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CONSEQUENCES OF INTERNAL HEAT FLOW IN RADIOISOTOPE HEAT SOURCES*

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ABSTRACT

Equations relating the maximum internal temperatures of various-shaped fuel forms and other source parameters are given. By using calculated results, the use of cermet is compared with the use of oxide-metal laminar structures as methods of decreasing the resistance to heat flow in the interior of heat sources and achieving higher surface heat fluxes without exceeding maximum allowable internal temperatures. The laminar composite fuel form is moderately superior.

The expected vapor phase migration and equilibrium distribution of a volatile radioactive fuel inside a heat source capsule is described. Redistribution does not always result in serious problems and can reduce the cost of source fabrication.

from the heat source and the size of some sources are limited by the maximum allowable internal temperature.

This report gives basic equations for the prediction of these limitations for simple cases. It is shown that the maximum available surface heat flux can be increased by the use of cermets or laminar metal structures when radioisotope compounds of poor thermal conductivity are used.

The existence of temperature differences within the capsule also leads to volatile transfer of radioactive material. The factors which determine the equilibrium distribution of the material are discussed, and a mathematical analysis is given for a simple case.

INTRODUCTION

A radioisotope heat source is generally considered to consist of a quantity of radioactive fuel and its container. The radioactive fuel can be in the form of a single compound of the radioisotope, or it can be associated with a matrix material of better thermal conductivity.

Large temperature differences can exist between the surface and the interior of the radioactive fuel (defined as the radioactive material with or without a diluent or matrix material) in heat sources. These gradients are created by the flow of heat, which is generated throughout the radioactive material, to the surface. Very high internal temperatures can result.

There are no specific rules for setting the maximum allowable internal temperature for a heat source. It must be set on the basis of the properties and use of the particular source. The melting point of the fuel has often been set as the maximum allowable temperature. Certainly, melting can in some cases result in a volume change which can damage the source. In other cases, it can allow an undesirable movement of material within the capsule. However, in other applications, melting in the interior of the radioactive mass of material does no harm. The maximum surface heat flux which can be obtained

EQUATIONS FOR FUEL FORMS OF SIMPLE GEOMETRIC SHAPE

Equations relating the difference between the surface temperature and the maximum internal temperature to various parameters for fuel forms of simple geometric shapes are given in Table 1. The symbols used in the equations are defined as follows:

ΔT = difference between the maximum allowable internal temperature and the surface temperature,

P = power density (rate of heat generation per unit volume) of the fuel,

h = distance from surface to plane of maximum temperature in a planar fuel form,

k = thermal conductivity of the fuel,

q = total rate of heat production by the source,

A = surface area through which heat leaves the fuel form,

D = diameter of a cylindrical or spherical fuel form,

l = length of a cylindrical fuel form,

q/A = surface heat flux.

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These equations were derived under the assumption that P and k are constant throughout the fuel and that the surface temperature is constant. Heat losses from the edges of planar fuel forms and from the ends of cylindrical fuel forms are neglected.

It can be seen that the maximum heat output of a spherical fuel form is limited by $\Delta T_{\max}$. The total power is not limited for flat or cylindrical fuel forms since the area or length respectively can be increased. The surface heat flux, q/A , is limited for all fuel forms.

The value of $\Delta T_{\max}$ is determined by the application in which the source is used as well as by the nature of the fuel materials. The surface temperature of the source is an important design parameter in such applications as thermionic and thermoelectric generators and is set in the design of the equipment. Since the required surface temperature may be quite high, $\Delta T_{\max}$ will be seriously restricted unless the melting point of the fuel material is very high or interior melting is allowed.

The maximum surface heat flux is an important parameter in certain heat source applications. It has been a limiting factor in the design of thermionic generating units. These units require a high heat flux delivered at a high temperature. The surface temperature of the fuel must be at least a little greater than the temperature at which the heat is delivered.

It can be seen that the maximum surface heat flux, q/A , is proportional to $(kP)^{1/2}$. Consequently, high thermal conductivity and high power density are desirable.

IMPROVEMENT OF THE THERMAL PROPERTIES OF HEAT SOURCES

The product kP can be improved for fuel of poor thermal conductivity by the addition of a metal of high conductivity. This can be done in two ways: (1) a cermet can be produced, and (2) oriented conductors of metal can be introduced into the body of the radioactive material.

The effectiveness of these methods in increasing the thermal conductivity is shown by the calculated curves in Figure 1. These curves assume that the thermal conductivity of the radioactive material is very small compared to that of the metal. Both phases are assumed to be 100% dense. The thermal conductivity of the cermets was calculated using three different equations⁽¹⁻³⁾ chosen from the many that various people have derived for the conductivity of aggregates. The oriented conductors give moderately better results.

Figure 2 is a chart which aids in the rapid calculation of kP for any combination of a highly conducting metal and a poorly conducting radioactive material providing the values of k and P for the pure materials are known.

Table 2 gives the maximum surface heat flux for fuel forms composed of an optimum laminar combination of tungsten metal and various radioactive materials. These data were calculated using the thermal conductivity of Jun and Hoch⁽⁴⁾ for tungsten. Power densities were taken from references 5 and 6. The thermal conductivities of the radioactive materials were all assumed to be 0.03 watt/cm °C. This value has little influence on the thermal conductivity of the composite fuel and exact values for many radioactive materials are not obtainable at the temperatures under consideration.

The proportional increase in the maximum surface heat flux of a laminar composite fuel form as compared to a pure radioactive material fuel form depends on the relative thermal conductivities of the metal and radioactive material. As an example, an increase of 3.5 fold could be achieved with a fuel of strontium oxide in combination with tungsten while an increase of only 2.9 fold could be achieved with one of curium sesquioxide and tungsten. This calculation assumes thermal conductivities of 0.02 and 0.03 watt/cm °C, respectively, for the oxides. The value for strontium oxide was obtained from E. E. Ketchen⁽⁷⁾ and that for curium sesquioxide was estimated.

THE EFFECTS OF VOLATILITY IN THERMIONIC HEAT SOURCES

There are limited data which indicate that certain radioactive fuels are moderately volatile under the proposed conditions of use. Consequently, there has been concern that these materials may leave the desired location by volatile transfer to other parts of the capsule.

It will be shown that this volatility will cause no severe problems with regard to the heat flow characteristics of a well designed source form and that volatile transfer will, in fact, tend to create a distribution of radioactive material within the capsule similar to the ideal distribution regardless of the initial physical form of the radioactive material. However, this does not preclude other problems caused by volatility such as the plugging of helium vents.

Recognition of this phenomenon can lead to a worthwhile reduction in the cost of fabrication of many sources. The material can be encapsulated as a loose powder instead of as a fabricated form. It will distribute itself in approximately the desired manner.

Figure 3-B shows a thermionic heat source which is typical of many proposed designs. The useful heat flow leaves through one end of the cylindrical container. The rest of the container is assumed to be insulated so as to reduce extraneous heat losses to a low level. The radioactive material is attached to the inner surface of the cylinder at the end from which the useful heat flow passes. The temperature of this end of the container is

"Superior numbers refer to similarly-numbered references at the end of this paper".

set in the design of the apparatus. The volume of the container is appreciably greater than the volume of the radioactive material in order to allow space for the helium released by the radioactive decay of an alpha emitting radioisotope.

It is assumed that the radioactive material vaporizes and condenses without change in its properties. It is also assumed that all of the heat is generated within the radioactive material. Thus, the reasoning will not necessarily apply if an appreciable fraction of the energy is emitted as gamma rays.

The volatile radioactive material will distribute itself in a continuous layer covering the inner surface of the capsule. No part of the surface of the capsule can remain bare so long as at least some heat escapes from all parts of the outer surface. This results from the fact that heat can flow only from a higher to a lower temperature. Furthermore, the heat leaving a bare surface can come only from radioactive material which must, therefore, be at a higher temperature than the bare surface. Transport of volatile material from the hotter to the cooler surface will occur.

The temperature of the inner surface of this layer will be uniform at equilibrium. The reason for this uniformity is apparent when one considers that if a difference in temperature did exist material would vaporize from the hotter surface and condense on the cooler surface.

For thin layers essentially all of the heat loss from any part of the radioactive layer passes through the outer surface of the layer. This rate of loss is essentially equal to the rate at which heat is generated within that part of the layer.

This equality exists because of the lack of other significant modes of heat loss or gain. The radioactive material layer cannot gain or lose heat by conduction or thermal radiation across the interior of the capsule because of the uniform inner surface temperature. Conduction within the layer parallel to the surface will be small as compared to flow from the outer surface of the layer so long as the thickness of the layer is small compared to the dimensions of the capsule.

It is apparent that the layer will be thick at the end from which the thermionic heat flow occurs and thin on the surfaces corresponding to the well insulated parts of the capsule. Thus, the radioactive material will tend to locate itself in the desired manner.

The thickness of this layer can be predicted mathematically by simple equations for the case in which the thickness of the layer of radioactive material is small as compared to the diameter and length of the capsule. This will be the case when an appreciable void volume has been allowed to contain helium generated by alpha decay. In this case the layer can be treated as a flat plate and the following equations from Table 1 will apply to any small area of the layer of radioactive material:

$$q = PAh \quad (1)$$

$$\text{or} \quad h = \frac{q/A}{P} \quad (2)$$

where q = rate of heat flow,

A = area,

P = power density,

h = thickness of radioactive layer.

The thickness is directly proportional to the rate of heat loss per unit area.

Application of the equation from Table 1 for the maximum internal temperature of a planar source form leads to Equation 3, which relates the outer surface of the radioactive layer to the constant inner surface temperature.

$$T_o = T_i - Ph^2/2k \quad (3)$$

where T_o = temperature of the outer surface of the radioactive layer,

T_i = temperature of the inner surface,

k = thermal conductivity of the fuel.

The other symbols have been defined. Equations 2 and 3 can be combined to produce Equation 4.

$$T_o = T_i - \frac{(q/A)^2}{2Pk} \quad (4)$$

It can be seen that T_o will vary and will be at a maximum when (q/A) is at a minimum. Thus, the outer surface of the radioactive material layer will be hotter for the well insulated parts of the capsule than at the end from which the thermionic heat flow occurs.

One consequence of the uniformity of the interior temperature, T_i , is that the lowering of T_i at any point will result in a general lowering of T_i throughout the source at equilibrium. One method of accomplishing this is to introduce oriented metallic conductors into the radioactive material layer on the thermionic end of the capsule (see Figure 4).

The change in T_i produced by this method can be of great practical importance as is shown by the following example. If the radioactive material layer at the thermionic end of the capsule is assumed to consist of pure $^{244}\text{Cm}_2\text{O}_3$ and the values of 50 watts/cm², 26.4 watts/cm³, and 0.03 watts/cm °C are used for q/A , P and k , respectively, T_i will exceed T_o by 1580°C according to Equation 4. If T_o is fairly high, part of the Cm_2O_3 will be molten. If an optimum combination of tungsten metal and $^{244}\text{Cm}_2\text{O}_3$ is used, the temperature difference will be only 187°C.

A high value of T_i has three undesirable consequences: (1) The stored helium gas is at the

temperature T_i and its pressure increases with temperature. (2) The metal wall of the capsule at a distance from the thermionic surface tends to increase in temperature as T_i increases. This leads to weakening and accelerated deterioration. (3) The high temperature of the capsule walls also will cause an increase in the heat loss through the insulation.

So far, the temperatures and heat flow have referred only to the layer of radioactive material. When it can be assumed that the flow of heat along the capsule walls parallel to the surface is negligible, (q/A) can be considered to be the heat flow from the surface of the capsule. However, in many cases this assumption will involve considerable error.

In the common case of a thick-walled metal capsule, the conduction of heat by the walls to the low temperature end of the capsule will have a substantial leveling effect on T_o , q/A , and h . The heat flow, q/A , from the radioactive material layer in any part of the hotter (insulated) part of the system will now equal the sum of the heat lost through the insulation and the heat conducted along the capsule walls. This greater flow of heat requires a thicker deposit of radioactive material to hold T_i constant. This is illustrated in Figure 4.

It should be noted, however, that the overall effects of the thermal conductivity of thick capsule walls are beneficial. The heat flowing along the walls flows to thermionic surface and is not lost. Heat losses are, in fact, reduced since the temperature of the insulated parts of the capsule are lowered toward that of the thermionic end which is fixed by the design of the device. This lowering of the temperature will also result in greater strength and less deterioration for the capsule metal. The movement of radioactive material away from the thermionic surface also reduces the thickness of the layer on this surface. Since T_o is fixed, this thinning of the radioactive layer causes a lowering of T_i which is the temperature of the helium storage volume. Thus, the pressure exerted by a given quantity of helium will be reduced.

Although this discussion was directed toward thermionic heat sources, the basic conclusions will apply qualitatively to any capsule geometry. The material will tend to concentrate on the surfaces from which the greatest heat flow occurs and T_i will be constant at equilibrium.

The characteristics of the ideal capsule for a volatile radioactive material in applications where internal temperature is a limiting factor can be summarized as follows: (1) It should have thick walls of a highly conducting metal. (2) Internal metallic heat paths should be used to conduct the heat to the desired surface.

A desirable distribution can be obtained even if the radioactive material is encapsulated as loose lumps or powder when comparatively fast migration toward the equilibrium distribution occurs. No

untoward situation will occur during the migration although the temperature in the center of a charge of loose powder might become very high. This would ordinarily do no more than cause rapid vaporization and migration to the cooler surfaces of the capsule. The material in contact with the capsule cannot reach a temperature much above the operating temperature because of rapid heat transfer to the capsule surface.

In many sources, the rate of volatile migration will not be rapid enough for an equilibrium distribution to be established during the life of the source, although at least some movement toward this distribution will always occur. The available vapor pressure data for most heat source compounds are not adequate for good estimations of migration rates.

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ILLUSTRATIONS

- Figure 1. Comparison of the Thermal Conductivities of Laminar Composite Fuels and Cermets.
- Figure 2. Comparison of Surface Heat Flux Factors of Laminar Composites and Cermets Fuels.
- Figure 3. Distribution of Radioactive Material Inside Capsule.
- Figure 4. Qualitative Representation of Fuel Distribution in Three Models.

Table 1. Equations for Fuel Forms of Simple Geometric Shape

Parameter	Flat Plate	Solid Cylinder	Solid Sphere
$\Delta T =$	$\frac{Ph^2}{2k}$	$\frac{PD^2}{16k}$	$\frac{PD^2}{24k}$
$q =$	PAh	$\frac{\pi PD^2 l}{4}$	$\frac{\pi PD^3}{6}$
$q_{\max} =$	$A(2kP\Delta T_{\max})^{1/2}$	$4\pi l k \Delta T_{\max}$	$\frac{\pi}{6} \left[\frac{(24k\Delta T_{\max})^3}{P} \right]^{1/2}$
$(q/A)_{\max} =$	$(2kP\Delta T_{\max})^{1/2}$	$(kP\Delta T_{\max})^{1/2}$	$\left(\frac{2kP\Delta T_{\max}}{3} \right)^{1/2}$

Table 2. Maximum Surface Heat Fluxes of Laminar Composite Planar Fuel Forms

Radioactive Compound	Power Density, watts/cc	Melting Point, °C	Surface Temp., °C	Maximum Surface Heat Flux, watts/cm ²
⁹⁰ SrO	2.08	2430	1250	35
			1500	31
			1800	25
¹⁴⁷ Pm ₂ O ₃	2.36	2350	1250	36
			1500	32
			1800	25
²⁴⁴ Cm ₂ O ₃	26.4	1950	1250	98
			1500	78
			1800	45
²³² UO ₂	33	2176	1250	125
			1500	106
			1800	78
			2000	53
²²⁸ ThO ₂	1270	3050	1250	1050
			1500	970
			1800	860
			2000	780

Comparison of the Thermal Conductivities of Laminar Composite Structures and Cermets.

FUELS

Comparison of Surface Heat Flux Factors of Laminar Composites and Cermets, *Figure 1*

DISTRIBUTION OF RADIOACTIVE
MATERIAL INSIDE CAPSULE

Fig. 3

ARROWS INDICATE DIRECTION OF HEAT FLOW.

ORNL-DWG. 67-268

T_0 : TEMPERATURE AT THE FUEL-CAPSULE INTERFACE (MAXIMUM AT INDICATED POINT)

T_1 : TEMPERATURE AT THE INNER SURFACE OF THE FUEL AT EQUILIBRIUM

$T_0 (A) > T_0 (B) > T_0 (C)$; $T_1 (A) > T_1 (B) > T_1 (C)$

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Fig. 27 Qualitative Representation of Fuel Distribution in Three Models.