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OF THE UNIVERSITY OF CALIFORNIA ○ LOS ALAMOS NEW MEXICO

BEHAVIOR OF SOME DELTA-STABILIZED PLUTONIUM-GALLIUM ALLOYS AT HIGH PRESSURES

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BEHAVIOR OF SOME DELTA-STABILIZED PLUTONIUM-GALLIUM ALLOYS AT HIGH PRESSURES

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by

R. O. Elliott
K. A. Gschneidner, Jr.

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ABSTRACT

A number of delta-stabilized plutonium-gallium alloys were subjected to pressures as high as 11,000 atm at 24°C. Transformation pressures and volumes, compressibilities, densities, and hardnesses of these alloys were determined as a function of composition, and the effect of gallium content on the stability of delta plutonium was studied and related to the plutonium-gallium phase diagram.

ACKNOWLEDGMENTS

The authors are grateful to Janet Clark for making the metallographic examinations; to F. E. Colvin and C. E. Westley for helping with some of the experiments and calculations; and to V. O. Struebing and P. McCrossen for preparing the alloys. In addition, they wish to thank the other members of LASL Group CMF-5 for various services; Group CMB-1 for the chemical analysis; and Group CMB-11 for machining the specimens. Lastly, the authors express their appreciation to F. W. Schonfeld and D. S. Hughes for their interest and helpful suggestions.

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FIGURES

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INTRODUCTION

The first studies of the effect of pressure on pure plutonium and plutonium alloys were made by Bridgman in 1945.[1] His data on pure plutonium have been recently declassified and published.[2] In 1956 Hughes[3] made some studies on two delta-stabilized plutonium-gallium alloys. Hughes' results showed that an alloy containing 3.4 a/o gallium underwent an almost completely reversible transformation between 8500 and 9000 atm and that an alloy containing 1.7 a/o gallium underwent an irreversible transformation at approximately 4700 atm.

It was hoped that a more detailed investigation of the effect of pressure on the delta-stabilized alloys might provide an explanation for the anomalous expansion behavior of the high-temperature delta phase in pure plutonium. Although such an explanation was not obtained, some interesting correlations between the high-pressure studies and the binary phase diagrams were observed.

To supplement Hughes' data, several more plutonium-gallium alloys and several other binary plutonium alloys were investigated. The results of the current investigation on plutonium-rich alloys containing copper, zinc, cadmium, aluminum, indium, germanium, tin, and cerium have been reported elsewhere.[4] The results of the investigation of the plutonium-gallium alloys are reported below.

Delta-phase plutonium can be easily retained to room temperature by a proper combination of alloying and heat treatment. Alloys of this type are generally referred to as delta-stabilized alloys, although the delta phase in many such alloys is actually metastable and will transform to a more stable phase or combination of phases when the alloy is thermally treated or compressed. In the limited range of our apparatus (maximum pressure 11,000 atm), some of the alloys that were studied were found to transform under compression although others did not. The transformation pressures and volumes, compressibilities, densities, and hardnesses of the alloys have been determined and plotted as a function of alloy composition.

APPARATUS AND TECHNIQUES

Alloy ingots were prepared by vacuum-melting and vacuum-casting weighed amounts of the component metals in an induction furnace. The cast ingots were about 1/2 in. in diameter and 1-3/4 in. long. These ingots were homogenized at 450°C for at least 200 hours, air-quenched to room temperature, and then machined to form right cylinders that were 0.434 in. in diameter and from 1.5 to 1.7 in. long. All alloy compositions given in this report are nominal.

All the alloys were subjected to metallographic and X-ray examinations before and after compression. Density measurements were also made before and after compression.* These tests were necessary because several alloys were found to be cored or otherwise inhomogeneous as a

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*Precision of density measurements is 0.03 gm/cm^3.

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result of being held for insufficient time at the heat-treatment temperature. Such alloys were given further heat treatment for an additional 200 to 400 hours.

The alloys were compressed in the simple piston and cylinder device shown in Fig. 1. The stationary piston was placed in one end of the high-pressure cylinder, and the specimen, glycerine, and movable piston were inserted at the opposite end in that order. The assembly was then fitted into the guide and protecting cylinder, and dial indicators were attached to the movable piston holder and guide.

The assembled apparatus is shown in Fig. 2. Force was applied to the movable piston by means of a 50-ton press. A Baldwin SR-4 load cell was used to measure the load, and the equipment was calibrated for pressure measurements at the solidification points of mercury, 8850 atm and 7.5°C.* The dial indicators located on opposite sides of the high-pressure cylinder made it possible to measure the linear displacement of the movable piston. Volume changes in the specimen, however, could not be obtained directly from the dial-indicator readings because the glycerine compressed, the cylinder expanded, and the pistons shortened under pressure. The data were corrected for the compression of the glycerine using values published by Bridgman,[5] and for the dimensional changes of the cylinder and pistons using calculations based on elastic theory. The volume measurements determined with this apparatus are estimated to

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*Accuracy of pressure measurements is estimated to be 200 atm.

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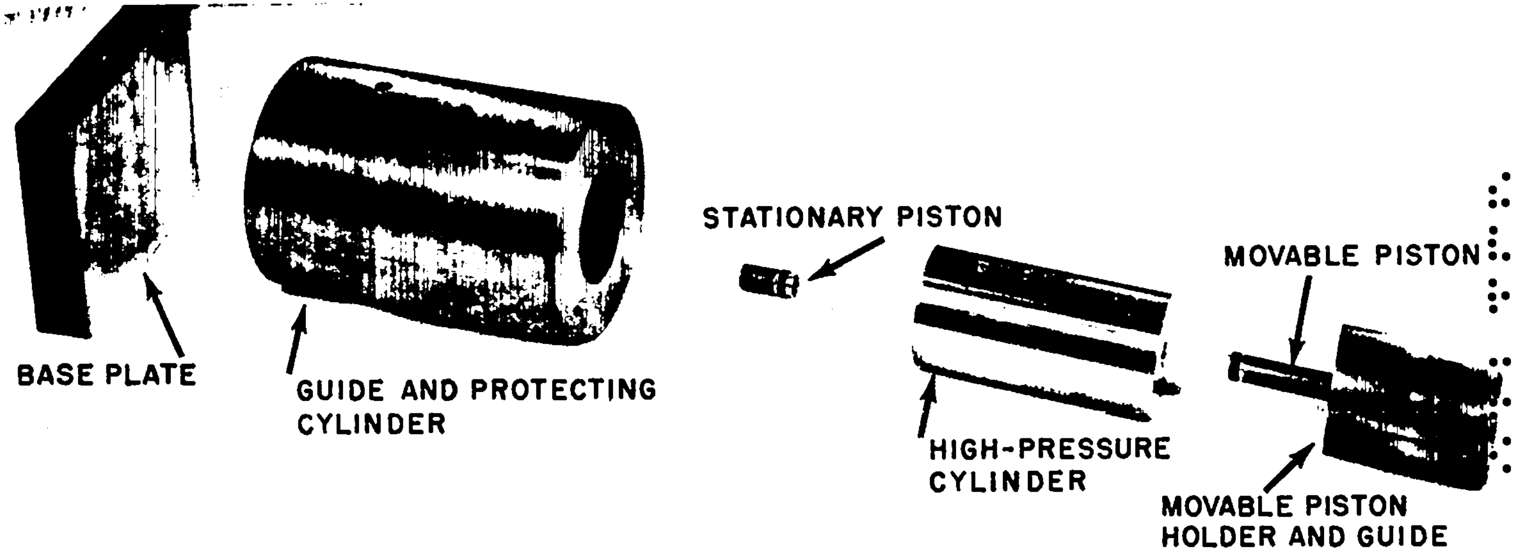

Fig. 1. Exploded View of 10-Kilobar High-Pressure Chamber

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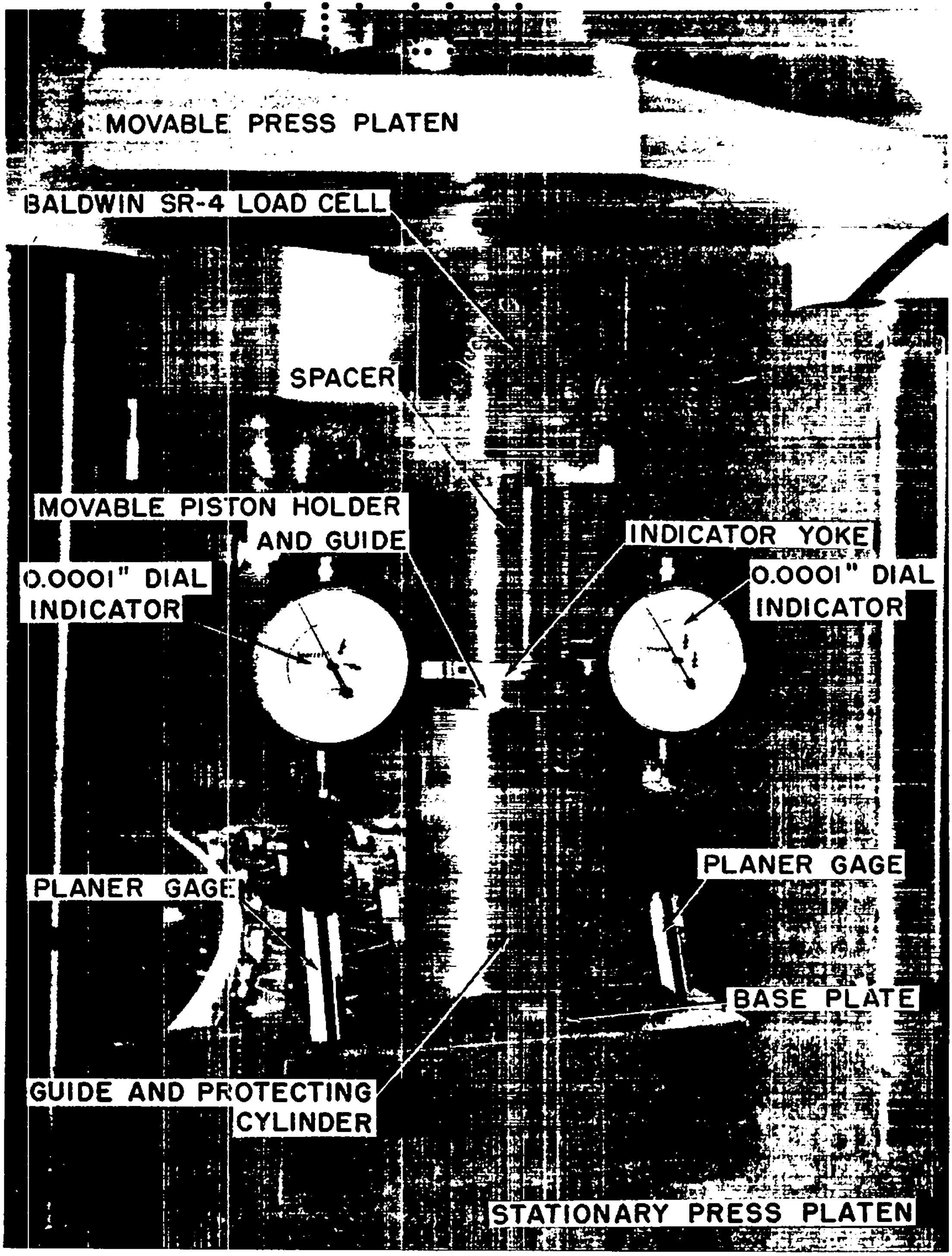

Fig. 2. Assembled View of 10-Kilobar High-Pressure Chamber

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be accurate to 10 per cent of the value reported. This estimate of the accuracy is based on the results obtained with a pure aluminum standard. All measurements were taken at 24°C.

RESULTS

The results of these compression studies are summarized in Table I. The compression curves of six plutonium-gallium alloys are shown in Fig. 3. It may be noted that the alloy containing 1.7 a/o gallium underwent an irreversible transformation (similar to that reported by Hughes[3]), and that the transformation in alloys having higher gallium content tended to become more reversible with increasing gallium content; i.e., the extent of reversibility increased. These irreversible transformations have been found to result from the change of metastable delta plutonium into alpha, or alpha plus a small amount of beta, with some untransformed delta usually remaining after compression. The amount of the delta phase remaining after compression increases with increasing gallium content. These data are in agreement with the results reported earlier by Hughes.[3] Figure 4 shows the variation of the transformation pressure and volume of transformation as a function of composition. The intersection of the transformation-pressure curve with the composition axis at about 0.6 a/o gallium indicates that this composition represents the minimum amount of gallium that is required to retain delta phase to room temperature under the heat-treatment conditions that were used. This interpretation has been verified for the plutonium-aluminum alloys.[4]

TABLE I. Summary of High-Pressure Data for Delta-Stabilized Plutonium-Gallium Alloys

Composition (a/o Ga)	Max. Press. (atm)	Trans. Press. (atm)	Vol. of Trans. (%)	Permanent Vol. Change[a] (%)	Compressibility x 10^6 (atm^{-1})	Density after Heat Treatment (gm/cm^3) Before Compress.	Density after Heat Treatment (gm/cm^3) After Compress.
1.7	9,100	2,810	16.7	16.0	3.6	15.81	18.81
2.5	10,000	6,360	12.0	8.7	3.6	15.78	17.57
3.5	10,600	8,140	10.3	3.9	3.3	15.72	16.43
3.75	11,000	8,700	8.8	3.1	3.8	15.75	16.10
4.0	10,060	b	-	-	4.1	15.73	15.76
5.0	8,920	b	-	-	3.7	15.70	15.84
10.0	9,100	b	-	-	3.7	15.24	15.55

[a] Values listed in this column were obtained by extrapolating the pressure-volume curves to 1 atm.

[b] No transformation was observed in these alloys.

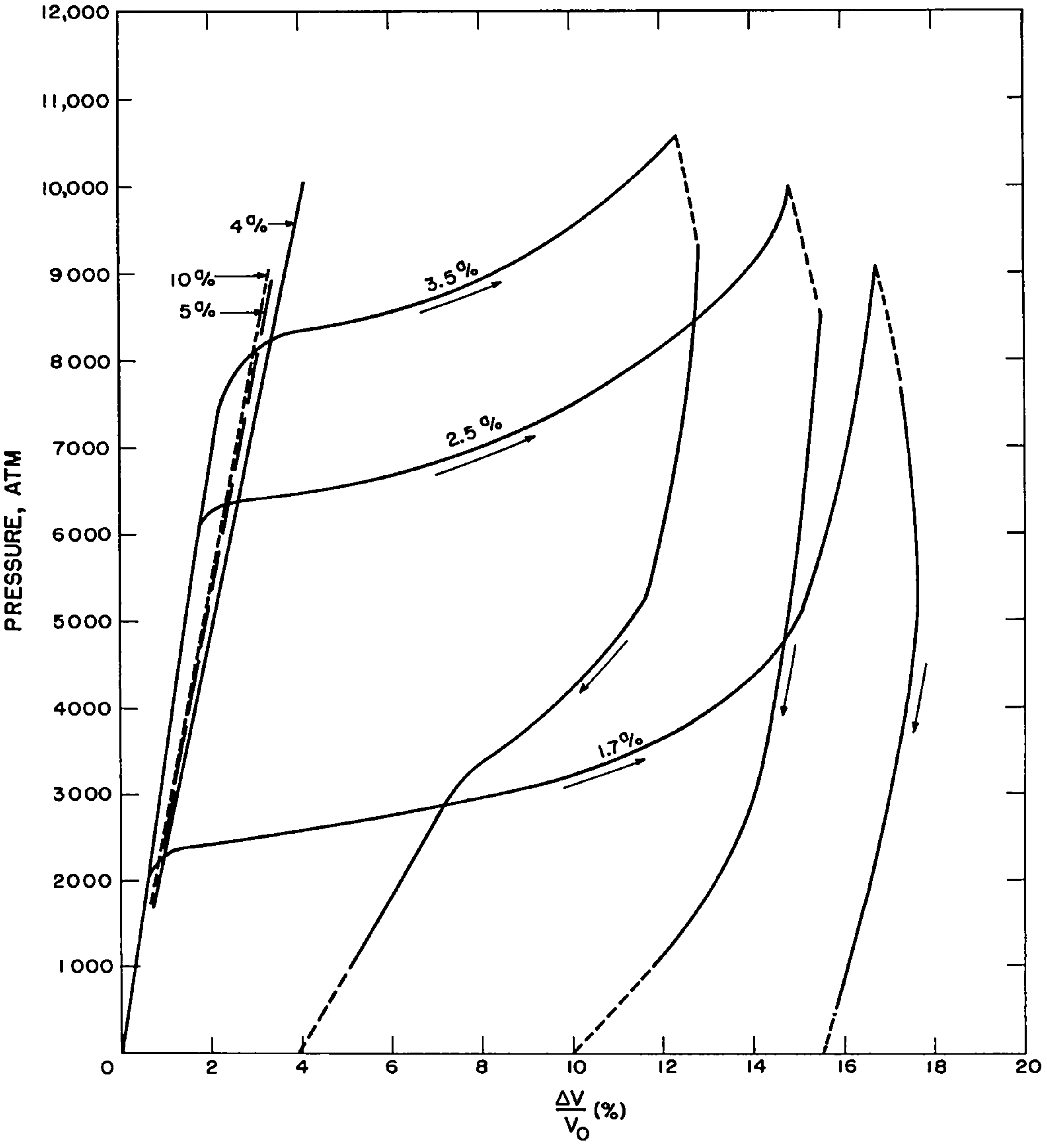

Fig. 3. Compression Curves of Six Plutonium-Gallium Alloys

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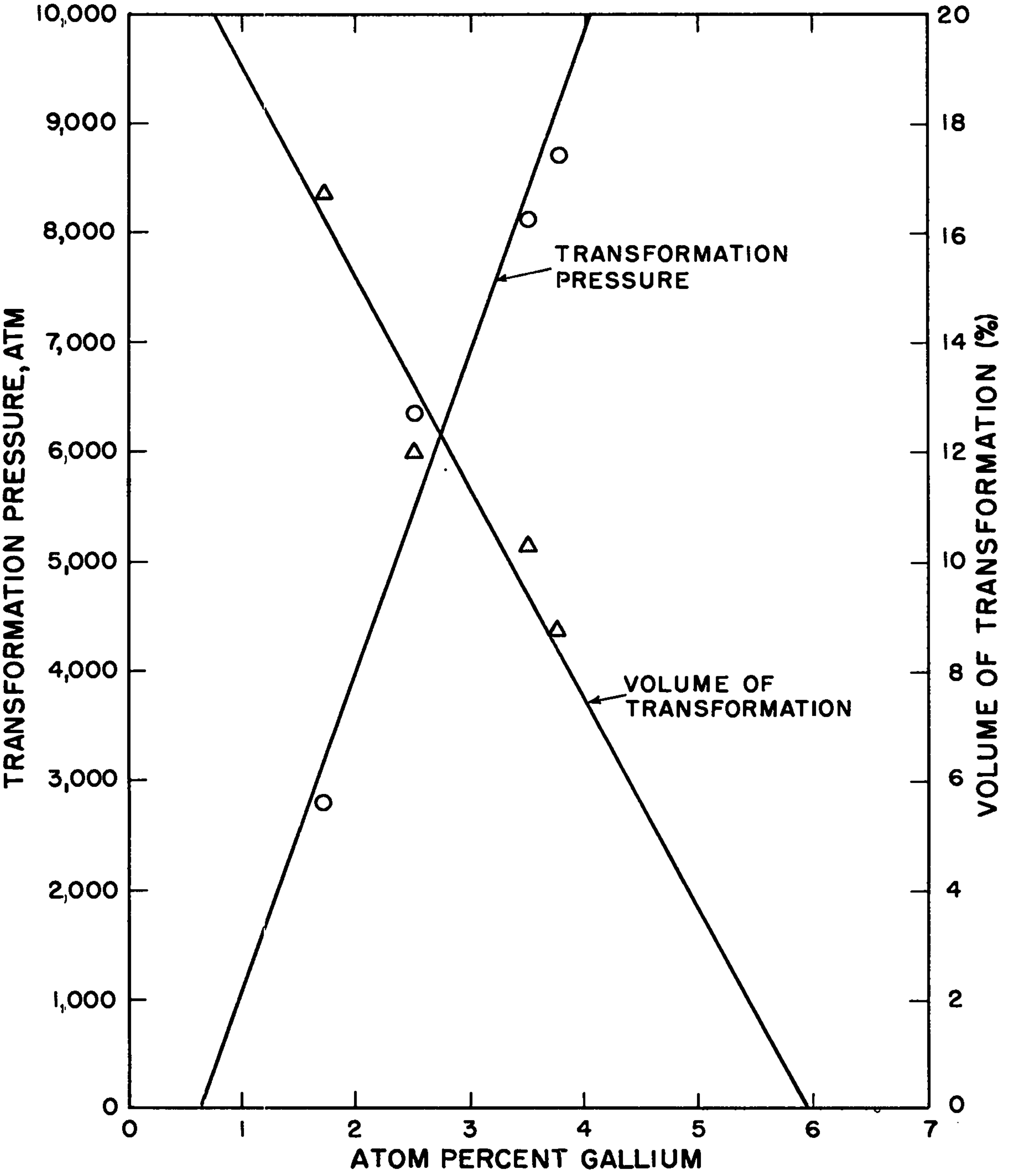

Fig. 4. Variation of Transformation Pressure and Volume of Transformation as a Function of Composition

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Figure 5 shows the densities of the alloys plotted against their gallium content. The intersection of the after-compression curves at 4.0 a/o gallium is interpreted to mean that all alloys containing less than 4.0 a/o gallium are metastable and should be expected to undergo some transformation reversibility, and that alloys containing more than 4.0 a/o gallium should be thermodynamically stable at room temperature and should undergo a completely reversible delta-to-alpha transformation. Although transformations were not observed for alloys containing more than 3.75 a/o gallium (probably because of the pressure limitation of the apparatus), the above interpretation is believed to be correct because the plutonium-cerium alloys that were studied showed complete reversibility at cerium compositions greater than that indicated at the intersection of the after-compression density curves.[4]

The compressibilities and hardnesses of the gallium alloys before and after compression are shown in Fig. 6. The amount of gallium appears to have little or no effect on the compressibility of the delta phase over the composition range for which measurements were taken. It should be noted that the compressibilities are believed to be accurate to 1.0×10^{-6} atm^{-1}. The after-compression hardness is seen to decrease rapidly with increasing gallium content until the composition of about 3.5 a/o gallium is attained. For alloys containing more than this amount of gallium only slight hardness changes occurred. The 3.5 a/o value is seen to agree reasonably well with the value obtained from the

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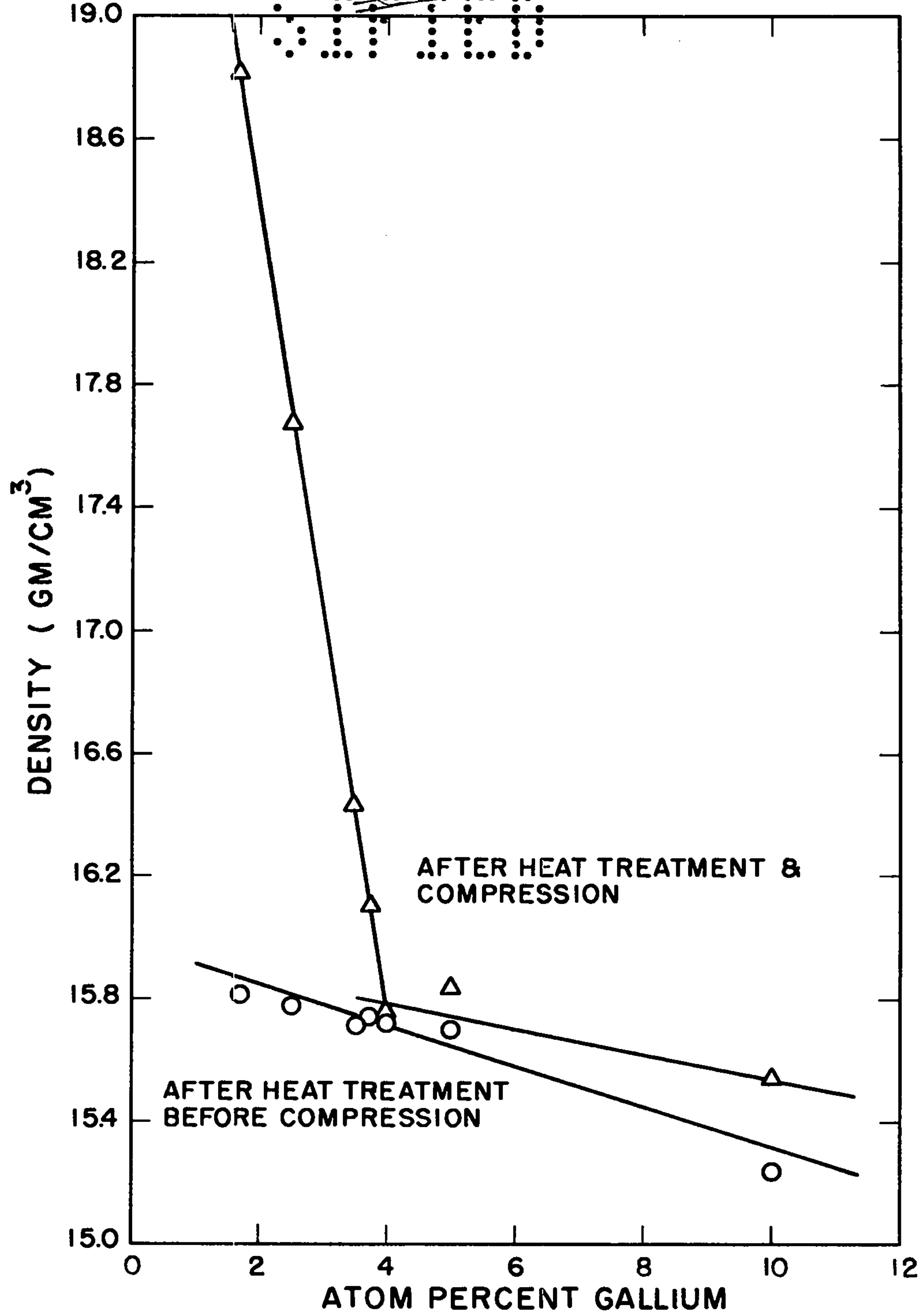

Fig. 5. Alloy Densities vs Gallium Content

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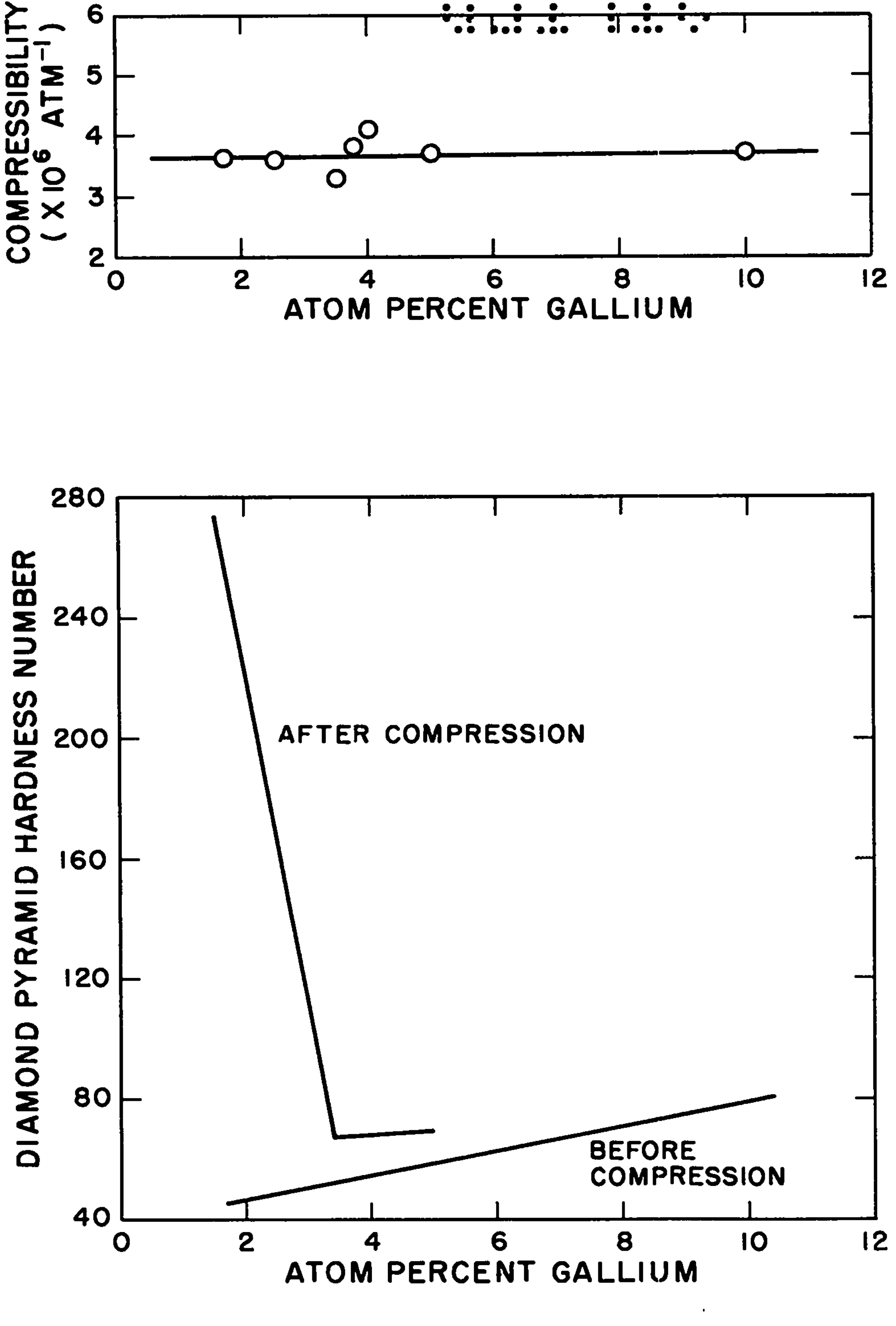

Fig. 6. Compressibilities and Hardnesses of Plutonium-Gallium Alloys

after-compression density curves (see Fig. 5). Without doubt, the value from the density curves is the more reliable.

The effects of cycling and of pressure on the densities of some of the alloys are shown in Fig. 7. For the 2.5 a/o gallium alloy the density is seen to increase linearly to a certain point as the number of cycles is increased. Beyond that point the density, for all practical purposes, remains constant. For the 3.5 a/o gallium alloy, the cycling experiment was not continued long enough to find the leveling-off point. Figure 7 also shows that the density increases with increased pressure. However, above some certain but undetermined pressure the increase in density would be expected to reach a maximum and then remain constant. The increase in density with increasing pressure indicates that the transformation becomes more irreversible as the pressure increases. It is expected that cycling and pressure would have no effect on alloys containing more than 4.0 a/o gallium. If this were not true, then the transformation in these alloys would not be truly reversible.

DISCUSSION

Some interesting correlations can be inferred from these studies. If the alloy composition indicated at the intersection of the density curves (4.0 a/o gallium) as shown in Fig. 5 can be assumed not to change with pressure, then this composition should indicate a point lying on the delta/delta-plus-alpha solvus line (at room temperature and pressure) for the plutonium-gallium phase diagram. This value (4.0 a/o gallium)

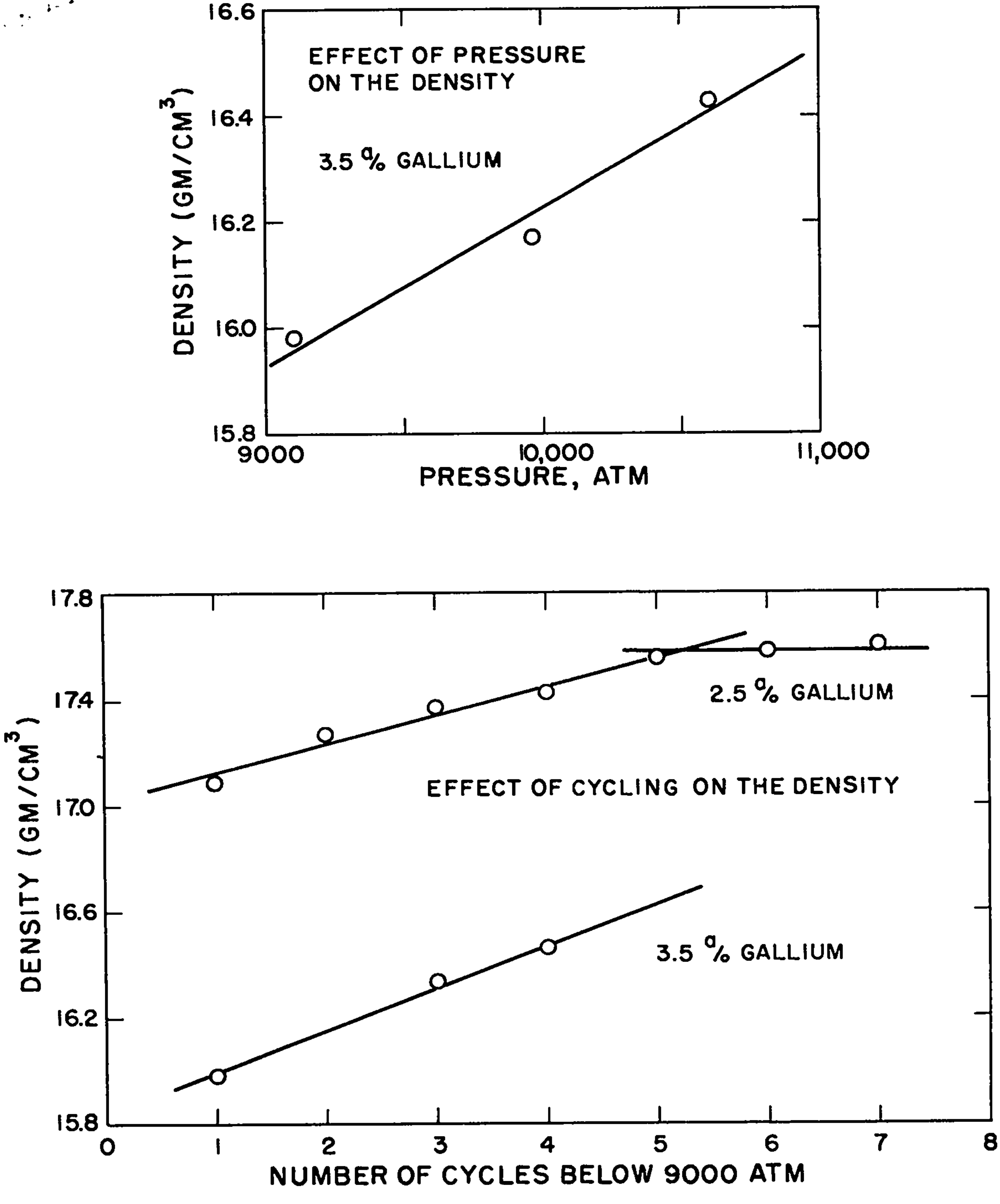

Fig. 7. Effects of Cycling and of Pressure on the Densities of Several Plutonium-Gallium Alloys

agrees reasonably well with the value of 5.0 ± 2.0 a/o gallium, which was obtained by extrapolating X-ray and density data from 200 to 250°C to room temperature in a study of the plutonium-gallium system. A tentative plutonium-gallium phase diagram is shown in Fig. 8 (Ref. 6).

One of the authors (K.A.G.) has calculated that, at elevated temperature, delta plutonium will transform to either the gamma or the delta-prime phase at 1300 atm; i.e., at pressures greater than 1300 atm no delta phase can be formed regardless of the temperature. As mentioned earlier, the delta phase in alloys containing more than 4.0 a/o gallium will transform reversibly to alpha plutonium. The pressures required to bring about this reversible transformation in alloys containing 4.0 a/o gallium must be at least 9900 atm, and for greater gallium content the pressure will be accordingly higher. Thus the addition of gallium to plutonium has the effect of extending the delta-field pressure-wise, which is similar to the effect produced by gallium in extending the delta-field temperature-wise (see Fig. 8).

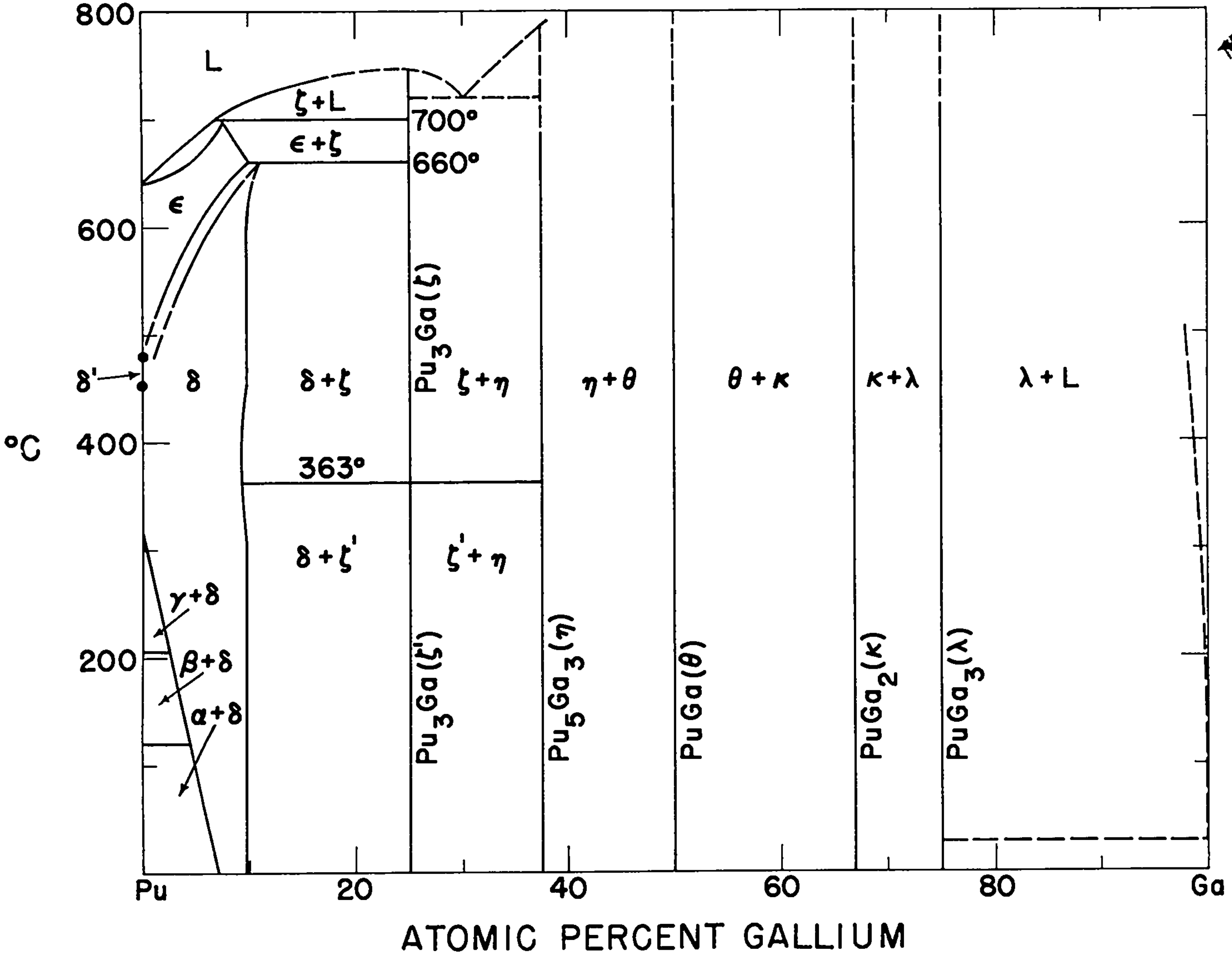

Fig. 8. Tentative Plutonium-Gallium Phase Diagram

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