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Martensite-Like Behavior in Actinide Metals and
Alloys Below Room Temperature**

by

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ANALYSIS OF EVIDENCE FOR AN IRREPRODUCIBLE MARTENSITE-LIKE BEHAVIOR IN
ACTINIDE METALS AND ALLOYS BELOW ROOM TEMPERATURE

by

Thomas A. Sandenaw

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ABSTRACT

Evidence is presented which suggests that a low-temperature, martensite-like behavior may be quite general in actinide metals and their alloys and compounds. There may be no metastable martensitic embryos in an α -phase structure of high-purity U, Np, and Pu formed by a diffusion-controlled $\beta \rightarrow \alpha$ transformation, and thus no evidence for low-temperature phases.

The effect of impurity content on observed low-temperature physical properties of these actinides is noted. It is proposed that impurities may be playing several roles. They may permit an electron redistribution in dilute alloys dependent upon the length of holding time. Experimentally determined values for the electronic contribution to heat capacity and the density of states of U, Np, and Pu should thus vary over a considerable range, as has been observed. Variations in interstitial ordering of impurity atoms with processing may yield stacking variants of each basic close-packed actinide metal structure and thus determine the number and structure of low-temperature phases.

I. INTRODUCTION

The proposal that a phase transformation in an actinide metal ($\delta \rightarrow \gamma$ phase Pu) could occur by a martensitic mechanism was made first by Pascard (1960).¹ Martensitic transformations in plutonium metal and other of its alloys have since been noted. They also have been reported to occur in uranium and neptunium metals.

One purpose of this paper is to present evidence that martensitic behavior may be quite general in actinide metals and their alloys and compounds. The great differences reported for the low-temperature physical property behavior of actinide metals and alloys can be explained if actinides sometimes undergo an unexpected martensitic transformation. Whether phase transformations in actinide materials are

martensitic or not may depend not only on cooling rate but also on prior thermal history, purity, and thermal cycling, as has been suggested by Goldberg and Massalski (1970).²

A second purpose is to point out the apparent effects of impurity content on the appearance or nonappearance of martensite-like transformations in uranium, neptunium, and plutonium metals on cooling below room temperature or on application of pressure. The actual impurity elements present and their quantity may play a critical role, depending upon how they affect the electrons per unit-cell ratio of the crystal structure and the subsequent electron-phonon interactions.

A third purpose is to review physical property evidence for both electron-phonon interactions and

Fermi surface - Brillouin zone boundary interactions in uranium and plutonium metals at low temperatures. A fourth purpose is to show the role that these interactions play in martensitic transformations and the development of antiphase-ordered structures.

It is suggested that the differing physical property behavior of actinide materials at low temperature may depend upon two factors: a) a redistribution of electrons between impurity atoms and actinide atoms in a dilute alloy held at low temperatures, b) a gain in f character in the valence band of the actinide atoms when the extremely dilute alloy is processed at low temperatures.

II. MARTENSITIC TRANSFORMATIONS IN URANIUM, NEPTUNIUM, AND PLUTONIUM METALS AND THEIR ALLOYS

A martensitic transformation induces defects. These defects can be stacking faults, dislocations, interstructural interfaces, and twin boundaries. Elastic strains can also result.

In their discussion of nonferrous martensites, Warlimont and Delaey (1974)³ note that there are three different types of fundamental martensitic structure. These are fcc, hcp, and close-packed orthorhombic with periodic stacking shifts. Although the structures of martensitic phases are all essentially close-packed, small orthorhombic and monoclinic distortions may result from complicated stacking sequences.

The martensitic nature of $\beta \rightarrow \alpha$ phase transformations in uranium, neptunium, and plutonium metals was discussed in surveys of phase transformations in actinide metals by Goldberg and Massalski (1970)² and Rehtien and Nelson (1973).⁴ The martensitic behavior of other phase transformations in uranium and plutonium metals and their alloys was also thoroughly reviewed by Goldberg and Massalski,² and Rehtien and Nelson⁴ also considered the transformation characteristics of uranium alloys.

The room-temperature alpha phase crystal structures of uranium and neptunium metals are orthorhombic, while that of plutonium is monoclinic. One can speculate that the room-temperature structures observed for these metals are caused by stacking variants resulting from martensitic $\beta \rightarrow \alpha$ phase transformations. The implication is that these elements are always capable of acting like martensites.

Heat evolution is a characteristic of the martensitic process. Because of this, Mogutnov and Shvartzman (1972)⁵ suggested that a calorimeter is an appropriate instrument for research on martensitic aging phenomena. The heat evolution shows up as dips in heat capacity versus temperature curves, as was shown by Mogutnov and Shvartzman for quenched iron-based alloys. The dips in heat capacity are sometimes extreme.

Abnormally large dips have been seen in the low-temperature heat capacity curves of low-purity, heavily radiation-damaged α phase plutonium by Sandenaw, Olsen, and Gibney (1960),⁶ and in similar curves for a δ -phase stabilized Pu-8at.% Al alloy by Sandenaw (1960).⁷ Dips have been shown by Taylor, Linford, and Dean (1963)⁸ in the low-temperature heat capacity curves of a δ -phase stabilized Pu-0.75 wt% Ga alloy. A drop-off in heat capacity curves of a Pu-10 at.% Fe eutectic alloy at low temperatures was reported by Sandenaw and Harbur (1973).⁹ Erratic low-temperature heat capacity curves, which were as extreme as any seen with iron-base martensites, were reported for $\text{PuC}_{0.80}$ by Sandenaw (1975).¹⁰ Thus a martensite-like behavior does not appear to be limited to plutonium metal and its alloys, but may also occur in the nonstoichiometric carbide.

III. OBSERVED EFFECTS OF IMPURITIES ON LOW-TEMPERATURE PHYSICAL PROPERTY BEHAVIOR OF URANIUM, NEPTUNIUM, AND PLUTONIUM METALS

A. Uranium

Although the α -phase uranium structure is classified as bc orthorhombic, it resembles that of a distorted hexagonal metal. A feature of the α -U structure is interstitial holes. Of the two types of interstitial holes, one has four nearest atoms and the other five nearest atoms. The locations and sizes of these holes have been tabulated by Opinsky (1954).¹¹ The question then arises whether impurity atoms can diffuse into these interstitial holes to give interstitially ordered solutions at room temperature with a surprisingly small concentration of solute.

In their discussion of ordering in unfamiliar systems, Richman and Davies (1973)¹² surveyed the ordering in uranium alloys. Uranium alloys show both chemical order and displacement order. The

alloys are γ -phase stabilized (bcc) by Nb, Mo, Ti, Re, or Ru. During quenching or subsequent low-temperature aging, the γ phase transforms to several metastable phases which include α' , α'' , ω , γ^0 , and γ^S . Richman and Davies note that the first two are orthorhombic and monoclinic distortions, respectively, of α -phase uranium and are formed martensitically together with tetragonal ω . γ^S is cubic and may be regarded as a displacement analog of short-range chemical order, while γ^0 (base centered tetragonal) is a long-range analog of chemical order.

Diffusion studies of Tudja and Ban (1973)¹³ revealed the existence of a new fcc phase at room temperature in the uranium-sulfur system having a composition ranging from $\sim\text{U}_{20}\text{S}$ to U_8S . The authors obtained the same fcc phase with ~ 9 at.% of Se, Te, P, As, Sb, Bi, C, and Si

The differences in physical property behavior of uranium noted at low temperatures by different experimenters may be due to the presence of different impurities. The elements listed by Tudja and Ban¹³ should tend to permit a low-temperature metastable fcc phase, while those elements stabilizing a γ -phase would tend to permit metastable α' , α'' , ω , γ^S , or γ^0 phases at low temperatures. The nature and extent of a martensitic transformation at ~ 43 K and the number of transformations noted below this temperature could depend upon the actual impurity elements present and the metastable phase that each element would tend to stabilize on cooling a specimen from the molten state.

The importance of impurities on the low-temperature physical properties of uranium was shown by Marples (1970).¹⁴ He studied the effect of alloying additions of Pu and Np on the lattice parameters of α -phase uranium between 4 and 300 K. The U-Np alloys became two-phase at the lowest temperature, while U-Pu alloys did not. The neptunium used for alloying contained considerably more impurities than either the uranium or plutonium, according to Marples.¹⁴ He suggested that a shear transformation could have occurred in some of the grains of the U-Np cast specimen, brought about by the strain produced by the anisotropic α - α' phase transition at 43 K.

Sandenaw (1975)¹⁵ found a reproducible heat capacity dip between ~ 36 K and 250 K in a ^{238}U specimen of nominal high purity. The impurity elements

present in this specimen were predominately Si and C, elements shown by Tudja and Ban¹³ to stabilize the fcc phase of uranium. Since Sandenaw¹⁵ did not see any evidence for low-temperature heat capacity peaks, the tendency must have only been for a martensitic transformation at ~ 36 K.

The heat capacity data of Flotow and Lohr (1960)¹⁶ for ^{238}U indicated that there was no physical property change below room temperature in high-purity metal. Experimental results thus seem to confirm that impurities are necessary for any low-temperature phase transformation in uranium.

B. Neptunium

Sandenaw (1965)¹⁷ and Lee, Mendelssohn and Sutcliffe (1970)¹⁸ have reported the results of low-temperature heat capacity measurements made on neptunium metal. Sandenaw's results¹⁷ were for impure metal. He observed a small peak at ~ 85 K, increased scatter of data from 75 K to 140 K, and a change of slope at 240 K. The heat capacity versus temperature curve of Lee et al.¹⁸ for purer neptunium was quite smooth. There was no clear-cut evidence from heat capacity measurements for a low-temperature martensitic transformation in neptunium.

C. Plutonium

The defect structure remaining after the β - α martensitic transformation in plutonium must be able to impart a physical property behavior reminiscent of a retained high-temperature (metastable) phase. This is suggested by the preliminary heat capacity behavior of a specimen of high-purity ^{242}Pu , as reported by Sandenaw and Gibney (1970).¹⁹

Four preliminary heat capacity runs between room temperature and 373 K were required before a smooth, reproducible C_p vs T curve was obtained with ^{242}Pu . If the evidence for a β - α transformation-induced metastable phase was so noticeable in extremely pure ^{242}Pu above room temperature, then retention of such a phase could be extremely easy in the case of impure plutonium metal.

Sandenaw et al. (1960)⁶ suggested that plutonium was alloy-like in its character and because of the hysteresis noted in measurable physical properties in almost every solid phase, that cooperative phenomena was responsible for a large part of the observed behavior.

There are eight crystallographically different atoms in the unit cell of α -phase plutonium. Pearson (1972)²⁰ classifies the α -phase structure of plutonium, as well as that of uranium and neptunium, as a metallic alloy. Fournier (1976)²¹ also notes that there are nonequivalent sites in the (α -phase) crystal structures of neptunium and plutonium and that nonequivalent sites correspond to different configurations. He points out that plutonium, in particular, should be regarded as an ordered alloy, and that this also explains the number of allotropic phases and their lack of stability.

The $\beta \rightarrow \alpha$ phase transformation of plutonium requires the ordering of crystallographically different atoms into the monoclinic structure of α phase. The rate of cooling through the $\beta \rightarrow \alpha$ transformation temperature could influence the formation of other, possibly distorted stacking variants of the monoclinic structure. This would explain the heat capacity results of Sandenaw and Gibney¹⁹ for ^{242}Pu . Several processing runs above room temperature may have been required to get the correct, stable α -phase structure.

The source of a martensite-like behavior in uranium and plutonium metals must be quite different at low temperatures. This is suggested by the different low-temperature electrical resistivity behavior of each element. No resistivity hump is seen with either pure or impure uranium. A maximum in the ρ vs T curve for plutonium metal is always seen at ~ 100 K. Only the resistivity value at the maximum appears to depend upon purity. The resistivity maximum suggests that there is always a long-period superlattice of defects present in the α -phase plutonium structure. This superperiod most likely results from a modulation of the basic close-packed structure by stacking fault boundaries remaining from the $\beta \rightarrow \alpha$ transformation. An examination of the resistivity, heat capacity, Hall coefficient, and other physical property results for plutonium metal at low temperatures has led Sandenaw and Harbur (1974)²² to conclude that an antiphase-ordered structure was responsible for observed behavior. Their analysis of results of physical property measurements suggested that similarities in the low-temperature properties of Pu metal and δ -phase stabilized alloy (fcc) could most likely be due to a type of long period superlattice.

It has been noted that martensitic transformations can lead to the development of a long-period antiphase structure as well as long-period stacking shift structures (Warlimont and Delaey (1974)).²³ An antiphase-ordered structure is thus completely compatible with martensitic transformations. Antiphase ordering could be responsible for the heat evolution noted in studies of quenched martensites.

The low-temperature heat capacity curve of pure ^{242}Pu did not show any evidence of phase transformations after preliminary processing, as shown by Sandenaw and Gibney.¹⁹ This suggests that interstitially ordered phases of impurities are also required for low-temperature phase transitions in plutonium as well as for uranium metal.

IV. SIMILARITIES BETWEEN PLUTONIUM AND CERIUM METALS

There are similarities in certain physical properties of plutonium and cerium, which have been attributed to promotion of an f electron. The two metals have bcc structures just below their respective melting temperatures and both exhibit the unusual behavior of contracting on melting. Jayaraman (1965)²⁴ has proposed that a $4f \rightarrow 5d$ electron promotion is responsible for the increase in density, i.e., the decrease in metallic radius of the cerium atom above the melting point. Wittenberg, Vaughn, and DeWitt (1970)²⁵ have suggested that the promotion of a $5f$ electron could account for the increased density of plutonium metal above its melting temperature.

Sandenaw, Olsen, and Gibney⁶ presented a low-temperature heat capacity curve for heavily self-damaged, impure plutonium metal. Two peaks were shown to occur below ~ 75 K, and a major peak noted at ~ 123 K. The specimen had been given considerable low-temperature processing. Attention is called to the similarity with the peak seen at ~ 165 K with cerium metal by Parkinson, Simon, and Spedding (1951).²⁶

Whether a new phase is observed in plutonium metal below room temperature may depend not only on purity but upon cycling and holding times at various temperatures, either below or above room temperature. Support for this hypothesis comes from the paper of Koskimaki, Gschneidner, and Panousis (1974).²⁷ They

showed that close to 100% β cerium could be prepared by repeated steps of thermal cycling of cerium samples between room temperature and liquid helium temperature followed by annealing at 75°C. They prepared single-phase α cerium by a pressure-cooling-depressurizing cycle.

The idealized temperature-pressure phase diagram for Ce metal which appears in the paper by Koskinaki et al.²⁶ shows the presence of β phase existing between ~150 K and ~350 K and from zero pressure to ~4 kbar pressure. One can speculate that the peaking in the heat capacity curve of Ce at ~165 K seen by Parkinson et al.²⁶ was evidence for the formation of some β -phase Ce. Similarities noted in the low-temperature heat capacity curve for impure α -Pu, as reported by Sandenaw et al.,⁶ could be evidence for at least one new low-temperature phase in plutonium metal below 200 K. The great differences in results reported for plutonium metal and its alloys by different experimenters could result from greatly differing cooling and cycling procedures particularly in the temperature region below 373 K.

V. POSSIBLE MECHANISMS FOR LOW-TEMPERATURE MARTENSITIC TRANSFORMATIONS IN ACTINIDE METALS AND ALLOYS

Weger and Goldberg (1973)²⁸ have suggested that two alternative channels in which electron-phonon interactions can manifest themselves are in superconductivity and martensitic transformations. There is considerable supporting evidence for the possibility of electron-phonon interactions in uranium and plutonium metals at low temperatures, although superconductivity has only been found in the case of uranium.

The stabilization of antiphase-ordered structures is considered to arise from the interaction of the Fermi surface and the Brillouin zone boundaries to lower the energy of conduction electrons. Toth and Sato (1968)²⁹ note that lowering of the energy of conduction electrons, due to creation of the extra period, was also indicated for the origin of stacking variants of close-packed structures, which appear in nonferrous martensites. It has been noted above that a forming antiphase structure or modulated stacking-order structure could account for some of the low-temperature physical property behavior of impure plutonium metal.

The gain in f character of uranium and plutonium metals under compression as well as on cooling toward 0 K, may play a role in the appearance of martensitic phases. This role may be through the effect of a gain in f character on electron-phonon interactions or Fermi surface-Brillouin zone boundary interactions.

An electron redistribution between impurity atoms and actinide element atoms could effectively change the electrons per unit cell ratio of the existing structure. The redistribution could require long-time holding at some low temperature. Completeness of the redistribution of electrons could be responsible for the completeness of any low-temperature phase transformation.

Electron-phonon interactions, Fermi surface-Brillouin zone boundary interactions, a gain in f character, or an electron redistribution appear to be closely interrelated in actinide materials. Each subject will now be discussed in greater detail.

A. Electron-Phonon Interactions

A disturbance in phonon distribution will result from any change in the behavior of conduction electrons. The phonon current dragged along and associated with the flow of conduction electrons can cause an extra contribution to thermoelectric power. A hump was seen below ~90 K in thermoelectric power versus temperature curve for α -Pu by Meaden and Lee (1962)³⁰ and Lallement (1963).³¹ A similar hump was noted by Sze and Meaden (1970)³² for α -U below ~50 K. A large hump in the low-temperature thermoelectric power for neptunium metal was also shown by Meaden and Lee.³⁰

The thermoelectric power behavior of $V_3\text{Ge}$ (a β -tungsten (A-15) structure material), reported by Sarachik, Smith, and Wernick (1963),³³ is very similar to that shown for α -Pu by Lallement.³¹ The maximum value for the thermoelectric power of the two materials appears at about the same temperature, i.e., about 60 K. The A-15 structure material, $V_3\text{Ge}$, supposedly does not undergo a martensitic transformation just above its superconducting transition temperature. The case for a martensitic transformation in α -Pu at ~60 K is thus questionable, based on thermoelectric power results.

The longitudinal attenuation behavior of ultrasonic waves has been shown to be essentially the

same at ~60 K for plutonium metal (Rosen, Erez, and Shtrikman, 1968),³⁴ δ -phase stabilized Pu alloy (Rosen, Erez, and Shtrikman, 1969),³⁵ V_3Ge (Rosen, Klimker, and Weger, 1969)³⁶ and at ~36 K in the case of uranium (Rosen, 1968).³⁷ The dips in heat capacity curves of plutonium metal and of δ -phase stabilized Pu alloy above ~60 K, as mentioned in an earlier section, seem to confirm the possibility of martensitic transformation in these materials.

Elastic constant results for uranium metal suggest a martensitic transformation at ~40 K. Weger and Goldberg (1973)³⁸ note that the martensitic transformation is brought about when the electronic contribution to the elastic constants just cancels the lattice contribution. They also note that in the case of superconductivity there is no such "accidental" balance between the two independent terms. The stiffness modulus, c_{11} , of single-crystal³⁹ " α -U" undergoes a discontinuity at ~40 K and a change of slope, as does the shear modulus, c_{44} (Fisher and McSkimin, 1961).⁴⁰ The same type of change in slope in the elastic constant, c_{11} , was shown at the martensitic transformation temperature for Nb_3Sn (a B-tungsten structure) by Rehwal (1968).⁴¹ A change of slope in the elastic constant, c_{44} , for V_3Si (a B-tungsten structure) was also seen at its martensitic transformation temperature by Testardi and Bateman (1967).⁴² The low-temperature elastic constant behavior of uranium metal thus seems to indicate the possibility of a martensitic transformation at ~40 K.

B. Fermi Surface-Brillouin Zone Boundary Interactions

Sandenaw and Harbur (1974)²² have suggested that FS-BZ boundary interactions could best explain the anomalous low-temperature behavior of plutonium metal and certain of its alloys. Changes in Brillouin zones are found to be associated with ordered interstitial sites in non-stoichiometric compounds, shear structures in oxides, many martensites, and long-period superlattices.

It seems plausible that changes in Brillouin zones could be associated with interstitial-ordered structures of impurity atoms in actinide metals and their extremely dilute alloys. This could play a role in low-temperature martensitic transformations. Variations in the interstitial ordering of impurity atoms could be responsible for differences in the

packing arrangement of close-packed planes. Modifications in cycling or processing of an actinide material specimen could yield different stacking variants of the basic close-packed structure, with a strong dependence of the variant on the particular impurity atoms present.

C. Gain in f Character

Calculations of Kmetko (1971)⁴³ indicated that U and Pu gained f character under compression. His calculations suggested that the relative change was small, and that the d character lost correspondingly. Jullien and Coqblin (1973)⁴⁴ studied the d-f hybridization of actinide metals and alloys. They noted that the localized model with a large d-f hybridization could account for the absence of localized magnetism in the strongly d-f hybridized uranium, neptunium, and plutonium metals. Later calculations of Hill and Kmetko (1975)⁴⁵ showed a surprising amount of s, d, and f hybridization occurring in the 6 p core band of the early actinides, with the f-like charge in this band completely delocalized.

It was proposed by Ward and Hill (1975)⁴⁶ that there is a deep involvement of the f electrons in the metallic bond of the early actinides. They noted that the hybridized bonding states did not "admit" to a definable valence, and that the net effect of these states on chemical, physical, and thermodynamic properties was far-reaching.

A disturbance in phonon distribution could result from a change in the behavior of conduction electrons due to a gain in f character. The thermoelectric power humps reported for both uranium and plutonium metals could be evidence for transfer of electrons to the 5f band at the noted temperatures on cooling. Such a transfer of electrons has been suggested by Marples¹⁴ as an explanation for the 43 K expansion anomaly in uranium. An expansion anomaly in plutonium metal below 40 K was also shown by Lallemand³¹ and could also be due to a transfer of electrons.

The differing physical property behavior found with actinide materials at low temperatures by experimentalists may be partially due to the extent of gain of f character in actinide atoms on processing.

D. Electron Redistribution

Slight variations in effective valence due to purity could account for the variability of physical property results for low-purity actinide metals. The effect of impurities may be through an electron redistribution between the impurity atoms and actinide element atoms. Such a redistribution may play the critical role in electron-phonon and FS-BZ boundary interactions and in gain in f character. Cycling procedures and holding time at a particular low temperature could significantly affect the completeness of an electron redistribution and the subsequent physical property behavior.

The low-temperature heat capacity results of Sandenaw and Gibney¹⁹ for high-purity ^{242}Pu suggested that the stable room temperature structure (monoclinic) remained unchanged on cooling to ~ 4 K. There was no indication of a phase transformation below room temperature, even after low-temperature cycling. There was thus no indication of a change in f character.

An electron redistribution in the presence of impurities seems to be a plausible explanation for the low-temperature physical property behavior of the actinide elements, U, Np, and Pu, upon low-temperature processing. A gain in f character should be a property of the actinide atoms themselves and be independent of any impurity atoms present in the metal. A possibility is that an electron redistribution favors a gain in f character after the actinide atoms in a structure have become approximately equivalent.

The values of γ , the electronic contribution to the heat capacity of U, Np, and Pu, could then be influenced greatly by the impurity elements present and the extent of electron redistribution due to processing. Determined values of the density of states of these actinide elements should vary considerably, as has been the case.

The variability in the thermoelectric power curves of plutonium, as shown in his Fig. 4 by Lallemand³¹ for the temperature range between 70 K and 150 K, could be demonstrating the dependence of electron-phonon interactions upon completeness of the electron redistribution between impurity atoms and plutonium atoms with processing. In the case of extremely pure material there may be no

redistribution and no apparent change in the electron per unit cell ratio.

The end result of low-temperature cycling of low-purity actinide metals may be as much to permit electron redistribution as to permit interstitial ordering of impurity atoms to crystal structures capable of undergoing phase (martensitic) transformations. The effect of interstitial ordering of impurities may be to modify the arrangement of close-packed planes. Although there may be identical atomic structures in the planes, the different impurities may yield different structures in the direction perpendicular to the stacking of planes.

There is an alternative explanation for the absence of low-temperature martensitic phase transformations in high-purity or even nominal-purity actinide metals. This answer is also suggested by the ^{242}Pu study of Sandenaw and Gibney.¹⁹ It is possible that the diffusion-controlled α -phase structure of ^{242}Pu was obtained on repeated heating from room temperature to ~ 373 K. One can theorize that there may be no metastable martensitic "embryos" in a completely diffusion-controlled α -phase structure. Such embryos may be required for phase transformations below room temperature in an actinide metal of any purity. Impurities may act as nucleation sites.

There is a plausible explanation why a non-stoichiometric compound like $\text{PuC}_{0.80}$ should show evidence for development of both an antiphase (martensitic) and an antiferromagnetic structure, i.e., multiple ordering, on holding for several days at ~ 75 K.¹⁰ Magnetic ordering (antiferromagnetism) often results in a compound when $5f$ electrons are essentially localized and the actinide atoms have the correct spacing. Fournier²¹ notes that up to plutonium, the hybridized $5f$ electrons are band electrons but that when actinide atoms are more separated in compounds, they can readily be localized.

There could be two consequences of holding $\text{PuC}_{0.80}$ at ~ 75 K. The longer the holding time, the greater could be the gain in f character due to an electron redistribution. The buildup of self-damage (swelling) with time could increase the interatomic distance between Pu atoms in the solid to the correct spacing for magnetism. Another

effect of radiation damage could be to form a distorted microstructure of point defects (vacancies and interstitials) which would easily permit a martensitic transformation in order to minimize the energy. The defects could be acting as nucleation sites (embryos) for a low-temperature martensitic transition.

VI. SUMMARY

Evidence is presented which suggests that a martensite-like behavior may be quite general in actinide metals and their alloys and compounds at low temperatures. Heat effects (heat evolution) are directly connected with the martensitic process. Heat capacity curves, which reveal temperature regions of heat evolution, are used as evidence for martensitic transformations in actinide materials.

Attention is called to certain similarities in the reported physical property behavior of cerium and plutonium. The published information for cerium metal shows that repeated steps of low-temperature cycling, with or without pressurizing, determine whether α -phase or β -phase cerium will be stabilized at room temperature. This behavior of cerium suggests that the anomalous low-temperature behavior of plutonium may be due to the irreproducible appearance of new phases, which depend not only on purity, but upon cycling and holding times at temperatures either below or slightly above room temperature.

Physical property evidence for electron-phonon interactions and Fermi surface-Brillouin zone boundary interactions in actinide metals is presented. Their relationship to martensitic transformations and antiphase-ordered structures is examined.

The effect of impurity content on observed low-temperature physical properties of the actinide metals, U, Np, Pu, is noted. If sufficiently pure, these actinide element metals do not appear to undergo any transformation below room temperature. It is proposed that impurities play several roles in determining the low-temperature physical property behavior of U, Np, and Pu. They may permit an electron redistribution in the dilute alloys at low temperatures, which results only from cycling or relatively long holding times at a particular temperature. Such a redistribution may play a

critical role in electron-phonon or FS-BZ boundary interactions and gain in f character. Variations in the interstitial ordering of impurity atoms with processing may yield different stacking variants of a basic close-packed actinide metal structure. The impurity element(s) present thus may determine the metastable structure(s) possible and the nature and number of low-temperature phase transformations.

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