Conf-720519--1

CURIUM-244 — A RADIOISOTOPIC POWER FUEL*

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ABSTRACT

The alpha-emitting radioisotope curium-244 is being developed by the USAEC as a power source for any of several conversion systems which produce electrical power. Projected costs and availability make this radioisotope especially promising. The cost of curium-244 which is produced in nuclear power reactors is estimated at $60/Watt (thermal) in quantities of 95 kW (thermal) per year by 1980. The efficient and safe use of this isotopic fuel requires that a suitable compound be prepared and fabricated into a suitable form with fully characterized physical and chemical properties. The task elements for accomplishing this program consist of compound preparation, source fabrication, compound characterization, source design, and prototype testing. Techniques developed for accomplishing these tasks and the results achieved to date are reviewed.

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* Research sponsored by tomic Energy Commission under contract with the Union Carbide Corporation.

INTRODUCTION

Electrical power requirements for orbiting satellites, remote weather stations, ocean buoys, etc. have indicated a need for compact, reliable power sources capable of long-term operations [1,2]. Radioisotopes are reliable sources of thermal energy that can be converted to electrical power by thermoelectric, thermionic, or mechanical conversion systems. Several radioisotopes are being studied in the U.S. Atomic Energy Commission's program for development of radioisotopic power [3]. Curium-244 is one of these radioisotopes. While the development of radioisotopic power was initiated in the U.S. in 1956 [2], curium-244 has been under development only since 1964. This paper reviews the current status in the development of curium-244 as a power fuel.

Curium has been chosen as a power fuel for several reasons: (1) high power density, (2) excellent properties of fuel compound, (3) low cost, and (4) availability. The high power density of curium compounds (28.4 W/cm^3 for $^{244}Cm_2O_3$) permits less costly and safer converter systems, particularly thermionic converters. Curium sesquioxide (Cm_2O_3) has been chosen as the primary fuel form for heat source applications because of its thermal stability and its compatibility with high temperature-resistant encapsulating materials. The high melting point (2265°C) and high power density of $^{244}Cm_2O_3$ afford a flexible range of power converter applications, including thermoelectric, thermodynamic cycle, and thermionic generators. The sesquioxide is prepared by precipitating curium oxalate and calcining to form CmO_2 followed by heating in an inert atmosphere at 1300°C to reduce the dioxide. Cylindrical pellets of near theoretical density ranging in size from 0.25- to 1.0-in. diameter have been prepared by hot pressing at 1450°C and 4000 psi.

Curium-244 will be available in large quantities as a by-product of power reactor operation in the last half of the 1970 decade. When curium recovery equipment is installed in conjunction with power reactor fuel reprocessing equipment, the only cost allocated to curium will be the processing cost. Various estimates of the future price of curium-244 based on large quantity production have ranged from \$30 to \$60/W_t of unencapsulated Cm $6m_2O_3$ powder [4]. To the estimate of curium-244 isotope cost should be added an estimated \$40/W_t for encapsulation costs for sources containing about 2000 W_t. One arrives at a conservative (high) \$100/W_t for an encapsulated source. It is estimated that encapsulated plutonium-238 heat sources will cost about \$500/W_t to fabricate in the future as compared to a present day \$700/W_t.

The predicted cost of electrical power from isotope fueled systems makes these systems cost-effective in a number of aerospace applications and competitive with solar cells, particularly in low-earth orbit missions. Predicted power costs of curium and plutonium fueled generators and solar cells are compared in Table I [4]. These figures include estimated power generator costs and curium-244 and plutonium-238 heat sources at \$100/W_t and \$500/W_t, respectively. The power costs for recovered fuel are based on \$50/W_t to reprocess plutonium-238 and \$20/W_t to reprocess curium-244 after the mission. No estimate is made for the cost of returning the fuel from space but it is expected such a capability will exist when personnel shuttles are developed.

Curium-244 is a reactor product that results from the neutron irradiation of uranium-238. Curium can be recovered directly from spent reactor fuel. The availability of this isotope is controlled by the quantity of reactors in operation in the these radioisotopes. While the development of radioisotopic power was initiated in the U.S. in 1956 [2], curium-244 has been under development only since 1964. This paper reviews the current status in the development of curium-244 as a power fuel.

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Currently, curium-244 for heat source applications is produced in AEC reactors. It is anticipated that eventually production of this isotope will be assumed by the private sector. Projected availability from commercial sources is shown in Table II [2]. The listed availability of curium-244 is 66% of the total to be available in the years 1975 through 1978 and 75% from 1979 through 1981. Curium-244 that would be available from the irradiation of recovered americium-243 has not been added to the table.

SOURCE COMPOUNDS AND FUEL FORMS

The safe and efficient use of radioisotopes in varied applications (extant, proposed, or envisioned) requires the selection of a suitable fuel compound with known physical and chemical properties and demonstrated fabrication technology. This paper is primarily concerned with the source compound $^{244}Cm_2O_3$ since it is the compound which has been studied extensively. Other compounds of curium-244, such as silicides [5], carbides, nitrides, sulfides, fluorides, oxysulfides, phosphides, phosphosulfides, and borides, were studied only to the extent necessary to indicate their suitability as source compounds [5-8].

Mixtures of Cm_2O_3 with metals offer advantages in safety and certain operating parameters. Fuel containment can be enhanced by mixing a relatively small proportion (20-30%) of $^{244}Cm_2O_3$ with an inert metal, such as iridium. The resultant cermet possesses sufficient power density for thermoelectric and thermodynamic generator applications while presenting essentially an inert face to environmental effects. Cermets of Cm_2O_3 also exhibit improved thermal conductivity and permit the use of larger individual fuel components while maintaining reasonable interior operating temperatures and reducing thermal stresses. Cermets ranging from 20 to 80 volume percent $^{244}Cm_2O_3$ to provide power densities ranging from 4 W_t/cm^3 to 16 W_t/cm^3 are presently being investigated for fabricability, uniformity, helium release, thermal conductivity, and impact effects. The percentage incremental cost due to iridium would range from 2.5% for 80 volume percent Cm_2O_3 cermet to 38.5% for 20% Cm_2O_3 cermet based on $40/$W_t$ for curium-244 and $5/g for iridium.

Advanced fuel forms of curium presently being investigated are the noble metal compounds of curium, $CmPt_3$, $CmRh_3$, and $CmIr_3$. Such compounds may have excellent high temperature properties as well as resistance to impact and environmental effects, particularly when alloyed with a noble metal [9]. Work at present involves studying the reactions to produce these compounds.

PROPERTIES OF CURIUM-244 SESQUIOXIDE

The production of curium-244 on a kilogram scale has been described by Groh *et al.*, [10] who also describe the radiochemical purity of $^{244}CmO_2$ prepared on a miniature scale. The purity [11] of a typical batch of $^{244}CmO_2$ prepared on a full-production scale is given in Tables III-V.

Curium, like the actinides plutonium and americium and the lanthanides cerium and praseodymium, can exist as an oxide in more than one oxidation state. Asprey *et al.* [12] prepared a black oxide by ignition of approximately 400°C and cooled it in air. A white or faint tan oxide was prepared by thermal decomposition of the efficient use of radioisotopes in varied applications (extant, proposed, or envisioned) requires the selection of a suitable fuel compound with known physical and chemical properties and demonstrated fabrication technology. This paper is primarily concerned with the source compound $^{244}Cm_2O_3$ since it is the compound which has been studied extensively. Other compounds of curium-244, such as silicides [5], carbides, nitrides, sulfides, fluorides, oxysulfides, phosphides, phosphosulfides, and borides, were studied only to the extent necessary to indicate their suitability as source compounds [5-8].

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Wallman [16] studied the effect of self-radiation on curium sesquioxide (bcc, a = 11.00 ± 0.01 Å) which he prepared by heating $^{244}CmO_2$ at 600°C in vacuum. He identified an A-type $CmCm_2O_3$ (hexagonal, a = 3.80 ± 0.02 Å, c = 6.00 ± 0.03 Å) produced by self-radiation. Haug [17] succeeded in preparing B-type Cm_2O_3 (monoclinic, a = 14.282 ± 0.008 Å, b = 3.65 ± 0.003 Å, c = 8.900 ± 0.005 Å, b = 100.31 ± 0.05°) by heating C-type Cm_2O_3 above 850°C. The A-type Cm_2O_3 was prepared by Mosley [14] by heating B-type Cm_2O_3 above 1600°C where a reversible transformation occurs. He reports the monoclinic form of Cm_2O_3 to be resistant to alpha radiation. Mosley reevaluated the lattice constants of all the forms of Cm_2O_3 (Table VI) and compared the transformation to those that occur in lanthanide oxides (Fig. 2). He concluded that B-type Cm_2O_3 is structurally best suited for a source form. Phase diagrams for the curium-oxygen system have been tentatively proposed by Smith [18] (Fig. 3) and Chikalla [15]. Smith's construction of a phase diagram is from the data of Mosley [14] and Posey [13] and is for the condition in air at atmospheric pressure. Chikalla's construction is from his thermal analysis data and covers the composition range of Cm_2O_3 to CmO_2.

The properties of a radioisotopic power fuel which must be known to predict the behavior of the fuel can be divided into two categories (Table VIIO: those which relate primarily to terrestrial safety and operational aspects, and the additional properties primarily for aerospace safety.

The properties of a radioisotope heat source inevitably change with time. The source is, at best, a pure compound only at the instant it is formed; thereafter, due to radioactive decay, it is a system of changing composition. In addition to the changes resulting from the accumulation of decay products, the effect of self-irradiation must also be considered. The changes in the properties produced by self-irradiation are primarily those which occur as a result of changes in the structure of the solid. Properties most likely to be affected include density, thermal conductivity, helium release, mechanical strength, and leach rate. Other properties such as vapor pressure, melting point, heat capacity, boiling point, heat of vaporization, and heat of fusion are essentially unaltered by radiation damage. As the temperature of a heat source increases, annealing of defects produced by radiation damage increases, so that at a given temperature steady state is established between the effects of annealing and those of radiation damage. At the temperatures at which heat sources will be used, steady state is established in a short time compared to the mission life of the source.

Properties inferred from measurements made on analogous nonradioactive materials (actinide and rare-earth stand-ins for curium fuels) are valid only if the property being determined is unaffected by radiation damage. Property measurements of nonradioactive samples can be assumed to represent the values at time zero when radiation damage is nil. To completely describe the physical properties of a heat source, the effects of radiation damage-thermal self-radiation. Haug [17] succeeded in preparing B-type Cm_2O_3 (monoclinic, a = 14.282 ± 0.008 Å, b = 3.65 ± 0.003 Å, c = 8.900 ± 0.005 Å, b = 100.31 ± 0.05°) by heating C-type Cm_2O_3 above 850°C. The A-type Cm_2O_3 was prepared by Mosley [14] by heating B-type Cm_2O_3 above 1600°C where a reversible transformation occurs. He reports the monoclinic form of Cm_2O_3 to be resistant to alpha radiation. Mosley reevaluated the lattice constants of all the forms of Cm_2O_3 (Table VI) and compared the transformation to those that occur in lanthanide oxides (Fig. 2). He concluded that B-type Cm_2O_3 is structurally best suited for a source form. Phase diagrams for the curium-oxygen system have been tentatively proposed by Smith [18] (Fig. 3) and Chikalla [15]. Smith's construction of a phase diagram is from the data of Mosley [14] and Posey [13] and is for the condition in air at atmospheric pressure. Chikalla's construction is from his thermal analysis data and covers the composition range of Cm_2O_3 to CmO_2.

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Density

The density of B-type (monoclinic) Cm_2O_3 as reported by Mosley [14] is 11.7 g/cm^3 at room temperature. This value was calculated from x-ray data, assuming six molecules per unit cell. The densities of A- and C-type Cm_2O_3 are given in Table VI.

Thermal Coefficient of Expansion

The thermal coefficient of expansion [14] of monoclinic Cm_2O_3 is shown in Fig. 4 for temperatures up to 1420°C. The density of monoclinic Cm_2O_3 at 1600°C (obtained by extrapolation) is 10.9 g/cm^3. The density of A-type Cm_2O_3 at 1650°C (see Table VI) is 11.4 g/cm^3. Moseley suggests that the change in density from B-type to A-type structure at approximately 1600°C may affect the integrity of fabricated Cm_2O_3.

Specific Power and Power Density

The specific power of elemental 100% curium-244 as calculated from decay energies and half-life (18.1 yr) is 2.84 W/g. The specific power of production quality $^{244}Cm_2O_3$ will approach 2.45 W/g depending on the level of ionic impurities and amount of decay. The theoretical power density of production quality $^{244}Cm_2O_3$ is 28.4 W/cm^3 at room temperature. Assuming 95% of theoretical density for pellets, the power density after fabrication is 27 W/cm^3.

Radiation and Shielding

The radiation characteristics of $^{244}Cm_2O_3$ product are given in ref. 20. The radiation characteristics of Savannah River Laboratory production $^{244}Cm_2O_3$ are summarized and compared with the radiation characteristics of $^{238}PuO_2$ in Table VIII. Arnold [21] has made calculations on the shielding requirement for curium isotope power fuels.

The radiation measurements made on 36.2 g of curium-244 in a stainless steel container with a wall thickness of 0.6 in. is given in Table IX.

Helium Release

The release of helium from $^{244}Cm_2O_3$ microspheres as well as 1- by 1-cm cylindrical pellets was studied [22] under steady-state conditions [23]. In the steady-state release experiments it was assumed that generation of helium within the material occurred at a rate, P (cm^3 of gas)/(cm^3 of solid)(sec), independent of time and position. At steady state the release rate of helium will equal the generation rate and under the foregoing assumptions the concentration distribution of helium within a releasing spherical unit is given by

$$c(r) = \frac{P}{6D}(a^2 - r^2) , \qquad (1)$$

where c(r) = concentration of helium at radius r, cm^3/cm^3,

a = effective radius of releasing unit, cm, and

D = effective diffusion coefficient, cm^2/sec.

The average concentration within the sphere, C_{av}, may then be calculated as [23]

$$C_{av} = \frac{P}{15D'} , \qquad (2)$$

where $D' = D/a^2$, the effective diffusion-release parameter, sec^{-1}. Experimentally, the

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$$C_{av} = \frac{P}{15D'} , \qquad (2)$$

where $D' = D/a^2$, the effective diffusion-release parameter, sec^{-1}. Experimentally, the sample is maintained at the required temperature in vacuum until the measured helium release rate equals the known generation rate. The average helium concentration is then determined by melting and degassing the sample, and an effective diffusion parameter, D′, can be calculated directly from Eq. (2).

The results of the steady-state helium release determination from 900 to 1800°C are plotted in Fig. 5. The diffusion parameter, D', is independent of the geometry of the sample, which indicates that the basic releasing unit is of constant dimensions.

At temperatures below 1620°C (the temperature of the monoclinic-hexagonal phase transition) the diffusion parameter, D', was controlled by two different processes. The value of D' is obtained by adding the values calculated from

$$D' = 7.87 \times 10^{-4} \exp(-16{,}248/RT°K) , \tag{3}$$

which is predominant at temperatures below 1300°C and the values calculated from

$$D' = 68.0 \exp(-53{,}080/RT°K), \tag{4}$$

which is predominant up to 1620°C. At temperatures above 1620°C, the release of helium is significantly different from that below 1620°C. This was presumably due to the characteristics of the hexagonal structure or to changes in the sample produced during the monoclinic-hexagonal transition. Figure 6 shows the rapid transient release of helium at the transition temperature in $^{244}Cm_2O_3$. The steady-state diffusion parameters in $^{244}Cm_2O_3$ above 1620°C are given by

$$D' = 4.65 \times 10^{7} \exp(-102{,}780/RT°K) . \tag{5}$$

The activation energies, -16,248 cal/mole and -53,080 cal/mole, are similar to that observed in $^{238}PuO_2$ [23].

Angelini [22] studied the effect of varying the radiation dose rate on the diffusion parameter (D/r^2) of helium in $^{244}Cm_2O_3$. To vary the radiation dose rate the $^{244}Cm_2O_3$ was diluted with Gd_2O_3. The classical diffusion model posits that the diffusion parameter is independent of helium concentration (or in this case the production rate of helium). The radiation dose rate, however, is a linear function of the helium production rate. Values of the steady-state helium concentration in the $^{244}Cm_2O_3$-Gd_2O_3 samples (from which diffusion parameters are calculated) were determined on 75 mole % Cm_2O_3-25 mole % Gd_2O_3 and 46 mole % Cm_2O_3-54 mole % Gd_2O_3 in the temperature range 1300-1700°C. Data for the diffusion parameter of helium have been previously reported. The data are plotted in Fig. 7. A power dependence of D'_M on radiation dose rate is inferred. This conclusion is consistent with the relative values of D' determined on $^{242}Cm_2O_3$, $^{244}Cm_2O_3$, and $^{238}PuO_2$. The assumption that Gd_2O_3 is inert is reasonable but not proven.

Thermal Conductivity

The thermal conductivity of $^{244}Cm_2O_3$ was measured [24] in a longitudinal heat flow thermal conductivity apparatus using an absolute method which was developed for use with highly radioactive samples and which had previously been used to measure the thermal conductivity of strontium-90 compounds. The thermal conductivity measurements which were made in the temperature range of 500-1100°C are plotted in Fig. 8. The data shown in Fig. 8 have been used to derive an equation to represent the thermal conductivity of 100% theoretical density $^{244}Cm_2O_3$ as a function of temperature (Eq. 6),

$$1/k = 37.795 + 0.01659T , \tag{6}$$

where k is in W/cm·°C and T is in °K.

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where k is in W/cm·°C and T is in °K.

Melting Point

The melting point of a sample $^{244}Cm_2O_3$ of unknown purity was reported by McHenry [25] to be 1950°C. Subsequently, measurements made on highly purified Cm_2O_3 (low plutonium content) gave higher melting points. Smith [26] reports 2255-2277°C for his most purified material and 2173-2186°C for material of composition similar to production quality

$^{244}Cm_2O_3$. Chikalla [27] made similar measurements of purified Cm_2O_3 and obtained melting temperatures of 2220-2240°C.

Critical Mass

The critical mass of $^{244}Cm_2O_3$ has been calculated [28] at Oak Ridge National Laboratory from cross-section data. The results (Table IX) indicate an underestimation of the critical mass, compared with replacement measurements made at Los Alamos [28].

Vapor Pressure and Heats of Vaporization and Formation

The vaporization rates of $^{244}Cm_2O_3$ have been measured by Smith [18] using a conventional Knudsen effusion technique. From Knudsen effusion data, the vapor pressures and heats of vaporization can be determined if the vapor species are known. Since the vapor species are not known (however, they are inferred by Smith from thermodynamic considerations to be curium plus oxygen), he arbitrarily expresses his vapor pressure date (Fig. 9) in terms of one mole of solid Cm_2O_3 vaporizing to one mole of gaseous (hypothetical) Cm_2O_3. He gives the heats of vaporization for the hypothetical condition of one mole of solid Cm_2O_3 vaporizing to 5 miles of gaseous atoms. When the vaporization reaction is determined, his vaporization rate data can readily be adjusted for the vaporization reaction to obtain true vapor pressures and heats of vaporization. The equation for the vapor pressure is

$$\log P_E \text{ (atm)} = 7.32\ (\pm 0.26) - [29{,}050\ (\pm 590)/T(°K)]. \qquad (7)$$

The equation was fitted to the data by least mean squares. Errors are standard deviation.

The heat of vaporization of one mole of Cm_2O_3 to 5 moles of gaseous atoms over the temperature range of 1800-2600°K was calculated to be 665 ± 13.5 kcal/mole of Cm_2O_3. The heats of vaporization were calculated to be 678.5 ± 8.0 and 670 ± 10 kcal/mole of Cm_2O_3 at 0°K as derived from the Third Law and Second Law of thermodynamics, respectively.

The heat of formation at 0°K was estimated by Smith [18] to be -429 ± 14 kcal/mole.

Vapor Transport of $^{244}Cm_2O_3$

The rate of transfer of Cm_2O_3 by volatilization and diffusion in a helium atmosphere has been measured by Posey [29]. These experiments simulated the conditions inside a heat source powered with $^{244}Cm_2O_3$. The rate of movement of Cm_2O_3 through a helium atmosphere in a tube of known length and constant cross section was determined. From these data the product of the vapor density times the diffusion coefficient was determined. A pellet of Cm_2O_3 was placed in one end of a 0.25-in.-ID tungsten tube of known length. The end of the tube containing the Cm_2O_3 was heated, while the other end was held at a lower temperature. The tube was in a vertical position with the hot end at the top to prevent thermal convection. Experiments were carried out at three temperatures. The details of the conditions are given in Table XI.

The results of the experiments described in Table XI are given in Table XII. The data in Table XII are reported at one atmosphere of helium by use of the equation (D)(C) =

$$(D)(C) = \frac{k}{p}, \qquad (8)$$

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$$(D)(C) = \frac{k}{p}, \qquad (8)$$

where D = diffusion coefficient, cm^2/sec,

p = helium pressure, atmospheres,

C = vapor density of Cm_2O_3, g/cm^3, and

k = constant.

2

The data in Table XII have been correlated by Eq. (9) so that the value of (D)(C) can be calculated for temperatures other than those at which the data were taken:

$$\log_{10}\left[\frac{(D)(C)}{T^{3/2}} \times 10^{10}\right] = 14.324 - (29,050/T), \tag{9}$$

where D = diffusion coefficient, cm^2/sec,

C = concentration of Cm_2O_3, g/cm^3, and

T = temperature, °K.

This equation was derived assuming that the vapor pressure varies with temperature according to the equation of Smith and Peterson [18]. The diffusion coefficient was assumed to vary as the 1.5 power of the absolut temperature. With the present data specific diffusion coefficients were not calculated. The vapor density, C, can be calculated directly using the vapor pressure data of Smith and Peterson [18]. Their data, however, were derived assuming a likely vapor specie. Any error in the assumption would have been reflected in the calculated value of "D". The product, (D)(C), is measured directly and is readily usable.

Solubility and Leach Rate of $^{244}Cm_2O_3$

The solution of Cm_2O_3 in air saturated distilled water and seawater has been investigated by Posey [30,31]. The solubility of $^{244}Cm_2O_3$ in seawater reached saturation in a few hours and did not change with time (up to 1032 hr). The solubility was $3.4 \pm 1.4 \times 10^{-3}$ mg/liter. Preliminary results on the solubility of $^{244}Cm_2O_3$ in air saturated distilled water as reported by Posey differ considerably from that in seawater. The solubility increased exponentially with time up to 1032 hr (the duration of the tests). The data were correlated with the equation:

$$S = 9.06 \times 10^{-4} t^{1.913} \tag{10}$$

where S = solubility in mg/liter, and

t = time, hr.

The equation was fitted by a method of least-squares. A plot of Eq. (10) is shown in Fig. 10. The saturation solubility of $^{244}Cm_2O_3$ in air saturated distilled waster was not determined. The difference in the solubility of $^{244}Cm_2O_3$ in seawater and distilled water was possibly attributable to the formation of a colloidal suspension in fresh water. Further studies are in progress by Posey.

Compatibility

The compatibility of $^{244}Cm_2O_3$ with the refractory metals and alloys Ta, Ta-10% W, T-111, Mo, TZM, W, and W-26% Re was studied by DiStefano and Lin [32] for times up to 10,000 hr at 1650 and 1850°C. The alloy W-26% Re showed the least interaction with $^{244}Cm_2O_3$; but Mo, TZM, and tungsten were acceptable, especially at 1650°C. Serious attack occurred *with Ta and Ta alloys. No reduction of* $^{244}Cm_2O_3$ *was observed.* The attack was believed to be by dissolution. Figures 11 and 12 show the several metals exposure to curium oxide at 1650 and 1850°C. Andelin and Watrous [33] are currently

T = temperature, °K.

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CURIUM SOURCES

A one-kilowatt heat source has been tested at ORNL for a period of 22 months. The objectives of this test were:

1. to develop the techniques for fabricating the fuel capsule, fuel pellets and fueling an all-refractory-metal heat source containing $^{244}Cm_2O_3$;
2. to investigate the performance of an open helium venting system; and
3. to investigate fuel pellet behavior in a full-size heat source.

The fuel capsule shown in Fig. 13 was fueled with 400 g of Cm_2O_3. The ~~assembly is show~~ test assembly is shown in Fig. 14. The source was tested for 22 months at an average temperature at the surface of the base of the source of 1100°C. The highest temperature was 1281°C.

The test assembly was disassembled in December 1972 and the fuel capsule was cut open. Some fuel migration was evident. Fuel was found between the bottom of the vent disk and the top of the fuel channels. An analysis of this material showed it to be about the same as the fuel pellets. A small amount of fine material (~2.7 g) was found above the vent disk; its heat output (~1.2 W/g) is substantially less than material chipped from the top of the fuel pellets (1.83 W/g). Analysis showed that this material contained Na, Ca, Mg, K, Al, and Ti in concentrations greater than the concentration in the parent fuel. A calorimetric measurement of the vent tube did not indicate that any curium-244 had reached the outside of the capsule.

The fuel pellets appeared to be intact and still in cylindrical form, but they were stuck tightly in the holes. The capsule pieces and vent tube were sealed in a large Hastelloy-C capsule and stored for metallurgical examination.

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Table [illegible]. Comparison of Space Electric Power Costs

System	Power (We)	Power Cost ($/We) Without Recovery	Power Cost ($/We) Recovered After 5 Yrs.
Plutonium - MHD	150	12,000	4,500
Curium - MHD	150	3,850	2,500
Plutonium-Brayton	1,000	2,900	650
Curium-Brayton	1,000	900	400
Solar Cells Low Earth Orbit	1,000	5,000	....
Solar Cells High Earth Orbit	1,000	2,000	....

Table [illegible]. Annual Projected Availability of ~~[illegible]~~ ~~and~~ Cm-244 in ~~Kilograms (KW)~~

Year	Plutonium-238 AEC	Plutonium-238 Commercial	Plutonium-238 Total	Curium-244 Commercial ( ) Kg KW(th)
1972	28		28 (16)	
1973	29		29 (16)	
1974	28		28 (16)	
1975	27	24	51 (28)	2 (5)
1976	29	58	87 (49)	4 (11)
1977	32	100	132 (74)	8 (22)
1978	32	148	180 (100)	12 (32)
1979	32	198	230 (130)	21 (53)
1980	36	260	296 (165)	36 (95)
1981	41	330	371 (210)	55 (145)

Table [illegible]. Isotopic Composition of SRL Curium-244 Product

Isotopic Mass	Isotope % by Weight November 1, 1968
^{242}Cm	0.0426
^{243}Cm	0.0238
^{244}Cm	95.08
^{245}Cm	0.729
^{246}Cm	4.02
^{247}Cm	0.0659
^{248}Cm	0.0381

IV

Table ~~5~~. Radioactive Impurities of SRL Curium-244 Product

Nuclide	Ci/g ^{244}Cm
^{103}Ru	<1 x 10^{-5}
^{106}Ru	7 x 10^{-5}
^{134}Cs	<1 x 10^{-5}
^{137}Cs	<1 x 10^{-5}
^{144}Ce	4.8 x 10^{-4}
^{144}Pr	<4.8 x 10^{-4}
^{95}Zr	<2 x 10^{-5}
^{95}Nb	<5 x 10^{-6}
^{154}Eu	<2 x 10^{-5}
^{243}Am	7.5 x 10^{-4}
^{252}Cf	1 x 10^{-4}

V

Table 5. Elemental Impurities of SRL Curium-244 Product

Element	Amount (ppm)	Element	Amount (ppm)	Element	Amount (ppm)	Element	Amount (ppm)
Li	<50	Cr	100	Sn	<25	Mo	<10
Na	10	Ni	100	Cd	<25	Mn	<10
Si	25	^{243}Am	3750	B	10	Co	<50
Zn	<250	Zr	150	As	<10	Pb	<10
Fe	500	Ca	<10	Cu	<10	Ta	<100
		Al	<25	Mg	<10		

Table 6. Structure of Cm_2O_3[a]

Composition: $CmO_{1.50} \pm 0.01$

C-type, body-centered cubic structure: a = 10.997 ± 0.005 Å

Density: 10.7 g/cm^3

C-B transformation: 800-1300°C (depends on crystallinity of C-type Cm_2O_3)

B-type, monoclinic structure:
a = 14.276 ± 0.008 Å
b = 3.656 ± 0.001 Å
c = 8.913 ± 0.004 Å
β = 100.39 ± 0.03°

Density: 11.7 g/cm^3
Six Cm_2O_3 molecules per unit cell
Space group C_{2h}^3 (C2/m)
Coefficient of linear thermal expansion
9.6 x 10^{-6}/°C (25-1000°C)
12.0 x 10^{-6}/°C (25-1400°C), slightly anisotropic

B-A transformation: 1600°C (reversible)

A-type, hexagonal structure (1650°C):
a = 3.845 ± 0.005 Å
c = 6.092 ± 0.005 Å

Density: 11.4 g/cm^3 (1650°C)

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[a] See reference [illegible]

Table 7. $^{244}Cm_2O_3$ FUEL PROPERTIES

BEING DEFINED IN GRNK PROGRAM

PROPERTY	USE SITUATION			
	OPERATION	REENTRY	LAUNCH PAD ABORT	TRANSPORTATION
Density	X			
Specific Power	X			
Power Density	X			
Radiation Properties	X			X
Radiation Stability	X			
Helium Release	X	X	X	
Thermal Conductivity	X	X	X	
Melting Point	X	X	X	
Critical Mass	X			
Vapor Transport	X	X	X	
Compatibility	I	I	I	
Coefficient of Expansion	X	X	X	
Vapor Pressure		X	X	
Heat Capacity		I	X	
Emissivity		O	O	
Heat of Fusion		O		
Heat of Vaporization		X		
Viscosity		O		
Surface Tension		O		
Impact Characteristics		O		
Leach Rates				I
Chemical Stability				I
Biological Health Aspects				I

X Complete
I Incomplete, in progress
O To be determined

Table 8. Summary of Radiation Characteristics of $^{244}Cm_2O_3$ and $^{238}PuO_2$

Energy		$^{244}Cm_2O_3$	$^{238}PuO_2$
		X-Ray Photons/sec/W	
0.017		1.7×10^{11}	1.5×10^{11}
		Gamma Photons/sec/W	
0.0-0.5		3×10^{8}	5.2×10^{8}
0.5-1.0		2×10^{7}	$\sim 5 \times 10^{5}$
1.0-2.0		4.9×10^{6}	
2.0-3.0		1.6×10^{6}	
3.0-4.0	S.F.	1.9×10^{5}	$\sim 1.1 \times 10^{4}$
4.0-5.0		6.4×10^{4}	
5.0-6.0		3.3×10^{4}	
6.0-7.9		5.6×10^{3}	
		Neutrons/sec/W	
S.F. Spectrum		4.6×10^{6}	5.6×10^{3}

Table 8A. Radiation from 36.2 Grams of Curium-244

Distance	N^f (mrem/hr)	γ (mR/hr)
18 inches	2500	200
1 meter	525	50

Table 9. Results of Critical Mass Calculations

Core		Reflector		Critical Mass (kg)	Critical Radius (cm)
Mixture[a]	Density (g/cm³)	Mixture	Thickness (cm)		
Cm_2O_3	10.60	Bare		21.1	7.8031
Cm_2O_3	9.01	Bare		29.2	9.1803
Cm_2O_3	10.60	Au-H_2O	4.0-15.0	11.9	6.4540
Cm_2O_3	10.60	Au-H_2O	2.0- 7.5	13.5	6.7188
Cm_2O_3	10.60	Au-H_2O	0.5- 2.0	16.5	7.1966

[a] 98.07 wt % ^{244}Cm; 1.93 wt % ^{241}Pu.

Table 10. Condition of Vapor Transport Experiments

Tube Number	Duration[a] (days)	Temp[b] (°C)	Pressure at Operating Temp[c] (Torr abs)
1	14 total	1904 average	116 average
2	7	2002	134
3	3	2100	177

[a] The times are accurate to ±5 min.

[b] The temperatures were determined by optical pyrometer. Corrections for window absorption and non black-body conditions were made using platinum and aluminum oxide melting point standards.

[c] The helium pressures are based on initial loading pressure plus calculated increase from curium decay.

Table 11. Rate of Diffusion of Cm_2O_3 in One Atmosphere Helium Pressure

Temp (°C)	(D)(C)[a] (g of Cm_2O_3 per cm·sec)
1903	3.50×10^{-8}
2002	14.5×10^{-8}
2100	76.0×10^{-8}

[a] D = diffusion coefficient in cm²/sec.
C = concentration of curium in the gas over the solid curium in g/cm³.