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DENSITIES OF SOME LOW MELTING PLUTONIUM ALLOYS*

Ralph H. Perkins, Louis A. Geoffrion, and John C. Biery

University of California
Los Alamos Scientific Laboratory
Los Alamos, New Mexico

1. Introduction

The Reactor Division of Los Alamos Scientific Laboratory is investigating the use of molten plutonium-containing fuels for fast reactor application. The molten fuel does not suffer from irradiation damage, so higher fuel burnup is feasible.

For the proper design and safe operation of a nuclear reactor, several of the physical properties of the fuel must be known to a high degree of accuracy. Two of the most important of these are the density of the fuel at operating temperature, and the change of density with temperature. Since all plutonium alloys that have been studied pass through a solid-liquid transition between room temperature and operating temperature, the change in density on melting must also be known.

The fuel used in LAMPRE (Los Alamos Molten Plutonium Reactor Experiment)

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was a Pu - 2.5 w/o (weight per cent) Fe eutectic alloy with a melting temperature of 411°C. However, limitations imposed on reactor design by this fuel caused consideration of other plutonium-containing alloys for use in future generations of this type of reactor. Several ternary alloys containing plutonium and cerium as two components have satisfactorily low melting points. The Pu-Ce-Co system at the present time appears to be the most acceptable. It exhibits little change in melting temperature with wide variation in plutonium concentration, which allows flexibility in reactor design. Other alloys that have received some consideration contain nickel, copper, and manganese as the third constituent.

These fuel alloys are difficult to handle experimentally since they oxidize readily, react with many solvents, and contain a poisonous fissionable material. The densities of these alloys must be known throughout the temperature range 25°-800°C and through the solid-liquid transition. The only experimental apparatus that appears suitable for measuring expansion coefficients of these materials under these experimental conditions is a volumeter.

While a volumeter is capable of measuring accurately the volume coefficient of expansion of materials, it cannot be used for absolute density measurements. Therefore, a density determination at a known temperature must be coupled with volumeter measurements to give the desired data. The weight loss technique using immersion in bromobenzene at room temperature proved to be satisfactory.

2. Equipment and Operating Procedure

The volumeter that was used in this investigation is described in detail elsewhere [1,2]. Briefly, it consists of two weld-sealed stainless steel containers of nearly equal volume located in a copper block inside a furnace. One container holds a tantalum crucible and the specimen being measured; the other contains a tantalum crucible and a tantalum specimen used as a control reference material. Stainless steel capillaries of equal length and volume connect each stainless steel container to a glass viewing capillary. A schematic diagram of this apparatus is shown in Fig. 1.

The working fluid that fills the specimen containers, the stainless steel capillaries, and portions of the glass capillaries is NaK (22 w/o Na - 78 w/o K). This alloy melts at -10°C and has a vapor pressure of 860 mm Hg at 800°C [3]. This relatively high vapor pressure at 800°C requires an overpressure of an inert gas to prevent boiling. The NaK and the fuel alloys that were studied are essentially insoluble in each other.

Fig. 1. Schematic Diagram of NaK Volumeter.

The room temperature densities of the specimens were determined by weight loss immersion in bromobenzene. All weighings were to ± 0.1 mg and the densities were accurate to ± 0.002 g/cc. The temperatures at which these measurements were made were known to $\pm 0.1^\circ\text{C}$.

Calibrated thermocouples were used for temperature measurements throughout the runs. The measurements made in the volumeter furnace have errors of less than $\pm 2^\circ\text{C}$ when all errors resulting from thermal stability and thermal asymmetries are considered.

The equations utilized in calculating the sample density are derived and presented in the paper by Knight [1]. The main equation necessary in these calculations is given below as Eq. 1.

$$C_S = - \{ \Delta V_D / \Delta T [1 - (T - T_G)(C_{M-C}) + (V_{FC} - V_{FS})(C_C) + (V_{KS} - V_{KC}) + (V_{MS} - V_{MC})(C_{M-h})] \} / V_S \quad (1)$$

The terms in this equation are defined as follows:

t = ambient, equilibrium temperature of the entire apparatus at time of initial data recording, $^\circ\text{C}$.

T = elevated temperature of container assemblies at time of observation, $^\circ\text{C}$.

ΔT = $T - t$, $^\circ\text{C}$.

T_G = temperature of glass capillaries and contained liquid medium at time of observation, $^\circ\text{C}$.

ΔV_C = observed volume displacement, between t and T , of liquid interface in capillary from control sample assembly, cm^3 .

ΔV_S = observed volume displacement, between t and T , of liquid interface in capillary from specimen container assembly, cm^3 .

ΔV_D = $\Delta V_C - \Delta V_S$, cm^3 .

V_{FC} = volume of steel control container at t , cm^3 .

V_{FS} = volume of steel specimen container at t , cm^3 .

V_{KC} = volume of tantalum crucible and tantalum sample in control container at t , cm^3 .

V_S = volume of specimen at t , cm^3 .

V_{KS} = volume of tantalum crucible in specimen container at t , cm^3 .

$$V_{MC} = V_{FC} - V_{KC} = \text{volume of NaK in } \underline{\text{control}} \text{ container at } t, \text{ cm}^3.$$

$$V_{MS} = V_{FS} - V_{KS} - V_S = \text{volume of NaK in } \underline{\text{specimen}} \text{ container at } t, \text{ cm}^3.$$

$$C_C = \text{cubic expansion coefficient of stainless steel container upon heating from } t \text{ to } T, \text{ cm}^3/\text{cm}^3/^\circ\text{C}.$$

$$C_K = \text{cubic expansion coefficient of tantalum upon heating from } t \text{ to } T, \text{ cm}^3/\text{cm}^3/^\circ\text{C}.$$

$$C_{M-h} = \text{cubic expansion coefficient of NaK upon heating from } t \text{ to } T, \text{ cm}^3/\text{cm}^3/^\circ\text{C}.$$

$$C_{M-c} = \text{cubic expansion coefficient of NaK upon cooling from } T \text{ to } T_G, \text{ cm}^3/\text{cm}^3/^\circ\text{C}.$$

$$C_S = \text{cubic expansion coefficient of specimen upon heating from } t \text{ to } T, \text{ cm}^3/\text{cm}^3/^\circ\text{C}.$$

The above equation with the required supporting equations and input data was processed on a 7094 digital computer. From C_S , the known temperature change, and the original sample density, the density of the sample was calculated from Eq. 2.

$$\rho_T(\text{specimen}) = \rho_t(\text{specimen}) / (1.0 + (T - t) C_S) \quad (2)$$

For many applications, a density coefficient is more useful than the volume coefficient. Therefore, the final density data were processed to produce coefficients which can be used in Eq. 3. Two densities at 0°C were

$$\rho_T = \rho_{0^\circ\text{C}} (1.0 - \alpha T) \quad (3)$$

calculated. A solid density for use with the solid phase portion of the curve was calculated by the linear extrapolation shown in Eq. 4.

$$\rho_{S0^\circ\text{C}} = \rho_t + (\rho_t - \rho_{T_1}) t / (T_1 - t) \quad (4)$$

In a similar manner, a pseudo liquid density at 0°C was calculated by extrapolating the liquid portion of the density curve back to 0°C with Eq. 5.

$$\rho_{L0^\circ\text{C}} = \rho_{T_N} + (\rho_{T_N} - \rho_{T_{N+1}}) (T_N) / (T_{N+1} - T_N) \quad (5)$$

In several of the experiments, small amounts of gas were evolved when the specimens became molten. This caused variations in height of NaK in the glass capillaries with changes in overpressure of helium above the NaK. The volume of this evolved gas at a given temperature could readily be determined by varying the pressure of the helium above the NaK between known values and observing the effect of this variation on the height of the NaK in the glass capillaries. The computer program was written so the final data were corrected for these evolved gases.

3. Results and Discussion

The apparatus was standardized and experience was gained in its operation by measuring the densities and coefficients of expansion of (1) nickel, (2) cerium, (3) Ce-Co alloys, (4) Ce-Ni alloys, (5) Ce - 15 w/o Cu alloy, and (6) Ce - 6 w/o Mn alloy. Cerium metal and all the binary cerium alloys that were studied except the Ce-Mn alloy increase in density on melting. The latter expands approximately 1% on becoming molten.

3.1 Plutonium Metal

Several investigators have measured the densities and phase transition temperatures of plutonium metal [1,4,5,6]. Unfortunately, the density of this metal is influenced by small amounts of impurities and by microcracks [6]. Since electrorefined plutonium containing less than 300 ppm impurities is now available, it appeared desirable to measure the density and coefficients of expansion of this material using the volumeter technique. The impurities detected in the plutonium that was used in this investigation are listed in Table I.

The density data for this material are plotted in Fig. 2. Experimental points were obtained on both heating and cooling. In most respects these data agree with the information obtained by other investigators [1,4,5,6]. The coefficients of expansion of all phases and the densities of the delta, delta prime, epsilon, and liquid phases are in excellent agreement with the best information that is available in the literature. Transition temperatures between the different phases also agree closely. However, the densities of the alpha, beta, and gamma phases that were obtained in this study are somewhat less than the values obtained by other investigators, apparently due to microcracks in the specimen [6].

The addition of gallium to plutonium metal stabilizes the delta phase at room temperature [7]. A specimen of Pu - 1 w/o Ga alloy was studied in the volumeter. The data are plotted in Fig. 2.

TABLE I
Analysis of Plutonium Metal

<u>Element</u>	<u>Concentration</u>
Pu	99.97%
Cu	5 ppm
Al	5 ppm
Am	123 ppm
Si	10 ppm

Fig. 2. Densities of Unstabilized and Stabilized (1 w/o Ga) Plutonium as Functions of Temperature.

At room temperature the specimen was almost completely delta plutonium, though a very small fraction existed as the alpha phase. As the temperature was raised this small amount of alpha was transformed successively to beta, gamma, and delta plutonium. At 325°C, a temperature at which unstabilized plutonium is in the delta phase, the densities of the unstabilized and stabilized materials are identical. However, the coefficients of expansion of the two materials are considerably different. While the coefficients of expansion of both unstabilized and stabilized are negative, the former exhibits a much larger (negative) coefficient.

The addition of gallium suppresses the transformation to the delta prime phase and makes the transformation to the epsilon phase sluggish and incomplete. While the delta prime - epsilon transition temperature is 480°C, a change in expansion coefficient of the stabilized plutonium did not become noticeable until a temperature of 500°C was reached. Between 500° and 585°C, the stabilized material exhibits a rather large negative coefficient of expansion. (The volume coefficient of expansion in this region is approximately -2.1×10^{-4} , as compared to -5.0×10^{-5} for unstabilized delta plutonium.)

In this temperature range, both delta and epsilon phases are stable. As the temperature is raised, more of the plutonium is converted to the epsilon phase, until at 585°C delta plutonium is not stable. Between 585° and 640°C, the epsilon Pu - 1 w/o Ga alloy exhibits a positive coefficient of expansion, though this expansion coefficient is somewhat less than that of unstabilized plutonium. The density of the stabilized material in this temperature range is also less than that of pure plutonium. This density difference is not due to the presence of Pu_3Ga , since the solubility of gallium in epsilon plutonium is 20%. However, the structure of the solid is apparently influenced by the gallium addition.

The Pu - 1 w/o Ga alloy begins to melt at $640 \pm 5^\circ\text{C}$, but the melting is not complete until a temperature of 665°C is reached. The density of the liquid at this temperature is 2% less than that of pure plutonium. Apparently the addition of gallium to plutonium not only produces an alloy that melts over a 25°C temperature range, but it also influences the structure of the liquid. This density difference between stabilized and unstabilized material becomes smaller as the temperature increases.

A small amount of gas came out of solution in the sample when it became molten. The height of NaK in the glass capillary connected to the sample container became dependent on the pressure of helium in the gas space above it. Compensations for this evolved gas were introduced into the calculations, and

data obtained in the liquid state were satisfactory. However, during cooling some of this evolved gas was trapped when the sample solidified. This entrapment caused the data points obtained on cooling to fall below those that were obtained on heating.

3.2 Pu-Fe

The fuel for LAMPRE was Pu - 2.5 w/o Fe alloy (10 a/o [atomic per cent] Fe) stabilized by the addition of 0.2 w/o Ga. The gallium was added to prevent damage to the tantalum container capsules by fuel expansion due to phase transitions during heating and cooling. In the solid state, this alloy is present principally as the intermetallic compound Pu_6Fe . Therefore, only 0.2 w/o gallium is required to stabilize the 30 a/o that is present as plutonium. Specimens of both stabilized and unstabilized Pu-Fe alloys were prepared for study in the volumeter. Analyses of these materials are shown in Table II.

The results of the volumeter runs on these materials are shown in Fig. 3. The curve obtained for the unstabilized alloy clearly shows the various phase transformations that occur in the solid state. This is a eutectic alloy, and it melts sharply at 411°C . The data obtained during the cooling portion of the cycle agree with those obtained on heating except in the regions where gamma and beta are the stable plutonium phases. Apparently the cooling rate was too rapid for the phase transitions to occur before the measurements were taken.

The fuel containing gallium stabilizer does not exhibit phase transitions in the solid state. The densities of the stabilized and unstabilized alloys are identical at 400°C . This stabilized alloy begins melting at 411°C , but it does not melt sharply. The alloy is not completely molten until a temperature of 432°C is reached. In addition, the density of the molten stabilized fuel at 435°C is 0.7% less than that of the stabilized material. Again the effect of gallium on density of plutonium appears in the liquid state as well as in the solid state. This difference in density between the stabilized and unstabilized liquid alloys becomes less as the temperature is raised (apparently due to destruction of the ordering effect attributed to the gallium).

The linear portions of the curves - the regions where the alloys are liquid, and the region between 25° and 325°C for the stabilized alloy - can be expressed mathematically in simple terms.

Unstabilized alloy:

$$410^\circ\text{C} < T < 800^\circ\text{C} \quad \rho_L = 16.94 - 1.64 \times 10^{-3} T$$

Stabilized alloy:

$$\begin{aligned} 25^\circ\text{C} < T < 325^\circ\text{C} \quad \rho_S &= 16.72 - 1.69 \times 10^{-3} T \\ 435^\circ\text{C} < T < 800^\circ\text{C} \quad \rho_L &= 16.77 - 1.42 \times 10^{-3} T \end{aligned}$$

TABLE II

Analyses of Stabilized and Unstabilized Pu-Fe Alloys

Element	Per Cent of Element	
	Unstabilized Alloy	Stabilized Alloy
Pu	97.55 $\pm$ 1	97.5 $\pm$ 1
Fe	2.38 $\pm$ 0.1	2.38 $\pm$ 0.1
Ga	-	0.19
Am	0.0288	0.0011
Al	0.0020	0.002
Ni	< 0.010	< 0.010
Cu	< 0.002	< 0.002
C	< 0.010	< 0.012

Fig. 3. Effects of Temperature on the Densities of Stabilized (0.2 w/o Ga) and Unstabilized Pu - 2.55 w/o Fe Alloys.

3.3 Pu-Co-Ce

The fuel for the first core of the FRCTF (Fast Reactor Core Test Facility) will be a Pu-Co-Ce alloy containing 6.2-8.0 g Pu/cc. If a large power reactor based on this fuel system is built, it must utilize a fuel of lower plutonium concentration - perhaps as low as 3.0 g Pu/cc. To obtain a complete picture of the densities of potential fuels in this system, volumeter samples of low-melting alloys containing nominally 3.0, 5.0, 6.2, and 8.0 g Pu/cc were prepared. Alloys containing higher concentrations were not studied since they are apparently too corrosive for the container materials that are presently being considered. Analyses of these alloys are shown in Table III.

The density data obtained on the alloy containing nominally 3.0 g Pu/cc are plotted in Fig. 4. There are no phase transitions in the solid state, and the experimental points fall on a straight line. Sufficient cerium is present to stabilize the plutonium in the delta phase. The alloy melts over a very narrow temperature range at $425 \pm 2^\circ\text{C}$. There is a density increase of 1.3% on melting. A straight line also fits the data points in the molten region. The density of this alloy at a given temperature can be calculated from the following equations:

$$\begin{aligned} 25^\circ\text{C} < T < 425^\circ\text{C} \quad \rho_S &= 9.080 - 6.80 \times 10^{-4} T \\ 430^\circ\text{C} < T < 800^\circ\text{C} \quad \rho_L &= 9.221 - 7.43 \times 10^{-4} T \end{aligned}$$

The specimen evolved a small amount of gas when it melted. While the sample was molten, the amount of gas could be calculated and compensations were made for it. However, when the sample was solidified, it trapped some of this gas, and the density readings were erroneously low. Additional lowering of the temperature cracked the sample in a number of places. These cracks contacted some of the gas bubbles, and the NaK filled these voids. At 25°C , the density agreed with that originally measured at this temperature.

Volumeter runs were made on two Pu-Co-Ce samples containing nominally 5.0 g Pu/cc. Although the two samples came from the same fuel batch, their densities at 25°C differed by 1.3%. The density curves for these two specimens are plotted in Fig. 5.

In the solid state, the two specimens exhibited the same coefficients of expansion. Although the two curves are displaced from one another, they are parallel. The data obtained with solid samples can best be fit with a slightly curved, rather than a straight, line. Both specimens began melting at $438 \pm 2^\circ\text{C}$. However, the less dense sample did not become completely molten until a temperature of 500°C was reached, while the other specimen was molten at 465°C . The slopes of the two curves in the liquidus region are somewhat different.

TABLE III
Analyses of Pu-Co-Ce Alloys

Element	Weight Per Cent of Element			
	3 g Pu/cc Alloy	5 g Pu/cc Alloy	6.2 g Pu/cc Alloy	8 g Pu/cc Alloy
Pu	34.2 $\pm$ 0.5	50.0 $\pm$ 1	61.0 $\pm$ 1	68.9 $\pm$ 0.7
Co	10.1 $\pm$ 0.2	9.5 $\pm$ 0.2	9.1 $\pm$ 0.2	8.0 $\pm$ 0.2
Ce	55.7 $\pm$ 0.8	41.7 $\pm$ 0.8	29.9 $\pm$ 0.6	23.0 $\pm$ 0.2
Am	0.0065	-	0.0035	0.0065
Fe	0.0200	0.0115	0.0117	0.0225
Al	0.0085	0.0040	0.0090	0.013
O	0.0025	0.005	0.042	0.058
Mg	0.017	0.016	0.017	0.011
C	0.006	0.0025	0.004	0.0015
Ni	0.011	0.0040	0.011	0.0220
Cr	0.0025	< 0.002	0.004	0.0055
Mn	0.015	0.013	0.009	0.010
H	-	0.001	0.006	0.004

Fig. 4. Density of a Pu-Co-Ce Alloy Containing Nominally 3.0 Grams Pu/cc as a Function of Temperature.

Fig. 5. Densities of Two Pu-Co-Ce Samples Containing Nominally 5.0 g Pu/cc as Functions of Temperature.

The more dense specimen increased in density by 1.8% on melting, while the less dense sample decreased in volume by 1.0%.

Linear expressions that closely approximate the solid and liquid regions for the more dense alloy are as follows:

$$\begin{aligned} 25^{\circ}\text{C} < T < 435^{\circ}\text{C} & \quad \rho_S = 10.40 - 7.80 \times 10^{-4} T \\ 465^{\circ}\text{C} < T < 800^{\circ}\text{C} & \quad \rho_L = 10.68 - 9.65 \times 10^{-4} T \end{aligned}$$

The two samples of nominally 6.2 g Pu/cc Pu-Co-Ce alloy behaved almost identically in the volumeter runs. At 25°C, their densities differed by only 0.12%, and they were essentially identical over the entire temperature range. The samples melted sharply at $440 \pm 2^{\circ}\text{C}$, and the alloy exhibited a 2.2% increase in density on melting. A plot of the data for these two tests is shown as Fig. 6.

While the density curve in the solid region is not linear below 100°C, the portion between 150° and 435°C can be approximated by a linear expression. The region between 450° and 800°C also is linear. These portions of the curve for the more dense sample fit the following equations.

$$\begin{aligned} 150^{\circ}\text{C} < T < 435^{\circ}\text{C} & \quad \rho_S = 10.944 - 8.37 \times 10^{-4} T \\ 450^{\circ}\text{C} < T < 800^{\circ}\text{C} & \quad \rho_L = 11.215 - 9.27 \times 10^{-4} T \end{aligned}$$

Again gas bubbles were entrapped by the samples on freezing and the density data obtained on cooling in the solid state were low.

Volumeter runs were also made on two samples of a Pu-Co-Ce alloy containing approximately 8.0 g Pu/cc. This information is plotted in Fig. 7. The densities of the two samples differed by 0.66% at 25°C, and the specimens exhibited very similar expansion characteristics throughout the runs. They both began melting at $415 \pm 3^{\circ}\text{C}$, but they did not become completely molten until a temperature of $445 \pm 3^{\circ}\text{C}$ was reached. Although both samples evolved gas on melting, apparently only a small fraction of this was trapped in the samples when they solidified. Therefore, the heating and cooling data are not significantly different.

Equations that fit the linear portions of the density curve for the most dense sample are as follows:

$$\begin{aligned} 200^{\circ}\text{C} < T < 410^{\circ}\text{C} & \quad \rho_S = 11.75 - 7.85 \times 10^{-3} T \\ 445^{\circ}\text{C} < T < 800^{\circ}\text{C} & \quad \rho_L = 12.164 - 8.78 \times 10^{-4} T \end{aligned}$$

3.4 Mn Additions to Pu-Co-Ce

Plutonium and the plutonium-containing alloys that were studied all expand on freezing. In the Pu-Co-Ce system, there is an increase in this expansion with increasing plutonium concentration. This expansion on freezing is a

Fig. 6. Densities of Two Pu-Co-Ce Samples Containing Nominally 6.2 g Pu/cc as Functions of Temperature.

Fig. 7. Densities of Two Pu-Co-Ce Samples Containing Nominally 8.0 g Pu/cc as Functions of Temperature.

potential source of trouble in a nuclear reactor. While it is intended that the fuel be molten at all times in an operating system, some unforeseen loss of power could result in freezing of the fuel in the reactor core. Under certain freezing conditions the container capsules might be bulged or ruptured. While this problem may be eliminated by controlling the freezing conditions or increasing the strength of the container material, it is more desirable to modify the fuel system to eliminate or reduce the expansion on freezing.

As was previously stated, the Ce-Mn alloy is the only binary cerium-containing alloy of those investigated that becomes more dense (by approximately 1%) on freezing. Alloys in the Pu-Mn-Ce system with plutonium concentrations less than 8 g/cc all melt at temperatures in excess of 600°C [8], so they are not suitable for use in a molten fuel system. An alternate approach is the addition of Mn to Pu-Co-Ce fuels to reduce their expansion on freezing.

Specimens of alloys containing 6.5 and 8.0 g Pu/cc with Mn additions were prepared and studied with the volumeter. The results of this investigation are summarized in Table IV.

The alloy containing 3.0 w/o Mn became completely molten in the narrow range 432-435°C. The alloys with higher manganese contents, however, did not melt completely until higher temperatures were reached. A radiograph of the capsule containing the 9.0 w/o Mn alloy at the conclusion of the volumeter study revealed that it had not become completely molten during the experiment. In all cases, however, the expansion on freezing occurred in a narrow temperature range just above the temperature where the sample began to melt.

Manganese additions reduce the expansion on freezing of Pu-Co-Ce alloys, but do not entirely eliminate it. A limited amount of differential thermal analysis studies indicates that larger additions of manganese to fuels significantly raises their melting temperatures [8]. Because of the relatively high temperatures at which these quaternary alloys become completely molten, they are not suitable fuels for nuclear reactors. Therefore, no additional studies are planned on this system.

4. Summary and Conclusions

Densities of unstabilized and stabilized (1 w/o Ga) plutonium were measured over the temperature range 25°-800°C. Gallium not only stabilizes the delta phase of plutonium in the solid state, but also influences its melting temperature and the density of the liquid metal. The Pu - 2.5 w/o Fe and the Pu - 2.5 w/o Fe - 0.2 w/o Ga alloys were also studied with the volumeter. The unstabilized alloy melts at $411 \pm 2^\circ\text{C}$ with a 1.5% decrease in

TABLE IV

Effects of Mn Addition on Expansion of Pu-Co-Ce Alloys on Freezing

Nominal g Pu/cc	Wt.% Pu	Wt.% Ce	Wt.% Mn	Melting Temp. (°C)	% Expansion on Freezing
6.5	60.8	30.3	0	440-442	2.3
6.5	60.2	26.3	3.0	432-435	2.0
6.5	59.1	22.2	5.5	440-442	1.5
6.5	58.7	19.2	9.0	440-442	1.0
8.0	68.1	23.9	0	415-445	3.0
8.0	67.1	15.2	7.5	420-435	1.8

volume. Gallium addition suppresses phase transitions in the solid state and affects melting temperature and density of the molten alloy.

Pu-Co-Ce alloys containing 3, 5, 6.2, and 8 g Pu/cc were studied with the volumeter. These alloys all melt in the range 425°-442°C, and they become more dense on melting. The change in density on melting increases with increasing plutonium content from 1.3% for the 3 g Pu/cc alloy to 3% for the 8 g/cc material. The addition of manganese to 6.2 and 8 g/cc alloys significantly reduced this expansion on freezing. However, manganese additions of 5.5 w/o and higher adversely affected the melting temperatures of these fuels.

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