PLUTONIUM ALLOYS CONTAINING CONTROLLED AMOUNTS OF PLUTONIUM ALLOTROPES OBTAINED BY APPLICATION OF HIGH PRESSURES

Fig. 1

WITNESSES:
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Fig. 2

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Reed O. Elliott
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BY
[Signature]
Fig. 4

TRANSFORMATION PRESSURE, ATMS

0 10,000
9,000
8,000
7,000
6,000
5,000
4,000
3,000
2,000
1,000

0 1 2 3 4 5 6

ATOM PERCENT ZINC

TRANSFORMATION PRESSURE

INVENTOR.
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Karl A. Gschneidner, Jr.

WITNESSES:
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Victor M. Land
PLUTONIUM ALLOYS CONTAINING CONTROLLED AMOUNTS OF PLUTONIUM ALLOTROPES OBTAINED BY APPLICATION OF HIGH PRESSURES

Filed April 18, 1960

Fig. 5

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Fig. 6
Fig. 7
Fig. 9

[Graph showing transformation pressure vs. atom percent cerium]
Fig. 11

H.T. & C. - AFTER HEAT TREATMENT AND COMPRESSION
H.T. - AFTER HEAT TREATMENT ONLY

(a) ALUMINUM

H.T. & C. (THROUGH BOTH TRANSFORMATIONS)
H.T. & C. (FIRST TRANSFORMATION ONLY)

(b) ZINC

H.T.

(c) CERIUM

ATOMIC PERCENT

DENSITY (GM/CM$^3$)

0 2 4 6 8 10 12 14

0 2 4 6 8 10 12 14

0 2 4 6 8 10 12 14
WITNESSES:

Alum

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Fig. 12
Fig. 14

WITNESSES:

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INVENTOR.

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BY

Roland C. Anderson
The present invention deals with alloys of plutonium containing minor amounts of delta stabilizing elements, and is particularly concerned with such alloys in which the final allotropic composition of the plutonium is a predetermined ratio dictated by the physical characteristics required in the end product, together with methods for obtaining such alloys. Such alloys are all useful, in the form of solid rods or other shapes, as nuclear reactor fuels in that type of reactor generally described as a "fast" reactor, i.e., one in which the neutrons causing fission have essentially the energies with which they are released in the fission process, and most are also useful in intermediate and thermal systems, i.e., those reactors in which the energies of the fission neutrons are degraded by collisions with moderator nuclei before combining with fissile nuclei.

As is now well established, unalloyed plutonium has six allotropic forms in the solid state, each form existing under equilibrium conditions at a particular range of temperatures and each having a distinct set of physical characteristics. Thus the alpha allotrope, the stable form at room temperatures, has the highest density, 19.7 g./cc., the greatest strength and hardness, and a positive coefficient of thermal expansion, while the delta allotrope has the lowest density, 15.9 g./cc., lower strength and hardness but greater ductility than alpha plutonium, and a negative volume coefficient of thermal expansion. The superior strength and hardness of alpha plutonium make it exceedingly difficult to fabricate by ordinary methods, though the same quantities are highly desirable in the final fuel piece. On the other hand, the superior ductility of delta plutonium renders it readily machinable, though it is inferior to the alpha form in strength and hardness, and has the further unfortunate characteristic of a negative coefficient of thermal expansion. The latter implies a contraction of the metal on heating, and is undesirable in a nuclear fission reactor fuel, as such a contraction leads to greater reactivity and thus imposes a greater control problem.

Past efforts to develop solid plutonium fuels which are capable of ready fabrication have been concentrated on methods of preserving or stabilizing the delta allotrope at room temperature. Since there is no method known for thus stabilizing the delta phase of unalloyed plutonium, such methods have invariably consisted in adding a minor amount of an element such as aluminum to the plutonium, heating the mixture to an elevated temperature to convert to the delta allotrope and insure a homogeneous mixture, and rapidly cooling the same room to room temperature to insure a maximum retention of the delta phase. This product is generally referred to as delta stabilized plutonium, and is apparently quite stable at room temperature and atmospheric pressure, provided it is not cold worked. The final step of the prior art is the machining of such a delta stabilized plutonium shape to final dimensions, after which it is either used directly or is given a second heat treatment to reconvert to the delta phase any plutonium transformed to other allotropes in the fabrication process.

While solid fuel rods thus containing plutonium entirely or predominantly in the delta state are satisfactory for some purposes, their employment is not altogether free of disadvantages. Small voids and cracks are usually present in the final product as a result of the quenching step and/or the inevitable partial conversion to the alpha phase, however small. Such flaws are undesirable because they make fabrication more difficult and, if retained in the final piece, because they decrease the nuclear efficiency of the reactor.

In addition, many such alloys preserve the negative coefficient of thermal expansion characteristic of delta plutonium, e.g., delta plutonium stabilized with aluminum requires a minimum addition of 2.5 atom percent (a/o) aluminum to insure a positive coefficient, and delta plutonium stabilized with cerium requires a minimum of 6.7 a/o cerium to achieve the same result.

Another disadvantage of such prior art fuel alloys is the fact that their densities are essentially fixed. Since the plutonium is essentially all of the delta phase and the additive is usually of low molecular weight or is added in a minor proportion, or both, the density is essentially that of the delta allotrope. This inflexibility has made it necessary for reactor designers to design to such density, whereas in many cases a higher density could conveniently have been exploited if it had been available.

As indicated above, the prior art delta stabilized plutonium obtained by the rapid cooling of the metal with a small alloying addition is apparently quite stable at room temperature and atmospheric pressure. The condition, however, is not one of equilibrium, as the delta allotrope transforms to other phases when the alloy is heat treated, compressed or worked, either hydrostatically or mechanically, as by rolling. The alloys are only metastable, and transform to a more stable, lower energy state under outside influences. (Although energy must be supplied to the alloy to initiate the delta-alpha transformation, the plutonium gives up more than this energy in the transformation.) Such transformations are undesirable in fuel rods during the course of operation, as the concomitant changes in physical properties will cause drastic changes in the mode of operation of the reactor, and may even produce conditions which necessitate a shutdown of the reactor. For one example, if in a jacketed solid fuel rod there is a sudden transformation from delta to alpha plutonium, the contraction may rupture the jacket and thus permit leakage of the fuel and fission products to the other parts of the reactor, a mishap which would require a lengthy shutdown for decontamination of such parts and replacement of the ruptured fuel rod.

Through a course of considerable experimental investigation by the present inventors, it has been discovered that high pressure techniques may be used to overcome many of the aforementioned defects of the prior art. The present inventors have found that most delta stabilized plutonium alloys may be partially transformed into the alpha phase (and one into beta) by pressure alone. In the course of such pressure treatment, any previously present voids and cracks are filled up, and new ones are introduced. In most such pressure treatments, the transformation is irreversible, i.e., no reverse transformation takes place when the pressure is released. For certain alloying additions, however, the present inventors have also discovered that with a certain mini-
The delta stabilized alloys prepared as above were subjected to high pressure in a high pressure cylinder having a bore approximately equal to the diameter of the specimens. The specimen was placed inside this bore on a stationary piston, and pressure was applied through a movable piston and transmitted hydraulically to the specimen by glycerin, the latter being used as the pressure-transmitting medium because it does not react with plutonium and because it is relatively incompressible. A Baldwin SR-4 load cell, calibrated for pressure measurement at the solidification point of mercury, 8850 atmospheres and 7.5°C, was used to measure the pressures. Dial indicators located on opposite sides of the cylinder were used to measure linear displacement of the movable piston. Volume changes in the specimens required not only calculation from the dial indicator readings, but also corrections for compression of the glycerin, expansion of the cylinder and contraction of the pistons, resulting in calculated volumes with an accuracy of ±10% of the values listed. Pressures required correction for the frictional resistance of the pistons.

The present invention may be more readily understood by referring to the attached drawings, in which:

FIGURE 1 is a set of experimental compression curves for various Pu–Al alloys in accordance with the method outlined above.

FIGURE 2 contains a curve showing the transformation pressure of such Pu–Al alloys as a function of the atom percent aluminum.

FIGURES 3 and 4 present the same type of compression information as in FIGURES 1 and 2 for various Pu–Zn alloys.

FIGURE 5 is a similar compression curve for two alloys of plutonium and indium.

FIGURE 6 presents the same type of compression characteristics as in FIGURES 1, 3 and 5 for one alloy each of Pu–Zr and Pu–Hf.

FIGURE 7 is a pressure-volume characteristic for one Pu–Er alloy.

FIGURE 8 presents compression curves for various Pu–Ce alloys, showing both the low pressure transformations and the high pressure transformations.

FIGURE 9 presents the transformation pressure of delta stabilized plutonium as a function of the atom percent cerium in the alloy for both transformations.

FIGURE 10 is a set of compression curves for Pu–Ce alloys, similar to those of FIGURE 8 but limited to the low pressure transformations.

FIGURE 11 presents the densities of plutonium alloys containing aluminum, zinc and cerium as a function of the atom percent alloying element, both before compression and after compression.

FIGURE 12 shows the dependence of hardness on atom percent of additive both before and after compression.

FIGURE 13 contains the phase diagrams of each of the three systems Pu–Al, Pu–Zn and Pu–Ce, and FIGURE 14 depicts the percent alpha and delta allotropes of plutonium as a function of the percent delta additive in the alloy, showing how a particular predetermined allotropic distribution can be obtained with various additions of such elements in various proportions.

In addition to presenting the results of the indicated experiments in graphical form in the attached drawings, such results plus additional results not presented graphically are listed below in Table I. Transformation pressures listed in this table were obtained by extrapolating the pressure-volume curves at the transformation breaks.

Volumes of transformation were also measured before and after compression, using a method which gave densities to a precision of ±0.03 g/cc. The hardnesses of the specimens were likewise measured before and after compression.
The same is true for the compression curves of the erbium (FIG. 7), for which the newer testing device was used, and the slight volume changes thereafter indicate that the volume begins to increase immediately after the pressure is released, and that the descending branch of each curve should be shifted slightly to the left. The same is true for the compression curves of the other alloys described below with the exceptions of those for indium (FIG. 5), zirconium and hafnium (FIG. 6) and erbium (FIG. 7), for which the newer testing device was used.

As can be seen in FIG. 2, the transformation pressure varies linearly with the atom percent aluminum in the specimen. These linear characteristics have been extended to zero pressure, and the extrapolated zero transformation pressure is interpreted as determining the minimum amount of aluminum, 1.0 a/o, required to retain delta phase plutonium at room temperature with the heat treatment being used—homogenization in the delta field of plutonium and an air quenching. To verify this assumption, two alloys containing 0.8 and 1.2 a/o Al were thus treated and examined. Upon examination, the 0.8 a/o Al alloy was found to consist mostly of alpha phase, and had a density of 11.7 g/cc. It is here noted that such minimum additive will vary somewhat with the size of the alloy specimen and with the quenching rate, e.g., a water quench or a quench in liquid nitrogen will stabilize the delta phase with a somewhat lower alloying addition than an air quench, whereas air quenching of a specimen several inches in each dimension will require a higher content of the delta stabilizing additive.

Turning to FIG. 3, it can be seen that all of the zinc alloys tested under compression, maximum 3.9 a/o Zn, transformed irreversibly under pressure. The data from FIG. 3 was used to obtain the graphs of FIG. 4 in the same manner as for the aluminum alloys, the extrapolated transformation pressure curve in this case indicating a minimum zinc content of about 1.1 a/o to stabilize the delta phase with the heat treatment used.

In FIG. 5, the compression curve for the Pu–In alloy containing 4.0 a/o In indicates another irreversible transformation of delta into a mixture of alpha plus delta, the transformation commencing at a pressure of 830 atmospheres. It should be noted (see Table 1) that the curve for the 2.5 a/o In alloy represents a test in which the plutonium prior to compression was essentially pure alpha phase. This was necessary because it is not possible to

### Table 1

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<tr>
<th>System</th>
<th>Composition (atomic percent additive)</th>
<th>Maximum Pressure (atm.)</th>
<th>Transformation Pressure (atm.)</th>
<th>Transformation Volume Change, a/o</th>
<th>Compressibility (X 10^9 atm.)</th>
<th>Before Compression (gm./cc.)</th>
<th>After Compression (gm./cc.)</th>
<th>Density</th>
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<tr>
<td>Pu-AI</td>
<td>1.7</td>
<td>8.840</td>
<td>2.940</td>
<td>16.8</td>
<td>16.0</td>
<td>4.1</td>
<td>15.73</td>
<td>18.67</td>
</tr>
<tr>
<td>Pu-Zn</td>
<td>2.5</td>
<td>8.820</td>
<td>4.580</td>
<td>11.6</td>
<td>10.9</td>
<td>3.8</td>
<td>15.69</td>
<td>17.70</td>
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<td>Pu-Ce</td>
<td>3.4</td>
<td>8.790</td>
<td>N.T.</td>
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<td>8.5</td>
<td>4.0</td>
<td>16.69</td>
<td>16.71</td>
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<td>4.0</td>
<td>10.050</td>
<td>N.T.</td>
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<td>10.9</td>
<td>3.4</td>
<td>15.50</td>
<td>15.45</td>
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<td>5.0</td>
<td>8.720</td>
<td>N.T.</td>
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<td>8.5</td>
<td>4.5</td>
<td>15.31</td>
<td>15.30</td>
</tr>
<tr>
<td>Pu-Zn</td>
<td>6.0</td>
<td>8.720</td>
<td>N.T.</td>
<td>8.5</td>
<td>8.5</td>
<td>4.5</td>
<td>15.31</td>
<td>15.30</td>
</tr>
<tr>
<td>Pu-Ce</td>
<td>7.5</td>
<td>8.700</td>
<td>N.T.</td>
<td>8.5</td>
<td>8.5</td>
<td>4.5</td>
<td>15.31</td>
<td>15.30</td>
</tr>
<tr>
<td>Pu-In</td>
<td>8.0</td>
<td>8.700</td>
<td>N.T.</td>
<td>8.5</td>
<td>8.5</td>
<td>4.5</td>
<td>15.31</td>
<td>15.30</td>
</tr>
<tr>
<td>Pu-Zr</td>
<td>9.0</td>
<td>8.700</td>
<td>N.T.</td>
<td>8.5</td>
<td>8.5</td>
<td>4.5</td>
<td>15.31</td>
<td>15.30</td>
</tr>
<tr>
<td>Pu-Zn</td>
<td>10.0</td>
<td>8.700</td>
<td>N.T.</td>
<td>8.5</td>
<td>8.5</td>
<td>4.5</td>
<td>15.31</td>
<td>15.30</td>
</tr>
</tbody>
</table>

* Values obtained by extrapolation of pressure-volume curves.

b N.T. means no transformation.
stabilize the delta phase with such a low fraction of additive, even by the most rapid quenching. It should also be noted that the other data in Table I, not graphically presented in FIG. 6, show irreversible pressure transformations of Pu-In alloys containing 3.4 a/o In and 5.0 a/o In. A transformation pressure-atomic percent indium characteristic similar to those in FIG. 2 for aluminum and FIG. 3 for zinc may be prepared from the data tabulated for indium in Table I. Density measurements of heat treated alloys shows that the minimum indium content necessary for retention of the delta phase at room temperature is 3.6 a/o with the heat treatment used.

In FIG. 6 there are presented one pressure-volume characteristic each for alloys of Pu-Zr and Pu-Hf with the plutonium stabilized in the delta phase by the heat treatment described above, i.e., homogenization at an elevated temperature in the delta field followed by a fairly rapid quench. While the one high zirconium alloy tested (10.0 a/o) exhibited only reversible transformation, it is fairly certain that alloys with lesser zirconium contents will transform irreversibly. Density measurements on a large number of heat treated but unpressurized specimens indicate that such alloys retain the delta phase plutonium at room temperature with zirconium contents from a minimum of 7.0 atomic percent (a/o) to at least 20.0 a/o, the density varying linearly with zirconium content in the interval. Alloys containing from 7.0 a/o to zero Zr increase abruptly in density, indicating almost complete conversion to alpha-Pu on cooling.

The pressure-volume curve for the delta-Pu 6.0 a/o Hf alloy of FIG. 6 shows an irreversible and complete transformation of the plutonium to the alpha phase, the transition beginning at a pressure of about 2350 atmospheres and being completed at a maximum pressure of 9900 atmospheres. Density data for various Pu-Hf alloys obtained by heat treating as above indicate the minimum hardness addition for stable retention of alpha-Pu under standard temperature and pressure to be about 4.5 a/o.

The one pressure-volume characteristic for a Pu–6.0 a/o Er alloy in FIG. 7 indicates a complete and irreversible transformation of plutonium from the delta phase to the alpha phase, the transformation pressure being about 1700 atmospheres and the maximum pressure 9900 atmospheres. Reference to Table I indicates that similar alloys containing erbium additions of 4.0 a/o and 8.0 a/o are likewise completely and irreversibly transformed under pressure. Density data obtained on heat treating a number of Pu-Er alloys indicate that the minimum erbium content of the binary alloy for stable retention of delta Pu at standard temperature and pressure is 4.5 ±0.5 a/o.

Alloys containing cerium in the range 1.0–4.5 a/o may be heat treated to stabilize plutonium in the delta phase, but are not thermodynamically stable, and will slowly transform, albeit imperceptibly, under standard temperature and pressure conditions.

The density curves for the cerium alloys in FIG. 11c show both the densities after compression through the first transformation and densities after the second, high pressure transformation. The intersection of the former with the before compression curve occurs at 7.1 a/o Ce. Delta stabilized alloys containing less than this amount of cerium transform irreversibly under compression into mostly beta phase, while delta stabilized alloys containing more than 7.1 a/o Ce are thermodynamically stable at room temperature and will transform reversibly under compression.

The extrapolated curves obtained before compression and after the high pressure transformation intersect at 16.3 a/o Ce, indicating alpha stabilized alloys containing more than 16.3 a/o Ce will transform from delta to alpha as the pressure is increased, and from alpha to beta as the pressure is decreased, i.e., both transformations are completely reversible. The critical points for these reversible trans-
formations of Pu–Al and Pu–Ce alloys are summarized in Table II. As indicated in FIG. 11b, all of the Pu–Zn alloys transformed irreversibly from delta to alpha at pressures below 10,000 atmospheres. Insufficient data is available for determination of the existence of critical points of the Pu–Zr, Pu–Hf, Pu–In and Pu–Er alloys.

### Table II

<table>
<thead>
<tr>
<th>Critical Composition</th>
<th>Transformation (Reversible)</th>
<th>Transformation Pressure (atm.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.5 a/o Al</td>
<td>delta to alpha</td>
<td>10,200</td>
</tr>
<tr>
<td>7.1 a/o Ce</td>
<td>delta to beta</td>
<td>3,800</td>
</tr>
<tr>
<td>10.3 a/o Ce</td>
<td>beta to alpha</td>
<td>20,500</td>
</tr>
</tbody>
</table>

The hardeneses of the various alloys as a function of atom percent additive both before and after compression are presented graphically in FIG. 12, the after compression values for cerium in this case being for the high pressure transformation. Hardness values for such cerium alloys after only the first transformation are intermediate between the values before the first transformation and after the second, e.g., 67 DPHN before compression, 118 DPHN after the first compression and 237 DPHN after the second transformation for an alloy containing 4.0 a/o Ce. The curves obtained for the delta stabilized alloys prior to compression indicate in general little variation in hardness prior to transformation, while the hardness values obtained after compression and transformation indicate rapid increases in hardness with decreasing content of the delta stabilizing element. These curves also indicate that hardeneses can be controlled over a limited range by selecting the proper combination of alloying element and pressure. They also indicate that for a particular preselect ed hardness of the transformed alloy the smallest dilution of the plutonium will be obtained with aluminum, followed by cerium and zinc in the order indicated.

The plutonium-rich portions of the phase diagrams of the Pu–Al, Pu–Zn and Pu–Ce systems are reproduced in FIG. 13. Effects of pressure on metastable, single-phase, binary alloys do not appear to have been determined previously. Some interesting correlations between the behaviors of such alloys under pressure and their respective phase diagrams can be inferred from the results of the present invention.

The intersection of the Pu–Al density curves at 4.5 a/o Al, referred to earlier in this specification, can be extrapolated to atmospheric pressure by assuming that this composition does not vary with pressure. This composition is then interpreted to be on the alpha plus delta/delta solvus phase boundary at room temperature (see FIG. 14a). This value, 4.5 a/o Al, is intermediate between the reported British value of 2.0±0.2 a/o Al and the Los Alamos value of 8.0 or 9.0 a/o Al, but these results are in disagreement with the Russian version of the Pu–Al diagram.

The latter version shows a eutectoid decomposition of delta phase into beta phase plus Pu5Al at about 175° C, and on cooling to room temperature alpha phase plus Pu5Al becomes the equilibrium state of such decomposed alloys. No evidence was found by the present inventors to substantiate this type of Pu–Al diagram. The metastable delta Pu–Al alloy transformed under pressure into either alpha or mixtures of alpha plus beta with some untransformed delta, and at no time was Pu5Al identified in any of the alloys by microscopic or X-ray methods, either before or after compression.

The intersection of the Pu–Ce density curves at 7.1 a/o Ce can also be extrapolated to atmospheric pressure in the same way as described above for the Pu–Al alloys. This composition represents the beta plus delta/delta solvus phase boundary, and it agrees reasonably well with the value of 5 a/o Ce, obtained by extrapolation of high-temperature X-ray and metallographic data (see FIG. 13c).

It had been calculated that pure delta plutonium will transform to either gamma or delta-prime at 1300 atmospheres and at elevated temperatures, i.e., at pressures greater than 1300 atmospheres no delta phase can be formed regardless of temperature. As can be seen in Table II, a delta-stabilized alloy containing 4.5 a/o Al will transform reversibly from delta to alpha at 10,000 atmospheres whereas a delta-stabilized alloy containing 7.1 a/o Ce will transform reversibly to the beta phase at 3800 atmospheres. These reversible transformation pressures increase with increases of alloy elements (see FIGS. 2 and 9). Thus, the addition of aluminum or cerium has the effect of extending the delta-phase field with respect to pressure, similar to the effect produced by these alloy additions in extending the delta-phase field with respect to temperature (see FIGS. 13a and 13c). The stability of beta phase Pu–Ce alloys after compression suggests that cerium is probably more soluble in beta than in the gamma or alpha phases.

The Pu–Zn diagram shows a eutectoidal decomposition of delta phase into gamma phase plus PuZn2 at about 235° to 243°, and alpha phase plus PuZn2 corresponds to the equilibrium state of decomposed alloys at room temperature (see FIG. 13a). Delta-stabilized Pu–Zn alloys are, therefore, actually metastable at room temperature and should transform under pressure to a more stable state. All delta-phase Pu–Zn alloys studied did transform under compression, substantially in agreement with this Pu–Zn phase diagram.

To enable those skilled in the art to practice the present invention, there is presented in FIG. 14 a series of graphs indicating the allotropic compositions of various delta stabilized plutonium alloys, each as a function of the atom percent delta stabilizing additive. These curves all represent the allotropic composition after maximum compression, i.e., compression to such a high pressure that virtually no change in density is obtained thereafter. Such curves have been derived from the density data of Table I and are easily reinterpreted into densities by the use of FIG. 11. If it is desired that the plutonium be equally distributed between the alpha and delta phases, FIG. 14 indicates that this can be done by alloying additions of any one of 2.9 a/o Al, 6.0 a/o Zn or 10.0 a/o Ce. These alloys, as well as those of In, Zr, Hf and Er, are prepared by the heat treatment outlined above, viz., homogenization in the delta field and air quenching to room temperature followed by the transformation under pressure as indicated in FIGS. 1, 3, 5, 6, 7 and 8. The choice of additive will depend to some extent upon the dilution which can be tolerated in the design and the hardness desired in the final product, and some compromise may be necessary. The hardness will be determined in accordance with FIG. 12, and the dilution by the neutron calculations of the designer. Regardless of the particular alloying elements selected, the final product will be free of cracks and voids, and will be stable at room temperature and temperatures at least within the alpha range of unalloyed plutonium and at pressures likely to be encountered in an operating reactor, e.g., pressures less than about 8000 atmospheres.

It is here pointed out that it is possible to obtain alloys of alpha and delta plutonium for a particular alloying addition other than the one used to obtain the appropriate curve of FIG. 14. This is accomplished by compressing the delta stabilized alloy of such composition to some maximum pressure intermediate between the initial transformation pressure and the pressure necessary for maximum transformation. Thus in FIG. 1, for example, if the delta stabilized alloy containing 4.5 a/o Al is compressed to some value such as 8000 atmospheres the permanent volume change will be somewhere in the neighborhood of 4%, indicating that only a fraction of the maximum possible transformation from delta to alpha
plutonium has been achieved. Such transformations are somewhat more difficult to accomplish than the maximum possible transformation at the peaks of the curves. They are easiest in the case of an alloy such as the 4.0 a/o iridium alloy indicated in FIG. 5 or the 8.0 a/o erbium alloy of FIG. 7 because of the greater slope of transformation as compared to the almost horizontal form of the transformation curves of aluminum, zinc or hafnium in FIGS. 1, 3 and 6.

It is also to be noted that the Ce–Pu alloys can be transformed into alloys in which the plutonium is predominantly of the beta phase, and thus obtain the intermediate physical characteristics of this phase (density = 17.6 g/cc., average linear coefficient of thermal expansion = $34.7 \times 10^{-6}/^\circ$ C.)

For instance, a 4.0 a/o Ce alloy compressed to 3600 atmospheres contained 77% beta phase, 14% delta phase, 9% gamma phase and no alpha.

What is claimed is:

1. A method for making stabilized plutonium alloys having a mixture of allotropes consisting essentially of alpha and delta and thereby obtaining in such alloys hardness, positive coefficient of expansion, and density which are intermediate those of undiluted delta phase and undiluted alpha phase plutonium, characterized by compressing said alloy to at least a pressure which produces rapid increase in density with a negligible increase in pressure, said alloys comprising binary alloys of plutonium containing about 4.5 a/o of an element selected from the class consisting of indium, erbium, and hafnium.

2. The method of claim 1 characterized by compressing said alloy to a range of pressures higher than all said pressures marked by said rapid increases of density and wherein the resulting volume decrease of said alloy with increasing pressure is negligible.

3. A method of treating a plutonium alloy containing delta phase plutonium stabilized at room temperature by the addition of about 4.5 a/o indium and rapid cooling from the delta field, characterized by compressing said alloy to at least 830 atmospheres.

4. A method of treating a plutonium alloy containing delta phase plutonium stabilized at room temperature by the addition of about 4.5 a/o erbium and rapid cooling from the delta field, characterized by compressing said alloy to at least 1700 atmospheres.

5. A method of treating a plutonium alloy containing delta phase plutonium stabilized at room temperature by the addition of about 4.5 a/o hafnium and rapid cooling from the delta field, characterized by compressing said alloy to at least 2350 atmospheres.

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<table>
<thead>
<tr>
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<th>Inventor(s)</th>
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<tbody>
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<td>MacLeod</td>
<td>Mar. 1, 1955</td>
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<td>2,789,072</td>
<td>White</td>
<td>Apr. 16, 1957</td>
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<td>2,827,404</td>
<td>Klein</td>
<td>Mar. 18, 1958</td>
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<tr>
<td>2,897,077</td>
<td>Coffinberry</td>
<td>July 28, 1959</td>
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<td>2,898,252</td>
<td>Zegler</td>
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<td>Schönfeld</td>
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<tr>
<td>2,929,706</td>
<td>Cramer et al.</td>
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