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PRELIMINARY DESIGN AND SAFETY
STUDIES OF LARGE RADIOISOTOPE
HEAT SOURCES FOR SPACE POWER (U)

Mid-Term Technical Progress Report

APRIL 15, 1965 through JULY 31, 1965

CONTRACT NO. AT(30-1)-3526

Prepared for:

UNITED STATES ATOMIC ENERGY COMMISSION
NEW YORK OPERATIONS OFFICE

376 Hudson Street
New York, New York 10014

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1. INTRODUCTION AND SUMMARY

1.1 INTRODUCTION

The mid term technical progress report being issued for the "Preliminary Design and Safety Studies of Large Radioisotope Heat Sources for Space Power Supplies", Contract AT(30-1)-3526, is presented in this document. The report covers the period from the initiation of the contract April 15, 1965, through July 31, 1965.

The "Large Radioisotope Heat Source" study is a six-month program to perform comprehensive preliminary design and safety studies of flexible heat source subsystems by considering various isotopes and their compounds, in conjunction with thermoelectric, Rankine and Brayton power conversion subsystems which would lead to safe and reliable power supplies for manned and unmanned missions requiring 1-10 KWe.

This study will develop and analyze overall heat source subsystems for the Pu-238, Po-210, and Cm-244 fuels in conjunction with power system-heat exchanger combinations. Consideration will be given to general safety criteria, as well as specific design and operational requirements imposed by power conversion systems and missions. Heat source trade-offs will be identified, and compromises indicated to permit flexibility of application to different fuels, power systems, and vehicles. Performance, weight, and efficiency data will be summarized for heat source configurations to permit assessment of relative advantages and disadvantages of the fuels in combination with other variables.

A fuel capsule preliminary design will be developed for specific heat source concepts and for each fuel compound. At a minimum the following fuel compounds will be considered:

Po-210 - As a compound with gadolinium or dysprosium

Pu-238 - Oxide, nitride and carbide

Cm-244 - Solid solution Cm_2O_3 - ThO_2

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The designs will seek to achieve maximum interchangeability, with appropriate consideration given to required fabrication and inspection methods, weight penalties, and compatibility of materials over the full range of temperature and stresses determined under the heat source and capsule analysis efforts. The effect of a helium release approach will be investigated and discussed. In addition, liaison will be maintained with government laboratories and other contractors to obtain data on fuel form properties, fuel availability, and encapsulation requirements.

Although this study is not primarily concerned with heat source subsystems, the interface between heat source and power conversion system, shielding, and fuel handling systems will influence optimization of heat source designs. The heat source subsystems will be designed so as to be adaptable with the following power conversion subsystems and their nominal operating temperatures:

- SiGe Thermoelectric, Hot Junction Temperature = 1500°F
- PbTe Thermoelectric, Hot Junction Temperature = 1100°F
- Argon Brayton Cycle, Turbine Inlet Temperature = 1550°F
- Mercury Rankine Cycle, Turbine Inlet Temperature = 1250°F
- Dowtherm Rankine Cycle, Turbine Inlet Temperature = 650°F

Parametric data and specifications will be prepared describing the various power conversion systems in terms of power level, radiator areas, temperatures, efficiency, etc., to permit optimization of heat source-heat exchanger weights and dimensions.

Preliminary shielding analyses will be performed to establish interactions between heat source design, fuel selection, vehicle configuration, etc. The results of these studies (presented parametrically) will provide guidance for the heat source design task. The missions considered will be non-specific in nature, but fall within general mission classifications.

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Pre-launch and in-space fuel handling operations for typical vehicle and heat source designs will be investigated to determine the influence upon heat source and shield design. Particular emphasis will be placed on the interface between handling tools and the fuel block and/or capsules. Fueling and recovery techniques will be described to the extent necessary to establish feasibility of heat source concepts and their compatibility with typical spacecraft applications.

The safety studies will establish fuel capsule and heat source safety design criteria. All safety considerations will be based on the following philosophy:

"Fuel capsule - fuel block combinations should be designed to provide total containment of the radioisotope under all conceivable conditions of ground handling, launch, preorbital conditions, orbital operation, re-entry and post re-entry"

Preliminary analyses of fuel criticality problems and re-entry heating will be carried out. Design requirements will be established for fuel containment, permissible source size, environmental temperatures, re-entry modes, etc.

In addition potential accidents will be identified for ground handling, launch, re-entry, and recovery phases of operation with large radioisotopic sources. Launch pad fires, hard impact, launch abort, criticality, random re-entry, sea water submergence, and soft earth burial will be analyzed for typical mission profiles. Relative hazards associated with fuels, fuel loadings, and representative mission applications will be discussed.

The study will ultimately evaluate the feasibility and practicality regarding the safe utilization of large radioisotopic heat sources, and the significant aspects of these feasibility questions will be identified within a recommended development plan. This development plan (as a result of this study) will define areas requiring further research and development which should properly precede a definitive design effort on the various isotope heat sources.

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1.2 SUMMARY

During the first quarter, work was initiated in all major technical areas. Initial design iterations have been made, and "reference" design concepts evolved. The most significant design requirements are:

- Preclude criticality under all conditions
- Emergency cooling provisions
- Re-entry survival

The above items must be completely incorporated to insure that the designs will be acceptable from the safety viewpoint.

The current status can be summarized as follows:

- Safety and Hazards (Section 2)

General safety criteria are established. A criticality problem exists for Pu-238, and very likely for Cm-244. The approach to preclude criticality utilizes poisons or diluents and considers the most pessimistic shape (spherical) in which a source can be deformed during accident conditions. Preliminary analyses indicate that criticality in the thermal region (for Pu-238) can be prevented by the addition of less than 20 percent (by volume) of suitable thermal neutron absorber. The prevention of fast fission criticality (of Pu-238) requires greater amounts of poisons or diluent. Calculations indicate that GdO_2 in a 1 for 1 (by volume ratio) will preclude criticality for a 50 KW_t compacted spherical geometry. Suitable diluents or poisons appear to preclude nuclear criticality under all conditions for PU-238.

Preliminary re-entry analyses and materials evaluations indicate feasibility of protection from random earth orbit re-entry by use of materials such as the

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foamed metals with appropriate filler materials which provide evaporative cooling. Complete protection of individual capsules can be accommodated, although protection of groups of capsules affords a significant weight saving. Graplute was originally considered for this application, but was found to be unacceptable due to excessive capsule temperatures following an orbital decay type of heat pulse. Controlled re-entry with recovery appears to be the only solution to prevent burial in the soil. Demonstrated recovery systems capable of providing this type of control are available.

- Systems Integration and Analysis (Section 3)

Potential applications and missions have been reviewed to provide design requirements. The manned missions are dominant, and the design requirements thus established are more demanding.

A strictly "all passive" emergency cooling system is not feasible. Accommodation of emergency cooling is however possible by taking into account redundancy in normal coolant loops, backed with either manual, or mechanical, ejection of exterior surface thermal insulation, or movement of the heat source out from the normal operating cavity.

The higher efficiencies associated with high power cycle temperatures, along with emergency operating conditions, dictate a high temperature capsule design. Operation at reduced temperatures permits consideration of additional fuel within the same volume, by allowing larger temperature drop from capsule to heat exchanger.

Power cycle studies indicate that a 25 KW_t heat source is the logical lower limit design point for the long half-life fuels. A common heat source (of 25 KW_t) capable of being integrated with either SiGe thermoelectrics, the Hg Rankine cycle, or the Brayton cycle will provide a power output range of $1.35 \text{ KW}_e - 4.75 \text{ KW}_e$.

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Fuel handling studies have been undertaken to identify major problem areas and possible avenues of solution.

- Concepts and Design Analysis (Section 4)

Design studies indicated feasibility of cylindrical heat sources. Concepts for Pu-238 (vented and non-vented capsules) were developed for a 25 KW_t fuel loading. A non-vented Po-210 heat source was also developed for an initial fuel loading of 56.25 KW_t (25 KW_t at end-of-life). The designs appear capable of precluding criticality as well as providing re-entry and emergency cooling protection and being adaptable to all power conversion systems. The vented capsule approach affords a significant weight saving for Pu-238 systems.

Detailed capsule design analyses were carried out to assess structural requirements, particularly for helium containment. Helium containment for Po-210 sources appears feasible with use of the superalloys, whereas refractories are necessary for non-vented Pu-238 capsules.

- Materials Evaluation and Selection (Section 5)

- • Nuclear (Section 5.1)

Po-210, Pu-238 and Cm-244, and their compounds were reviewed, and liaison carried out with Mound Laboratory and Oak Ridge National Laboratories. Due to past emphasis on the element plutonium, the majority of information was available for this element (although specific data for Pu-238 and its compounds is still meager). Information on Cm-244 is of a preliminary nature, with the polonium technology being still in the early developmental stage.

Basic properties and behavioral data such as thermal conductivity, thermal expansion, solubility and melting temperature; compatibility of nuclides and

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their compounds with encapsulating materials; chemical and dimensional stability; and effects of radiation in accelerating capsule corrosion in various media are technology areas where more work must be done.

Available fuels data is summarized in Section 5.1; a detailed topical report was issued June 18, 1965.

Another item worthy of mention is the inclusion of the first known theoretical calculation and prediction of the cross-sections of Cm-244 by A. Prince of General Electric Co. - Nuclear Materials and Propulsion Operations by means of nuclear systematics. The details are contained in Appendix A.

- • Non-Nuclear (Section 5.2)

A literature review for data was undertaken to evaluate the chemical compatibility of potential materials for heat source designs, and to assure system materials compatibility. The design under consideration contains material interfaces expected to operate at or about 1800⁰ F for periods of 5 years or more. Appendices B and C contain the detailed results of beryllium and graphite material and compatibility searches.

Although cladding materials of principal interest in present designs are TZM, Haynes 25 and Hastelloy X, data was also sought for the major constituents of these alloys and for other similar alloys.

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2. SAFETY

The utilization of radioisotopic fuels for space power necessitates consideration of safety in all handling and operational facets from isotope encapsulation through mission completion. The large quantities of fuel required by this application demands that special emphasis be placed on heat source designs to insure radioisotope containment. Safety criteria and hazards evaluations dealing with potential accidents are presented in the following sections, with particular emphasis being placed on those situations which can lead to volatilization and widespread dispersion of the fuels.

2.1 SAFETY CRITERIA

The following general safety criteria will provide the framework for design requirements.

- Procedures which will be used in the manufacture, storage, and transportation of the fuel capsule, or heat source prior to the actual mission shall conform to applicable AEC and government regulations.
- The radioactive fuel capsule and heat source assemblies shall be designed to provide for complete containment through all normal mission operations and accident situations, except under conditions where fuel dispersal does not subject the populace to undue hazards.

The first of these criteria represents normal practice for nuclear equipment, and there are well established regulations and practices for shipping and handling large radioactive sources. These criteria are expanded in Table 2-1.

The second criterion, that of complete containment, provides for the primary safety control of hazardous material, and in effect couples design requirements to hazards categories.

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TABLE 2-1. GROUND HANDLING CRITERIA

- Dose rates at a surface shall not exceed 200 mrem/hr or 10 mrem/hr at 1 meter.
- Dose rates in uncontrolled areas shall not exceed 2 mrem/hr. This implies an appropriate controlled exclusion area around the shipping configuration.
- The surface of the shipping configuration accessible to handling personnel shall not exceed 180°F.
- The surface of the cask container which may contact any surface (floor or wall) shall not exceed 350°F.
- The shipping configuration shall maintain substantial shielding integrity and prevent fuel capsule clad failure in the event of an impact corresponding to a thirty foot drop on a non-yielding flat surface and a three and a half foot drop on a six-inch diameter mandrel, all followed by the standard fire.
- The shipping configuration shall maintain substantial structural integrity and prevent fuel capsule clad failure while submerged in a standard 1 hour fire.
- For planned operations prior to launch, personnel doses shall not be in excess of 1-1/4 rem/quarter.

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These hazard categories (in order of decreasing magnitude) can be defined as follows:

- I. Substantial release of small fuel particles (less than 3 microns) into the earth's atmosphere or earth's surface at low altitudes (unavoidable inhalation).
- II. Release of larger particles (approximately 100 microns) at high altitudes causing widespread surface contamination.
- III. Immersion of exposed fuel in shallow sea water.
- IV. Localized contamination (i.e., launch pad, or along flight trajectory).
- V. Local hazard due to external gamma and neutron radiation effects, fire hazard, localized containment, impact, etc.

These hazard categories are used to assess design requirements in Section 4.

The vaporization of fuel presents the largest potential hazard, and any operational mode or situation leading to this situation must be prevented. As a result the following key items can be immediately identified as being primary design considerations:

- Criticality
- Re-entry (random)
- Emergency cooling
- Burial

The hazards potential afforded by a nuclear excursion (and subsequent fuel dispersion) is so high that this situation must be prevented under any situation. Immersion of a heat source, or any of its components in any moderating materials (i.e., chemical fuel, water, liquid hydrogen, etc) must not result in a critical configuration. In addition physical deformations under any conditions must not result in an excursion; nor may a combination of these two

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effects be allowed to produce a critical assembly. The details of criticality analyses and hazards are covered in Section 2.2.1.

Re-entry heating, loss of coolant, and burial can result in excessive temperatures. Release of fuel during re-entry necessitates proper re-entry thermal protection (particularly for random re-entry), while emergency cooling must be provided when the normal cooling loop is inoperative. Either burial or an undue hazard resulting from burial must be prevented.

2.2 HAZARDS

The isotopes being considered for this application emit radiation which can be hazardous in the event of accidents or inadequate handling procedures. The energy of the different nuclear radiation per disintegration for each nuclear radioisotope considered is:

Average Available Energy per Disintegration (Mev)

	Alpha	Gamma	Fission Neutrons (α)	Neutrons*
Po-210	5.30	1.0×10^{-5}	0	1.8×10^{-7}
Pu-238	5.48	1.2×10^{-5}	5.5×10^{-9}	2.3×10^{-7}
Cm-244	5.79	2.5×10^{-5}	3.9×10^{-6}	4.3×10^{-7}

where the predominance of the alpha radiation is evident. However, during all stages of normal use, except that of individual fuel element assembly, the alpha energy is entirely absorbed within the fuel itself and its encapsulating materials. Thus, when encapsulated, the radiation hazard is associated with the much smaller amounts of gamma and neutron radiation. Also, since the disintegration energies are approximately equal, the number of curies per heat source of a given rating will be to a rough approximation independent of fuel.

In addition it is necessary to evaluate those properties of each fuel which give rise to biological hazards. These can be summarized as follows:

* Calculated assuming an oxygen compound which is realistic for Pu and Cm but not for Po.

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Biological Parameters of Fuel Forms

Fuel	Po-210	Pu-238	Cm-244
$MPC_w - \mu c/cc$	7×10^{-6}	5×10^{-6}	7×10^{-5}
$MPC_a - \mu c/cc$	2×10^{-10}	7×10^{-13}	3×10^{-12}
Maximum permissible body burden (critical organ) - μc	.03 (spleen)	.04 (bone)	0.1 (bone)
Maximum permissible short time inhalation - μc	.21	.17	.47
Maximum permissible short time ingestion - μc	1.0	1.4×10^3	1.2×10^3

There is no significant difference in permissible body burdens, even though the critical organ for Po-210 is the spleen, and the bones for Pu-238 and Cm-244. The last two rows give maximum permissible short time inhalation and ingestion limits, considering the different solubilities of the materials in the body. On this basis, Po-210 would appear to be more hazardous, but allowance should be made for its relatively short half life, which would mean that the maximum permissible body burden would exist for only a short time.

To fully complete this comparison it is necessary to evaluate fuel form characteristics (such as solubility, vapor pressure, stability, etc) along with accident environments and the associated complex interactions of the heat source and its components. Detailed, comprehensive analyses will be deferred until the next reporting period. Criticality and re-entry are covered in the following sections.

2.2.1 CRITICALITY

Criticality can result in widespread fuel vaporization and release, and the design must prevent all possibilities of this occurring. Until very recently, the possibility of a critical accident involving the even-even nuclides 94 Pu-238 and 96 Cm-244 was of no practical

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significance because only small quantities of the materials were available. The SNAP isotopic power programs project the use of kilogram quantities of both materials. Work by Hanford (Hw-84369) based on the experimental fast neutron cross-section data of Butler and Sjoblom (ANL) for Pu-238, gave an unreflected critical spherical mass of around 5 kg, about half that for the Pu-239 nuclide. Stubbins et al confirmed the fissionability of Pu-238 by critical experiment using the Jezebel critical assembly. The computed critical mass, however, was computed to be roughly equal to that of Pu-239. With the results of the Pu-238 studies in view, ORNL has arranged for critical mass studies of Cm-244 at Los Alamos and at the ORNL critical facilities, but no results have been reported to date.

The hazards associated with an excursion phenomena are widespread. For example, a critical excursion of a 25 KWt source containing 7.1×10^4 gms of Pu-238 O_2 , assuming a resulting energy release of 100 MW seconds, will result in a vaporization and release to the atmosphere of about 12 percent of the fuel. This release, amounting to about 10^5 curies would be the source of widespread inhalation and considerable ground contamination.

Calculations on the extent of hazard for this magnitude of release have been made assuming inversion type weather conditions with:

Wind Velocity, $u = 3$ meters/sec

Sutton's parameters $n = 0.333$

$C_y = 0.2$

$C_z = 0.05$

Effective height, $h = 100$ meters of release

Deposition coefficient $V_{d/u} = 3.4 \times 10^{-4}$

The resulting body burdens along the center line of the cloud are shown in Figure 2-1. The maximum permissible body burden of $0.04 \mu c$ would be exceeded out to distances of 335 miles. The total area involved at or above this level could be about 2300 square miles. This results in ground contamination (Figure 2-2) amounting to more than the AEC limit for unconditional release and could extend to distances of 7800 miles covering an area of 720,000 square miles.

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Figure 2-1. Body Contamination From 100 Mw-Sec Pu^{238} Excursion

Figure 2-2. Ground Contamination From 100 Mw-Sec Pu^{238} Excursion

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The distances over which these body burdens and ground contamination levels are calculated to exist are extremely large and it is most unlikely that the atmospheric conditions assumed would last for the time necessary for the emitted cloud to move that far. However, the calculated areas are probably more nearly correct and give a reasonable estimate of the extent of contamination possible. The necessity of precluding criticality under any condition is evident.

Because of the very large possible hazard, it was decided that a critical excursion involving Pu-238 and Cm-244 was not permissible under any condition and a method of positively preventing a critical accident was sought.

Preliminary criticality studies were carried out on a nominal 50 Kw(th) Pu-238 heat source. To evaluate the worst cases for all the condition, an homogenized spherical configuration was used. The four cases computed and the assumptions involved are given in Tables 2-2 and 2-3. The cross-sections data for Pu-238 were obtained from BNL for thermal and fast neutrons and in the resonance region, cross-section data for Pu-240 were used (normalized to the Pu-238 levels at 0.1 Mev. The cross-section data is shown in Figure 2-3. Cross-sections data for the other Pu isotopes and for the non-fissile, clad, coolant, and structural materials were standard values taken from BNL-325. Table 2-3 summarizes the calculated reactivities.

The fourth case presented included approximately 100 kg of Gd_2O_3 intimately mixed with the PuO_2 , which then forces the reactivity of the homogenized spherical source below, $k = .75$. Since a homogenized sphere represents an extreme deformation, or accidental geometry case, the addition of this poison and/or diluent has freed the heat source from both geometric and accidental criticality constraints.

The assessment of the criticality of Cm-244 was deferred until later due to the lack of cross-section data. Analytical techniques were employed to predict the cross-sections and are presented in detail in Appendix A. Calculations using this information will be made later, as well as reviewing the amount of poison needed in several reference designs.

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TABLE 2-2. ASSUMPTIONS FOR CRITICALITY STUDIES

FUEL INVENTORY	50 KILOWATTS THERMAL (126 Kg)	
FUEL ISOTOPIC CONTENT, %	Pu-238	80.0
	Pu-239	16.5
	Pu-240	2.5
	Pu-241	0.8
	Pu-242	0.08
FUEL COMPOUND	PuO ₂	
FUEL DENSITY, g/cc	10.25 (90% OF THEORETICAL 11.46)	
$\bar{v}$ Pu-238	2.33	
CROSS-SECTIONS		
THERMAL FISSION	16.9 BARNS	
FAST FISSION	2.6 BARNS	

2.2.2 RE-ENTRY

In normal operation, the isotopic heat source will not be exposed to unprotected re-entry conditions; with the heat source assembly being placed in either a manned or un-manned vehicle for launching and recovery. However, the possibility exists that the heat source could experience random re-entry resulting from an accident involving break-up of the launch vehicle. Therefore, back up heat protection must be provided for the heat source and/or fuel capsules.

Re-entry analyses were commenced with aerothermodynamic effects of earth orbital re-entry being analyzed by means of a GE-MSD digital computer code (the "Round Earth-Point Mass Code"), which calculates instantaneous heating rates based on a computed trajectory and employs body drag subroutines dependent on density and Mach number relationships coupled to a reference area. An instantaneous stagnation heating rate (based on a reference sphere of one foot diameter) is computed according to the relationship

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TABLE 2-3. REACTIVITY OF HEAT SOURCES*

CASE NUMBER	MATERIAL	VOLUME, LITERS	WEIGHT, Kg	REACTIVITY
1	PuO ₂	12.5	126	1.182
	VOID	NONE	-	
2	PuO ₂	12.5	126	0.60
	GRAPHITE	63.7	121	
	HAYNES-25	28.3	252	
	STRUCTURE	5.8	13	
	SODIUM	10.8	11	
	VOID	62	-	
3	PuO ₂	12.5	126	0.946
	HAYNES-25	10.9	98	
	VOID	NONE	-	
4	PuO ₂	12.5	126	0.75
	HAYNES-25	10.9	98	
	Gd ₂ O ₃	15	110	
	VOID	NONE	-	

* 50 KW_t Pu-238 O₂

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$$\dot{q}_{\text{stagnation}} = \frac{17,600}{\sqrt{R_N}} \left[\frac{\rho_{\infty}}{\rho_s} \right]^{1/2} \left[\frac{V_{\infty}}{V_c} \right]^3 \quad (\text{Eq. 2-1})$$

where

- R_N = Reference nose radius
- ρ_{∞} = Free stream density
- V_{∞} = Free stream velocity
- ρ_s = Sea level density
- V_c = Orbital velocity at re-entry

Figure 2-4 illustrates typical time dependent computed heating rates as a function of $W/C_D A$ (the body-weight-to-drag-area product parameter). A re-entry velocity of 25,600 ft/sec, re-entry altitude of 350,000 feet, and path angle of 90° (essentially an orbital decay type of re-entry) were employed for this portion of the study.

Since the majority of heat source configurations considered to date were of a cylindrical geometry, the computed spherical stagnation heating rates were modified by the multipliers shown in Table 2-4 to assess various body re-entry modes. For the case of a cylinder in the "bombing" mode, the heating rate is also a function of cylinder radius (a factor of 1.7 on the cylinder centerline stagnation heating rate exists at the cylindrical sharp corner), and a multiplier of 1.14 (a commonly accepted integrated heating rate factor) was employed to calculate material thickness required at the cylindrical ends. Drag coefficients based on cylinder lateral area (L.D) were computed based on the following relationships:

Case	Free Molecular	Laminar Continuum
Side on	2.0	0.667 (2-K)
End on	1.57 D/L	0.714 D/L (2-K)
End-over-end tumbling	1.273 + D/L	(0.283 + 0.303 D/L)(2-K)
Random Tumbling	1.57 + .785 D/L	(0.393 + 0.198 D/L)(2-K)

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TABLE 2-4. GEOMETRIC "TRANSFORMATION" MULTIPLIERS

<u>To Obtain</u>	<u>Multiply Spherical Stagnation Heating Rate By</u>
$\dot{q}_{\text{hemisphere}}$	$\sqrt{\frac{24}{D_{\text{Reference}}}}$
$\dot{q}_{\text{stagnation}}$ line (2 Dimensional) Flow	$\frac{1}{2} \sqrt{\frac{24}{D_{\text{Ref}}}}$
$\dot{q}_{\text{stagnation}}$ (Bombing Mode)	$\frac{1}{\sqrt{3}} \sqrt{\frac{24}{D_{\text{Ref}}}}$
$\dot{q}_{\text{tumbling cylinder}}$	$\frac{.554}{\sqrt{2}} \sqrt{\frac{24}{D_{\text{Ref}}}}$
$\dot{q}_{\text{tumbling andspinning cylinder}}$	$\frac{1}{\pi} \frac{.554}{\sqrt{2}} \sqrt{\frac{24}{D_{\text{Ref}}}}$

Figure 2-4. Calculated Heating Rates (Reference Sphere 1' Dia.)

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C_D is based on $A_{RD} = LD$

A_R = Reference area (ft^2)

D = Cylinder diameter (Ft)

L = Cylinder length (ft)

K = Density ratio across normal shock (ρ_∞ / ρ_2)

Early heat source concepts considered graphite for the heat protection shield. Subsequent analyses of random re-entry from an orbital decay mode revealed that the graphite functioned as a heat sink rather than as an ablator. A typical re-entry temperature-time history is shown in Figure 2-5 for a heat source utilizing graphite. Since graphite reaction temperatures are normally in the 4000-4500^oF range (depending on back pressure effects), the melting temperatures of superalloy clad materials were exceeded.

As a result alternate heat shield materials were investigated which would limit capsule temperature to 2200-2300^oF, during the re-entry period. Foamed metal systems impregnated with suitable additives offer a potential answer. Pertinent properties are shown in Table 2-5. Initial evaluations were oriented at individual fuel capsule protection, and aluminum trifluoride appeared a likely candidate. Figure 2-6 shows a typical temperature-time history curve for a situation where an inadequate thickness of heat shield was employed.

The heating rates employed to calculate average heat shield thicknesses were based on the tumbling mode (cylinders with an L/D approaching unity will very likely tumble), and the heat shield material used in the evaluation was a porous nickel impregnated with aluminum trifluoride (90% AlF_3 + Ni by volume). The AlF_3 vapor pressure-temperature characteristics, indicate a potential sublimator with a heat of vaporization of 1632 BTU/Lb and a subliming temperature of 2200-2300^oF. This heat shield absorption capability, and a body re-radiation at 2200^oF with an effective emissivity of 0.7, resulted in the heat shield weights and thicknesses shown in Table 2-6. Heat shielding-to-total heat source weight fractions of 18-25 percent resulted, with the specific body L/D ratios significantly affecting the drag coefficients.

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HEAT SOURCE CONFIGURATION

Figure 2-5. Heat Source Temperature with Graphite Heat Protection

Figure 2-6. Heat Source Temperature with Foamed Metal Ablative Heat Protection AlF_3 and Ni

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TABLE 2-5. CANDIDATE ABLATIVE MATERIALS

	MELTING POINT (°F)	HEAT OF FUSSION (Btu/Lb.)	BOILING POINT (°F)	HEAT OF VAPORIZATION (Btu/Lb.)	DENSITY gm/cm ³
Lithium					
LiF	1556	164	3057	3540	2.30
Li ₂ O	3150				2.00
* LiN	decomposes at about 1832° F			2530	
Beryllium					
BeF ₂	1466	229	2430	1530	2.01
BeO	4622				3.00
BeN	3980				
Boron					
BF ₃			-150		
B ₂ O ₃	842	142	3680	1810	1.85
BN	sublimes		5430		
Sodium					
NaF	1818	334	3010	1290	2.80
Na ₂ O	1684		4040		2.30
Magnesium					
MgF ₂	2485				2.79
MgO	5252				3.65
Aluminum					
AlF ₃	sublimes		2320	1650	2.88

* Liberated Li oxidizes with high exothermic heat reaction

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TABLE 2-6. REQUIRED HEAT PROTECTION FOR HEAT SOURCE CONFIGURATION

Drawing	Weight (Less re-entry Material) (Lbs.)	L _{Bare} (In.)	D _{Bare} (In.)	Thickness of Ablator (Inches)	Weight of Heat Shield Material (Lbs)	Total Weight (Lbs.)
LHS-010 ⁽¹⁾	454	29	14.5	0.75	146	600
LHS-005 ⁽²⁾	724	20	15	1.01	166	890
LHS-009 ⁽³⁾	235	12	13	0.76	67	299
LHS- (4)	404	12	10	1.26	127	531

- (1) Non-vented Pu-238 capsule, 25 KW_t Sodium filled module
- (2) Non-vented Pu-238 capsules, 25 KW_t, individual capsule loading
- (3) Vented Pu-238 capsules, 25 KW_t, Sodium filled module
- (4) Vented Pu-238 capsules, 25 KW_t, individual capsule loading

Protection of the individual capsules for the complete duration of re-entry resulted in heat shield weights (based on an AlF₃ impregnated metal foam) equal to, or even twice the capsule weight depending on the W/C_D characteristics. As a result the concepts evolved were based on a heat protection material wrapped around the total heat source volume.

Additional work is being carried on to assess more thoroughly potential heat shield materials as well as different re-entry trajectories. If heats of vaporization on the order of 2500 BTU/Lb could be achieved, individual capsule protection might be feasible.

2.2.3 BURIAL

Soft earth, or sand burial of fuel capsules following an abort can lead to high temperatures, and in some cases the melting of clad and/or fuel. A still more severe case is the burial of entire or partial pieces of heat sources. Since the evaluation of these accidents is de-

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pendent not only on accidents postulated, but analyses methods and assumptions, the conclusions reached are subject to question. In general, calculations based on final steady state temperatures will result in some melting, with the only positive means of preventing this situation being controlled re-entry.

A poor conducting medium (such as dry sand) could lead to melting of the surface clad material (if the superalloys are employed) with the material melting and then re-solidifying to provide a more favorable heat transfer path. Continued temperature rises could occur until reaction between the environmental media and capsule material or fuel itself occurred. Table 2-7 contains typical melting points of fuel compounds and other materials. It is evident that lower temperature (below fuel compound melting) reactions could take place to slow down the temperature excursion, as well as stabilize the reaction at a lower temperature.

A double walled capsule design employing a refractory pressure shell (to contain the helium) as well as an external oxidation resistant material, affords significant impact and burial protection. Accidents leading to burial are less prone to capsule fracture, and vice-versa. A postulation of exposure of all, or of significant portions of the fuel on impact appears unlikely.

TABLE 2-7. MATERIAL MELTING TEMPERATURES

PuO_2	=	2300°C (4172°F)	Molybdenum	=	4750°F
PuN	=	2600°C (4712°F)	Niobium	=	4470°F
PuC	=	1650°C (3002°F)	Haynes-25	=	2350°F
Cm_2O_3	=	1950°C (3542°F)	Platinum	=	3220°F
CmO_2	=	600°C (1112°F)	Nickel	=	2650°F
GdPo	=	1675°C^* (3050°F)			
SiO_2	=	$1400\text{--}1600^{\circ}\text{C}$ ($2350\text{--}2900^{\circ}\text{F}$)			
(Sand)					

* This is a dissociation temperature for the polonide.

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In either event, the presence of a mixed fuel-soil material is of no particular hazard (assuming no volatilization of fuel) except as a radiation source (which would be partially shielded by the soil) or as a source of thermal energy (as to a bare foot) unless the resulting compound has an increased solubility and potable water has access to it. In general, flow of water through soils is relatively slow (often measured in feet per year) and in addition most soils possess a considerable exchange capacity for materials like Pu, and would not be expected to transmit much to actual water supplies. Counter measures including a search and possible recovery (search for radiation and heat emission); and monitoring of water and food supplies in general area where a heat source or capsules are presumed to have been lost are certainly possible and should be part of emergency plans in the event of an abort.

Selective burial conditions will be analyzed in the next reporting period, particularly those related to the Po-210 compounds. The need for supporting experiments and tests is self-evident even at this stage. Compound interactions and materials property and compatibilities are also required to verify these studies.

2.2.4 GENERAL HAZARDS

To facilitate general hazards evaluations from the launch pad through final operation, a series of "event" charts (see Figures 2-7 through 2-9) has been prepared. These "hazard trees" will be used to assess the overall safety and hazard situation.

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Figure 2-7. Event Chart I

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Figure 2-8. Event Chart II

Figure 2-9. Event Chart III

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3. SYSTEMS

A multipurpose radioisotopic heat source must not only adapt to various fuels, power conversion systems, and power output ratings, but also integrate readily with a variety of spacecraft and mission requirements. The following sections review the work performed on

- System & Mission Integration
- Shielding
- Power Conversion Subsystems
- Fuel Handling

3.1 SYSTEM AND MISSION REQUIREMENTS

A review of mission and system requirements was made to determine the influence of manned versus unmanned demands. Table 3-1 illustrates typical potential missions. The first requirement (that of power level) is condensed in Table 3-2, with the unmanned power requirements shown in Table 3-3. A composite isotope demand curve is shown in Figure 3-1, with the manned isotope demands clearly dominant. Early requirements call for utilization of a short half-life isotope, with eventual utilization of the longer half-life fuels. The above information is the result of liaison with NASA personnel.

Other vehicle and mission interfaces and requirements are summarized in an "applications" matrix shown in Figure 3-2. Manned systems are again dominant and present more stringent design requirements. A Saturn V launch environment is applicable to Pu-238 and Cm-210 heat sources, with the possibility of using a SIB-Centaur combination for early Po-210 missions. In addition redundancy is required for heat source cooling, as well as emergency cooling survival with the heat source re-useable after power plant shutdown and re-start.

The area of fuel recovery and integration with spacecraft systems was investigated.

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TABLE 3-1. POTENTIAL MISSIONS FOR LARGE
ISOTOPE POWER SYSTEMS

MANNED

- EXTENDED APOLLO OR EARLY ORBITAL SPACE STATION
- LARGE ORBITAL SPACE STATION
- LUNAR SHELTER MODULE
- LUNAR ROVING VEHICLE
- INTERPLANETARY TRAINING MODULE
- PLANETARY LANDER

UNMANNED

- TEST SATELLITES
- FM AND TV BROADCAST SATELLITE
- EARLY OUTER PLANET FLYBY PROBES
- VOYAGER

Figure 3-1. Estimated Isotope Demand

TABLE 3-2. SUMMARY OF MANNED APPLICATIONS OF ISOTOPES
PROJECTED MISSIONS

<u>Mission</u>	<u>Power (KW_e)</u>	<u>Duration</u>	<u>Flights/Yr.</u>	<u>Total Flights Through 1980</u>	<u>Notes</u>
<u>Extended Apollo</u>	1.5	30-90 days	6-8	30-40	Potential use in Earth Orbit, Lunar Orbit, Crew Training & Shuttle, etc.
<u>Lunar Exploration Shelter Module</u>					
14 day Shelter	2	14-28 days	2-3	6-9	Fixed Site Exploration
84 day Shelter	4	90 days	1-2	2-4	Fixed Site Exploration
2 year Shelter	5	2 years	1-2	4-6	Fixed Site Exploration and Scientific Support
<u>Roving Vehicles</u>					
14 day mission	3	14 days	2-3	6-9	14 day Lunar Exploration
14-140 day mission	3-15	14-140 days	1	4	Versatile Range of Lunar Traverse
<u>Space Stations</u>					
National Multi-purpose space station (NMSS)	7-10	2-5 years (90 day re-supply)	2-3	6-8	Large variety of missions, including near Earth Orbit, Polar Orbit, synchronous.
<u>Interplanetary Mission Module</u>					
Training & Checkout	10-20	90-120 days	3-5	20-30	Continuous operation with extremely high reliability required.
<u>Lander</u>	10-20	400-1000 days	As req'd	As req'd	To be deployed to planetary orbits or surfaces for unmanned operation after encounter.

TABLE 3-3. UNMANNED SCIENTIFIC MISSIONS (REF. OSSA PROSPECTUS 1964)

	1967	1968	1969	1970	1971	1972	1973	1974	1975	1976	1977	1978	1979	1980	
1. Adv. Meteorological Satellite	-	-	-	40	50	-	100	100	-	-	-	-	-	-	
2. Nimbus	50	50	250	250	250	250	250	250	-	250	250	250	250	-	
3. Synchronous Meteorological Satellite	-	-	-	-	500	500	-	500	500	-	-	500	500	-	
4. OGO	-	-	-	-	350/500	350/500	350/500	350/500	350/500	-	-	-	-	-	
5. SGM	-	-	-	500/750	500/750	500/750	500/750	500/750	-	-	-	-	-	-	
6. AOA0	-	-	-	-	-	-	-	-	750/1000	-	750/1000	-	-	-	
7. Radioastronomy Observatory	-	-	-	-	250/500	250/500	-	250/500	-	-	-	-	-	-	
8. Manned Orbiting Telescope	-	-	-	-	-	-	-	-	-	-	-	-	-	1000/1500	
9. Voyager	-	-	-	-	600	-	600	-	600	-	600	-	-	-	
10. Mercury Flyby	-	-	-	-	-	-	400	-	-	-	-	400	-	-	
11. Jupiter Flyby	-	-	-	-	-	1000/1200	-	1000/1200	-	-	1000/1200	-	-	-	
12. Comet Intercept	-	-	-	-	-	-	-	-	-	30	-	-	-	-	
13. Direct Broadcast (FM Radio)	-	-	-	-	2000/3000	2000/3000	-	-	-	-	-	-	-	-	
14. Surveyor	25	40	40	40	-	-	-	-	-	-	-	-	-	-	
<hr/>															GRAND TOTAL
TOTALS Electric (Watts)	75	90	290	840/1090	4650/6350	4850/6700	2850/3800	3150/4250	2600/3300	830/1280	3400/4050	1700/2150	1300/1750	1250/2000	-
Thermal* (KW _p)	1.5	1.8	5.8	16.8/21.8	93/127	97/134	57/76	63/85	52/66	17/26	68/81	34/43	26/35	25/40	588-744

* 5% conversion efficiency assumed

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APPLICATIONS

	EXTENDED APOLLO OR EARLY MANNED SPACE STATION (MOD. MOBILE ETC)	LARGE SPACE STATION	LUNAR SHELTER MODULE	LUNAR ROVING VEHICLES	INTERPLANETARY MISSION MODULE (TRAINING)	PLANETARY LANDER (DELIVERED BY MANNED VEHICLE)	NIMBUS BROADCAST TEST SATELLITE	FM AND TV BROADCAST SATELLITE	FIRST OUTER PLANET FLY BY PROBE	VOYAGER	MANNED			UNMANNED		
	PB 210, PU 23B, CM 244	PU 23B, CM 244	PU 23B, CM 244	PU 23B, CM 244	PU 23B, CM 244	PU 23B, CM 244	PB 210, PU 23B, CM 244	PU 23B, CM 244	PU 23B, CM 244	PU 23B, CM 244	PB 210	PU 23B	CM 244	PB 210	PU 23B	CM 244
1 CANDIDATE ISOTOPIES																
2 LAUNCH ENVIRONMENT	TITAN 3C	SATURN IB	SATURN IB	SATURN IB	SATURN IB	SATURN IB	TITAN 3C	TITAN 3C	SATURN IB CENTAUR	SATURN IB CENTAUR	SATURN IB CENTAUR	SATURN IB CENTAUR	SATURN IB CENTAUR	SATURN IB CENTAUR	SATURN IB	SATURN IB
2.1 FIRE AND EXPLOSION																
2.2 TERMINAL VIBRATION																
2.3 SHOCK AND VIBRATION																
2.4 RANDOM RE ENTRY																
3 MISSION SHIELDING (1)																
3.1 γ DOSE / YEAR	75 REM / YR	25 REM / YR	25 REM / YR	150 REM / YR	75 REM / YR	12.5 REM / YR	3 x 10 ² mR / YR	3 x 10 ² mR / YR	10 ² mR / YR	3 x 10 ² mR / YR	3 x 10 ² mR / YR	3 x 10 ² mR / YR	3 x 10 ² mR / YR	3 x 10 ² mR / YR	3 x 10 ² mR / YR	3 x 10 ² mR / YR
3.2 γ DOSE / YEAR							3 x 10 ² RAD / YR	3 x 10 ² RAD / YR	10 ² RAD / YR	3 x 10 ² RAD / YR	3 x 10 ² RAD / YR	3 x 10 ² RAD / YR	3 x 10 ² RAD / YR	3 x 10 ² RAD / YR	3 x 10 ² RAD / YR	3 x 10 ² RAD / YR
3.3 SEPARATION DISTANCE	6 TO 40 FEET	20 TO 100 FEET	20 TO 1000 FEET	4 TO 20 FEET	20 TO 100 FEET	40 TO 300 FEET	4 TO 40 FEET	4 TO 40 FEET	20 TO 100 FEET	3 TO 10 FEET	3 TO 10 FEET	3 TO 10 FEET	3 TO 10 FEET	3 TO 10 FEET	3 TO 10 FEET	3 TO 10 FEET
3.4 SHIELD SOLID ANGLE	0.11 TO 2.11	0.0511 TO 1.11	0.0511 TO 1.11	0.11 TO 2.11	0.0511 TO 1.11	0.0511 TO 1.11	0.11 TO 0.411	0.11 TO 0.411	0.0511 TO 0.411	0.11 TO 0.411	0.11 TO 0.411	0.11 TO 0.411	0.11 TO 0.411	0.11 TO 0.411	0.11 TO 0.411	0.11 TO 0.411
3.5 GROUND SCATTER	NO	NO	YES	YES	NO	YES	NO	NO	NO	YES	NO	YES	YES	NO	YES	YES
3.6 EQUIV DOSE AT 1 METER																
3.6.1 γ DOSE / YEAR	250 TO 1000 REM / YR	750 TO 2500 REM / YR	750 TO 2500 REM / YR	250 TO 1000 REM / YR	250 TO 1000 REM / YR	250 TO 1000 REM / YR	4.5 x 10 ² TO 4.5 x 10 ² mR / YR	4.5 x 10 ² TO 4.5 x 10 ² mR / YR	4.5 x 10 ² TO 4.5 x 10 ² mR / YR	4.5 x 10 ² TO 4.5 x 10 ² mR / YR	4.5 x 10 ² TO 4.5 x 10 ² mR / YR	4.5 x 10 ² TO 4.5 x 10 ² mR / YR	4.5 x 10 ² TO 4.5 x 10 ² mR / YR	4.5 x 10 ² TO 4.5 x 10 ² mR / YR	4.5 x 10 ² TO 4.5 x 10 ² mR / YR	4.5 x 10 ² TO 4.5 x 10 ² mR / YR
3.6.2 γ DOSE / YEAR							4.5 x 10 ² TO 4.5 x 10 ² RAD / YR	4.5 x 10 ² TO 4.5 x 10 ² RAD / YR	4.5 x 10 ² TO 4.5 x 10 ² RAD / YR	4.5 x 10 ² TO 4.5 x 10 ² RAD / YR	4.5 x 10 ² TO 4.5 x 10 ² RAD / YR	4.5 x 10 ² TO 4.5 x 10 ² RAD / YR	4.5 x 10 ² TO 4.5 x 10 ² RAD / YR	4.5 x 10 ² TO 4.5 x 10 ² RAD / YR	4.5 x 10 ² TO 4.5 x 10 ² RAD / YR	4.5 x 10 ² TO 4.5 x 10 ² RAD / YR
4 PLANNED EARTH RE ENTRY AND RECOVERY	CONTROLLED MAN ASSISTED	CONTROLLED MAN ASSISTED	NOT REQUIRED	NOT REQUIRED	CONTROLLED MAN ASSISTED	NOT REQUIRED	NOT REQUIRED	NOT REQUIRED	NOT REQUIRED	NOT REQUIRED	NOT REQUIRED	CONTROLLED MAN ASSISTED	CONTROLLED MAN ASSISTED	NOT REQUIRED	NOT REQUIRED	NOT REQUIRED
5 ACCIDENTAL EARTH RE ENTRY FROM DEBIT	INTACT											INTACT (1)	INTACT (1)			
6 ACCIDENTAL PLANETARY ENTRY OR IMPACT	NOT APPLICABLE	NOT APPLICABLE	INTACT DESIRABLE	INTACT DESIRABLE	NOT APPLICABLE	INTACT DESIRABLE	NOT APPLICABLE	NOT APPLICABLE	INTACT DESIRABLE	INTACT DESIRABLE	DESIRABLE THAT FUEL SURVIVE ENTRY AND IMPACT			INTACT, HOWEVER, NOT A DESIGN REQUIREMENT		
7 CRITICALITY											NOT APPLICABLE	SUBCRITICALITY ASSURED BY FUEL CAPSULE		NOT APPLICABLE	SUBCRITICALITY ASSURED BY FUEL CAPSULE	
8 SEA WATER SUBMERGENCE																
8.1 SHALLOW RECOVERABLE	CONTAINMENT REQD 1 YR															
8.2 DEEP NON RECOVERABLE	CONTAINMENT DESIRABLE															
9 BUREL IN EARTH OR SAND																
10 OPERATION DURING LAUNCH																
11 COOLING																
12 HEAT SINK TEMPERATURE	26°F	26°F	140°F	140°F	26°F	100°F (MARS)	26°F	26°F	-250°F (JUPITER)	100°F	26°F	26°F	26°F TO 140°F	26°F	26°F TO 140°F	26°F
13 HEAT REJECTION SYSTEM MEAN TIME TO FAILURE	3 x 10 ⁵ HRS	1 x 10 ⁶ TO 5 x 10 ⁶ HRS	1 x 10 ⁶ TO 5 x 10 ⁶ HRS	1 x 10 ⁶ HRS	1 x 10 ⁶ HRS	5 x 10 ⁶ HRS	3 x 10 ⁶ HRS	3 x 10 ⁶ HRS	3 x 10 ⁶ HRS	3 x 10 ⁶ HRS	3 x 10 ⁶ HRS	3 x 10 ⁶ HRS	3 x 10 ⁶ TO 5 x 10 ⁶ HRS	3 x 10 ⁶ HRS	3 x 10 ⁶ TO 5 x 10 ⁶ HRS	3 x 10 ⁶ HRS
14 METEOROID FLUX	GEOCENTRIC															
15 RESTART CAPABILITY	REQUIRED						NOT REQUIRED							REQUIRED		NOT REQUIRED
16 NUMBER OF THERMAL CYCLES (NORMAL TO EMERGENCY)	6	50	50	50	100	50	5	5	5	5	6	100	100	5	5	5
17 DAMAGED CAPSULE HANDLING IN SPACE	REQUIRED						NOT REQUIRED							REQUIRED		NOT REQUIRED
18 COST OPTIMIZATION PARAMETERS																
18.1 INITIAL FUEL COST											200 \$/H	500 1000 \$/H	500 1000 \$/H	200 \$/H	500 1000 \$/H	500 1000 \$/H
18.2 RECOVERY VALUE OF FUEL											0	500 1000 \$/H	500 1000 \$/H	0	500 1000 \$/H	500 1000 \$/H
18.3 PROB OF FUEL RECOVERY											0	0.90	0.90	0	0.90	0.90
18.4 INTEREST CHARGE % / YR											0	6 % / YR	6 % / YR	0	6 % / YR	6 % / YR
18.5 MISSION DURATION, YR											0.25 YR	0.25 - 5 YR	0.25 - 5 YR	0.25	0.25 - 5 YR	0.25 - 5 YR
18.6 POWER PLANT HARDWARE COST \$ / LB											100 500 \$ / LB	1000 - 3000 \$ / LB	1000 - 3000 \$ / LB	100 500 \$ / LB	1000 - 3000 \$ / LB	1000 - 3000 \$ / LB
18.7 TRANSPORTATION COST \$ / LB																

NOTE 1 - MANNED DOSE BASED ON 25 REM / MISSION
UNMANNED DOSE 10¹² mR AND 10² RAD / YR

Figure 3-2. "Mission Applications" Matrix

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Figure 3-3. General Arrangement Command Module

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A parametric study for manned re-entry was carried out utilizing two capsule-heat source concepts - that of single capsules and integral capsule/heat source integration into a manned orbital re-entry vehicle of the Apollo configurations.

Figure 3-3 illustrates the general arrangement of the Apollo command module.

A reference 25 KW_t heat source was considered and effects were analyzed to assess integration problems. The following isotope capsule characteristics were considered:

- Thermal Characteristics

The effect of the thermal source on the vehicle environmental control system and the effect of the higher temperatures on the re-entry shield and other heat sensitive components such as parachutes, pyrotechnics etc, was considered

- Radiation Characteristics

Shielding of the source from personnel and radiation sensitive components was studied. Geometry of the Apollo vehicle was evaluated to determine approximate distances for calculating permissible dosages.

- Weight and Size

Consideration was given to the weight and geometry of the source for the effect on the vehicle attitude control during orbit, and stability during re-entry. The reference capsule, for purposes of this study, was a cylinder with a diameter of 2.5 inches, a length of 12.5 inches and a weight of 12 pounds. The modular heat source was a cylinder 14 inches in diameter, 13 inches in length, and weighing 300 pounds.

A time period for the manned re-entry mission was selected as twenty hours which was considered as the maximum period for the vehicle to attain a favorable de-orbit position

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from a nominal low earth orbit. This time period allows the vehicle thirteen orbits and would cover the entire earth.

Integration with the command module is predicated partially on available volume. Figure 3-4 illustrates the secondary structure schematically. A concept based on the ability to remove capsules from the heat source assembly is shown in Figure 3-5, with the capsules distributed evenly within the skirt region volume. A drawback to the individual capsule loading concept is the prolonged loading time, as well as extra-vehicular maneuvering required. This arrangement however is favorable to the vehicle since symmetry exists about the roll axis and the weight is forward of the normal command module C.G., thus avoiding aerodynamic unbalance problems during re-entry. Also, positioning the capsule equally about the roll axis would not grossly affect the attitude control system.

Minimum radiation shielding (app 3" of LiH) would be needed since the distance from the capsules to personnel averages 5 feet. Approximately 99 percent of the heat can be dissipated by fin radiators leaving only 1.1 KW entering the vehicle to affect components and the environment control system (Table 3-4).

Incorporation of a singular heat source module into an Apollo configuration poses significant integration problems. The only location on the vehicle where space could be made available is in the recovery equipment area, around the main chute area. However, incorporation of the weight of a 300-500 Lb heat source at aft C.G. would cause gross instability during re-entry resulting in tumbling and fishtailing. The preferable location is up forward, behind the heat shield, but this is impractical due to the location of the present personnel compartment.

Assuming that the personnel locations could be moved, it is then possible to locate the heat source in a location behind the heat shield (Figure 3-6). However, due to the concentrated heat load an active thermal control loop is required to dissipate the source heat. A typical radiator area required would be approximately 50 square feet and would dissipate 98 percent of the thermal energy, with only 0.6 KW entering the command module, raising local

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TABLE 3-4. INDIVIDUAL CAPSULE CONCEPT SUMMARY WEIGHTS

Beryllium Fins	
Area 40 ft ² , Thickness 0.18"	70 Lbs
Beryllium Capsule Holders	
Length 14", Thickness 0.25", Diameter 2.5"	40
Shielding	
4 cm LiH	275
"Min K" Insulation	
40 square feet, 3" thickness	100
Miscellaneous Hardware	30
Capsules	265
Total Weight	780 Lbs

Thermal

Beryllium Fin	
Fin Base Temp	850° F
Fin Efficiency	50%
Emissivity	0.8
Area	40 Ft ²
Heat Load into Spacecraft	0.35 KW

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Figure 3-4. Command Module Secondary Structure

Figure 3-5. Cell Concept

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temperatures and taxing the environmental control subsystem. Pump power for the active radiator loop can be supplied from thermoelectric elements integral with the module receptacle. Table 3-5 describes the active radiator loop parameters.

Because of the required high heat rejection temperature Apollo heat shield would be required (such as GE Elastomeric Shield Material - Series 100, which has a demonstrated useability after thermal soaking at 600°F). In addition the ablation shield immediately adjacent to the heat source would have to have an even higher temperature capability (such as the advanced heat shield systems considered for the X-20 program). The potential integration and vehicle modification problems are tremendous, and logically lead to the consideration of alternate methods such as unmanned recovery vehicles. The weight and thermal summary is shown in Table 3-6.

Utilization of an unmanned recovery system tailored to the heat source, or capsule requirements offers a potential solution to problems other than vehicle integration, particularly in the safety areas if directed re-entry is employed. The technology and reliability associated

Figure 3-6. Modular Concept

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TABLE 3-5. RADIATOR DESIGN PARAMETERS FOR MODULAR CAPSULE

Radiator inlet temperature	800 ^o F
Mean radiator temperature	750 ^o F
Emissivity	0.8
Panel Efficiency	60%
Radiator Area	50 Ft ²
Thermoelectrics	
Overall efficiency	2%
Available power	500 watts
Pump efficiency	25%
Hydraulic power	125 watts
Allowable pressure drop	2-4 psi
Radiator fluid	Dowtherm ET-378*
Radiator material	Beryllium with steel liners

* Initial decomposition temperature = 835^oF; for short term operations (several days) this fluid should be satisfactory.

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TABLE 3-6. WEIGHT SUMMARY MODULAR CONCEPT

Radiator (including coolant)		
50 Ft ² Be Radiator	35 Lbs	
Feed lines	<u>5</u>	
	40 Lbs	40 Lbs
Pumps (2)		20
Insulation (Min K)		
Area 26 Ft ² , Thk. = 3 in.		75
Radiation Shield (LiH) 12 cm		445
T/E Power Module (500 watts elec)		50
Capsule(s)		600
Miscellaneous Hardware		30
		<hr/>
	Total Weight	1260 Lbs

THERMAL PERFORMANCE MODULAR CONCEPT (Table 3-6 Cont.)

Radiator - Be/ss	
Operating Temp	800 ^o F
Thermal Capacity	25 KW
Area	50 Ft ²
Sink Temp	27 ^o F
Emissivity	.8
T/E Power Module (SiGe)	
Hot Side Temp	1500 ^o F
Cold Frame Temp	800 ^o F
Power Output	500 Watts
Heat Load to C/M	
With 3 in. ins. on module	.2 KW
With 3 in. under radiator	<u>.4 KW</u>
Total	.6 KW

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with manned systems has received much consideration, and much publicity. There is however a very comparable and demonstrated technology and capability existing in unmanned recovery systems (such as the Discovery vehicles). A very condensed re-entry/recovery synopsis is shown in Table 3-7 which is based on GE-MSD programs. (These comprise about 60 percent of the total related programs in the United States.) The 96 percent recovery record is quite significant since it includes all vehicle, including developmental programs since 1960. The variety of re-entry vehicles involved, and associated development paths is illustrated in Figure 3-7. A more complete presentation of associated programs and details is not possible due to program classification restrictions. There is however, a very well developed technology available for heat source recovery.

A study to evaluate this potential was undertaken and a re-entry/recovery vehicle concept evolved as shown in Figure 3-8. This unmanned recovery vehicle (R/V) is patterned almost identically after the NERV (Nuclear Emulsion Recovery Vehicle) which has been flown and recovered successfully from orbit. The concept of a split (or breaking) shield which allows the center nose piece to be extended forward for experiments in space, and is then retracted for re-entry has been demonstrated several times, with a photo of a recovered vehicle used for obtaining detailed heat transfer-re-entry data shown in Figure 3-9.

The NERV vehicle has sufficient volume to accomodate the vented Pu-238 heat source module described in Section 4.0, with the heat source being loaded into the R/V in the externally deployed condition, and the retro rocket package (coupled with space station position) providing a positive means to accurately return the payload. Upon re-entering, the R/V nose portion will be drawn in to provide a continuous heat shield (it may be possible to employ aerodynamic loads to insert the heat source)- nose section until the system has decelerated. A chute system then deploys, the nose pops out, and the system then water impacts (the interior foam provides floatation capability), with the recovery telemetry allowing for positive retrieval.

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**MARK 2/
MARK 2 SPACE LAB/
RECOVERABLE
DATA CAPSULE**

- IR SENSOR
- MAGNETOMETER
- EXOSPHERIC CONTROL
- ATTITUDE STABILIZED
- DATA CAPSULE

ABLE

RVX-1

- MATERIAL EXPERIMENTS
- PARACHUTE RECOVERY

RVX-2/2A

- LARGE VEHICLE PARACHUTE RECOVERY
- MATERIAL EXPERIMENTS
- SPHERE-CONE AERO-AND THERMO

MARK 3

- ABLATION
- FLAP EXPERIMENTS

MARK 3C TP

- ATTITUDE STABILIZATION
- REACTION CONTROL
- RADAR SYSTEM

MARK 6

- HONEYCOMB STRUCTURE
- HIGH CAPACITY DATA SYSTEM

NIKE ZEUS TVX

- NOSE ANTENNA
- SHARP BODY AERO AND THERMO
- TRANSMISSION THRU PLASMA

RMV

- STABLE PLATFORM
- SHARP BODY

MARK 12

- HIGH ACCURACY
- PENETRATION

MBRV

- NAVIGATION
- FLAP CONTROL
- STABLE PLATFORM
- RECOVERY

STRATEGIC
RE-ENTRY
SYSTEMS
RE-ENTRY
TEST
VEHICLES

DISCOVERER

- RETURN FROM ORBIT
- DATA CAPSULE
- RETRO ROCKET
- PARACHUTE RECOVERY

NERV

- MEASURE RADIATION IN VAN ALLEN BELT

BIOS I

- EARLY BIOLOGICAL EXPERIMENTS IN SPACE

BIOSATELLITE

- BIOLOGICAL PAYLOAD
- MICROMINIATURIZATION
- 30 DAY ORBIT
- LIFE SUPPORT
- TWO GAS LIFE SUPPORT SYSTEM

ADVANCED BIOSATELLITE

- LARGE PRIMATES
- 180 DAY ORBIT
- LIFE SUPPORT

OMBRV

OMRV
START

PRECISE LANDING

LIFTINGBODY
MANEUVERING
RETURN-FROM
ORBIT

SCIENTIFIC
RECOVERABLE
SYSTEMS

MARS VENUS LANDER STUDIES

VOYAGER LANDERS

- UNDERSTAND SOLAR SYSTEM
- LIFE DETECTION ON PLANETS

INTERPLANETARY
LANDERS

Figure 3-7. Evolution of Unmanned Re-entry/Recovery Vehicles

Figure 3-8. Re-entry/Recovery Vehicle

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TABLE 3-7. RE-ENTRY/RECOVERY HISTORY

270 Successful Flights out of 276
174 Ballistic Type Vehicles out of 176
96 Recoveries out of 100
29 Different ballistic entry and orbital entry/
recovery programs

Table 3-8 is a detailed weight breakdown for a 25 KW_t (vented Pu-238 heat source) based on flight qualified and flown (except for the heat source) hardware. A very attractive payload fraction of 50% emerges. Table 3-9 compares the weights of integrating individual modules versus single heat source with an Apollo command module. A comparison of manned versus unmanned fuel recovery is shown in Table 3-10. Excluding shield weight, either approach appears to produce the same weight. Additional studies will be carried out in this area, particularly since recovery of the long-lived isotopes has significant economical and national effort overtones.

Figure 3-9. NERV Vehicle

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TABLE 3-8. WEIGHT OF UNMANNED RE-ENTRY VEHICLE FOR HEAT SOURCE

<u>COMPONENT</u>	<u>WEIGHT (LB.)</u>
HEAT SHIELD AND STRUCTURE	180
FOAMED INSULATION	15.0
ELECTRICAL POWER SYSTEM	250
RE-ENTRY BATTERY	10.0
POWER CONTROLLER	1.0
INVERTER	3.0
SWITCHES	3.0
HARNESS	8.0
TRACKING, TELEMETRY, AND COMMAND SYSTEM	11.0
SENSORS & ACCELEROMETER	1.0
ANTENNA	1.0
TRANSMITTER	.5
CONVERTER CONTROLLER	.5
RECORDER	4.5
COMMUTATOR	1.0
MULTICODER	2.5
DEORBIT (RETRO) SYSTEM	75.0
RECOVERY SYSTEM	10.0
SEAMARKER (2)	2.0
PROGRAMMER	3.3
G. SWITCH	.9
FLASHING LIGHT CONTROLLER	.7
BEACON	1.0
FEEDLINE DISCONNECT	.8
FLASHING LIGHT	.3
ANTENNA	1.0
EMPTY R/V WEIGHT	316
HEAT SOURCE WEIGHT	300
TOTAL R/V - HEAT SOURCE WEIGHT	616 Lbs

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TABLE 3-9. WEIGHT SUMMARY FOR INTEGRATION OF HEAT SOURCE
WITH APOLLO SPACECRAFT

	24 INDIVIDUAL PuO ₂ FUEL CAPSULES 1.04 KW _t EACH PASSIVE COOLING	ONE 24 KW _t PuO ₂ FUEL ACTIVE COOLING
RADIATOR & PUMPS	-	60
PASSIVE FIN	70	-
CAPSULE HOLDERS	40	-
THERMAL INSULATION	100	200
RADIATION SHIELDING	275	445
T/E POWER MODULE	-	50
MISC. HARDWARE	30	30
TOTAL WEIGHT OF INTEGRATION COMPONENTS,	515	785
FUEL CAPSULE(S)	265	300
TOTAL WEIGHT ADDED TO VEHICLE	780 Lbs	1085 Lbs

TABLE 3-10. COMPARISON OF MANNED AND UNMANNED
RE-ENTRY VEHICLES FOR HEAT SOURCES

	MANNED APOLLO SPACECRAFT	UNMANNED RE-ENTRY VEHICLE
HEAT SOURCE	300 Lbs	300 Lbs
WEIGHT OF INTEGRATION COMPONENTS EXCLUSIVE OF RADIATION SHIELD	340	316
WITH RADIATION SHIELD	785	

* 25 KW_t PuO₂ CYLINDRICAL HEAT SOURCE MODULE

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3.2 SHIELDING

Shielding requirements of large heat sources are dictated by

- Allowable dose rates
- Vehicle configuration
- Fuel form
- Power loading

The allowable dose rate is in turn related to mission requirements (for example, military missions or emergency conditions could allow high, short time exposure rates). Recalling that industrial practices (based on a working period of 50 years) allow a yearly dose rate as follows:

$$\text{Dose Rate} = 5 (\text{Persons Age} - 18 \text{ years}) (\text{rem/year})$$

with maximum of 12 rem/year allowed (3 rem/quarter) if records are kept. Assuming an astronaut's exposure to radiation would be limited to about 10 years (estimated training and mission time), it would probably be acceptable to allow him to accept 25 rem/year or even 25 rem/missions) if the number of missions were kept low and spaced years apart. To permit shield design assessments a nominal value of 25 rem/year (exclusive of solar flares, Van Allen radiation, etc) was assumed.

To relate dose rate to vehicle interaction, a typical MORL vehicle was investigated (see Figure 3-10), with the heat source located at the rear extremity. Since the duty cycle calls for a variety of activities, the astronaut's location position and time of stay varies. An average duty cycle as related to position is shown in Table 3-11, with the net effect being a variation in allowed shield design dose at a distance of 1 meter from the heat source. Weight studies carried out in Section 3.3 were based on a 200 rem/year at 1 meter dose rate for conservatism in preliminary weight estimates.

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TABLE 3-11. AVERAGE CREW DUTY STATIONS

LOCATION NO.	PURPOSE	SEPARATION DISTANCE INCHES	PERCENT OF MISSION
1	SLEEP	210	33
2	RECREATION	150	18.3
3	DUTY	255	47
4	INSIDE MAINTENANCE	45	1.5
5	OUTSIDE MAINTENANCE	125	0.2
	MISSION DOSE, REM	MISSION DURATION, MONTHS	DESIGN DOSE RATE AT 1 METER, REM/YR
	25	3	5250
	25	6	2620
	25	12	1310

To facilitate shield thickness and weight evaluations, parametric shielding analyses were carried out for Pu-238, Cm-244 and Po-210.

The thickness of shields that will be required for large isotope heat sources (25 to 200 KWT) for both manned and unmanned missions were evaluated. The shielded dose rates are determined by the mission comparison evaluation which shows that manned applications will require limitations on dose rates (1 meter) ranging from 200 to 20,000 rem/year; whereas, unmanned applications will require limitations on dose rates to electronics ranging from 2×10^{11} to 1×10^{15} nvt/yr and 2×10^6 to 1×10^{10} rad/yr gamma, all at 1 meter. Based on ORNL-3576*, the allowable unmanned application dose rates are such that no shielding will be required. Thus, shielding will not be a consideration in the design of heat sources for unmanned missions, unless special restrictions are imposed by some item of payload

* E. D. Arnold, "Handbook of Shielding Requirements and Radiation Characteristics of Isotopic Power Sources for Terrestrial, Marine and Space Applications," ORNL-3576, April, 1964.

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equipment, (e.g. photographic film).

3.2.1 PLUTONIUM-238

The thickness of lithium hydride required to shield Pu-238 O_2 isotope sources is shown in Figure 3-11 as a function of power level and allowable dose rate. The gamma contribution from Pu-238 is sufficiently low that the 1 cm of high Z material assumed to be included in the source will provide adequate gamma shielding.

3.2.2 CURIUM-244

The thickness of lithium hydride and tungsten required to shield $Cm_2^{244}O_3$ sources is shown in Figure 3-12. The radiation level from the sources is sufficient to require significant thickness of both neutron and gamma shielding. The dose is divided evenly between the neutron and gamma contributions, although an optimization indicates that a slightly lower total thickness (2 percent) can be obtained with a division of 75 percent dose due to gamma and 25 percent due to neutron. Gamma self-shielding is significant as evidenced by Figure 3-12. In a compact, cylindrical array of 5 KWt sources, the capsules within the array are partially shielded by the surrounding sources and a lower thickness of tungsten is required. The reduction in thickness varies from 0.5 to 1.2 cm.

3.2.3 POLONIUM-210

The thickness of lithium hydride and tungsten required to shield Gd Po sources is shown in Figure 3-13. There is some gamma selfshielding as in the case of Cm-244. However, because of the generally smaller size of the isotope sources, the effect is relatively small and is neglected. Neutron shielding is not required at dose rates above 2000 rem/yr, and even at 2000 rem/yr, shielding is not required for power levels below 40 KWt. The apparent discontinuity in tungsten thickness at a dose rate of 2000 rem/yr is a result of the decrease in thickness that results from the decreased neutron dose rate, and the therefore higher allowable gamma dose rate.

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Figure 3-11. Pu O₂ Neutron Shielding

Figure 3-12. Cm₂₄₄ O₃ Neutron and Gamma Shielding

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Figure 3-13. Po^{210} Neutron and Gamma Shielding

3.2.4 SHIELD WEIGHT

The weight of the shield is determined by the configuration and volume of the heat source, the thickness of the shield, and the solid angle subtended by the payload. The shield thicknesses are defined in the previous sections; but, there are many possible combinations of the other parameters. In order to minimize shield weight, it is desirable to arrange the heat source in a compact array as shown in Figure 3-14a. In the early manned applications (MORL, MOL, Apollo-X, etc.), the heat source will likely be located within several meters of the manned compartment and a 2π shield of the shape shown will be required. On the other hand, to enhance passive emergency heat rejection, it is desirable to provide a heat source arrangement with a large surface area as shown in Figure 3-14b. The 2π shield required in manned applications is also shown.

In latter missions, a deployment capability will likely be developed and a simple conical shield such as that shown in Figure 3-14c can be utilized. A compact fuel source is provided

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A. Compact Cylindrical Array Fuel Source

B. Flat Array Fuel Sources

C. Conical Shielded Compact Fuel Source

Figure 3-14. Shield/Heat Source Configuration

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to minimize the shield weight; however, the shield will not enclose the heat source and passive heat rejection will not be impaired.

Shield weights were calculated for these heat source/shield arrangements with the results shown in Figure 3-15 through 3-23. The heat source volume is included as the ratio of the total heat source volume to the actual fuel volume required at each power level. The fuel power densities on which the fuel volume is based are also given for the various isotopes. The weight of shields for 2π shielded cylindrical arrays is given in Figures 3-15, 3-16, 3-17 as functions of the volume ratio, dose rate at 1 meter, and the beginning of life power level. The fuel source is a right circular cylinder with a length to diameter ratio equal to 1.0.

The weights of shields for the 2π shielded flat array fuel sources are given in Figures 3-18 through 3-20. The exposed side of the fuel source provides sufficient area to reject the isotope heat with a surface temperature of 1500°F and an emissivity of 0.9 (6.6 KWt/ft^2).

The weights of shields for conical shielded compact array fuel sources are given in Figures 3-21 through 3-23. The fuel source is a right circular cylinder with a length to diameter ratio equal to 1.0. The shield half-angle is 15 degrees, which is approximately the minimum that can be obtained in realistic vehicle integrations.

The heat source volume ratio is given in Figure 3-24 for all three isotopes as a function of heat transfer power density and power level. A low volume ratio is desirable because it minimizes fuel source and shield weight; however, the minimum ratio that can be obtained is limited by the maximum "packing" capability of the fuel source or by the maximum heat transfer capability. In addition to the fuel, void, capsule material, structure and internal coolant piping, the volume ratio includes an allowance for 2 inches of insulation around the heat source and a 3-inch section at the end of the source for coolant piping.

The design of the Po-210 fuel source is limited by a maximum heat transfer capability of about 300 KWt/ft^3 which results in volume ratios of 40 to 60. The Cm-244 source is limited, by the maximum packing capability of the fuel source, to power densities of about

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50 KWt/ft³ which results in volume ratios of 30 to 40. These equivalent volume ratios indicate that a direct exchange of Po-210 and Cm-244 is possible in a heat source. The Pu-238 fuel source can provide power densities of only about 10 KWt/ft³ with volume ratios of 15 to 20.

The shield weight penalty resulting from the use of a flat array fuel source designed for passive emergency heat rejection versus a compact fuel source designed for minimum shield weight is given in Figure 3-25. The weight penalty is given for all three isotopes for the fuel source power densities shown and for dose rates that are most typical for manned applications.

The weight penalties of compact versus flat capsule arrays are summarized in Table 3-12. Cm-244 shielding penalties are most significant ranging from 1000-1350 Lbs at a 25 KWt loading, into thousands of pounds at the higher power levels. The penalty for Po-210 is somewhat reduced but still significant. The Pu-238 difference is small at the lower power levels (several hundred pounds) but climbs steadily with power and becomes on the order of 1000-2000 Lbs for a 200 KWt loading. Results of these shielding calculations indicate the strong incentive for developing compact heat source configurations, as opposed to flat arrays.

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Figure 3-15. Weight of Shields for 2π Shielded Compact Cylindrical Pu²³⁸O₂ Fuel Source

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Figure 3-16. Weight of Shields for 2π Shielded Compact Cylindrical $\text{Cm}_2^{244}\text{O}_3$ Fuel Source

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Figure 3-17. Weight of Shields for 2 π Shielded, Compact Cylindrical Gd Po Fuel Source

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Figure 3-18. Weight of Shields for 2π Shielded, Flat Array $\text{Pu}^{238}\text{O}_2$ Fuel Source

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Figure 3-19. Weight of Shields for 2π Shielded, Flat Array $\text{Cm}_2^{244}\text{O}_3$ Fuel Source

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Figure 3-20. Weight of Shields for 2π Shielded, Flat Array Gd Po Fuel Source

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Figure 3-21. Weight of Shields for Conical, Shielded, Compact Cylindrical
Pu²³⁸ O₂ Fuel Source

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Figure 3-22. Weight of Shields for Conical Shielded Compact
Cylindrical $\text{Cm}^{244}\text{O}_2$ Fuel Source

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Figure 3-23. Weight of Shields for Conical Shielded, Compact Cylindrical
Gd Po Fuel Source.

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Figure 3-24. Heat Source Volume Ratio

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Figure 3-25. Weight Penalty of Shield for Compact Versus Flat Array
Fuel Source

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TABLE 3-12. SHIELD WEIGHT PENALTY OF COMPACT VERSUS FLAT ARRAY

	Power Level (Kwt)	Dose Rate (@ 1 meter)	PuO ₂ (Lbs ²)	Cm ₂ O ₃ (Lbs) ³	GdPo* (Lbs)
Compact	25	200	300	1350	1100
Flat	25	2000	0	1000	580
Compact	50	200	560	3300	2500
Flat	50	2000	220	2300	1045
Compact	100	200	1050	7500	5600
Flat	100	2000	530	5200	3500
Compact	200	200	2100	17500	-
Flat	200	2000	1035	11200	-

* Po-210 estimates based on estimate where beginning of life power = 2 x (end of life power).

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3.3 EMERGENCY COOLING

For truly passive emergency heat rejection, without the aid of heat pipes or coolant loops, the isotope heat source must be configured with a high surface to volume ratio and with sufficient surface area exposed to space to reject the total isotope heat without exceeding the limiting fuel temperature. This results in a fuel array shape such as that shown in Figure 3-26a.

Continuous Passive Heat Rejection

The ultimate in reliability is attained by designing the heat source for continuous heat rejection to space with only a fraction of the total energy given up to the power conversion system. In the emergency condition, the heat source temperature increases and all of the energy is rejected to space. However, such a concept involves an isotope penalty, in that more power must be provided than is required by the power conversion system. The amount of excess isotope is strongly affected by the difference between the normal operating temperatures and the maximum allowable fuel temperature in the emergency condition. This relationship is shown in Figure 3-27. Typical emergency and normal operating temperatures may be 2000°F and 1800°F , respectively, thus resulting in a temperature ratio of 1.09. This ratio results in a Q_T/Q_{PCS} value of 3.3 as shown by Figure 3-27 and a total power requirement of 82.5 KWt for an input of 25 KWt to the PCS. With larger temperature ratios, the excess isotope is significantly reduced; however, ratios above 1.2 are not attainable. Thus, the continuous heat rejection concept is not applicable because of the severe penalty on isotope inventory.

Evaporative or Sublimative Insulations

The excess isotope required for continuous heat rejection can be eliminated by protecting the heat source with an insulating material. For high reliability, however, it is undesirable to require an insulation that must be removed mechanically upon detection of an over temperature in the heat source. Either the temperature sensor or the mechanical mover may

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A. CONTINUOUS PASSIVE HEAT REJECTION

B. EVAPORATIVE OR SUBLIMATIVE INSULATIONS

C. INSULATIONS RETAINED BY LOW MELTING ALLOYS

Figure 3-26. Passive Emergency Cooling-Direct Radiation from Fuel Block

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Figure 3-27. Total Heat Source Loading for Passive Emergency Heat Rejection

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fail. As illustrated in Figure 3-26b, these inadequacies may be overcome by selecting an insulation material whose inherent properties are such that if the heat source exceeds the normal operating temperature, the insulation will either evaporate or sublime, thus exposing the heat source surface for cooling.

Materials of this type are being investigated for use as a coating on space nuclear power-plant radiator to prevent freeze-up of the radiators prior to plant start-up.

The principal short-coming of this concept is that the emergency heat dump would operate only once. This would be a severe restriction in a manned system application as a man would be required to don a spacesuit and perform extra-station operations to replace the insulation prior to a restart. This could involve a significant time delay and therefore increase back-up power source requirements. Also, the manned approach to the high temperature, high radiation source, isotope systems would be difficult.

Insulations Retained by Low Melting Alloys

Development will likely be required in the selection of an insulation with the exact evaporative or sublimative qualities required in the systems discussed above, in which case, it is also possible to employ a conventional insulation that is retained by structural members designed to melt if the fuel source exceeds a specified temperature. The spring loaded insulation would be ejected upon overheating to allow direct cooling.

This concept simplifies the insulation development problems; unfortunately, however, it is restricted by the same shortcoming of operation only once and of requiring manned replacement.

Insulated Heat Pipes

With the successful development of heat pipes, it will be possible to provide passive heat rejection and simultaneously free the heat source design from the requirement for high

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surface area for direct heat rejection. This is illustrated in Figure 3-28. The compact fuel array is advantageous because it has inherent advantages in:

- Minimizing shield weight
- Allowing location of the heat source remotely from the direct heat rejection surfaces of the vehicle
- Easing the fuel source/spacecraft integration difficulties.

The two concepts previously discussed of utilizing insulation to limit heat rejection in normal operating conditions can also be adapted to the heat pipes as shown in Figure 3-28. The same limitations are incurred, however, of one time use and manned replacement of the insulation. These limitations can be overcome with the development of a heat pipe that is automatically triggered by an increase in temperature and conversely, deactivated by a decrease in temperature. This is particularly attractive because the "on"--"off" characteristic of the heat pipe is controlled thermodynamically and therefore, provides extremely high reliability. The "on"--"off" characteristic must not necessarily be sharp, as a change in heat rejection rate of 10 to 20 would involve an isotope penalty of only 10 or 5 percent, respectively.

Semi-Active Heat Rejection

Semi-active heat rejection systems that utilize the expansion of a bi-metallic, of a gas cylinder, or of a bourdon type tube to effect movement, can be used to uncover or expose the heat source for cooling. These systems are illustrated in Figure 3-29. With proper design, the increase of fuel temperature can be used to provide the force required for operation of the system, thus eliminating the need for an outside power source and for detection of fuel temperature.

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- EVAPORATIVE OR SUBLIMATIVE INSULATION ON HEAT PIPE

- INSULATIONS RETAINED BY LOW MELTING ALLOYS
- NO INSULATION - HEAT PIPE COOLING TRIGGERED AUTOMATICALLY BY INCREASE IN FUEL TEMPERATURE

Figure 3-28. Passive Emergency Cooling Radiation from Heat Pipe

The shutters can be applied to a heat block for direct heat rejection or to a heat pipe radiator panel. The shutters in this latter case serve as the "on"--"off" control for the heat pipes and the heat pipes allow the use of a compact fuel array.

The principal criticisms of the shutters are that:

- With direct heat rejection, the large fuel array results in a shield weight penalty,
- With heat pipes, much greater reliability can be attained with a self-regulating heat pipe,
- The use of shutters with a compact array is contingent upon the successful demonstration of the heat pipe principle, and

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• AUTOMATICALLY ACTUATED SHUTTERS

• AUTOMATIC EXPOSURE OF FUEL CAPSULES

Figure 3-29. Semi-Active Emergency Cooling

- Shutters, because of their mechanical nature, likely cannot provide the desired reliability level.

The same mechanical movers used to open and close shutters can also be used to expose the fuel directly as illustrated in Figure 3-29. This concept would not pass a critical safety review, however, because of the danger of failure of one of the actuators in the exposed conditions and possible subsequent loss of the fuel.

Active Heat Rejection

Heat rejection can be provided reliably by circulating coolant loops as illustrated in Figure 3-30. This type of system provides the advantages of a compact heat source and the advantage of no moving mechanical parts. The disadvantages, of course, are the weight of the cooling equipment and the susceptibility of the loops to a loss of coolant. This latter disadvantage can be overcome by the use of multiple loops; however, a method must be devised for actuating the pump when the fuel array exceeds the normal operating temperature. For high reliability, this actuation should not rely upon temperature sensing equipment or mechanical actuators. A concept utilizing an EM pump with zero ΔT across the thermoelectrics when the PCS's are operating is worthy of investigation.

Recommendations

Primary emphasis should be placed on high reliability, redundant active coolant loops. However, in addition, provision should be made to either remove a thermal insulation covering the heat source, or eject the heat source assembly, to permit passive radiation cooling to occur. Concepts to accomplish this are discussed in Section 4.

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PUMPED COOLANT LOOPS

Figure 3-30. Active Emergency Cooling

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3.4 POWER CONVERSION SUBSYSTEMS

The power conversion subsystems which were to be considered are as follows:

<u>SYSTEM</u>	<u>NOMINAL HOT SIDE TEMPERATURE</u>
PbTe Thermoelectrics	1100 ^o F Hot Junction
SiGe Thermoelectrics	1500 ^o F Hot Junction
Hg Rankine Cycle	1250 ^o F Turbine Inlet Temperature
Brayton Cycle (with Argon)	1550 ^o F Turbine Inlet Temperature
Dowtherm Rankine	600 ^o F Turbine Inlet Temperature

During the first portion of the study period all systems except the Dowtherm Rankine were analyzed.

3.4.1 THERMOELECTRICS

The PbTe thermoelectric property data utilized was that derived from the SNAP-17B Study (Contract AT (30-1)-3308), and is shown in Figures 3-31 through 3-34 as a function of temperature. The Seebeck coefficient, electrical resistivity, thermal conductivity, and Figure of Merit (Z) data for SiGe were obtained through liaison with the RCA Thermoelectrics Products Group (Harrison, New Jersey), and are shown in Figures 3-35 through 3-42.

The material theoretical efficiency is, however, actually dependent on the ZT product, where

$$\eta_{\text{theoretical material}} = \left[\frac{T_H - T_C}{T_H} \right] \left[\frac{\sqrt{1 + ZT} - 1}{\sqrt{1 + ZT} + T_C/T_H} \right] \quad (\text{Eq 1})$$

The material efficiency potential (ZT) variation with temperature is illustrated in Figure 3-43. The theoretical material efficiencies of PbTe and SiGe are plotted as a function of the hot and cold junction temperatures (T_H and T_C) in Figures 3-44 and 3-45.

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Figure 3-46 is a composite curve illustrating the maximum material efficiency potential for the tellurides and the silicides based on a SiGe hot junction temperature of 1550° F, where the cascaded theoretical efficiency being can be expressed as

$$\eta_{\text{cascaded theoretical}} = \left(1 - \eta_{\text{PbTe}}^{\text{Theo}}\right) \left(1 - \eta_{\text{SiGe}}^{\text{Theo}}\right) \quad (\text{Eq. 2})$$

Since "practicality" aspects do not permit complete attainment of the material theoretical efficiency, a predicted thermopile, or device, efficiency is shown in Figure 3-47.

Attainment of 90 percent theoretical efficiency is feasible for SiGe, with a corresponding value of 85 percent for the PbTe (although this might take some careful engineering for a pressure loaded system). The resulting specific fuel loading ($\text{KW}_{\text{thermal}}$ per $\text{KW}_{\text{electrical}}$) is shown in Figure 3-48. A SiGe system would require 160 KW_t to produce 10 KW_e , while a cascaded SiGe - PbTe system would drop the fuel requirement to 125 KW_t . Other system considerations, such as pump power requirements, indicate that 200 KW_t might be representative of a SiGe thermoelectric conversion system to produce 10 KW_e .

3.4.2 MERCURY RANKINE CYCLE

The Hg Rankine cycle affords a power conversion efficiency improvement over the thermoelectrics, as well as having undergone component and hardware development. Cycle studies were carried out by means of a GE-MSD computer code in which machinery characteristics representative of the SNAP-2 and Sunflower turbomachinery were employed. Figure 3-49 (obtained from TRW-Cleveland) was used to define the turbine characteristics, and Figure 3-50 illustrates the cycle schematically.

Computed cycle efficiencies and power outputs (for a fixed 25 KW_t heat source input) are shown in Figure 3-51 and 3-52 for superheats of 200° F and 250° F respectively (increased superheat affords little gain in efficiency). Net power outputs of 2.5 - 3.0 KW_e are possible with a 25 KW_t heat source in the 100-200 psia turbine inlet pressure range (characteristic

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Figure 3-31. Variation of Seebeck Coefficient with Temperature

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Figure 3-32. Variation of Electrical Resistivity with Temperature

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Figure 3-33. Variation of Thermal Conductivity with Temperature

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Figure 3-34. Variation of Figure of Merit with Temperature

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Figure 3-35. N-Type Seebeck Coefficient versus Temperature
Silicon-Germanium Alloy

Figure 3-36. P-Type Seebeck Coefficient versus Temperature
Silicon-Germanium Alloy

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Figure 3-37. N-Type Electrical Resistivity versus Temperature
Silicon-Germanium Alloy

Figure 3-38. P-Type Electrical Resistivity versus Temperature
Silicon-Germanium Alloy

Figure 3-39. N-Type Thermal Conductivity versus Temperature
Silicon-Germanium Alloy

Figure 3-40. P-Type Thermal Conductivity versus Temperature
Silicon-Germanium Alloy

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Figure 3-41. Figure of Merit versus Temperature N-Type SiGe

Figure 3-42. Figure of Merit versus Temperature P-Type SiGe

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Figure 3-43. Variation of ZT Product with Temperature

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Figure 3-44. Material Theoretical Efficiency versus Hot and Cold Junction Temperature
P-LEG = Pb Sn Te
N-LEG = LTN 120

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Figure 3-45. Maximum Thermoelectric Theoretical Efficiency versus Hot and Cold Junction Temperature (Silicon-Germanium)

of SNAP-2). Presently demonstrated experience indicates extended operational times if boiling temperatures are held to the 1000-1050°F range, or lower. A Sunflower unit, for example, has undergone an endurance run in excess of 3500 hours. Cycle performance improvement due to improved turbine performance related to increased flow rates are shown in Figure 3-53.

A growth potential is afforded the Hg Rankine cycle by use of a SiGe "topping" unit which would initially receive the energy from a heat source and then dump to a Hg Rankine cycle as illustrated in Figure 3-54.

A comparison of the cycle efficiency of a "topped" Hg unit versus a Hg unit is shown in Figure 3-55. The greatest cycle efficiency improvement occurs at the lower turbine inlet pressures (with correspondingly lower boiling temperatures) to provide not only an improved cycle performance, but also the inclusion of a static energy conversion device to back up

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Figure 3-46. Material Theoretical Efficiency versus Cold Junction Temperature

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Figure 3-47. Predicted Device Efficiency versus Cold Junction Temperature

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Figure 3-48. Variation of Specific Fuel Loading versus Cold Junction Temperature

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Figure 3-49. Mercury Turbine Efficiency versus Turbine Shaft Power

Figure 3-50. Mercury Rankine Cycle Schematic

Figure 3-51. Mercury Rankine Cycle Performance Power Output and Efficiency versus Turbine Inlet Pressure

Figure 3-52. Mercury Rankine Cycle Performance Power Output and Efficiency versus Turbine Inlet Pressure

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Figure 3-53. Mercury Rankine Cycle Performance

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Figure 3-54. Cascaded SiGe - Hg Rankine Cycle

Figure 3-55. Comparison of Hg Rankine Cycle Efficiency with a SiGe Topped Hg Rankine Cycle

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a rotating unit. Figures 3-56 through 3-59 illustrate the power and efficiency potential available with SiGe being operated up to an 1800⁰ F hot junction temperature.

The Hg component weights utilized to arrive at a power conversion specific weight are shown in Table 3-13 through 3-15. Weights ranged from an upper value of 140 Lbs/KW_e (based on current component demonstration at 3.5 KW_e) down to 85 Lbs/KW_e (for uprated systems at 5.0 KW_e). These weights were used to derive overall system weights.

3.4.3 BRAYTON CYCLE

Brayton cycle studies (utilizing Argon) were carried out to determine cycle performance characteristics. The variation of cycle efficiency with compressor pressure ratio and turbine inlet temperature is typified with the results given in Figure 3-60, with cycle efficiency maximizing at a pressure ratio of 2.0-2.5. The effect of turbine pressure ratio to compressor ratio is quite evident, the implication being that direct gas cooling of the heat source will not be attractive due to excessive pressure drop in a volume restricted heat source.

The component efficiencies employed in the parametric studies are based on demonstrated hardware performance, and are summarized in Table 3-16. A nominal attainable value of 80 percent and 85 percent for compressors and turbines respectively was used to estimate minimum cycle performance. Figure 3-61 illustrates maximum cycle efficiencies as a function of compressor and turbine inlet temperatures, as well as other cycle parameters.

Figure 3-62 shows the significant efficiency increase resulting from compressor and turbine efficiencies of 83 percent and 88 percent respectively. A composite efficiency curve illustrating the significance of turbine-to-compressor pressure ratio is shown in Figure 3-63. A liquid metal cooled heat source is coupled through an intermediate liquid metal-to-gas heat exchanger to the Brayton loop.

The resulting specific fuel loading (KW_t/KW_e) is shown in Figure 3-64. A 10 KW_e output with 50 KW_t isotopic fuel loading appears feasible. Higher component efficiencies could lower this by 10-15 KW_t.

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Figure 3-56. Power Output and Efficiency of a Two Loop Mercury Rankine Cycle Topped by a SiGe Thermopile

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Figure 3-57. Power Output and Efficiency of a Two Loop Mercury Rankine Cycle Topped by a SiGe Thermopile

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Figure 3-58. Power Output and Efficiency of a Two Loop Mercury Rankine Cycle Topped by a SiGe Thermopile

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Figure 3-59. Power Output and Efficiency of a Two Loop Mercury Rankine Cycle Topped by a SiGe Thermopile

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TABLE 3-13. SNAP-2 COMPONENT WEIGHTS FOR LHS

A. PCS

CRU	60 Lbs
Regulator Tank	24 Lbs
Hg Inventory	165 Lbs
Hg Startup Gear	40 Lbs
Hg Piping	<u>10 Lbs</u>
	299 Lbs

B. Controls

Parasitic Load	64 Lbs
Load Switch	38 Lbs
Power Distribution	<u>44 Lbs</u>
	146 Lbs

C. Condensing Heat Exchanger

Based on 3.5 KWe	35 Lbs
------------------	--------

D. Totals and Specific Weights

PCS	299 Lbs
Controls	146 Lbs
Condenser	<u>35 Lbs</u>
	480 Lbs

$$\text{Specific Weight} = \frac{480}{3.5} = 137 \frac{\text{Lb}}{\text{KW}}$$

Notes:

1. Data from Page 21 NAA SR 10491 (Boiler not included)
2. Current Design Components

TABLE 3-14. SNAP-2 COMPONENT WEIGHTS FOR LHS

A. PCS

CRU	87 Lbs
Hg Startup Comp	150 Lbs
Hg Steady State Comp	<u>125 Lbs</u>
	362 Lbs

B. Controls

Parasitic Load Control	} 27 Lbs
Power Distribution	
Prototype Package	

C. Condensing Heat Exchanger

Based on 5.0 KWe	50 Lbs
------------------	--------

D. Totals and Specific Weight

PCS	362 Lbs
Controls	27 Lbs
Condenser	<u>50 Lbs</u>
	439 Lbs

$$\text{Specific Weight} = \frac{429}{5.0} = 88 \frac{\text{Lb}}{\text{KW}}$$

Notes:

1. Data from Page 12 of NAA SR 10491 (Boiler not included)
2. Weights are for current developmental components in a 5.0 KWe system (control package is questionable)

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TABLE 3-15. SNAP-2 COMPONENT WEIGHTS FOR LHS

A. PCS

Total Subsystem 350 Lbs

B. Controls

Power Distribution & Controls 100 Lbs

C. Condensing Heat Exchanger

Based on 5.0 KWe 50 Lbs

D. Totals and Specific Weight

PCS 350 Lbs

Controls 100 Lbs

Condenser 50 Lbs

500 Lbs

$$\text{Specific Weight} = \frac{500}{5} = 100 \frac{\text{Lb}}{\text{KW}}$$

Notes:

1. Data from Page 13 NAA SR 9056
2. Weights are objectives for a proposed 5.0 KWe uprated SNAP-2 system

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Figure 3-60. Cycle Efficiency Variation with Compressor Pressure Ratio

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TABLE 3-16. DEMONSTRATED BRAYTON CYCLE HARDWARE PERFORMANCE

COMPRESSORS:

Program	Diameter (inches)	Design Speed (RPM)	Test Fluid	Design Pressure Ratio	Design Inlet Temperature °F	Design Inlet Pressure (psia)	Target Efficiency (total/total) (%)	Test Efficiency (%)
USAF	3.2	64,000	Argon & Air	2.06	80	31.3	80.0	80.5
AiResearch	4.125	45,000	Argon & Air	2.0	140	15.0	82.0	82.6
NASA	6.0	38,500	Argon	2.38	76	6.0	80.0	81.6
<u>TURBINES:</u>								
AiResearch	4.6	64,000	Argon	2.044	1540	10.0	84.78	---
NASA	6.0	38,500	Argon	1.560	1480	13.2	88.0	89.0 (Cold Testing)

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Figure 3-61. Brayton Cycle Performance Variation of Maximum Cycle Efficiency; Working Fluid = Argon

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Figure 3-62. Brayton Cycle Performance Variation of Maximum Cycle Efficiency; Working Fluid = Argon

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Figure 3-63. Brayton Cycle Performance Variation of Maximum Cycle Efficiency; Working Fluid = Argon

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Figure 3-64. Variation of Specific Fuel Loading versus Compressor Inlet Temperature

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3.4.4 RADIATOR PARAMETRIC STUDIES

A radiator parametric study was undertaken to determine primary power conversion subsystem radiator characteristics for a 40-250 KWt heat rejection level. Studies were carried out by means of a GE-MSD Space Radiator Design and Optimization digital computer code which permits the steady state design and optimization of fin-tube-header radiator matrices employing either fluids or gases. Inputs to the program include thermal requirements (i.e., heat rejection), environmental factors, geometric design factors, physical property data, material armor criteria, pressure drop, and heat transfer characteristics.

The radiators examined were assumed to be in an earth orbit, cylindrical in shape, made of aluminum with stainless steel tube liners, and using liquid NaK as the flowing fluid. A fixed header length and number of panels were considered which was compatible with vehicles of 5, 10, and 20 foot diameters.

Three families of radiators (non-area limited) for heat rejection levels of 50 KWt, 100 KWt and 200 KWt were weight optimized for various NaK inlet temperatures, and fluid temperature drops (ΔT_{Rad}) across the radiator. Figures 3-65 through 3-67 contain plots of optimum radiator specific weight ($\text{Lb/KW}_{\text{Rej}}$) as a function of radiator inlet temperature and ΔT_{Rad} . The corresponding specific areas ($\text{Ft}^2/\text{KW}_{\text{Rej}}$) curves are contained in Figures 3-68 through 3-70, and the specific pump work ($\text{KW}_{\text{Hydraulic}}/\text{KW}_{\text{Rejected}}$) shown in Figures 3-71 through 3-73.

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Figure 3-65. Parametric Radiator Study Using NaK and 50 KW_t Heat Rejection
 Radiator Specific Weight Versus Radiator Inlet Temperature

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Figure 3-66. Parametric Radiator Study Using NaK and 100 KW_e Heat Rejection
Radiator Specific Weight Versus Radiator Inlet Temperature

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Figure 3-67. Parametric Radiator Study Using NaK and 200 KWt Heat Rejection
Radiator Specific Weight Versus Radiator Inlet Temperature

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Figure 3-68. Parametric Radiator Study Using NaK and 50 KWt Heat Rejection
Radiator Specific Area Versus Radiator Inlet Temperature

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Figure 3-69. Parametric Radiator Study Using NaK and 100 KW_e Heat Rejection
Radiator Specific Area Versus Radiator Inlet Temperature

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Figure 3-70. Parametric Radiator Study Using NaK and 200 KW_e Heat Rejection
Radiator Specific Area Versus Radiator Inlet Temperature

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Figure 3-71. Parametric Radiator Study Using NaK and 50 KW_t Heat Rejection
Radiator Specific Pump Work Versus Radiator Inlet Temperature

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Figure 3-72. Parametric Radiator Study Using NaK and 100 KWt Heat Rejection
Radiator Specific Pump Work Versus Radiator Inlet Temperature

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Figure 3-73. Parametric Radiator Study Using NaK and 200 KWt Heat Rejection
 Radiator Specific Pump Work Versus Radiator Inlet Temperature

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These curves can be used to match a radiator to a cycle by simply using the heat exchanger coolant outlet temperature as the radiator NaK inlet temperature and the heat exchanger coolant temperature rise as the NaK temperature drop through the radiator. The data from each radiator family is probably accurate for ± 10 percent of a given heat rejection level.

3.4.5 SYSTEM WEIGHT EVALUATION

Preliminary system weight estimates were made for heat source systems employing 25 KWt of Pu-238 coupled with the following power conversion subsystems:

- SiGe Thermoelectrics - $T_H = 1550^\circ\text{F}$
- Hg Rankine - Turbine Inlet = 1225°F
- Brayton Cycle - Turbine Inlet = 1550°F (Gas-cooled radiator)

A typical optimization curve for SiGe is shown in Figure 3-74, where the effect of cold junction temperature and radiator ΔT is shown for three thermal loadings. The significance of radiator ΔT (due to the increased pumping requirements) with increasing power level is evident. An optimum SiGe cold junction temperature of $400\text{--}600^\circ\text{F}$ appears to exist.

Detailed weight summaries are presented in Tables 3-17 through 3-19. An overall total weight summary comparison for the various power conversion subsystems is contained in Table 3-20. System specific weights of $700\text{--}1100 \text{ Lb/KW}_e$ for the SiGe thermoelectrics down to $400\text{--}500 \text{ Lb/KW}_e$ for the Brayton cycle result for a 25 KWt Pu-238 system (vented and non-vented capsules respectively).

System weight studies and overall cycle optimization will be carried out in detail during the latter part of the study based on the detailed parametric data obtained to date. An improvement in system specific weight should occur with increased fuel loading and power output.

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SYSTEM SPECIFIC WEIGHT & NET POWER OUTPUT
versus
COLD JUNCTION TEMPERATURE
SiGe, $T_H = 1550^\circ\text{F}$

Note: Does not include shield weights

Figure 3-74. System Specific Weight and Net Power Output Versus Cold Junction Temperature,
SiGe, $T_H = 1550^\circ\text{F}$

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TABLE 3-17. HEAT SOURCE SYSTEM WEIGHT SUMMARY - THERMOELECTRICS - SiGe

	<u>NON-VENTED TORUS</u>	<u>NON-VENTED BLOCK</u>	<u>VENTED TORUS</u>	<u>VENTED BLOCK</u>
HEAT SOURCE	600	890	300	530
SHIELD	440	390	265	260
RADIATOR AND RAD. STRUCTURE	285	285	285	285
POWER MODULE	50	50	50	50
PIPING & HEAT EXCHG.	25	25	25	25
EM PUMPS	<u>35</u>	<u>35</u>	<u>35</u>	<u>35</u>
TOTAL WEIGHT	1435 LBS	1675 LBS	960	1185
SPECIFIC WEIGHT	1060 $\frac{\text{LBS}}{\text{KW}_e}$	1240 $\frac{\text{LBS}}{\text{KW}_e}$	710 $\frac{\text{LBS}}{\text{KW}_e}$	880 $\frac{\text{LBS}}{\text{KW}_e}$
THERMAL LOADING = 25 KW _t (Pu-238)				
NET POWER OUTPUT = 1.35 KW _e				

TABLE 3-18. HEAT SOURCE SYSTEM WEIGHT SUMMARY— MERCURY RANKINE CYCLE

	NON-VENTED TORUS	NON-VENTED BLOCK	VENTED TORUS	VENTED BLOCK
HEAT SOURCE	600	890	300	530
SHIELD	440	390	265	260
RADIATOR & RAD. STRUCTURE	204	204	204	204
PIPING & HEAT EXCH.	25	25	25	25
POWER UNIT (PCS + CONTROLS + CONDENSER)	480	480	480	480
Em PUMPS	35	35	35	35
	<hr/>	<hr/>	<hr/>	<hr/>
TOTAL WEIGHT	1784 LBS	2024 LBS	1309 LBS	1534 LBS
SPECIFIC WEIGHT	775 LBS/KWe	925 LBS/KWe	570 LBS/ KWe	680 LBS/ KWe

THERMAL LOADING = 25 KWt (Pu-238)

NET POWER OUTPUT = 2.3 KWe

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TABLE 3-19. HEAT SOURCE SYSTEM WEIGHT SUMMARY — BRAYTON CYCLE

	NON-VENTED TORUS	NON-VENTED BLOCK	VENTED TORUS	VENTED BLOCK
HEAT SOURCE	600	890	300	530
SHIELD	440	390	265	260
RADIATOR	540	540	540	540
BRAYTON MACHINERY	350	350	350	350
PIPING & HEAT EXCHANGE	50	50	50	50
EM PUMPS	35	35	35	35
TOTAL WEIGHT	2015 Lbs	2255 Lbs	1540 Lbs	1765 Lbs
SPECIFIC WEIGHT	425	475	325	370

THERMAL LOADING = 25 KW_t (Pu-238)

NET POWER OUTPUT = 4.75 KW_e

TABLE 3-20. COMPARISON OF SYSTEM TOTAL WEIGHT VERSUS POWER CONVERSION SUBSYSTEM

	NET POWER (KW _e)	SPECIFIC WEIGHT (LBS/ KW _e)	TOTAL WEIGHT
THERMOELECTRICS (SiGe)	1.35	1060	1435
	1.35*	710	960
Hg RANKINE	2.3	775	1785
	2.3*	570	1310
BRAYTON	4.75	485	2015
	4.75*	385	1540

NOTE: BASED ON 25 KW_t INPUT

* VENTED CAPSULES

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4. CONCEPTS AND DESIGN ANALYSIS

4.1 CONCEPTS

The previously discussed over-riding requirements imposed by:

- Criticality
- Accidental Re-entry
- Emergency Cooling

are only three of the many design considerations applicable to large heat source assemblies. Table 4-1 presents an overall summary of design requirements. Primary emphasis was placed on compact arrangements to effect a saving of shield weight, and to allow more freedom in manned vehicle integration. Therefore, two basic approaches were taken, that of designing a single heat source module versus individual fuel capsules. These two concepts were dubbed the "modular" and "cell" concepts respectively.

The modular approach is shown in Figure 4-1, where the entire fuel load is contained within the fuel capsules (16 capsules) which are within a graphite block. Since this concept was evolved prior to a proper criticality and re-entry material assessment, the graphite was to serve both as a geometric (nuclear) spacer, and provide re-entry heat protection. The graphite was eventually discarded for reasons previously discussed. The concept does however provide a single heat source package for handling, and interfacing with a cylindrical heat exchanger (tube and fin) matrix, as shown in Figure 4-2.

The "cell" approach is illustrated in Figure 4-3 (graphite shown again since this was an early version), affords individual capsule flexibility. The power conversion heat exchanger flow channels are integral with the hexagonal structure. In addition, the potential for scheduled refueling (if Po-210 is employed initially) is possible without a complete loss of power. For example a 1/3 re-fueling allowance could be provided for. However, accommodation of individual fuel capsule random re-entry results in a weight penalty;

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Figure 4-1. Heat Source Module/25 KWt Pu²³⁸O₂ Graphite Matrix

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Figure 4-2. Heat Source - Heat Exchanger Interface

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TABLE 4-1. TYPICAL DESIGN CONSIDERATIONS

- MATERIALS COMPATIBILITY
- FUEL FORMS DATA
- FUEL CONTAINMENT
- FUEL AVAILABILITY AND COST
- CRITICALITY
- VEHICLE INTEGRATION
- LAUNCH ABORT
- ACCIDENTAL/PLANNED RE-ENTRY
- CAPSULE IMPACT
- SEA WATER SUBMERGENCE
- EMERGENCY COOLING
- POWER CONVERSION SYSTEM TYPES
- FABRICATION
- LAUNCH ENVIRONMENT

in addition, emergency cooling is more difficult if one needs to rely only on heat conduction paths leading to the lateral surfaces.

To facilitate design evaluation, a fixed 25 KW_t fuel loading (at end of life) was chosen, and Pu-238 as well as Po-210 fuels were utilized since they represent the maximum spread in thermo-mechanical matching requirements. Figure 4-4 illustrates a reference design $25 \text{ KW}_t \text{ PuO}_2$ heat source module, with Figures 4-5 and 4-6 illustrating the Po-210 concept which has 8 KW_t individual modules (at beginning of life) to produce 25 KW_t at end-of-life. These design concepts are discussed in detail in the following sections.

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Figure 4-3. Heat Source Cells/25 Kw Pu²³⁸O₂ Graphite Matrix

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Figure 4-4. 25 KWt Pu O₂ Module

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Figure 4-5. 25 Kwt Po^{210} Heat Source

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Figure 4-6. Individual Heat Source Module/8 KWt Gd Po²¹⁰

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4.1.1 MODULAR CONCEPT

The modular concept shown in Figure 4-4 is basically a hollow cylindrical heat source which is loaded into the spacecraft heat exchanger volume as a singular unit. The fuel capsules (22 capsules for a 25 KW_t load) are on a common diameter and immersed in sodium to enhance heat transfer characteristics. The fuel capsules (cylinders with ends down) with an L/D of 5.5 employ a composite wall. Helium pressure containment necessitates an inner refractory (TZM) pressure vessel, with a superalloy (Haynes-25) outercladding to provide oxidation resistance. The PuO₂ is encapsulated in Tantalum - 10% Tungsten. Internal capsule positioning is done by means of alumina end caps on the fuel capsules. The sodium filler provides another service, that of hydrostatic impact cushioning. Hydrostatic cushioning has been demonstrated in capsule impact testing carried out at GE-MSD.

The sodium containment vessel is Columbium-1% Zircornium for compatibility reasons, and employs domed toroidal ends consistent with impact requirements. The random re-entry heat protection (in this case earth orbital) material covers the exposed surface, and blocks the inner cylindrical cavity at one end.

The unit weighs 382 pounds without re-entry protection, and 502 pounds with a protective heat shield. Other design specifics are shown in Table 4-2. Nominal capsule wall temperatures of 1700-1750^oF are predicted. The unit can also operate under emergency cooling conditions with all fluid loops dead provided the lateral cylindrical surface can be exposed to space. Figure 4-7 illustrates a typical temperature map in the emergency mode, with temperatures lower than during nominal operating.

Vented capsules offer a means of reducing unit size and volume, and a comparative system is shown in Table 4-2, with the total weight dropping below 300 pounds. Emergency cooling conditions still require evaluation, as does the effect on volume of poisoning or diluting the fuel to prevent criticality. Venting is also attractive in that it allows use of Haynes-25 throughout the capsule (including fuel containment) averting a potential refractory-superalloy compatibility problem.

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Figure 4-7. Modular Concept - Typical Emergency Cooling Condition

4.1.2 INDIVIDUAL CAPSULES ("CELL") CONCEPT

The reference design for the cell concept is detailed in Figure 4-8, and borrows on the original cell structure illustrated in Figure 4-3. Removal of the graphite has permitted a volume shrinkage, and an hexagonal block (Columbium-1% Zirconium) with circular holes to receive individual capsules contains the heat-exchanger flow passages tying to the power conversion system. This concept allows for loading fuel capsules either on the pad, or in space; as well as a potential for re-fueling.

Six coolant passages are located equally around each fuel capsule with each passage containing a concentric tube which forms a return flow heat exchanger. Heat from the capsules is radiated to the surface of the heat block and then conducted through the heat block wall to the coolant passages. Manifolds located on the aft face of the heat block (non-loading end) join the inlet and outlet passages from each capsule. Several interconnection patterns are

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Figure 4-8. Twenty-Four Capsule Cell Concept - Long-Life
Non-vented Fuel Capsule

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possible to provide two or more parallel fluid loops to the power conversion system. Such multiple loop combinations can provide redundancy or permit stepwise reduction in power if failure in the power conversion system occurs. Columbium-1% Zirconium would be utilized for all tubing and manifolds.

The non-vented fuel capsule used in this concept would be similar to the one previously described with the exception that a "handling spoiler" would be located on one end of the capsule. Inconel X capsule retaining spring clips utilized to hold the capsule firmly in place during loading and unloading are attached to the "handling spoiler". Inconel X pressure disc retainers are attached to a beryllium cover plate which positions and secures the alumina insulating pressure discs and Inconel X Belleville spring washers to the cover plate.

Upon insertion of the fuel capsule into the block, the fuel capsule's domed end is inserted into an alumina retainer. Three retaining spring clips located at the other end of the capsule then lock onto the inside surface of the beryllium retaining gate, holding the capsules in place until the cover makes contact. Securing the cover forces the capsule further into the block through use of the Belleville spring washer and alumina insulating pressure disc, thus releasing the force on the spring clips. The cover plate could feature a rotational movement with a guide pin, an axial motion, a hinged motion, or a combination of several, to facilitate in-space fuel loading. Removal of fuel capsules would be accomplished in the reverse order. Burnup protection material is required on external surfaces.

The vented design would be identical to the one just described except it would be smaller and weigh less. The weights and other design details are shown in Table 4-2.

4.1.3 Po-210 DESIGN

The 25 KW_t (at end-of-life) design shown in Figures 4-5 and 4-6 permits mating through radiative coupling to a heat exchanger matrix (much like the modular Pu-238 concepts). However, the vast difference in fuel characteristics (a heating rate of 825 watts/cc theoretical and a 186 day half life) resulted in the investigation of a different design concept.

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TABLE 4-2. SUMMARY OF DESIGN CHARACTERISTICS

GAS CONTAINMENT	NON-VENTED	NON-VENTED (MODULAR)	NON-VENTED (CELL)	VENTED (MODULAR)	VENTED (CELL)
FUEL	Gd-Po-210	Pu-238 O ₂	Pu-238 O ₂	Pu-238 O ₂	Pu-238 O ₂
THERMAL OUTPUT, KW _t	25 (56.2) ⁽¹⁾	25	25	25	25
NUMBER OF MODULES	7	1	1	1	1
MODULE DIMENSIONS					
LENGTH, INCH	13.375	28.8	15.25	13.6	13.25
DIAMETER, INCH	4.875	14.3	17.2	14.3	12.5
NUMBER OF CAPSULES (TOTAL)	252	22	24	16	24
CAPSULE DIMENSIONS					
LENGTH, INCH	3.895	12.9	14.1*	11.3	11.9 ⁽²⁾
DIAMETER, INCH	0.625	2.64	2.32	1.92	1.7
WALL THICKNESS, INCH	0.050	0.120	0.120	.080	.080
CAPSULE MATERIAL	HAYNES-25	HAYNES-25 AND TZM	HAYNES-25 AND TZM	HAYNES-25	HAYNES-25
CAPSULE TEMPERATURES					
NORMAL OPERATION, °F	1977	1790	1770	1820	1780
MODULE WEIGHT, LBS (EXCLUDING RE-ENTRY PROTECTION)	222	382	743	223	453
TOTAL HEAT MODULE WEIGHT, LBS	334	503	909	282	580

(1) As loaded

(2) Includes handling spoiler

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The capsules (36 per module for a total of 252 capsules) due to heat transfer restrictions employed an effective void volume of approximately 95 percent thus allowing the utilization of Haynes-25 with the associated isotope short half life. Tantalum-10% Tungsten was chosen for final encapsulation (per Mound Laboratory experience). The capsules are in turn contained within perforated tubes (or "fingers") (80-85 percent porosity) which allow thermal radiation coupling to the interlocking heat exchanger surfaces.

The heat exchanger coolant passages are located equally around each of the twelve "fingers" of the module with tapered inlet and exit passages thereby forming a return flow heat exchanger. Heat from the capsules is radiated out through the holding tube to the surface of the heat exchanger and then conducted through the heat exchanger walls to the coolant passages. Manifolds, located at the aft face of the heat source, join the inlet and exit passages from each finger. Several interconnection patterns are possible to provide parallel fluid loops or redundancy to the power conversion system. Columbium-1% Zirconium is utilized for all heat exchanger tubing and manifolds.

The seven 8 KW_t modules then are located in a two-three-two array with three fusible link type bolts located about the center module (see Figure 4-5) with support being provided by one of the manifolds which also serves as the basic plate. They are retained by the hexagonal cartridge retainer plate which is secured to the base plate by the three fusible link bolts. Overheating of the center cartridge results in separation of the fusible links (probably a copper composite with reinforcing whiskers) whereupon the cartridges, cartridge retainer plate, and insulation pan are ejected by means of springs which are located at the cool extremities of the heat source. Inertia forces free the modules when the pressure plate is stopped by the cartridge ejection devices. The limited mating area of the cartridge retainer plate would then allow the exterior cartridges to fall free of the center cartridge. Emergency cooling conditions are shown in Figure 4-9 for the beginning-of-life conditions.

All of the designs discussed so far have been highly dependent on the fuel capsule design and technology requirements. Accommodation of severe emergency cooling conditions coupled with potential re-entry necessitate a high capsule reliability. As a result, the next section (Section 4.2) will deal entirely with capsule design aspects.

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$T_{\text{SINK}} = 120^{\circ}\text{F}$

Figure 4-9. Po-210 Concept Emergency Cooling Temperatures (50 KWt Total Heat Load at BOL)

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4.2 FUEL CAPSULE DESIGN

Maintenance of complete fuel containment under all plausible eventualities is the main aim in the capsule design. Consistent with this objective, the design parameters are optimized to best meet system performance objectives and minimum system weight. Primary factors affecting sizing of the capsules are:

- Thermal performance
- Gas pressure build-up
- Impact survival
- Materials compatibility with the environment.

These are covered in detail in this section. Additional factors affecting design, but not necessarily size, are compatibility interactions between the capsule materials, and fabrication considerations. Compatibility will be discussed in Section 5 and fabrication will be treated in this section as it relates to specific design features.

4.2.1 CONFIGURATION ANALYSIS

Before analyzing the capsule design problems, it is appropriate to establish a physical picture of the capsule and its qualitative features. Prior experience is a helpful starting point and Figure 4-10 illustrates an impact resistant concept resulting from SNAP-17B experience coupled with Company sponsored development activities. From this we identify the capsule as a fairly thick walled cylindrical shell constructed in two deep-formed halves joined by welding at roughly the axial mid-point. Contained within this shell is a sealed, thin-walled liner containing the isotopic fuel. The purpose of the inner liner is to protect the integrity of the main outer shell from the possible damaging effects of chemical attack by the fuel; it is also extremely helpful, if not essential, in the fabrication process, easing the problems of decontaminating the external surface of the outer shell for safe handling of the finished fuel capsule.

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NOTE

LINER WAS NOT INCLUDED FOR IMPACT TESTING

Figure 4-10. Impact Resistant β -fuel Capsule

The capsule shown in Figure 4-10 was aimed at a β -particle emitting fuel. Thus, the first departure for containing curium-244, plutonium-238, or polonium-210 is to provide for the evolution of helium gas. Void volume within the capsule is required to accommodate the gas, and the radius and thickness dimensions of the shell must be adjusted to withstand the pressure build-up. The cylindrical geometry is near optimum for pressure containment, but the ends must now be configured to those of a pressure vessel. Figure 4-11 illustrates the conceptual result. As shown in this figure, the inner liner is now configured as an annular container to provide void volume and to keep the fuel at the heat transfer surface. Typically these capsules will operate at high temperatures with a likely requirement for refractory materials to form the pressure vessel. This being the case, an impact resistant outer clad of superalloy or noble metal is a conceptual requirement to protect the refractory metal against oxidation when located within the Earth's atmosphere (during pre-launch handling or after an abort).

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Figure 4-11. α -fuel Capsule Concept

From a fabrication and heat transfer viewpoint, the resultant composite capsule shell shown in Figure 4-11 is undesirable. Recognizing the build-up of pressure as the underlying cause of the complication, there is a strong incentive to seek a means of venting the gas without compromising the overall safety objectives. Many means for achieving venting have been identified; the main problem lies in establishing, with full confidence, the ability of those schemes to meet the venting requirement for all plausible conditions during the required fuel containment period. Thus, without having a proven solution to the problem of venting, only the implications to system design, of such an approach, have thus far been considered in this study.

Actual capsule dimensions relate to the system requirements mainly through heat transfer considerations. Sufficient surface area must be available on the capsules to permit design heat flow at reasonable capsule temperatures; furthermore, dimensions must be selected consistent with the desired shape of the assembled heat source which is affected by radiation shielding and emergency cooling considerations. It is difficult to parametricize this type of

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analysis because of the numerous variables involved. However, some constraints may be placed on the variables to simplify the optimization process. For example, capsule L/D has been restricted to values in the neighborhood of 6 for this study. As this parameter increases, the capsule weight decreases for a given capsule radius and wall thickness. GE-MSD impact testing was conducted on capsules having L/D in the 2-3 range; however, other investigators have exceeded, somewhat, a value of 6. Pending further experimental evaluation, there is reason to believe that large L/D could be detrimental from an impact standpoint. Therefore L/D = 6 has been assumed to be an upper limit.

Assessment of the safety and handling requirements dictates radiative heat transfer between the capsules and the thermodynamic cycle heat receiver. Thus the radius, length, and number of capsules can be related to the heat source thermal rating and operating temperatures by the equation,

$$r^2 N \frac{L}{D} \cong \frac{Q_T}{4\pi K (T_1^4 - T_2^4)} \quad (4.1)$$

where K accounts for surface emissivities, view factors, and Stefan-Boltzmann constant; r and N are the capsule radius and number of capsules; and Q_T , T_1 , and T_2 are the total heat source thermal rating, capsule temperature, and heat receiver temperature. While equation (4.1) establishes a relationship among the capsule design parameters to assure sufficient heat transfer area, it does not guarantee sufficient internal volume to contain the fuel. Assuming the capsule, sized for heat transfer, is more than large enough to enclose the fuel, a certain amount of void volume will be present within the capsule. An approximate equation for this is,

$$V_v = \pi r^2 L - \frac{W_{Tf}}{N \gamma_f}, \quad (4.2)$$

where W_{Tf} is the total fuel weight and γ_f is its density. Replacing the fuel weight by,

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$$W_{Tf} = \frac{Q_T}{k_w},$$

and the number of capsules by,

$$N = \frac{A_T}{2 \pi r L},$$

the void volume becomes

$$V_v = \pi r^2 L - \frac{Q_T}{k_w A_J \gamma_f} \cdot 2 \pi r L$$
$$\text{or } V_v = \pi r^2 L - \frac{2 \pi r L K (T_1^4 - T_2^4)}{k_v} \quad (4.3)$$

In this equation k_v is the fuel specific power per unit volume. Expressed as a fraction of total capsule volume, the void is given by,

$$\frac{V_v}{V_T} = 1 - \frac{2K (T_1^4 - T_2^4)}{rk_v} \quad (4.4)$$

Clearly, there are combinations of temperature, radius, and fuel for which V_v/V_T could be negative. Such cases infer that fuel containment volume is the limiting design constraint rather than heat rejection area.

Equation (4.4) is presented graphically in Figure 4-12. Fuel specific power for plutonium is based on plutonium dioxide and that for polonium is based on gadolinium polonide suspended in a tantalum matrix. It is noteworthy that most of the polonium fuel volume is accounted for by the tantalum in establishing the value of specific power used in the figure. Despite this, the void volumes for polonium are seen to be very large compared to the plutonium. The

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effect of this can be represented in a more revealing manner as a limitation on equation (4.1). This is shown in Figure 4-13 for both polonium and plutonium where the heat receiver temperature is 1550°F , the end of life thermal rating is 25 KW, the capsule L/D is 6, the surface emissivities are 0.75, and the view factor is unity. Because of the short half-life of polonium-210, achievement of a given thermal rating at end of life requires a substantial increase in the initial fuel loading (in kilowatts) compared to plutonium-238. For a 90 day mission following a 48 day period from isotope separation to launch, double the thermal loading for plutonium would be required in a polonium design as reflected in Figure 4-13. In addition to showing the relationship between radius, number of capsules, and temperature, the figure shows the zero void volume radius cut-off. Thus it is seen that most of the range of variables for plutonium capsules is lost and the designs are restricted to the larger, lower temperature capsules with fewer of them required. In fact, capsule designs right on the cut-off line would have to be vented since this represents the zero void volume limit. To allow for gas pressure build up, the limit would have to be raised. Reducing the effectiveness of heat transfer, by lowering the emissivities or the view factor, yields some additional maneuvering room for detailed sizing of plutonium capsules, but there is a weight penalty associated with this. On the other side of the figure, polonium is seen to involve no such limitations. The problem here is more a matter of providing enough heat transfer area, hence there is ample volume. Curium would fall between the limits of plutonium and polonium; the latter therefore bracket the design problems and have received the bulk of the attention in this study to date.

Up to this point, the constraints on capsule design have had nothing to do with strength considerations. Wall thickness obviously has much to do with this, but so also do the void volume, fuel loading, radius and temperature. The evidence of Figure 4-13 suggests the importance of stress considerations in the plutonium capsules due to the squeeze on available void volume imposed by the fuel and heat transfer characteristics. Furthermore, the large disparity in power per molecule of gas evolved between polonium and plutonium implies some fundamental differences in the comparative pressure-stress conclusions. To close the loop on configuration analysis, the effect of gas pressure build-up must therefore be carefully examined.

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Figure 4-12. Variation of Void Volume Fraction with Capsule Temperature and Radius For Plutonium and Polonium Fuel.

Figure 4-13. Effect of Heat Transfer Requirements on Capsule Design Parameters for Plutonium and Polonium Capsules.

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4.2.2 GAS PRESSURE BUILD-UP

For an isotope which decays by alpha particle emission to either a stable element or a very long half-life daughter, the total number of gas molecules evolved is equal to the initial number of fuel molecules present. Examination of Table 4-3 shows that the three isotopes

Table 4-3. Comparison of The Three Fuels

ISOTOPE	POLONIUM-210	CURIUM-244	PLUTONIUM-238
Half-Life	138 d	17.6 y	89 y
Decay Particle	α	α	α
Daughter	Lead - 206	Plutonium 240	Uranium 234
Half-Life	Stable	6760 y	248,000 y

under consideration meet this criterion; hence decay follows a simple exponential law.

Figure 4-14 shows the history of gas evolution expressed as a fraction of the total number of moles available and plotted against time in half-lives.

Energy is released with each alpha particle; therefore, energy release also follows an exponential law. Thus,

$$q = ke^{-0.693f} \quad (4.5)$$

where f is time expressed in half lives, and K is a constant depending upon the particular isotope and the available quantity. The fuel will reach an initial equilibrium temperature depending on the heat transport mechanisms and parameters of the surroundings, and the temperature will decay to that of the environment. For the case of heat removal by conduction or convection, the instantaneous temperature may be expressed as a fraction of the initial equilibrium temperature (T_O) and the environment temperature (T_A) by,

$$\frac{T}{T_O} = \left[1 - \frac{T_A}{T_O} \right] e^{-0.693f} + \frac{T_A}{T_O} \quad (4.6)$$

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Figure 4-14. Helium Gas Evolution History

If the mechanism of heat removal is purely radiation, the corresponding temperature decay equation is,

$$\frac{T}{T_0} = \left\{ \left[1 - \left(\frac{T_A}{T_0} \right)^4 \right] e^{-0.693f} + \left(\frac{T_A}{T_0} \right)^4 \right\}^{1/4} \quad (4.7)$$

Both of these equations are plotted in Figure 4-15 for two choices of the heat sink temperature (T_A) to initial temperature (T_0) ratio. The substantial variation in the temperature histories, for low ambient levels, associated with these two modes of heat transfer is clearly shown in this figure.

Pressure build-up within a fuel capsule results from the combined effect of gas evolution and temperature decay. Modifying the perfect gas law to a dimensionless form,

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Figure 4-15. Capsule Temperature Decay History

$$\frac{PV}{RT_0 n_0} = \frac{n}{n_0} \cdot \frac{T}{T_0}, \quad (4.8)$$

the pressure history can readily be obtained in a very generalized manner. Thus, for convection and radiation, the two corresponding pressure relationships are,

$$\frac{PV}{RT_0 n_0} = \left\{ 1 - e^{-0.693f} \right\} \left\{ \left[1 - \frac{T_A}{T_0} \right] e^{-0.693f} + \frac{T_A}{T_0}^4 \right\}, \quad (4.9)$$

and

$$\frac{PV}{RT_0 n_0} = \left\{ 1 - e^{-0.693f} \right\} \left\{ \left[1 - \left(\frac{T_A}{T_0} \right)^4 \right] e^{-0.693f} + \left(\frac{T_A}{T_0} \right)^4 \right\}^{1/4}. \quad (4.10)$$

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In these equations, P is pressure V is gas volume, R is the universal gas constant, T_o is the initial gas temperature, and n_o is the number of moles of fuel. Typical pressure histories using these equations are illustrated in Figure 4-16.

In a real mission, step changes in the pressure history can occur. For instance, by suitable constraints in the power conversion equipment design, the receiver temperature can be held constant as the power decays. This causes the gas temperature to decay as though a high environment temperature (T_A) exists. After completion of the useful mission, the system could go onto an emergency cooling mode for which the initial equilibrium temperature would have been higher than the initial operating temperature. As an additional possibility, the fuel capsule could then be ejected and come to rest on the surface of the Earth in a cooling mode having an initial equilibrium temperature lower than the initial operating temperature. To examine such a sequence, the pressure history must be normalized about some reference temperature most logically the initial operating temperature. Figure 4-17 illustrates the example described above assuming radiative heat transfer. The curves are suitably normalized and the dashed line illustrates the life history path.

Figure 4-16. Typical Generalized Gas Pressure Histories

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Figure 4-17. Pressure History Example Normalized About The Initial Operating Temperature

4.2.3 STRESS RUPTURE ANALYSIS

The effect of the gas pressure is to stress the fuel capsule walls. Since the temperature level is high, the stress is associated with combined time and temperature effects and the mode of failure is expected to be stress rupture. To identify a satisfactory design it is necessary to determine the geometric parameters for which a 100% stress rupture life coincides with the desired mission duration. Since Larson-Miller stress rupture data is not necessarily presentable in a simple mathematical relationship, the general nature of the analysis deteriorates at this point, and design must be more specifically oriented. With the variety of materials, temperatures, fuels, and pressure history choices coupled with the tedious job of accumulatively analyzing stress rupture life, electronic data processing is a virtual necessity. A suitable capsule analysis computer program has been developed by GE-MSD, and its essential features are illustrated in Figure 4-18.

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Figure 4-18. Capsule Stress Rupture Life Analysis

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For a specific fuel and initial operating temperature, the pressure parameter becomes $\frac{PV}{W}$ as shown, where W is the fuel loading by weight. A geometric parameter $\frac{rW}{tV}$ can be identified which when multiplied by the pressure parameter yields hoop stress. For a given point on the $\frac{PV}{W}$ curve and a given value of the $\frac{rW}{tV}$ parameter, the corresponding value of hoop stress identifies a value for the Larson-Miller time-temperature parameter P. Corresponding to the $\frac{PV}{W}$ value is a temperature which, together with the value of P, identifies a corresponding stress rupture life t. The time increment dt associated with this analysis then identifies the fraction of the stress rupture life used, dt/t. Pursuing this line of reasoning along the pressure history curve, and accumulating the fractions of stress rupture life, a time will eventually be reached for which 100% of the life is used. To achieve a longer life, the analysis must be repeated for smaller values of $\frac{rW}{tV}$ or vice versa. Thus, by sufficient application of the process a curve of $\frac{rW}{tV}$ versus time can be obtained which represents a locus of values for which 100% of the stress rupture life is expended. By selecting a desired life span from this curve, the corresponding capsule $\frac{rW}{tV}$ limit is obtained. Specific values of r, W, t, and V may then be selected such that this limit is not exceeded, while meeting the design constraints presented earlier.

Many $\frac{rW}{tV}$ versus time plots have been generated for various fuel, capsule material, and temperature combinations. Figure 4-19 illustrates some representative examples. A TZM (molybdenum alloy), plutonium fueled capsule is shown for an initial operating temperature of 1800°F, and a Haynes 25 polonium fueled capsule is included for a 2000°F initial temperature. In each case the data is presented for a 0°F and 1550°F ambient temperature. For plutonium, it is assumed that mission lives would not exceed fifteen years which amounts to 0.174 half lives. As seen in Figure 4-16, the capsule pressure is quite low at this point, and there is no noticeable difference between the high and low ambient temperature cases. Therefore, a switch (at f = 0.174) to the emergency cooling mode has essentially the same effect as designing for this condition from time zero. On this basis then the TA = 1550°F curve for plutonium has no practical significance.

In the case of polonium, a different argument ensues. Since the operational life terminates at a fuel half-life of unity, the high ambient pressure case dominates the design. One significant

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Figure 4-19. Examples of $\frac{rW}{tV}$ for 100% Stress Rupture
Life Related To $\frac{tV}{\text{Capsule Life}}$

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finding in all cases of significance is that an asymptotic value of $\frac{rW}{tV}$ is approached in the range of 1-2 half-lives such that designs adequate for this condition are also good for 10 or more half-lives. This stems from the temperature decay, and the tremendous leverage of temperature on time in the Larson-Miller parameter.

Judging strictly from stress-rupture considerations, a reasonable design constraint on the $\frac{rW}{tV}$ parameter can be obtained from the asymptote values. These are plotted against initial temperature in Figure 4-20 for various combinations of material, fuel, and ambient temperature. Plutonium designs are shown for both Haynes 25 and TZM for the condition of decay to a 0°F ambient temperature. In the case of polonium, the 1550°F ambient temperature condition is appropriately shown. At first glance, this data comparison implies a greater stress rupture problem for the polonium capsules than the plutonium due to the lower allowable values of $\frac{rW}{tV}$ for a given capsule material choice. However, the polonium fuel loading is very low due to the high power density and the void volume fraction is fundamentally high as shown in Figure 4-12; therefore, any comparative conclusions on the stress rupture limitations must first consider the constraints imposed on $\frac{rW}{tV}$ by considerations other than stress rupture.

It has already been pointed out that the polonium capsules require twice the heat rejection surface area compared to plutonium capsules because of the rapid power decay. Therefore, where subscript (u) refers to plutonium and subscript (o) refers to polonium,

$$\frac{N_o L_o r_o}{N_u L_u r_u} \cong 2 \quad (4.11)$$

including the beginning of life power ratio effect, the fuel loading ratio for these two materials is roughly 100:1, therefore,

$$\frac{W_o N_o}{W_u N_u} \cong 0.01 \quad (4.12)$$

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Figure 4-20. Comparative Analysis of Polonium and Plutonium Capsule Design Stress-Rupture Characteristics

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As shown in Figure 4-12 there is a fundamental void volume disparity which is affected by temperature and radius. If a quantity ξ is designated which represents the ratio of polonium void fraction to plutonium void fraction, then

$$\frac{V_o}{V_u} \cong \xi \frac{r_o^2 L_o}{r_u^2 L_u} . \quad (4.13)$$

Finally, the wall thickness ratio may be designated by,

$$\frac{t_o}{t_u} = \eta . \quad (4.14)$$

Combining these four equations, it is found that,

$$\frac{\left(\frac{rW}{tV}\right)_u}{\left(\frac{rW}{tV}\right)_o} \cong 200 \xi \eta \quad (4.15)$$

To achieve a void volume ratio factor (ξ) of unity or less requires very extreme radius choices as shown in Figure 4-12. It can reasonably be stated that this factor will want to be greater than one. For a fair comparison, $\eta = 1$ may be assumed, and it becomes apparent that over-all design factors tend to set a ratio of at least 200 between the plutonium and polonium $\frac{rW}{tV}$ parameters. This is shown in Figure 4-20 by the shaded region. The lower boundary shows polonium designs lying within the stress rupture limit of the superalloy for the appropriate high ambient temperature condition associated with an operating life out to ten half-lives. The upper boundary shows plutonium designs lying beyond the stress rupture limit of the refractory metal for the appropriate low ambient temperature condition associated with an emergency cooling mode. It is therefore clear that stress rupture becomes a dominating factor in designing the plutonium capsules, and it is necessary to deviate from an optimum thermal design to satisfy this condition. Even more important from a practical design standpoint,

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where a polonium design can be comfortably defined within the limits of superalloy materials, the corresponding plutonium design must use refractory metals, and simultaneously requires roughly four times the wall thickness.

4.2.4 OTHER FACTORS AFFECTING GAS CONTAINMENT

For the preceeding analysis, all values of initial temperature should properly apply to the gas and not the capsule outer wall temperature. In practice there will probably be a difference between these two temperatures, but there is a great incentive to minimize the difference by suitable internal design. One obvious mitigating effect is associated with the ends of the capsules. Where the inner wall of the fuel annulus will undoubtedly be hotter than the capsule exterior, the gas will be able to transfer this heat to the cooler end domes thereby averaging down the effective gas temperature.

Another effect concerns the possible entrainment of helium within the fuel matrix; the fraction so held will not contribute to the pressure build-up problem. There are many uncertainties associated with evaluating the magnitude of this effect and its stability, particularly during re-entry where a tendency to bake-out may exist for a short time. On this basis, no attempt to take credit for this effect has been taken in this study.

To control the problems of criticality in the plutonium fueled versions of the heat source, nuclear poisons have been identified as a likely requirement. These will occupy space within the capsule and have the practical effect of reducing the fuel specific power measured on a volume basis, and will lower the values of the void volume fractions identified in Figure 4-12. Adverse implications to stress-rupture and capsule weight logically follow. This effect has not yet been factored into the capsule design analysis.

It is suspected that sea water corrosion will impair the integrity of the capsule walls after prolonged immersion, but this is not identified as a critical problem despite the major uncertainties associated with defining long term corrosion criteria. Assuming that data for Stellite is typical of the superalloys, it is found that polonium achieves ten half-lives of decay

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before a noticeable fraction of the wall is removed by corrosion. Since the available data is for periods well in excess of the corresponding 3.78 years, the significance of the effect should be very low in this case. For plutonium designs, the corrosion data must be extrapolated by two orders of magnitude which is obviously rash. In order to maintain surface continuity during impact, a superalloy outer cladding of 0.040 in thickness has been rather arbitrarily selected to provide oxidation protection for the refractory metal capsules. According to the available data, this thickness will be removed completely by sea water corrosion after six plutonium half-lives. At this time the temperature level is well below the refractory oxidation threshold and the pressure is reduced by a factor of at least four; hence, the refractory metal should be amply able to take care of itself for the rest of the way, in or out of water.

By far the most critical "other factor" concerning gas pressure build-up is impact. It is well known that terminal velocity impact substantially alters the capsule geometry, and the degree of alteration is somewhat proportional to capsule size. Since stress-rupture calculations are based on a carefully shaped pressure vessel configuration, and the post impact configuration is undefinable, impact may be expected to precede eventual rupture with a high degree of certainty for the larger capsules. This is of particular concern to the plutonium designs which are critical in this respect anyway. Figure 4-21 illustrates a typical deformed capsule cross-section for angle (45°) and side-on impact conditions as obtained on large capsules. In both cases, the deformation is accompanied by severe local working of the material which can adversely affect ductility characteristics. Capsule temperature decay after impact is associated with an initial equilibrium temperature of about 1200°F for exposure to air temperatures on the Earth's surface; hence, internal pressure build-up is less than that obtained under operating conditions, and stress rupture life is improved. However, in the case of a plutonium capsule, this is only expected to reduce the required wall thickness by a factor of two, and it is doubtful that the degree of margin thus inferred is sufficient to compensate for the shape changes illustrated in Figure 4-21.

In the case of the polonium capsule the severity of the post impact situation is greatly reduced. Overall design conditions dictate a much smaller capsule and a high operating temperature. The effect on stress-rupture of the much lower post impact temperature history is to drastically

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45° ANGLE IMPACT DEFORMATION EXAMPLE

SIDE-ON IMPACT DEFORMATION EXAMPLE

Figure 4-21. Typical Impact Deformed Capsule Cross-Sections

reduce the theoretically required wall thickness. Figure 4-22 compares the 100% used life curves of a Haynes 25 polonium fueled capsule for both a 0°F ambient and a 1550°F ambient temperature. The divergence of the curves at lower initial temperatures is directly translatable into a comparison of required wall thickness. For an abort immediately following launch, the ratio of thicknesses is seen to exceed an order of magnitude. Obviously this ratio would decrease as the operational life increases due to the rapid expenditure of stress rupture life during operation; however, it is quite apparent that a polonium capsule has a fundamental margin which may reasonably be expected to permit successful design against post impact rupture.

4.2.5 COMPARATIVE SUMMARY OF VENTING ARGUMENTS

The foregoing discussions have progressively intimated the desirability of a scheme for venting helium gas from the capsules; they have also implied that the desirability of such an approach differs in magnitude between polonium and plutonium fueled capsules. An assessment

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Figure 4-22. Effect of Post-Impact Environment On
Polonium Fueled Capsule Thickness Requirements

of the relative merits and need for capsule venting is summarized in Figure 4-23 on the basis of ten design factors.

1. Current state-of-the-art in the isotope fuel capsule field does not include a proven, acceptable means for venting; hence, there is a philosophical reason to remain with unvented designs for both fuels.
2. Stress rupture requirements impose relative limitations on the design scope for plutonium capsules compared to polonium, and the result is a strong incentive to vent the plutonium designs but not the polonium.
3. To contain gas pressure, plutonium designs require refractory metals in the temperature range under consideration, and this introduces the complication of an outer oxidation protection. Many unknowns are introduced by this composite concept both in the area of materials compatibility and design interactions. Venting eliminates

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	UNVENTED		VENTED	
	POLONIUM	PLUTONIUM	POLONIUM	PLUTONIUM
State-of-Art	●	●		
Stress Rupture	●			●
Materials Selection	●			●
Fabrication	●			●
Weight				●
Criticality Interactions				●
Sea Water Corrosion			●	●
Impact Survival			●	●
Life History	●			●
O ₂ /Fuel Effect	●			

Figure 4-23. Comparative Summary of Venting Arguments

this for plutonium designs, but is probably not required for polonium containment since the superalloys can reasonably contain the pressure.

4. Following directly from the materials selection, fabrication of a vented plutonium capsule should be far simpler than an unvented design assuming an acceptable vent can be made. Again, the unvented polonium design does not require a composite capsule and venting is not desirable on this basis.
5. Substantial weight savings are realized by venting a plutonium capsule, but are not in the case of polonium
6. Criticality is not a problem with polonium; however, the volume required for poisons in the plutonium designs competes with void volume requirements and aggravates the stress-rupture problem.

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7. Sea water corrosion is not identified as a serious issue affecting the venting choice; however, uncertainties in the available data and the long term effect on material properties leaves a minor incentive to vent in both cases.
8. Impact survival has been discussed above. Although there is some reason for confidence in the case of polonium to resist post-impact rupture, this issue would be eliminated by venting in both fuel choices.
9. Predictability and control of events, be it by human action or machinery reliability, deteriorates with time. Stress-rupture survival in the case of plutonium depends on predicting the pressure-temperature history for hundreds of years or guaranteeing retrieval. For polonium, the time period is a maximum of 3.8 years. On this basis then, the desirability of venting applies mainly to plutonium.
10. Oxidation of polonium fuel forms can produce gaseous or vapor forms by which part of the fuel may escape through a vent, as discussed in Section 5. Thus there is an incentive to maintain the sealed integrity of the capsule for this fuel. This issue does not apply to plutonium fuel forms.

Summarizing the inferences drawn above, it may be concluded that the complication of venting is not required for polonium fuel capsules and the possible weight savings would probably be trivial. On the other hand, venting will yield substantial weight savings for plutonium capsules and may prove to be required if all containment criteria are to be met.

4.2.6 IMPACT CONSIDERATIONS

Design of the capsules for impact survival cannot be accomplished on purely theoretical grounds; furthermore, impact testing programs have only scratched the surface for a comprehensive understanding of the problem. Testing at the GE-MSD High Explosive Field Test Facility has explored a wide range of variables as shown in Figure 4-24, and the following general conclusions have been drawn.

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SIZES	-	3 1/2 2 1/2
MATERIALS	-	STAINLESS, HAYNES 25, HASTELLOY N
END CLOSURES	-
MID CLOSURES	-
END TREATMENTS	-
THICKNESSES	-	0.206, 0.188, 0.120, 0.080
TEMPERATURES	-	RT AND 1100-1200°F
VELOCITIES	-	250-550 fps
ATTITUDES	-
HYDROSTATIC	-	
IMPACT SURFACE	-	GRANITE

Figure 4-24. Scope of GE-MSD Capsule Impact Testing Experience

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- Welds should be avoided at the end regions of the capsule
- High ductility is a more important impact survival property than high strength
- Heavy wall gages are not desirable, and there is probably an optimum value for a particular size and L/D.
- Material cross-section should be minimized to approach the primary wall thickness at shape discontinuities
- Closure welds at the mid-span point along the capsule axis are extremely reliable
- Energy absorbing end treatments are generally unnecessary
- Drag spoilers should be included on dome ended capsule configurations.

The most severe impact condition identified was a 45° angle impact with the capsule at elevated temperature (1100°F). Success was achieved at these conditions with a wall thickness as low as 0.080 inch and velocities in the neighborhood of 300 fps using Haynes 25. From these tests, energy absorbed has been correlated against wall cross-sectional area, and this has been used as a guide line for sizing capsules for impact resistance. Determination of impact energy is achieved by means of a computer program having the following inputs and analysis options.

Inputs

length
diameter
weight
initial altitude
configuration
fall mode

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Configuration Options

flat ended cylinder
recess ended cylinder
hemispherical ended cylinder

Fall Mode Options

tumbling
end-on

Typical data is shown in Figure 4-25 relating velocity, energy, and altitude to elapsed time for both a tumbling mode and an end-on mode. Actually, the capsule impact analysis may be somewhat academic for most cases since the heat source designs have generally evolved to modular concepts where the capsules are contained in multiple arrangements. However, the conditions which pertain for modular impact may still require a capsule essentially designed to survive impact alone.

4.2.7 INTERIM CAPSULE REFERENCE DESIGNS

Typical fuel capsule designs are illustrated, in some detail, in Figure 4-26. Included are an unvented plutonium capsule consistent with the 22 capsule, modular heat source concept shown in Figure 4-4; a vented plutonium capsule sized for the 24 capsule, individual cell heat source design of Figure 4-8, and an unvented polonium capsule for the 252 capsule, modular heat source design of Figure 4-5.

The large, unvented plutonium fueled capsule is sized to reject 1.14 KW at a surface operating temperature of 1790°F. Assuming heat transfer from the lateral surface only, these numbers are consistent with the combined conductive and radiative heat transfer path to the heat receiver. Configured as a cylindrical pressure vessel with ellipsoidal end domes shaped to exclude bending stresses at the change in contour, the capsule contains an annular shaped polonium dioxide fuel charge enclosed in a tantalum liner. The pressure vessel itself is composed of a double walled shell, being TZM (molybdenum) on the inside and Haynes 25

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VELOCITY - 10^2 FPS; ALTITUDE - 10^4 FT; ENERGY - 10^4 FT-LBS

Figure 4-25. Typical Results of Impact Velocity and Energy Analysis

TYPICAL DESIGNS

Figure 4-26. Interim Reference Capsule Designs.

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(cobalt based superalloy) on the outside. The TZM is selected for stress-rupture considerations and the Haynes 25 is for oxidation protection. Since the outer clad must not be broken by impact, a thin coating was eliminated because of the possibility of abrasive removal under a glancing impact onto an angled surface. The choice of 0.040 inch for the thickness of this layer is an arbitrary one based on judgment and manufacturing handling considerations.

Referring to Figure 4-20, the indicated rW value for stress rupture survival is 440. On this basis the unvented plutonium capsule^{tV} should have a pressure vessel wall thickness of 0.24 inch, or a total of 0.28 inch including the outer cladding. Even if weight were not a major concern, such a wall thickness is of questionable suitability for resisting impact. Some relief could be obtained by manipulating the radius and void volume, but the effect is to substantially enlarge the overall design with the associated heat source and radiation shielding weight penalties. The preferred answer is venting; however, retrieval is another out. For manned applications, where capsule retrieval is a planned event, a conductive or convective mode of heat transfer can be applied after operational life. In fact, the equilibrium temperature for simple exposure as Earth pushes the design rW value well over 1000. The only condition for which the large thickness is required is for decaying, within the heat source, for several hundred years, in space. For decay to 0°F by conduction or convection, the required wall thickness is 0.080 inch. Applied to the radiation heat transfer case, this thickness would provide a stress rupture life of 19 years which is judged to be well in excess of the likely operational life. Since this thickness also represents a favorable value for impact resistance, it was selected for the reference design pending further examination of the safety ramifications, and design schemes to circumvent the problem.

Choice of the fuel liner material depends on compatibility with the fuel and the capsule wall. TalOW was identified as a suitable choice (Section 5), and a 0.020 inch thick liner of this material is shown in the design. The liner actually consists of a fabricated closed cylinder with a concentric inner cylinder supported by rabbets on the end plates. Helium leakage around the rabbets permits access to the void volume. Final closure of the liner after insertion of the fuel is achieved by first placing a ring of tantalum foam on top of the fuel and then welding the end plate to the outer cylinder wall. The foam ring serves as a thermal and

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physical barrier between the fuel and the weld. Venting of one or both of the liner end plates would permit helium gas to utilize the void volume in the end domes. The approach and dimensions described for the liner are representative of current technology; several such liners have been fabricated in stainless steel, with depleted uranium as a simulated fuel, in connection with Company funded fabrication development studies.

After insertion of the assembled liner into the two TZM pressure vessel halves, present indications suggest diffusion bonding or electron beam welding as the mid-span closure joining technique. After annealing, weld inspection could be accomplished by such non-destructive methods as ultrasonic, eddy current, or infra-red testing. Alternatively, the presently used dummy weld verification technique could be used. Effective application of the Haynes 25 outer cladding is a critical problem because of the large difference in the expansion coefficient between this material and the refractory metal. Shrink fitting has been successfully applied to accomplish this assembly, but it requires heating the Haynes perilously close to the melting point, and the use of a suitable press to speed the actual insertion process. By this means a light interference fit at operating temperature can be obtained. Some form of metallurgical bonding between the two shells would be preferable and several manufacturing techniques can be identified to achieve this including coextrusion, pressure bonding, and explosive bonding. Diffusion across the interface may be a problem in such composite designs, in which case suitable barrier layers will require development.

The unvented plutonium design is rated at 1.04 KW per capsule and operates at a capsule surface temperature of 1780° F. In this case heat transfer is purely by radiation and the surface emissivities are assumed to be 0.85. Elimination of the void volume permits a much more compact capsule design as shown in Figure 4-26. More important, the shell can now be made of Haynes 25 and, for the sake of compatibility, so can the fuel liner. The 0.080 inch wall thickness is selected based on impact experience. Simplification of the fabrication over the previous example is obvious. A vent is called out in the figure but no attempt is made to describe such a device. Many schemes have been devised involving vitreous molecular sieves, mechanical sieves, or combinations. The problems of letting helium out and nothing else in are manifold, and the design implications are very much tied

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up with safety considerations. Conceptual work in this area has not yet identified a sufficiently acceptable design concept.

On the same scale as the plutonium capsule, Figure 4-26 also illustrates the reference polonium design. Its very small size is quite evident. Each capsule is rated at 224 watts output at the beginning of life and the surface temperature is 1980°F. The fuel is gadolinium polonide suspended in a tantalum matrix. As with the unvented plutonium capsule, the liner is Ta1OW and fabrication would follow the same pattern. A Haynes 25, single wall outer shell is shown consistent with the findings of the stress-rupture analysis. According to Figure 4-20, a design $\frac{rW}{tV}$ value of 17.2 is indicated for this temperature and ten half-lives containment, the indicated thickness is then found to be about 0.050 inch. Actually, since 0.3 half-lives are spent on Earth before launch at substantially lower than operating temperature, and since an emergency cooling mode of decay will be used after one half life, this thickness value is conservative from a stress-rupture view point. However, reductions below this value become suspect from both impact and fabrication considerations.

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There are significant thermal, mechanical, and chemical interfaces and interactions between the isotopic fuels, fuel cladding materials, and the heat source structural materials. The materials considered, available data, and their interrelations will be reviewed in the following sections. Much of the following information is based on up-to-date liaison with Mound Laboratory and Oak Ridge National Laboratory.

5.1 ISOTOPIC FUELS

The choice of fuel form is based on both functional and safety criteria. Functional criteria are influenced by mission constraints, weight limitations, operational conditions, and useable life. The safety criteria reflect shielding and handling requirements; and the desired behavior of the fuel under all operational conditions and under system failure conditions.

The fuel form characteristics related to functional criteria are:

- Power Density
- Allowable Temperature
- Compatibility
- Physical and Chemical Stability
- Fuel Synthesis and Encapsulation
- Availability
- Radiological Hazards

Power Density

The power density of the fuel must be sufficient to insure adequate thermal flux to the heat exchanger. To avoid excessive heating at the beginning of life, short-lived contributors to the decay heat must be excluded. To avoid excessive dilution of the thermal power, inert materials must be held to the minimum. In all cases it is desired to attain the highest isotopic purity of the heat source nuclides.

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Allowable Temperature

The fuel form must be capable of operating throughout the mission life with heat exchanger temperatures not less than 1600⁰F. In addition it must remain solid at the highest temperature attained within the fuel under emergency cooling conditions as well as at operational temperatures.

Compatibility

The fuel form must be chemically inert in contact with the primary encapsulating materials over the entire range of operating temperatures, for an indefinite period of time, without changes resulting from radiolysis, pyrolysis, spontaneous or induced fission, neutron bombardment, decay product in-growth or any combination of these.

Physical and Chemical Stability

As a result of radiolysis, pyrolysis, spontaneous or induced fission, neutron bombardment, decay product in-growth, or any combination of these, the fuel form must remain chemically and physically stable. In addition, it must not undergo any change in macroscopic structure which might rupture the cladding or cause shifting of the fuel in such a manner that hot spots would be created. The alpha particles emitted by the three fuels pick up free electrons to become neutral helium atoms. The pressure buildup resulting from helium evolution is factored into the capsule design, or is avoided entirely in the case of a vented capsule. However, the fuel form must not evolve any additional gas, or gaseous or volatile compound.

Fuel Synthesis and Encapsulation

The fuel form must be producible under normal glove box/hot cell conditions from available feed stocks. The methods of preparing the desired chemical and physical forms must be able to produce, routinely and consistently, material of the required purity, configuration, physical structure, and density in the environment of the glove box/hot cell.

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Availability

The selected fuel forms must be such that current technology exists at a sufficiently advanced level to insure an adequate supply of the desired fuels to meet the mission dates.

Two classes of hazards to man are associated with the use of radioisotopes:

- Those which may arise from excessive exposure to the penetrating radiation produced by the nuclide when it is outside the body, and
- Those which may arise from excessive uptake of the material into the system by inhalation and ingestion.

Shielding to the level required to prevent overexposure to radiation must be provided where and when needed. The radioisotope fuel must be completely contained to prevent uptake into the biosphere. Extrapolation of the properties of the encapsulating materials to time periods orders of magnitude longer than the duration of the tests on proposed materials, raises a significant question. (Consequently, it is not possible to predict the long term containment of long-lived fuel forms.)

The chemical composition and physical form of the fuel must be so stable under any of the credible failure conditions that it will not react either chemically or physically to damage or destroy the containment. If the containment is eventually breached by external corrosive agents, the fuel should have the minimum solubility and should be of such form that its uptake into the biosphere is as small as possible. Many of the functional criteria have application also for safety. At least one of the fuel materials, Pu-238, is known to be fissionable. Curium-244 may also be fissionable. It is imperative that critical accidents be precluded.

The nuclear, chemical, and physical characteristics of three radionuclides plutonium 238, polonium 210, and curium 244 and selected compounds were reviewed in detail in "Preliminary

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Fuels Data" DIN #4952-05-2, 18 June 1965. Data available to that date were reported and summarized. Additional data are included in Appendix A of this report.

5.2 POLONIUM AS AN ISOTOPIC FUEL

Fuel Forms

Polonium can be used in a heat source as the elemental metallic material or as a compound. Polonium resembles lead and bismuth in its physical characteristics but behaves chemically like its lighter homologue, tellurium. (References 1 through 7) Elemental polonium is limited in its applicability as a heat source because of its relatively low melting and boiling temperatures. Although the results of research and development programs now in progress may eventually permit use of Po-210 in its vapor phase at temperatures above 962°C (1765°F), it does not appear desirable to recommend its use for the Large Heat Source application. Polonium forms compounds with both the metals and the non-metals. Most of these compounds including oxides, halides, nitrides, and organics, are characterized by their instability at temperatures above 500°C . (References 5, 6, 7) One group of compounds, the polonides, formed by polonium with various metals and analogous to the metal sulphides, selenides, and tellurides, shows greater stability at high temperature. The rare earth polonides, including yttrium and the whole of the naturally occurring lanthanide series, have been synthesized by Mound Laboratory and some of their more significant characteristics determined. (References 8 through 12) Of these, gadolinium polonide with a melting / dissociation temperature of 1675°C appears as the most promising form for an isotopic heat source. (Reference 11) A list of pertinent properties of the metal and the gadolinium polonide is given in Table 5-1.

Gadolinium polonide is prepared by the direct reaction of the elements in an inert atmosphere or under vacuum at temperature of 600°C or above (Reference 12). A powered form of the compound can thus be produced. An alternative method is to mix the gadolinium or other rare earth metal as a powder with tantalum powder which is then pressed and sintered.

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TABLE 5-1. PHYSICAL PROPERTIES OF POLONIUM

	Elemental Polonium (Reference 5, 11)	Gadolinium Polonide (Reference 11)
Density, G Cm ⁻³	9.4	10.0
Melting Temp., °C	252 ± 2	1675 ± 50
Boiling Temp., °C	962 ± 2	Dissociates @ 1675
Heat of Vaporization $\frac{\text{KCAL}}{\text{MOLE}}$	24.597	?
Linear Expansion Coefficient Cm-Cm ⁻¹ - °C ⁻¹ (-196°C to + 30°C)	2.35 x 10 ⁻⁵	?
Vapor Pressure @ 25°C	5 x 10 ⁻¹⁵ TORR	?
Vapor Pressure 438°C to 745°C	LOGp = $\frac{(-5377.8)}{T} + 7.2345$	?
Specific Power W-G ⁻¹	144.1	82.4
Power Density, W-Cm ⁻³	1340	824.0
Thermal Conductivity	?	?

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The sintered matrix whose density can be controlled over quite broad limits, is then exposed to polonium vapor which reacts with the gadolinium, but not with the tantalum (Reference 5), to form the compound GdPo in situ. The power density can be controlled closely by adjusting the amount of gadolinium and excess polonium can be driven off as vapor at temperatures well below the dissociation/melting temperature of the GdPo. The tantalum matrix can be shaped to match the desired form of the encapsulation. The thermal expansion and thermal conductivity of the source thus produced are effectively those of the tantalum.

Problem Areas

The tendency of polonium compounds to dissociate with the production of free polonium vapor raises questions of the release of this vapor to the air and of compatibility of the vapor with fuel cladding materials. In the event of heat source burial and meltdown, free Po vapor will almost certainly be formed at some point in the thermochemical process. However, the possible release of this vapor to the air and the extent of the resulting contamination are very difficult to evaluate in a real situation, because of the multiplicity of factors affecting the interaction of the vapor with air, with molten constituents of heat source structure and capsule, and with the materials in which burial occurs. A rare earth polonide when exposed to the air will oxidize rapidly. (Reference 13). The polonium oxide thus formed will pyrolyze to Po vapor and oxygen around 850°C . As the temperature decreases, this Po vapor will recombine with oxygen and this ultimately will react with nitrogen oxides (Reference 5) to form the basic nitrate, $\text{PoO}(\text{NO}_3)_2$ which has a solubility in water around $2 \text{ mg} - \text{liter}^{-1}$. Additional studies which simulate the burial-meltdown accident and the oxidation conditions under careful control, are needed to permit a valid assessment of the biological hazard.

The compatibility of the fuel forms with encapsulating materials is of direct concern because it is necessary to design the containment to withstand all credible operational, environmental, and accidental stresses without release of the isotopic fuel. Data on the interaction of Po metal and GdPo with various encapsulating materials are being accumulated by Mound Laboratory. It has been reported (Reference 12) that tantalum and (Reference 13) tantalum-10% tungsten alloy are suitable materials for use in contact with both the metal and the GdPo at temperatures up to 1000°C . The metal attacks the Ta slightly but the reported

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depth of penetration (0.004 inch) is not coupled with information on the time of exposure; the GdPo appears less active than the metal and as noted above can be prepared by reaction between the Po as vapor and the Gd as metal in a Ta matrix, at around 1000°C. It is not sufficient, however, to consider only the interaction of polonium with its encapsulation; the behavior of the decay product must also be taken into account. Po-210 decays to Pb-206 which is radioactively stable.

In a practical heat source, by the end of 138 days, half of the initial loading of Po-210 has decayed to lead. Short term tests, 5 hours, at 1500°C showed that while lead wet tantalum slightly, it did not attack or alloy with it (Reference 12). Later reports indicate that exposure of the tantalum to lead for 1000 hours at 1800°C produced no significant corrosion of the tantalum. The behavior of the ternary system Po + Pb + Gd in contact with the Ta matrix/capsule material must be studied. Provisionally, tantalum is accepted as the primary encapsulating material for operations limited to 1000°C and below.

Development

The temperature at which the GdPo is formed may be of considerable importance to its stability. Although phase equilibria for the Gd-Po system are lacking, by analogy to the rare earth tellurides, and selenides it is anticipated that polonium will form other compounds than the monopolonide, including a sesquipolonide, a dipolonide, and a tripolonide (Reference 15).

The melting/dissociation temperatures for these compounds should be lower than that of the monopolonide and the resulting products are expected to be GdPo and free polonium as vapor or liquid. The existence of these compounds must be determined and their effect on the fuel encapsulation must be evaluated.

The other rare earth polonides especially dysprosium polonide deserve further investigation because their dissociation temperatures are somewhat higher than that of GdPo. Moreover, the possible use of materials other than tantalum for matrices should be examined. Extended tests at temperatures up to 1500°C with Ta as well as other candidate materials are needed.

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5.3 PLUTONIUM AND ITS CHARACTERISTICS

Plutonium is a synthetic element whose isotopes are produced by neutron irradiation of various isotopes of uranium. Its chemical and physical properties are generally similar to the other actinide elements with some notable exceptions (References 16 through 20).

The properties and behavior of plutonium have been extensively investigated for the nuclide, Pu-239, because of its use first as a material for nuclear weapons and more recently as a fuel for nuclear reactors. In discussing the behavior of the nuclide Pu-238, it has been assumed that many of the data generated for Pu-239 can be used without distinction for Pu-238. The typical isotopic content of production grade Pu-238 is shown in Table 5-2.

Plutonium 238 has been used in the form of the elemental metal as the heat source material for the SNAP-9 series of thermoelectric generators. The alloys of plutonium and its compounds can also be considered as heat source materials.

5.3.1 PLUTONIUM METAL

The high power density of Pu-238 metal makes it a natural candidate for use as heat source material. However, the applicability of the metal is limited to systems requiring relatively low temperatures. The melting point of the pure metal is around 630°C (1166°F) and small quantities of impurities (0.2%) such as iron and nickel lower the melting point to around 425°C . (Reference 13) Molten plutonium is corrosive to almost all practical capsule materials so that it is necessary to restrict the use of the metal to operating temperatures around 400°C . (Reference 12) For the Large Heat Source the use of Plutonium as the metal is not recommended.

5.3.2 PLUTONIUM ALLOYS

Several binary alloys of plutonium have been studied. The Pu-Zr combination has received fairly intensive attention. Although these alloys do increase the melting temperature, their

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applicability for systems requiring temperatures around 1000°C must be considered marginal. For the Pu-Zr alloy a 50 a/o dilution of Pu is required to reach a solidus temperature of 1050°C . (Reference 21 and 22)

TABLE 5-2. ISOTOPIC PURITY OF Pu-238
(Reference 13)

Plutonium Isotope	%	Half Life Years
238	80.0	86.4
239	16.5	2.44×10^4
240	2.5	6.6×10^3
241	0.8	13.2
242	0.08	3.8×10^5

5.3.3 PLUTONIUM COMPOUNDS

The fuel material must be usable to 1000°C (1832°F). This review of plutonium compounds has, consequently, been limited to those compounds whose melting temperature is not less than 1200°C (2192°F). A list of these materials with some of their pertinent characteristics is shown in Table 5-3 (Reference 12).

Not all of these materials appear as promising candidates for further study. The compounds with Al, Bi, Ni, Co, and Si, have the common disadvantage of being extremely difficult to prepare in stoichiometric compositions as is evidenced by the multiple compounds listed for these materials. Non-stoichiometric composition implies an excess of plutonium or the other constituent with consequent uncertainties in fuel characteristics and compatibility. The compounds with boron, beryllium, and fluorine which have high melting temperatures, will not be further considered at this time because of the very large neutron production rate, greater than $10^6 \text{ n-g}^{-1}\text{-sec}^{-1}$, arising from the (alpha, neutron) (α , n) reaction.

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TABLE 5-3. CHARACTERISTICS OF PLUTONIUM COMPOUNDS WITH $T_M > 1200^\circ\text{C}$
(References 12 and 44)

Compound	Crystal Structure	Density g per cm ³	Plutonium Density g per cm ³	Melting Point °C	Hardness (DPHN)
PuAl ₂	Fcc, Cu ₂ Mg type	8.06	6.57	1540 ± 50	550
PuAl ₃	Hexagonal, new structure type	6.67	4.98	1220 ± 4	495
PuAs	Fcc, NaCl type	10.39	7.9	2000	155
PuB	Fcc, NaCl type	≈13.85	≈13.25	2030 ± 50	---
PuB ₂	Hexagonal, AlB ₂ type	---	---	1825 ± 50	---
PuB ₄	Tetragonal	---	---	2050 ± 50	---
PuB ₆	Cubic, CaB ₆ type	---	---	2100 ± 50	---
PuBe ₁₃	Fcc, NaZn ₁₃ type	4.35	2.92	≈1700	565
PuBi	Fcc, NaCl type	11.62	6.2	≈1240	50-60
PuC	Fcc, NaCl type	13.6	13.1	1650	600-750
Pu ₂ C ₃	Bcc	12.7	11.8	2050	800-900
PuC ₂	(Unknown)	---	---	2250	---
PuCo ₂	Fcc, Cu ₂ Mg type	13.35	8.94	1320	625
PuCo ₃	Rhombohedral, PuNi ₃ type	---	---	≈1200	420
Pu ₂ Co ₁₇	Hexagonal, Th ₂ Ni ₁₇ type	10.10	3.14	1210	620
PuF ₃	Hexagonal, LaF ₃ type	9.32	7.52	1425	---
PuN	Fcc, NaCl type	14.25	13.47	2800	580
PuNi ₂	Fcc, Cu ₂ Mg type	13.1	8.78	1210	580
PuNi ₃	Rhombohedral unit cell	11.8	6.79	1220	485
PuNi ₄	Monoclinic	11.3	5.72	1250	225
PuNi ₅	Hexagonal, CaZn ₅ type	10.8	4.85	1320	525
Pu ₂ Ni ₁₇	Hexagonal, Th ₂ Ni ₁₇ type	10.3	3.23	1230	500
Pu ₂ O ₃	Hex, or Bcc	11.47	10.42	2085	235
PuO ₂	Fcc	11.46	10.1	2360	105
PuP	Fcc, NaCl type	9.87	8.74	2730	235
PuS	Fcc, NaCl type	10.60	9.36	2460	---
Pu ₂ S ₃ -Pu ₃ S ₄	Bcc, Th ₃ P ₄ type	8.41	6.98	1750 ± 10	---
Pu ₅ Si ₃	---	---	---	1377	369-484
Pu ₃ Si ₂	---	---	---	1445	586-693
PuSi	Orthorhombic, FeB type	10.15	9.08	1577	533
Pu ₂ Si ₃	Hexagonal, pseudo-AlB ₂ type	8.77	7.47	1750 ± 20	---
PuSi ₂	Bct, ThSi ₂ type	9.08	7.35	1795 ± 23	447-505

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(References 23, 24 and 25) The remaining compounds are binary compositions with the 5B and 6B elements of the periodic table. These appear to have real promise as fuel materials. It should be emphasized, however, that there are regrettably limited experimental data on these materials for many of the characteristics having direct engineering significance. Some of the nuclear and thermal properties of these candidate fuel materials are listed in Table 5-4.

The Arsenide, the Phosphide, and the Sulphides of Plutonium

Very little experimental information is available on the characteristics of PuAs, PuP, PuS, and Pu₂S₃. (Reference 12) As indicated in Table 5-4 all these materials have high melting temperatures, reasonable power densities, and acceptable levels of neutron production from (α , n) reactions. Their thermal conductivities have not been reported, but they are expected to be similar to those of the lanthanide congeners and close to the nitride. No data are available on the compatibility of PuAs and PuP with cladding materials. Recent work at Argonne National Laboratory (Reference 26) indicates that the plutonium sulphides are compatible with refractory metal such as W, Ta, and Mo at temperatures up to 2000°C (3632°F). The arsenide and the phosphide are suspected to have high vapor pressures as they approach their melting temperatures. This might limit their usefulness as a fuel. Synthesis of PuAs and PuP is at the laboratory stage of development. However, ANL has recently developed a pilot level process for the sulphide using uranium as a stand-in material. The resulting product of this process is a tough, machinable ceramic and it is thought that the plutonium sulphides will behave similarly. (Reference 26)

Plutonium Carbide

Plutonium monocarbide, PuC, has been studied extensively for the Pu-239 nuclide as a possible reactor fuel. The technology of its production is, consequently, reasonably well developed.

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TABLE 5-4. NUCLEAR AND THERMAL PROPERTIES OF PLUTONIUM COMPOUNDS
WITH $T_M > 1200^\circ\text{C}$

Material	Theoretical Crystalline Density	Melting Temperature		Thermal Conductivity	Neutron Production	Specific Thermal Power	Thermal Power Density
	g -cm^{-3}	$^\circ\text{C}$	$^\circ\text{F}$	$\text{cal-sec}^{-1}\text{-cm}^{-1}\text{-}^\circ\text{C}^{-1}$	$\text{n-sec}^{-1}\text{-g}^{-1}$	watts-g^{-1}	watts-cm^{-3}
Metal	15.5	630	1166	0.0098	3×10^3	0.46	7.13
PuAs	10.39	2000	3632	?	3×10^3	0.35	3.64
Pu ₂ O ₃	12.7	2050	3722	0.030	5×10^4	0.427	5.43
PuC	13.8	1650	3002	0.03	5×10^4	0.438	6.05
PuN	14.25	2600	4712	0.05	3×10^3	0.434	6.17
PuO ₂	11.46	2240	4064	0.01 (?)	4×10^4	0.396	4.51
Pu ₂ O ₃	11.47	2085	3795	?	4×10^4	0.417	4.78
				?			
				?			
PuP	9.87	2730	4946	?	3×10^3	0.41	4.0
Pu ₂ S ₃	8.41	1725(?)	3137(?)	?	6×10^4	0.38	3.2
PuS	10.6	2460	4460	?	6×10^4	0.41	4.3

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The melting point of 1650°C (3002°F) is lower than the nitride and oxide but is still high enough for heat source use. The compound is stable under radiation and neutron bombardment and will produce no gaseous or volatile compounds by daughter in-growth. The carbide will react with air or water and consequently must be handled in an inert atmosphere. Its power density is somewhat less than the metal and nitride but slightly higher than the oxide. Plutonium carbide reacts with many of the common encapsulating materials to form metal carbides. Although thorough testing at operating temperatures is needed to determine its long term compatibility, this carbide attack raises a problem which must be solved before it can be chosen for use as a heat source. The neutron production by the (α, n) reaction is slightly greater than that produced by the oxide and is a factor of 10 higher than the nitride. Plutonium also forms the sesquicarbide, Pu_2C_3 , and the dicarbide, PuC_2 , which differ from the monocarbide and from each other in power density and melting point. Data are lacking to permit a full evaluation at this time. (Reference 44)

Plutonium Nitride

The plutonium nonnitride, PuN , has a high melting point (around 2600°C (4712°F)). It may be prepared by the reaction of nitrogen with PuH_2 at 600°C . Its power density is second only to the metal. The neutron production via the (α, n) reaction is approximately equal to the spontaneous fission production, since the threshold for this reaction is above 6 Mev for both the isotopes. (Reference 27) Unfortunately, the techniques of producing this compound are on a laboratory level and many of the important physicochemical and thermal properties are unknown. The material must be handled in an inert atmosphere because of its pyrophoricity. Upon exposure to air or to water it is rapidly converted to the oxide. Data on its compatibility with possible encapsulating materials are limited. Although its high power density, good thermal conductivity, and high melting temperature make it potentially useful, it does not at this time appear to be suitable for the LHS application without substantial additional work on methods of preparation, compatibility, and definition of basic properties.

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Plutonium Dioxide

Plutonium forms three oxides; PuO , Pu_2O_3 , and PuO_2 . (References 16 through 22 and 28 through 34) The dioxide is the most stable form and has been studied extensively, for the Pu-239 nuclide, as a reactor fuel usually mixed with uranium or thorium oxides or both. Its preparation from feed solution such as the nitrate can be accomplished by precipitation as the oxalate and subsequent calcination to the oxide. Because of the continuing interest in the oxide as a reactor fuel, its technology is well developed. For design purposes the melting temperature of 2240°C (4064°F) will be used (References 17, 19, 21, 22, 32, 42). Melting temperatures ranging from 2170°C (Reference 22) to 2400°C (Reference 42) have been reported. The melting temperature is known to be affected by impurities in the processed materials, and also by exposure of the compound to neutral, reducing, or oxidizing conditions.

Studies on the compatibility of PuO_2 with various candidate encapsulating materials are being carried on by Mound Laboratory at temperatures around 1000°C (1832°F) which closely approximates the proposed capsule operating temperature. (References 12 and 13) The tests already completed were relatively short term but several materials after exposure for various periods up to 8000 hours appear to suffer little or no attack. (Reference 34) Among the materials displaying acceptable corrosion resistance are several of the superalloys and nickel. It has been reported that both Ta and W reduce PuO_2 at higher temperatures around 1600°C (2912°F) (Reference 34). It should be noted that the results thus far obtained, although sufficient for initial design are inconclusive and additional work is needed in this area. Special emphasis on the effects of small quantities of impurities in the fuel or cladding is required.

The stability of PuO_2 to pyrolysis has been studied under neutral, reducing, and oxidizing conditions (References 36 through 41). There is some volatilization of the oxide probably as a suboxide under neutral and reducing conditions. At very high temperatures around 2100°C deoxygenation to the sesquioxide may occur, a reflection of the greater stability of the lower oxidation states of plutonium (Reference 18). In oxidizing atmospheres the

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degree of volatilization is dependent on the partial pressure of the oxygen. Since this mechanism does not become significant at temperatures below 1600°C (2912°F), PuO_2 appears to have satisfactory stability for the LHS application. Further study of the behavior in the neutral atmosphere created by the evolution of helium by alpha decay is required.

Experience with plutonium-239 dioxide as a constituent of mixed oxide reactor fuels indicates a high degree of dimensional and chemical stability under the neutron and gamma fluxes and fission product in-growth occurring in reactor operation. The long term stability of the dioxide (one half-life of Pu-238 - 86.4 years) to alpha particles and neutrons, to fission, and to the ingrowth of the decay products and the products of spontaneous and induced fission needs further evaluation.

As noted in the preceding paragraph, the resistance of PuO_2 to release of oxygen by radiolysis seems well-confirmed by the reactor fuels experience. The immediate decay chain products appear equally stable under the operating conditions.

Further study is needed of the decay chains of the other plutonium nuclides and evaluation of the quantities of gaseous and volatile nuclides and their compounds produced by spontaneous and induced fission, to attempt assessment of the total gas evolution over even one half-life: 86.4 years.

The most conspicuous source of gas evolution is the alpha decay of Pu-238. Subsequent to its emission from the nucleus, the alpha particle picks up to electrons to become a neutral helium atom. If this neutralization occurs within the solid PuO_2 , some or all of the decay helium may be retained in the interstitial vacancies of the oxide crystal.

It must be concluded interstitial retention when it occurs is a short-term phenomenon and over long periods of time greater than 5 years, the release of helium, fission gases, and volatiles, must be regarded as complete. Cladding design objectives must consequently be directed either to release or containment of these gaseous materials.

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Plutonium dioxide like most of its congeners among the actinide and lanthanide elements forms a highly refractory material when prepared at high temperatures. The reactivity is strongly dependent on the temperature of calcination and the physical size of the particles.

The material calcined at temperatures above 900°C and low temperature calcined material subsequently fired and sintered at high temperatures is chemically very inert. Dissolution of high fired plutonium is achieved only with difficulty using reagents such as H_3PO_4 (85% at 100°C), 10M HNO_3 + 0.05 HF, and 5-6M HI. Refluxing is necessary in all cases and the dissolution rate is slow (References 12 and 18).

Solubility rates in sea and fresh water, which are of biological significance, have not been studied although some preliminary data are given in Reference 12 and 34. Moreover, the solubility rates depend upon many fortuitous factors which cannot be assessed with certainty for a real situation.

At this time PuO_2 appears as the most acceptable plutonium fuel material. The preliminary designs which involve power density, reactivity suppression, cladding compatibility and weight are based on the use of this compound.

5.3.4 CHOICE OF PHYSICAL FORM

The PuO_2 can be prepared in a number of physical forms including the low and high temperature calcined powder, pressed and sintered pellets, and high fired granules and spherules with diameters in the micron range. In addition, the oxide can be mixed with other oxides, coated with metal vapor, pressed, and fired, and mixed with metal powder, pressed and sintered. All of these forms have advantages for different applications and the ability to fabricate these forms is well established in current plutonium technology. The loose calcined powder presents a large surface area, and because of its greater surface activity may present problems of compatibility with the container; it will not be further considered as a fuel form.

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Two of the possible physical fuel forms deserve further consideration to meet the current functional and safety criteria:

- Pressed and high temperature sintered pellets
- High-fired particles metal-coated, then pressed to give a substantially continuous metal phase with 80 percent PuO_2 content

Pellets are prepared by compacting and sintering low temperature calcined material under controlled atmosphere conditions and at high temperatures. This process yields a product with better than 90 percent of theoretical density. Pellets can be prepared in almost any geometric shape and in sizes which are limited only by the capability of the presses currently available in the glove boxes. The sintered pellets exhibit about the same resistance to dissolution as the microspheres but they offer a much smaller surface area. The technology of pelletizing PuO_2 is well developed at Mound Laboratory and in a number of commercial installations.

The thermal conductivity of PuO_2 has not been reported but it is expected to lie in the range from 0.001 to 0.01 $\text{cal-cm}^{-1}\text{-sec}^{-1}\text{-}^\circ\text{K}^{-1}$. Although a solid sintered body provides a better heat conduction path than a bed of spherules, there will be a substantial temperature gradient between the center and the surface depending on the size of the fuel body (Reference 43). It is desirable to have a good conductive contact between the fuel body and the cladding especially when the fuel material has a low thermal conductivity. Current technology for the production of plutonium ceramic forms does not permit close tolerances on fired bodies (Reference 34). Hot pressed materials can be better controlled dimensionally but the resulting body may not achieve the desired density. Machining of the plutonium ceramic creates problems which are amenable to solution but which have not been extensively studied. Autoclaving or other pressing techniques can be used to effect intimate contact between fuel body and clad (Reference 44).

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Techniques developed for power reactor fuels production give promise of providing a fuel form with many desirable characteristics. A high-temperature fired particle of small size: roughly 100 to 200 microns, is produced by furnace firing at 2100°C or by plasma torch. The former method yields particles which are smooth but slightly irregular in shape and which have density very close to theoretical. The latter method produces nearly perfect spherules with somewhat lower density because voids frequently form inside the spheres. The fired particles are then coated with almost any metal or alloy or with another oxide followed by a metal coat. The coating can be made dense and substantially impermeable. The coated particles are then pressed to the desired shape and sintered if necessary (Reference 44).

Cermets of this kind have been made with a continuous metal phase and with about 80 percent PuO_2 by volume. The resulting cermet fuel pellets have several characteristics which make them appear useful as heat source fuel forms:

- A good thermal and even a metallurgical bond can be formed between the cladding and the fuel body
- The thermal conductivity is at least a factor of 10 better than the oxide fuel by itself
- The metal coating forms a barrier to minimize dissolution of the oxide if it is exposed to water, and restricts the surface area exposed at any given time.
- The additional inert material provides diluent helping to reduce the fissile reactivity
- The processes are well-developed

Plutonium dioxide in a cermet form has been identified as the fuel form for the preliminary design effort.

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5.3.5 PROBLEM AREAS

Plutonium-238 is susceptible to fission from fast neutrons. The fast cross sections reported by Butler and Sjoblom and BNL are shown in Figure 5-1. Experiments carried out using the Jezebel critical assembly have confirmed the fissionability of Pu-238 (Reference 45) and calculations by Hanford (HW-84369) give a bare critical mass about half that for Pu-239. Table 5-5 lists some preliminary estimates of critical mass for Pu-238 metal and oxide. It is considered necessary to preclude the formation of a critical mass through any deformation of the Large Heat Source by providing sufficient diluent/poison intimately mixed with the fuel to maintain the reactivity substantially below 1.0 under all credible conditions. This problem is discussed further in the Safety section of this report. At this time the most promising diluent poison appears to be gadolinium oxide which has been used by ANL in quantities up to 40 percent by volume in UO₂ fuels operating at 2000°C (Reference 26). An alternative material is hafnium oxide which has slightly larger cross-sections in the fast region; no data have been reported on the use of this material in a fuel form. Other materials which appear promising for absorption and scattering may introduce problems of compatibility and in some cases create significant nuclear problems; boron and beryllium increase the neutron yield via the (α , n) reaction by about 100 (Reference 23, 24, 25).

Radiation measurements on typical encapsulated sources of Pu-238 indicate the presence of measurable amounts of Tl-208 a decay product of the Pu-236 chain having energetic gammas at various levels including 2.6 Mev. (Reference 12) It is estimated that this will reach its peak after about 20 years when it will contribute about 50 percent of the total gamma radiation from the source (Reference 46).

The neutron contribution from the (α , n) reaction on oxygen-17 and oxygen-18 is nearly 15 times that resulting from spontaneous fission. Contributions of this kind may also arise from other light element impurities such as fluorine, boron, and the like. It is important to insure that the process development effectively excludes the light element impurities to the greatest practical extent (Reference 46).

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Figure 5-1. Fast Fission Cross Sections - Pu-238

TABLE 5-5. MINIMUM CRITICAL MASSES (SPHERICAL)

Fuel Form		Mass Kilograms	Thermal Power Kilowatts	Reference
Metal	Pu-239 - bare	16.5	NA	Koch and Paxton Ann. Rev. Nuc. Sci. Vol. 9 1959
	Pu-239 - reflected	5.8	NA	
	Pu-238 - bare	7.0	3.3	Reference 45
	Pu-238 - reflected	4.0	1.9	Stubbins et al.
	Pu-238 - bare	5.6	2.6	HW-84369
	Pu-238 - reflected	3.0 ?	1.4 ?	
Plutonium dioxide				
	Pu-238 O ₂ Bare	14.9	6.9	Reference 45,
	Pu-238 O ₂ Reflected	10.5	4.8	Stubbins et al.
	Pu-238 O ₂ Bare	11.9	5.5	Estimated from
	Pu-238 O ₂ Reflected	7.9	3.6	HW-84369

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As noted previously, the compatibility of the dioxide fuel with capsule and coating materials has been studied but the data are limited with respect to operating temperatures, exposure times, and materials. In a later paragraph the reported data are discussed and tabulated. This remains a critical problem area.

The need to contain the fuel under all credible operational and accident conditions presents very practical problems in the case of Pu-238. The desired containment time is ten half-lives, about 900 years. It is unrealistic to expect that data on corrosion resistance and mechanical stress can be extrapolated to time periods, orders of magnitude greater than the empirically established information. As discussed below the preliminary studies indicate that none of the capsule materials are capable of containing the helium pressure for more than ten years. This conclusion leads to the vented capsule which itself presents problems, especially the solubility of the fuel, which must be further investigated.

5.3.6 DEVELOPMENT AREAS

The oxide fuel forms require much additional investigation to establish their use as the Pu fuel form for the Large Heat Source. There are deficiencies in the fundamental physical data. Quantity production methods for determining physical forms to specified tolerances on size, density, and shape must be developed. The long term stability of the oxide to the various stresses and internal changes must be undertaken promptly since it is doubtful that any "accelerated" testing can provide accurate simulation of the long term effects. It is possible that a surrogate for Pu-238 might be used but the only materials which appear useful are Pu-236, Pu-241, and Cm-242. There are various technical objections to the use of these materials in addition to the difficulty of obtaining them in a sufficient quantity and purity to be effective. Retention of helium in the proposed cermet fuel form requires more study.

As noted previously, data on compatibility are limited. The behavior of various candidate materials for coating and primary fuel cladding in contact with the oxide require extensive experimental work at temperatures up to the maximum expected for the emergency operational conditions: around 2300° F.

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Cross-section data for fission, absorption, and scattering for Pu-238 have been determined for some portions of the neutron energy spectrum. Empirical determination of the resonance cross-sections and verification of the cross-sections already reported should be undertaken as soon as possible.

Current criticality studies rely on normalized values based on the resonance cross-sections of Pu-240. The value of the coefficient for the average number of neutrons produced per fission, $\bar{\nu}$ (nu bar) is known to vary significantly with the neutron energy. It is of obvious practical importance to determine the correct value which should be used in determinations of reactivity. The selection of a material which can effectively damp the fissile reactivity of the source depends on many factors which have not yet been critically evaluated. Criticality experiments using the normal production grade of Pu-238 with its usual isotopic impurities should be performed in thermal, intermediate and fast critical assemblies. The possible reduction of the neutron contribution from the (α, n) by elimination of light element impurities through more selective processing should be considered. ANL has indicated the ability to produce Pu whose total non-isotopic impurities are less than 50 ppm (Reference 47). In addition, the use of oxygen-16 for the synthesis of the oxide should be reviewed in the context of the mission weight capability and the cost of providing reagents and controlled atmosphere having the O-17 and O-18 depleted by a factor of 10 or more (Reference 48).

5.3.7 ALTERNATE FUEL FORMS

The preliminary choice of the dioxide in a metal matrix as the fuel form reflects the state-of-the-art today. The development of plutonium reactor fuels may well provide significant advances applicable to isotopic fuels and much more attention should be given to these developments. The carbide and nitride technology offer particular promise. In addition, at least one other binary compound plutonium sulphide deserves serious consideration. The experience at ANL with the uranium sulphide congener indicates some very attractive properties (Reference 26).

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Little consideration has been given to other than binary compounds of Pu, generally because more complex compounds tend to dissociate more readily. In view of the need to provide diluent/poison to suppress reactivity, further consideration of the more complex compounds appears warranted.

As previously noted, the alloys of Pu offer little promise of yielding a high temperature fuel form. Some consideration should be given to the possible alloys with refractories like rhenium. By analogy to the Pu-Zr alloy, the Pu-Hf alloy should exhibit desirable properties and have the additional advantage of providing in situ, one of the best diluent/poison materials obtainable.

5.4 CURIUM-244 AND ITS CHARACTERISTICS

Of the three isotopes under consideration for the Large Heat Source, the least information is available for curium-244. Data on the characteristics of Cm-244 are still at the research level. Nuclear and radiation properties are reasonably well-known with some very important exceptions (Reference 49). The characteristics and behavior of the elemental material and of the putative binary compounds with B, C, N, O, F, Si, P, S, Cl, and Br are extrapolated from the lanthanide and actinide homologs (Reference 50). No information is reported on the possible complex compounds and the alloys.

Experimental data are extremely meager and currently extended to some initial information on the oxides and the fluoride (Reference 50 and 51). Consequently, even the tentative and preliminary discussion here and in the report on "Preliminary Fuels Data" are subject to significant revision as new empirical data become available.

Curium-244 is produced by five successive neutron captures starting with Pu-239, as the target. The yield from a typical irradiation is shown in Table 5-6, (Reference 52), and the build-up of Cm-244 from the target material versus time is shown in Figure 5.2

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TARGET MATERIAL: Pu-239
FLUX: 3×10^{14} n-cm⁻¹-sec⁻¹

Figure 5-2. Yield of Cm-244

Curium and its Compounds

Work on elemental curium metal has been carried on by a number of investigators (References 53, 54, and 55). Curium metal, unlike Po-210 and Pu-238, has a reported melting temperature, $1340^{\circ}\text{C} \pm 40^{\circ}\text{C}$ (2450°F), sufficiently high to permit use of metal directly as a high temperature heat source.

No data are reported for the thermal properties; the density is estimated from the lattice parameters as around 13 g-cm^{-3} . Compatibility with encapsulating materials is as yet undetermined.

Two oxides of curium have been identified: CmO_2 and Cm_2O_3 . Recent experimental work by Butler, McHenry et al at ORNL (ORNL-TM-1036) has confirmed the earlier work of Asprey et al (Reference 56), that the dioxide is unstable in neutral or reducing atmospheres

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above 600°C. The sesquioxide has been shown to be the stable form and has a melting temperature around 1950°C (3542°F). The sesquioxide, Cm_2O_3 is expected to behave like its lanthanide and actinide congeners. Experimental data on stability, compatibility, solubility and other chemical-physical properties and reactions are lacking. The initial tests with iridium cermets and with W plus 2 percent Re alloy capsules are of such short duration and are for such small quantities of material that definitive conclusions are not warranted on the basis of the experiments. It is probable, however, that the curium oxide will react in a manner similar to plutonium oxide and that the technology developed for Pu will be generally applicable to Cm.

TABLE 5-6. YIELD OF CURIUM
(Reference 52)

Nuclide	Percent
Cm-242	0.02
Cm-243	0.03
Cm-244	95.5
Cm-245	1.6
Cm-246	2.7
Cm-247	0.04
Cm-248	0.04
Am-243	0.0009
Cf-252	1×10^{-4}

The synthesis of curium fluoride and its identification as CmF_3 has been reported. Early studies (Reference 57) had given a melting temperature of around 600°C (1112°F) which appeared anomalous when compared to other actinide trifluorides. The current investigation has shown the melting temperature to be consistent with that of PuF_3 , around 1400°C (2552°F). No other data are reported for the fluoride (Reference 51).

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5.4.1 PROBLEM AREAS

Like all the transuranium elements, curium-244 undergoes spontaneous fission; its S. F. half-life is around 1.4×10^7 years. Since no cross-section data for Cm-244 were available, the cross-sections have been calculated over the neutron energy spectrum from 0.001 ev to 10 Mev on the basis of nuclear theory and systematics. The results are shown in Figure A-5 and a detailed discussion is given in Appendix A. Although formal calculations of criticality have not yet been performed it appears probable that Cm-244 will be fissile to fast neutrons and will have a finite critical mass. The problem created by this is directly comparable to that for Pu-238. The problems of stability, and cladding and coating compatibility discussed for Pu-238 apply also to Cm-244.

The neutron production from spontaneous fission on Cm-244 is so large approximately 2.1×10^7 neutrons- $g^{-1} \text{ sec}^{-1}$, that the problems arising from neutron production via the (α, n) reaction involving light element impurities and O-17 and O-18 are of no consequence. However, as Table 5-6 shows the normal production of Cm-244 contains traces of Cf-252. Californium 252 has a spontaneous fission rate of around 10^{12} n-sec $^{-1}$ -g $^{-1}$ thus the contamination of Cm-244 by as little as 100 micrograms of Cf-252 per gram of Cm-244 would increase the neutron production by a factor of ten.

5.4.2 DEVELOPMENT AREAS

Development areas discussed for Pu-238 are generally applicable to Cm-244, but much more effort is required to achieve a comparable level of knowledge. Curium-244 until recently has been a laboratory material with no engineering applications, whereas plutonium has been under intensive study for years.

As indicated in the preceding section, the effort to reduce neutron production by the removal of the heavy oxygen isotopes is not significant for Cm-244.

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For curium-244 a thorough investigation of neutron cross-section, and determination of empirical values for ν over the spectrum are required. Criticality studies with the production materials are also needed.

Given the present state of curium technology, it appears premature to discuss alternate fuel forms. The planned experimental and development work at ORNL and the activities being carried on at Savannah River are directed to a comprehensive study of curium and its compounds. The results of these efforts may yield better fuel material than the metal and the oxide.

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5.5 NON-NUCLEAR MATERIALS

The non-nuclear materials discussed here include the fuel matrix and primary clad; the outer clad; the heat block materials, including the heat sink, structure, liquid metals if used to enhance internal heat transfer, spacers, supports and other miscellaneous pieces; external cladding, if used; and heat shielding to provide protection against re-entry heating. All these materials must satisfy at least the following general requirements:

- Be compatible chemically and mechanically with the materials with which they interface
- Maintain their physical, chemical, thermal, and mechanical properties within acceptable limits, over the entire range of the temperatures expected and under operational, environmental and credible accidental stress.

The Large Heat Source is a high temperature system and the materials used in it must be serviceable over the temperature range from ambient, 20°C (68°F) up to 1250°C (2282°F). The materials usable up to this maximum temperature are limited to a small group of base metals, the superalloys, the refractory metals and their alloys, and some ceramics. The number of candidate materials is further reduced by other considerations including:

- Strength
- Corrosion resistance: in liquid and gaseous media
- Compatibility
- Fabrication

5.5.1 COMPATIBILITY OF FUEL-MATRIX-CLADDING MATERIALS

It appears probable that all of the fuel compounds selected in this preliminary study will be used in metal matrices which will in turn be enclosed by the primary cladding. It is therefore necessary to establish the compatibility of the fuels with the matrix materials and of the fueled

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TABLE 5-7a. COMPATIBILITY TEST DATA FOR
GADOLINIUM POLONIDE (GdPo)

Matrix/Clading Materials	Exposure Time - Hours	Temperature °C °F	Atmosphere	Remarks
Tantalum	0.5	1850 3362	Hydrogen	Cladding failed
	2000	1000 1832	Hydrogen	Slight attack (0.004" penetration ?)
	1000	1800 3272	Hydrogen	With lead (Po-210 decay product) only, no attack
Tungsten + 25% Rhenium	1	1700 3092	Hydrogen	With free gadolinium metal: slight attack; no Po
Rhenium	1	1700 3092	Hydrogen	With free gadolinium metal: slight attack; no Po

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TABLE 5-7b. COMPATIBILITY TEST DATA
FOR PLUTONIUM DIOXIDE (PuO₂)

Matrix/Cladding Material	Exposure Time (hr)	Temperature °C	°F	Atmosphere	Remarks
Stainless Steel					
AISI - 304	2	1177	2150	hydrogen	No Reaction
316	2	1177	2150	hydrogen	No Reaction
318	2	1177	2150	hydrogen	No Reaction
347	2	1177	2150	hydrogen	No Reaction
Inconel	2	1177	2150	hydrogen	No Reaction
L-605 (Haynes 25)	8000	1000	1832	oxygen?	No Reaction
Zircaloy-2	2	1093	2000	argon	extensive reaction, discoloration and crumbling
Molybdenum	4	1593	2900	hydrogen	None
	20	1371	2500	hydrogen	None
Tungsten	4	1593	2900	hydrogen	apparent reaction due possibly to formation and volatilization of WO ₃
	20	1371	2500	hydrogen	
Niobium	4	1593	2900	argon	no apparent reaction
	0.1	2300	4172	argon-helium	no apparent reaction
Vanadium	4	1593	2900	argon	no apparent reaction
Titanium	4	1593	2900	argon	microscopic precipitate at PuO ₂ -Ti interface
Chromium	4	1316	2400	hydrogen	No apparent reaction
Tantalum	7	1704	3100	argon	No apparent reaction
Al ₂ O ₃	4	1593	2900	hydrogen	PuAlO ₃ formation
MgO	4	1593	2900	hydrogen	No Reaction
ZrO ₂	4	1593	2900	hydrogen	PuO ₂ -ZrO ₂ solid solution formation
Th	1.5	1000	1832	vacuum	β Pu ₂ O ₃ formation complete
U	2	900	1670	vacuum	β Pu ₂ O ₃ formation complete
Zr	?	1000	1832	vacuum	some reduction and ZrO ₂ formed
Mg	2	500	932	vacuum	α Pu ₂ O ₃ formation complete

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TABLE 5-7c. COMPATIBILITY TEST DATA
FOR CURIUM OXIDE (Cm_2O_3)

Matrix/Cladding Materials	Exposure Time - Hours	Temperature		Atmosphere	Remarks
		$^{\circ}\text{C}$	$^{\circ}\text{F}$		
Iridium	4	2000	3632	Oxygen	No evidence of intergranular attack

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matrix with the primary clad. Table 5-7 shows the results of compatibility tests on the various matrix and clad materials exposed to the three selected fuel compounds: GdPo, PuO_2 , and Cm_2O_3 . It is evident from the table that the data are with few exceptions short-term and do not provide a satisfactory answer to the problems of fuel-matrix-clad compatibility. The work thus far reported, however, provides a sound basis for planning a comprehensive program for compatibility testing. In addition, it is important that any testing program realistically simulate not merely the static temperatures representing a fixed operating point but also the transient temperatures associated with a non-catastrophic excursion occasioned by temporary loss of cooling. Changes in the behavior of the super-alloys and the refractory metals are known to be triggered by such excursions and a previously non-reactive material can become reactive.

The effect of the decay product in-growth on compatibility must be determined by simulation with non-radioactive materials if necessary. For example, the in-growth of Pb-206 from the decay of Po-210 may affect the phase relationships of the GdPo and the possible interaction of the Pb-Gd-Po-Ta system should be determined experimentally. Build-up of the decay products of Pu-238 and Cm-244 is much slower and is probably of less consequence, because the decay products as long lived members of the actinide group should form compounds homologous with the starting materials.

The effect of impurities in the fuels and in the matrix and cladding materials is not usually considered but may be of direct engineering significance. Large amounts of impurities are known to change thermal conductivity, expansion coefficient, vapor pressure, and melting temperature. The impact on compatibility is difficult to guess. The behavior of the oxides in neutral atmospheres and in oxygen or air is considerably different. For this reason compatibility testing should simulate the internal helium atmosphere to insure that the test conditions are representative of the real situation.

Preliminary choice of matrix and primary cladding materials has been made on the basis of the compatibility test results. For Po-210, the matrix and primary cladding is tantalum. For $^{238}\text{PuO}_2$ the material with the most extensive history is L-605 alloy (Haynes 25) which is chosen for matrix and primary clad. TZM alloy (Mo + 0.5% Ti + 0.5% Zr) is an alternate.

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In the absence of firm data it is assumed that $^{244}\text{Cm}_2\text{O}_3$ will use the same materials as PuO_2 .

5.5.2 OUTER CLADDING (CAPSULE) MATERIALS

The outer cladding must provide a number of characteristics including:

- High impact and creep strength at operational temperatures of 1000°C (1832°F) throughout design mission life and for short periods up to 1250°C (2282°F).
- Corrosion resistance in air and water
- Compatibility with primary clad and with the heat block
- Fabricability in a hot cell/glove box environment

To achieve these objectives the outer cladding may be a multiple-layer capsule using a refractory metal for high strength and a superalloy for oxidation resistance (see Section 4). Refractory metals including Nb, Mo, Ta, W, and Re and their alloys provide high creep strength as shown in Figures 5-3 and 5-4. Other properties of these materials are shown in Table 5-8. The refractory metals and alloys are quite susceptible to oxidation and must be protected from contact with air and other oxidizing media above the temperatures shown in Table 5-8 and Figure 5-5. Adherent ceramic coatings which are frequently used do not appear adequate for this application because of their inability to withstand impact and thermal cycling. Consequently, when refractories are used, they must be protected by a complete outer clad. Noble metals such as platinum were considered but because of the finite reciprocal solid solubilities of Pt and the refractory metals at temperatures as low as 1000°C (1832°F), were rejected as protective cladding. The superalloys whose compositions are shown in Table 5-9 and whose strength with temperature is shown in Table 5-10 appear as promising candidates for the protective clad. The oxidation resistance of Haynes 25 is good up to the allowable maximum temperature of 1250°C (2282°F) (See Figure 5-5). Its good resistance to thermal fatigue and acceptable impact strength at lower temperatures: 1000°C (1832°F), should provide a high integrity cladding under impact conditions. Little is known about the long term compatibility between Haynes 25 and refractory metals. Preliminary indications of migration of some alloy constituents into the refractory metal, identify a need for

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TABLE 5-8. PROPERTIES OF REFRACTORY METALS

Element	Melting Point (°F)	Density (gm-cm ⁻³)	Recrystal- lization Temperature (°F)	Temperature Limit in Air (°F)
Niobium	4350	8.57	1785-2100	750
Tantalum	5432	16.6	2200-2400	570
Molybdenum (TZM)	4730	10.2	2500-2600	930
Tungsten	6170	19.3	2200-3000	700
Rhenium	5738	21.0	2200-2750	1100

TABLE 5-9. TYPICAL CHEMICAL COMPOSITION OF SELECTED HIGH TEMPERATURE SUPERALLOYS (%)

Element	Haynes 25	19-9 DL	Hastelloy C	Hastelloy X	Hastelloy N	Rene 41
Aluminum	—	—	—	—	—	1.5
Carbon	0.10	0.30	0.08	0.10	0.10	0.10
Chromium	20.0	19.0	15.0	22.0	7.0	19.0
Cobalt	Balance	—	2.5	—	—	11.0
Columbium	—	0.40	—	—	—	—
Iron	1.5	Balance	5.0	18.0	5.0	5.0
Manganese	1.6	0.5	0.5	0.5	0.5	0.1
Molybdenum	—	1.5	16.0	9.0	17.0	10.0
Nickel	10.0	9.0	Balance	Balance	Balance	Balance
Silicon	0.5	0.5	0.5	0.5	0.5	0.5
Titanium	—	0.3	—	—	—	3.0
Tungsten	15.0	1.5	4.0	1.0	—	—

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TABLE 5-10. TYPICAL MECHANICAL PROPERTIES OF SELECTED HIGH TEMPERATURE SUPERALLOYS (%)

Tensile Ultimate Strength (psi)						
Temperature	Haynes-25	19-9 DL	Hastelloy C	Hastelloy X	Hastelloy N	Rene 41
RT	146,000	120,000	121,000	110,000	110,000	196,000
1000	116,000	89,000	99,000	95,000	90,000	175,000
1200	103,000	75,000	97,000	85,000	80,000	170,000
1400	66,000	43,000	79,000	60,000	60,000	140,000
1600	47,000	--	56,000	36,000	47,000	80,000
Elongation (%)						
Temperature	Haynes-25	19-9 DL	Hastelloy C	Hastelloy X	Hastelloy N	Rene 41
RT	64	55	48	40	51	14
1000	59	43	52	40	47	14
1200	35	34	55	37	38	14
1400	12	36	59	40	25	11
1600	30	--	47	50	25	19
Charpy "V" Impact (ft-lbs)						
Temperature	Haynes-25	19-9 DL	Hastelloy C	Hastelloy X	Hastelloy N	Rene 41
RT	160	46	21-23 (Izod "V")	50	85	--
1000	170	53	--	--	90	--
1200	160	52	--	--	80	--
1400	140	56	--	--	75	--
1600	110	--	--	50	70	--

References: (1) "High Temperature Alloys Handbook," Union Carbide Stellite Co.

(2) "High Temperature Alloys Handbook," Universal Cyclops Steel Corp.

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Figure 5-3. Temperature Vs. Tensile Strength

Figure 5-4. Ten-thousand-hour Rupture Strength of Alloys

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MATERIALS SELECTION

Figure 5-5. Comparative Oxidation Characteristics of Materials in Air

further investigation and may require the development of a satisfactory diffusion barrier which does not impose an unacceptable thermal barrier.

Long-term corrosion resistance in water is particularly difficult to assess. Only limited data are available for the sea water corrosion rates of the superalloys. Table 5-11 shows the available data. The data for Stellite 6 are thought to be representative of the behavior of L-605 (Haynes 25) although L-605 contains 20 percent chromium vs 30 percent in stellite 6 and L-605 contains 15% W vs 5% W in Stellite 6.

Whether or not the local corrosion in cracks and crevices experienced with this alloy would also occur with L605 cannot be stated with any assurance.

Contact with personnel at International Nickel Company revealed that they have not tested L605. To the best of their knowledge, no sea water data are available for this alloy.

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Galvanic Corrosion

When dissimilar metals are in close proximity, accelerated corrosion may occur if they are in contact through an electrolyte. The amount of corrosion depends upon the potential between the two metals. In general, the farther apart the two metals are in the galvanic series, such as shown in Table 5-12, the greater the corrosion. The metal near the anodic end of the series will be the one that corrodes.

Table 5-11. Corrosion Rates in Sea Water for Cobalt and Nickel Base Alloys

	Form	Exposure Time (yr)	Corrosion Rate (in/yr)	Depth of Pitting (in)	Remarks
Hastelloy C	Plate	6	0.00005	0.000	
Stellite 6 (similar to Haynes 25)	Plate	2	0.0001	0.0007 max.	Local corrosion in cracks or crevices

Table 5-12. Galvanic Series in Sea Water

1. Magnesium Alloys	11. Nickel (active)
2. Zinc	12. Brass
3. Aluminum	13. Copper
4. Cadmium	14. Titanium
5. Aluminum alloys	15. Monel
6. Low Carbon Steel	16. Silver Solder
7. Austenitic Stainless Steels (active)	17. Nickel (passive)
8. Hastelloy C	18. Austenitic Stainless Steel (passive)
9. Lead	19. Silver
10. Tin	20. Graphite

It is apparent that an alloy such as Hastelloy C may be subject to galvanic corrosion because of the many and varied metals which are more cathodic. If vehicle structural materials are also considered, the problem becomes more complex since sacrificial magnesium and

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aluminum alloys will be present as well as additional titanium and stainless steels. Impact with the ocean will probably result in disintegration in the vehicle so this may not be a problem. The place of L-605 in this series is not defined.

Another potential problem area, considering the possibility of several hundred years exposure to sea water, is the galvanic potential between the base alloy and a weld metal deposit. If the weld is more anodic than the capsule, concentrated corrosion could occur at a localized area within the weld. Essentially, no data exist to define this problem.

As noted in Table 5-9, some alloys contain chromium which is preferentially depleted in vacuum under operating temperatures. Test data on the depletion from Haynes 25 at 1750^oF indicates a loss rate which decreases with time. After five years operation, the depth of loss is in the order of one to five mils. The use of a coating as described below, however, will inhibit chromium depletion.

Capsule surface emissivity is an important factor in determining clad operating temperature; if heat transfer is primarily by radiation. In general, some form of surface coating must be employed to increase emittance. For example, the emittance of a cleaned Haynes-25 surface is about 0.25 at the fuel capsule clad operating temperature. A surface treatment in which the Haynes-25 is cleaned, silicon carbide blasted, oxidized in a wet hydrogen atmosphere for four hours at 1000^oC, followed by oxidation in still air for 2 hours at 1000^oC, was developed at General Electric.* The resulting oxidized surfaces possess total hemispherical emittances in excess of 0.8. Coatings are stable in high vacuum at temperatures below 1050^oK. At higher temperatures, the emittance could be expected to decrease from values above 0.8 to somewhere between 0.70 and 0.75. Stability of the oxide coating can be enhanced by introducing a small quantity of titanium or zirconium oxide to form interoxidic compounds with the substrate metal oxides. Tests indicate that such coatings possess emittances in excess of 0.85 at temperatures above 1250^oK in high vacuum.

*SNAP-17B Power Supply Development Program (Final Report - Appendix B) GEMS-NS-102

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All five refractory metals can be joined satisfactorily using mechanical fasteners, brazing, and resistance, fusion, or electron beam welding. Good ductility is retained in the fusion welded joints of Nb, Ta, and high Re alloys of Mo and W. Mo and W, however, generally produce coarse grained fusion welds. There is a tendency for Mo and W welds to crack when heavy sections are joined. The problems of handling refractory metals are intensified by the need to perform the operations in the hot cell/glove box environment. Techniques including autoclaving, magnaforming, and other pressure methods for obtaining a diffusion bond may be satisfactory for the refractory claddings. Extensive GE experience with refractory metal reactor fuel elements indicates the potentialities of these methods.

5.5.3 HEAT BLOCK MATERIALS

Two general heat block concepts have been evolved in this study. These are described in detail and their relative merits are discussed in Section 4.1. The modular concept consists of a graphite torus sheathed in Haynes alloy into which the capsules are fitted in individual cylindrical holes. Ceramic supports and spacers prevent the capsules from touching the graphite and the graphite from touching the Haynes 25 sheath. The voids around the capsules and between the graphite and the external sheath are filled with liquid sodium.

The cell concept uses a niobium alloy (Nb +1% Zr) as the capsule support structure and as an integral heat exchanger. Relevant properties of this alloy and other candidate materials appear in Table 5-13. The capsules are inserted into the structure but ceramic spacers prevent direct contact between the Haynes 25 capsules and the niobium alloy. The possible compatibility problem between superalloy and refractory metal mentioned previously is thus avoided. Heat transfer in this structure is by radiation.

Sodium, lithium and NaK produce grain boundary corrosion in the superalloys at 1000°C (1832°F). Direct contact between the capsule and the graphite block must also be avoided because of the severe intergranular carburization that will occur. The compatibility problems among the constituents of the modular concept heat block are discussed in detail in Appendix B.

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Table 5-13. Fuel Block Material Data

	(Hot Pressed Block) Beryllium	Graphite	TZM Molybdenum	Tungsten	Ta-10W	Haynes 25	Hastelloy C	Udimet 700	Niobium
K-Thermal Conductivity Btu/hr/sq ft/°F/ft	@ 1800°F 39.6	@ 1800°F 15	@ 1652°F 61.2	@ 1800°F 66.5	@ 1800°F 28.8	@ 1600°F 15	@ 1600°F 12	@ 1600°F 14.1	@ 1600°F 35
ρ density - lb/ft ³	115.8	112.2	634	1203	1050	570	558	494	535
K/ ρ	0.342	.134	.0965	.0548	.0274	0.0264	0.0216	0.0286	0.065
Melting Point °F	2345	3700	4730	6170	5495	2425-2570	2320-2380	2300-2600	4350
Cp Btu/lb/°F	@ 1800°F .79	@ 1800°F .45	@ 1800°F .066	@ 1800°F .043	.043	0.092	0.092	0.10	@ 1800°F 0.08
Latent Heat of Fusion Btu/lb	470	-	-	-	-	-	-	-	-
Impact Str. Ft-Lb					Charpy Keyhole @ 900°F 143	Charpy @ 1600°F 120	Izod V @ 100°F 22		-
Coefficient of linear expansion 10 ⁻⁶ in/in/°F	R. T. -1600°F 9.0	R. T. 0.6-2.2	R. T. -1652°F 3.36	R. T. -1600°F 2.7	R. T. -1600°F 3.8	R. T. -1600°F 9.1	R. T. -1600°F 8.3	R. T. -1800°F 9.8	R. T. -1800°F 4.3
Ult. Tensile Strength (psi)	@ 1200°F 24,000		@ 1800°F 90,000	@ 1800°F 60,000	@ 1800°F 80,000	@ 1800°F 70,000	@ 1500°F 44,000	@ 1800°F 52,000	@ 1800°F 60,000
Modulus of Elasticity (psi x 10 ⁻⁶)	@ 1200°F 28.0		@ 1800°F 27.8	@ 1800°F 38	@ 1800°F 17.0	@ 1600°F 24.0	@ 1600°F 22.0	@ 1500°F 23.5	@ 1500°F 14

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5.5.4 RE-ENTRY PROTECTION MATERIALS

The individual fuel capsules or the entire heat block must be protected against the effects of re-entry heating. There is wide experience in the protection of re-entering bodies which are themselves at low initial temperatures. The protection of a body like the fuel capsule or the heat block which may be at 1000°C (1832°F) initially, presents a new range of problems.

Beryllium has been used for re-entry protection and has been proposed for use on bodies whose initial temperatures are around 400°C (752°F). Consequently, Be was evaluated as a possible heat block material and as a final clad for the capsules and for the heat block. A brief investigation, however, disclosed that Be is highly reactive with other metals and above 800°C (1472°F) the reactions progressively accelerate. Moreover, Be has a high vapor pressure in vacuum at 850°C and above. The current status of beryllium for high temperature applications is reviewed in Appendix C.

Transpiration cooling of rocket nozzles by impregnating their tungsten structures with copper and silver-copper alloys has proved quite effective. A similar approach is under investigation for application to the individual capsule or the heat block. Foamed materials in a variety of metals and ceramics and in a wide range of porosities and effective densities have become available. Table 5-14 shows a few of these materials with the pore sizes and densities available. These low density structures can be infiltrated with salts having large heats of vaporization, to approximately 90 percent of the available voids. Aluminum trifluoride, AlF_3 , which sublimates at 1270°C (2318°F) with a heat of transition of $76 \text{ kcal-mole}^{-1}$ is a possible infiltrant. Other salts and mixtures of salts are under study. Additional investigations of how this foamed and infiltrated material can be applied integrally to the capsules and heat blocks are now in progress.

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Table 5-14. Foamed Available Structures

<u>Material</u>	<u>Pore</u>	<u>Sizes</u>		<u>Nominal(inches)</u>		<u>Density Range</u>
	.010	.020	.050	.080		
Stabilized Zirconia		x	x	x		4-15%*
Alumina		x	x	x		4-15%
Nickel	x	x	x	x		4-15%*
Copper	x	x	x	x		4-15%
Nichrome V		x	x			4-15%
Hastelloy X		x	x			4-15%
Molybdenum		x	x			4-15%
Tungsten		x	x			4-15%
Tantalum	x	x	x			4-15%

*Eighty mil pore size results in about 3-4% density. As pore size decreases, density increases. Ten mil pore size can be produced in densities from 7-15% of theoretical.

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APPENDIX A.

NEUTRON CROSS-SECTIONS FOR CM-244

A.1 INTRODUCTION

Neutron cross-sections for Cm^{244} have been calculated from thermal to 14 mev using a combination of nuclear theory and systematics.

This report discusses the possible reactions of neutrons with Cm^{244} , the theoretical assumptions made along with their justification and reasons for choosing certain recommended values.

A.2 DETAILED ANALYSIS

Table A-1 gives the thresholds for the various reactions of neutrons with Cm^{244} up to 15 mev.

Table A-1. (Ref. 1)

Reaction	Threshold (mev)
n, γ	exothermic
n, n'	0.043
n, 2n	6.67
n, 3n	12.36
n, p	0.72
n, d	3.65
n, t	3.69
n, α	exothermic

The first excited state of Cm^{244} is at 43 kev, thus inelastic scattering ($\sigma_{nn'}$) does not occur below this energy. (Atomic effects in the lower region are not included.)

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The cross-sections in the range thermal to 14 mev are given as

$$\sigma_T = \sigma_{\text{non}} - \sigma_{\text{el}}$$

$$\sigma_{\text{non}} = \sigma_{\text{nn'}} + \sigma_{\text{nf}} + \sigma_{\text{n}\gamma} + \sigma_{\text{n2n}} + \sigma_{\text{n3n}}$$

where the (n, p), (n, α) etc reactions are considered to be small due to Coulomb barrier effects.

A.3 RESONANCE PARAMETERS

The resonance parameters for Cm^{244} were taken from the recent work of Cote² et al

Table A-2.

E_R (ev)	Γ_n (10^{-3} ev)	Γ_γ (10^{-3} ev)	Γ (10^{-3} ev)
7.73	10.3 ± 0.5	37.2 ± 3.3	47.5 ± 2.5
16.9	2.0 ± 0.05	37.0 ± 3.0	39 ± 3.0
22.9	0.97 ± 0.05		
35.0	4.03 ± 0.20	43.5 ± 8.0	47.5 ± 8.0
52.8	0.70 ± 0.05		
69.9	0.48 ± 0.06		
86.0	20.0 ± 1.3		
96.0	6.2 ± 3.1		
133.0	15.1 ± 1.8		
182.0	8.8 ± 2.4		
197.0	22.0 ± 2.7		
211.0	42.7 ± 3.8		
222.0	39 ± 4		
231.0	18.3 ± 3.0		
273.0	43 ± 14		
360.0			
419.0			
445.0			
516.0			
650.0			
768.0			

Cote² et al obtained an s wave strength function $S_0 = \frac{\langle \Gamma_n^0 \rangle}{\langle D \rangle} = 0.76 \frac{+0.35}{-0.17} \times 10^{-4}$ for Cm^{244} and a $\Gamma_\gamma = 37 \times 10^{-3}$ ev.

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Using the resonance parameters in Table A-2, an infinitely dilute resonance integral for the resolved resonances was computed by a single-level Breit-Wigner approximation

$$I_{\infty, x} = \int_{E_c}^{\infty} \sigma_x \frac{dE}{E} \quad (1)$$

where

$$\sigma_x(E) = \frac{0.65 \times 10^6}{\sqrt{E}} \sum_i \frac{g_i \Gamma_{ni} \Gamma_{\gamma i}}{\sqrt{E_{Ri}} \left[(E_{Ri} - E)^2 + \left(\frac{\Gamma_i}{2} \right)^2 \right]} \quad (2)$$

where $x = c$ (radiative capture)

$x = f$ (fission)

i = resonance number

g_i = statistical weight factor

Γ_n = neutron width

Γ_{γ} = capture width

Γ_f = fission width

$\Gamma = \Gamma_f + \Gamma_n + \Gamma_{\gamma}$ = total width

E_R = resonance energy

Assuming a bound level at $E = -15$ ev then a total absorption integral ($I_{\infty a}$) was found to be

$$I_{\infty a} = 780 \text{ barns}$$

where the absorption cross section is given by $\sigma_a = \sigma_{n\gamma} + \sigma_{nf}$

A.4 LOW ENERGY CROSS SECTION ($0.001 \text{ ev} \leq E < 1.0 \text{ kev}$)

A.4.1 CAPTURE CROSS SECTION

Using Eq. 2 and resonance parameters from Table A-2, yielded a value of $\sigma_{n\gamma} = 9.9$ barns at $E = 0.0253$ ev due to the resolved levels. An additional 0.1 barn was assumed to be contributed by the higher levels. Thus

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$$\sigma_{n\gamma} = 10 \text{ barns}$$

This value was compared to quoted values given in the literature

BNL 325 -	$\sigma_{n\gamma} = 15 \pm 10 \text{ barns}$
A. H. Jaffey ³ -	$\sigma_{n\gamma} = 20 \text{ barns}$
C. M. Stevens ⁴ -	$\sigma_{n\gamma} = 25 \pm 10 \text{ barns}$
S. E. Vandenbosch ⁵ et al	$\sigma_{n\gamma} = 32 \text{ barns}$

These values refer to pile neutrons, therefore the capture cross section at $E = 0.0253$ should be somewhat smaller.

When the -15.0 ev level was taken into consideration ($\Gamma_n = 11.55 \times 10^{-3} \text{ ev}$) then

$$\sigma_{n\gamma} = 12 \text{ barns}$$

From 0.001 ev to the first resonance the capture cross section was assumed to follow the

$\frac{1}{v}$ law given by

$$\sigma_{n\gamma}(E) = \frac{\sigma(0.0253) \sqrt{0.0253}}{\sqrt{E}} \quad (3)$$

In the energy region from 30 ev to 1 kev the resonance structure of the cross-sections were ignored and treated as constant with $\sigma_{nf} = 25 \text{ mb.}$

$$\sigma_{n\gamma} = 0.2 \text{ b.}$$

$$\sigma_T = 9.5 \text{ b.}$$

(This is analogous to the treatment of Pu^{240} by J. S. Story et al⁶)

A.4.2 FISSION CROSS-SECTION

From Hyde⁷ (Volume III, p. 50) Cm^{244} shows a positive energy difference between the binding energy B_n and the activation energy E_a where

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$$E_a = E_b - 0.9 \text{ mev} \quad (4)$$

and

$$E_b = \left(19 - \frac{0.36 Z^2}{A} + \epsilon \right) \text{ mev} \quad (5)$$

For Cm^{244}

$$\begin{aligned} B_n &= 5.642 \text{ mev (Hillman)}^8 \\ E_b &= 5.86 \text{ mev} \\ E_a &= 4.96 \text{ mev} \end{aligned}$$

$$\therefore B_n - E_a = 0.682 \text{ mev}$$

Thus Cm^{244} should have a thermal fission cross-section greater than 1 barn.

With this information it was possible to calculate a quantitative thermal fission cross section from Hyde (Volume III, p. 58).

$$\log \frac{\sigma_{nf}}{\sigma_{n\gamma}} = -1.2 \quad \text{for } B_n - E_a = 0.7$$

since $\frac{\sigma_{nf}}{\sigma_{n\gamma}} = \frac{\Gamma_f}{\Gamma_\gamma}$

then $\sigma_{nf} = 0.1 \sigma_{n\gamma}$ at thermal energy which yields $\sigma_{nf} = 1.2$ barns ($\sigma_{n\gamma} = 12$ b).

This value of σ_{nf} is consistent with Vandenbosch and Seaborg's analysis.⁹ Assuming

$$\alpha = \frac{\sigma_{n\gamma}}{\sigma_f}$$

to be constant, then σ_{nf} is expected to follow the $1/v$ law of $\sigma_{n\gamma}$ up to the first resonance.

A.4.3 ELASTIC CROSS SECTION σ_{e1} $0.001 \text{ ev} < E < 7 \text{ ev}$

This cross-section is assumed to be constant at $\sigma_{e1} = 10$ barns which is somewhat higher

than that quoted by Summer¹⁰ for U²³⁸ in the same energy range, and approximately the same as that quoted for Pu²⁴⁰ by Parker et al, and that quoted by Joanou et al¹² for U²³⁸.

A.5 HIGH ENERGY CROSS-SECTIONS ($E \geq 1$ kev)

A.5.1 CAPTURE CROSS SECTION

Following the theory of Lane and Lynn¹³, the capture cross section averaged over many resonances is given by

$$\langle \sigma_{\gamma}(J, \ell) \rangle = 2 \pi^2 \frac{g_J}{\langle D(J) \rangle} \sum_j \left\langle \frac{\Gamma_n(J, \ell_j) \Gamma_{\gamma}(J)}{\Gamma(J)} \right\rangle \quad (6)$$

ℓ = neutron orbital angular momentum

j = entrance channel spin

$\langle D(J) \rangle$ = average spacing between resonances of given spin J

$\langle \rangle$ indicates an average overall resonances of spin J.

The strength function for s wave neutrons for U²³⁸ is $S_0 = \frac{\langle \Gamma_n^0 \rangle}{\langle D \rangle} = 0.94 \times 10^{-4}$
while for Cm²⁴⁴ $S_0 = 0.76 \times 10^{-4}$

Comparisons of the level structure of Cm²⁴⁴ and U²³⁸ as shown in Table A-3 suggest that $\langle D \rangle_{244} = \langle D \rangle_{238} = 18.5$ ev. (See Rosen¹⁴)

No attempt has been made to determine which levels are p wave.

Table A-3.

Resonance Levels (ev)

U ²³⁸ (Ref. 15)	Cm ²⁴⁴ (Ref. 2)
6.65	7.73
10.2	16.9
21.0	22.9

Table A-3. (Cont.)

Resonance Levels (ev)

U^{238} (Ref. 15)	Cm^{244} (Ref. 2)
36.4	35.0
66.1	52.8
81.0	69.9
90.0	86.0
103.0	96.0
117.0	133.0
146.0	182.0
165.0	197.0
190.0	211.0
209.0	222.0
etc.	etc.

Now $\Gamma_{\gamma}(U^{238}) = 25 \times 10^{-3}$ ev (Ref. 14)

$\Gamma_{\gamma}(Cm^{244}) = 37 \times 10^{-3}$ ev (Ref. 2)

Therefore, from Equation (6) since for s wave neutrons

$$\langle \sigma_{\gamma} \rangle \propto \frac{\langle \Gamma_n^0 \rangle}{\langle D \rangle} \Gamma_{\gamma} \quad (7)$$

(Γ_{γ} is constant)

then

$$\frac{\sigma_{n\gamma}(U^{238})}{\sigma_{n\gamma}(Cm^{244})} = \frac{s_o \Gamma_{\gamma}(U^{238})}{s_o \Gamma_{\gamma}(Cm^{244})} \quad (8)$$

or

$$\sigma_{n\gamma}(Cm^{244}) = 1.2 \sigma_{n\gamma}(U^{238}) \quad (9)$$

Another method of estimating the capture cross-section of Cm^{244} is in terms of the binding energy of the last neutron in Cm^{245} as compared to that of U^{239} .

$$\text{i.e., } \frac{\sigma_{n\gamma}(\text{Cm}^{244})}{\sigma_{n\gamma}(\text{U}^{238})} = \frac{5.642}{4.696} = 1.2$$

$$\text{or } \sigma_{n\gamma}(\text{Cm}^{244}) = 1.2 \sigma_{n\gamma}(\text{U}^{238}) \quad (10)$$

which is the same as that given in Equation (9). The capture cross-sections for U^{238} were taken from Parker¹⁶ and Gibbons¹⁷ et al

A.5.2 FISSION CROSS-SECTION

The fast fission cross-section as a function of energy is assumed to have the familiar step-like character shown in Figure A-1.

Figure A-1. Fast Fission Cross-section (σ) Versus Energy (E_{mev})

Figure A-1 shows that the fission cross-section increases suddenly at definite thresholds. The second step occurs when Cm^{244} undergoes an (n, n') reaction and is left with enough excitation energy to undergo fission. The next step occurs at the (n, 2n) threshold (fission threshold for Cm^{243}). By knowing σ_c (cross-section for compound nucleus formation) one can determine σ_{nf} . The fission cross-section at 3 mev for Cm^{244} was determined by 3 different methods

1. The $\frac{Z}{A}^{4/3}$ empirical relation (see Hyde Volume III, p. 73) gave a value $\sigma_{nf} = 2.5$ barns (Figure A-2)
2. At 3 mev the fission cross-section was assumed to be approximately equal to σ_c since $\sigma_{nn'}$ and $\sigma_{n\gamma}$ are very small and $\sigma_{n,2n}$ likewise this yielded $\sigma_{nf} = 2.57$ barns

Figure A-2. Variation of Plateau with $\frac{Z^{4/3}}{A}$

3. From Vandenbosch and Seaborg⁹

$$\sigma_{nf} = \sigma_c \left[\frac{\Gamma_f}{\Gamma_T} \right] = \sigma_c \left[\frac{1}{\frac{\Gamma_n}{\Gamma_f} + 1} \right] \quad (11)$$

$$\frac{\Gamma_n}{\Gamma_f} = 0.15, \quad \sigma_c = 2.95 \text{ b} \quad (\text{Optical Model Calculation})$$

$$\therefore \sigma_{nf} = \frac{2.95}{1.15} = 2.57$$

Comparing the 2-3 mev values of Pu^{238} , U^{238} and Pu^{240} as given in more recent literature (Refs 18-24) with that calculated for Cm^{244} permitted normalization by analogy to produce σ_{nf} up to 7 mev.

A similar normalization was made at 10 mev where

σ_{nf} for Cm^{244} was given by⁹

$$\sigma_{nf} = \sigma_c \left[\left(\frac{\Gamma_f}{\Gamma_T} \right)_{A+1} + \left(1 - \frac{\Gamma_f}{\Gamma_T} \right)_{A+1} \left(\frac{\Gamma_f}{\Gamma_T} \right)_A \right] \quad (12)$$

Values of $\frac{\Gamma_f}{\Gamma_T}$ for Cm^{245} were taken from Halpern²⁵

$$\sigma_{nf} (10 \text{ mev}) = 2.9 \text{ barns}$$

A similar calculation could be made at the third step ($E \sim 18 \text{ mev}$) using

$$\sigma_{nf} = \sigma_c \left[\left(\frac{\Gamma_f}{\Gamma_T} \right)_{A+1} + \left(1 - \frac{\Gamma_f}{\Gamma_T} \right)_{A+1} \left(\frac{\Gamma_f}{\Gamma_T} \right)_A + \left(1 - \frac{\Gamma_f}{\Gamma_T} \right)_{A+1} \left(1 - \frac{\Gamma_f}{\Gamma_T} \right)_A \left(\frac{\Gamma_f}{\Gamma_T} \right)_{A-1} \right] \quad (13)$$

where

$$\left(\frac{\Gamma_f}{\Gamma_T} \right)_{A-1} = \left(\frac{\Gamma_f}{\Gamma_T} \right)_{\text{Cm}^{243}} = 0.1 \text{ from Ref. 9.}$$

A.6 TOTAL, ELASTIC AND INELASTIC CROSS SECTIONS

The total, elastic and inelastic cross-sections were computed with a program ABACUS II,²⁶ based on the "Optical Model" of the nucleus²⁷ ABACUS II uses the "cloudy crystal ball" model of the nucleus to calculate the "shape elastic" scattering cross-section σ_{SE} and reaction cross-section σ_R (σ_c in this report) and the statistical theory of Hauser and Feshbach* to calculate the compound elastic σ_{cE} , and inelastic σ_{nn} , cross-sections.

*W. Hauser and H. Feshbach "Inelastic Scattering of Neutrons" Phys. Rev. 87 (1952) 366.

One also obtains the total elastic angular distribution, total elastic, total scattering, partial and total inelastic, and differential inelastic scattering cross-sections.

The optical model parameters chosen were those used by Wilmore and Hodgson²⁸ who following a suggestion by Perey and Buck²⁹ found equivalent local potentials to the non-local model of Perey et al.

The real well depth is given by

$$V_{RE} = 47.01 - 0.267 E - 0.00118 E^2 \quad (E \text{ in Mev})$$

The imaginary well depth

$$V_{IM} = 9.52 - 0.53 E$$

The real part of the spin-orbit potential

$$V_{SR} = 7.0 \text{ mev}$$

The well radius for the real well

$$R_1 = 1.322 - 0.00076 A + (4 \times 10^{-6}) A^2 - (8 \times 10^{-9}) A^3 \text{ fermi}$$

The imaginary well radius by

$$R_2 = 1.266 - 0.00037A + (2 \times 10^{-6}) A^2 - (4 \times 10^{-9}) A^3 \text{ fermi}$$

A = mass number

Well diffuseness

$$a = 0.66 \text{ fermi}$$

Imaginary well width

$$b = 0.48 \text{ fermi}$$

The form of the real non-spin dependent part of the nuclear potential had the familiar Saxon-Woods form, while imaginary part had the derivation Saxon-Woods form factor.

A spin orbit potential was included (real part only) and was of the Thomas form. (For a more complete treatment of the above the reader is referred to Ref. 30.)

The energy level structure of Cm^{244} used in the optical model calculations are shown in Table A-4. Also shown for comparison are the partial level structures of Pu^{240} , P^{238} and U^{238} .

Table A-4.

Cm^{244}		Pu^{238}		Pu^{240}		U^{238}	
E (kev)	J^π	E (kev)	J^π	E (kev)	J^π	E (kev)	J^π
1042	6+	1029	2+	1020	?	1039	2+
502	8+	514	8+	597	1-	679	1-
296	6+	303.6	6+	292	6+	308	6+
142.3	4+	145.07	4+	142	4+	148	4+
42.9	2+	44.07	2+	43	2+	45	2+
0	0+	0	0+	0	0+	0	0+

The similarity in level structure of these even-even nuclei in addition to their rotational collective status ($\sigma = 0, 2, 4, 6 \dots$) is a good indication that certain analogies employed in determining cross-sections are well founded. One sees in fact that the level structure of Cm^{244} and Pu^{238} are indeed close.

Figures A-3, A-4 and A-5 show the cross-sections for Cm^{244} as calculated for this report. While Tables A-5 and A-6 give the tabulated values. In general the difference in structure of Cm^{244} from that of say U^{238} or Pu^{240} is such that the detailed form of the partial cross-sections e.g., σ_{nf} , $\sigma_{n\gamma}$, σ_{nn} , will differ but the overall characteristics essentially are very similar.

It is also interesting to note that in the Mev region, the Cm^{244} total cross-section is smaller than that of U^{238} which has a smaller mass number.

Figure A-3. Low Energy Cross-Sections for Curium - 244

An example is at 3 mev $\sigma_T (\text{Cm}^{244}) = 6.743 \text{ b}$ while $\sigma_T (\text{U}^{238}) = 7.8 \text{ barns}$. However, if one looks at the values for the respective nuclear radii $R = r_0 A^{1/3}$ for

$$\left. \begin{array}{l} \text{U}^{238} \quad r_0 = 1.536 \\ \text{Cm}^{244} \quad r_0 = 1.499 \end{array} \right\} \text{Hyde Volume I, p. 250}$$

This systematic decrease of r_0 with increased Z and A tends to substantiate the optical model calculations.

Now the capture and fission cross-sections in the range 50 kev to 2 mev are significant and must be considered in the optical model calculations. Thus it was necessary to correct the Hauser-Feshbach calculations of the inelastic and compound elastic cross-sections by a correction factor

$$F = \left[1 - \frac{\sigma_{ny} + \sigma_{nf}}{\sigma_c} \right]$$

Figure A-4. Partial Inelastic Cross-sections for Curium-244

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Figure A-5. Cross-Sections for Curium²⁴⁴

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Table A-5. Inelastic Cross-Section of Cm^{244} Up to 2 Mev

E (Mev)		Energy in Mev of Excited States					Total $\sigma_{nn'}$
	0.043	0.1423	0.296	0.502	1.042	Barns	
0.05	0.4					0.4	
0.1	2.095					2.095	
0.2	2.592					2.592	
0.3	2.452	0.045				2.497	
0.4	2.076	0.094				2.17	
0.5	1.65	0.14				1.79	
0.6	1.299	0.18	0.0009			1.48	
0.7	1.06	0.208	0.003			1.271	
0.8	0.921	0.234	0.006			1.161	
1.0	0.789	0.285	0.0159			1.09	
1.25	0.7332	0.341	0.036	0.0001	0.0007	1.111	
1.5	0.6293	0.3407	0.0518	0.0004	0.0058	1.028	
2.0	0.3877	0.2478	0.0568	0.0019	0.0228	0.717	

Table A-6. Calculated Cross-Sections for Cm^{244}
From 1 kev to 14 mev

E (Mev)	σ_{nf} (b)	σ_e /(b)	$\sigma_{\text{nn'}}$ (b)	$\sigma_{\text{n}\gamma}$	$\sigma_{\text{n' } 2\text{n}}$ (b)	σ_{tot} (b)
0.001	0.00642	12.894		4.5		17.4
0.0015	0.0109	14.149		2.44		16.4
0.002	0.0154	14.185		1.8		16
0.0025	0.021	14.369		1.51		15.9
0.003	0.0279	14.285		1.387		15.7
0.0035	0.0361	14.164		1.3		15.5
0.004	0.0492	14.101		1.25		15.4
0.0045	0.0725	14.018		1.21		15.3
0.005	0.1135	13.887		1.2		15.2
0.0055	0.18	13.775		1.195		15.15
0.006	0.268	13.645		1.187		15.1
0.0065	0.345	13.535		1.17		15.05
0.007	0.413	13.457		1.15		15.02
0.0075	0.475	13.390		1.135		15
0.008	0.54	13.345		1.45		15
0.0085	0.592	13.308		1.1		15
0.009	0.641	13.264		1.095		15
0.0095	0.688	13.232		1.08		15
0.01	0.72	13.210		1.07		15
0.015	0.9	13.179		0.921		15
0.02	0.962	13.183		0.825		14.97
0.025	0.962	13.246		0.742		14.95
0.03	0.895	13.320		0.685		14.9
0.035	0.818	13.442		0.64		14.9
0.04	0.76	13.545		0.595		14.9
0.045	0.72	13.491	0.03	0.559		14.8
0.05	0.685	13.181	0.4	0.525		14.791
0.055	0.66	12.945	0.69	0.495		14.79
0.06	0.642	12.738	0.94	0.47		14.79
0.065	0.628	12.557	1.16	0.445		14.79
0.07	0.619	12.356	1.39	0.425		14.79
0.075	0.61	12.193	1.54	0.407		14.75
0.08	0.603	12.048	1.7	0.389		14.74
0.085	0.6	11.935	1.82	0.375		14.73
0.09	0.602	11.838	1.9	0.36		14.70
0.095	0.608	11.743	1.99	0.349		14.69
0.10	0.62	11.631	2.095	0.335		14.681
0.15	0.652	10.761	2.43	0.257		14.1
0.20	0.71	10.326	2.592	0.211		13.839
0.25	0.76	9.557	2.6	0.183		13.1
0.30	0.83	9.429	2.497	0.166		12.922

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Table A-6. Calculated Cross-Sections for Cm²⁴⁴
From 1 kev to 14 mev (Cont'd)

E (Mev)	$\sigma_{nf}(b)$	$\sigma_e(b)$	$\sigma_{nn'}(b)$	$\sigma_{n\lambda}(b)$	$\sigma_{n,2n}(b)$	$\sigma_{tot}(b)$
0.35	0.93	8.964	2.35	0.156		12.4
0.40	1.03	8.687	2.17	0.15		12.037
0.45	1.22	8.371	1.96	0.149		11.7
0.50	1.44	7.877	1.79	0.15		11.257
0.55	1.69	7.527	1.61	0.153		10.98
0.60	1.87	7.069	1.48	0.16		10.579
0.65	1.97	6.821	1.34	0.169		10.3
0.70	2.09	6.467	1.271	0.173		10.001
0.75	2.1	6.179	1.196	0.176		9.65
0.80	2.19	5.970	1.161	0.177		9.498
0.85	2.195	5.731	1.1	0.174		9.2
0.90	2.2	5.559	1.07	0.715		9.0
0.95	2.22	5.401	1.06	0.169		8.85
1.0	2.25	5.181	1.09	0.165		8.686
1.5	2.342	3.956	1.028	0.09		7.416
2.0	2.40	3.640	0.715	0.052		6.807
2.5	2.465	3.676	0.475	0.032		6.65
3.0	2.522	3.880	0.317	0.0245		6.743
3.5	2.53	4.270	0.181	0.0189		7.0
4.0	2.533	4.567	0.110	0.015		7.225
4.5	2.525	4.884	0.068	0.0128		7.5
5.0	2.535	5.007	0.045	0.011		7.598
5.5	2.533	5.108	0.0299	0.0096		7.68
6.0	2.53	5.040	0.021	0.00855		7.6
6.5	2.53	4.977	0.0154	0.00765		7.53
7.0	2.53	4.673	0.012	0.007	0.012	7.234
7.5	2.61	4.339	0.0098	0.00645	0.055	7.02
8.0	2.755	3.860	0.008	0.006	0.08	6.709
8.5	2.85	3.543	0.0072	0.00566	0.0945	6.5
9.0	2.917	3.242	0.0062	0.0054	0.109	6.28
9.5	2.96	3.009	0.00555	0.00513	0.12	6.10
10.0	2.985	2.775	0.005	0.00495	0.13	5.9
10.5	3.0	2.652	0.00453	0.00478	0.139	5.8
11.0	3.007	2.457	0.0042	0.0046	0.147	5.62
11.5	3.003	2.366	0.0039	0.00447	0.153	5.53
12.0	3.002	2.241	0.0037	0.00435	0.1585	5.41
12.5	3.0	2.211	0.00349	0.00425	0.161	5.38
13.0	3.0	2.108	0.0033	0.00418	0.165	5.28
13.5	3.0	2.035	0.00315	0.0041	0.168	5.21
14.0	3.0	1.971	0.003	0.004	0.17	5.148

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where σ_c (cross-section for compound nucleus formation) is taken from ABACUS II.

A.7 n-2n CROSS-SECTION

The n-2n cross-section was obtained by systematics using the relation

$$\sigma_{n,2n} = \sigma_{\text{non}} - (\sigma_{n\gamma} + \sigma_{nf} + \sigma_{nn'})$$

where the n, 2n threshold was taken as 6.67 mev. The n, 3n threshold of 12.36 predicated that this cross-section would be insignificant and was arbitrarily set equal to zero.

A.8 MEAN NUMBER OF NEUTRONS PER FISSION AND AVERAGE KINETIC ENERGY OF FISSION FRAGMENTS

Several values of $\bar{\nu}$ for Cm^{244} decaying by spontaneous fission have been reported (See Hyde Volume III, p. 217). These values range from $\bar{\nu} = 2.60$ to $\bar{\nu} = 2.84$. The most recent value of $\bar{\nu}$ has been given by Bol'shov et al³¹ who quotes a value

$$\bar{\nu} = 2.71 \pm 0.04 \text{ for } \text{Cm}^{244}$$

in comparison with Pu^{240} , Bol'shov states that

$$\frac{\bar{\nu}(\text{Cm}^{244})}{\bar{\nu}(\text{Pu}^{240})} = 1.250 \pm 0.013$$

The average K.E. of fission fragments from Cm^{244} has been given by Smith et al³² as

$$E_K = 185.5 \pm 4 \text{ mev}$$

whereas Bol'shov³¹ quotes

$$E_K = 182.3 \pm 2.3 \text{ mev}$$

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A.9 SUMMARY

The neutron cross-sections of Cm^{244} have been calculated from thermal energies to 14 Mev. The calculations have been cross checked in most case by as many as three independent theoretical approaches giving a high degree of confidence in the results. It appears desirable, however, to verify these calculations by performing cross-section experiments at an early date. An estimation of the minimum critical mass based on these cross-sections, will be made in the final portion of this study to determine the extent of the criticality problem in the use of Cm^{244} as a heat source.

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EVALUATION OF HEAT BLOCK DESIGN

The literature has been reviewed for data to evaluate the chemical compatibility of materials chosen for the GE Large Radio-isotope Heat Source heat block designs, and to provide the basis for suggested changes in design to improve system compatibility. Compatibility between lithium and sodium and various cladding metals and alloys and also between cladding materials and graphite was considered. In general, data resulted from experiments of from several hours' to several thousand hours' duration.

The design now under study includes the following interfaces expected to operate at about 1800 F for periods of five years or more:

TZM alloy radioisotope heat source cladding vs Al_2O_3

Al_2O_3 vs Haynes 25 or Hastelloy X cladding for graphite heat block

Haynes 25 vs sodium or lithium thermal bond

Al_2O_3 vs sodium

Al_2O_3 vs graphite

Graphite vs sodium.

Although the cladding materials of principal interest in the present heat block design are TZM, Haynes 25 and Hastelloy X, data was also sought for the major constituents of these alloys and for other similar alloys.

METAL AND ALLOY-GRAPHITE COMPATIBILITY

Geras and Mallet⁽¹⁾ studied the compatibility of several metals and alloys with graphite at 1650 F and 1850 F for 250 and 1000 hours at

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carbon diffusion through the protective layer without the formation of copper or nickel carbides. A 4-mil chromium plate on 316 SS was about as effective in reducing carburization of the stainless steel. Although carbide precipitates were observed in the steel, only a slight increase in alloy hardness occurred.

Only the nickel alloys were tested for 1000 hours at 1850 F. Of these Inconel X and Alnico were superior to the others and to the stainless steels. Even the Inconel X and Alnico, however, showed deep carbon penetration, extensive hardening and probably lowered ductility. Copper foils 1.5 and 8 mils thick did not prevent carburization of the Inconel X and Alnico. In addition to carbon diffusion through the foils, the copper also diffused into the alloys and lost its integrity.

In similar work by Bokros,⁽²⁾ austenitic and ferritic stainless steels, nickel base alloys and several refractory metals and alloys were tested in contact with graphite in helium-filled capsules at interface contact pressures of 2000 psi and at temperatures up to 1850 F for 1000-2000 hours.

The rates of carburization of Mo and Nb were sensitive to the purity of the helium in the reaction capsule. After 2000 hours at 1850 F in ungettered helium the Mo_2C reaction layer formed in the Mo-C couple was about 3.5-5 mils thick. When zirconium-gettered helium was used, the Mo_2C layer thickness was 0.4-1 mil similar to the above findings of Gerds and Mallet. Reaction layer thickness for graphite and niobium and niobium + 1

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Zr alloys at 1850 F for 2000 hours in gettered helium was about 1.5 mils. Use of ungettered helium increased reaction layer thickness by 50-100 per cent. The Nb + 1 Zr alloy was less reactive in the ungettered helium apparently because the Zr in the alloy acted as an internal getter and prevented carbon transfer in the gaseous CO phase. The iron-chromium and iron-chromium-aluminum alloys were exposed only at temperatures up to 1500 F and will not be discussed here.

Bokros found that nickel and Monel were compatible with graphite up to 1850 F, for times up to 2000 hours. Carbon dissolved in the nickel at 1700 F, however, reprecipitated during aging in the form of graphite nodules. The room temperature ductility of aged nickel and Monel was the same as the unaged and uncarburized ductility. The ultimate tensile strength increased with aging temperature up to 600 F, and then decreased to slightly above the uncarburized strength for aging temperatures up to 1200 F. The addition of copper to nickel in the Monel apparently was the reason for a decreased tendency toward graphitization during aging because of lowered carbon diffusion into the alloy.

Nickel alloys with chromium, molybdenum, and iron exhibited the least compatibility with graphite. Extensive grain boundary carbides occurred in these alloys at temperatures as low as 1300 F. In addition, considerable internal oxidation was observed when unpurified helium was used as the exposure environment. Intergranular carbides in Inconel X penetrated 10 mils in 2000 hours at 1300 F. Inconel, Inconel 702 and Nichrome V were even less compatible. Hastelloy B carburized rapidly at 1500 F.

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Copper, aluminum, chromium and molybdenum protective barriers on stainless steels were also evaluated. The copper diffused rapidly into 304 and 430 SS and offered no protection even at 1500 F. Chromium plating, 1 mil thick, protected 304 and 430 SS only up to 1500 F, but diffused rapidly into the stainless steel in 1000 hours at 1700 F.

A 5-mil molybdenum foil on 316 SS appeared to prevent carburization of the stainless steel at 1600 F but the results are inconclusive because of carburization of the unprotected edges of the stainless steel.

Research conducted at Battelle^(3,4) on refractory metal and alloy compatibility with graphite and uranium carbide fuel alloys indicates Nb, Nb-1Zr and Mo carburization rates at 1800 F which are comparable to those reported by Bokros⁽²⁾ (about 1/4 as high as rates reported by Bokros but probably within reasonable limits for experimental scatter).

No specific information has been obtained on compatibility between Haynes 25 alloy and graphite. Some data^(5,6,7,8) has been found, however, on solubility and diffusion rates of carbon in several constituents of the alloy and in Ni-Cu, Ni-Co, and Fe-Co alloys. This information will require further analysis along with additional thermodynamic data on carbide stability of the various constituents of Haynes 25 alloy to determine whether carburization rates in this alloy are tolerable for the heat block cladding application.

METAL AND ALLOY-LIQUID METAL COMPATIBILITY

As in the case of the metal-graphite compatibility data, the metal-liquid metal compatibility data is for exposure times from several hours to a

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maximum of several thousand hours--considerably less than the 50,000 hours compatibility and strength requirement for the proposed heat block application. At the present time, predictions of the long-term compatibility can only be made by extrapolating the short-term compatibility data.

Reynolds⁽⁹⁾ reported that in nine sodium-filled Nb-1Zr reflux capsules corrosion was negligible after 5000 hours at 2200 F. Similar satisfactory results were obtained for tantalum and molybdenum; reaction layer and sodium penetration for the three materials was less than 0.5 mil. Other BNL research⁽¹⁰⁾ extended the time and temperature limits for sodium compatibility with Nb-1Zr to 8000 hours at 1315 C. For the same conditions Nb-10W-10Ta and Ta-10W alloys suffered severe attack.

Nevsorov⁽¹¹⁾ reported on reaction between flowing sodium and nickel-based alloys (Ni:60-75%, Cr:16-20%, Mo:5%, Ti:1.0-2.5%, W:10%) and austenitic steels (up to 20% Cr and 14% Ni) at temperatures up to 930 C. The nickel-based alloys suffered intergranular attack for exposure times as low as 300 hours.

The austenitic steels appeared to be unaffected under the same time-temperature conditions. The intergranular attack of the nickel-based alloys was believed to result in part from grain boundary oxidation as a result of oxygen impurity (oxygen concentration, $5 \times 10^{-3}\%$) in the sodium.

In a 3600-hour 600 C exposure to flowing sodium (O_2 concentration $2-4 \times 10^{-3}\%$) Nb was severely attacked while Mo was unaffected. At even higher oxygen contamination levels in the sodium (2×10^{-3} to $10^{-2}\%$) the Mo appeared to be unaffected by nearly 400 hours exposure at 900 C.

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Similar observations on the effect of interstitial contamination in the test material and oxygen contamination in the liquid metal have been made by several investigators.

DiStefano and Hoffman^(12,13,14) reported that in the refractory metals, corrosion resistance is generally excellent up to 1700 F or 1800 F provided that interstitial contamination in the refractory metal and oxygen contamination in the liquid metal is kept low.

Niobium containing less than 100 ppm O₂ was completely resistant to lithium corrosion in 300 hour tests at temperatures up to 1800 F and in flowing lithium at temperatures up to 1600 F. At higher oxygen levels, the niobium was subject to intergranular attack at rates depending on the oxygen concentration. In other tests of Inconel, stainless steels, molybdenum, tantalum, vanadium, and zirconium all but the Inconel and stainless steels were corrosion resistant at 1500 F for 100-hour to 3000-hour test periods.

Tantalum, like niobium, suffered intergranular corrosion by lithium at oxygen levels of several hundred parts per million. Vanadium, titanium and zirconium were not affected in this way even at oxygen concentrations greater than 2000 ppm. Additions of 1 w/o zirconium to niobium eliminated the grain boundary penetration by lithium presumably because the zirconium formed ZrO₂ and prevented Li₂O formation in the grain boundaries.

Limited static testing at 1500 F⁽¹³⁾ indicates that lithium corrosion resistance of Haynes 25 alloy is not as good as the Hastelloy alloys and Inconel.

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In studies of the nickel-base alloys it was found that Cr, Fe, and Mo improve the corrosion resistance of nickel in 100-hour 1500 F tests. Copper and aluminum additions to nickel (Monel and Duranickel) resulted in more severe attack than on unalloyed nickel because of preferential solution of copper and aluminum.

Studies at BMI on the use of refractory metals in thermionic converter systems showed that many of the refractory metals and alloys remained resistant to lithium attack at temperatures above 1800 F.

Lithium did not attack TZM, Mo-50Re, W, W-.9Nb, W-10Mo, W-10Re, or W-25Re in 1000 hours at 2500 F, but Ta-12W and T-111 were measurably attacked by 2500 F lithium in 100 hours.

At temperatures above 2500 F, most of the alloys were noticeably attacked. TZM and Mo-50Re remained compatible with lithium at 3000 F. When attack occurred it was generally intergranular corrosion resulting from grain boundary impurities or impurities in the liquid metal. Several instances of dissimilar metal corrosion were observed in three component systems.

Results of compatibility studies of liquid metal vs metal oxide and metal carbide, reported by Hoffman,⁽¹⁴⁾ are limited to 1500 F and 100-hour exposure. TiC, ZrC, Cr₃C₂, were not measurably attacked either in lithium or sodium or lead. MgO, Al₂O₃ (single crystal), and ZrO₂ (CaO stabilized) did not react with sodium or lead but were severely attacked by lithium under the same time and temperature conditions.

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DISCUSSION AND CONCLUSIONS

The quantitative compatibility data, although limited, are sufficient to permit several conclusions on the feasibility of a long lived RTG using the liquid metal-bonded graphite heat block.

The superalloys and stainless steels do not appear to be satisfactory for cladding application at 1800 F for periods of 5 years or longer. In the first place, most of the nickel based and cobalt based alloys (Inconels, Hastelloys, Haynes 25) and the stainless steels suffer severe intergranular carburization, with corresponding loss of ductility. Secondly, these alloys are subject to grain boundary corrosion by lithium and sodium at temperatures considerably lower than the proposed 1800 F RTG operating temperature. In addition to the separate reactions with graphite and liquid metal, the superalloy would probably be more severely attacked when in simultaneous contact with graphite and liquid metal.

Although Monel is the single nickel-base alloy exhibiting sufficient compatibility with graphite for this application, it would not be suitable in a lithium bonded system because of attack by the liquid metal. It might be a satisfactory cladding material, however, in a system consisting of graphite but without the liquid metal.

The several barrier materials considered in research to date are of marginal value in preventing carburization at 1800 F and one barrier, copper, would be of no value in liquid lithium because of its reported high solubility in that material.

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In general, the refractory metals and their alloys would be more satisfactory materials in the liquid metal-graphite system with Cb-1Zr and TZM at the present time being the most promising alloys in the group. Their compatibility with graphite appears to be satisfactory for the proposed time and temperature conditions, and their compatibility with lithium and sodium are more than adequate. It must be remembered, however, that the attractive compatibility with graphite and liquid metal of the refractory metals and alloys depend on low oxygen and nitrogen impurity levels in the system. The problems of maintaining adequate purity levels in the liquid metal and in the cladding alloy and of outgassing the graphite before heat block assembly would no doubt require considerable attention. Moreover, the effects of dissimilar metal corrosion in systems of three or more component materials are not well known.

It is likely that such effects exist, and that the presence of a liquid metal accelerates the dissimilar metal reactions by enhancing the rate of transport of reaction products. Consequently, a more attractive system would have the desired heat transfer properties without the liquid metal component.

The compatibility data also indicate that the Al_2O_3 spacers proposed in the present heat block design would be compatible with liquid sodium but not with liquid lithium. The high vapor pressure of the sodium thermal bond (reported to be about two atmospheres at 1800 F) might rule out the use of

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sodium in any practical heat block design. If spacers are required, several of the refractory metal carbides could be used in the refractory metal-lithium-graphite system. TaC, ZrC, WC are sufficiently compatible with refractory alloy cladding and with graphite and liquid metal. It would be as advantageous from the standpoint of problems of dissimilar material compatibility to eliminate spacer materials as it would be to eliminate the liquid metal thermal bond.

These simplifications, that is, the elimination of the liquid metal and the spacers appear to be possible if the refractory metal-clad graphite combination is used. Further, it would be worth considering the elimination of the graphite from the system provided that desired heat transfer, heat capacity and re-entry ablation characteristics could be obtained. Use of a refractory metal clad in this manner would introduce a new "compatibility" problem, that is, the oxidation of the refractory in air. The effectiveness of protective coatings under conditions of ground handling and test must be evaluated.

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BERYLLIUM AS HEAT BLOCK MATERIAL IN CONTACT WITH
HIGH TEMPERATURE HEAT SOURCES

Beryllium is being considered for use as a heat block material in radioisotope-powered thermoelectric generators. Its low density, high thermal conductivity and high heat capacity are ideal for this application. It seems feasible, too, that cladding techniques can be developed for various cladding materials to protect the beryllium from evaporation or oxidation at elevated temperatures.

A major obstacle to its use, however, is its chemical reactivity with most metallic cladding materials. The following data indicate that beryllium reaction rates with most metal or alloy claddings would be prohibitively high at the anticipated operating temperatures (1800 F).

TABLE 1. BATTELLE DATA ON BERYLLIUM COMPATIBILITY
WITH METALS AND ALLOYS (Reference BMI-1545(U))

Metal or Alloy	Temperature, °F	Time, hour	Reaction Layer Thickness, mils
Ti Name1	1525	4	1-4
Ti Name1	1650	4	"
Mild Steel	1650	4	"
Stainless Steel	1550	4	"
" "	1650	1	"
" "	1650	4	"
Molybdenum	1550	1	"
"	1650	4	"
Tungsten	1650	4	"
"	1750	4	"
Tantalum	1650	4	"
Niobium	1650	4	"
Nickel	1650	4	" (a)

a) Data originally reported for Nickel in BMI-1545(U) was later found to be incorrect. Correct reaction rate 1-4 mils in 4 hours.

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TABLE 2. P&W DATA ON BERYLLIUM COMPATIBILITY WITH METALS
AND ALLOYS AT 1400 F (Reference PWAC 1018, 1019(C))

Metal	Number of Reaction Zones	Reaction Layer Thickness (mils)	
		800 hours	10,000 hrs ^(a)
Ag	1	1	3
Co	2	3	15
Cr	1	2.3	6
Cu	6	100	250
Fe	1	4	20
Mo	1	.4	4
Ni	2	28	80
Pd	2	9-12	30
Pt	1	44	
Ta			4
Ti	1	2	10
V	1	.7	5
W	1	.5	5
Zr	1	22	90
316 Stainless Steel	3	10	20
Nb + 1 Zr	1	1.3	7
Nb + 20 Ti	1	1.5	

(a) 10,000-hour data extrapolated from 3000-hour tests.

A complete assessment of beryllium usefulness in this application must look beyond the high degree of reactivity of beryllium. The evaluation should examine the ultimate effects of the reaction, and whether the effects can be tolerated or whether system design can be changed to minimize the compatibility problem.

Several questions which need further study are:

(a) What are the high-temperature long-time reaction effects?

The data in Tables 1 and 2 are for relatively short times at temperatures several hundred degrees below the proposed

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operating temperature. The reaction rates would be expected to increase with increasing temperature and the thickness of reaction zones would be expected to increase for longer exposure times. However, the exact temperature and time dependence in the various systems is not known. Additional research is necessary under the more severe time-temperature conditions to determine if diffusion of beryllium through the reaction zones continues at a constant or at a decreasing or increasing rate as the reaction layer grows. It would also be necessary to determine if any reaction products formed under the more severe conditions act as barriers to beryllium diffusion.

- (b) Can barrier materials be developed to prevent beryllium reaction with the cladding? The barrier material would have to be inert with respect to the beryllium and to the cladding, have a thermal conductivity comparable to that of beryllium and also perhaps satisfactory physical and mechanical properties, such as thermal expansion, ductility and strength.
- (c) In the event that the compatibility problem cannot be completely solved, how much beryllium-clad reaction can be

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tolerated? What is the effect of reaction products on the clad strength? If the reaction products were ductile and as strong as the beryllium then mechanical integrity might not be impaired. The extent to which the clad thickness could be increased to minimize the effects of low strength, low ductility reaction products is dependent on limitations on system weight and thermal efficiency.

- (d) If beryllium diffuses completely through the clad what is the controlling factor for beryllium loss at the clad surface: beryllium diffusion rate or evaporation rate? It might be possible to develop claddings or barriers through which beryllium would diffuse at a tolerable rate.

In the Battelle and Pratt and Whitney work referenced above it was found that, in addition to the generally poor compatibility, with few exceptions no interface bonding could be maintained. The ideal situation for heat transfer would be the formation of sound bonds between the beryllium and the claddings. Thermal cycling failure of the bonds because of lower ductility reaction products or because of differences in thermal expansion reduces the effective conductivity of the system and thus leads to higher temperatures. In this connection additional study would be required to:

- (a) Develop beryllium-clad-barrier systems which form sound interface bonds.

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- (b) Study the physical and mechanical properties of various intermetallic reaction layer materials- thermal expansion, thermal conductivity, ductility and strength.
- (c) Examine beryllium-clad geometry to minimize the effect of differential thermal expansion, and low strength and ductility reaction layers. With proper selection of cladding and barrier materials and proper geometry and fabrication method it might be possible to maintain contact between the beryllium and cladding even though their thermal expansion coefficients are different and reaction products have low strength and low ductility.

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