction constant k is a function of temperature, T , so that for a fixed T and a fixed CO_2 a plot of $\ln C_{CO_2}$ versus τ should yield a straight line.

Equation (D-6) has been tested this way for three cases:

Astroquartz....Oxidized at 900°F, 21% O_2 , 10 cfh air

Astrosil.....Oxidized at 1800°F, 10% O_2 , 4 cfh

Refrasil Paper.Oxidized at 900°F, 5% O_2 , 4 cfh

and the results have been plotted in Figures D-1, D-2, D-3, and D-4. The instrument response, has been accounted for by the following method.

A relation between the CO_2 concentration leaving the furnace, C , and the CO_2 concentration reading, C_r , of the analyzer may be obtained by assuming that the rate of change of C_r with respect to time is proportional to the difference in the two concentrations C and C_r :

$$\frac{dC_r}{dT} = P(C - C_r) \quad (D-7)$$

where P is a proportionality constant. Then

$$C = \left[C_r + \frac{1}{P} \frac{dC_r}{dT} \right] \quad (D-8)$$

FIGURE D-1

FIGURE D-2

FIGURE B-3

FIGURE D-4

If P can be determined, C can be obtained by applying (D-8) (determining dC_r/dT by a graphical differentiation). Equation (D-7) may be solved for C_r using equation (D-6) for C; but the result is too complicated for practical usage.

The constant P has been determined by applying a step function to the instrument:

$$\begin{aligned} C &= 0 & \tau < 0 \\ & & \tau > T \\ C &= 50 \text{ ppm} & T \leq \tau \leq 0 \end{aligned}$$

Solving equation (D-7) yields:

$$\begin{aligned} \ln \left[1 - \frac{C_r}{500} \right] &= -P\tau & \text{for } T \geq \tau \geq 0 \\ \ln \left[\frac{C_r}{500} \right] &= -P(\tau - T) & \text{for } \tau > T \end{aligned}$$

These equations have been used in conjunction with an experimental step function to obtain a value for P of $+3.51 \text{ min}^{-1}$ for the instrument used. P should be a function of experimental conditions; however, the effect of P on the correction (D-8) is not great, and $P = 3.51 \text{ min}^{-1}$ appears satisfactory when used. The correction was used only in the detailed analysis of figures and not in the correlation of Figure D-5. Figures D-1, D-3 and D-4 represent short reaction times, since the reaction is nearly complete after several minutes. Reliable data for times up to one hour were available for the Astrosil test of Figures D-2 and D-3. Figure D-2 represents the reaction from 2 to 60 minutes, while Figure D-3 represents the reaction from 0 to 2 minutes. The dotted lines of Figure D-2 represent the straight sections of the curve and include those extrapolated from Figure D-2.

It is noted that the curves do not contain a single straight line but several straight lines. This would indicate that the reaction is a complex one and is not accurately described by equation (D-6). The complexity of the reaction is supported by a chemical analysis of the spacers which indicates that a variety of substances are present. These include:

For J. P. Stevens	inorganic sulfates
9073 binder	NH^+ ion
	aliphatic hydrocarbons
	ester
	acid
	dimethyl polysilosane

For Dexter Aluminum cellulose
opacified paper

Though equation (D-6) is limited in its use, it will be useful as a guide to a correlation of the data.

Since it is desired to correlate the reaction's degree of completion with the time required for the completion, a new variable may now be defined:

$$f = \frac{C_{CO_2}}{\bar{C}_{CO_2}}$$

Here $\bar{C}_{CO_2}$ is an arbitrary CO_2 concentration which for convenience will be the carbon dioxide equivalent of the original carbon content of the spacer divided by the volume of the furnace tube. Thus,

$$\begin{aligned}\bar{C}_{CO_2} &= \frac{(x \text{ lb. carbon})}{\text{lb. spacer}} \frac{(\rho \text{ lb. spacer})}{\text{ft}^2 \text{ spacer}} \frac{(\bar{V} \text{ ft}^3 \text{ air})}{(A \text{ ft}^2 \text{ spacer}) (\text{mol air})} \\ &= \frac{(12 \frac{\text{lb. carbon}}{\text{mol carbon}})}{(12 \frac{\text{lb. carbon}}{\text{mol carbon}})} \frac{(1 \frac{\text{mol carbon}}{\text{mol } CO_2})}{(V \text{ ft}^3)} \\ &= \left(\frac{x \rho A \bar{V}}{12 V} \right) \frac{\text{mol } CO_2}{\text{mol air}}\end{aligned}$$

where

- x = wt. fraction of carbon in binder
- ρ = areal density of spacer
- A = area of spacer
- $\bar{V}$ = volume of the furnace
- V = molar volume of air at the temperature of the furnace

Denoting the time for completion as τ_f , equation (D-6) becomes:

$$\ln \left[\frac{S \bar{V} \bar{C}_f}{2 k C_{O_2}^a C_B^{ob} L_w} \right] = -k \left[C_{O_2}^a \tau_f \right] \quad (D-9)$$

Since a is unknown and b is strongly suspected of being equal to unity, it will be convenient to arbitrarily set both a and b equal to unity. While this will somewhat affect the validity of (D-9), the validity of the correlation will be unaffected since only the parameters in brackets are to be correlated and not the equation.

Noting the

$$\frac{\bar{C}}{C_B^0} = \frac{\frac{x_p a}{12V} \frac{\text{mol carbon}}{\text{ft}^3 \text{ air}}}{\frac{x_p}{12} \frac{\text{mol carbon}}{\text{ft}^2}}$$

$$= \frac{A}{V} \text{ ft}^{-1}$$

Substituting and rearranging

$$\ln \left[\left(\frac{SA}{LV_w} \right) \left(\frac{v}{C_{O_2}} \right) f \right] = \ln 2k - k(C_{O_2} \tau)$$

Therefore a correlation obtained between

$$\left[\left(\frac{SA}{LV_w} \right) \left(\frac{v}{C_{O_2}} \right) f \right] \text{ and } [C_{O_2} \tau] \quad (D-10)$$

should be a function of temperature alone.

It should also be noted that:

$$\begin{aligned} \left(\frac{SA}{LV_w} \right) \left(\frac{v}{C_{O_2}} \right) f &= \left(\frac{Q}{VC_{O_2}} \right) f \\ &= \left(\frac{12Q}{x_p A V C_{O_2}} \right) \end{aligned}$$

where Q is the volumetric flow through the furnace and should be at the temperature of the furnace. However,

$$\frac{Q}{V} = \frac{Q_{NTP}}{V_{NTP}}$$

where the subscript NTP refers to the volume at normal temperature (70°F) and pressure (1 atm.), and no temperature correction will be needed.

The results of the correlation (D-10) are presented in Figure D-5. Points have been taken for varying degrees of reaction completion and varying conditions of oxidation.

The results appear to be reasonably independent of the particular spacer material used.

Because of the scatter of data of Figure D-5 the temperature dependence cannot be determined. It has been determined, however, by residual carbon content analyses before and after processing, that carbon content decreases with increased reaction (processing) temperature. The residual carbon contents of the spacers are significantly reduced by processing and the effectiveness of processing increases with increasing temperature. Residual carbon data to support these statements are presented in Section 3.4.1.

The optimum conditions used for processing would then be the maximum service temperature of the spacer, and the oxygen concentration of air....21 per cent by volume.

NOMENCLATURE

a	- exponent
A	- area of spacer (ft ²)
b	- exponent
C	- CO ₂ concentration
C _B	- areal concentration of carbon in spacer (mol/ft ²)
C _{CO₂}	- CO ₂ concentration
C _{O₂}	- O ₂ concentration (vol fraction)
$\bar{C}$	- a reference CO ₂ concentration
$\bar{C}_{CO_2}$	- same as $\bar{C}$
f	- degree of reaction completion (dimensionless)
k	- reaction constant
L	- length of spacer (ft)
N _c	- CO ₂ flux (mol/ft ² min.)
P	- proportionality constant (min ⁻¹)
Q	- flow rate of oxidizing stream (ft ³ /hr)
S	- internal cross-sectional area of furnace tube (ft ²)
t	- time (min.)
T	- temperature (°F)
v	- velocity of oxidizing stream
V	- volume of furnace (ft ³)
$\bar{V}$	- molar volume of oxidizing stream (ft ³ /mol)
w	- width of spacer (ft)
x	- weight fraction of carbon in spacer (lb-carbon/lb-spacer)
y	- distance coordinate

Greek Letters

θ	- total reaction time (min.)
ρ	- spacer density
τ	- a time variable (min.)

Subscripts

NTP	- normal temperature and pressure (70°F, 1 atm.)
B	- binder

Superscripts

o	- refers to T = 0
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APPENDIX E
OFFGASSING DATA

Tables E-I and E-II present a summary of the insulation materials offgassing tests and empty chamber recalibration tests, respectively.

Figures E-1 through E-9 present the offgassing data for each candidate insulation material as a function of bakeout time.

TABLE E-1

OFFGASSING TESTS SUMMARY - INSULATION MATERIALS

Test No.	Furnace ***	Processing Temp. (°F)	Offgassing Temp. (°F)	Area One Side (ft ²)	Date Started	Material
* ST 7	L	900	900	80	10/22/66	Aluminum Foil
ST 8	L	UP**	900	80	10/26/66	Aluminum Foil
ST 9	H	UP	1800	80	11/12/66	Tantalum Foil
ST 11	L	1000	900	64	11/15/66	Glass Fiber Paper
ST 12	H	1640	1640	42	11/18/66	Nickel Foil
ST 13	L	1000	900	80	11/18/66	Aluminum Flake Opacified Paper
ST 14	H	UP	1640	80	11/28/66	Nickel Foil
ST 16	H	1700	1800	80	12/1/66	Quartz Cloth
ST 17	L	1550	1270	80	12/5/66	Copper Foil
ST 18	H	1700	1800	80	12/5/66	Quartz Paper
ST 19	L	1200	1270	80	12/12/66	Copper Flake Opacified Paper
ST 20	H	1550	1400	57.5	12/27/66	Quartz Paper
ST 21	L	1000	100	80	12/27/66	Glass Fiber Paper
ST 22	L	900	100	80	12/30/66	Aluminum Foil
ST 23	L	1550	1400	80	1/9/67	Quartz Paper
ST 24	H	1700	1800	80	1/16/67	Quartz Cloth
ST 25	H	1400	1270	80	1/19/67	Copper Flake Opacified Paper
ST 26	L	1700	1700	80	2/8/67	Quartz Paper
ST 28	L	1700	1700	71.5	2/13/67	Quartz Cloth
ST 29	L	1700	1800	71.5	2/16/67	Quartz Cloth
ST 30	L	1700	1800	62.5	2/23/67	Quartz Paper

* ST = Sample Test

** UP = Unprocessed

*** L = Stainless Steel Furnace

H = Inconel 600 Furnace

TABLE E-IIOFFGASSING TESTS SUMMARY - EMPTY CHAMBER RECALIBRATIONS

<u>Test No.</u>	<u>Furnace</u>	<u>Temp. (°F)</u>	<u>Date Started</u>
* RC 11	L **	900	10/19/66
RC 13	H ***	1800	11/9/66
RC 14	L	900	11/9/66
RC 15	H	1640	11/15/66
RC 16	L	900	11/21/66
RC 17	H	1640	11/21/66
RC 18	L	1270	12/1/66
RC 19	L	1270	12/8/66
RC 20	H	1800	12/12/66
RC 21	L	1270	12/15/66
RC 22	H	1400	12/20/66
RC 24	L	100	12/20/66
RC 25	H	1400	1/3/67
RC 26	L	100	1/3/67
RC 27	L	1400	1/5/67
RC 28	H	1800	1/10/67
RC 29	L	1400	1/12/67
RC 30	L	1270	1/16/67
RC 31	L	1270	1/24/67
RC 33	L	1700	2/1/67
RC 34	L	1800	2/20/67

* RC - Recalibration of Chamber

** L - Stainless Steel Furnace

*** H - Inconel 600 Furnace

Figure E-3

E-6

Figure F-4

E-7

OFFGASSING RATE, MICRON LITERS/FT² HR

ADSORPTION RATE

Figure E-8

E-11

Figure E-9

E-12

APPENDIX F

VACUUM ENCLOSURE DATA

I. MATERIALS SELECTION

II. INCONEL-600 SPECIMEN INSPECTION

I. VACUUM ENCLOSURE MATERIALS SELECTION CRITERIA

The following is a discussion of the properties listed in Table XXII, Criteria list, section 3.4.3.

(1) Availability

It is important that candidate materials considered be available in forms which most likely would be required for fabrication of an enclosure. Table F-I shows that with one exception, Rene-41 tubing, all five of the candidates are available in sheet, strip, bar, tubing and welding rod.

(2) Compatibility

The enclosure will be in contact with the insulation materials and their offgassing constituents and therefore must be unaffected by physical contact. Since nickel is a constituent of all of the candidate materials, it is reasonable that all will be compatible with nickel. The compatibility of the remaining foil and spacer insulation materials with the candidate super alloys was investigated during the vacuum enclosure specimen tests.

(3) Diffusion of the components of air

A limited amount of data was found for the diffusion rates of the components of air through the super alloys at high temperatures. The only reference obtained referred to hydrogen diffusion through Inconel, Type 446 stainless steel and an Fe-Cu-Al alloy. A diffusion rate of $.00406 \text{ cm}^3 \text{ STP/min cm Atm}^{1/2}$ for hydrogen through Inconel-600, was calculated based on this reference.³² Based on this diffusion rate and the capacity of Linde getter for hydrogen, 100,000 micron liters/gram, it can be seen that this rate of hydrogen diffusion through Inconel-600 would not require a large amount of getter. Nitrogen will not diffuse through nickel but does diffuse through other constituents of the super alloys such as iron, chromium and molybdenum.¹¹

A diffusion rate equivalent to that of nitrogen through unoxidized iron¹¹ would be high enough to present a problem if this same rate existed in a typical super alloy enclosed insulation system but it is anticipated that the diffusion rate of nitrogen through a super alloy with an oxide surface will be much less than nitrogen through unoxidized iron.

(4) Elongation

Haynes-25 and Rene-41 exhibit relatively low elongation at rupture in the temperature range of 1400 to 1500°F. The short time tensile elongation for Rene-41 at 1500°F is only four per cent at rupture as shown in Table F-II. Elongation is considered an important property because a material with a relatively high elongation can more readily stress relieve in areas of stress concentration without failure

TABLE F-IAVAILABILITY

<u>Alloy</u>	<u>Sheet and Strip</u>	<u>Plate</u>	<u>Bar</u>	<u>Wire</u>	<u>Tubing</u>	<u>Forging Stock</u>	<u>Welding Rod</u>	<u>Investment Castings</u>	<u>Sand and Resin Shell Mold Casings</u>
Hastelloy-X	x	x	x	x	x	x	E	x	x
Haynes-25	x	x	x	x	x	x	E	x	---
Inconel-600	x	x	x	x	x	x	x	---	---
Inconel-625	x	x	x	x	x	x	x	---	---
Rene-41	x	x	x	x	---	x	x	x	---

Note: x indicates availability
 --- signifies unavailability
 E indicates coated welding electrodes

TABLE F-IIELONGATION

<u>Alloy</u>	<u>Lowest Elongation (Actual Data) 70-1800°F</u>	<u>Temperature</u>	<u>Form</u>	<u>Reference</u>
Hastelloy-X	37%	1200°F and 1400°F	.109" sheet, heat treated at 2150°F rapid air cooled	Pg. 3 Haynes-Stellite F-30, 0370
Haynes-25	12%	1400°F	.109" sheet, heat treated at 2250°F rapid air cooled	Pg. 5 Haynes-Stellite F-30, 041C
Inconel-600	39%	1200°F	Hot rolled sheet	Pg. 10 Internat'l Nickel 7500-8-64T-7
Inconel-625	39%	1000°F	Not defined	Pg. 21 Internat'l Nickel 25M 1-65, S-9
René-41	4%	1500°F	.062" sheet solution heat treated at 2150°F air cooled; aged 4 hrs. at 1650°F air cooled.	Pg. 5 Haynes-Stellite F-30, 155B

than a material with a low elongation. Haynes-25 and Rene-41 exhibit lower elongation behavior than the other candidates and thus would appear more susceptible to cracking.

Per cent elongation for each of the candidate super alloys in the form described in Table F-II is shown as a function of temperature in Figure F-1.

(5) Fatigue strength at 1800°F

Fatigue strength data available at 1800°F for the candidate super alloys under consideration are very limited. The data obtained have been plotted vs. cycles of endurance as shown in Figure F-2.

(6) Machineability

In order to satisfy the enclosure fabrication requirements, the candidate material must be machineable. It is rather difficult to compare machineability of one super alloy with that of another because vendors employ different bases for rating. An effort to compare the five candidate alloys is summarized in Table F-III. All of the candidate alloys are machineable.

(7) Melting practice

It is of significance to note melting practice of the candidate super alloys as described by Table F-IV. Joining of the same alloy of different melting practice has been found to create problems during welding. Thus, in procuring different forms of the same alloy, it is important that melting practice be consistent.

(8) Notch sensitivity

Charpee V notch impact strength has been utilized to compare notch sensitivity of the candidate super alloys. The results are shown in Table F-V. Only a limited amount of data was found at 1800°F. Of the two candidates for which data are available, it can be seen that the Haynes-25 alloy exhibits an impact strength of approximately twice that of the Hastelloy-X depending upon the material conditions.

(9) Oxidation resistance

Most of the data available compare results of oxidation experiments using the weight-gain or weight-loss method, but this method may not indicate the full extent of oxidation damage. Most super alloys are susceptible to intergranular oxide penetration,⁵³ and this phenomena could be a serious problem with a thin wall portion of an enclosure. It is possible for surface oxidation to be practically nil; however, intergranular penetration at the same time could be deep enough to cause failure.⁵³

FIGURE F-1

FIGURE F-2

TABLE F-III
MACHINEABILITY

<u>Alloy</u>	<u>Machineability</u>	<u>Reference</u>
Hastelloy-X	Machineable	P. 25 Haynes-Stellite F-30, 0370
Haynes-25	Machineable Machineability rating* 10% (solution treated)	P. 29 Haynes-Stellite F-30, 041C P. 2 DMIC-134
Inconel-600	Slightly more machineable than 304SS and slightly less machine- able than 303SS	P. 27 Internat'l Nickel 6500-8-64, T-7
Inconel-625	Not as machineable as Inconel-600	Internat'l Nickel
René-41	Machineability rating* 6% (aged)	P. 2 DMIC-134

*Machineability indexes, using carbide tools, .009 - 1 pr. feed, .010-inch depth of cut and .015-inch wear level.

TABLE F-IVMELTING PRACTICE

<u>ALLOY</u>	<u>MELTING PRACTICE</u>	<u>REFERENCE</u>
Hastelloy-X	Air melted	Vendor
Haynes-25	Air melted	"
Inconel-600	Air (vacuum melted may be special ordered)	"
Inconel-625	Air (vacuum melted may be special ordered)	"
René-41	Vacuum melted	"

TABLE F-V.NOTCH SENSITIVITYCHARPY V-NOTCH IMPACT STRENGTH

<u>Alloy</u>	<u>Material Conditions</u>	<u>Test Tempera- ture</u>	<u>Typical Impact Strength</u>	<u>Reference</u>
Hastelloy-X	Plate-heat treated at 2150°F water quenched	70°F	54.1 ft-lb	Haynes-Stellite F-30,0370 ↓
	" " " "	1500°F	58.0 ft-lb	
	Plate-heat treated at 2150°F and aged 500 hr. at 1800°F	70°F	20.0 ft-lb	
	Plate-heat treated at 2150°F and aged 168 hr. at 1500°F	1500°F	29.0 ft-lb	
Haynes-25	Plate-heat treated at 2225°F water quenched	70°F	193 ft-lb	Haynes-Stellite F-30, 041C ↓
		500°F	219 ft-lb	
		1000°F	201 ft-lb	
		1200°F	170 ft-lb	
		1400°F	143 ft-lb	
		1600°F	120 ft-lb	
Inconel-600	Notch ductile	1800°F	106 ft-lb	Internat'l Nicke ↓
Inconel-625	Notch ductile			
René-41				

The oxidation resistance, Table F-VI includes values of intergranular oxide penetration for different time periods.⁵³ The data which were obtained were for a maximum of 1000 hours. The depth of intergranular penetration in Rene-41 is given as greater than 0.003 inch for continuous exposure of 1000 hours at 1800°F, and 0.002 inch for Inconel-600 with cyclic exposure. The depth of penetration after 10,000 hours should be greater than the 1000 hour penetration depth.

ORNL indicated that oxidation penetration in superalloys may be independent of pressure in some cases. Stellite Division and Oak Ridge personnel indicated that the oxidation resistance of Hastelloy-X and Inconel-625 is the best of the superalloys. Precipitation hardenable alloys are particularly susceptible to intergranular oxide penetration.

(10) Phase Change Embrittlement

Some super alloys are subject to phase change embrittlement after long periods of exposure to high temperatures. The data obtained from the International Nickel Company indicated that Inconel-600 does not undergo phase change embrittlement. Inconel-625 undergoes a Gamma-prime phase change after long periods of exposure at 1200°F.

(11) Strength, creep

Since vendors employ various means of comparing creep stress, candidates were compared by plotting initial stress to produce one per cent total elongation as shown in Figure F-3. Data on this basis was available for Haynes-25 and Hastelloy-X only. For 10,000 hours' service, extrapolation of the Hastelloy-X curves indicates an initial stress to produce one per cent total elongation of less than 1000 psi.

(12) Strength, rupture

Stress rupture data for sheet stock at 1800°F is plotted vs. time for each of the candidate super alloys in Figure F-4. Little data are available for 10,000 hours, and extrapolation of existing data, indicate a stress rupture value of 2000 psi or less for all super alloy candidates under consideration.

(13) Alloy thermal conductivity

Thermal conductivity is of significant importance in feed-through and support considerations of an enclosure system. A low thermal conductivity is desirable in order to realize the full performance advantages of a Super Insulation system. Alloy thermal conductivities vs. temperature are shown for each of the candidate materials in Figure F-5.

TABLE F-VIOXIDATION RESISTANCE

(Intergranular Oxide Penetration in Inches)

<u>Alloy</u>	<u>1 hr.</u>	<u>10 hr.</u>	<u>100 hr.</u>	<u>1000 hr.</u>	<u>10,000 hr.</u>	<u>Reference</u>
Hastelloy-X	$.17 \times 10^{-3}$ (33)*					DMIC-214
Haynes-25	$.21 \times 10^{-3}$ (33)*	$.70 \times 10^{-3}$ (35)*	1.60×10^{-3} (35)*			DMIC-214
Inconel-600			$.9 \times 10^{-3}$ (2000°F) (27*)	$2 \times 10^{-3**}$		DMIC-214 + Internat'l Nickel
Inconel-625						DMIC-214
Rene-41	$.30 \times 10^{-3}$ (33)*	$.75 \times 10^{-3}$ (31)*	1.7×10^{-3} (31)*	$>3.0 \times 10^{-3}$		DMIC-214

*DMIC-214 reference number

**Cyclic exposure (Huntington Alloy Products Division Data)

ALLOY THERMAL CONDUCTIVITY vs TEMPERATURE

(14) Vaporization rate

Super alloy constituents such as chromium, manganese, iron, nickel, cobalt, carbon and silicon are lost at high temperature in vacuum due to vaporization.⁵ Vaporization rates of super alloys increase with chromium and manganese concentration and temperature.

It is difficult to calculate a vaporization rate for an alloy because exposed alloy surface composition is continually changing. As concentration of the more volatile constituents decrease due to vaporization, the overall evaporation rate decreases. Data obtained by D. T. Bourgette⁵ and R. C. Bates³ are the basis for the vaporization rate curves of Figure F-6. These curves were derived assuming a uniform surface loss, based on weight loss. However, the actual material loss may not be confined to the surface but might result in a loss throughout the thickness, affecting the density and soundness of the structure. Calculations based on work by R. C. Bates³ indicate the loss from a Hastelloy-X surface at 1800°F for 10,000 hr to be greater than .004 inch (assuming a uniform surface loss). In an enclosure design with a support which might typically be 0.008 to 0.012 inch thick, this could present a severe problem.

Loss of alloy constituents due to vaporization could result in the following effects:

- . Loss of material, reducing cross-sectional area of stressed components.
- . The effect of voids or stress risers due to material loss on physical properties of material that remains.
- . The effect of change in composition due to loss of more volatile constituents on properties of the material that remains.

It was first believed that vaporization rates were independent of pressure in the 10^{-3} to 10^{-4} mmHg pressure range. This was based on a comparison of the work by Bates³ at 10^{-3} mmHg and Bourgette⁵ in the 5×10^{-7} to 1×10^{-9} mmHg range.

However, experiments conducted at ORNL in the pressure range of 10^{-3} to 10^{-5} mmHg, the pressure level anticipated in typical vacuum enclosures, indicated that if sufficient oxygen is present in the vacuum environment, oxide formation at this pressure level may protect super alloys from vaporization. It was believed that any residual oxygen or carbon dioxide present in the vacuum enclosure at this pressure level might oxidize the surface on the vacuum side.

During testing of the Inconel 600 and Hastelloy-X specimens, Sections 3.4.3 and 3.4.3- respectively, mass spectrometer analysis showed no oxygen to be present in the vacuum enclosure specimen environment and therefore no protective oxide formation on the superalloy can be expected.

VAPORIZATION RATE OF ALLOY SURFACE IN VACUUM AT 1800°F

Figure F-6

Experiments were conducted at ORNL on the behavior of Haynes-25 in high vacuum (5×10^{-7} - 1×10^{-9} mmHg) for a long period of time. The Haynes-25 alloy was exposed to hard vacuum for test periods up to 5000 hours at 980°C. Based on extrapolation of these data to 10,000 hours, total vaporization of Haynes-25 over this period is 17.5 mg. per sq. cm. or a 0.378 mil thickness reduction from each surface of the specimen if uniform surface loss is assumed.

In order to avoid the superalloy vaporization problem the use of a refractory metal as a candidate vacuum enclosure material was explored with ORNL. Since the coating of refractory metals for service in an oxidizing atmosphere for long periods has not yet been completely successful, this was not pursued further. The possibility of the use of an inert gas purge on the air side of the enclosure in order to protect against oxidation was discussed. ORNL personnel believed that the purity of the purge gas, typically argon, required to avoid a problem in the use of refractory would be greater than that which could be measured. Also, in using a controlled inert atmosphere, the impurities of the purge and contaminants within the refractory and the power system enclosure could react to cause a problem with the refractory metal.

The possibility of a vacuum wall refractory - air wall superalloy was also explored. In general, personnel at Stellite and Oak Ridge indicated many problems in the use of a bimetallic refractory superalloy wall. ORNL suggested that platinum and its alloys be considered for vacuum enclosure requirements.

(15) Weldability

A single weldability test common to all vendors for the evaluation of all super alloy weldability does not exist. This survey indicated, however, that all of the super alloy candidates can be welded for vacuum service. Hastelloy-X and Inconel-625 appear more readily weldable than the other candidates.

(16) General Considerations

Effect of Long Time Exposure to High Temperature

Experiments at ORNL have indicated that exposure of Haynes-25 alloy to 1600°F for periods of 1000 hours results in a phase change embrittlement. Both Haynes Stellite Division and ORNL also recommended avoiding the use of the precipitation hardenable superalloys such as Rene-41, Inconel-X, Inconel-700, and Inconel-W because of the change in their properties over periods of time in the temperature range of 1400 to 1800°F.

Materials Compatibility of Superalloys

ORNL indicated that dissimilar metal contact with the hot vacuum enclosure should be avoided. This situation could exist if the

radiation shields of the multi-layer insulation system were not carefully installed. It was indicated that dissimilar metal contact at these temperatures of 1400-1800°F could cause formation of highly undesirable bimetallic compounds. Some of these bimetallics could have melting points below the operating temperature of the enclosure and in addition could be very brittle, causing cracking. At Stellite Division it was indicated that the superalloys are not compatible with a pure nitrogen atmosphere at 1800°F, and nitriding of the superalloy would occur at this temperature. However, testing at Stellite has indicated that one-half percent of oxygen or more is sufficient to form a protective layer, eliminating the nitriding effect.

II. INCONEL-600 SPECIMEN EXAMINATION

After 300 hours of testing at 1800°F, a leak developed in the specimen and the test was terminated. The specimen was removed from the test apparatus and examined.

In order to determine exact leakage areas, a helium mass spectrometer leak detection was conducted. Approximately 10 pinhole leaks, both in the insulation covered portion and exposed portion of the specimen, were found. Photographs, 20 power magnification, of the vacuum side specimen surface for areas both beneath the insulation and exposed are shown in Figure F-7 and Figure F-8, respectively. Evidence of vaporization and internal oxidation can be seen in the exposed area, Figure F-8, by the dark areas which are void spaces. The long void in the center was found to be a leakage area.

Specimen Microstructures

Microstructures are shown for the specimen before and after test in Figures F-9 and F-10 respectively. Figure F-9 (before test) is shown with 100-power magnification, and Figure F-10 (after test) is shown with 200 power magnification. In Figure F-10 (after test) oxidation can be noted on the surface of the specimen exposed to the air (right side in photograph). Surface oxidation appears to be less than 1 mil although this would be expected in the short exposure time of the test. Surface voids probably caused by vaporization of constituents can be noted on the side of the specimen exposed to the vacuum environment (left side in photograph). Although the before and after photographs are of different magnifications, an apparent grain growth from grain size 6 (28-48 grains/in²) to grain size 2 (1-1/2 to 3 grains/in²) can be observed. This grain growth is believed to have occurred because the specimen was subjected to a temperature equal to or slightly above the annealing temperature for the 300-hour period.

Microstructures, 100 power magnification, are shown for the specimen longitudinal weld area before and after test in Figure F-11 and F-12, respectively. The general metallurgical condition of the weld

Figure F-7 Inconel-600 Test Specimen, Vacuum Surface
Beneath Astroquartz Cloth

Figure F-8 Inconel-600 Test Specimen,
Vacuum Surface Exposed

Figure F-9 Inconel-600 Test Specimen,
Microstructure Before Testing

Figure F-10 Inconel-600 Test Specimen
Microstructure After Testing

Figure F-11 Inconel-600 Test Specimen Weld Zone
Microstructure Before Testing

Figure F-12 Inconel-600 Test Specimen Weld Zone
Microstructure After Testing

area appears to be equal to or better than that of the parent metal. The subsurface oxidation, dark crevice at upper left of Figure F-12, may have been caused by stress concentrations at this point. Dendrites can be seen on the left of the specimen in Figure F-11.³⁴ Dendrites can be formed when rapid cooling such as would occur in a weld zone takes place.

Component Vaporization and Stress Level

Initial discussions with Oak Ridge National Laboratory, indicated that a low partial pressure of oxygen on the enclosure vacuum side would tend to retard vaporization of the superalloy components. Mass spectrometer analysis conducted during the 300 hour test disclosed that no oxygen was present. Thus the gaseous constituents of the vacuum environment do not appear to offer possibilities for protective oxide formation on the super alloy. Later discussions with Oak Ridge National Laboratory concerning analysis of the Inconel-600 test indicated that component vaporization may also be a function of stress level and that stresses may cause accelerated vaporization of manganese and chromium from superalloys. Specimen stress level was high, 1300 psi, which is 150 psi greater than the 10,000 hour rupture test of 1150 psi. A stress level based on a 1% creep in 10,000 hours, a commonly used design criteria, would be about 550 psi.

Diffusion of Air Components

During the test period mass spectrometer gas analysis of the vacuum environment indicated that diffusion of air through Inconel-600 does not appear to be a problem. This test period was only 300 hours, however, and a longer test duration, 2000 hours, would be required to substantiate this initial observation.

Insulation Materials Compatibility

At the conclusion of the test the materials compatibility fixture was disassembled and insulation samples examined visually for evidence of incompatibility. No evidence of incompatibility with the specimen enclosure, Inconel-600, or vacuum environment could be detected, Section 3.5.1.

Specimen Creep

Evaluation of specimen creep was made by comparing dimensions of the specimen before and after testing. Specimen diameter measurements indicated that an elongation of approximately 1 to 2-1/2 per cent occurred over the portion of the specimen covered by insulation and an elongation of 3 to 4 per cent occurred in the exposed portion. Based on the stress level, 1300 psi, an elongation of about 1.4% would be predicted from published data. These results indicate that limited structural support may have been gained from the Astroquartz-tantalum

insulation wrapped over the specimen. However, the effect of any insulation structural support would have to be very carefully investigated before it could be used with any confidence for design.

APPENDIX G
INSULATION DYNAMIC TESTS

In order to determine the layer densities of the insulation combinations at various loadings, the Load Density tester shown in Figure G-1 was used. Curves for compressive load vs. insulation layer density of each insulation combination were generated based on the data obtained in the range of 0.01 to 0.1 psi. The resulting curves are shown in Figure G-2 through G-6.

Figures G-7 through G-11 show samples of the insulations before and after vibrational testing.

Figure G-1 Load Density Tester

LOAD VS DENSITY
ALUMINUM FOIL - GLASS FIBER PAPER

LOAD VS DENSITY
ALUMINUM FOIL - ALUMINUM FLAKE OPACIFIED GLASS FIBER PAPER

FIGURE G-3

LOAD VS DENSITY
COPPER FOIL-REFRASIL QUARTZ PAPER

FIGURE G-4

LOAD VS DENSITY
NICKEL FOIL - ASTROQUARTZ CLOTH

FIGURE G-5

LOAD VS DENSITY
COPPER FLAKE OPACIFIED QUARTZ FIBER PAPER (PROCESSED)

FIGURE G-6

G-8

Figure G-7 Aluminum Foil-Glass Fiber Paper Composite
Before and After Vibrational Testing

Before Vibrational Testing

After Vibrational Testing

Figure G-8 Aluminum Foil-Aluminum Flake Opacified Glass Fiber Paper Composite Before and After Vibrational Testing

Figure G-9 Copper Foil-Refrasil Quartz Fiber Paper Composite Before and After Vibrational Testing

Figure G-10 Nickel Foil-Astroquartz Cloth Composite Before and After Vibrational Testing

Before Vibrational Testing

After Vibrational Testing

Figure G-11 Copper Flake Opacified Refrasil Quartz Fiber Paper Composite Before and After Vibrational Testing

7.0 Contract Reports

Quarterly Progress Report of Research and Development in a Thermal Insulation Study, April-June 1966, ALO-3632-4

Quarterly Progress Report of Research and Development in a Thermal Insulation Study, July-September 1966, ALO-3632-7

Quarterly Progress Report of Research and Development in a Thermal Insulation Study, October-December 1966, ALO-3632-11

Quarterly Progress Report of Research and Development in a Thermal Insulation Study, January-March 1967, ALO-3632-14

Quarterly Progress Report of Research and Development in a Thermal Insulation Study, April-June 1967, ALO-3632-17