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Constitution of Plutonium Alloys

by

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TABLE OF CONTENTS

Introduction		1	
Binary Systems		3	
<i>System</i>	<i>Page</i>	<i>System</i>	<i>Page</i>
Pu-Ag	3	Pu-Na	67
-Al	5	-Nb	67
-Am	9	-Nd	69
-As	11	-Ni	71
-Au	13	-Np	73
-B	15	-O	75
-Ba	17	-Os	77
-Be	19	-P	79
-Bi	21	-Pb	81
-C	23	-Pd	83
-Ca	25	-Pr	85
-Cd	25	-Pt	87
-Ce	27	-Rb	89
-Co	29	-Re	89
-Cr	31	-Rh	91
-Cs	33	-Ru	93
-Cu	35	-S	95
-Dy	37	-Sc	97
-Er	37	-Se	101
-Eu	37	-Si	103
-Fe	39	-Sm	105
-Ga	41	-Sn	107
-Gd	43	-Sr	109
-Ge	45	-Ta	109
-H	47	-Tb	111
-Hf	49	-Te	111
-Hg	51	-Th	113
-Ho	53	-Ti	115
-In	53	-Tl	117
-Ir	55	-Tm	119
-K	57	-U	121
-La	57	-V	123
-Li	59	-W	125
-Lu	59	-Y	127
-Mg	59	-Yb	129
-Mn	61	-Zn	129
-Mo	63	-Zr	131
-N	65		
Ternary and Higher Systems		133	
<i>System</i>	<i>Page</i>	<i>System</i>	<i>Page</i>
Pu-Al-Ga	133	Pu-Fe-U	153
-Ce-Co	135	-Fs(Fz)-U	155
-Ce-Co-Mn	137	-Mn-U	161
-Ce-Cu	139	-Mo-U	163
-Ce-Fe	141	-Nb-U	169
-Ce-Ga	143	-Th-U	173
-Ce-Mn	145	-Ti-U	179
-Ce-Ni	147	-U-Zr	183
-Cu-Sn	151		

LIST OF FIGURES

Fig. 1.	Plutonium-silver phase diagram. (Redrawn from Ref. 2.)	3
Fig. 2.	Plutonium-aluminum phase diagram.	6
Fig. 3.	Plutonium-ameridium phase diagram. (Redrawn from Ref.1.)	9
Fig. 4.	Plutonium-gold phase diagram.	14
Fig. 5.	Plutonium-beryllium phase diagram. (Courtesy Ellinger, Land, and Gschneidner and by permission of Gordon and Breach, Science Publishers, Inc., from <i>Plutonium Handbook</i> , Vol. 1, p. 198, 1967.)	20
Fig. 6.	Plutonium-bismuth phase diagram. (Courtesy Ellinger, Land, and Gschneidner and by permission of Gordon and Breach, Science Publishers, Inc., from <i>Plutonium Handbook</i> , Vol. 1, p. 199, 1967.)	21
Fig. 7.	Plutonium-carbon phase diagram.	23
Fig. 8.	Plutonium-cadmium phase diagram. (Redrawn from Ref. 1.)	25
Fig. 9.	Plutonium-cerium phase diagram.	27
Fig. 10.	Plutonium-cobalt phase diagram (Courtesy Ellinger, Land, and Gschneidner and by permission of Gordon and Breach, Science Publishers, Inc., from <i>Plutonium Handbook</i> , Vol. 1, p. 203, 1967.)	29
Fig. 11.	Plutonium-chromium phase diagram.	31
Fig. 12.	Plutonium-copper phase diagram.	35
Fig. 13.	Plutonium-iron phase diagram.	39
Fig. 14.	Plutonium-gallium phase diagram.	41
Fig. 15.	Plutonium-hydrogen phase diagram. (Courtesy Ellinger, Land, and Gschneidner and by permission of Gordon and Breach, Science Publishers, Inc., from <i>Plutonium Handbook</i> , Vol. 1, p. 207, 1967.)	47
Fig. 16.	Plutonium-hafnium phase diagram. (Redrawn from Ref. 1.)	49
Fig. 17.	Plutonium-mercury phase diagram.	51
Fig. 18.	Plutonium-indium phase diagram. (Redrawn from Ref. 1.)	53
Fig. 19.	Plutonium-lanthanum phase diagram.	58
Fig. 20.	Plutonium-magnesium phase diagram.	59
Fig. 21.	Plutonium-manganese phase diagram. (Courtesy Ellinger, Land, and Gschneidner and by permission of Gordon and Breach, Science Publishers, Inc., from <i>Plutonium Handbook</i> , Vol. 1, p. 213, 1967.)	61
Fig. 22.	Plutonium-molybdenum phase diagram. (Redrawn from Ref. 1.)	63
Fig. 23.	Plutonium-niobium phase diagram.	67
Fig. 24.	Plutonium-neodymium phase diagram.	69
Fig. 25.	Plutonium-nickel phase diagram. (Courtesy Ellinger, Land, and Gschneidner and by permission of Gordon and Breach, Science Publishers, Inc., from <i>Plutonium Handbook</i> , Vol. 1, p. 216, 1967.)	71
Fig. 26.	Plutonium-neptunium phase diagram. (Redrawn from Ref. 1.)	73
Fig. 27.	Plutonium-oxygen phase diagram.	76
Fig. 28.	Plutonium-osmium phase diagram. (Redrawn from Ref. 1.)	77
Fig. 29.	Plutonium-lead phase diagram. (Courtesy Ellinger, Land, and Gschneidner and by permission of Gordon and Breach, Science Publishers, Inc., from <i>Plutonium Handbook</i> , Vol. 1, p. 211, 1967.)	81
Fig. 30.	Plutonium-palladium phase diagram. (Redrawn from Ref. 1.)	83
Fig. 31.	Plutonium-praseodymium phase diagram.	85
Fig. 32.	Plutonium-platinum phase diagram. (Redrawn from Ref. 1.)	87
Fig. 33.	Plutonium-rhodium phase diagram. (Redrawn from Ref. 1.)	91
Fig. 34.	Plutonium-ruthenium phase diagram. (Courtesy Ellinger, Land, and Gschneidner and by permission of Gordon and Breach, Science Publishers, Inc., from <i>Plutonium Handbook</i> , Vol. 1, p. 219, 1967.)	93
Fig. 35.	Plutonium-scandium phase diagram. (Redrawn from Ref. 1.)	97

Fig. 36.	Plutonium-scandium phase diagram.	98
Fig. 37.	Plutonium-silicon phase diagram. (Redrawn from Ref. 1.)	103
Fig. 38.	Plutonium-samarium phase diagram.	105
Fig. 39.	Plutonium-tantalum phase diagram.	109
Fig. 40.	Plutonium-thorium phase diagram. (Courtesy Ellinger, Land, and Gschneidner and by permission of Gordon and Breach, Science Publishers, Inc., from <i>Plutonium Handbook</i> , Vol. 1. p. 221, 1967.)	113
Fig. 41.	Plutonium-titanium phase diagram.	115
Fig. 42.	Plutonium-uranium phase diagram.	121
Fig. 43.	Plutonium-vanadium phase diagram.	123
Fig. 44.	Plutonium-tungsten phase diagram.	125
Fig. 45.	Plutonium-yttrium phase diagram. (Redrawn from Ref. 1.)	127
Fig. 46.	Plutonium-zinc phase diagram.	129
Fig. 47.	Plutonium-zirconium phase diagram.	131
Fig. 48.	Plutonium-cerium-cobalt system liquidus surface. (Redrawn from Ref. 1.)	135
Fig. 49.	Plutonium-cerium-cobalt system 400°C isotherm. (Redrawn from Ref. 1.)	136
Fig. 50.	Plutonium-cerium-copper system liquidus surface. (Redrawn from Ref. 1.)	139
Fig. 51.	Plutonium-cerium-iron system liquidus surface. (Redrawn from Ref. 1.)	141
Fig. 52.	Plutonium-cerium-nickel system liquidus surface. (Redrawn from Ref. 1.)	147
Fig. 53.	Plutonium-cerium-nickel system 430°C isotherm. (Redrawn from Ref. 1.)	148
Fig. 54.	Plutonium-cerium-nickel system liquidus surface and 400°C isotherm. (Redrawn from Ref. 3.)	149
Fig. 55.	Effect of fission on the solidus temperature of uranium-20 wt%-plutonium alloys (Redrawn from Ref. 1.)	156
Fig. 56.	Solidus temperatures in the uranium corner of the plutonium-fission-uranium sys- tem. (Redrawn from Ref. 1.)	156
Fig. 57.	Isopleth showing equilibrium phases and transformations in the pseudo-ternary plutonium-fission-uranium system at 20 wt% plutonium. (Redrawn from Ref. 1.)	157
Fig. 58.	Plutonium-fission-uranium isothermal sections showing matrix phase equilibria and invariant reactions between 620 and 475°C. (Redrawn from Ref. 1.)	158
Fig. 59.	Plutonium-molybdenum-uranium partial liquidus projection and fields of primary crystallization. (Redrawn from Ref. 1.)	164
Fig. 60.	Plutonium-molybdenum-uranium partial solidus projection. (Redrawn from Ref. 1.)	164
Fig. 61.	Plutonium-molybdenum-uranium 1000°C partial isothermal section. (Redrawn from Ref. 1.)	165
Fig. 62.	Plutonium-molybdenum-uranium 900°C partial isothermal section. (Redrawn from Ref. 1.)	165
Fig. 63.	Plutonium-molybdenum-uranium 800°C partial isothermal section. (Redrawn from Ref. 1.)	166
Fig. 64.	Plutonium-molybdenum-uranium 700°C partial isothermal section. (Redrawn from Ref. 1.)	166
Fig. 65.	Plutonium-molybdenum-uranium 450°C partial isothermal section. (Redrawn from Ref. 1.)	167
Fig. 66.	Plutonium-molybdenum-uranium phases retained at room temperature by quench- ing from the gamma-phase region. (Redrawn from Ref. 1.)	167
Fig. 67.	Plutonium-niobium-uranium 900°C isothermal section (tentative).	170
Fig. 68.	Plutonium-niobium-uranium 630°C isothermal section showing peritectoidal forma- tion of ζ by the reaction $\beta U + \eta + \gamma U \rightarrow \zeta$ (tentative).	171
Fig. 69.	Plutonium-niobium-uranium 600°C isothermal section (tentative).	171
Fig. 70.	Plutonium-niobium-uranium 590°C isothermal section (tentative), showing the class II solid state reaction $\beta U + \gamma U \rightarrow \alpha U + \zeta$	172
Fig. 71.	Plutonium-niobium-uranium 350°C isothermal section (tentative).	172

Fig. 72.	Plutonium-thorium-uranium liquidus projection and fields of primary crystallization. (Redrawn from Ref. 1.)	174
Fig. 73.	Plutonium-thorium-uranium 1000°C isothermal section. (Redrawn from Ref. 1.)	174
Fig. 74.	Plutonium-thorium-uranium 900°C isothermal section. (Redrawn from Ref. 1.)	175
Fig. 75.	Plutonium-thorium-uranium 715°C isothermal section, showing peritectoidal formation of η (U-Pu) phase by the reaction $\alpha\text{Th} + \beta\text{U} + \gamma\text{U} \rightarrow \eta$. (Redrawn from Ref. 1.)	175
Fig. 76.	Plutonium-thorium-uranium 700°C isothermal section. (Redrawn from Ref. 1.)	176
Fig. 77.	Plutonium-thorium-uranium 610°C isothermal section, showing peritectoidal formation of ζ (U-Pu) phase by the reaction $\alpha\text{Th} + \beta\text{U} + \eta \rightarrow \zeta$. (Redrawn from Ref. 1.)	176
Fig. 78.	Plutonium-thorium-uranium 575°C isothermal section. (Redrawn from Ref. 1.)	177
Fig. 79.	Plutonium-thorium-uranium isopleth at 10 at% plutonium. (Redrawn from Ref. 1.)	178
Fig. 80.	Plutonium-titanium-uranium solidus projection in the uranium corner of the system.	180
Fig. 81.	Plutonium-titanium-uranium gamma solid solution decomposition temperatures.	181
Fig. 82.	Plutonium-titanium-uranium partial isopleth at 33.3 at% titanium.	181
Fig. 83.	Plutonium-uranium-zirconium solidus projection in the uranium corner of the system.	184
Fig. 84.	Plutonium-uranium-zirconium solidus and liquidus projections in the plutonium corner of the system. (Redrawn from Ref. 5.)	184
Fig. 85.	Plutonium-uranium-zirconium gamma solid solution decomposition temperatures.	185

LIST OF TABLES

I.	Crystal Structures and Transformation Temperatures of the Plutonium Allotropes	2
II.	Crystal Structure Data for Plutonium-Silver Alloys	4
III.	Crystal Structure Data for Plutonium-Aluminum Alloys	7
IV.	Crystal Structure Data for Plutonium-Arsenic Alloys	11
V.	Crystal Structure Data for Plutonium-Gold Alloys	14
VI.	Crystal Structure Data for Plutonium-Boron Alloys	15
VII.	Crystal Structure Data for Plutonium-Beryllium Alloys	20
VIII.	Crystal Structure Data for Plutonium-Bismuth Alloys	22
IX.	Crystal Structure Data for Plutonium-Carbon Alloys	24
X.	Solubility of Carbon in Liquid Plutonium	24
XI.	Crystal Structure Data for Plutonium-Cadmium Alloys	26
XII.	Crystal Structure Data for Plutonium-Cobalt Alloys	30
XIII.	Solubility of Chromium in Liquid Plutonium	32
XIV.	Crystal Structure Data for Plutonium-Copper Alloys	36
XV.	Crystal Structure Data for Plutonium-Iron Alloys	40
XVI.	Crystal Structure Data for Plutonium-Gallium Alloys	42
XVII.	Solubility of Plutonium in Liquid Gallium	42
XVIII.	Crystal Structure Data for Plutonium-Germanium Alloys	45
XIX.	Crystal Structure Data for Plutonium-Hydrogen Alloys	48
XX.	Crystal Structure Data for Plutonium-Hafnium Alloys	50
XXI.	Crystal Structure Data for Plutonium-Mercury Alloys	52
XXII.	Solubility of Plutonium in Mercury	52
XXIII.	Crystal Structure Data for Plutonium-Indium Alloys	54
XXIV.	Crystal Structure Data for Plutonium-Iridium Alloys	55
XXV.	Crystal Structure Data for Plutonium-Magnesium Alloys	60
XXVI.	Crystal Structure Data for Plutonium-Manganese Alloys	62
XXVII.	Solubility of Manganese in Liquid Plutonium	62
XXVIII.	Solubility of Molybdenum in Liquid Plutonium	64
XXIX.	Crystal Structure Data for Plutonium-Nitrogen Alloys	65

XXX.	Solubility of Niobium in Liquid Plutonium	68
XXXI.	Crystal Structure Data for Plutonium-Nickel Alloys	72
XXXII.	Crystal Structure Data for Plutonium-Neptunium Alloys	74
XXXIII.	Crystal Structure Data for Plutonium-Oxygen Alloys	76
XXXIV.	Crystal Structure Data for Plutonium-Osmium Alloys	78
XXXV.	Crystal Structure Data for Plutonium-Phosphorus Alloys	79
XXXVI.	Crystal Structure Data for Plutonium-Lead Alloys	82
XXXVII.	Crystal Structure Data for Plutonium-Palladium Alloys	84
XXXVIII.	Crystal Structure Data for Plutonium-Platinum Alloys	88
XXXIX.	Crystal Structure Data for Plutonium-Rhenium Alloys	89
XL.	Solubility of Rhenium in Liquid Plutonium	89
XLI.	Crystal Structure Data for Plutonium-Rhodium Alloys	92
XLII.	Crystal Structure Data for Plutonium-Ruthenium Alloys	94
XLIII.	Crystal Structure Data for Plutonium-Sulfur Alloys	95
XLIV.	Crystal Structure Data for Plutonium-Scandium Alloys	99
XLV.	Crystal Structure Data for Plutonium-Selenium Alloys	101
XLVI.	Crystal Structure Data for Plutonium-Silicon Alloys	104
XLVII.	Crystal Structure Data for Plutonium-Tin Alloys	107
XLVIII.	Solubility of Plutonium in Liquid Tin	107
XLIX.	Solubility of Tantalum in Liquid Plutonium	110
L.	Crystal Structure Data for Plutonium-Tellurium Alloys	111
LI.	Crystal Structure Data for Plutonium-Thorium Alloys	114
LII.	Solubility of Titanium in Liquid Plutonium	116
LIII.	Crystal Structure Data for Plutonium-Thallium Alloys	117
LIV.	Solubility of Thulium in Liquid Plutonium	119
LV.	Crystal Structure Data for Plutonium-Uranium Alloys	122
LVI.	Solubility of Vanadium in Liquid Plutonium	124
LVII.	Solubility of Tungsten in Liquid Plutonium	126
LVIII.	Crystal Structure Data for Plutonium-Zinc Alloys	130
LIX.	Solubility of Zirconium in Liquid Plutonium	132
LX.	Crystal Structure Data for Plutonium-Zirconium Alloys	132
LXI.	Crystal Structure Data for Plutonium-Cerium-Cobalt Alloys	136
LXII.	Lattice Constants of Gamma-Naught and Gamma Phases in Uranium-20 wt% Plutonium-5, 10, and 15 wt% Fissium Alloys Quenched from Various Temperatures [1].	159
LXIII.	Lattice Constants of ZrRu Phase in Plutonium-Fissium- Uranium Alloys Quenched from Various Temperatures [1].	159

CONSTITUTION OF PLUTONIUM ALLOYS

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INTRODUCTION

This report summarizes the information that was available to the authors prior to July 1967 pertaining to the phase relationships and crystal structures of the intermediate phases in 75 binary and 17 ternary, or higher, alloy systems of plutonium with other elements. The alloy systems are presented in alphabetical order according to the chemical symbols of the nonplutonium elements.

The information given here is mainly in the form of constitutional diagrams and crystal structure tables. Only brief descriptions accompany the diagrams. In general, these descriptions are intended to document information sources, point out any significant differences that may exist between different versions of the diagrams, and clarify certain details of the phase relationships that are known but may not be readily apparent from the diagrams. Many of the diagrams are composites based on the results of more than one group of investigators.

All the binary systems have been drawn

with plutonium as the base. i.e., at the left, and compositions are given in atomic per cent (at.%). Standard nomenclature has been retained for the terminal phases; Greek letters are used to designate intermediate phases in order of increasing content of the nonplutonium alloying elements.

Although a considerable volume of information on at least 50 ternary and higher alloy systems with plutonium, exclusive of any systems involving the metalloids and gases, has been reported in the literature, most of that work relates to the results of engineering test studies (corrosion and thermal expansion data, for example) and is inappropriate for inclusion in this report on the constitution of plutonium alloys. The smaller amount of work that sheds light on the phase relationships in these ternary and higher systems follows the section on binary systems.

Information about the allotropic modifications of plutonium, which may be helpful to the reader in understanding and using the equilibrium diagrams in this report, is summarized in Table I.

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TABLE I. CRYSTAL STRUCTURES AND TRANSFORMATION TEMPERATURES OF THE PLUTONIUM ALLOTROPES^a

Allotrope	Temperature range of stability, °C	Space lattice and space group	Unit cell dimensions, Å	Atoms per unit cell	X-ray density, g/cm ³
α	below ~ 115	primitive monoclinic P2 ₁ /m	(21°C) a = 6.183 b = 4.822 c = 10.963 β = 101.79°	16	19.86
β	~ 115 - ~ 200	body-centered monoclinic I2/m	(190°C) a = 9.284 b = 10.463 c = 7.859 β = 92.13°	34	17.70
γ	~ 200 - 310	face-centered orthorhombic Fddd	(235°C) a = 3.159 b = 5.768 c = 10.162	8	17.14
δ	310 - 458	face-centered cubic Fm3m	(320°C) a = 4.6371	4	15.92
δ'	458 - 480	body-centered tetragonal I4/mmm	(465°C) a = 3.34 c = 4.44	2	16.00
ϵ	480 - 641	body-centered cubic Im3m	(490°C) a = 3.6361	2	16.51

^a From W. H. Zachariasen and F. H. Ellinger, *Acta Cryst.*, **16**: 780 (1963); *ibid.* p. 369; W. H. Zachariasen and F. H. Ellinger, *Acta Cryst.*, **8**: 1431 (1955); and F. H. Ellinger, *Trans Met. Soc. AIME*, **206**: 1256 (1956).

BINARY SYSTEMS

Pu-Ag

Plutonium-Silver

Early work by [1] revealed the existence of immiscible liquids and at least one intermediate phase in this system. That work has been verified and extended by [2], who have published the diagram shown in Fig. 1. Crystal structure data in Table II for PuAg_3 have been determined by [3].

[1] E. M. Cramer, Los Alamos Scientific Laboratory, unpublished work.

[2] V. I. Kutaitsev, N. T. Chebotarev, I. G. Lebedev, M. A. Andrianov, V. N. Konev, and T. S. Menshikova, "Phase Diagrams of Plutonium with the Metals of Groups IIA, IVA, VIIB and IB," in *Plutonium 1965*. A. E. Kay and M. B. Waldron (Eds.). pp. 429-430, 446, Chapman and Hall, London (1967).

[3] O. J. C. Runnalls, "The Crystal Structures of Some Intermetallic Compounds of Plutonium." *Can. J. Chem.*, **34**: 133-145 (1956).

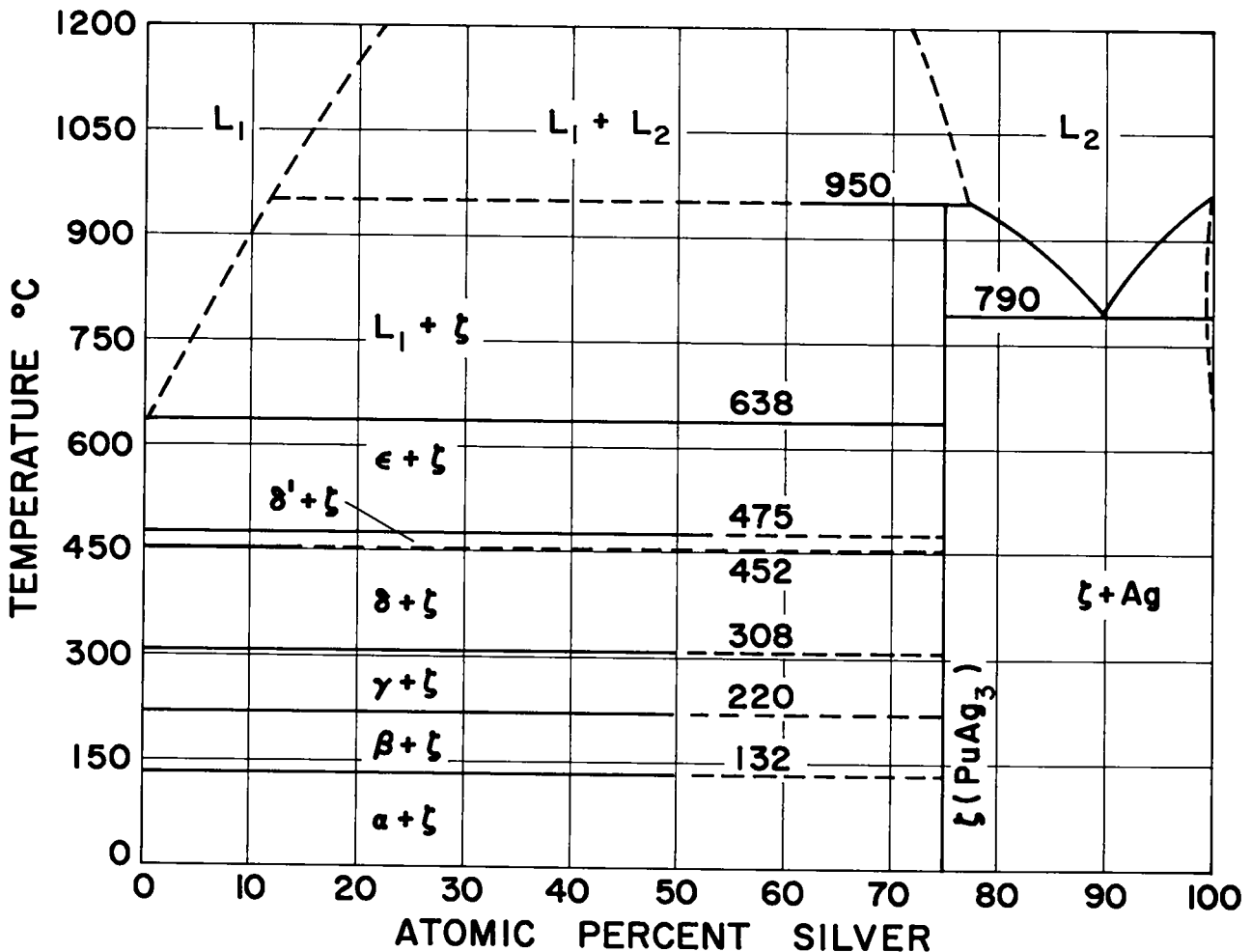


Fig. 1. Plutonium-silver phase diagram. (Redrawn from Ref. 2.)

TABLE II. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-SILVER ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
PuAg ₃		hexagonal	P6 ₃ , P6 ₃ /m, or P6 ₃ 22	12.730		9.402	16	11.33	[3]

Pu-Al □ Plutonium-Aluminum

The phase diagram shown in Fig. 2 is based on the thermal, microstructural, and x-ray data of [1], [2], [3], [4], and [5]. Most of the solid-state phase relationships were identified by [1] and [2], but they did not obtain data adequate to locate the solidus and liquidus boundaries accurately. The results reported by [3] are in agreement with the diagram shown here, with five exceptions: (1) eutectoid decomposition of δ at 175°C; (2) temperatures of 530 and \sim 540°C, respectively, for the PuAl and Pu₃Al peritectoid horizontals; (3) a eutectic temperature of 635°C; (4) some indicated solubility of Al in the γ and β phases; and (5) absence of phase transformations in PuAl₃ and PuAl₄. The high-pressure work of [6] supports the conclusion that δ does not decompose eutectoidally, and the results of [7] show that less than 0.25 at.% Al is soluble in δ' . [8] has suggested that the solid solubility of Pu in Al is approximately 0.26 wt% (0.03 at.%) at 600°C, whereas [9] has reported a maximum solubility of 0.05 wt% (0.0056 at.%) at 650°C. [10] have confirmed the result of [9]. In phase equilibria studies, [5] has observed a phase transformation in PuAl, and several polymorphic transformations in PuAl₃, and has provided crystallographic data. [11], [12], [13], and [14] have also provided crystallographic data on the intermediate compounds (see Table III).

- [1] R. D. Moeller and F. W. Schonfeld, "Alloys of Plutonium with Aluminum," USAEC Report LA-1000, Los Alamos Scientific Laboratory (Feb. 13, 1950).
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- [4] F. H. Ellinger, C. C. Land, and W. N. Miner. "The Solubility Limits of Aluminum in Delta Plutonium and Some Revisions of the Plutonium-Aluminum Phase Diagram," *J. Nucl. Mater.*, **5(2)**: 165-172 (1962).
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- [14] A. C. Larson, D. T. Cromer, and C. K. Stambaugh, "The Crystal Structure of PuAl₃," *Acta Cryst.*, **10**: 443-446 (1957).

TABLE III. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-ALUMINUM ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.	
				a	b	c				
Pu ₃ Al ζ	SrPb ₃	tetragonal	P4/mmm	4.499		4.536	1	13.45	[3]	
				4.499		4.538		13.45	[12]	
	cubic		4.500					[4]		
PuAl η		cubic	I	10.76			29	10.25	[3]	
	10.769					10.253		[4]		
PuAl ₂ θ	Cu ₂ Mg	cubic	Fd3m	7.831			8	8.09	[11]	
				7.840 (Pu-rich)					[3]	
				7.836 (Al-rich)					[3]	
				7.874					[4]	
				7.838 (Pu-rich)					8.06	[12]
				7.848 (Al-rich)					8.095	[12]
			7.833			[5]				
PuAl ₃ ι	PuAl ₃	hexagonal	P6 ₃ /mmc	6.08		14.40	6	6.8	[11]	
				6.084		14.427			[3]	
				6.10		14.47			[14]	
			cubic (3H)	Pm3m	4.262				6.604	[5]
			hexagonal (6H)	P6 ₃ /mmc	6.083		14.410		6.643	[5]
			rhombohedral (9H _β)	R3m	7.879; α = 45.94°				6.657	[5]
			rhombohedral (9H _α)	R3m	7.901; α = 45.81°				6.634	[5]
PuAl ₄ κ	UAl ₄	orthorhombic	Imma	4.42	6.26	13.66	4	6.02-6.11	[11]	
				4.387	6.262	13.714			[3]	
				4.41	6.29	13.79			[13]	
α-PuAl ₄		orthorhombic	Imma	4.396	6.266	13.708		5.680	[5]	
β-PuAl ₄		orthorhombic	Imma	4.396	6.266	13.708		5.680	[5]	



Pu-Am □
Plutonium-Amercium

The phase diagram shown in Fig. 3 was constructed by [1] from microstructural and x-ray data. The phase boundaries for the Am-rich portion of the diagram are only approximated, because few such alloys were available. Although no intermediate phases were observed, [1] do not ex-

clude the possibility of an ordering reaction in the δ phase, which they could not have detected because of the nearly equal scattering factors of the two components.

- [1] F. H. Ellinger, K. A. Johnson, and V. O. Struebing, "The Plutonium-Amercium System," *J. Nucl. Mater.*, **20(1)**: 83-86 (1966).

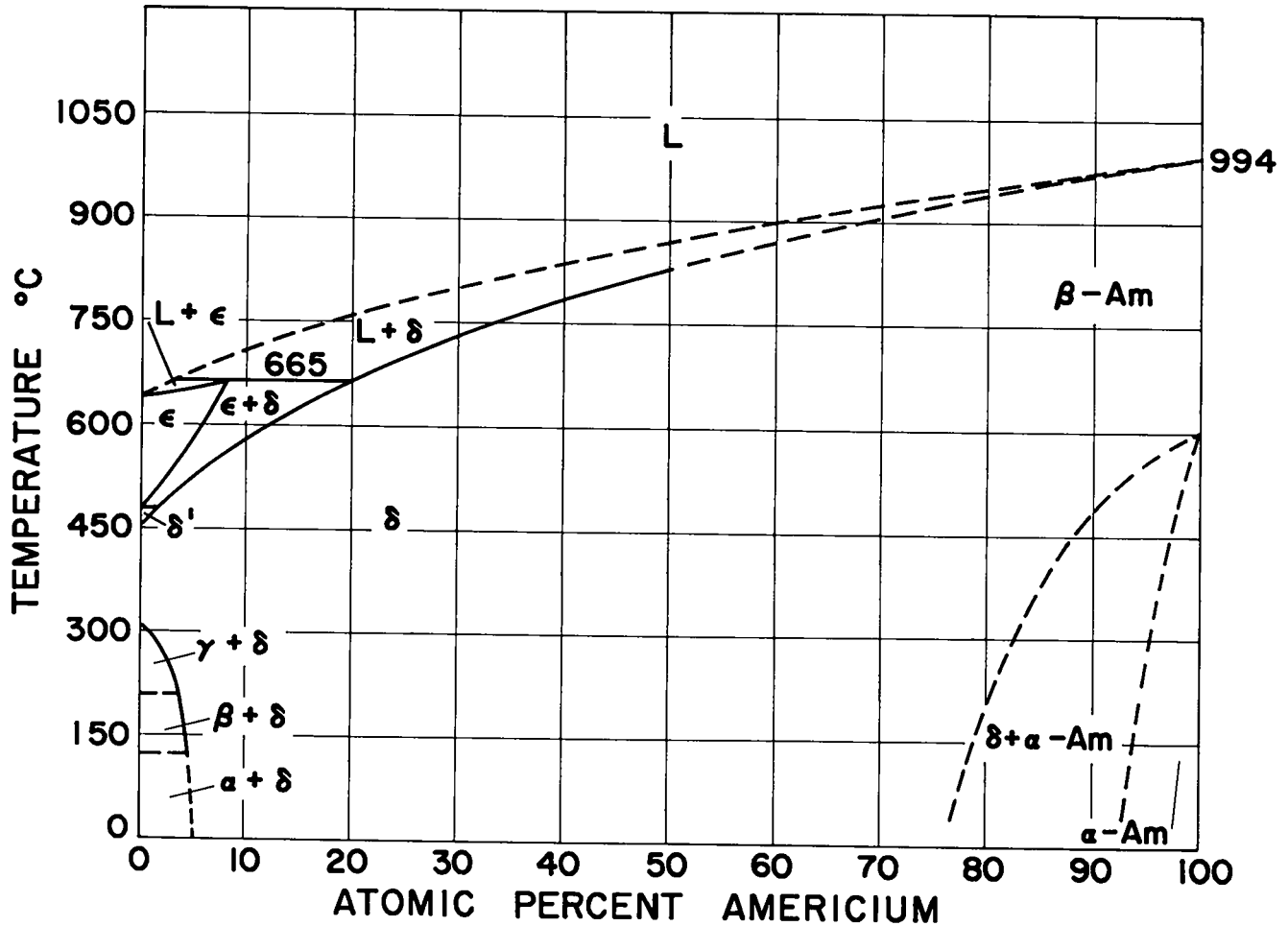
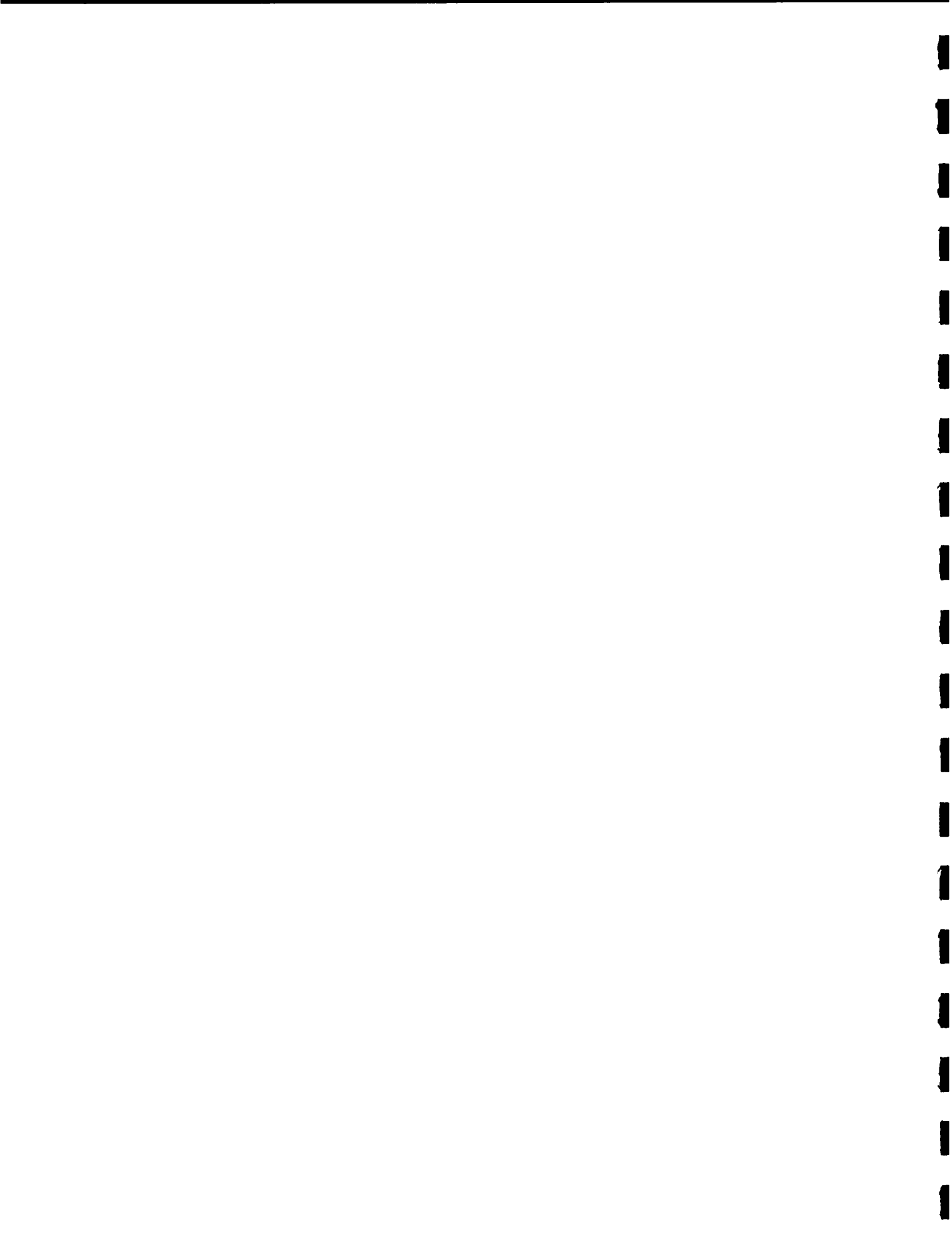


Fig. 3. Plutonium-amerium phase diagram. (Redrawn from Ref. 1.)



Pu-As □**Plutonium-Arsenic**

One compound in this system, PuAs, has been prepared by [1] through induction melting of the components in vacuum and in helium, and was found to decompose at about 2000°C. Attempts by [1] to prepare other compounds were unsuccessful, but [2] have reported that several additional compounds may exist. Crystal structure data for PuAs are given in Table IV.

- [1] A. E. Gorum, "The Crystal Structures of PuAs, PuTe, PuP, and PuOSe," *Acta Cryst.*, **10**: 144 (1957).
- [2] W. M. Pardue, V. W. Storhok, R. A. Smith, and D. L. Keller, "An Evaluation of Plutonium Compounds as Nuclear Fuels, USAEC Report BMI-1968, Battelle Memorial Institute (Oct. 29, 1964).

TABLE IV. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-ARSENIC ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
PuAs	NaCl	cubic	Fm3m	5.855			4	10.39	[1]

Plutonium-Gold

Phase diagrams for this system have been published by [1] and [2]. That by [1] is considered to be preliminary and is based on thermal, dilatometric, microstructural, x-ray, and electron microprobe data. The diagram of [2] is based on thermal, dilatometric, microstructural, and x-ray data. In addition, preliminary results of thermal analysis, metallography, and electron microprobe analysis have been reported by [3]. The results of these investigators are in fair agreement as to such general characteristics of the system as low terminal solid solubilities, a multiplicity of intermediate phases, an Au-rich eutectic, and a maximum liquidus temperature at about 75 at.% Au, but differ considerably in detail. [4] estimated that the most Pu-rich phase contains more than 50 at.% Au, but [5], as well as [2] and [3], has concluded that this phase is PuAu, and [1] have found evidence of a phase containing about 65 at.% Pu. [2] observed the melting point of Pu to be lowered to 635°C by the addition of Au, but [1] and [3] found it to be increased to 652°C, and [6] found the solidus temperature in 5- and 10-at.%-Au alloys to be about 650°C. [1] found eight intermediate phases in the system (see Table V), while [2] and [3] have identified five. [1] suggest that PuAu and PuAu₂ melt congruently, but [2] report that these phases are formed by peritectic reactions. The lattice spacing measurements of [7], made on the Au phase in Au-rich alloys homogenized at 600°C, indicate that the solubility of Pu in Au at that temperature is negligibly small. The dia-

gram shown in Fig. 4 is a composite based mainly on the work of [1], [2], and [3]. The inset box in the diagram outlines the major area of disagreement between [1] and [2].

- [1] B. Hocheid, A. Tanon, S. Bedere, J. Despres, S. Hay, and F. Miard, "Studies of the Binary Systems Plutonium-Gold, Plutonium-Gallium and Plutonium-Indium," in *Plutonium* 1965, A. E. Kay and M. B. Waldron (Eds.), pp. 321-330, Chapman and Hall, London (1967).
- [2] V. I. Kutaitsev, N. T. Chebotarev, I. G. Lebedev, M. A. Andrianov, V. N. Konev, and T. S. Menshikova, "Phase Diagrams of Plutonium with the Metals of Groups IIA, IVA, VIIIA and IB," *Ibid.*, pp. 430-431, 447.
- [3] J. E. Selle, "The Plutonium-Gold System," *Ibid.*, p. 450.
- [4] R. G. Cope, J. N. Lowe, and D. C. Miller, "Studies of Alloys of Plutonium with Transition Elements and Gold," UKAEA Report AWRE 0-36/61 (1961).
- [5] E. M. Cramer, Los Alamos Scientific Laboratory, unpublished work.
- [6] W. N. Miner, Los Alamos Scientific Laboratory, unpublished work.
- [7] F. H. Ellinger, Los Alamos Scientific Laboratory, unpublished work.

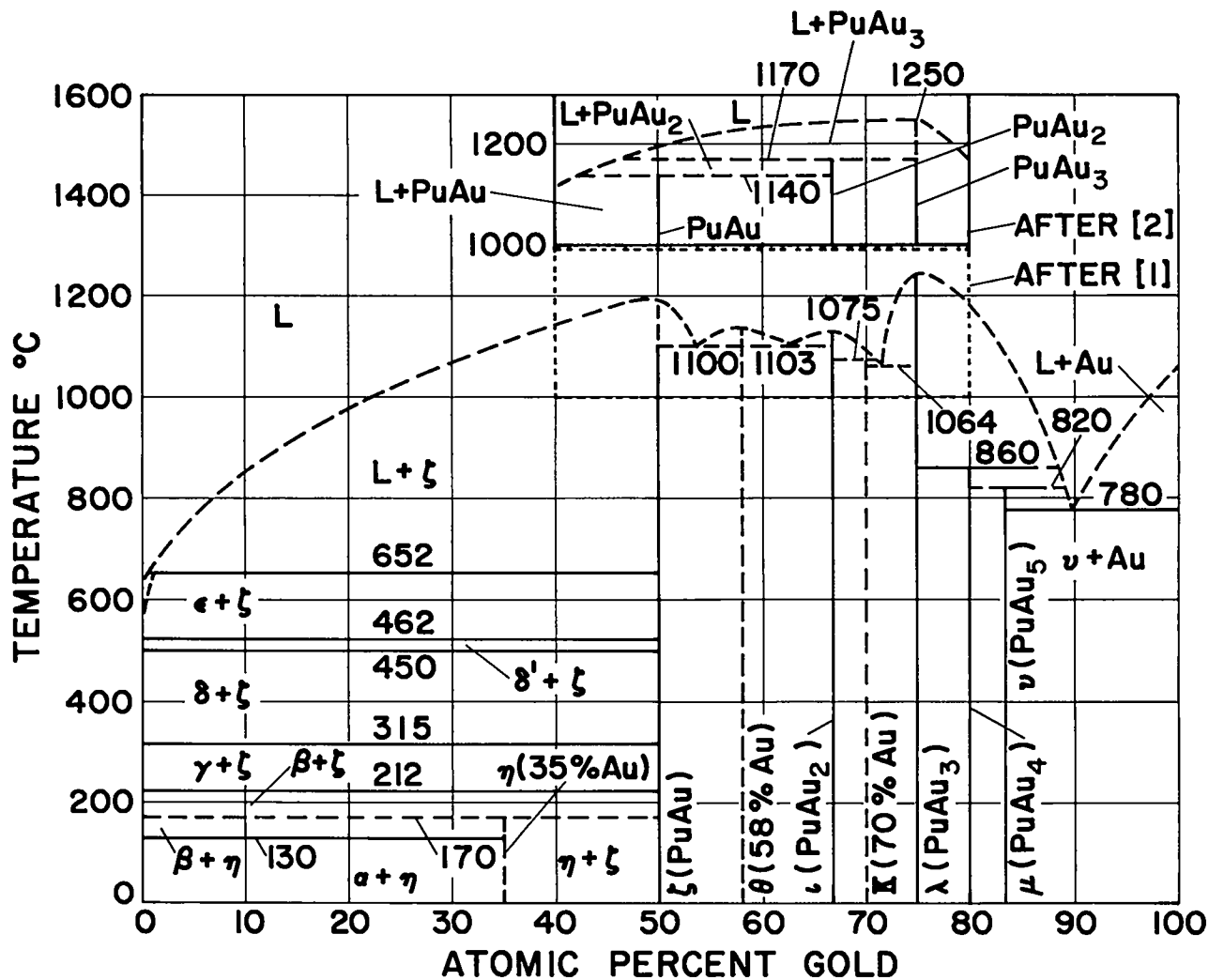


Fig. 4. Plutonium-gold phase diagram.

TABLE V. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-GOLD ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
η (?)									
PuAu	unknown								
ζ									
θ (?)									
PuAu ₂	unknown								
ι									
κ (?)									
PuAu ₃		hexagonal		12.710	9.210	16	17.11	[2]	
λ									
PuAu ₄	unknown								
μ									
PuAu ₅	unknown								
ν									

Pu-B □

Plutonium-Boron

Several compounds are known in this system. PuB₂, PuB₄, and PuB₆ have been reported by [1], [2], and [3]; PuB₁₂ and "PuB₁₀₀" by [3]; and PuB by [1]. However, [2] found no unequivocal evidence for PuB and [3] did not observe it by either x-ray or microstructural examinations. [2] suggest that PuB₄ and PuB₆ are not thermodynamically stable at room temperature. The microstructural evidence of [3] indicates that PuB₄ melts congruently; that PuB₂, PuB₆, PuB₁₂, and "PuB₁₀₀" melt peritectically; and that a eutectic at a composition greater than 95 at.% B occurs between PuB₁₂ and "PuB₁₀₀". The melting points of PuB₄, PuB₆, and PuB₁₂ appear to be above

2150°C, according to [3]. [3] was unable to reproduce the variation in lattice constant reported by [1] and [2] for PuB₆, and found that PuB₂ formed at temperatures as high as 1750°C, in contrast to [1]'s observation that this phase formed at 800°C but not at 1200°C. Crystal structure data are given in Table VI.

- [1] B. J. McDonald and W. I. Stuart, "The Crystal Structures of Some Plutonium Borides," *Acta Cryst.*, **13**(5): 447 (1960).
- [2] R. E. Skavdahl, T. D. Chikalla, and C. E. McNeilly, "Phase Relations in the Plutonium-Boron System," *Am. Nucl. Soc. Trans.*, **7**: 403-404 (1964).
- [3] H. A. Eick, "Plutonium Borides," *Inorg. Chem.*, **4**(8): 1237-1239 (1965).

TABLE VI. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-BORON ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
PuB ₂	AlB ₂	Hexagonal	P6/mmm	3.1857	3.9485	1	12.68	[3]	
				3.18				3.9	[1]
PuB ₄	ThB ₄	Primitive tetragonal	P4/mbm	7.1018	4.0028	4	9.27	[3]	
				7.10				4.014	[1]
PuB ₆	CaB ₆	Primitive cubic	Pm3m	4.1134	4.115 to 4.140	1	7.26 to 7.11	[3]	
				4.115 to 4.140				[1]	
				4.112 to 4.140				[2]	
PuB ₁₂	UB ₁₂	face-centered cubic	Fm3m	7.4843		4		[3]	
"PuB ₁₀₀ "		cubic		23.43				[3]	



Pu-Ba □

Plutonium-Barium

[1], [2], and [3] have attempted to alloy Pu with Ba, Sr, and Ca by direct melting of the components. The results obtained suggest that these elements are immiscible in both the liquid and solid states. [4] state that, except for Be and Mg, the alkaline-earth elements do not react with Pu in the solid state.

- [1] E. M. Cramer, Los Alamos Scientific Laboratory, unpublished work.
- [2] F. W. Schonfeld, "Plutonium Phase Diagrams Studied at Los Alamos," Chap. XXII in *The Metal Plutonium*, A. S. Coffinberry

and W. N. Miner (Eds.), p. 251, The University of Chicago Press, Chicago (1961).

- [3] V. O. Struebing, Los Alamos Scientific Laboratory, unpublished work.
- [4] A. A. Bochvar, S. T. Konobeevsky, V. I. Kutaitsev, T. S. Menshikova, and N. T. Chebotarev, "Interaction Between Plutonium and Other Metals in Connection with Their Arrangement in Mendeleev's Periodic Table," *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958*, Vol. 6, p. 184, United Nations, Geneva (1958).



Plutonium-Beryllium

The reports of studies of Pu-Be alloys by [1], [2], and [3] are in general agreement. The phase diagram shown in Fig. 5 conforms largely with the results of those authors but includes modifications based on the work of [4], [5], and [6]. The high-melting intermediate phase PuBe₁₃ is useful as a neutron source having a predictable neutron yield for a given weight of compound. [4] found the melting point of PuBe₁₃ by optical pyrometer measurements to be 1950 ± 50°C, but [3] gives the melting point as 1700°C. The melting point of Pu was observed by [6] to be lowered a maximum of 10°C at the eutectic horizontal, while [3] found the amount of lowering to be 45°C. Microstructural work of [7] tends to confirm the result of [6]. There appears to be negligibly small solubility of Be in the Pu allotropes, according to [3]. Crystal structure data in Table VII have been reported by [1], [7], and [8].

- [1] A. A. Bochvar, S. T. Konobeevsky, V. I. Kutaitsev, T. S. Menshikova, and N. T. Chebotarev, "Interaction Between Plutonium and Other Metals in Connection with Their Arrangement in Mendeleev's Periodic Table," *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958*, Vol. 6. pp. 184-186, United Nations, Geneva (1958).
- [2] F. W. Schonfeld, "Plutonium Phase Diagrams Studied at Los Alamos," Chap. XXII in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), p. 241, The University of Chicago Press, Chicago (1961).
- [3] S. T. Konobeevsky, "Equilibrium Diagrams of Certain Systems of Plutonium," Session on the Peaceful Uses of Atomic Energy, Section on Chemical Sciences, III: pp. 362-374, Academy of Sciences, Moscow (1955).
- [4] V. O. Struebing, Los Alamos Scientific Laboratory, unpublished work.
- [5] R. D. Moeller and C. R. Tipton, Jr., Los Alamos Scientific Laboratory, unpublished work.
- [6] R. O. Elliott, Los Alamos Scientific Laboratory, unpublished work.
- [7] O. J. C. Runnalls, "The Crystal Structures of Some Intermetallic Compounds of Plutonium," *Can. J. Chem.*, **34**: 133-145 (1956).
- [8] F. H. Ellinger, "A Review of the Intermetallic Compounds of Plutonium," Chap. XXV in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), pp. 286-287, The University of Chicago Press, Chicago (1961).

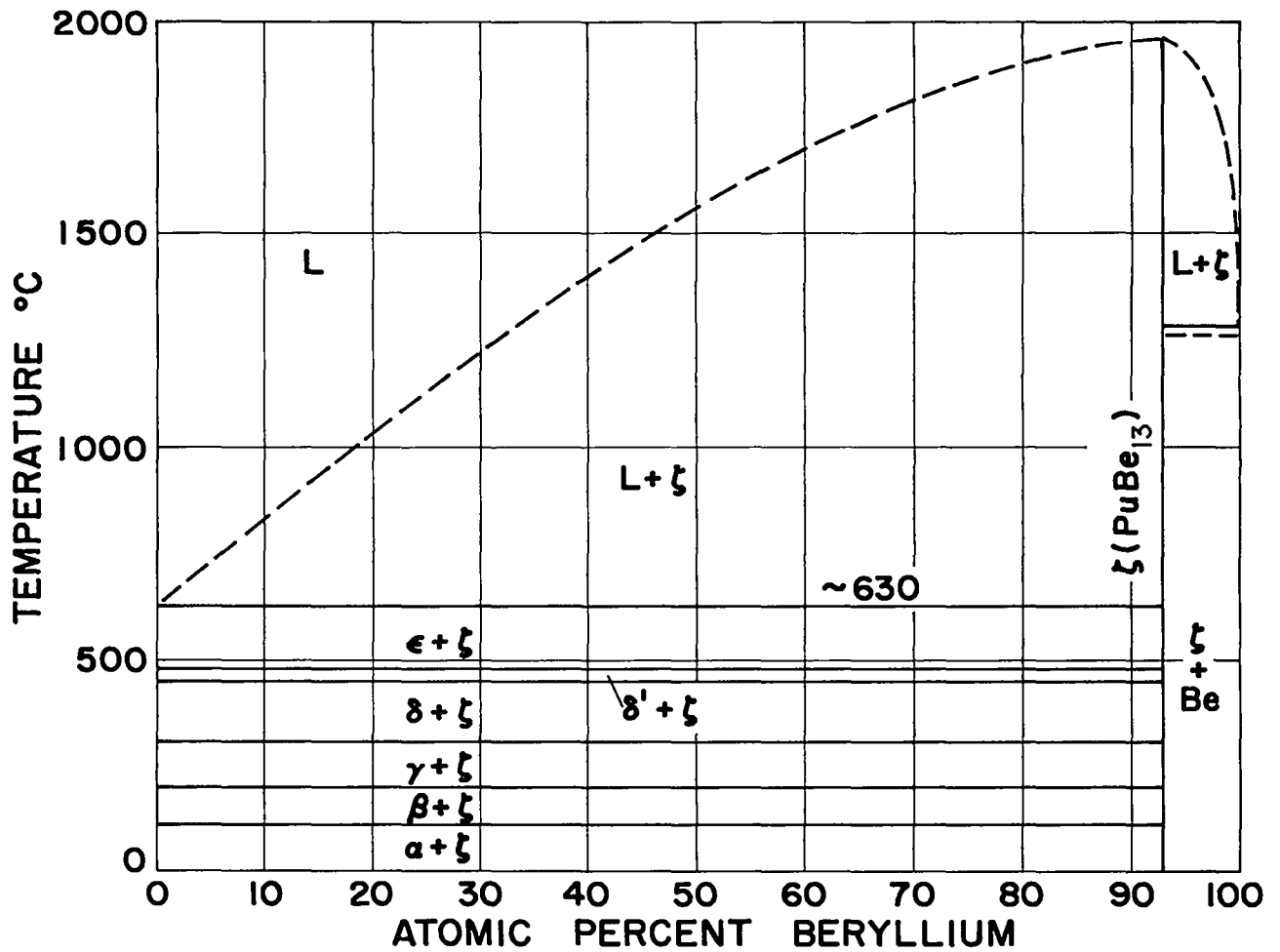


Fig. 5. Plutonium-beryllium phase diagram. (Courtesy Ellinger, Land, and Gschneidner and by permission of Gordon and Breach, Science Publishers, Inc., from *Plutonium Handbook*, Vol. 1, p. 198, 1967.)

TABLE VII. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-BERYLLIUM ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
PuBe ₁₃	NaZn ₁₃	Cubic	Fm3c	10.284 (Be-Rich)			8	4.35	[7]
ζ				10.278 (Pu-Rich)					
				10.274				4.36	[1]
				10.282 (Be-Rich)				4.35	[8]

Pu-Bi □

Plutonium-Bismuth

Partial phase diagrams for this system have been reported by [1] and [2]. The liquidus between 80.8 and 100 at.% Bi has been determined by [3], and the melting point of PuBi was placed at approximately 1300°C by [4]. The diagram shown in Fig. 6 is based on the combined results of these investigators. The major point of difference between the diagram of [2] and that shown here is the temperature of the peritectic formation of PuBi₂, which [2] give as 528°C. Crystal structure data have been reported by [2] and [5] and are given in Table VIII.

[1] F. W. Schonfeld. "Plutonium Phase Diagrams Studied at Los Alamos," Chap. XXII in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), p. 241. The University of Chicago Press, Chicago (1961).

[2] A. A. Bochvar, S. T. Konobeevsky, V. I.

Kutaitsev, T. S. Menshikova, and N. T. Chebotarev, "Interaction of Plutonium and Other Metals in Connection with Their Arrangement in Mendeleev's Periodic Table." *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958*, Vol. 6, p. 184-193, United Nations, Geneva (1958).

[3] R. O. Elliott and E. M. Cramer, Los Alamos Scientific Laboratory, unpublished work.

[4] V. O. Struebing, Los Alamos Scientific Laboratory, unpublished work.

[5] F. H. Ellinger, "A Review of the Intermetallic Compounds of Plutonium," Chap. XXV in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), p. 287, The University of Chicago Press, Chicago (1961).

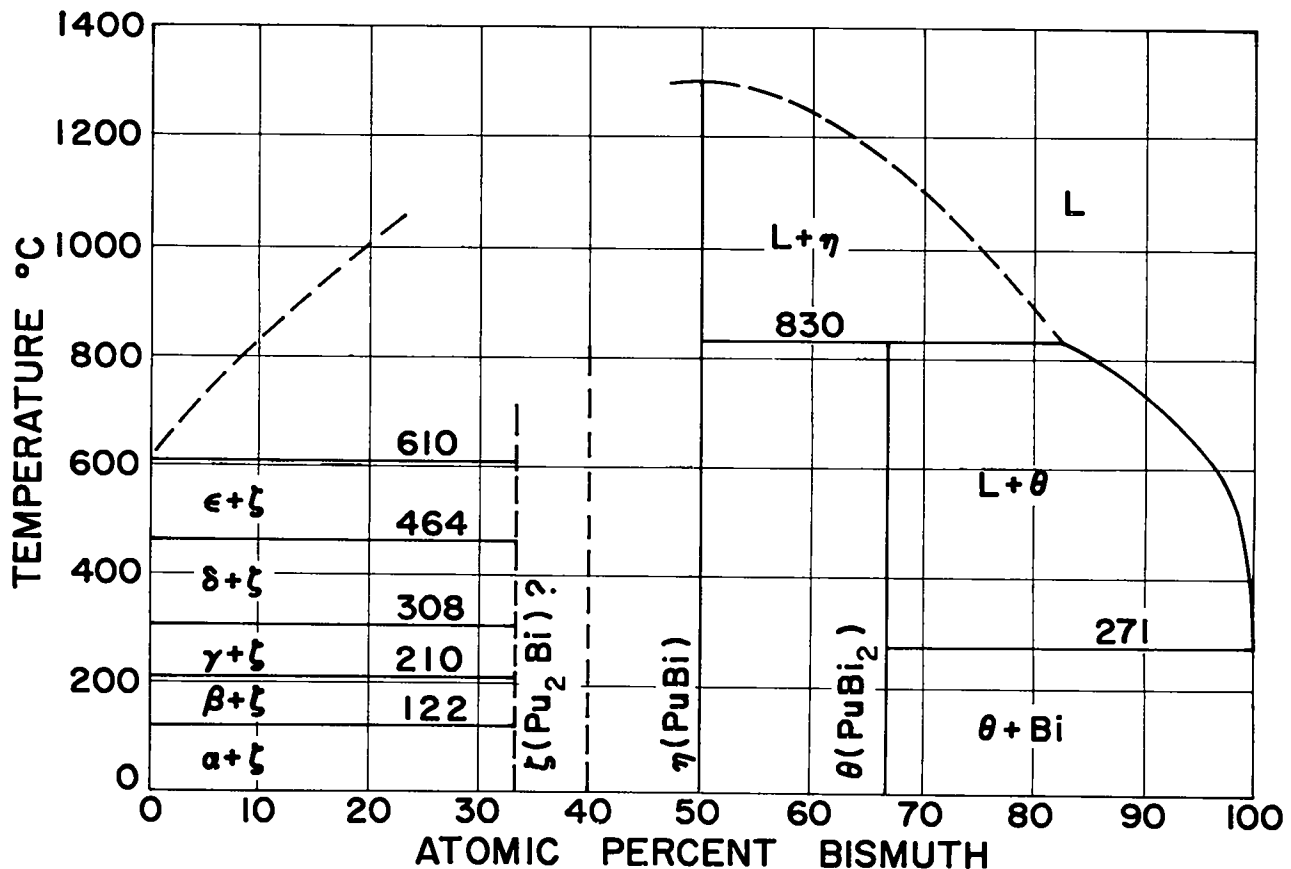


Fig. 6. Plutonium-bismuth phase diagram. (Courtesy Ellinger, Land, and Gschneidner and by permission of Gordon and Breach, Science Publishers, Inc., from *Plutonium Handbook*, Vol. 1, p. 199, 1967.)

TABLE VIII. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-BISMUTH ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
Pu ₂ Bi(?) ξ	unknown								
PuBi η	NaCl	face-centered cubic	Fm3m	6.348 6.350			4	11.63 11.62	[2] [5]
PuBi ₂ θ	unknown								

Pu-C □

Plutonium-Carbon

The phase diagram shown in Fig. 7 is mainly that of [1] but includes minor modifications based on the later work of [2], [3], and [4]. Additions of C lower the melting point of Pu about 10°C; however, [1] did not observe a eutectic microstructure in either quenched or slowly cooled alloys containing between 0.5 and 30 at.% C. The eutectic temperature has been reported as 637°C by [3]. Thermal analysis results of [1] show that C additions have little or no effect on the solid state transformations of Pu, and thus indicate negligibly small solubility of C in the Pu allotropes. Also, [5] found no significant solubility of C in δ' or ϵ Pu. The temperature of the peritectoid reaction ($\epsilon + \eta \rightarrow \zeta$) was placed at 575°C by [1], between 577° and 585°C by [2], and at 558°C by [3]. The monocarbide, commonly referred to as PuC, exists over an appreciable range of composition and, according to [6], has a defect structure containing at saturation only 89% of the theoretical number of C atoms. The

maximum C content of PuC has been reported by [2], [3], [4], and [6] to be near 48 at.%, and the minimum has been reported by [2], [3], and [4] to be near 42.5 at.%. The volume fraction analysis data of [2] indicate that Pu₂C₃ has a solubility range of about 1 at.% above 855°C, and [4] has suggested that Pu₂C₃ has a small range of homogeneity. PuC and Pu₂C₃ are found in as-cast alloys but Pu₃C₂ and PuC₂ are found in significant proportion only in alloys that have been appropriately heat treated. Crystal structure data for PuC and Pu₂C₃ have been determined by [1], [6], [7], [8], and [9] and are given in Table IX. The structures of Pu₃C₂ and PuC₂ are unknown. The solubility of C in liquid Pu, as determined by [10] is given in Table X.

- [1] R. N. R. Mulford, F. H. Ellinger, G. S. Hendrix, and E. D. Albrecht, "The Plutonium-Carbon System," in *Plutonium 1960*, E. Grison, W. B. H. Lord, and R. D. Fowler (Eds.), pp. 301-311. Cleaver-Hume Press. Ltd., London (1961).
- [2] K. A. Johnson, "Metallographic Prepara-

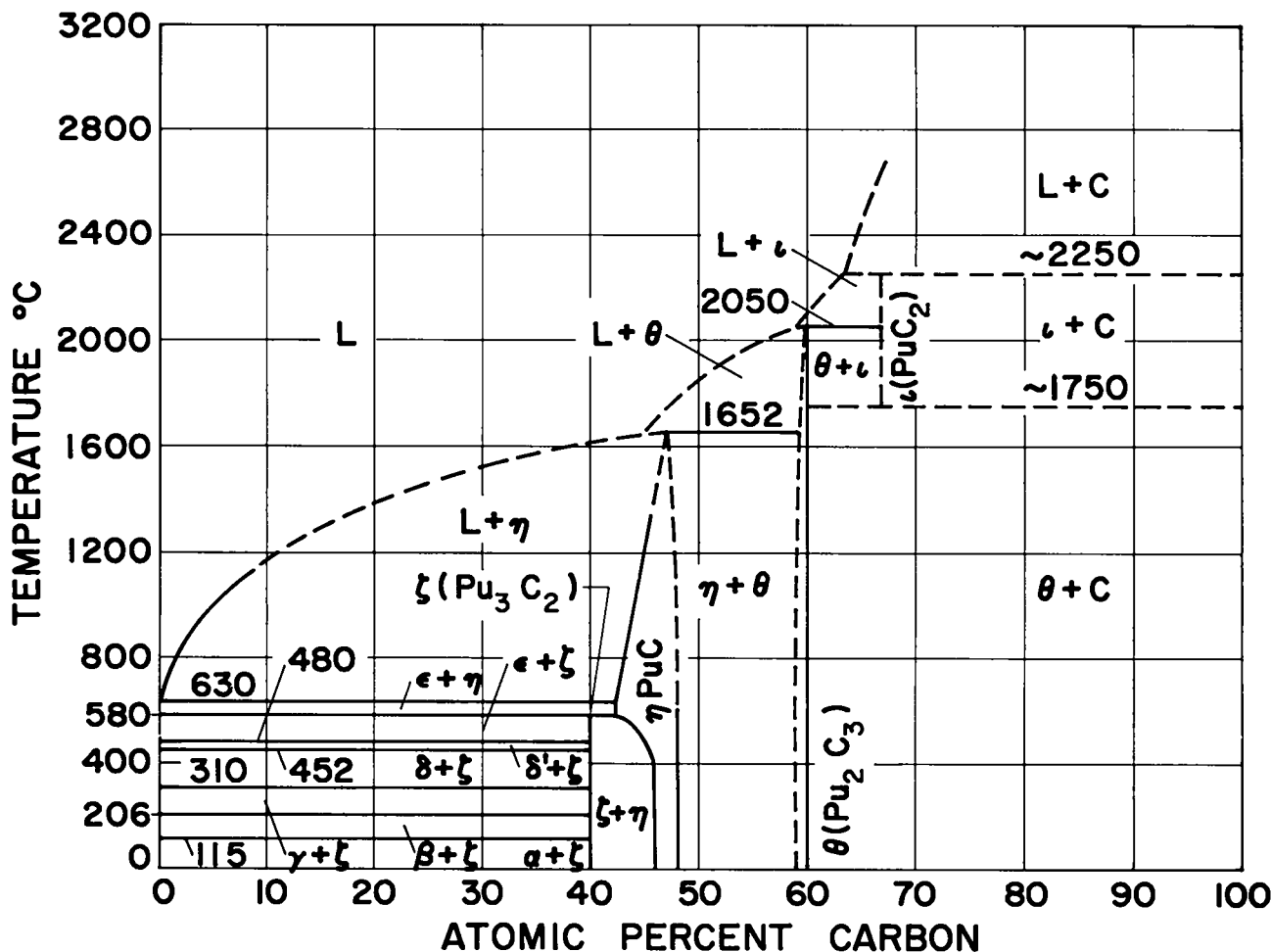


Fig. 7. Plutonium-carbon phase diagram.

- tion and Observations of Some Plutonium-Carbon Alloys," USAEC Report LA-3191-MS, Los Alamos Scientific Laboratory (November 1964).
- [3] S. Rosen, M. V. Nevitt, and A. W. Mitchell, "Metallographic and X-ray Observations of Pu-C Alloys," USAEC Report TID-17867, Argonne National Laboratory (November 1962).
- [4] O.L. Kruger, "Phase Studies on Arc-Melted Plutonium-Carbon Alloys Near the Monocarbide Composition," *J. Am. Ceramic Soc.*, **46**(2): 80-85 (1963).
- [5] R. O. Elliott and A. C. Larson, "Delta-Prime Plutonium," Chap. XXIV in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), pp. 265-280, The University of Chicago Press, Chicago (1961).
- [6] R. Pascard, "Etudes preliminaries sur le systeme plutonium-carbone et les solutions solides carbure d'uranium-carbure de plutonium." (in French), in *Powder Metallurgy in the Nuclear Age — Plansee Proceedings 1961*, F. Benesovsky (Ed.), pp. 387-419, Springer-Verlag, Vienna (1962).
- [7] W. H. Zachariasen, "Crystal Chemical Studies of the 5f-Series of Elements: XII. New Compounds Representing Known Structure Types," *Acta Cryst.*, **2**: 388-390 (1949).
- [8] J. L. Drummond, B. J. McDonald, H. M. Ockenden, and G. A. Welch, "The Preparation and Properties of Some Plutonium Compounds: Part VII. Plutonium Carbides," *J. Chem. Soc.*, 1957 (Part IV): 4785-4789 (1957).
- [9] W. H. Zachariasen, "Crystal Chemical Studies of the 5f-Series of Elements: XV. The Crystal Structure of Plutonium Sesquicarbide," *Acta Cryst.*, **5**: 17-19 (1952).
- [10] D. F. Bowersox and J. A. Leary, "The Solubilities of Carbon, Tantalum, Tungsten, and Rhenium in Liquid Plutonium. *J. Nucl. Mater.*, **21**: 219-224 (1967).

TABLE X. SOLUBILITY OF CARBON IN LIQUID PLUTONIUM

Temperature, °C	Solubility, at. % C
700	1.36 ± 0.03
750	1.61 ± 0.06
800	2.26 ± 0.14
850	2.67 ± 0.08
900	3.57 ± 0.13
950	4.47 ± 0.27

TABLE IX. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-CARBON ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
Pu ₃ C ₂ ξ	Unknown								
PuC η	NaCl	face-centered cubic	Fm3m	4.958 (Pu-rich) 4.974 (C-rich) 4.959 (Pu-rich) 4.973 (C-rich) 4.968 (Pu-rich) 4.978 (C-rich)		4	13.6	[1, 7] [1] [8] [8] [6] [6]	
Pu ₂ C ₃ θ	Pu ₂ C ₃	body-centered cubic	I43d	8.1258 (Pu-rich) 8.1317 (C-rich)		8	12.70	[1, 9] [1]	
PuC ₂ ι	Unknown								

Pu-Ca **Plutonium-Calcium**

See Pu-Ba (Plutonium-Barium).

Pu-Cd **Plutonium-Cadmium**

A complete diagram for this system has been published by [1], and a partial diagram, relative to Cd-rich alloys, has been published by [2]. With one exception, the diagrams are in reasonable agreement in the area in which they overlap. The compound judged by [1] to be PuCd_4 , on the basis of quantitative metallography, is designated as PuCd_3 by [2], on the basis of mixed x-ray diffraction patterns. The diagram of [1]

is shown in Fig. 8. Crystal structure data are given in Table XI.

- [1] D. E. Etter, D. B. Martin, D. L. Roesch, C. R. Hudgens, and P. A. Tucker, "The Plutonium-Cadmium Binary System," *Trans. Met. Soc. AIME*, **233**: 2011-2013 (1965).
- [2] I. Johnson, M. G. Chasanov, and R. M. Yonco, "Pu-Cd System: Thermodynamics and Partial Phase Diagram," *Trans. Met. Soc. AIME*, **233**: 1408-1414 (1965).

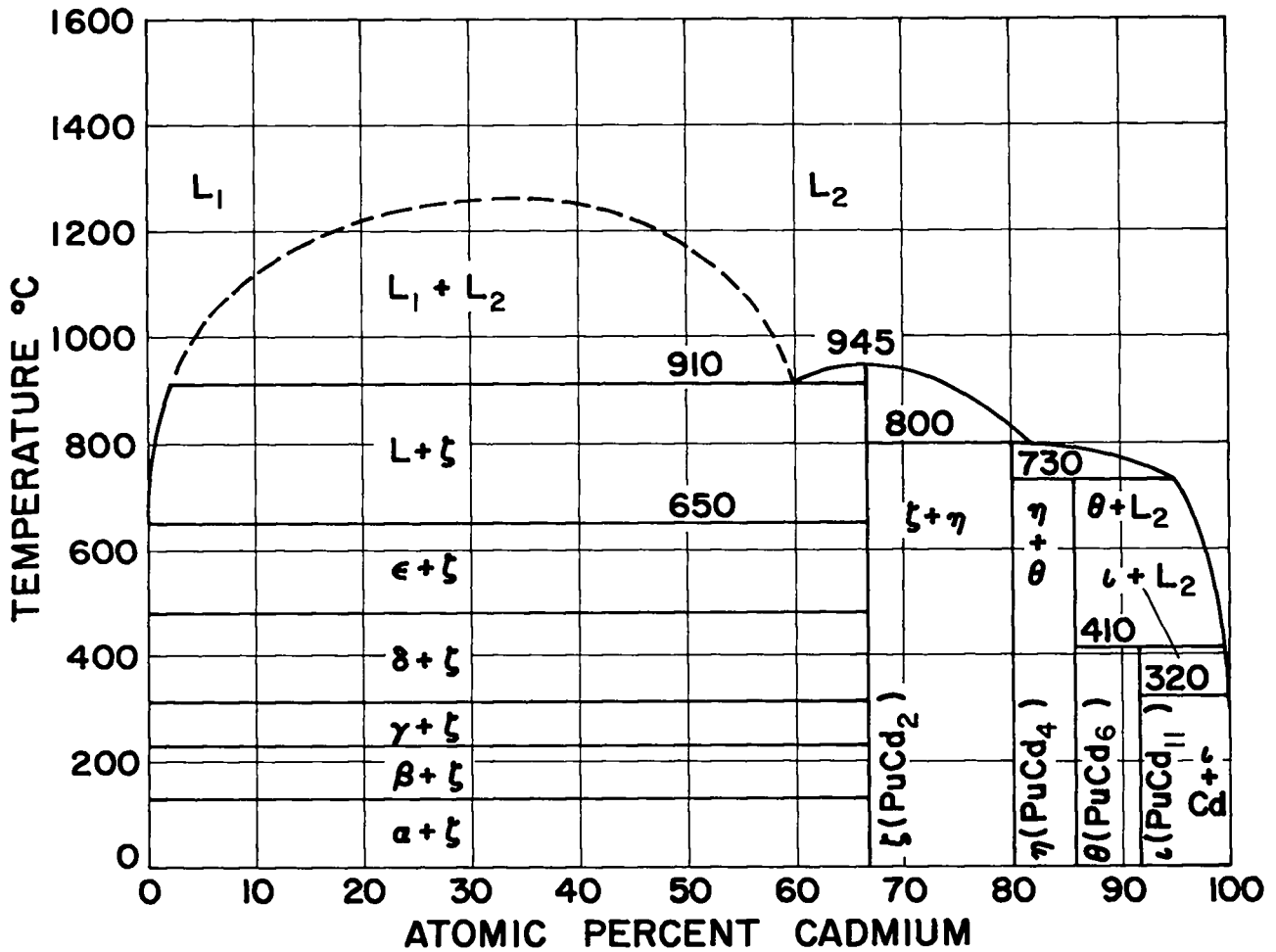


Fig. 8. Plutonium-cadmium phase diagram. (Redrawn from Ref. 1.)

TABLE XI. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-CADMIUM ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
PuCd ₂ ξ	Unknown								
PuCd ₄ (?) η	Unknown								
PuCd ₆ θ		Cubic		15.59					[2]
PuCd ₁₁ ι		Cubic		9.282					[2]

Pu-Ce □

Plutonium-Cerium

Complete diagrams for this system have been published by [1] and [2]. These are in general agreement except for the shape of the liquidus boundary and the mode of transformation of the high-temperature Ce-rich solid solution, which are not yet definitely established. Also, [2] report 613°C instead of 592°C for the temperature of the peritectoid reaction $\epsilon + \gamma$ Ce \rightarrow δ Pu. The diagram shown in Fig. 9 is based

on the results of these two groups of investigators.

- [1] F. H. Ellinger, C. C. Land, and E. M. Cramer, "Plutonium-Cerium Phase Diagram," in *Extractive and Physical Metallurgy of Plutonium and Its Alloys*, W. D. Wilkinson (Ed.), pp. 149-167, Interscience Publishers, New York (1960).
- [2] J. E. Selle and D. E. Etter, "The Plutonium-Cerium System," *Trans. Met. Soc. AIME*, **230**: 1000-1005 (1964).

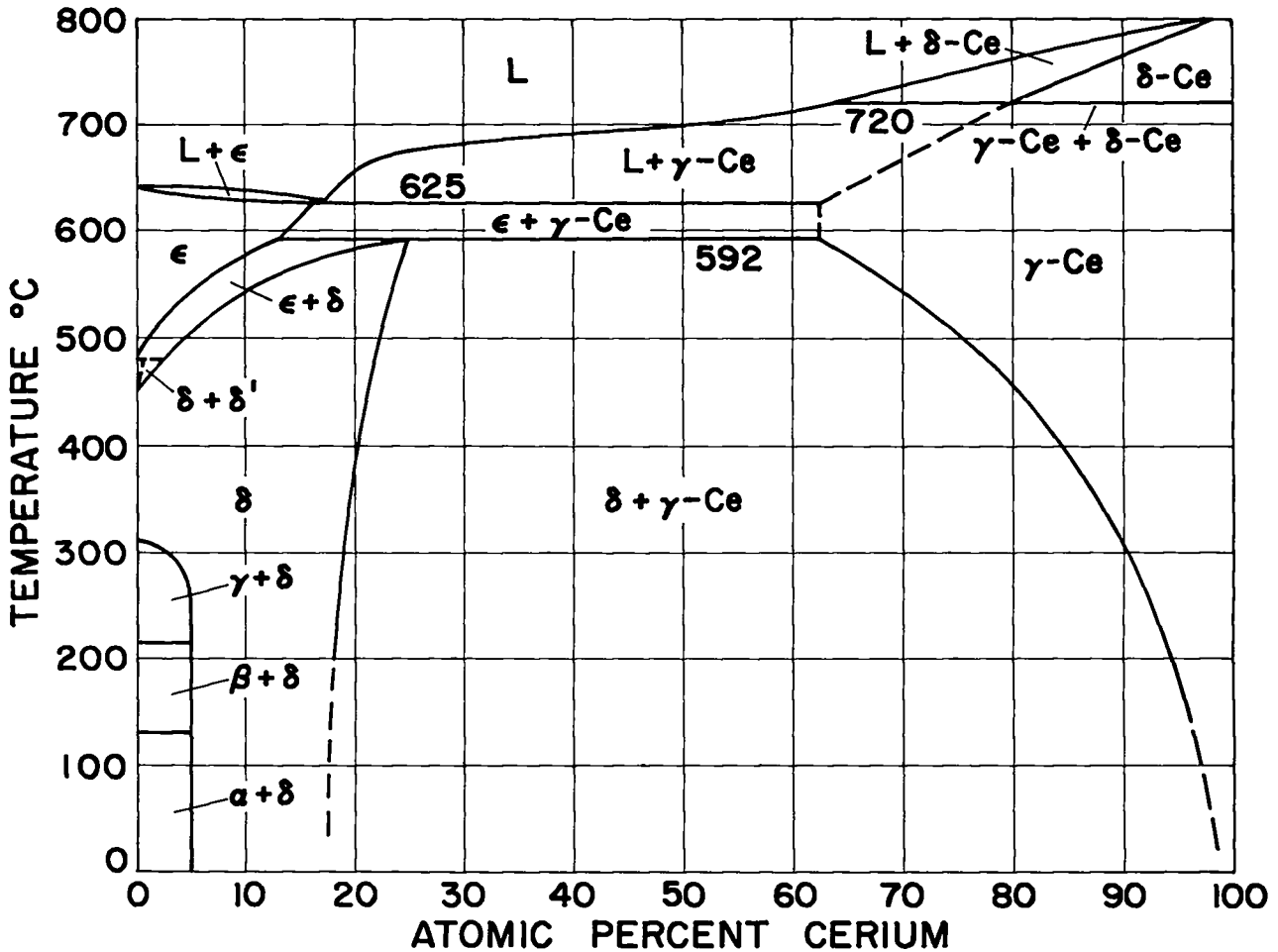


Fig. 9. Plutonium-cerium phase diagram.



Plutonium-Cobalt

A nearly complete diagram for this system has been reported by [1], a partial diagram has been published by [2]. the phase relationships and solubilities in Pu-rich alloys have been studied by [3]. and liquidus temperatures have been determined by [4]. The diagram shown in Fig. 10 is based on the combined results of these investigators. Crystal structure data given in Table XII have been reported by [1]. [5]. [6]. [7], and [8].

- [1] D. M. Poole and J. L. Nichols, "The Plutonium-Cobalt System." UKAEA Report AERE-R 3609 (1961).
- [2] F. W. Schonfeld, "Plutonium Phase Diagrams Studied at Los Alamos." Chap. XXII in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), p. 242. The University of Chicago Press, Chicago (1961).
- [3] R. O. Elliott and A. C. Larson, "Delta-Prime Plutonium," Chap. XXIV. Ibid., pp. 265-280.

- [4] C. C. Land, Los Alamos Scientific Laboratory. unpublished work.
- [5] A. A. Bochvar, S. T. Konobeevsky, V. I. Kutaitsev, T. S. Menshikova, and N. T. Chebotarev, "Interaction of Plutonium and other Metals in Connection with Their Arrangement in Mendeleev's Periodic Table," *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958*, Vol. 6, pp. 187-188, United Nations, Geneva (1958).
- [6] F. H. Ellinger, "A Review of the Intermetallic Compounds of Plutonium," Chap. XXV in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), pp. 288-290, The University of Chicago Press, Chicago (1961).
- [7] A. C. Larson, D. T. Cromer, and R. B. Roof, Jr., "The Crystal Structure of Pu_3Co ," *Acta Cryst.*, **16**: 835-836 (1963).
- [8] O. J. C. Runnalls, "The Crystal Structures of Some Intermetallic Compounds of Plutonium," *Can. J. Chem.*, **34**: 133-145 (1956).

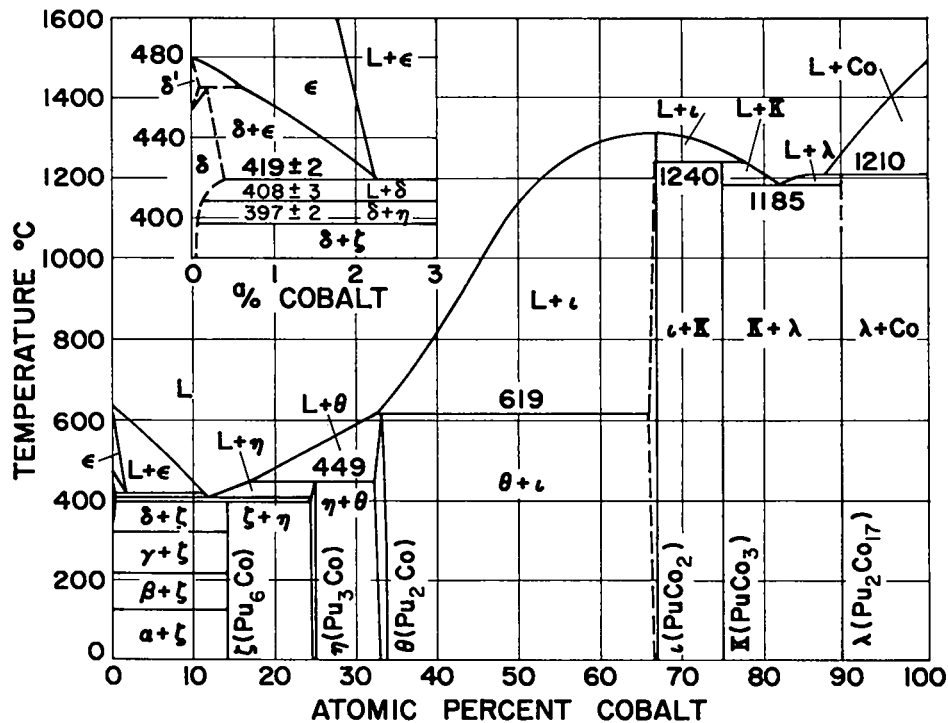


Fig. 10. Plutonium-cobalt phase diagram (Courtesy Ellinger, Land, and Gschneidner and by permission of Gordon and Breach, Science Publishers, Inc., from *Plutonium Handbook*, Vol. 1, p. 203, 1967.)

TABLE XII. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-COBALT ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
Pu ₆ Co ζ	U ₆ Mn	tetragonal	I4/mcm	10.45		5.32	4	17.06	[5]
				10.46		5.33		17.00	[6]
				10.475		5.340		[1]	
Pu ₃ Co η	Al ₂ CuMg	orthorhombic	Cmcm	3.501	11.03	9.23	(Pu-rich)	14.7	[1]
				3.477	10.99	9.20	(Co-rich)		[1]
				3.470	10.939	9.196	4	14.76	[6]
				3.475	10.976	9.220		14.65	[7]
Pu ₂ Co θ	Fe ₂ P	hexagonal	P321	7.902	(Pu-rich)	3.549	3	14.0	[6]
				7.763	(Co-rich)	3.648			[6]
				7.803	(Pu-rich)	3.606			[1]
				7.732	(Co-rich)	3.654			[1]
PuCo ₂ ι	Cu ₂ Mg	cubic	Fd3m	7.075			8	13.39	[8]
				7.083	(Pu-rich)				[5]
				7.066	(Co-rich)				[5]
				7.081	(Pu-rich)				[6]
				7.095	(Pu-rich)				[1]
				7.023	(Co-rich)				[1]
PuCo ₃ κ	PuNi ₃	rhombohedral (in hexagonal system)	R $\bar{3}$ m	8.635; α = 33°40'			3	11.74	[1]
				5.003	24.42				
Pu ₂ Co ₁₇ λ	Th ₂ Ni ₁₇	hexagonal	P6 ₃ /mmc	8.325		8.104	2	10.10	[6]
				8.327		8.107			[1]

Pu-Cr **Plutonium-Chromium**

The phase diagram of [1] and the data of [2], [3], and [4] are in general agreement as to phase relationships but differ in details (see Fig. 11). [1] have placed the eutectic temperature at 615°C, and the composition at less than 2.2 at.% Cr; whereas [2] found the temperature to be 626°C, and [3] did not find metallographic evidence for a eutectic structure in alloys containing 1 and 2 at.% Cr. The data of [4] suggest that the maximum solubility of Cr in δ Pu is about 1 at.%. Solubility data determined by [5] for Cr in liquid Pu are given in Table XIII.

Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958, Vol. 6, pp. 184-193, United Nations, Geneva (1958).

- [1] A. A. Bochvar, S. T. Konobeevsky, V. I. Kutaitsev, T. S. Menshikova, and N. T. Chebotarev, "Interaction of Plutonium and Other Metals in Connection with Their Arrangement in Mendeleev's Periodic Table,"
- [2] W. N. Miner, Los Alamos Scientific Laboratory, unpublished work.
- [3] E. M. Cramer, Los Alamos Scientific Laboratory, unpublished work.
- [4] F. H. Ellinger, Los Alamos Scientific Laboratory, unpublished work.
- [5] D. F. Bowersox, "The Solubilities of Selected Elements in Liquid Plutonium: XII. Chromium." USAEC Report LA-3850, Los Alamos Scientific Laboratory (March 11, 1968).

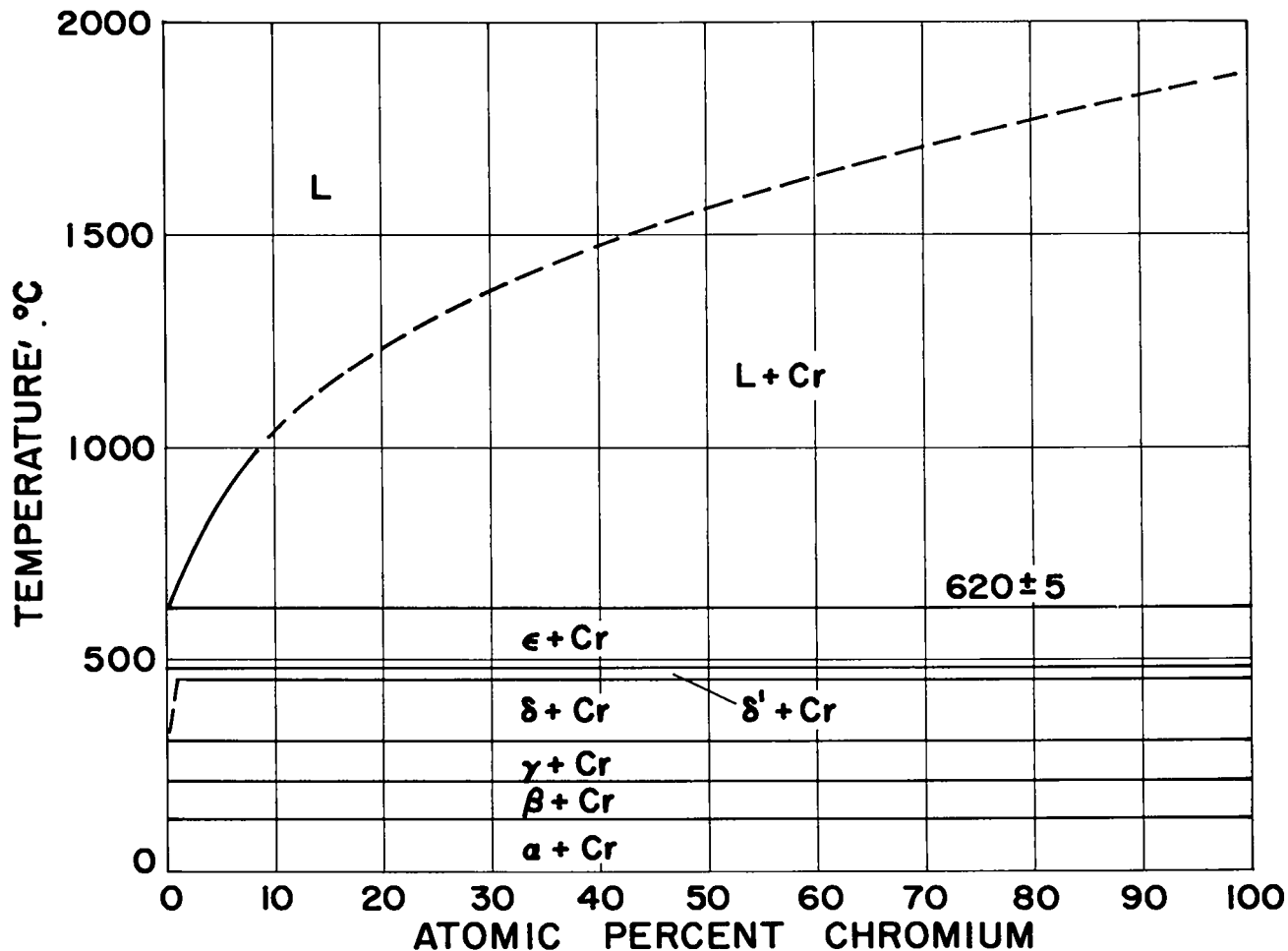


Fig. 11. Plutonium-chromium phase diagram.

TABLE XIII. SOLUBILITY OF CHROMIUM
IN LIQUID PLUTONIUM

Temperature, °C	Solubility, at. % Cr
700	1.62 ± 0.03
750	2.23 ± 0.00
800	3.26 ± 0.01
850	4.35 ± 0.01
900	5.59 ± 0.00
950	6.94 ± 0.01
1000	8.58 ± 0.01

Pu-Cs □

Plutonium-Cesium

Pu appears to be completely immiscible with the alkali metals in both the liquid and solid states, according to [1], and [2] have stated that the alkali metals do not react with Pu in the solid state.

[1] F. W. Schonfeld, "Plutonium Phase Diagrams Studied at Los Alamos," Chap. XXII in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), p. 251. The Uni-

versity of Chicago Press, Chicago (1961).

[2] A. A. Bochvar, S. T. Konobeevsky, V. I. Kutaitsev, T. S. Menshikova, and N. T. Chebotarev, "Interaction Between Plutonium and Other Metals in Connection with Their Arrangement in Mendeleev's Periodic Table," *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva*, 1958, Vol. 6, p. 184, United Nations, Geneva (1958).



Pu-Cu □

Plutonium-Copper

Phase diagrams for this system have been proposed by [1], [2], and [3]. With the exception of the number and composition of the intermetallic compounds, the results of the three investigations are in good agreement. [1] reported the existence of PuCu_2 , PuCu_4 , and PuCu_6 ; [2] found evidence for PuCu_2 , PuCu_4 , $\text{Pu}_4\text{Cu}_{17}$, and $\text{Pu}_2\text{Cu}_{11}$; and [3] identified PuCu_2 , PuCu_4 , $\text{Pu}_4\text{Cu}_{17}$, and PuCu_6 , and stated that a structure determination confirmed the composition PuCu_6 , with no supporting evidence for $\text{Pu}_2\text{Cu}_{11}$. The diagram shown in Fig. 12 is based mainly on the work of [2] and [3]. Crystal structure data are given in Table XIV.

[1] A. A. Bochvar, S. T. Konobeevsky, V. I. Kutaitsev, T. S. Menshikova, and N. T. Chebotarev, "Interaction of Plutonium and Other Metals in Connection with Their Ar-

rangement in Mendeleev's Periodic Table." *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958*, Vol. 6, pp. 184-193, United Nations, Geneva (1958).

[2] T. B. Rhinehammer, D. E. Etter, and L. V. Jones, "The Plutonium-Copper Phase Diagram," in *Plutonium 1960*, E. Grison, W. B. H. Lord, and R. D. Fowler (Eds.), pp. 289-300, Cleaver-Hume Press, Ltd., London (1961).

[3] V. I. Kutaitsev, N. T. Chebotarev, I. G. Lebedev, M. A. Andrianov, V. N. Konev, and T. S. Menshikova, "Phase Diagrams of Plutonium with the Metals of Groups IIA, IVA, VIIIA and IB," in *Plutonium 1965*, A. E. Kay and M. B. Waldron (Eds.), pp. 429 and 445. Chapman and Hall, London (1967).

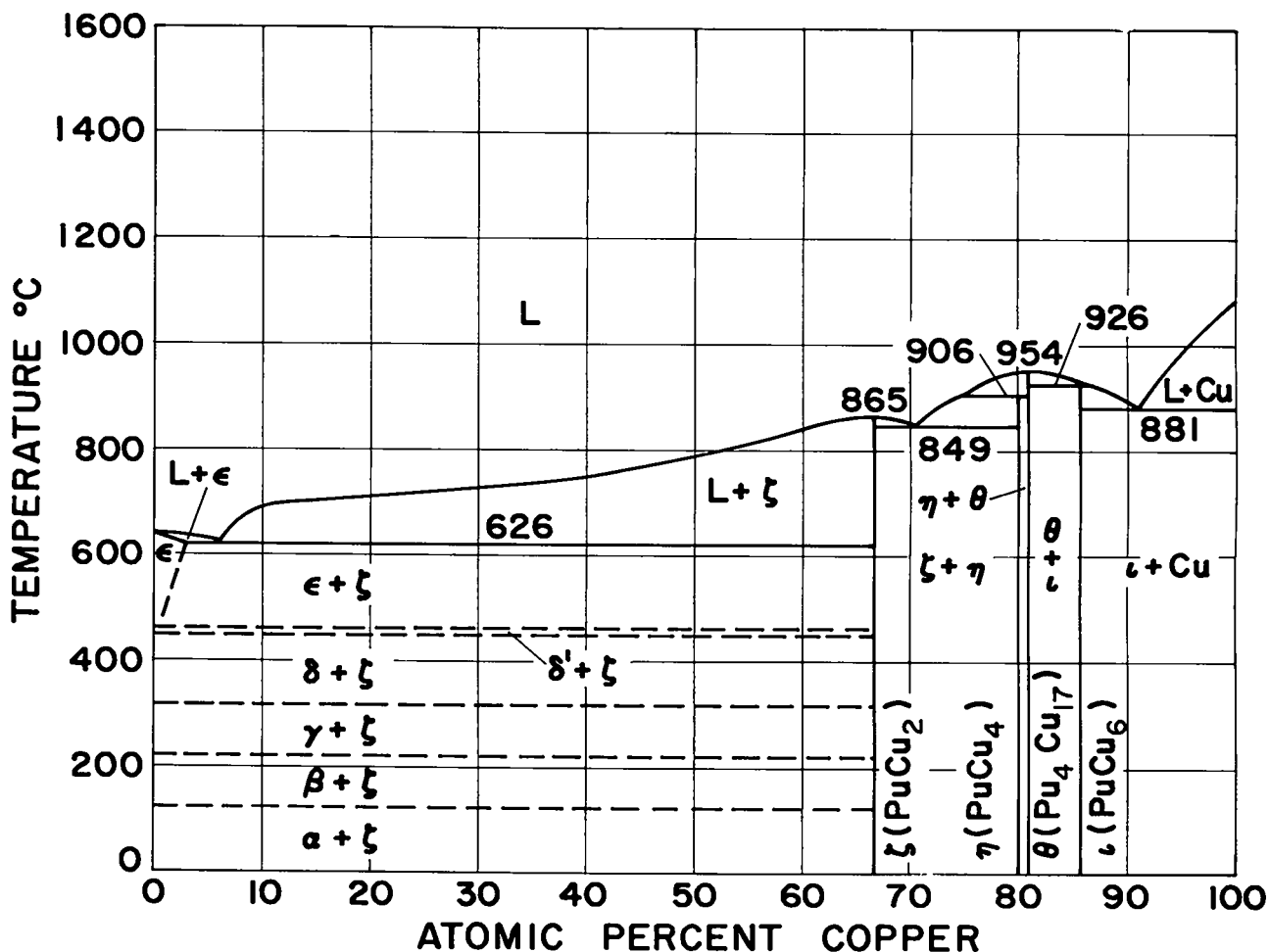


Fig. 12. Plutonium-copper phase diagram.

TABLE XIV. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-COPPER ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
PuCu ₂ ζ	unknown								
PuCu ₄ η	unknown								
Pu ₄ Cu ₁₇ θ	unknown								
PuCu ₆ ε	CeCu ₆	orthorhombic	Pnma	8.50	5.025	10.059	4	10.12	[3]

Pu-Dy **Plutonium-Dysprosium**

See Pu-Gd (Plutonium-Gadolinium).

Pu-Er **Plutonium-Erbium**

See Pu-Gd (Plutonium-Gadolinium).

Pu-Eu **Plutonium-Europium**

It has been found by [1] that liquid Pu is completely immiscible with liquid Eu and Yb.

[1] F. H. Ellinger, C. C. Land, and K. A. Johnson, Los Alamos Scientific Laboratory, unpublished work as of June 1967.



Pu-Fe □

Plutonium-Iron

Phase diagrams for this system have been proposed by [1], [2], and [3]. In addition, the phase relationships in the δ' Pu region have been studied by [4]. liquidus points for alloys containing between 15 and 30 at.% Fe have been determined by [5], and [6] has suggested minor modifications to the published diagrams in the Fe-rich region between Pu_2Fe and Fe. In general, the results of the various investigators differ only in details. [1] did not report the inverse peritectic decomposition of the ϵ Pu solid solution, as observed by [2] and confirmed by [4]. [2] placed the Pu-Pu₆Fe eutectic temperature at 413°C. rather than at 410°C. the eutectic temperature found by [1] and [3]. Also, [2] found somewhat higher solubilities of Fe in ϵ and δ Pu (> 2.5

and 1.5 at.%) than did [4] (> 2 and 0.6 at.%). The temperature of the peritectic formation of Pu₆Fe has been reported variously, as 430°C by [1], 428°C by [2], and 425°C by [7]. [6] has reported the existence of a hexagonal modification of PuFe₂ between 760 and 1020°C and has provided solubility data with respect to Pu in γ Fe and Fe in PuFe₂. The diagram shown in Fig. 13 is a composite based on the results of the various investigators. Crystal structure data have been reported by [1], [2], [6], [8], and [9] (see Table XV.)

- [1] S. T. Konobeevsky, "Equilibrium Diagrams of Certain Systems of Plutonium." Session on the Peaceful Uses of Atomic Energy, Section on Chemical Sciences, III: pp. 362-374. USSR Academy of Sciences, Moscow (1955).

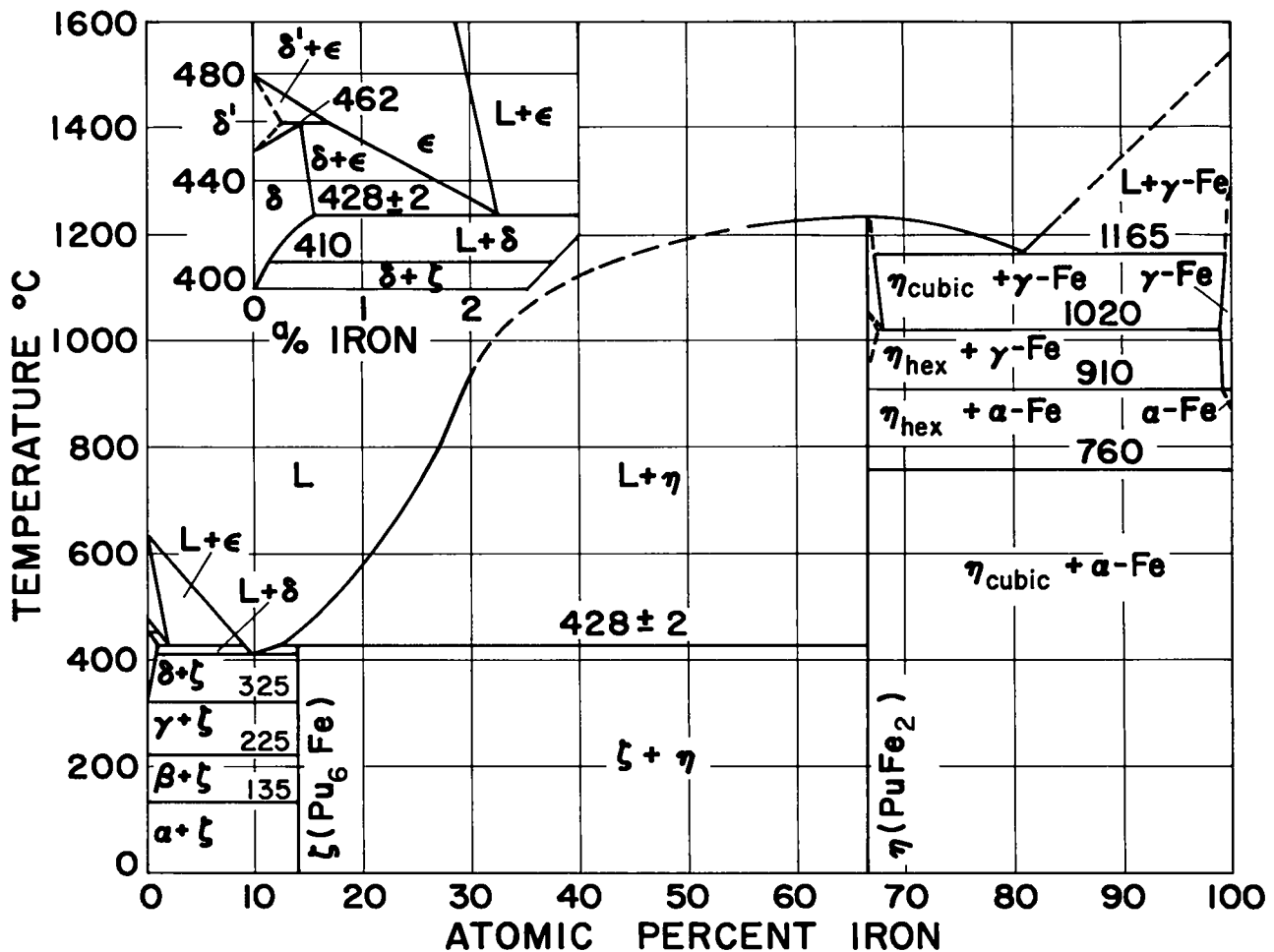


Fig. 13. Plutonium-iron phase diagram.

- [2] P. G. Mardon, H. R. Haines, J. H. Pearce, and M. B. Waldron, "The Plutonium-Iron System," *J. Inst. Metals*, **86**: 166-171 (1957).
- [3] F. W. Schonfeld, "Plutonium Phase Diagrams Studied at Los Alamos," Chap. XXII in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), p. 243, The University of Chicago Press, Chicago (1961).
- [4] R. O. Elliott and A. C. Larson, "Delta-Prime Plutonium," *Ibid.*, Chap. XXIV, pp. 265-280.
- [5] C. C. Land, in "Quarterly Status Report on Plutonium Reactor Fuel Development for Period Ending February 20, 1964," USAEC Report LAMS-3057, p. 13, Los Alamos Scientific Laboratory, (Mar. 25, 1964).
- [6] E. Avivi, "Études d'alliages plutonium-fer et d'alliages uranium-plutonium-fer" (in French), Centre d' Études Nucléaires de Fontenay-aux-Roses, French Report CEA-R2444 (Jan. 27, 1964).
- [7] W. N. Miner, Los Alamos Scientific Laboratory, unpublished work.
- [8] F. H. Ellinger, "A Review of the Intermetallic Compounds of Plutonium," Chap. XXV in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), pp. 292-294. The University of Chicago Press, Chicago (1961).
- [9] O. J. C. Runnalls, "The Crystal Structures of Some Intermetallic Compounds of Plutonium," *Can. J. Chem.*, **34**:133-145 (1956).

TABLE XV. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-IRON ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
Pu ₆ Fe ζ	U ₆ Mn	tetragonal	I4/mcm	10.404		5.355	4	17.07	[8]
				10.403		5.347		17.10	[1]
				10.405		5.349			[2]
PuFe ₂ η (cubic)	Cu ₂ Mg	cubic	Fd3m	7.150 (preparation "a")			8	12.74	[9]
				7.190 (preparation "b")				12.53	[9]
				7.191				12.53	[8]
				7.178				12.59	[1]
				7.189					[2]
				7.184 (Pu-rich)					[2]
			7.189				[6]		
PuFe ₂ η (hex)		hexagonal		5.64		18.37		[6]	

Plutonium-Gallium

Two versions of this phase diagram have been published. That of [1] is based on thermal, dilatometric, microstructural, and x-ray data. The diagram of [2] is supported by similar data and, in addition, by results of electron-beam microprobe analysis. Both diagrams are in satisfactory agreement for the most part although there are certain important differences. Information about alloys containing more than 75 at.% Ga has also been reported by [3], and additional crystal structure data have been given by [4], [5], and [6] (see Table XVI.) The diagram shown in Fig. 14 is mainly that of [1] as modified by the work of [3]. There is no significant solubility of Ga in α , β , and γ Pu, and the solubility of Ga in δ 'Pu is indicated to be less than 0.25 at.%. A filtration method was used by [1] to determine the solubility of Pu in liquid Ga. These data are given in Table XVII. The following are the main points of difference between the published work of [2] and the phase diagram shown here. According to [2], δ is formed from ϵ and Pu_3Ga_3 , whereas [1] found δ to be formed from ϵ and η . The maximum solubility of Ga in ϵ was found by [2] to be near 13 at.% instead of the 20 at.% solubility reported by [1]. η is shown by [2] to

decompose to form ϵ and Pu_3Ga , and by [1] to form δ and Pu_3Ga . Also, [2] found the 18-at.%-Ga alloy to be single phase (η) when annealed at 690°C, which is within the $\epsilon + \eta$ field of [1]. Pu_3Ga_3 forms peritectically according to [1] but congruently according to [2]. [2] suggest that Pu_3Ga_3 may have a high-temperature transition. The existence of Pu_2Ga_3 was not confirmed by [2]. PuGa_2 was found by [2] to have a range of solubility, in the region above 750°C, which reaches the maximum width of about 5 at.% at 1105°C.

- [1] F. H. Ellinger, C. C. Land, and V. O. Struebing, "The Plutonium-Gallium System." *J. Nucl. Mater.*, **12(2)**:226-236 (1964).
- [2] B. Hocheid, A. Tanon, S. Bedere, J. Despres, S. Hay, and F. Miard, "Studies of the Binary Systems Plutonium-Gold, Plutonium-Gallium and Plutonium-Indium." in *Plutonium 1965*, A. E. Kay and M. B. Waldron (Eds.), pp. 330-336. Chapman and Hall, London (1967).
- [3] C. C. Land, F. H. Ellinger, and K. A. Johnson, "The Intermetallic Compounds PuGa_4 and PuGa_6 ." *J. Nucl. Mater.*, **16(1)**: 87 (1965).

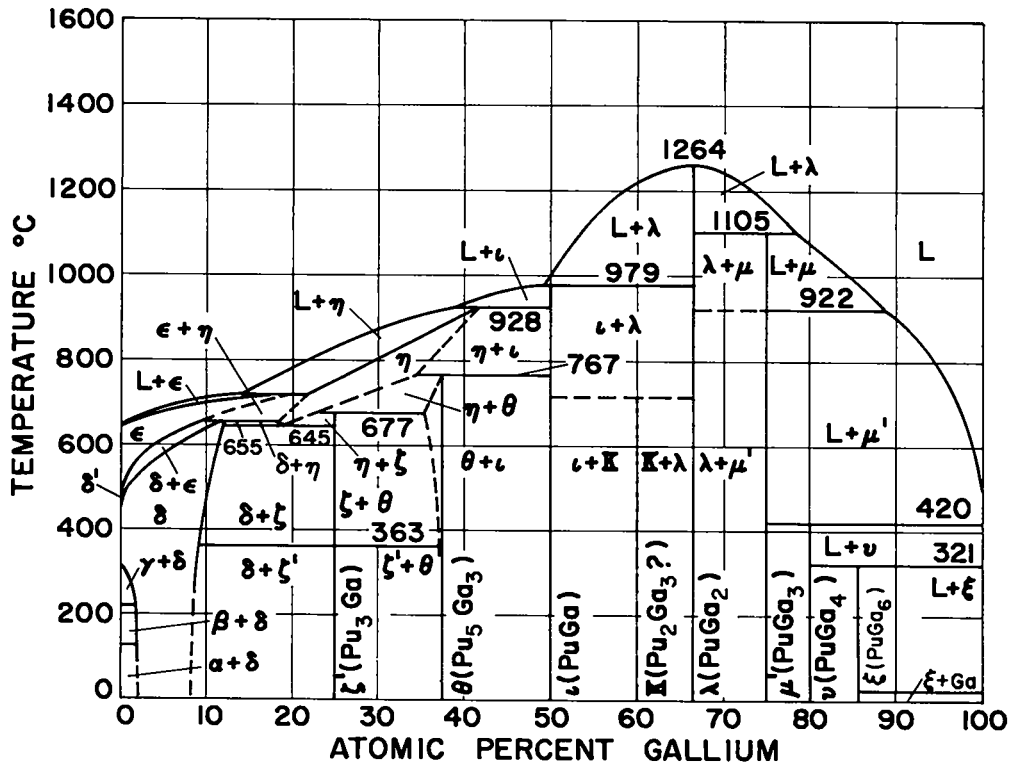


Fig. 14. Plutonium-gallium phase diagram.

[4] B. Hocheid, A. Tanon, and J. Despres, "Observations sur la structure des composés intermétalliques des systèmes binaires Pu-Ga et Pu-In" (in French). *J. Nucl. Mater.* **15(3)**: 241-244 (1965).

[5] A. C. Larson, D. T. Cromer, and R. B. Roof, Jr., "The Crystal Structure of the High-Temperature Form of PuGa₃." *Acta Cryst.*, **18(2)**: 294-295 (1965).

[6] F. H. Ellinger and W. H. Zachariasen. "The Crystal Structures of PuGa₄ and PuGa₆." *Acta Cryst.* **19**: 281-283 (1965).

TABLE XVII. SOLUBILITY OF PLUTONIUM IN LIQUID GALLIUM

Temperature ±5°C	Pu in solution at. %
496	0.24
500	0.17
597	1.01
706	3.2
811	6.4
907	10.6

TABLE XVI. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-GALLIUM ALLOYS

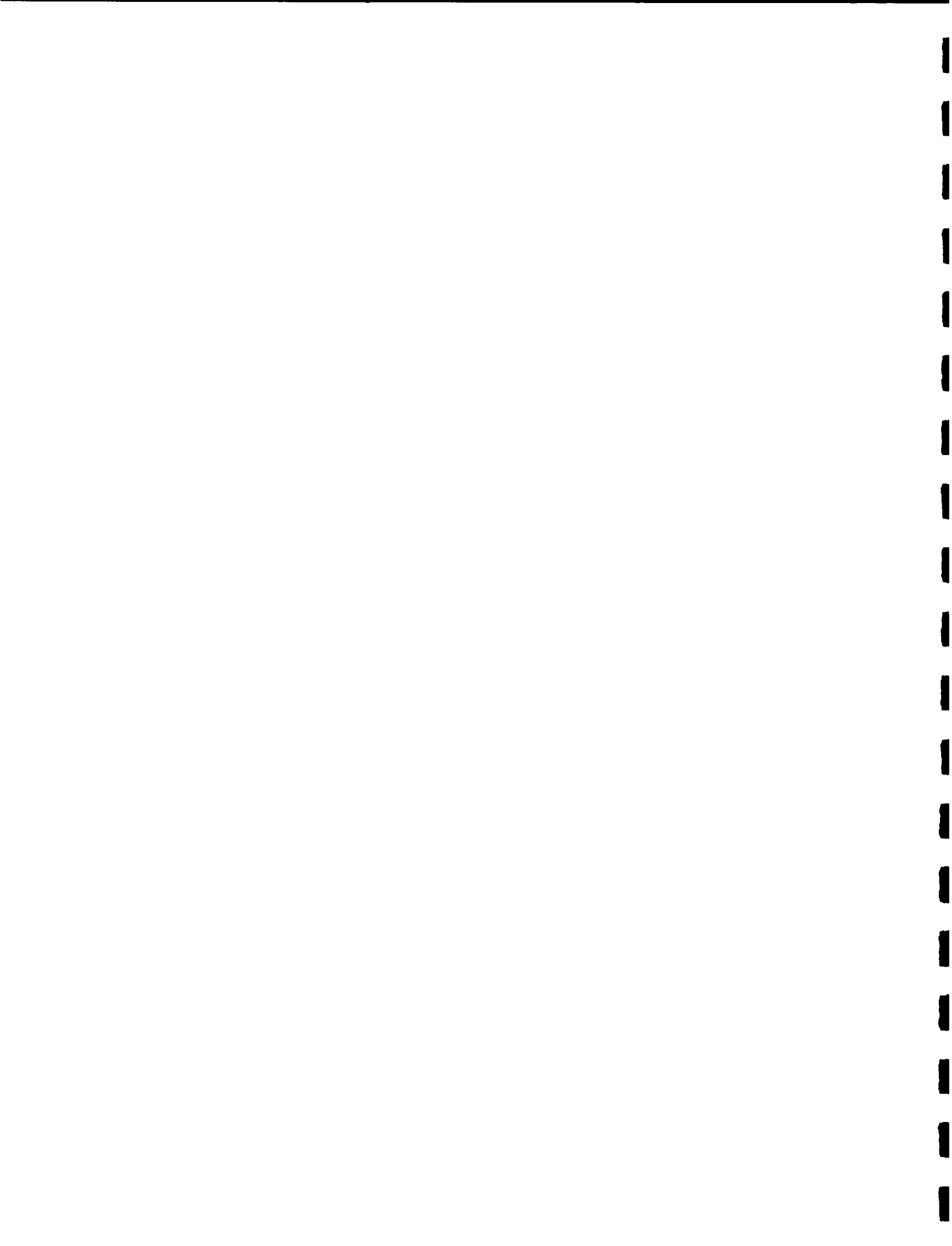
Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
Pu ₃ Ga ζ	AuCu ₃	cubic	Pm3m	4.507			1	14.27	[1]
				4.500					[4]
Pu ₃ Ga ζ'	SrPb ₃	tetragonal	P4/mmm	4.469		4.527	1	14.45	[1]
				4.492		4.555			[4]
η		body-centered pseudo-cell	cubic	3.58					[1]
Pu ₅ Ga ₃ θ	W ₅ Si ₃ ?	tetragonal face-centered cubic	I4/mcm	11.735		5.511	4	12.29	[1]
				5.570					[4]
PuGa ι		tetragonal body-centered cubic high temperature form	I	6.640		8.066	8	11.53	[1]
				3.53					2
Pu ₂ Ga ₃ (?) κ		unknown							[1]
PuGa ₂ λ	AlB ₂	hexagonal	P6 ₃ /mmm	4.248		4.120	1	9.76	[1]
				4.258		4.120			[4]
PuGa ₃ μ		rhombohedral (in hexagonal system)	R3m	10.001; α = 35° 59'		28.031	4	9.63	[5]
PuGa ₃ μ'	Ni ₃ Sn	hexagonal	P6 ₃ /mmc	6.300		4.514	2	9.59	[1]
PuGa ₄ ν	UAl ₄	body-centered orthorhombic	Imma	4.380	6.290	13.673	4	9.13	[3,6]
PuGa ₆ ξ	PuGa ₆	tetragonal	P4/nbm	5.942		7.617	2	8.11	[3,6]

Pu-Gd □
Plutonium-Gadolinium

The systems Pu-Gd through Pu-Lu, with the exception of Pu-Yb, have been found by [1] to display limited solution of the rare-earth element in δ Pu and ϵ Pu and more than 20 at.% solubility of Pu in each rare-earth element. [2], on the other hand, report as much as 15 at.% Gd soluble in δ Pu and ϵ Pu, and only about 10 at.% Pu soluble in α Gd. [1] have found an intermediate phase with the Sm-type structure in the Pu-Tb and Pu-Dy systems. It contains approximately 30 at.% Pu and is stable at elevated

temperatures only. There is also some indication that this intermediate phase will be found in other heavy rare-earth systems with Pu.

- [1] F. H. Ellinger, K. A. Johnson, and C. C. Land, Los Alamos Scientific Laboratory, unpublished work as of June 1967.
- [2] V. I. Kutaitsev, N. T. Chebotarev, I. G. Lebedev, M. A. Andrianov, V. N. Konev, and T. S. Menshikova, "Phase Diagrams of Plutonium with the Metals of Groups IIA, IVA, VIIIA and IB," in *Plutonium 1965*. A. E. Kay and M. B. Waldron (Eds.), pp. 420-449. Chapman and Hall, London (1967).



Pu-Ge □
Plutonium-Germanium

The results of a limited investigation of Pu-Ge alloys by microstructural and x-ray diffraction methods have been reported by [1] and [2]. There are five intermediate phases, Pu₃Ge, Pu₃Ge₂, Pu₂Ge₃, PuGe₂, and PuGe₃ (see Table XVIII), and a eutectic between PuGe₃ and Ge.

[1] F. H. Ellinger, "A Review of the Inter-

metallic Compounds of Plutonium," Chap. XXV in *The Metal Plutonium*. A. S. Coffinberry and W. N. Miner (Eds.), pp. 290-291. The University of Chicago Press, Chicago (1961).

[2] F. W. Schonfeld, "Plutonium Phase Diagrams Studied at Los Alamos," *Ibid.*, Chap. XXII, p. 250.

TABLE XVIII. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-GERMANIUM ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
Pu ₃ Ge	unknown								
Pu ₃ Ge ₂	unknown								
Pu ₂ Ge ₃	defect AlB ₂	hexagonal	P6/mmm	3.975		4.198	1	10.1	[1]
PuGe ₂	ThSi ₂	body-centered tetragonal	I4/amd	4.102		13.81	4	10.98	[1]
PuGe ₃	AuCu ₃	cubic	Pm3m	4.223			1	10.07	[1]



Pu-H □ **Plutonium-Hydrogen**

The pressure-temperature-composition relationships in this system have been determined by [1] and [2] and were used in constructing the phase diagram given in Fig. 15, which shows the phase equilibria at atmospheric pressure. Dissociation pressure vs temperature data for PuH_2 are also included in the diagram. It should be realized that the equilibria shown in the diagram cannot be attained unless the alloys are contained in a closed system to prevent dissociation of the hydride phases. Crystal structure data for PuH_2 and PuH_3 have been reported by [3] and are given in Table XIX.

- [1] R. N. R. Mulford and G. E. Sturdy. "The Plutonium-Hydrogen System. I. Plutonium Dihydride and Dideuteride." *J. Am. Chem. Soc.* **77**: 3449-3452 (1955), and "The Plutonium-Hydrogen System: II. Solid Solution of Hydrogen in Plutonium Dihydride." *J. Am. Chem. Soc.* **78**: 3897-3901 (1956).
- [2] R. N. R. Mulford, Los Alamos Scientific Laboratory, unpublished data.
- [3] F. H. Ellinger. "A Review of the Intermetallic Compounds of Plutonium." Chap. XXV in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), pp. 291-292, The University of Chicago Press, Chicago (1961).

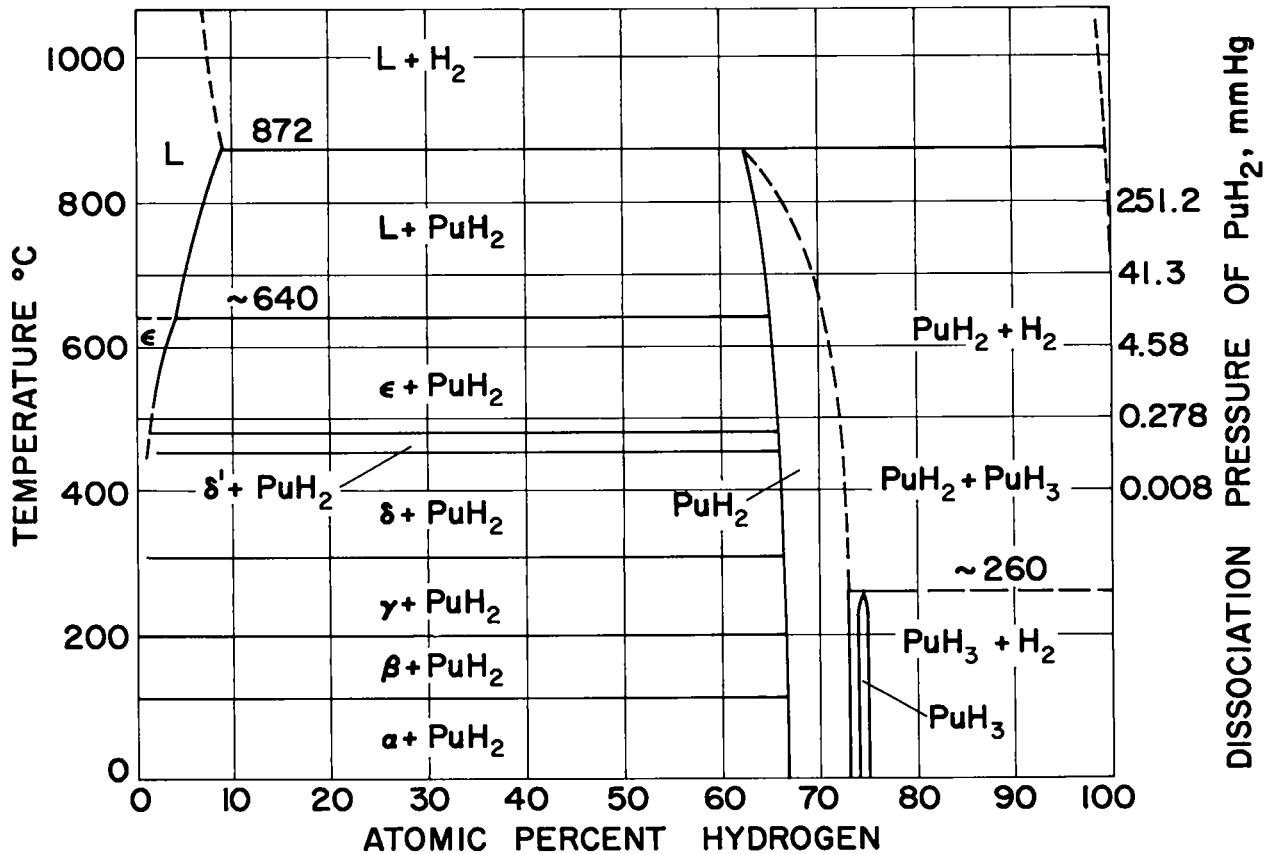


Fig. 15. Plutonium-hydrogen phase diagram. (Courtesy Ellinger, Land, and Gschneidner and by permission of Gordon and Breach, Science Publishers, Inc., from *Plutonium Handbook*, Vol. 1, p. 207, 1967).

TABLE XIX. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-HYDROGEN ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
PuH ₂	CaF ₂ (for PuH ₂)	Face-centered cubic	Fm3m	5.359(PuH ₂)			4	10.40	[3]
PuH _{2.75}	PuH ₂			5.34(PuH _{2.5})					
PuH ₃		Hexagonal	P6 ₃ /mmc	3.78	6.76		2	9.61	[3]

Plutonium-Hafnium

A complete phase diagram of this system has been published by [1], and is shown in Fig. 16. X-ray data have been reported by [2], and the microstructures of cold rolled and annealed (420°C) alloys containing 9 and 12 at.% Hf have been examined by [3]. The solubility of Hf in ϵ Pu ranges from about 11 at.% at 765°C to 5.5 at.% at 520°C, and in δ Pu decreases from about 7 at.% at 520°C to 3.5 at.% at 290°C, according to [1]. The microstructural work of [3] suggests a slightly higher solubility, about 10 at.% in δ Pu. [4] has found somewhat higher liquidus temperatures for the Pu-rich alloys (5 at.% Hf, 783°C; 10 at.% Hf, 814°C) than those shown by [1]. Also, [4]'s values for the $L + \alpha\text{Hf} \rightleftharpoons \epsilon$ reaction (749°C), the $\epsilon + \alpha\text{Hf} \rightleftharpoons \delta$ reaction (503°C) and the equilibrium temperature for the decomposition of θ (between 300° and 325°C) are all lower than the corresponding temperatures given by [1]. The solubility of Pu in Hf is given as about 10 at.% at 1150°C and less than 1 at.% at 500°C by [1], while [2] have suggested that the solid solubility of Pu in Hf may be as much as 5 at.%. δ Pu may be retained metastably to room temperature by quenching but transforms to α Pu when cold-

worked. There are two intermediate phases (see Table XX). ζ exists over a limited temperature range in alloy compositions between about 3 and 4.5 at.% Hf, and θ has a homogeneity range that extends from about 4 to 10 at.% Hf.

- [1] V. I. Kutaitsev, N. T. Chebotarev, I. G. Lebedev, M. A. Andrianov, V. N. Konev, and T. S. Menshikova. "Phase Diagrams of Plutonium with the Metals of Groups IIA, IVA, VIIIA and IB," in *Plutonium 1965*, A. E. Kay and M. B. Waldron (Eds.), pp. 424-425. 440. Chapman and Hall. London (1967).
- [2] M. B. Waldron, J. Garstone, J. A. Lee, P. G. Mardon, J. A. C. Marples, D. M. Poole, and G. K. Williamson, "The Physical Metallurgy of Plutonium," *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958*, Vol. 6, p. 166, United Nations. Geneva (1958).
- [3] S. E. Bronisz, Los Alamos Scientific Laboratory. unpublished work.
- [4] F. H. Ellinger. Los Alamos Scientific Laboratory. unpublished work.

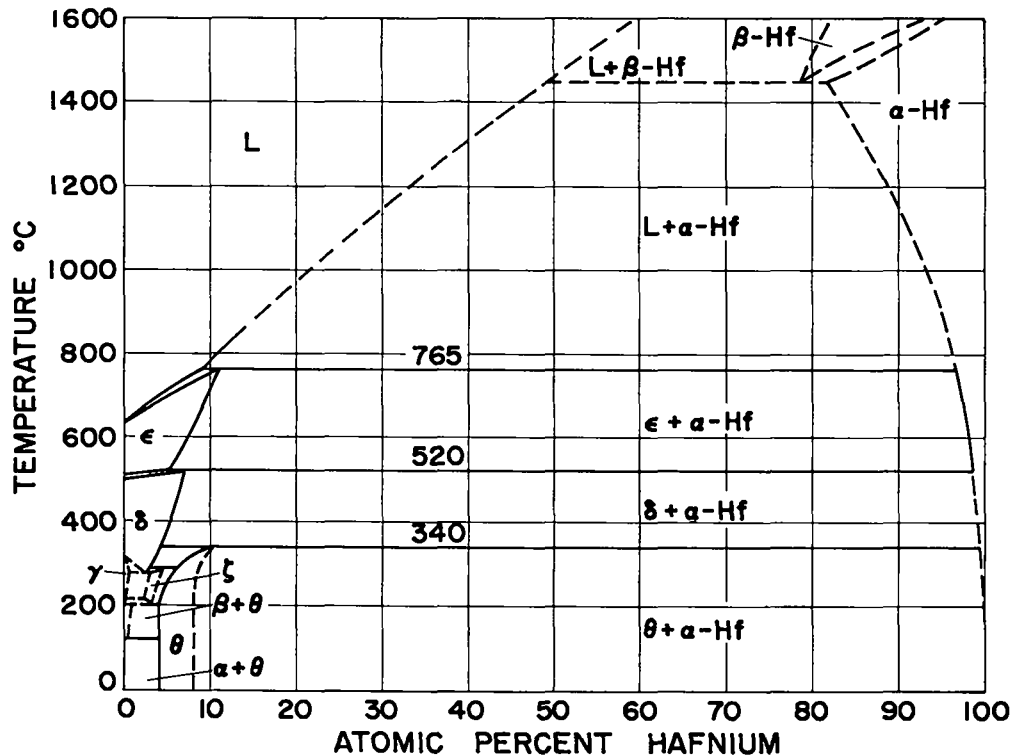


Fig. 16. Plutonium-hafnium phase diagram. (Redrawn from Ref. 1.)

TABLE XX. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-HAFNIUM ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
ζ	unknown								
θ		orthorhombic		10.415	10.428	11.245		17.7	[1]

Pu-Hg □ **Plutonium-Mercury**

X-ray data obtained by [1] and [2] have revealed the existence of two or possibly three Hg-rich compounds in this system (see Table XXI). The most Hg-rich compound was found by them to be PuHg_4 , isostructural with UHg_4 . Another compound appeared to be isostructural with hexagonal UHg_3 , but the identification is considered uncertain. In a subsequent study, [3] identified PuHg_4 more closely as $\text{Pu}_5\text{Hg}_{21}$ (γ -brass structure) and suggested also that UHg_4 is probably γ -phase U_5Hg_{21} . The solubility of Pu in Hg in the range 21 to 325°C has been determined by [4]. Their results are given in Table XXII. The partial phase diagram given in Fig. 17 is a slightly modified version of the one published by [5].

[1] F. H. Ellinger, "A Review of the Intermetallic Compounds of Plutonium," Chap.

XXV in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), p. 296, The University of Chicago Press, Chicago (1961).

[2] R. N. R. Mulford, Los Alamos Scientific Laboratory, unpublished work.

[3] A. F. Berndt, "A Gamma-phase in the Plutonium-Mercury System," *J. Less-Common Metals*, **11(3)**: 216-219 (1966).

[4] D. F. Bowersox and J. A. Leary, "The Solubility of Plutonium in Mercury," *J. Inorg. Nucl. Chem.*, **9**: 108-112 (1959).

[5] F. W. Schonfeld, "Phase Diagrams Studied at Los Alamos," Chap. XXII in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), p. 248, The University of Chicago Press, Chicago (1961).

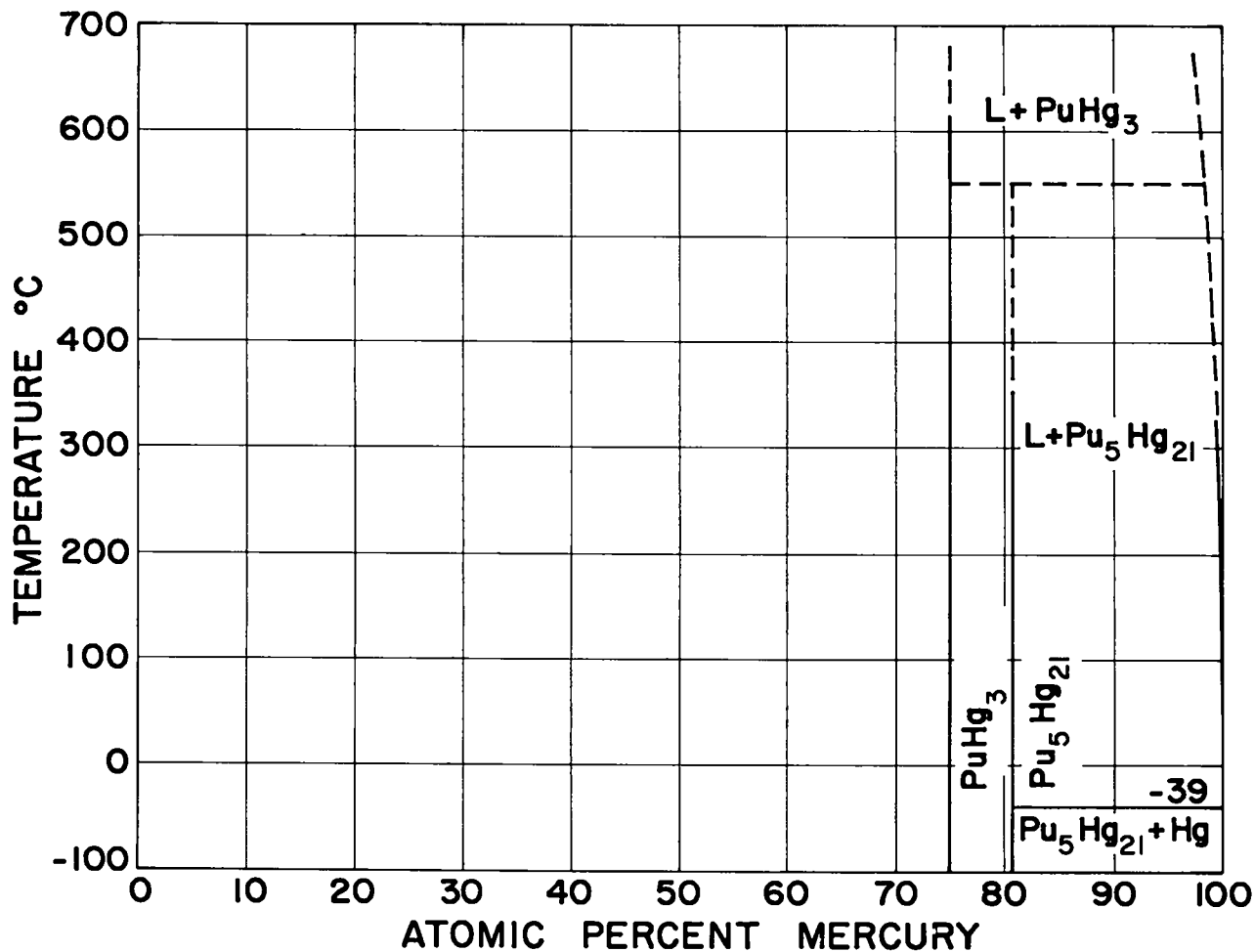


Fig. 17. Plutonium-mercury phase diagram.

TABLE XXI. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-MERCURY ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
PuHg ₃ (?)	Apparently isostructural with UHg ₃ , but a satisfactory diffraction pattern has not been obtained							[1]	
PuHg ₄		pseudo-cubic		3.61				[1]	
or Pu ₃ Hg ₂₁	γ-brass	distorted body-centered cubic	D8 ₁₋₃	21.78			16	13.90	[3]

TABLE XXII. SOLUBILITY OF PLUTONIUM IN MERCURY

Temperature. °C	at. % Pu
21	0.0131
24	0.0161
50	0.0255
100	0.0625
150	0.126
190	0.182
200	0.190
225	0.275
260	0.380
280	0.421
300	0.496
325	0.561

Pu-Ho **Plutonium-Holmium**

See Pu-Gd (Plutonium-Gadolinium).

Pu-In **Plutonium-Indium**

A complete phase diagram, based on thermal filtration, x-ray, and microstructural data, has been published by [1], and is given in Fig. 18. The region from 0 to 25 at.% In has been studied by [2], whose results concerning several details differ somewhat from those of [1], and the crystal structures of Pu_3In and PuIn_3 have been reported by [3] (see Table XXIII). The solubility of In in ϵ and δPu is limited to about 2 at.%, and is negligible in γ , β , and αPu . There appears to be little, if any, solid solubility of Pu in In. The work of [2] indicates that In additions between about 0.6 and 4 at.% stabilize δPu to room temperature. However, [1] have reported that, although δPu can be readily retained to room temperature, it is not thermodynamically stable below 300°C. They

show the eutectoid decomposition of δPu at about 300°C. The thermal data of [2] suggest to them the decomposition of Pu_3In at 678°C, but [1] give the peritectic reaction $L + \eta \rightleftharpoons \text{Pu}_3\text{In}$ at 962°C. The peritectic reaction $L + \text{Pu}_3\text{In} \rightleftharpoons \epsilon\text{Pu}$ is placed at 660°C (or 666°C) by [2] and at 676°C by [1], and, while [2] show the peritectoid formation of δPu from ϵPu and Pu_3In at 500°C, [1] show this reaction at 527°C.

- [1] F. H. Ellinger, C. C. Land, and K. A. Johnson, "The Plutonium-Indium System," *Trans. Met. Soc. AIME*, **233**: 1252-1258 (1965).
- [2] B. Hocheid, A. Tanon, S. Bedere, J. Despres, S. Hay, and F. Miard, "Studies of the Binary Systems Plutonium-Gold, Plutonium-Gallium and Plutonium-Indium." in *Plutonium*

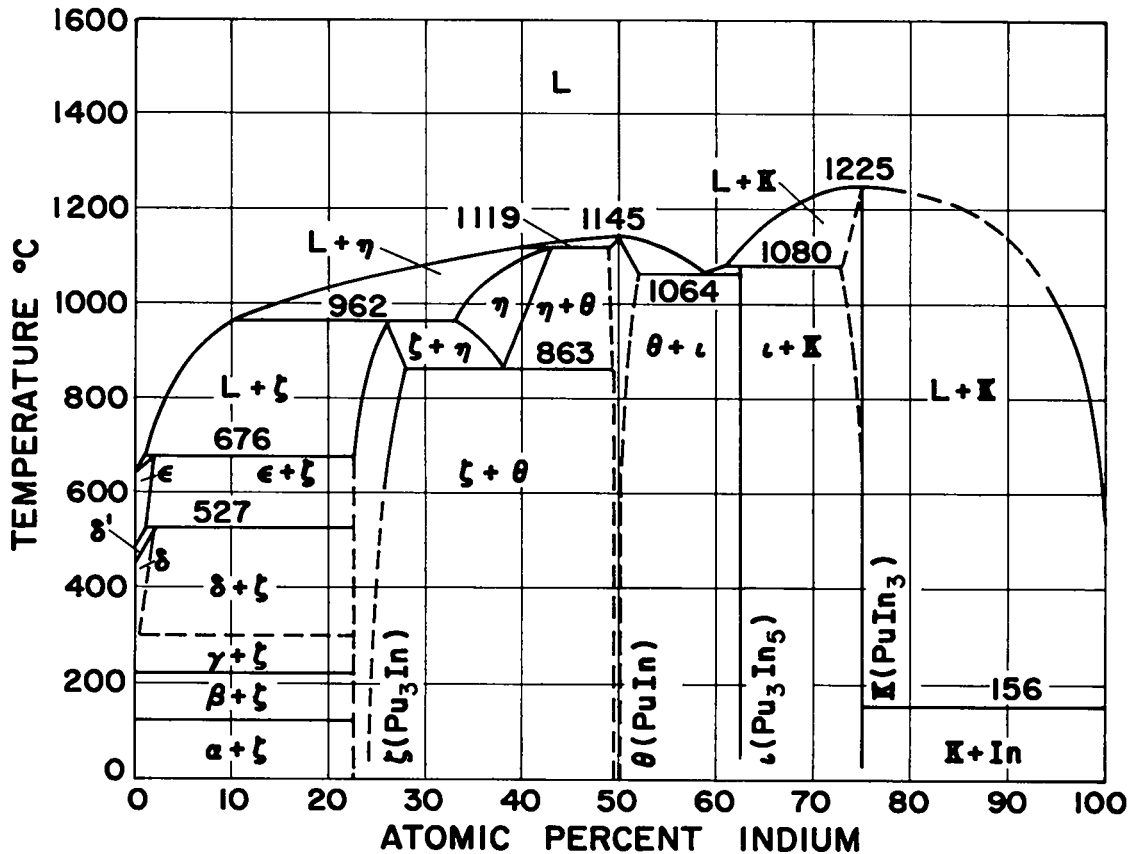


Fig. 18. Plutonium-indium phase diagram. (Redrawn from Ref. 1.)

1965, A. E. Kay and M. B. Waldron (Eds.). pp. 321, 336-339. Chapman and Hall. London (1967).

- [3] A. A. Bochvar, S. T. Konobeevsky, V. I. Kutaitsev, T. S. Menshikova, and N. T. Chebotarev. "Interaction Between Plutoni-

um and Other Metals in Connection with Their Arrangement in Mendeleev's Periodic Table," *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958*, Vol. 6, p. 186. United Nations. Geneva (1958).

TABLE XXIII. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-INDIUM ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
Pu ₃ In ξ	AuCu ₃	cubic	Pm3m	4.702			1	13.3	[3]
				4.705 (Pu-rich)				13.34	[1]
				4.722 (In-rich)				12.96	[1]
η	unknown								
PuIn θ	AuCu	tetragonal	P4/mmm	4.811		4.538	2	11.19	[1]
Pu ₃ In ₅ ι	unknown								
PuIn ₃ κ	AuCu ₃	cubic	Pm3m	4.607			1	9.9	[3]
				4.6096					[1]

Pu-Ir □

Plutonium-Iridium

X-ray examinations by [1] have revealed the existence of three intermetallic compounds in this system (see Table XXIV). PuIr₂ has been identified, but Pu₅Ir₄ and Pu₅Ir₃ are tentative determinations because their crystal structures have not been solved. However, Pu₅Ir₄ was found to be isostructural with Pu₅Rh₄, and Pu₅Ir₃ was found to be isostructural with Pu₅Rh₃ and Pu₅Os₃. There

appears to be little, if any, solubility of Ir in ε and δPu. The solubility of Pu in Ir is about 1 at.% at 1000°C.

- [1] V. I. Kutaitsev, N. T. Chebotarev, I. G. Lebedev, M. A. Andrianov, V. N. Konev, and T. S. Menshikova, "Phase Diagrams of Plutonium with the Metals of Groups IIA, IVA, VIIIA and IB," in *Plutonium 1965*, A. E. Kay and M. B. Waldron (Eds.), p. 428, Chapman and Hall, London (1967).

TABLE XXIV. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-IRIDIUM ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
Pu ₅ Ir ₃	unknown								
Pu ₅ Ir ₄	unknown								
PuIr ₂	Cu ₂ Mg	face-centered cubic	Fd3m	7.512-					[1]
				7.528					



Pu-K **Plutonium-Potassium**

See Pu-Cs (Plutonium-Cesium).

Pu-La **Plutonium-Lanthanum**

The phase diagram shown in Fig. 19 as determined by [1] is in agreement with the tentative results of [2]. The limits of the liquid miscibility gap have not yet been completely fixed. [1] give lattice parameter *vs.* composition data for the β La solid solution.

[1] F. H. Ellinger, C. C. Land, and K. A. John-

son, "The Plutonium-Lanthanum System." *Trans. Met. Soc. AIME*, **239(6)**: 895-898 (1967).

[2] D. M. Poole, M. G. Bale, P. G. Mardon, J. A. C. Marples, and J. L. Nichols, "Phase Diagrams of Some Plutonium Alloy Systems," in *Plutonium* 1960, E. Grison, W. B. H. Lord, and R. D. Fowler (Eds.), pp. 267-280, Cleaver-Hume Press, Ltd., London (1961).

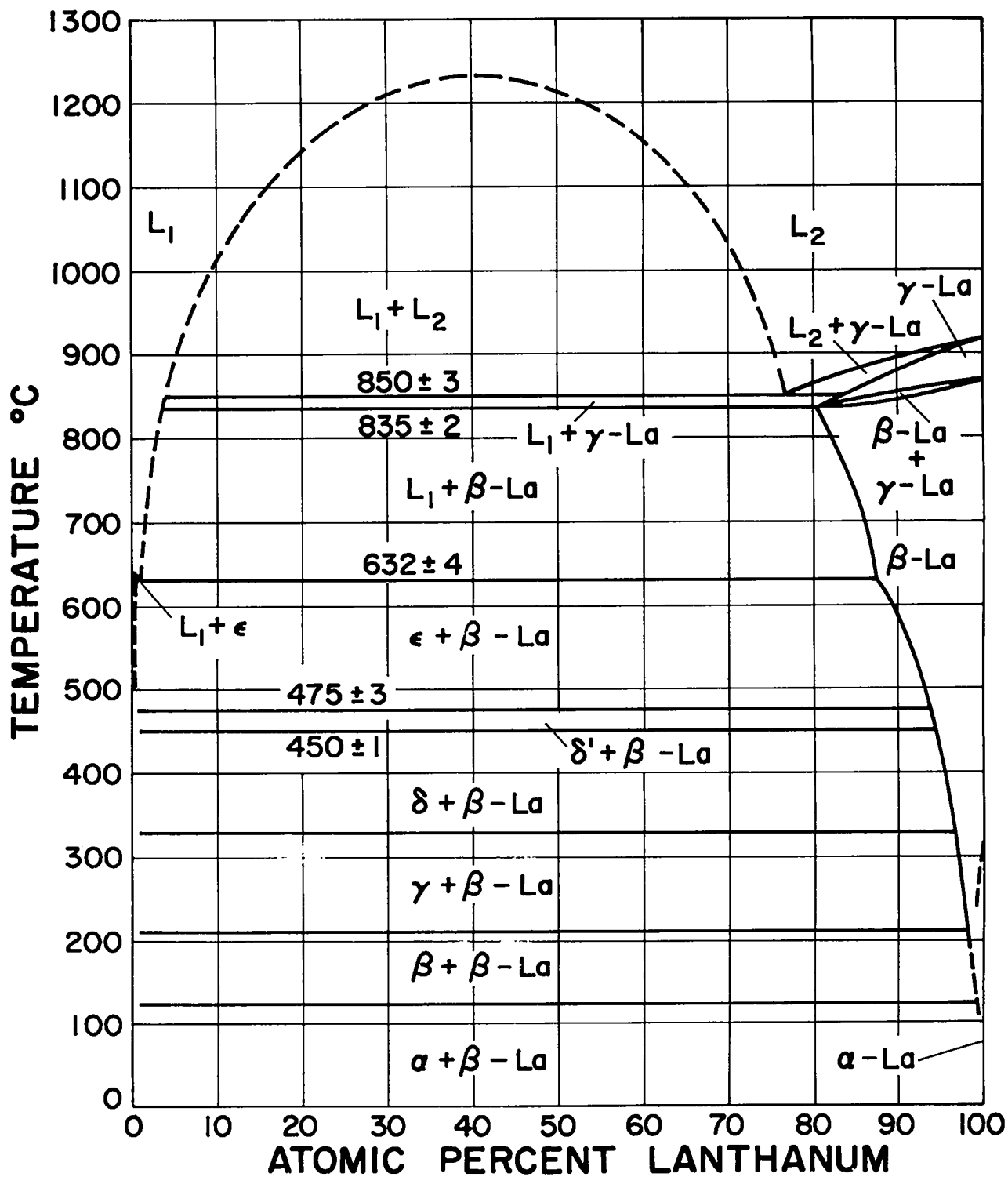


Fig. 19. Plutonium-lanthanum phase diagram.

Pu-Li **Plutonium-Lithium**

See Pu-Cs (Plutonium-Cesium).

Pu-Lu **Plutonium-Lutetium**

See Pu-Gd (Plutonium-Gadolinium).

Pu-Mg **Plutonium-Magnesium**

A phase diagram for this system, based on a limited amount of thermal, x-ray, and microstructural data, has been published by [1], and [2] have investigated the extent of the region of liquid immiscibility by a filtering technique. Also, diffusion studies by [3] and [4] have provided information about the solubility of Pu in Mg. The diagram shown in Fig. 20 takes into account the

combined results of these investigators. The maximum solubility of Pu in Mg appears to be of the order of 4 at.%. There seems to be no extensive solubility of Mg in Pu, although some solubility is indicated by the retention of δ and β Pu to room temperature. Crystal structure data in Table XXV for PuMg_2 and PuMg_{2+x} have been reported by [5].

[1] F. W. Schonfeld, "Plutonium Phase Diagrams Studied at Los Alamos," Chap. XXII

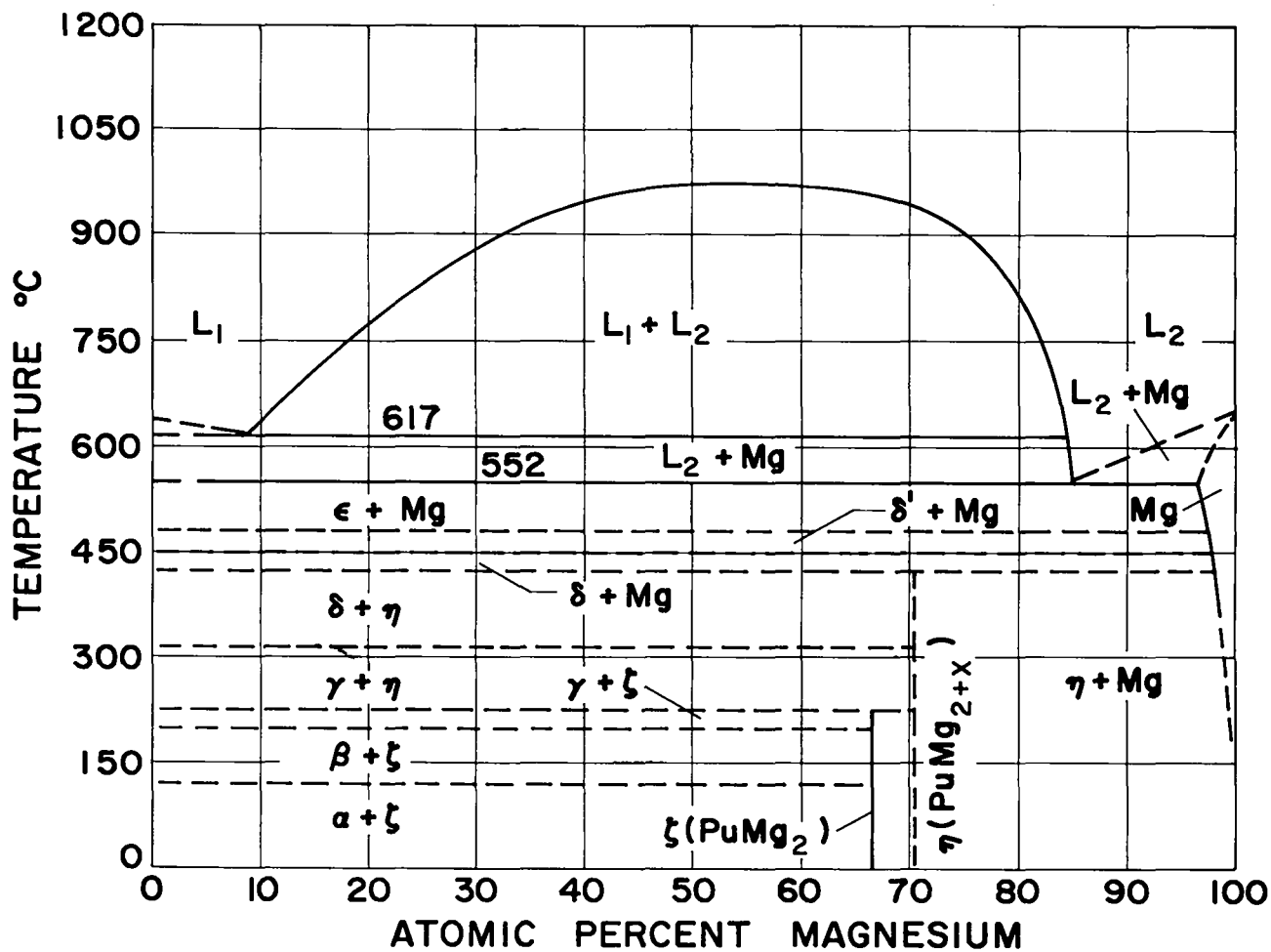


Fig. 20. Plutonium-magnesium phase diagram.

- in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), p. 244. The University of Chicago Press. Chicago (1961).
- [2] J. D. Schilb and R. K. Steunenberg, "Liquid Immiscibility in the System Pu-Mg," in "Argonne National Laboratory Chemical Engineering Semiannual Report. July-December, 1964." USAEC Report ANL-6925. pp. 48-49 (May 1965).
- [3] D. J. Hodkin and P. G. Mardon. "The Diffusion of Plutonium in Magnesium and Magnesium Alloys." *J. Nucl. Mater.*, **16**: 271-289 (1965).
- [4] D. Calais, M. Dupuy, M. Mouchnino, A. Y. Portnoff, and A. Van Craeynest. "Diffusion of Plutonium in the Solid State." in *Plutonium 1965*, A. E. Kay and M. B. Waldron (Eds.), pp. 359-366. 371-372. Chapman and Hall, London (1967).
- [5] F. H. Ellinger. "A Review of the Inter-metallic Compounds of Plutonium," Chap. XXV in *The Metal Plutonium*. A. S. Coffinberry and W. N. Miner (Eds.), p. 295. The University of Chicago Press. Chicago (1961).

TABLE XXV. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-MAGNESIUM ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
PuMg ₂ ζ	CaF ₂	face-centered cubic	Fm3m	7.34			4	4.83	[5]
PuMg _{2+x} η		hexagonal		13.8		9.7			[5]

Pu-Mn
Plutonium-Manganese

A complete phase diagram for this system has been published by [1], and some Pu-rich alloys have been studied by [2] and [3] (see Fig. 21). The latter work has placed the ϵ Pu/PuMn₂ eutectic at 525°C instead of 510°C as found by [1]. Also, the eutectoid temperature was found to be 446°C by [4], whereas [1] found it to be 430°C. ϵ Pu will dissolve a maximum of about 4 at.% Mn. The solubility of Mn in the other Pu allotropes is quite limited. The solubility of Pu in the Mn allotropes has not been determined, but [1] noted that the lattice parameter of β Mn is expanded from 6.293 to 6.317 Å by the solution of Pu. [1], [5], and [6] have reported crystal structure data for PuMn₂ (see Table XXVI). Solubility data determined by [7] for Mn in liquid Pu are given in Table XXVII.

- [1] S. T. Konobeevsky, "Equilibrium Diagrams of Certain Systems of Plutonium," Session on the Peaceful Uses of Atomic Energy, Section on Chemical Sciences, III: pp. 362-374, USSR Academy of Sciences, Moscow (1955).
- [2] W. N. Miner, Los Alamos Scientific Laboratory, unpublished work.
- [3] E. M. Cramer, Los Alamos Scientific Laboratory, unpublished work.
- [4] R. O. Elliott and A. C. Larson, "Delta-Prime Plutonium," Chap. XXIV in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), pp. 265-280, The University of Chicago Press, Chicago (1961).

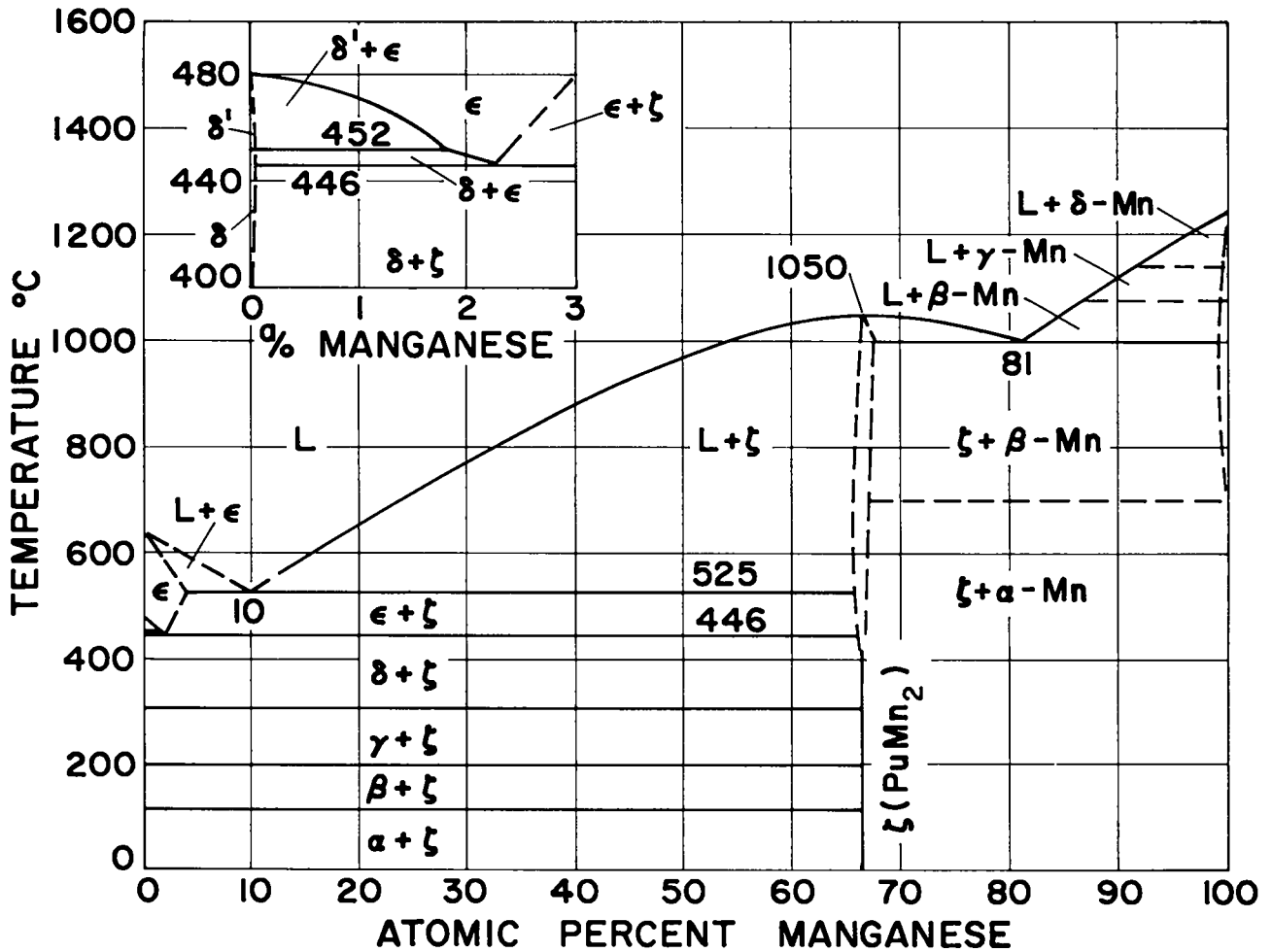


Fig. 21. Plutonium-manganese phase diagram. (Courtesy Ellinger, Land, and Gschneidner and by permission of Gordon and Breach, Science Publishers, Inc., from *Plutonium Handbook*, Vol. 1, p. 213, 1967.)

- [5] O. J. C. Runnalls, "The Crystal Structures of Some Intermetallic Compounds of Plutonium," *Can. J. Chem.*, **34**: 133-145 (1956).
- [6] F. H. Ellinger, "A Review of the Intermetallic Compounds of Plutonium," Chap. XXV in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), pp. 295-296. The University of Chicago Press. Chicago (1961).
- [7] D. F. Bowersox, Los Alamos Scientific Laboratory, to be published.

TABLE XXVI. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-MANGANESE ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
PuMn ₂	Cu ₂ Mg	face-centered cubic	Fd3m	7.290 (Pu-rich)			8	11.96	[5]
				7.292 (Pu-rich)					[6]
				7.29 (Pu-rich)					[1]
				7.26 (Mn-rich)					[1]

TABLE XXVII. SOLUBILITY OF MANGANESE IN LIQUID PLUTONIUM

Temperature, °C	Solubility, at. % Mn
850	30.8 ± 0.3
950	45.7 ± 1.3
1000	53.6 ± 2.2

Pu-Mo
Plutonium-Molybdenum

A phase diagram of the simple eutectic type for this system (see Fig. 22) has been published by [1]. The eutectic composition is close to Pu, and the eutectic temperature is given as 590°C. However, [2] found the eutectic temperature to be 613°C. The absence of intermediate phases in as-cast alloys has been verified by [3] and [4], and a liquidus point at 26 at.% Mo and 1750°C was found by [3] and [5] who equilibrated liquid Pu in the presence of excess Mo and then measured, by areal analysis, the proportion of primary dendrites of Mo in the alloy. The solubility of Mo in liquid Pu between 700 and 1000°C, as determined by [6], is given in Table XXVIII.

[1] A. A. Bochvar, S. T. Konobeevsky, V. I. Kutaitsev, T. S. Menshikova, and N. T. Chebotarev, "Interaction Between Plutonium

and Other Metals in Connection with Their Arrangement in Mendeleev's Periodic Table," *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958*, Vol. 6, p. 191, United Nations, Geneva (1958).

[2] P. G. Mardon, J. P. Evans, D. J. Hodkin, J. M. North, and J. H. Pearce, "The Constitution and Fabrication of Uranium-Molybdenum-Plutonium Fuels," in *Plutonium 1960*, E. Grison, W. B. H. Lord, and R. D. Fowler (Eds.), pp. 329-352, Cleaver-Hume Press, Ltd., London (1961).

[3] E. M. Cramer, Los Alamos Scientific Laboratory, unpublished work.

[4] F. H. Ellinger, Los Alamos Scientific Laboratory, unpublished work.

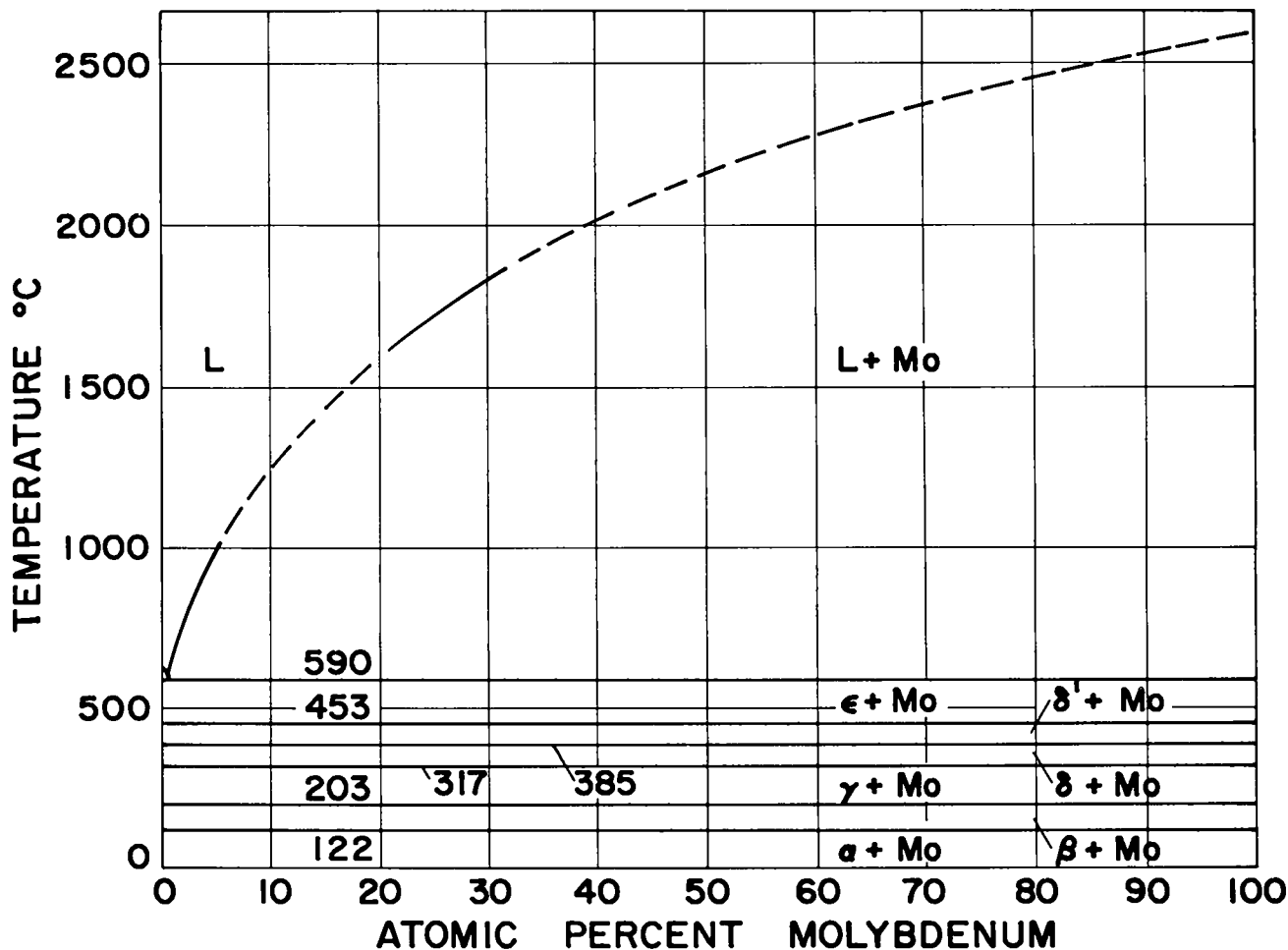


Fig. 22. Plutonium-molybdenum phase diagram. (Redrawn from Ref. 1.)

- [5] V. O. Struebing, Los Alamos Scientific Laboratory, unpublished work.
- [6] D. F. Bowersox and J. A. Leary, "The Solubilities of Selected Elements in Liquid Plutonium: VIII. Molybdenum," USAEC Report LA-3523, Los Alamos Scientific Laboratory (May 24, 1966).

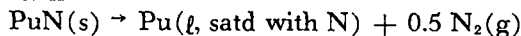
TABLE XXVIII. SOLUBILITY OF MOLYBDENUM IN LIQUID PLUTONIUM

Temperature, °C	Solubility, at. % Mo
700	1.56 ± 0.04
750	2.08 ± 0.04
800	2.75 ± 0.06
850	3.06 ± 0.02
900	3.68 ± 0.10
950	4.53 ± 0.04
1000	5.00 ± 0.12

Pu-N □

Plutonium-Nitrogen

One compound, PuN, is known in this system. It was first identified by [1]. PuN formed by heating Pu and PuH₂ in N₂ has been examined by [2], and that formed by melting Pu in N₂ has been studied by [3] and [4]. According to [4], congruent melting of PuN occurs only under pressures in excess of 24.5 atm, the upper limit that could be attained in their apparatus. At lower pressures, PuN decomposes according to the reaction



The decomposition pressures observed by [4] range from about 0.01 atm at 2300°C to 24.5 atm at 2770°C. The apparent melting point of PuN under 1 atm of N₂ has been reported as 2750 ± 75°C by [5], but the corresponding value determined by [4] is 2584 ± 30°C. Crystal structure data for PuN are given in Table XXIX.

[1] W. H. Zachariasen, "Crystal Chemical

Studies of the 5-f Series of Elements: XII. New Compounds Representing Known Structure Types," *Acta Cryst.*, **2**: 388-390 (1949).

[2] F. Brown, H. M. Ockenden, and G. A. Welch, "The Preparation and Properties of Some Plutonium Compounds: Part II. Plutonium Nitride," *J. Chem. Soc.*, 1955 (Part IV): 4196-4201 (1955).

[3] F. H. Ellinger, Los Alamos Scientific Laboratory, unpublished work.

[4] W. M. Olson and R. N. R. Mulford, "The Decomposition Pressure of Plutonium Nitride," *J. Phys. Chem.*, **68**: 1048-1051 (1964).

[5] D. F. Carroll, "Synthesis and Properties of Plutonium Mononitride," USAEC Report HW-SA-2755, Hanford Atomic Products Operation (1962).

TABLE XXIX. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-NITROGEN ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
PuN	NaCl	face-centered cubic	Fm3m	4.9075			4	14.2	[3]
				4.905					[1]
				4.905					[2]
				4.9055					[4]



Pu-Na □ **Plutonium-Sodium**

See Pu-Cs (Plutonium-Cesium).

Pu-Nb □ **Plutonium-Niobium**

According to [1], Pu and Nb do not form intermediate phases (see Fig. 23). This has been confirmed by the microstructural work of [2] and the x-ray examinations of [3] on alloys containing 10 and 50 at.% Nb. The solubility of Nb in liquid Pu at about 2000°C was found to be about 60 at.% Nb by [2] and [4], and for the temperature range 700 to 950°C the values as determined by [5] are given in Table XXX.

Chebotaev, "Interaction Between Plutonium and Other Metals in Connection with Their Arrangement in Mendeleev's Periodic Table," *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958, Vol. 6, p. 191, United Nations. Geneva (1958).*

[1] A. A. Bochvar, S. T. Konobeevsky, V. I. Kutaitsev, T. S. Menshikova, and N. T.

[2] E. M. Cramer, Los Alamos Scientific Laboratory, unpublished work.

[3] F. H. Ellinger, Los Alamos Scientific Laboratory, unpublished work.

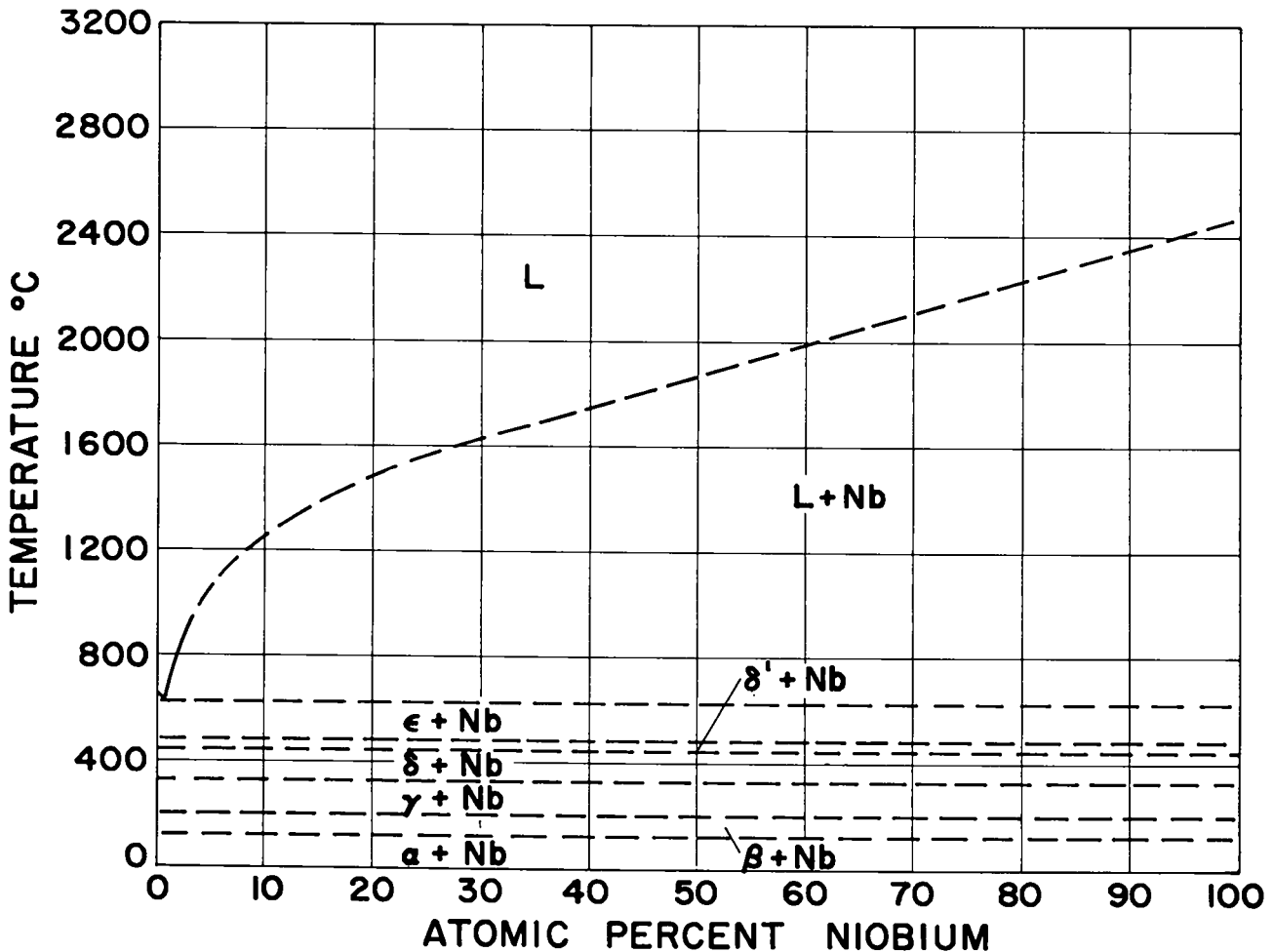


Fig. 23. Plutonium-niobium phase diagram.

[4] V. O. Struebing, Los Alamos Scientific Laboratory, unpublished work.

[5] D. F. Bowersox and J. A. Leary, "The Solubilities of Refractory Elements in Liquid Plutonium," *Am. Nucl. Soc. Trans.*, **9(1)**: 7 (1966); also **10(1)**: 106 (1967).

TABLE XXX. SOLUBILITY OF NIOBIUM IN LIQUID PLUTONIUM

Temperature, °C	Average solubility, at. % Nb
700	1.10
750	1.44
800	1.69
850	2.05
900	2.68
950	3.14
1000	3.71

Pu-Nd **Plutonium-Neodymium**

The phase diagram shown in Fig. 24 has been constructed by [1] on the basis of thermal, micrographic, and x-ray diffraction data. The interpretation of the data was straightforward; there appear to be no major uncertainties in the diagram. [2] have published a partial preliminary Pu-Nd diagram that shows about 8 at.% Nd soluble in δ Pu and in ϵ Pu. It also shows a β Nd phase (fcc) field extending down to about 200°C.

- [1] F. H. Ellinger, C. C. Land, and K. A. Johnson, unpublished work as of June 1967.
- [2] V. I. Kutaitsev, N. T. Chebotarev, I. G. Lebedev, M. A. Andrianov, V. N. Konev, and T. S. Menshikova, "Phase Diagrams of Plutonium with the Metals of Groups IIA, IVA, VIIIA and IB," in *Plutonium 1965*, A. E. Kay and M. B. Waldron (Eds.), pp. 420-449, Chapman and Hall, London (1967).

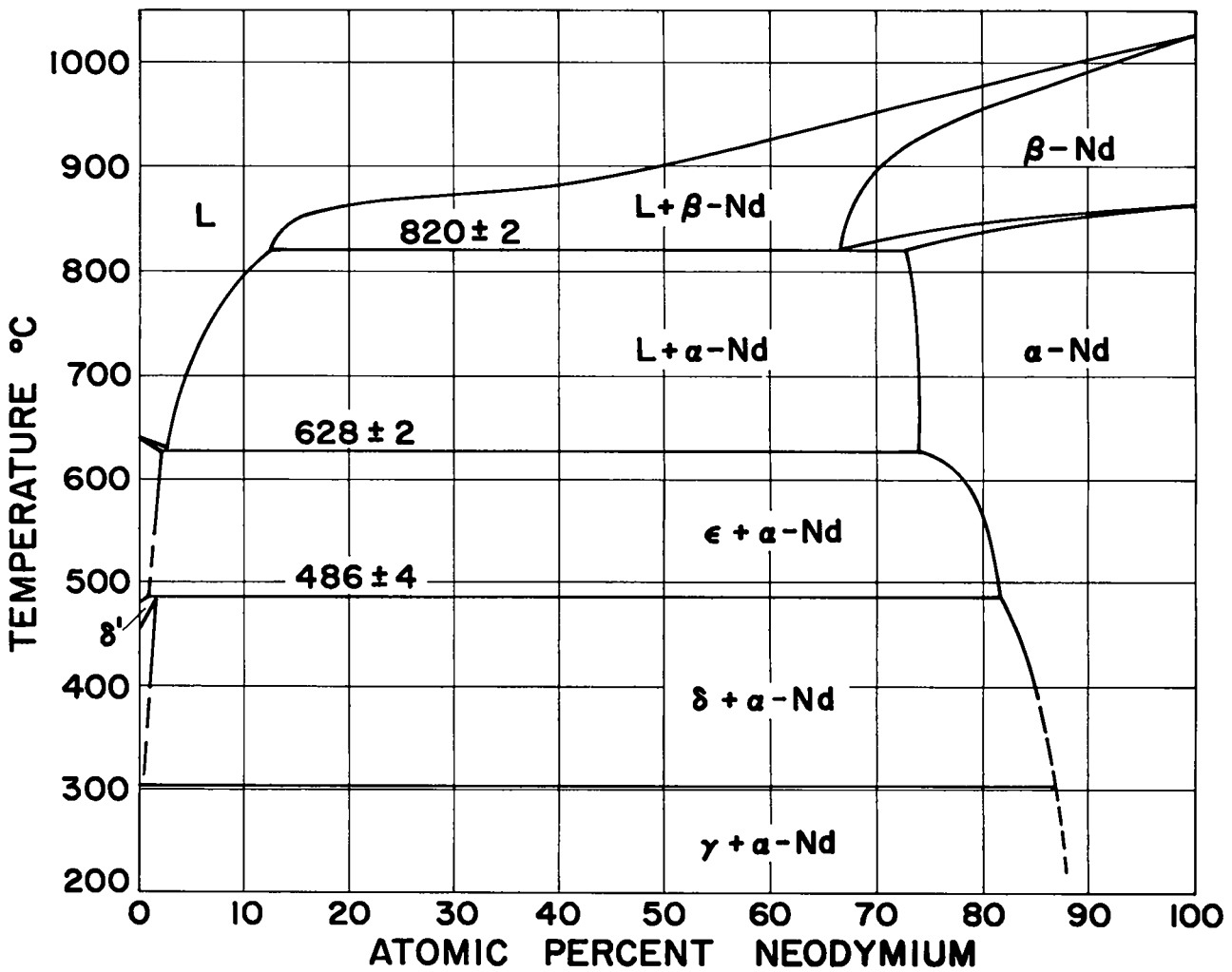


Fig. 24. Plutonium-neodymium phase diagram.



Complete phase diagrams for this system have been published by [1] and [2], and the effect of Ni additions on the phase equilibria associated with δ' Pu has been reported by [3]. The diagram of [2] is in general agreement with that of [1], except that it does not include the intermediate phase $\text{Pu}_2\text{Ni}_{17}$. The diagram given in Fig. 25 is based on the work of [1] and [3]. There are two eutectics: one at 12.5 at.% Ni and 465°C, the other at 92 at.% Ni and 1210°C. ϵ Pu dissolves a maximum of 4.3 at.% Ni at 450°C. The solubility of Ni in the other Pu allotropes is very restricted. The maximum solubility of Pu in Ni is about 1.8 at.% Pu at 1210°C. Crystal structure data have been determined for all six of the intermediate phases and are given in Table XXXI.

[1] G. W. Wensch and D. D. Whyte, "The Nickel-Plutonium System," USAEC Report

[2] S. T. Konobeevsky, "Equilibrium Diagrams of Certain Systems of Plutonium," Session on the Peaceful Uses of Atomic Energy, Section on Chemical Sciences, III: pp. 362-374, USSR Academy of Sciences, Moscow, 1955.

[3] R. O. Elliott and A. C. Larson, "Delta-Prime Plutonium," Chap XXIV in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), pp. 265-280, The University of Chicago Press, Chicago (1961).

[4] D. T. Cromer and R. B. Roof, Jr., "The Crystal Structure of PuNi." *Acta Cryst.*, **12**: 942-943 (1959).

[5] O. J. C. Runnalls, "The Crystal Structures of Some Intermetallic Compounds of Plu-

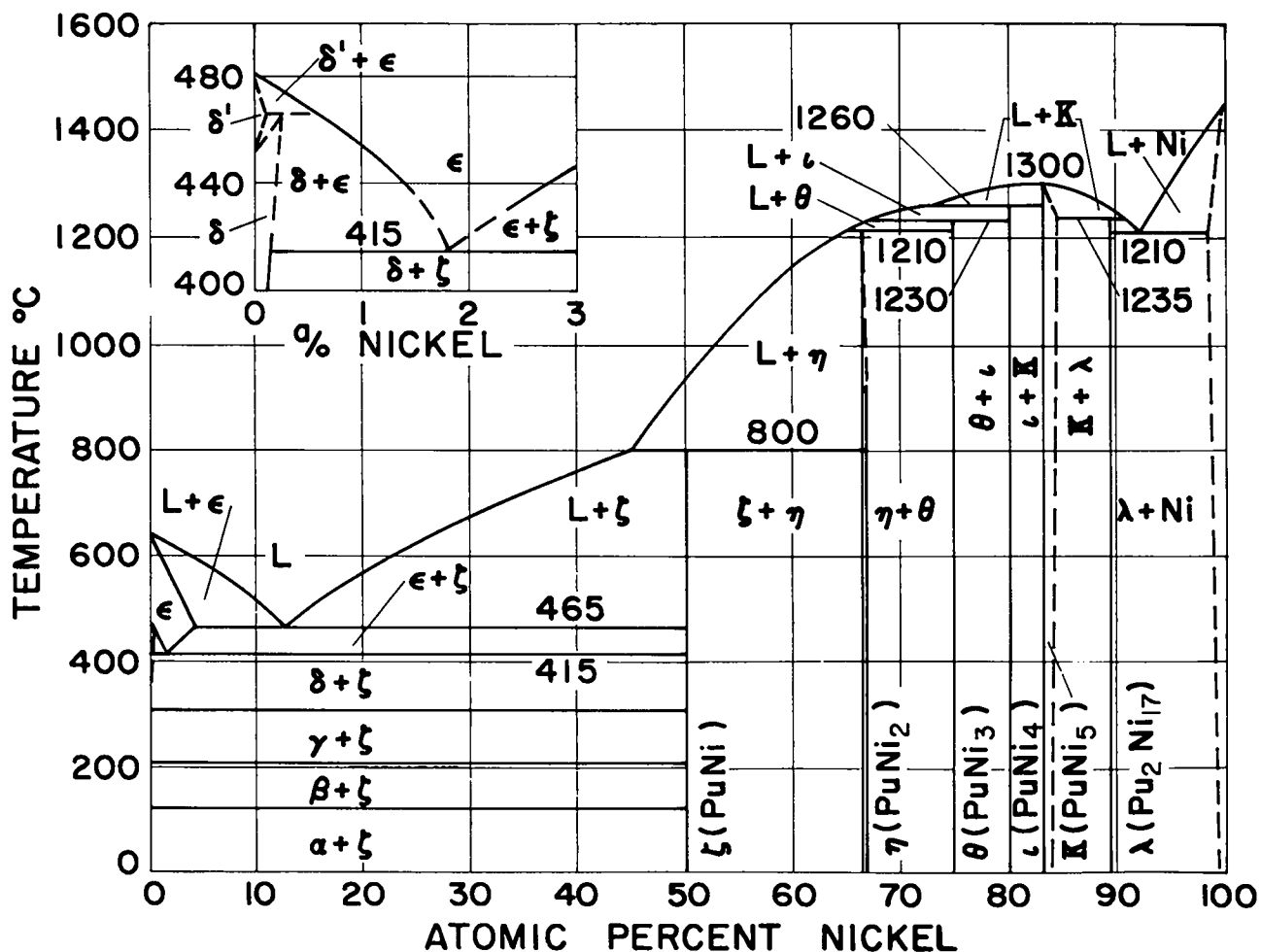


Fig. 25. Plutonium-nickel phase diagram. (Courtesy Ellinger, Land, and Gschneidner and by permission of Gordon and Breach, Science Publishers, Inc., from *Plutonium Handbook*, Vol. 1, p. 216, 1967.)

- tonium," *Can. J. Chem.*, **34**: 133-145 [7] D. T. Cromer and C. E. Olsen, "The Crystal Structures of PuNi₃ and CeNi₃," *Acta Cryst.*, **12**: 689-694 (1960).
- [6] F. H. Ellinger, "A Review of the Inter-metallic Compounds of Plutonium," Chap. XXV in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), pp. 296-297. The University of Chicago Press, Chicago (1961).
- [8] D. T. Cromer and A. C. Larson, "The Crystal Structure of PuNi₄," *Acta Cryst.*, **13**: 909-912 (1960).

TABLE XXXI. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-NICKEL ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
PuNi ξ	TII	orthorhombic	Cmcm	3.59	10.21	4.22	4	12.9	[4]
PuNi ₂ η	Cu ₂ Mg	face-centered cubic	Fd3m	7.16 7.141 (Pu-rich) 7.115 (Ni-rich) 7.14			8	13.1	[5] [6] [2] [2]
PuNi ₃ θ	PuNi ₃	rhombohedral	R3m	8.615; α = 33°44'			3	11.8	[7]
PuNi ₄ ι	PuNi ₄	monoclinic	C2/m	4.87	8.46	10.27	6	11.3	[8]
PuNi ₅ κ	CaZn ₅	hexagonal	P6/mmm	4.875		3.970	1	10.8	[5]
				4.872 (Pu-rich)		3.980			[6]
				4.861 (Ni-rich)		3.982			[6]
Pu ₂ Ni ₁₇ λ	Th ₂ Ni ₁₇	hexagonal	P6 ₃ /mmc	8.30		8.00	2	10.3	[5]
				8.29		8.01			[6]

Pu-Np □ **Plutonium-Neptunium**

This system has been investigated by [1], who used thermal, dilatometric, and x-ray techniques, and by [2], who studied alloys in the composition range from 0 to 25 at.% Np by dilatometry and thermal analysis. The results of both groups of investigators are essentially in agreement, except that the extensive solubility of Np in β Pu was not clearly revealed in the work of [2]. The diagram shown in Fig. 26 is that of [1]. Np is highly soluble in α and β Pu, the maximum amounts being, respectively, 96 at.% at 275°C and about 85 at.% at 540°C, and ϵ Pu is completely miscible with γ Np. The maximum solubilities of Np in γ , δ , and δ' Pu are limited to about 2 at.%. β Np will dissolve about 10 at.% Pu at 540°C and about 2 at.% at 280°C. α Np will

dissolve about 20 at.% Pu at room temperature, the solubility decreasing with increasing temperature to 0 at.% at the α Np \rightleftharpoons β Np transition temperature at about 285°C. The single intermediate phase, η , (see Table XXXII) has a composition range from 3 to 50 at.% Np.

- [1] P. G. Mardon, J. H. Pearce, and J. A. C. Marples, "Constitution Studies of the Neptunium-Plutonium Alloy System," *J. Less-Common Metals*, **3**: 281-292 (1961).
- [2] R. G. Cope, D. G. Hughes, R. G. Loasby, and D. C. Miller, "The Plutonium-Ruthenium and Plutonium-Neptunium Binary Phase Diagrams," in *Plutonium 1960*, E. Grison, W. B. H. Lord, and R. D. Fowler (Eds.), pp. 280-289, Cleaver-Hume Press, Ltd., London (1961).

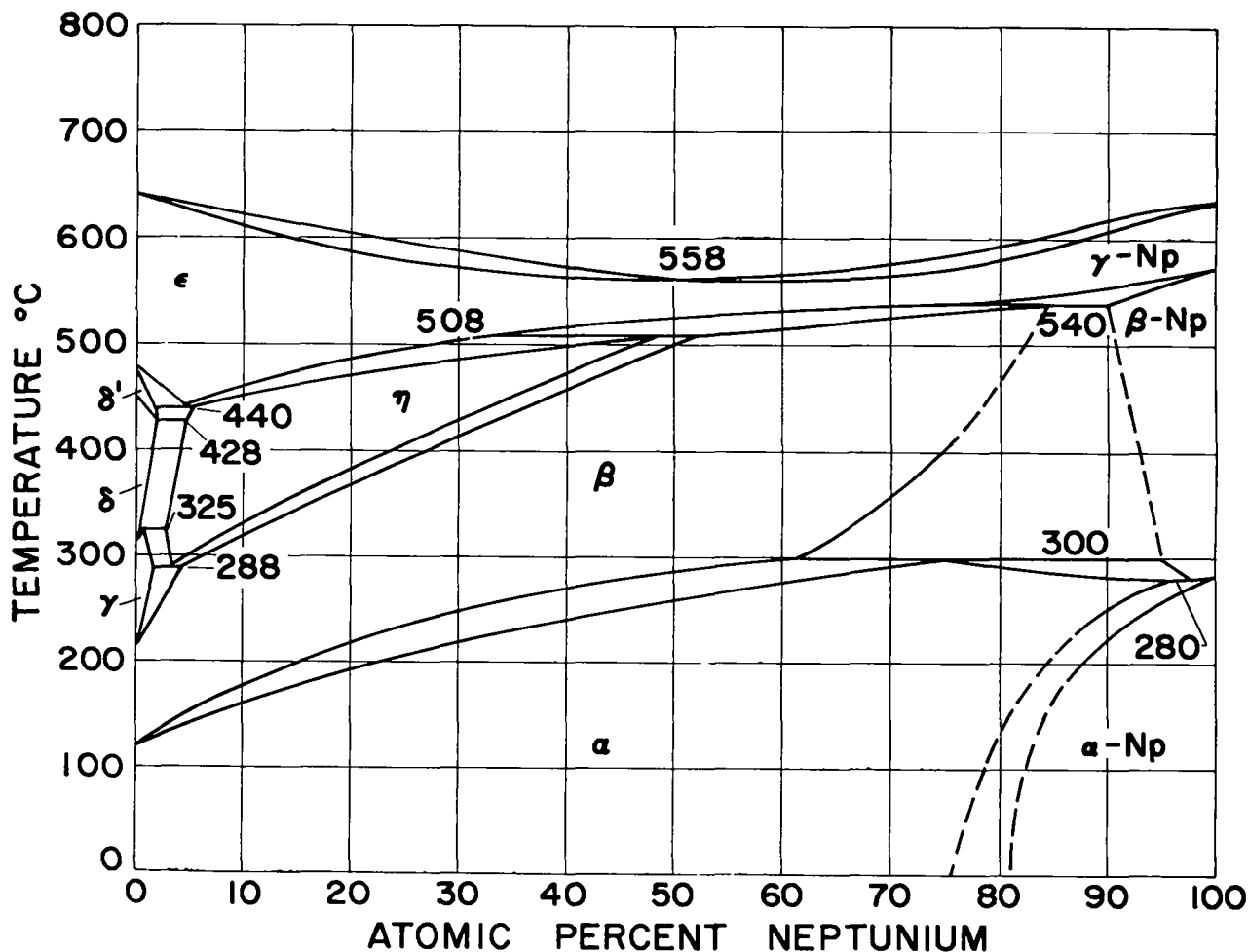


Fig. 26. Plutonium-neptunium phase diagram. (Redrawn from Ref. 1.)

TABLE XXXII. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-NEPTUNIUM ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
PuNp η		orthorhombic (tentative)		10.86	10.67	10.43	54 atoms		[1]

Plutonium-Oxygen

Several versions of the phase diagram for this system have been proposed. The earliest was presented by [1] in 1958. Since then the studies of [2], [3], and [4], in particular, have provided further information about Pu-O phase relationships. A complete diagram, however, is not well established and future work will very likely modify the composite diagram given here in Fig. 27. The region of that diagram below about 60 at.% O is due mainly to [1]; the high-temperature region between about 60 and 67 at.% O, to [2]; and the low-temperature region in that range, to [3] and [4]. Although there is evidence for the existence of PuO, for example in the product resulting from the reduction of PuO₂ by C, as reported by [5], and as a surface film on Pu metal, and although crystal structure data for the monoxide have been reported by [6], no pure bulk samples of PuO have yet been produced. It appears at present that PuO may be stabilized by impurities or by surface energy but that it does not occur as a solid, equilibrium phase. Therefore, the monoxide has not been included in the phase diagram shown in Fig. 27. Hexagonal Pu₂O₃, frequently termed β-Pu₂O₃, is a definite phase having the A-type rare-earth sesquioxide structure. Its upper limit of O content has been placed at about 60.2 at.% (O/Pu = 1.510) by [3], and its congruent melting point was found to be 2085°C by [2]. Body-centered cubic PuO_{1.52}, designated also as α-Pu₂O₃, is stable only below about 300°C. It has the C-type rare-earth sesquioxide structure. The exact composition of PuO_{1.61} is in doubt, and its true structure has not been definitely established but is believed to be closely related to that of PuO_{1.52}. PuO₂, at temperatures above about 650°C, can exist with considerable O deficiency, down to at least 63 at.% (O/Pu = 1.7) and perhaps as far as 61.7 at.% (O/Pu = 1.61) according to [10]. Removal of O expands the fcc lattice. Although PuO_{2-x} apparently cannot be retained to room temperature by quenching, [7] have observed that Ta or W impurity will stabilize PuO_{2-x} with an enlarged lattice parameter (a = 5.44 Å) at 25°C. [8] and [9] have also reported crystal structure data for the Pu oxides, and [10] have reviewed and summarized all the information that was available to them relative to the Pu-O system prior to September 1967. Crystal structure data are given in Table XXXIII.

[1] C. E. Holley, Jr., R. N. R. Mulford, E. J. Huber, Jr., E. L. Head, F. H. Ellinger, and C. W. Bjorklund, "Thermodynamics and Phase Relationships for Plutonium Oxides." *Proceedings of the Second United Nations*

International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958, Vol. 6, pp. 215-220, United Nations, Geneva (1958); also, Progress in Nuclear Energy, Series V, Vol. 3, pp. 128-138, Pergamon Press, London (1961).

- [2] T. D. Chikalla, C. E. McNeilly, and R. E. Skavdahl, "The Plutonium-Oxygen System," *J. Nucl. Mater.*, **12(2)**: 131-141 (1964).
- [3] E. R. Gardner, T. L. Markin, and R. S. Street, "The Plutonium-Oxygen Phase Diagram," *J. Inorg. Nucl. Chem.*, **27**: 541-551 (1965).
- [4] C. Sari, U. Benedict, and H. Blank, "Metallographic and X-Ray Investigations in the Pu-O and U-Pu-O Systems," Institut für Transurane, EURATOM, Karlsruhe, Paper SM 98/3 presented at the Symposium on Thermodynamics of Nuclear Materials with Emphasis on Solution Systems. IAEA, Vienna, Sept. 4-8, 1967.
- [5] R. E. Skavdahl, in "Quarterly Progress Report, Research and Development Programs Executed for the Division of Reactor Development, July, August, September, 1962." USAEC Report HW-75914, pp. 3.11-3.15. Hanford Atomic Products Operation (December 1962).
- [6] R. C. L. Mooney and W. H. Zachariasen. "Crystal Structure Studies of the Oxides of Plutonium," Paper 20.1 in *Transuranium Elements, Part II*. National Nuclear Energy Series, Div. IV, Vol. 14B, G. T. Seaborg, J. J. Katz, and W. M. Manning (Eds.), pp. 1442-1447. McGraw-Hill Book Co., Inc., New York (1949).
- [7] R. N. R. Mulford and L. E. LaMar. "The Volatility of Plutonium Oxide," in *Plutonium 1960*, E. Grison, W. B. H. Lord, and R. D. Fowler (Eds.), pp. 411-421, Cleaver-Hume Press, Ltd., London (1961).
- [8] F. H. Ellinger. "A Review of the Intermetallic Compounds of Plutonium," Chap. XXV in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), pp. 298-299. The University of Chicago Press, Chicago (1961).
- [9] D. H. Templeton and C. H. Dauben, "The Crystal Structures of NpC and Pu₂O₃," USAEC Report UCRL-1886. University of California. Radiation Laboratory (July 14, 1952).
- [10] Report of IAEA Panel Meeting on Plutonium Oxides, to be published by IAEA.

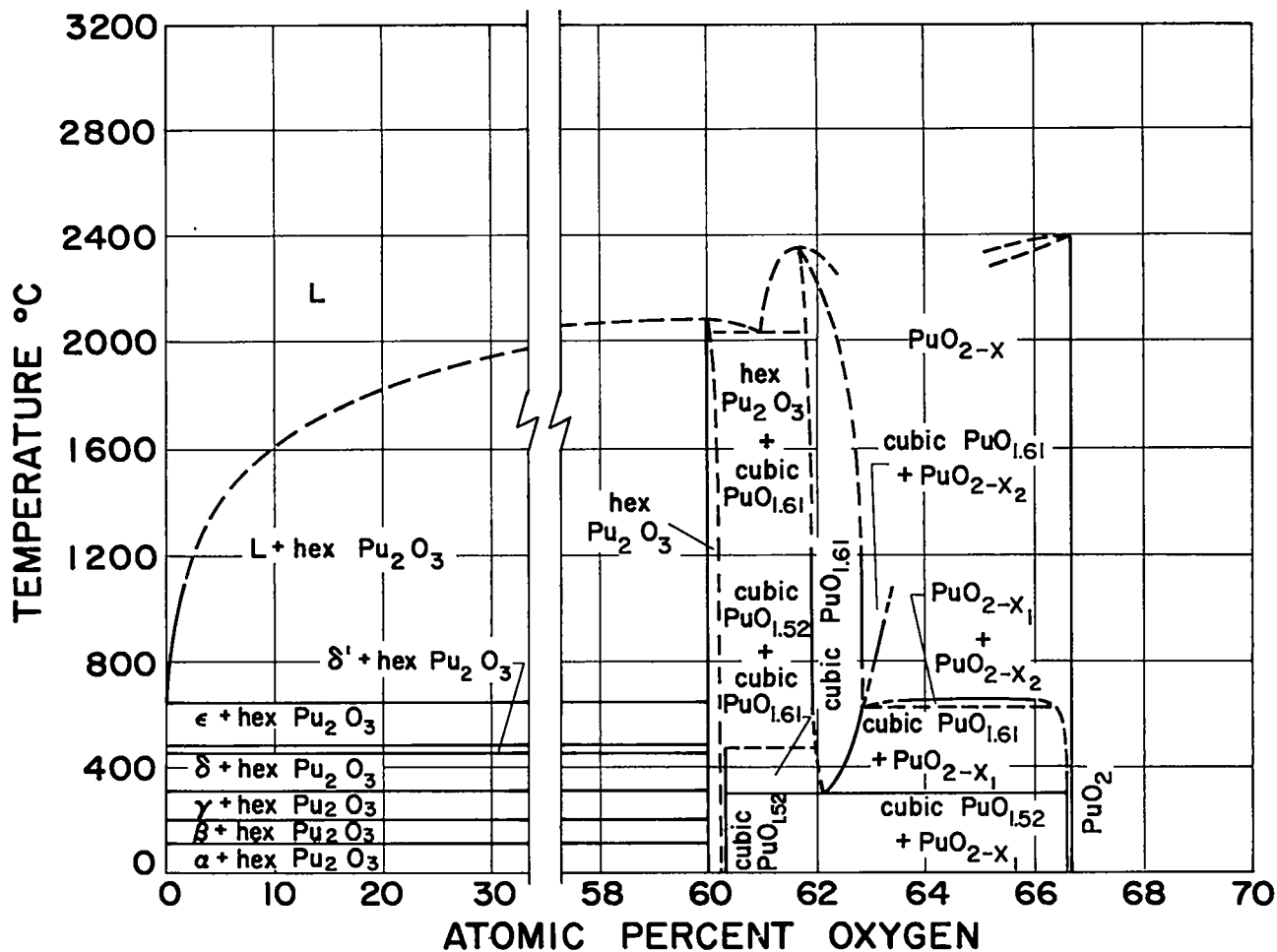


Fig. 27. Plutonium-oxygen phase diagram.

TABLE XXXIII. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-OXYGEN ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
hexagonal Pu ₂ O ₃	hexagonal	hexagonal	P $\bar{3}$ m1	3.841 3.840		5.958 5.957	1	11.47	[1] [9]
cubic PuO _{1.52}	Mn ₂ O ₃	body-centered cubic	Ia3	11.050 11.051			16	10.2	[10] [3]
cubic PuO _{1.61}		body-centered cubic		11.00 - 11.03					[10]
PuO ₂	CaF ₂	face-centered cubic	Fm3m	5.3960 5.3952			4	11.46	[8] [4]

Pu-Os □ **Plutonium-Osmium**

The partial phase diagram shown in Fig. 28 is based on the diagram published by [1], who used thermal, metallographic, and x-ray methods in studying Pu-Os alloys in the composition range from 0 to 66.7 at. % Os. The eutectic between ϵ Pu and η ($\sim \text{Pu}_3\text{Os}$) is placed at about 11 at. % Os and 495°C. ϵ Pu will dissolve a maximum of 6.8 at. % Os at that temperature, and the solid solution decomposes eutectoidally at 2.45 at. % Os and 410°C. The solubility of Os is less than 0.4 at. % in δ Pu and less than 0.3 at. % in γ , β , or α Pu. The ζ phase

($\sim \text{Pu}_{19}\text{Os}$), formed by peritectic reaction at 425°C, exists over a homogeneity range from 3.4 to 7.4 at. % Os. Crystal structure data are given in Table XXXIV.

- [1] A. A. Bochvar, S. T. Konobeevsky, V. I. Kutaitsev, T. S. Menshikova, and N. T. Chebotarev. "Interaction Between Plutonium and Other Metals in Connection with Their Arrangement in Mendeleev's Periodic Table." *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958*. Vol. 6, p. 191. United Nations, Geneva (1958).

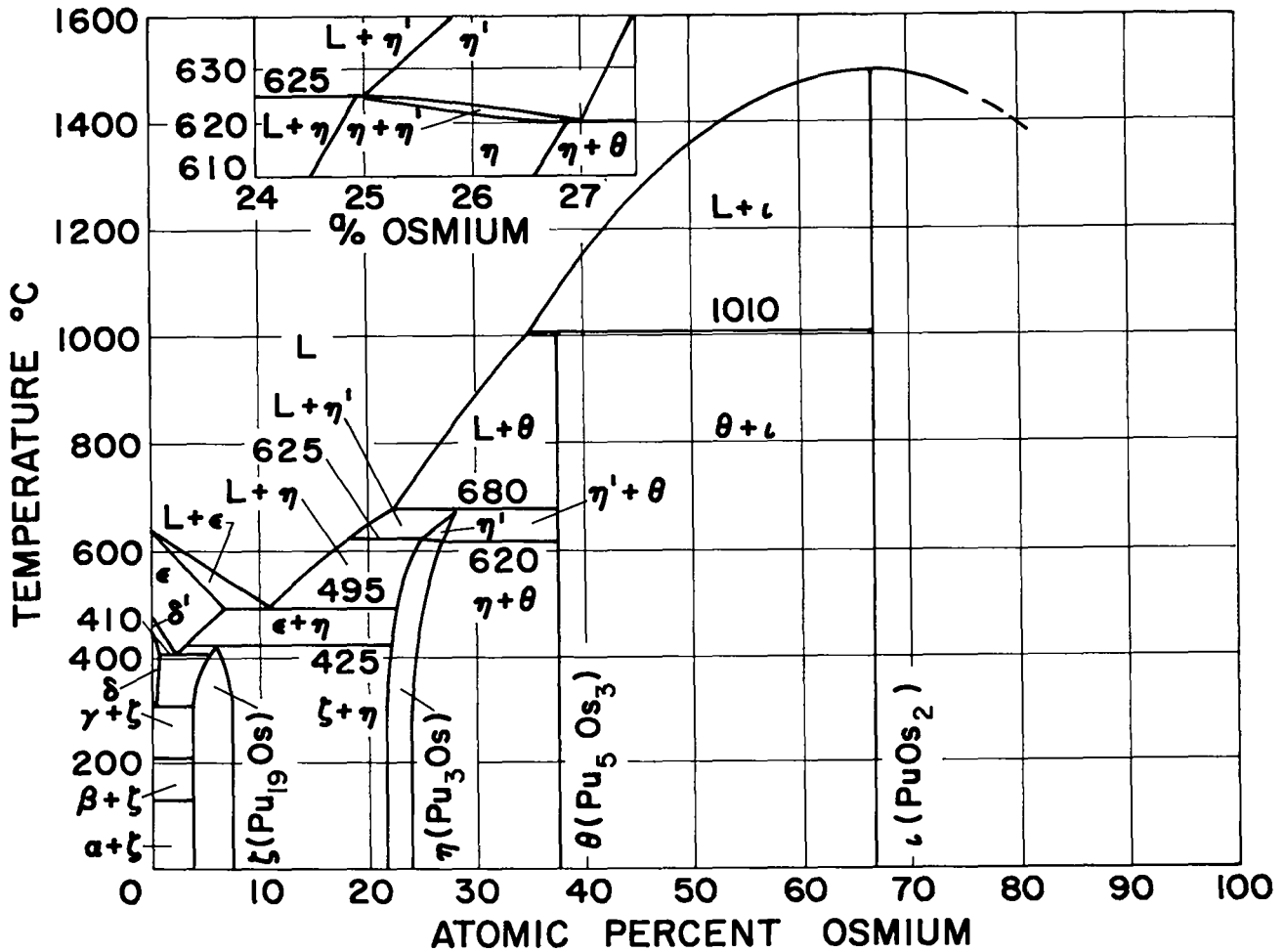


Fig. 28. Plutonium-osmium phase diagram. (Redrawn from Ref. 1.)

TABLE XXXIV. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-OSMIUM ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
~ Pu ₁₉ Os	unknown								
ζ									
~ Pu ₃ Os	unknown								
η									
~ Pu ₃ Os	unknown								
η'									
Pu ₅ Os ₃	unknown								
θ									
PuOs ₂	MgZn ₂	hexagonal	P6 ₃ /mmc	5.337		8.683	4	19.2	[1]
ι									

Pu-P □

Plutonium-Phosphorus

PuP has been prepared by induction melting of the components by [1] and by reaction of phosphine gas with powdered, decomposed PuH₂ by [2]. No higher phosphides of Pu were found. [1] reported the decomposition of PuP in the vicinity of 2000°C under slightly less than 1 atm of He, and [2] observed rapid decomposition at 2600°C under 2 atm of Ar. Crystal structure data for PuP are given in Table XXXV.

- [1] A. E. Gorum, "The Crystal Structures of PuAs, PuTe, PuP and PuOSe." *Acta Cryst.*, **10**: 144 (1957).
- [2] J. H. Handwerk, O. L. Kruger, and J. B. Moser, "Preparation and Properties of Some Groups V and VI Compounds of Plutonium," in *Plutonium 1965*, A. E. Kay and M. B. Waldron (Eds.), pp. 739-750. Chapman and Hall, London (1967).

TABLE XXXV. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-PHOSPHORUS ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
PuP	NaCl	face-centered cubic	Fm3m	5.664			4	9.87	[1]
				5.6514 ^a				9.89	[2]
				to					
				5.6598					

^aIn equilibrium with Pu.



Pu-Pb □

Plutonium-Lead

The phase diagram given in Fig. 29 is due largely to the work of [1] but has been modified to show the peritectic formation of the Pu-rich ζ phase, as indicated by the microstructural studies of [2]. The portion of the diagram reported by [1] between 35 and 60 at.% Pb has been omitted here because the phase relationships associated with congruent formation are not compatible with the peritectic formation of ζ . Addition of a small amount of Pb results in the eutectic lowering of the 640°C melting point of Pu to 610°C. Little, if any, solubility of Pb in the allotropes of Pu is indicated, and the same is true of the solid solubility of Pu in Pb. Crystal structure data have been reported for PuPb_3 by [3] but the crystal structure of the ζ phase, which is very pyrophoric, is not known (see Table XXXVI).

- [1] A. A. Bochvar, S. T. Konobeevsky, V. I. Kutaitsev, T. S. Menshikova, and N. T. Chebotarev, "Interaction Between Plutonium and Other Metals in Connection with Their Arrangement in Mendeleev's Periodic Table." *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958*, Vol. 6, pp. 188-189. United Nations, Geneva (1958).
- [2] E. M. Cramer, Los Alamos Scientific Laboratory, unpublished work.
- [3] F. H. Ellinger, "A Review of the Intermetallic Compounds of Plutonium." Chap. XXV in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), p. 295. The University of Chicago Press, Chicago (1961).

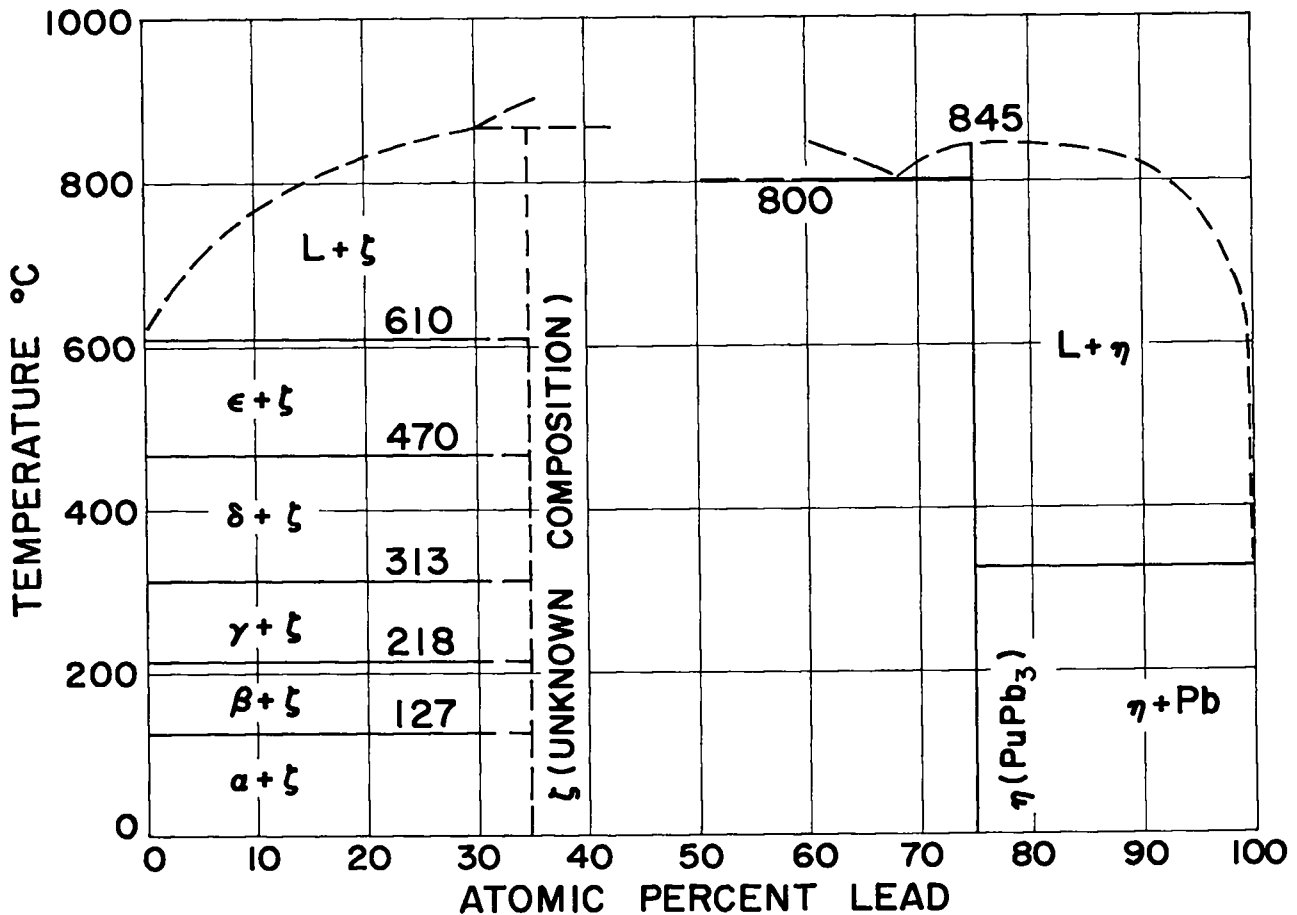


Fig. 29. Plutonium-lead phase diagram. (Courtesy Ellinger, Land, and Gschneidner and by permission of Gordon and Breach, Science Publishers, Inc., from *Plutonium Handbook*, Vol. 1, p. 211, 1967.)

TABLE XXXVI. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-LEAD ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
ζ	unknown								
PuPb ₃	Probably ordered AuCu ₃	cubic	Pm3m	4.81 4.808			1	12.86	[1] [3]

Pu-Pd □
Plutonium-Palladium

Apparently, the only complete diagram for this system has been published by [1] (see Fig. 30). They report four compounds: Pu_5Pd_4 , formed by peritectic reaction at 970°C ; PuPd , formed peritectically at 1150°C and stable down to 950°C where, on further cooling, it decomposes into Pu_5Pd_4 and Pu_3Pd_4 ; Pu_3Pd_4 , which melts congruently at $\sim 1190^\circ\text{C}$; and PuPd_3 , which has a homogeneity range of about 5 at.% and melts congruently at $\sim 1500^\circ\text{C}$. They also found three eutectics: ~ 1 at.% Pd at 630°C , 62 at.% Pd at

1060°C , and ~ 84 at.% Pd at 1240°C . Pd solubility in ϵ and δ Pu is indicated to not exceed 1 at.%, and the solid solubility of Pu in Pd was found to increase from 13.6 at.% Pu at room temperature to 14.3 at.% Pu at 1000°C . Crystal structure data are given in Table XXXVII.

[1] V. I. Kutaitsev, N. T. Chebotarev, I. G. Lebedev, M. A. Andrianov, V. N. Konev, and T. S. Menshikova. "Phase Diagrams of Plutonium with the Metals of Groups IIA, IVA, VIIIA and IB." in *Plutonium 1965*. A. E. Kay and M. B. Waldron (Eds.), pp. 420-449, Chapman and Hall, London (1967).

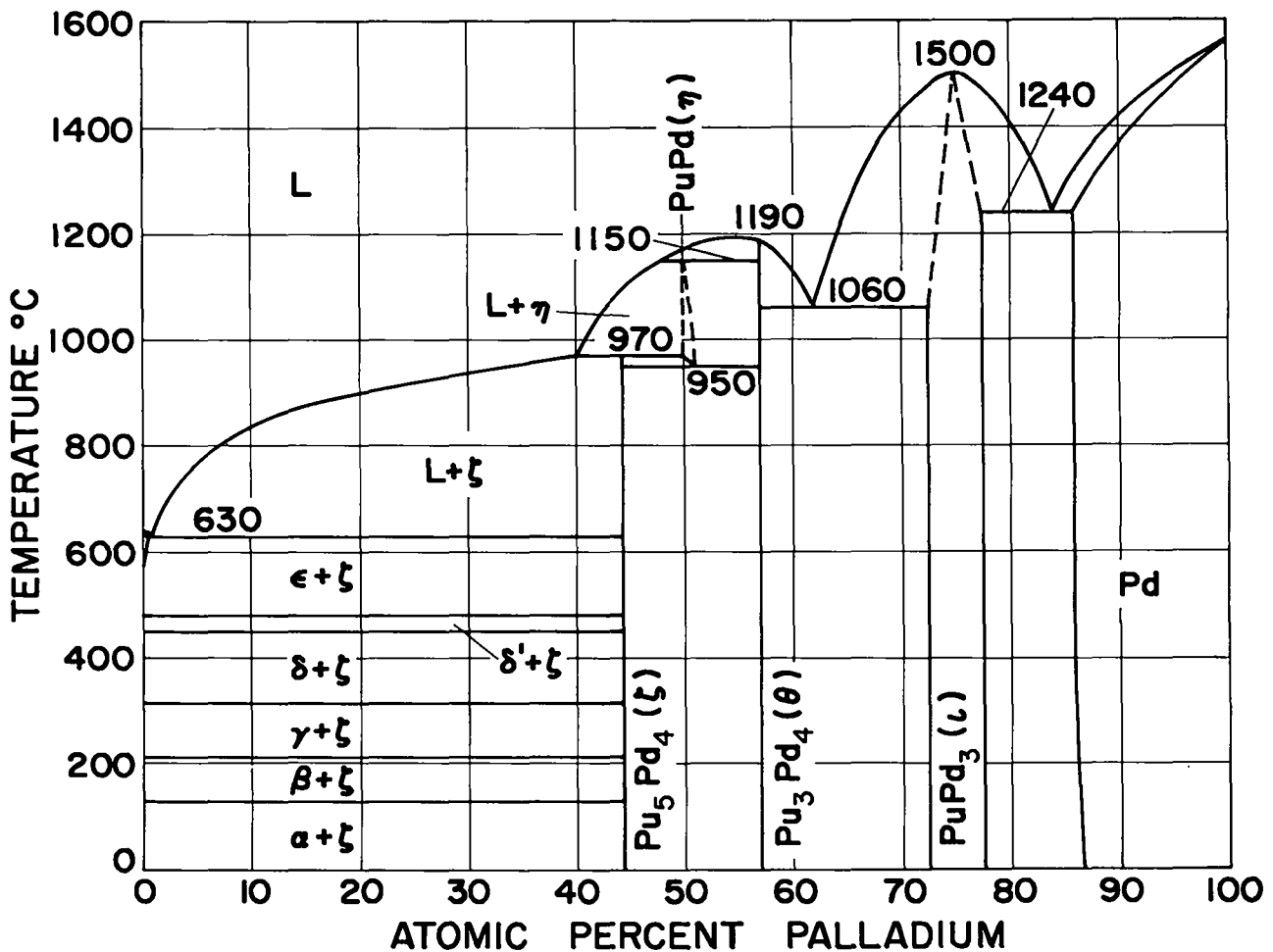


Fig. 30. Plutonium-palladium phase diagram. (Redrawn from Ref. 1.)

TABLE XXXVII. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-PALLADIUM ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
Pu ₅ Pd ₄ ξ	unknown								
PuPd η	unknown								
Pu ₅ Pd ₄ θ	rhombohedral (in hexagonal system)			7.916; α = 114.2° 13.304	5.783	2	12.8	[1]	
PuPd ₃ ι	AuCu ₃	cubic	Pm3m	4.077 to 4.119		1	13.41 for 75 at. % Pd	[1]	

Pu-Pr □
Plutonium-Praseodymium

The phase diagram of Fig. 31 has been constructed from the thermal, dilatometric, micrographic, and x-ray diffraction data of [1]. The only questionable feature of the diagram relates to the α' Pr phase which is an fcc form of the α Pr solid solution. It is not known whether α' Pr is an authentic Pu-Pr phase or one that is stabilized by impurities. [2] have published a partial preliminary diagram which shows as much as 10 at.% Pr soluble in δ Pu and in ϵ Pu. Also, it shows

a β Pr (fcc) field extending down to approximately 350°C.

- [1] F. H. Ellinger, C. C. Land, and K. A. Johnson, unpublished work as of June 1967.
- [2] V. I. Kutaitsev, N. T. Chebotarev, I. G. Lebedev, M. A. Andrianov, V. N. Konev, and T. S. Menshikova, "Phase Diagrams of Plutonium with the Metals of Groups IIA, IVA, VIIIA and IB," in *Plutonium 1965*. A. E. Kay and M. B. Waldron (Eds.), pp. 420-449. Chapman and Hall, London (1967).

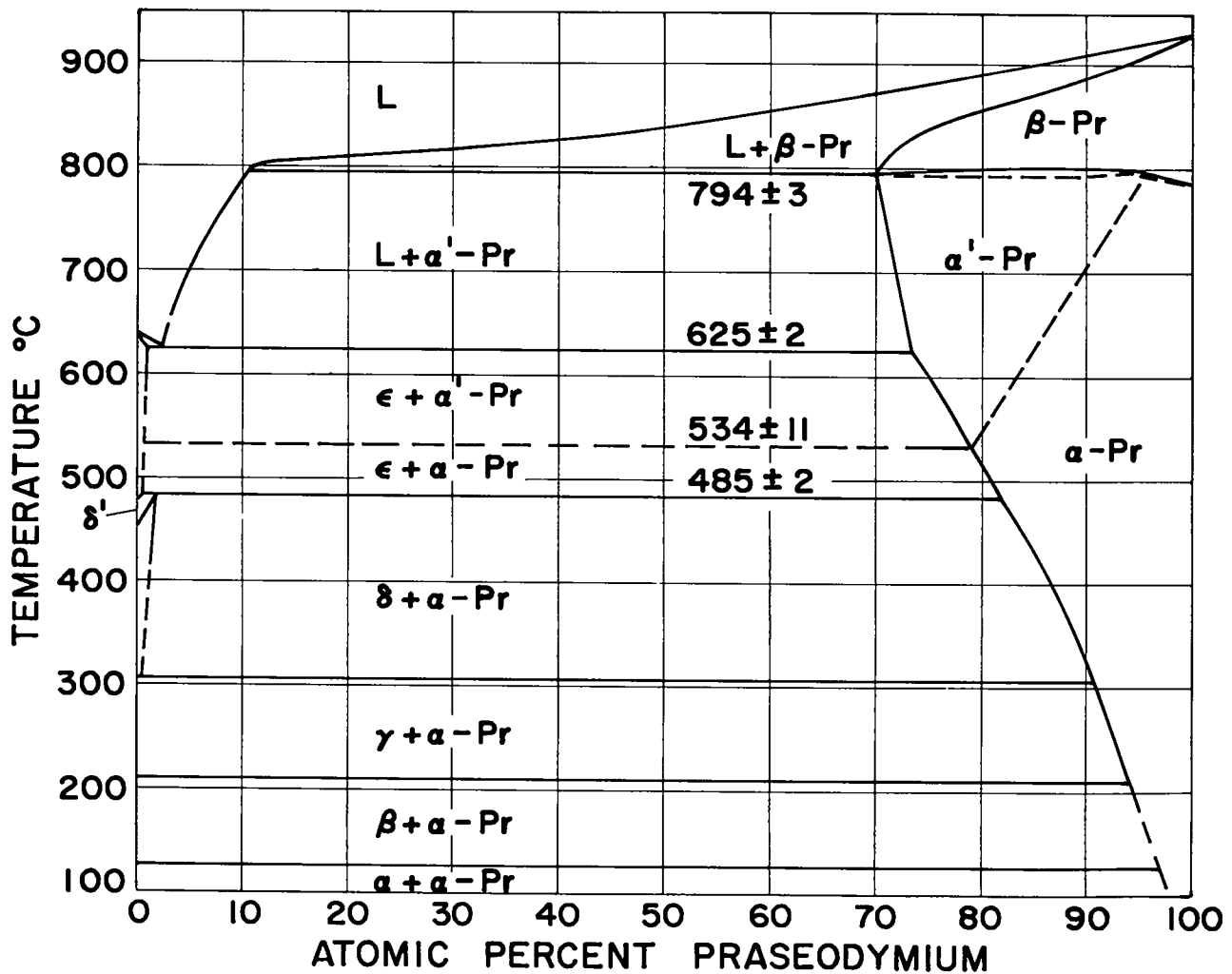
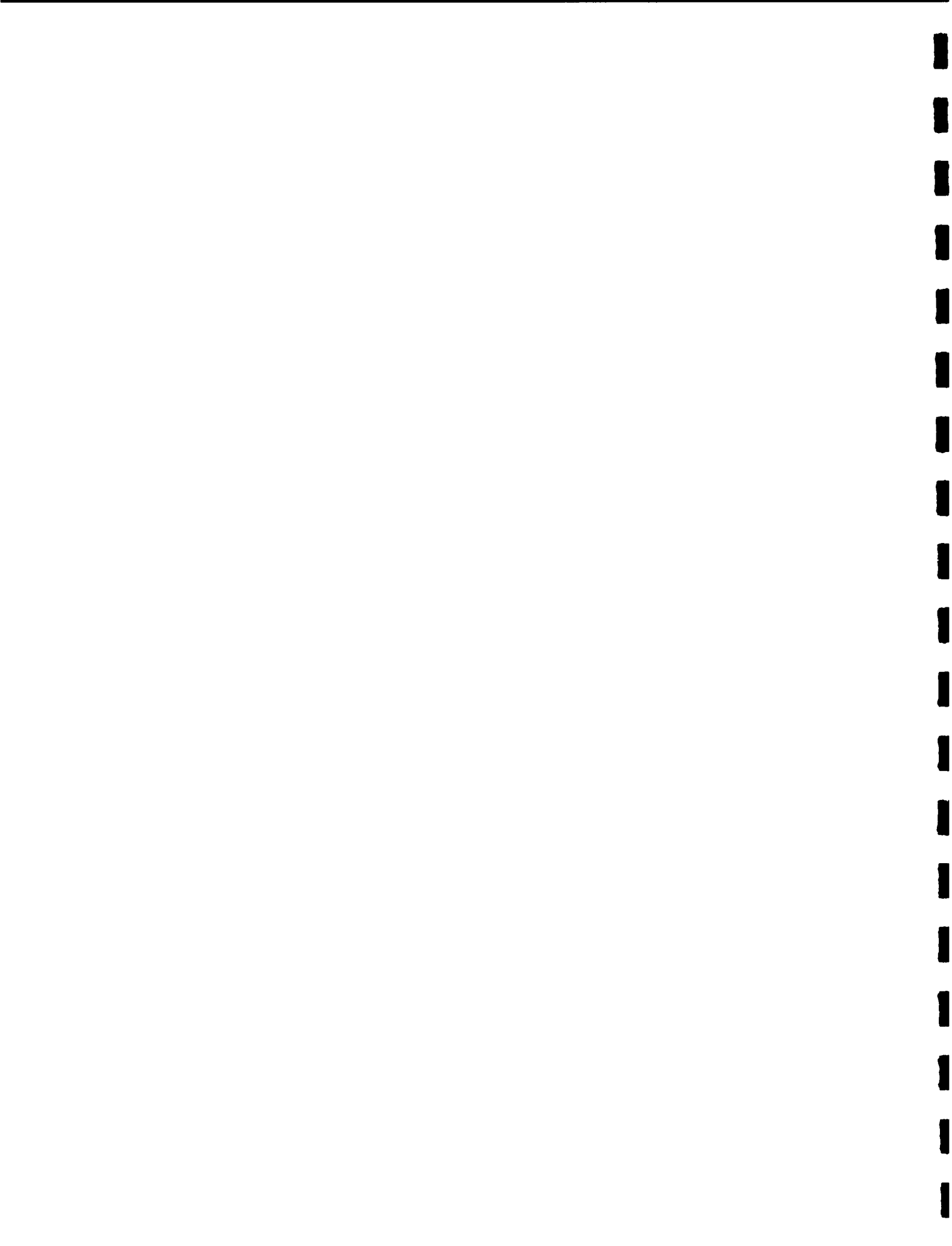


Fig. 31. Plutonium-praseodymium phase diagram.



Pu-Pt □
Plutonium-Platinum

significant. Crystal structure data are given in Table XXXVIII.

A complete diagram for this system has been published by [1], and is the basis for the diagram shown in Fig. 32. Also, the results of a cursory investigation by [2] have been reported. There appear to be three eutectics: the first at high Pu content and 622°C. probably near 1 at.% Pt, which is about the maximum solubility of Pt in ϵ Pu; the second between PuPt_3 and PuPt_5 ; and the third between PuPt_5 and Pt. The temperatures of the latter two eutectic reactions are unknown but are indicated to be well above 1000°C. The solubility of Pt in δ Pu is less than 1 at.%, and the solubility of Pu in Pt is so low as to be in-

[1] V. I. Kutaitsev, N. T. Chebotarev, I. G. Lebedev, M. A. Andrianov, V. N. Konev, and T. S. Menshikova. "Phase Diagrams of Plutonium with the Metals of Groups IIA, IVA, VIIIA and IB," in *Plutonium 1965*. A. E. Kay and M. B. Waldron (Eds.), pp. 420-449. Chapman and Hall, London (1967).

[2] "Reactor Fuels and Materials Development Plutonium Research: July-September, 1964," USAEC Report MLM-1220 Mound Laboratory, pp. 9-10. (Sept. 30, 1964).

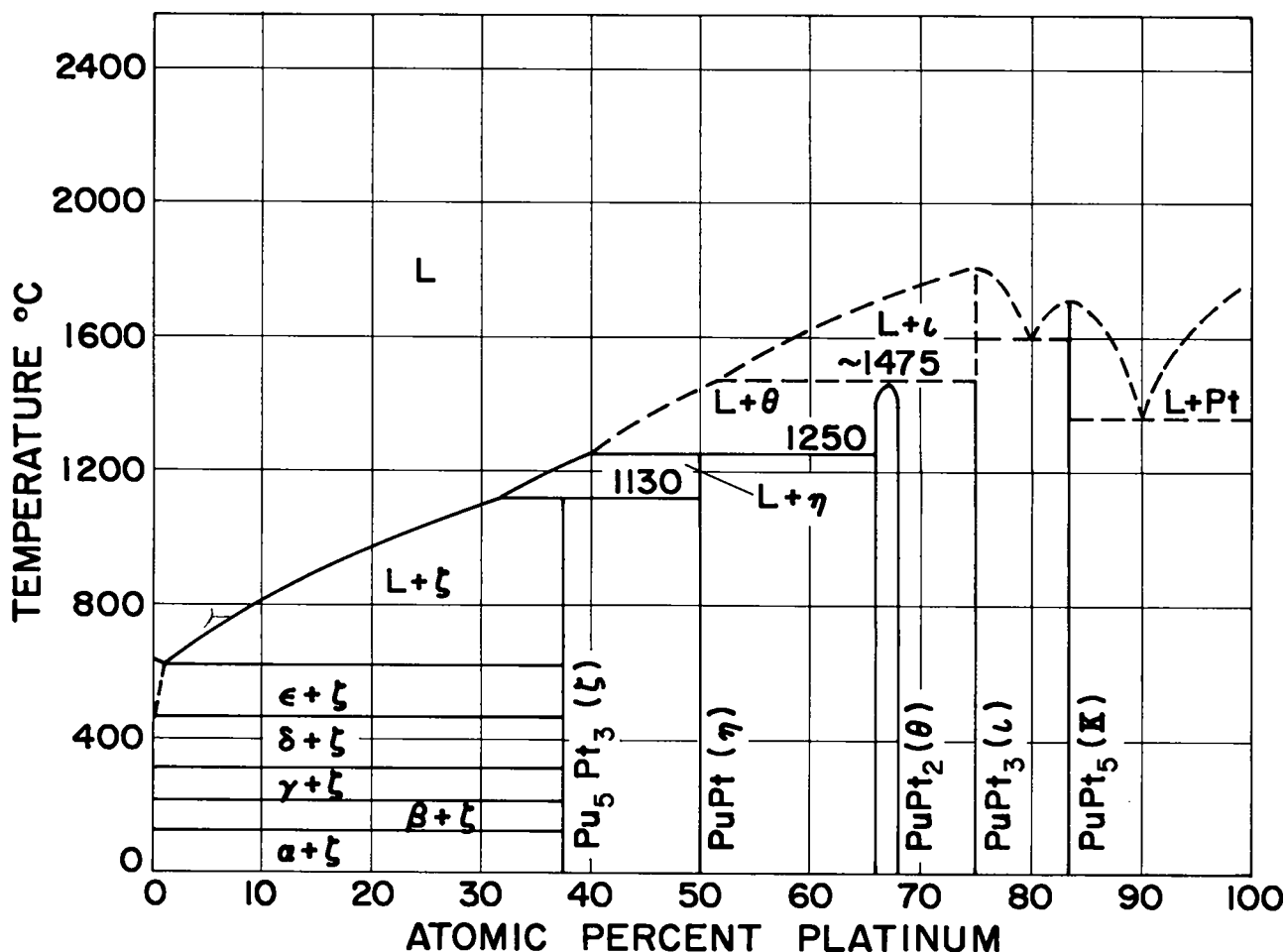


Fig. 32. Plutonium-platinum phase diagram. (Redrawn from Ref. 1.)

TABLE XXXVIII. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-PLATINUM ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
Pu ₅ Pt ₃ ξ	unknown								
PuPt η	TaB	orthorhombic		3.816	10.694	4.428		15.95	[1]
PuPt ₂ θ	Cu ₂ Mg	face-centered cubic	Fd3m	7.631 to 7.653			8	18.69	[1]
PuPt ₃ ι	AuCu ₃	cubic	Pm3m	4.107			1	19.75	[1]
PuPt ₆ κ	unknown								

Pu-Rb **Plutonium-Rubidium**

See Pu-Cs (Plutonium-Cesium).

Pu-Re **Plutonium-Rhenium**

One compound in this system has been identified by [1] as PuRe₂ (see Table XXXIX), and microstructural evidence of [2] indicates the existence of a eutectic between ϵ Pu and PuRe₂. The solubility of Re in liquid Pu between 700° and 950°C has been determined by [3] and is given in Table XL.

[1] F. H. Ellinger. "A Review of the Inter-metallic Compounds of Plutonium." Chap.

XXV in *The Metal Plutonium*. A. S. Coffinberry and W. N. Miner (Eds.). pp. 299-300. The University of Chicago Press. Chicago (1961).

[2] E. M. Cramer, Los Alamos Scientific Laboratory. unpublished work.

[3] D. F. Bowersox and J. A. Leary. "The Solubilities of Carbon, Tantalum, Tungsten, and Rhenium in Liquid Plutonium." *J. Nucl. Mater.*, **21(2)**: 219-224 (1967).

TABLE XXXIX. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-RHENIUM ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
PuRe ₂	MgZn ₂	hexagonal	P6 ₃ /mmc	5.396		8.729	4	18.45	[1]

TABLE XL. SOLUBILITY OF RHENIUM IN LIQUID PLUTONIUM

Temperature, °C	Solubility, at. % Re
700	1.57
750	1.89
800	2.35
850	2.75
900	3.27
950	4.05

Pu-Rh □

Plutonium-Rhodium

A complete phase diagram for this system has been published by [1] and is shown in Fig. 33. They report seven compounds (see Table XLI), two of which melt congruently: PuRh and PuRh₃, and the remaining five which form by peritectic reactions: Pu₂Rh at ~ 940°C, Pu₅Rh₃ at 980°C, Pu₅Rh₄ at 1180°C, Pu₃Rh₄ at 1310°C, and PuRh₂ at 1340°C. The maximum solubility of Rh in εPu is given as about 2 at.% at 590°C. The solubility

of Rh in δPu is significantly lower, and the solubility of Pu in Rh is not greater than 2 at.%.

[1] V. I. Kutaitsev, N. T. Chebotarev, I. G. Lebedev, M. A. Andrianov, V. N. Konev, and T. S. Menshikova, "Phase Diagrams of Plutonium with the Metals of Groups IIA, IVA, VIIIA and IB," in *Plutonium* 1965, A. E. Kay and M. B. Waldron (Eds.), pp. 420-449, Chapman and Hall, London (1967).

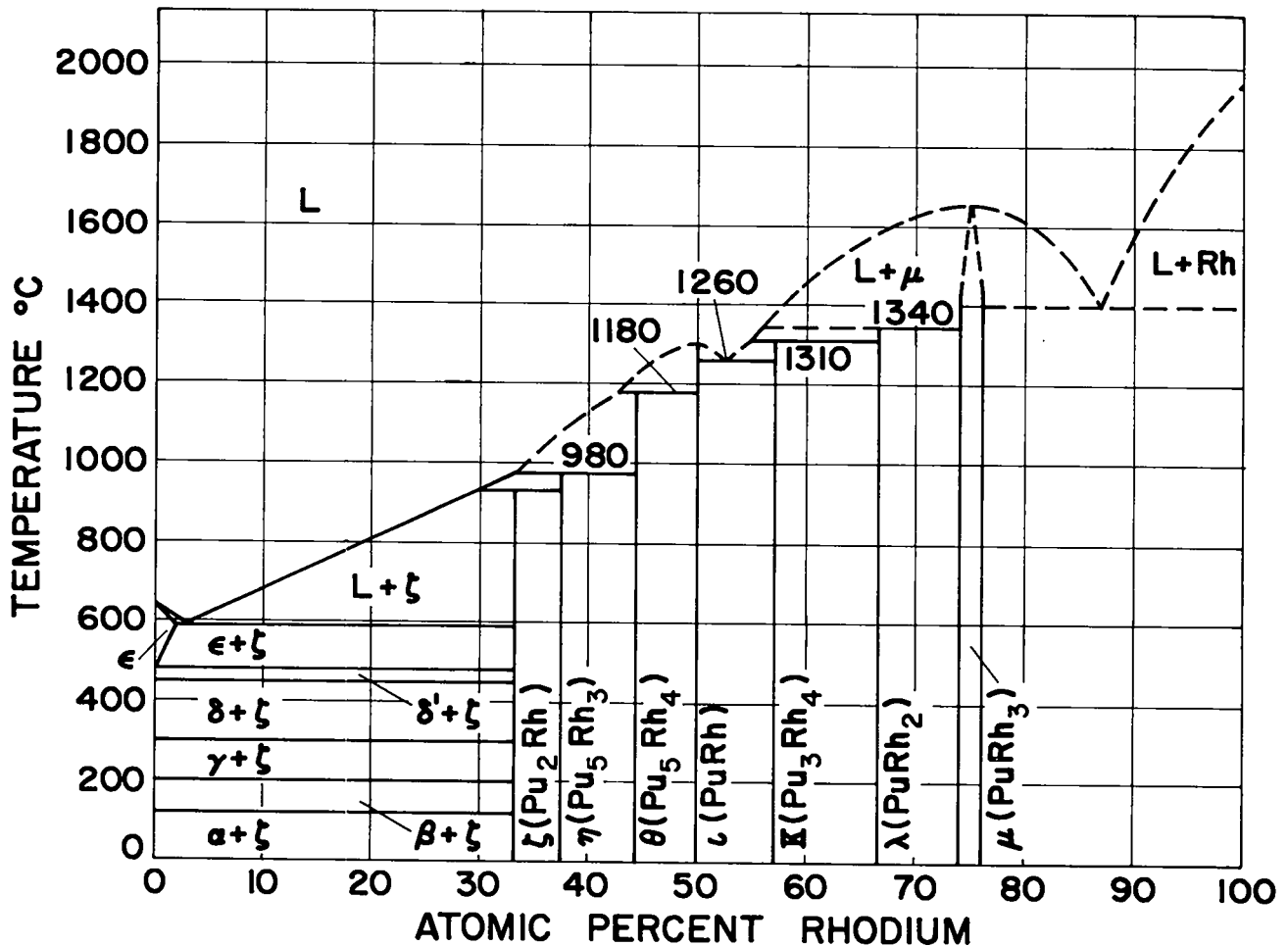


Fig. 33. Plutonium-rhodium phase diagram. (Redrawn from Ref. 1.)

TABLE XLI. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-RHODIUM ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
Pu ₂ Rh ζ	unknown								
Pu ₅ Rh ₃ η	unknown								
Pu ₅ Rh ₄ θ	unknown								
PuRh ι	unknown								
Pu ₃ Rh ₄ κ	unknown								
PuRh ₂ λ	Cu ₂ Mg	face-centered cubic	Fd3m	7.488			8	14.07	[1]
PuRh ₃ μ	AuCu ₃	cubic	Pm3m	4.009 to 4.040			1	13.95	[1]

Pu-Ru
Plutonium-Ruthenium

Partial phase diagrams for this system have been published by [1], [2], and [3]. The diagram shown in Fig. 34 is based mainly on the investigations of [1] and [2]. The work of [1] is supported by thermal, x-ray, and microstructural data; that of [2], which is restricted to the region from 0 to 25 at.% Ru, by thermal, dilatometric, and density measurements; and that of [3] by thermal, dilatometric, and x-ray data. There is general agreement as to the form of the diagram from 0 at.% Ru to about Pu_5Ru_3 and from PuRu_2 to 100 at.% Ru, although there are a few differences concerning some phase boundary details. [2] found the maximum solubility of Ru in ϵPu to be 4.5 at.%. [3] have reported this solu-

bility to be about 3 at.%, and [4] found the limit to be slightly greater than 5 at.%. The solubility of Ru in $\delta'\text{Pu}$ is less than 1 at.% according to [5], in δPu about 1 at.% according to [2], and is negligibly small in γ , β and αPu . [2] found ζ to have a homogeneity range extending from 3 to 6 at.% Ru and placed the eutectic point at 4.2 at.% Ru. Near the central region of the diagram, [3] have reported that Pu_5Ru_3 and PuRu are formed by peritectic reaction at 1025° and 1250°C , respectively. On the other hand, a small amount of microstructural data of [5] indicates that Pu_5Ru_3 and PuRu melt congruently and that the eutectics $\text{Pu}_3\text{Ru}_3/\text{PuRu}$ and $\text{PuRu}/\text{PuRu}_2$ are formed. Crystal structure data (see Table XLII) for only PuRu and PuRu_2 have been reported by [3] and [6].

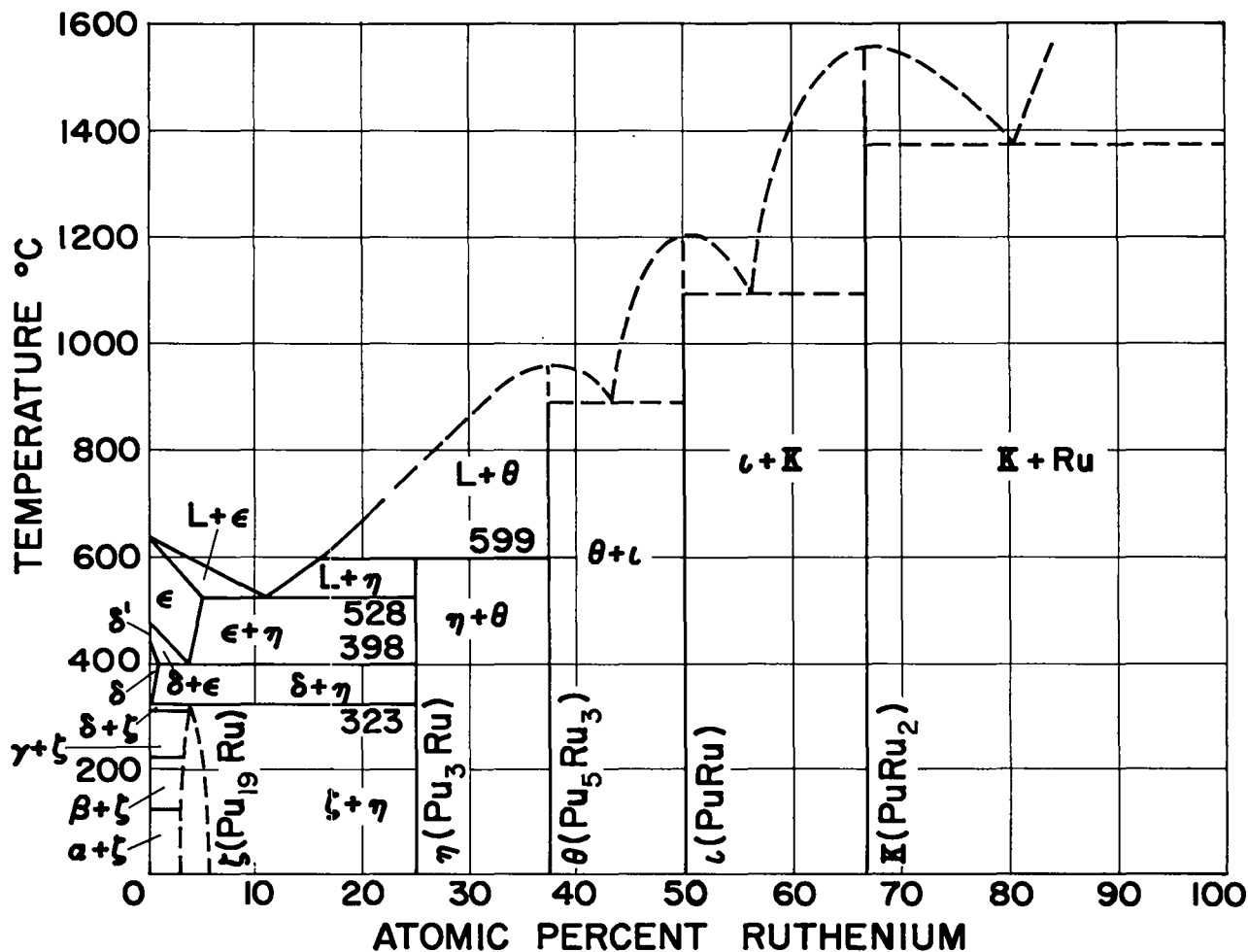


Fig. 34. Plutonium-ruthenium phase diagram. (Courtesy Ellinger, Land, and Gschneidner and by permission of Gordon and Breach, Science Publishers, Inc., from *Plutonium Handbook*, Vol. 1, p. 219, 1967.)

- [1] F. W. Schonfeld. "Plutonium Phase Diagrams Studied at Los Alamos," Chap. XXII in *The Metal Plutonium*. A. S. Coffinberry and W. N. Miner (Eds.), p. 246. The University of Chicago Press, Chicago (1961).
- [2] R. G. Cope, D. G. Hughes, R. G. Loasby, and D. C. Miller. "The Plutonium-Ruthenium and Plutonium-Neptunium Binary Phase Diagrams." in *Plutonium 1960*. E. Grison, W. B. H. Lord, and R. D. Fowler (Eds.), pp. 280-286. Cleaver-Hume Press, Ltd., London (1961).
- [3] V. I. Kutaitsev, N. T. Chebotarev, I. G. Lebedev, M. A. Andrianov, V. N. Konev, and T. S. Menshikova. "Phase Diagrams of Plutonium with the Metals of Groups IIA, IVA, VIIIA and IB." in *Plutonium 1965*. A. E. Kay and M. B. Waldron (Eds.), pp. 420-449. Chapman and Hall, London (1967).
- [4] E. M. Cramer. Los Alamos Scientific Laboratory. unpublished work.
- [5] F. H. Ellinger, Los Alamos Scientific Laboratory. unpublished work.
- [6] F. H. Ellinger, "A Review of the Intermetallic Compounds of Plutonium." Chap. XXV in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), p. 300. The University of Chicago Press, Chicago (1961).

TABLE XLII. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-RUTHENIUM ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
Pu ₁₀ Ru ζ	unknown								
Pu ₃ Ru η	unknown								
Pu ₅ Ru ₃ θ	unknown								
PuRu ι	CsCl	cubic	Pm3m	3.363 3.3635			1	14.87 14.84	[6] [3]
PuRu ₂ κ	Cu ₂ Mg	face-centered cubic	Fd3m	7.476 7.472 to 7.476			8	14.06 14.03	[6] [3]

Pu-S □

Plutonium-Sulfur

The compounds PuS and Pu₂S₃-Pu₃S₄ were first identified by [1]. Crystal structure data for PuS₂ and Pu₂S₃ have been reported by [2], and for PuS and Pu₂S₃ by [3] (see Table XLIII). According to [2], a range of homogeneity for PuS₂ is indicated by an observed decrease in the *a* dimension of the tetragonal unit cell when S is removed without the formation of a new phase. Also, [2] have reported the existence of three modifications of Pu₂S₃, which they designate as

α (stable up to 1200°C), β (stable between 1200 and 1400°C), and γ (stable above 1400°C). [4] has reported that Pu₂S₃ melts congruently at 1725 ± 10°C under a vacuum of 10⁻² torr. In Ar, however, he found this compound to be stable to 2300°C. the maximum temperature attained. The melting point of PuS is given as 2330 ± 30°C by [3].

[1a] W. H. Zachariasen, "Crystal Chemical Studies of the 5f-Series of Elements: X. Sulfides and Oxysulfides," *Acta Cryst.*, 2: 291-296 (1949).

TABLE XLIII. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-SULFUR ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
PuS	NaCl	face-centered cubic	Fm3m	5.536			4	10.60	[1a]
				5.540				10.59	[3]
				5.5409 ^a					
Pu ₂ S ₃ α		orthorhombic		7.69	8.41	15.15			[2]
Pu ₂ S ₃ β		orthorhombic		7.18	10.50	12.98			[2]
Pu ₂ S ₃ γ		cubic		8.4546					[2]
Pu ₂ S ₃ - Pu ₃ S ₄	Th ₃ P ₄	body-centered cubic	I43d	8.4182			4	8.526	[3]
				8.4543				8.4	[1b]
PuS ₂		tetragonal		7.962		7.962			[2]
PuS _{2-x} ^b		tetragonal		7.886		7.962			[2]

^aIn equilibrium with Pu₂S₃.

^bIn equilibrium with Pu₂S₃ (where x corresponds to the range of homogeneity).

- [1b] W. H. Zachariasen, "Crystal Chemical Studies of the 5f-Series of Elements: VI. The Ce_2S_3 - Ce_3S_4 Type of Structure." *Ibid.*, pp. 57-60.
- [2] J. P. Marcon and R. Pascard, "Sulfures et seleniures superieurs de plutonium et des lanthanides" (in French). *J. Inorg. Nucl. Chem.*, **28**: 2551-2560 (1966).
- [3] J. H. Handwerk, O. L. Kruger, and J. B. Moser, "Preparation and Properties of Some Groups V and VI Compounds of Plutonium." in *Plutonium 1965*. A. E. Kay and M. B. Waldron (Eds.), pp. 739-750. Chapman and Hall, London, (1967).
- [4] Y. B. Katayama, in "Ceramics Research and Development Operation, Quarterly Report, October — December, 1962," USAEC Report HW-76300. p. 2.1. Hanford Atomic Products Operation (May 16, 1963).

Pu-Sc □ **Plutonium-Scandium**

Comparison of the phase diagram of [1] (Fig. 35) with that of [2] (Fig. 36) reveals

overall agreement with the exception of the fcc form of Sc shown by [1]. This form of Sc is known to be due to impurities. The δ Pu and α Sc solvus lines differ significantly. Crystal structure data are given in Table XLIV.

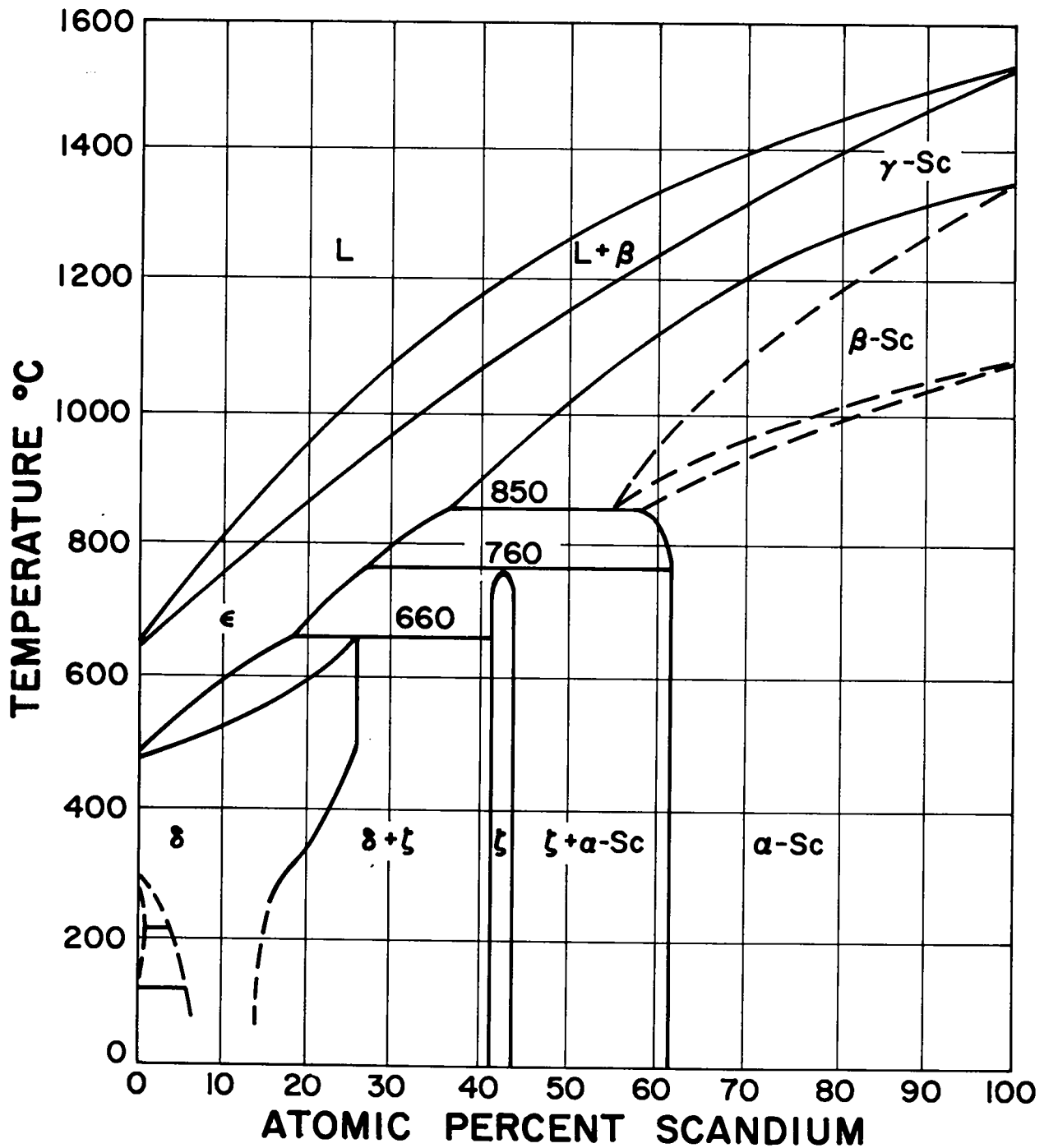


Fig. 35. Plutonium-scandium phase diagram. (Redrawn from Ref. 1.)

[1] V. I. Kutaitsev, N. T. Chebotarev, I. G. Lebedev, M. A. Andrianov, V. N. Konev, and T. S. Menshikova, "Phase Diagrams of Plutonium with the Metals of Groups IIA, IVA, VIIIA and IB," in *Plutonium* 1965. A. E. Kay and M. B. Waldron (Eds.). pp.

420-449, Chapman and Hall, London (1967).

[2] F. H. Ellinger, K. A. Johnson, and C. C. Land, Los Alamos Scientific Laboratory, unpublished work as of June 1967.

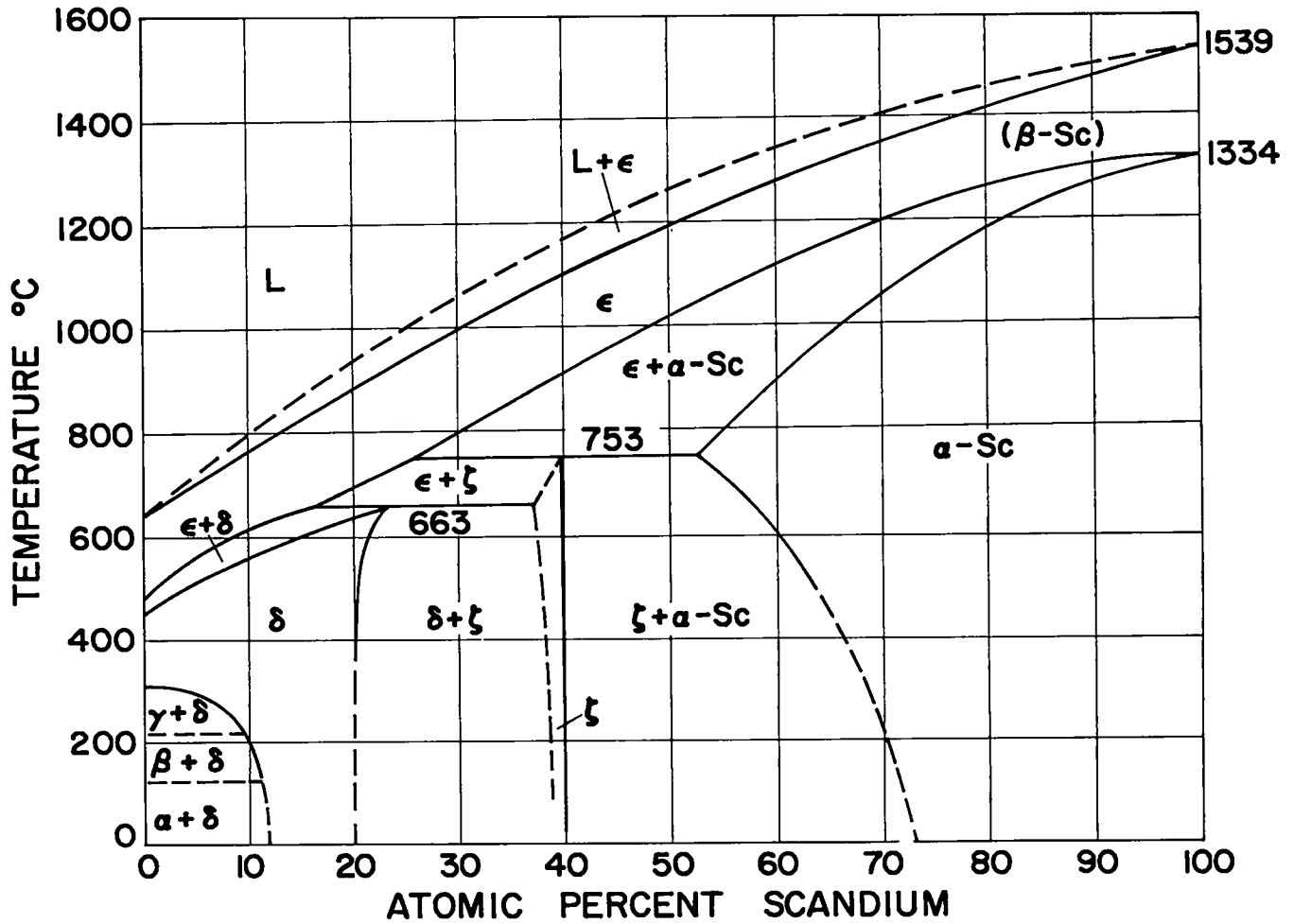
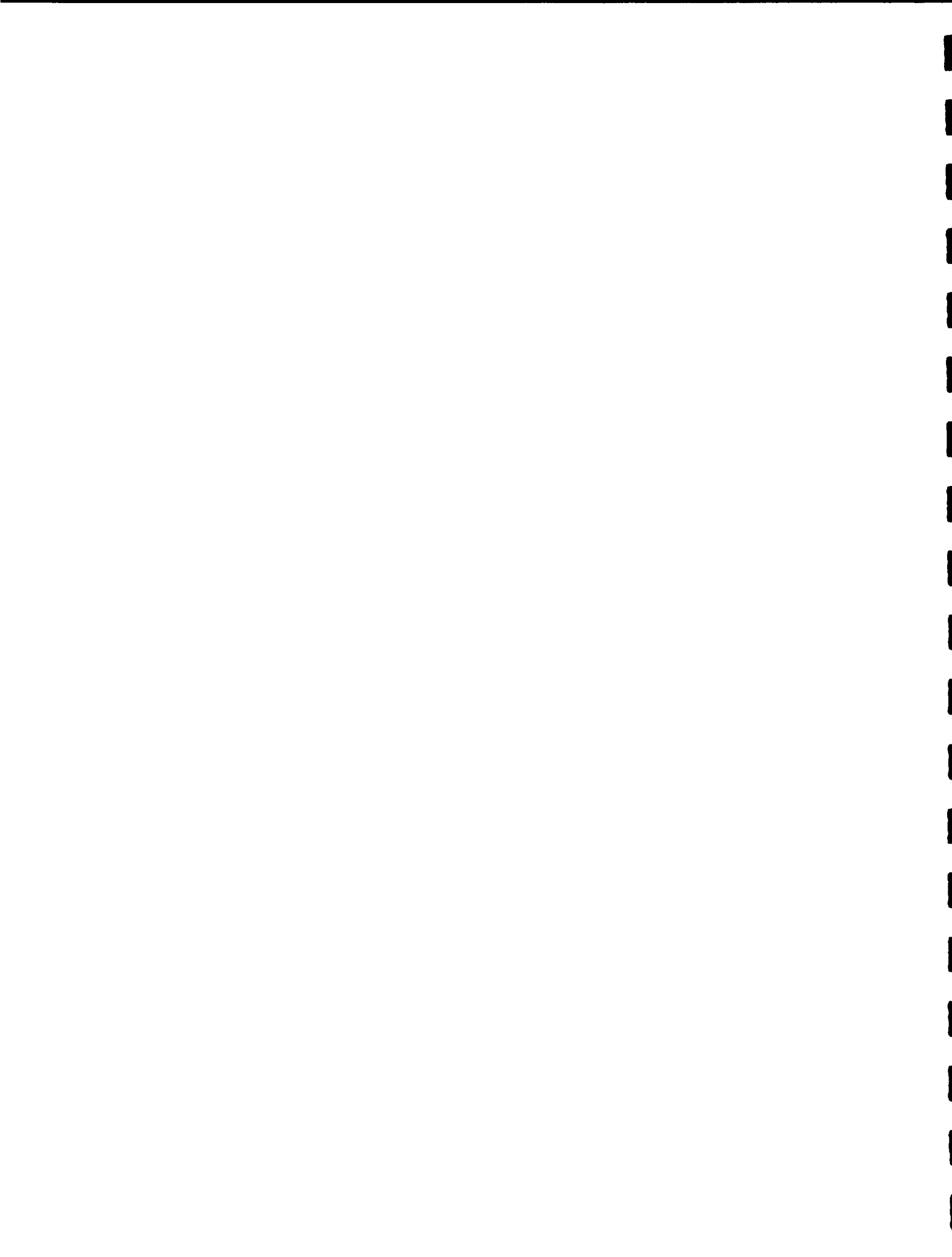


Fig. 36. Plutonium-scandium phase diagram.

TABLE XLIV. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-SCANDIUM ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
ζ (~ 40 at.% Sc)	hexagonal		P6 ₃ /mmc	3.310 (Pu-rich)	10.717	4 atoms	10.5	[2]	
				3.308 (Sc-rich)	10.709			[2]	
				3.310	10.715				
				to 3.307	to 10.718			[1]	



Pu-Se □

Plutonium-Selenium

The compounds PuSe_2 and Pu_2Se_3 have been described by [1]. PuSe_2 appears to have a range of homogeneity, since its lattice parameters were found to vary according to whether the compound was pure or contained Pu_2Se_3 . Pu_2Se_3 exists in two forms, η and γ , which are isomorphous with their

rare earth homologues (see Table XLV). The orthorhombic η form when heated in vacuum at 1400°C was found to transform to fcc γ .

- [1] J. P. Marcon and R. Pascard. "Sulfures et seleniures superieurs de plutonium et des lanthanides" (in French), *J. Inorg. Nucl. Chem.*, **28**: 2551-2560 (1966).

TABLE XLV. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-SELENIUM ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm^3	Ref.
				a	b	c			
Pu_2Se_3 η	U_2S_3	orthorhombic		4.10	11.10	11.32			[1]
Pu_2Se_3 γ		cubic		8.787					[1]
PuSe_2		tetragonal		8.33	8.41				[1]
PuSe_{2-x}^a		tetragonal		8.198	8.364				[1]

^aIn equilibrium with Pu_2Se_3 (where x corresponds to the range of homogeneity).



Pu-Si □

Plutonium-Silicon

A complete diagram of the system, based on thermal, dilatometric, microstructural, and x-ray data has been published by [1] (see Fig. 37). Crystal structure data for the five intermediate compounds have been reported: Pu_5Si_3 (ζ) by [1] and [2]. Pu_3Si_2 (η) by [1]. PuSi (θ) by [3]. $\sim\text{Pu}_3\text{Si}_5$ (ι) by [1], [3] and [4], and $\sim\text{PuSi}_2$ (κ) by [3] and [5] (see Table XLVI). Both ι and κ appear to have homogeneity ranges of a few percent; the reactions of these phases with the liquid phase have not been established conclusively. Only a few liquidus points were determined; consequently, most of the liquidus appears as a broken line in the diagram. Very lit-

tle solubility of Si in α , β , and γPu is indicated. The solubility limit of Si in δPu at 350°C is about 0.5 at.% and in the ϵ phase at 550°C is about 0.6 at.%. The maximum solubility in ϵPu is approximately 1 at.% at 590°C . [6] have shown that less than 0.5 at.% Si eliminates the δ' phase in these alloys. Although δPu is not stabilized to room temperature by the addition of Si, the δ phase can be retained metastably at that temperature if alloys containing at least 0.75 at.% Si are cooled sufficiently rapidly from the δ phase temperature region, e.g., by water quenching. Information on the solubility of Pu in Si is lacking.

- [1] C. C. Land, K. A. Johnson, and F. H. Ellinger, "The Plutonium-Silicon System." *J. Nucl. Mater.*, **15**(1): 23-32 (1965).

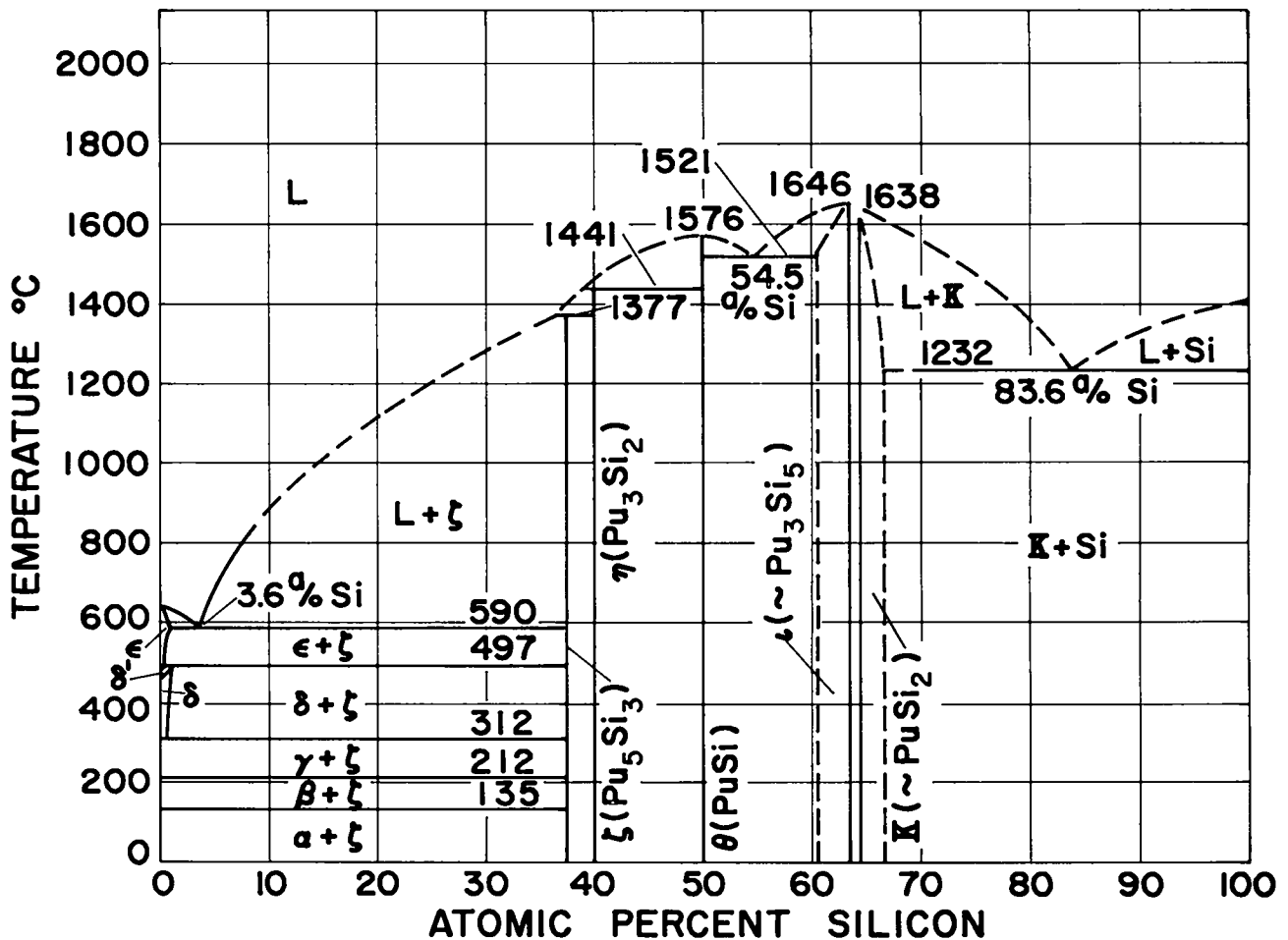


Fig. 37. Plutonium-silicon phase diagram. (Redrawn from Ref. 1.)

- [2] D. T. Cromer, A. C. Larson, and R. B. Roof, Jr., "The Crystal Structure of Pu_5Si_3 ," *Acta Cryst.*, **17**: 947-950 (1964).
- [3] F. H. Ellinger. "A Review of the Inter-metallic Compounds of Plutonium." Chap. XXV in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), pp. 300-302, The University of Chicago Press, Chicago (1961).
- [4] O. J. C. Runnalls and R. R. Boucher, "The Crystal Structure of βPuSi_2 ," *Acta Cryst.*, **8**: 592 (1955).
- [5] W. H. Zachariasen. "Crystal Chemical Studies of the 5f-Series of Elements: VIII. Crystal Structure Studies of Uranium Silicides and of CeSi_2 , NpSi_2 and PuSi_2 ." *Acta Cryst.*, **2**: 94-99 (1949).
- [6] R. O. Elliott and A. C. Larson. "Delta-Prime Plutonium," Chap. XXIV in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), p. 272. The University of Chicago Press, Chicago (1961).

TABLE XLVI. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-SILICON ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
Pu_5Si_3 ζ	W_5Si_3	body-centered tetragonal	I4/mcm	11.407		5.444	4	12.00	[1]
				11.409		5.448		11.98	[2]
Pu_3Si_2 η	U_3Si_2	tetragonal	P4/mbm	7.483		4.048	2	11.33	[1]
PuSi θ	FeB	orthorhombic	Pbnm	5.727	7.933	3.847	4	10.15	[3]
$\sim \text{Pu}_3\text{Si}_6$ ι	Defect AlB_2	hexagonal	P6/mmm	3.875		4.102	1	8.96	[1]
				3.876		4.090		8.8	[3]
				3.884		4.082			[4]
$\sim \text{PuSi}_2$ κ	ThSi_2	tetragonal	I4 ₁ /amd	3.967		13.72	4	9.08	[3]
				(excess Si) 3.98		13.58			[5]
				(unknown composition)					

Pu-Sm
Plutonium-Samarium

According to [1], the general form of the Pu-Sm phase diagram resembles that of Pu-Nd (see Fig. 38). Sm additions lower the melting point of Pu by about 3°C. Less than 5 at.% Sm is soluble in δ Pu and ϵ Pu. A maximum of about 33 at.% Pu is soluble in bcc β Sm, and a maxi-

imum of about 30 at.% Pu is soluble in α Sm. However, the Sm structure (9-fold stacking sequence of close-packed layers) was found to change to the double-hexagonal close-packed structure with the solution of about 5 at.% Pu.

[1] F. H. Ellinger, C. C. Land, and K. A. Johnson, Los Alamos Scientific Laboratory, unpublished work as of June 1967.

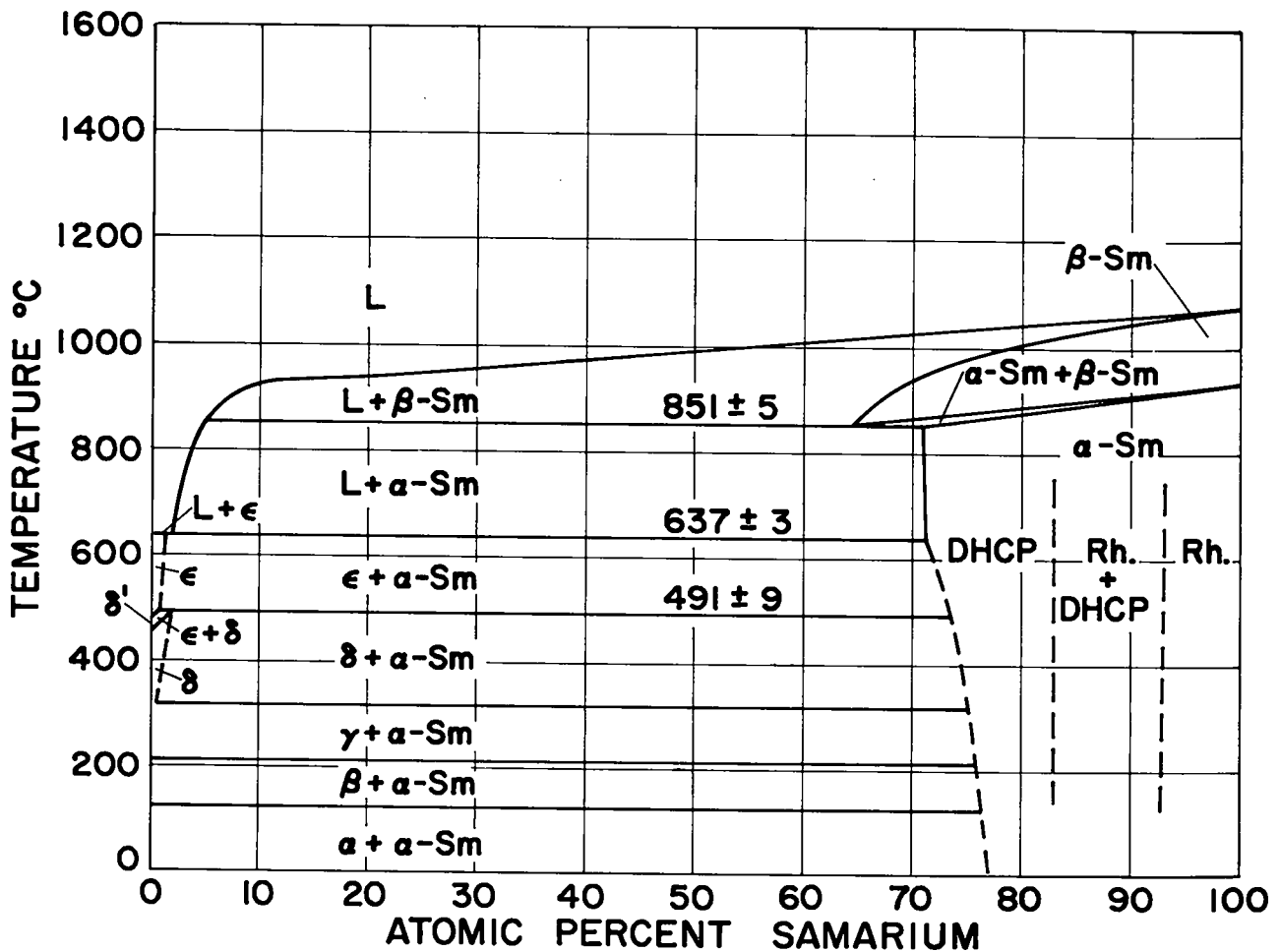


Fig. 38. Plutonium-samarium phase diagram.



Pu-Sn □

Plutonium-Tin

Two intermediate phases, Pu₃Sn and PuSn₃, in the system have been identified by [1-3] by x-ray and microstructural studies of a few alloys (see Table XLVII). [4] reports that terminal solid solubilities are low, approaching zero, and that a eutectic is indicated at about 13 at.% Sn. Solubility data determined by [5] for Pu in liquid Sn are given in Table XLVIII. Also [5] found the melting point of PuSn₃ to be 1198 ± 5°C.

[1] E. M. Cramer, Los Alamos Scientific Laboratory. unpublished data.

[2] F. H. Ellinger. "A Review of the Interme-

tallic Compounds of Plutonium." Chap. XXV in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.). p. 303, The University of Chicago Press. Chicago (1961).

[3] F. H. Ellinger. Los Alamos Scientific Laboratory. unpublished work.

[4] F. W. Schonfeld, "Plutonium Phase Diagrams Studied at Los Alamos," Chap. XXII in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.). p. 250. The University of Chicago Press. Chicago (1961).

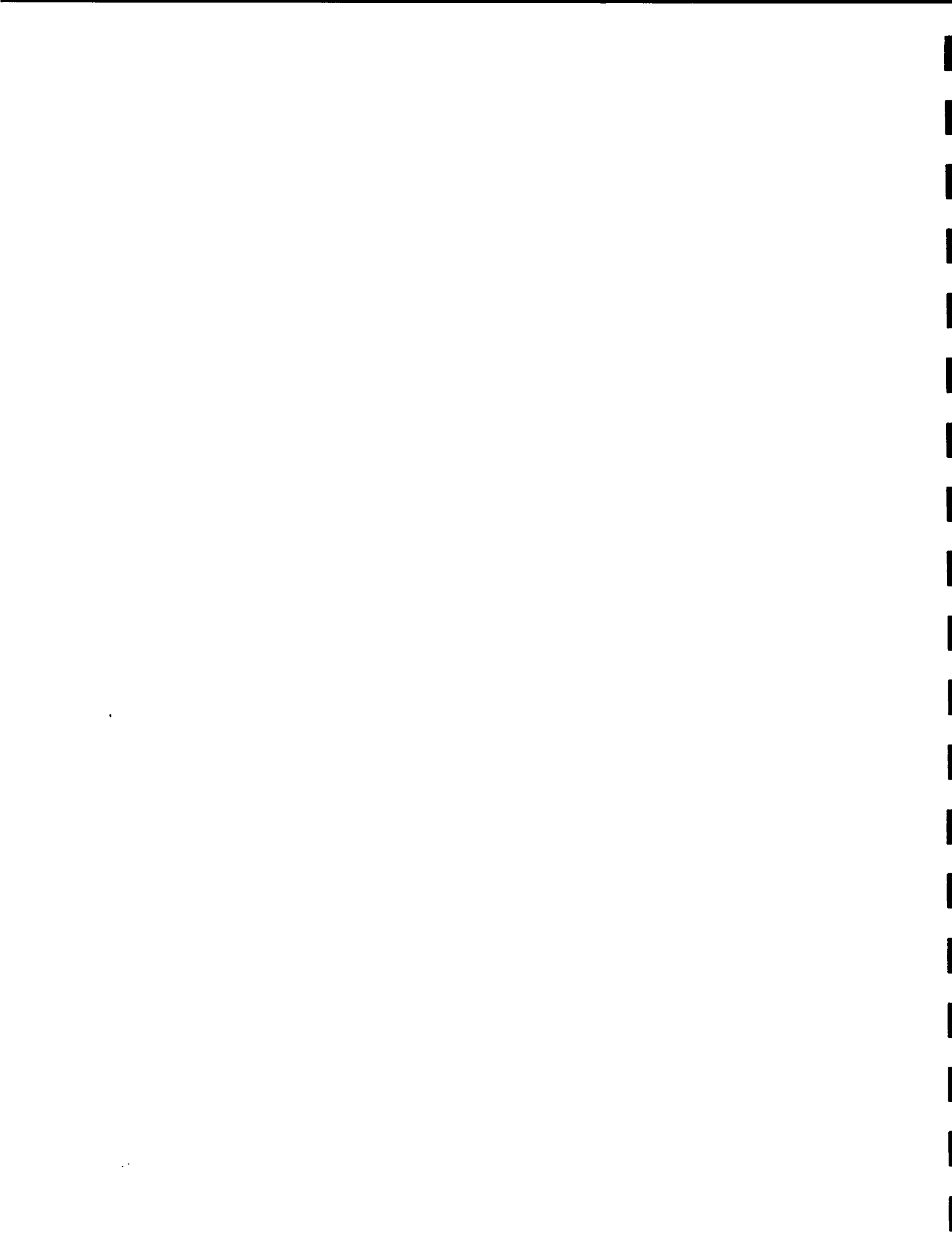
[5] J. W. Ward and R. N. R. Mulford, "Solubility of Plutonium in Liquid Tin," *J. Nucl. Mater.*, **12(3)**: 335-336 (1964).

TABLE XLVII. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-TIN ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
Pu ₃ Sn	AuCu ₃ (?)	face-centered cubic		4.680					[3]
PuSn ₃	AuCu ₃	primitive cubic	Pm3m	4.630			1	9.96	[2]

TABLE XLVIII. SOLUBILITY OF PLUTONIUM IN LIQUID TIN

Temperature, °C	Solubility, at. % Pu
400	0.0032
499	0.0198
601	0.0979
698	0.290
800	0.905
903	2.06
1001	4.41
1102	8.21



Pu-Sr □ **Plutonium-Strontium**

See Pu-Ba (Plutonium-Barium).

Pu-Ta □ **Plutonium-Tantalum**

The solubility of Ta in liquid Pu at several temperatures between 750 and 950°C has been determined by [1] and is given in Table XLIX. At 2000°C, about 16 at.% Ta is soluble in Pu according to the work of [2] and [3], and [4] has found the solubility at 1000°C to be of the order of 0.5 at.% Ta. The Pu-Ta phase diagram is shown in Fig. 39.

and Rhenium in Liquid Plutonium." *J. Nucl. Mater.*, **21(2)**: 219-224 (1967); also, *Am. Nucl. Soc. Trans.*, **10(1)**: 106 (1967).

[1] D. F. Bowersox and J. A. Leary, "The Solubilities of Carbon, Tantalum, Tungsten

[2] E. M. Cramer. Los Alamos Scientific Laboratory. unpublished work.

[3] V. O. Struebing, Los Alamos Scientific Laboratory, unpublished work.

[4] J. R. Hancock, Los Alamos Scientific Laboratory, unpublished work.

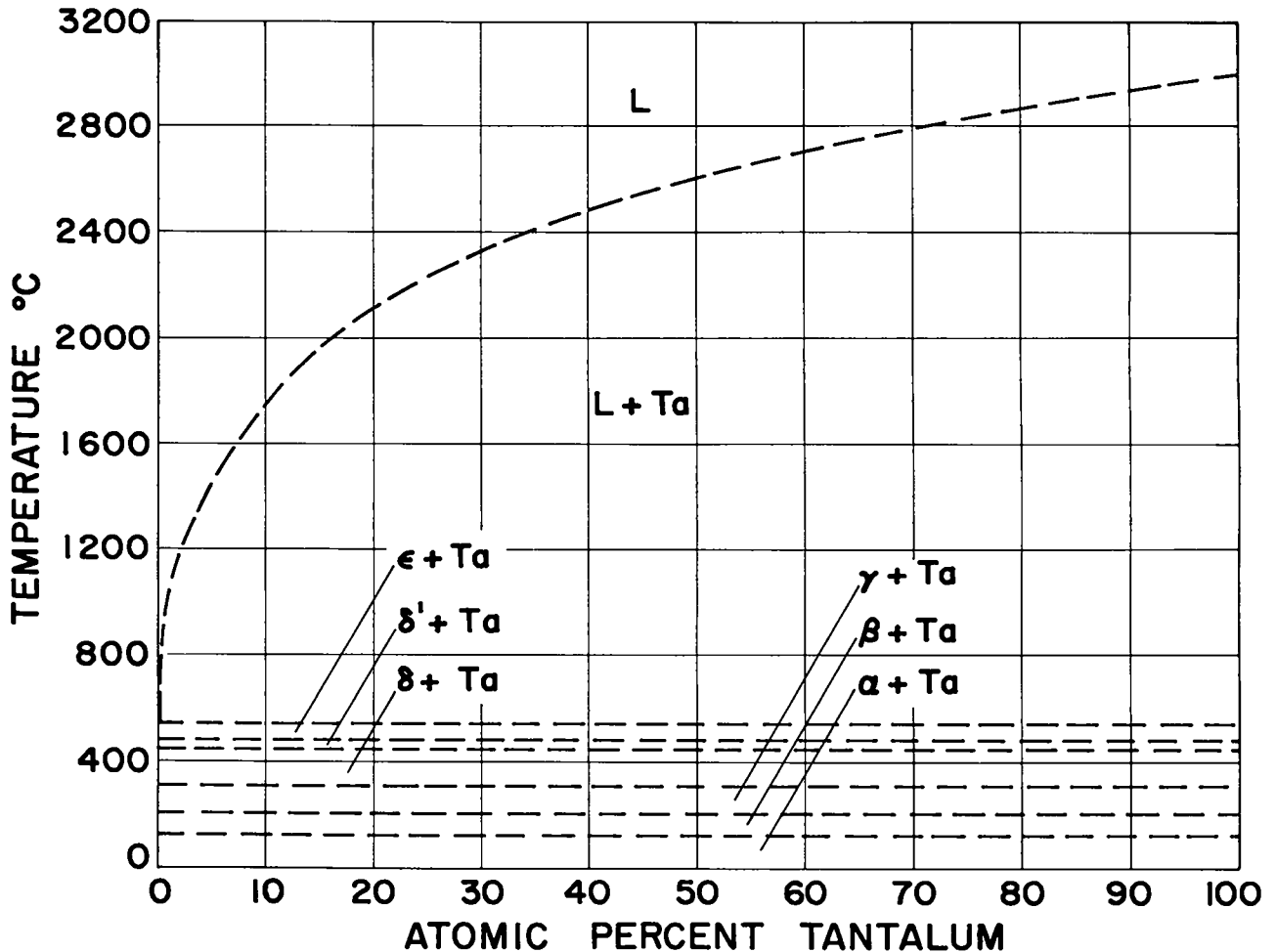


Fig. 39. Plutonium-tantalum phase diagram.

TABLE XLIX. SOLUBILITY OF TANTALUM IN LIQUID PLUTONIUM

<u>Temperature. °C</u>	<u>Solubility. at. % Ta</u>
750	0.085 ± 0.002
800	0.119 ± 0.002
850	0.190 ± 0.008
900	0.267 ± 0.010
950	0.352 ± 0.013

Pu-Tb **Plutonium-Terbium**

See Pu-Gd (Plutonium-Gadolinium).

Pu-Te **Plutonium-Tellurium**

components in vacuum and in helium.

One intermediate phase, PuTe, is known in the system (see Table L). It has been prepared by [1] by induction melting of the elemental

[1] A. E. Gorum. "The Crystal Structures of PuAs, PuTe, PuP, and PuOSe." *Acta Cryst.*, **10**: 144 (1957).

TABLE L. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-TELLURIUM ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
PuTe	NaCl	face-centered cubic	Fm3m	6.183			4	10.33	[1]



Pu-Th □ **Plutonium-Thorium**

The phase diagram shown in Fig. 40 was constructed from the combined results of [1], [2], and [3]. The β -Th solid solution field described by [2] has not been reported by [1] or [3], however, and thus is not shown. Therefore, the α Th/(L + α Th) solidus of [1] rather than the β Th/(L + β Th) solidus of [2] has been drawn in the diagram. The liquidus on the Th-rich side of the ϵ Pu/ ζ eutectic point is based on the work of [2], but according to the microstructural results of [1] the solubility limit of Th in

liquid Pu appears to be no more than 10 at.% Th at 825°C. The maximum solubility of Th in ϵ Pu has been reported as 5.6 at.% by [2] and as 5 at.% by [1] and by [3], at the eutectic temperature. The composition of the single Pu-Th intermediate phase, ζ , (see Table LI) in the system has been bracketed between 30 and 33 at.% Th by [1] and [4]. Under equilibrium conditions, ζ is formed by the peritectic reaction, L + α Th \rightleftharpoons ζ . Under normal conditions, however, the peritectic reaction is suppressed on cooling, and ζ is formed by a nonequilibrium peritectoid reaction subsequent to eutectic solidification. The solubility limit of Pu in α Th has also been determined by [5].

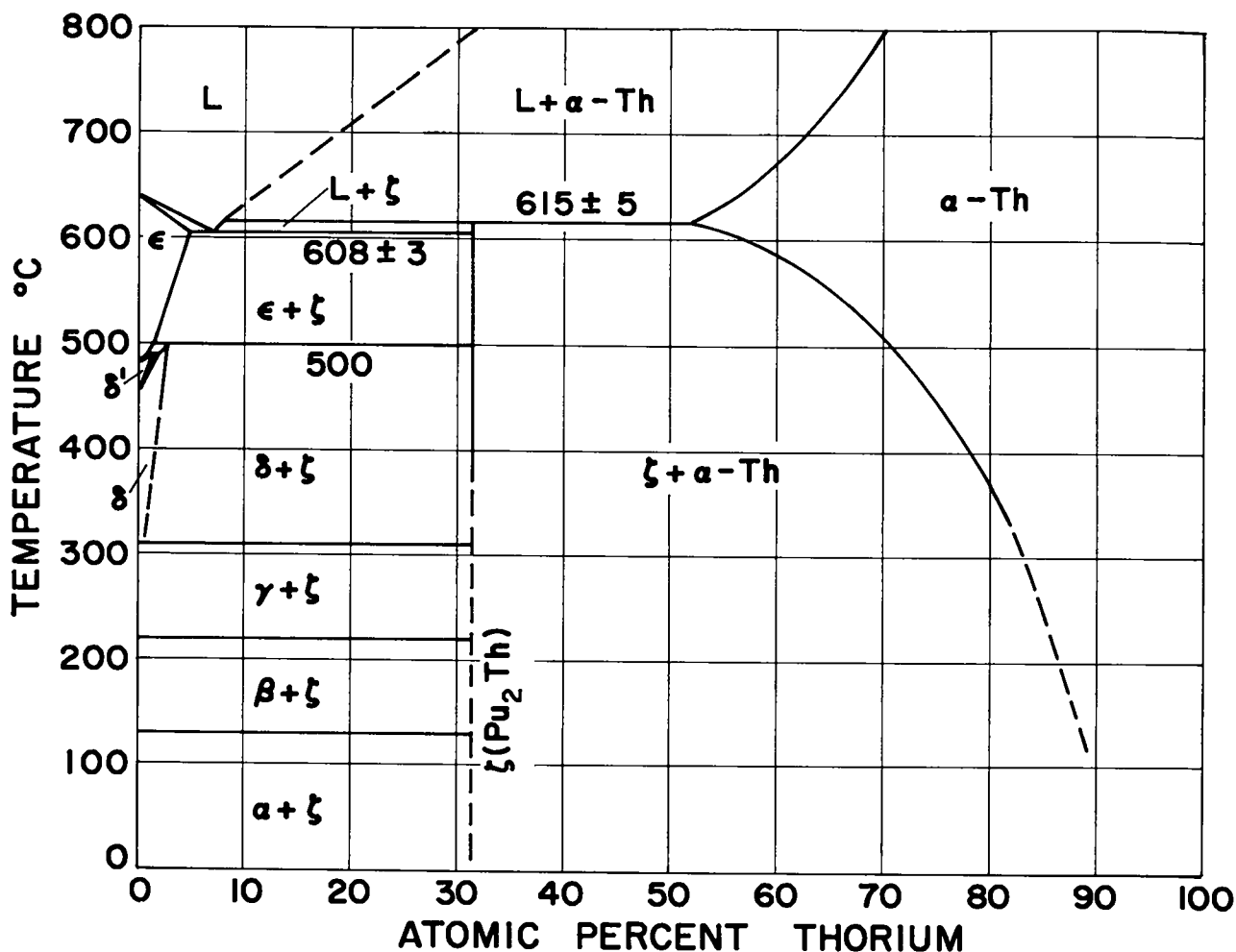


Fig. 40. Plutonium-thorium phase diagram. (Courtesy Ellinger, Land, and Gschneidner and by permission of Gordon and Breach, Science Publishers, Inc., from *Plutonium Handbook*, Vol. 1, p. 221, 1967.)

Their results are in agreement with those of [1] and [3] from 600 to 500°C. Between 500 and 400°C, however, [5] found the solubility limit to remain constant at 29.6 at.% Pu, instead of decreasing with decreasing temperature as shown in the diagram.

- [1] D. M. Poole, G. K. Williamson, and J. A. C. Marples, "A Preliminary Investigation of the Plutonium-Thorium System," *J. Inst. Metals*, **86**: 172-176 (1957-58).
- [2] A. A. Bochvar, S. T. Konobeevsky, V. I. Kutaitsev, T. S. Menshikova, and N. T. Chebotarev, "Interaction Between Plutonium and Other Metals in Connection with Their Arrangement in Mendeleev's Periodic Table." *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva*, 1958, Vol. 6, pp. 191-192. United Nations, Geneva (1958).
- [3] F. W. Schonfeld. "Plutonium Phase Diagrams Studied at Los Alamos," Chap. XXII in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), p. 262-263. The University of Chicago Press, Chicago (1961).
- [4] K. Imlah. Los Alamos Scientific Laboratory. unpublished work.
- [5] D. Calais, M. Dupuy, M. Mouchnino, A. Y. Portnoff, and A. Van Craeynest. "Diffusion of Plutonium in the Solid State," in *Plutonium 1965*, A. E. Kay and M. B. Waldron (Eds.), p. 387. Chapman and Hall, London (1967).

TABLE LI. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-THORIUM ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
Pu ₂ Th ζ		orthorhombic(?)		9.820	8.164	6.681	6		[1]
				7.90	8.43	9.79	8		[2]

Pu-Ti □ **Plutonium-Titanium**

A complete diagram of this system has been published by [1], tentative results have been reported by [2], and details of the phase equilibria involving δ' Pu have been determined by [3]. The diagram shown in Fig. 41 is based on the results of these three groups of investigators. With a few exceptions, the diagram of [1] is in satisfactory

agreement with the work of [2] and with the diagram shown here. These points of difference, taken from the work of [2], include the following: The peritectic temperature, ϵ Pu \rightleftharpoons L + β Ti, is placed at 730°C; the solubility of Ti in ϵ Pu is given as 14 at.% Ti, and of Pu in β Ti as 12 at.% Pu, at the peritectic temperature; and the eutectoid reaction, β Ti \rightleftharpoons α Ti + ϵ Pu, is placed at 6 at.% Pu and 580°C. Solubility data determined by [4] for Ti in liquid Pu are given in Table LII.

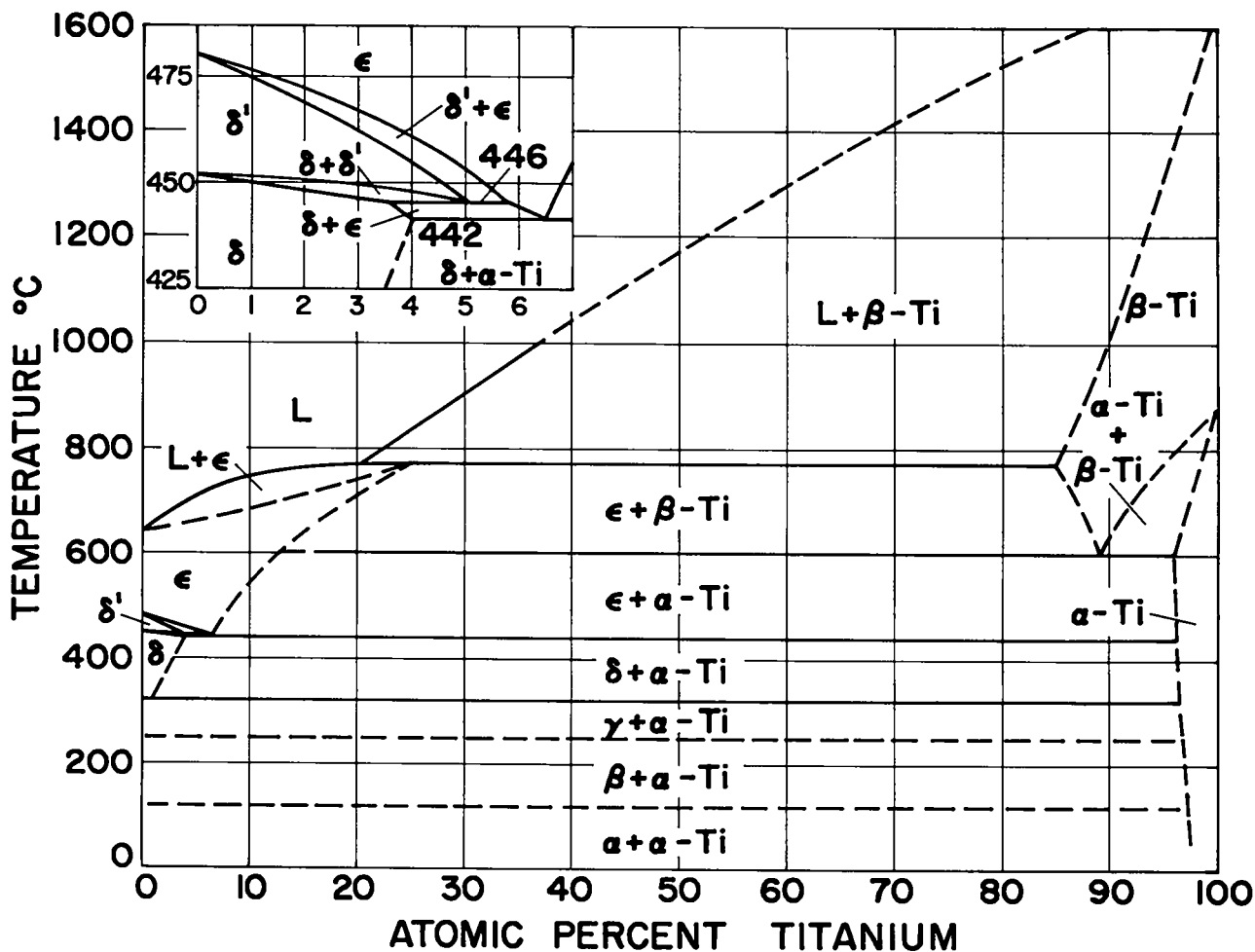


Fig. 41. Plutonium-titanium phase diagram.

- [1] V. I. Kutaitsev, N. T. Chebotarev, I. G. Lebedev, M. A. Andrianov, V. N. Konev, and T. S. Menshikova, "Phase Diagrams of Plutonium with the Metals of Groups IIA, IVA, VIIIA and IB," in *Plutonium* 1965, A. E. Kay and M. B. Waldron (Eds.), pp. 420-449, Chapman and Hall, London (1967).
- [2] D. M. Poole, M. G. Bale, P. G. Mardon, J. A. C. Marples, and J. L. Nichols. "Phase Diagrams of Some Plutonium Alloy Systems," in *Plutonium* 1960, E. Grison, W. B. H. Lord, and R. D. Fowler (Eds.), pp. 277-278, Cleaver-Hume Press, Ltd., London (1961).
- [3] R. O. Elliott and A. C. Larson. "Delta-Prime Plutonium." Chap. XXIV in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), pp. 265-280, The University of Chicago Press, Chicago (1961).
- [4] D. F. Bowersox, Los Alamos Scientific Laboratory, to be published.

TABLE LII. SOLUBILITY OF TITANIUM IN LIQUID PLUTONIUM

Temperature, °C	Solubility, at. % Ti
700	5.44 ± 0.10
750	9.72 ± 0.20
800	22.50 ± 1.0
850	26.07 ± 0.5
900	28.81 ± 0.6
950	33.31 ± 0.6
1000	37.17 ± 1.0

Pu-Tl □

Plutonium-Thallium

Examination of a few Pu-rich alloys by [1-3] has revealed that the solubility of Tl in δ Pu is less than 1 at.% and that the most Pu-rich intermediate phase is Pu_3Tl . Crystal structure data (see Table LIII) have been reported by [4] for Pu_3Tl and PuTl_3 .

[1] F. H. Ellinger, Los Alamos Scientific Laboratory, unpublished work.
 [2] C. C. Land, Los Alamos Scientific Laboratory, unpublished work.

tory, unpublished work.

[3] K. Imlah, Los Alamos Scientific Laboratory, unpublished work.
 [4] A. A. Bochvar, S. T. Konobeevsky, V. I. Kutaitsev, T. S. Menshikova, and N. T. Chebotarev. "Interaction Between Plutonium and Other Metals in Connection with Their Arrangement in Mendeleev's Periodic Table," *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958*, Vol. 6, pp. 186-187, United Nations, Geneva (1958).

TABLE LIII. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-THALLIUM ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
Pu_3Tl	AuCu_3	primitive cubic		4.723				14.5	[4]
PuTl_3	Disordered Mg	hexagonal		3.458	5.519			12.4	[4]



Pu-Tm **Plutonium-Thulium**

See Pu-Gd (Plutonium-Gadolinium).

The solubility of Tm in liquid Pu between 700 and 1000°C, as determined by [1], is given

in Table LIV.

- [1] D. F. Bowersox and J. A. Leary, "The Solubilities of Selected Elements in Liquid Plutonium: X. Thulium," USAEC Report LA-3623, Los Alamos Scientific Laboratory (Nov. 4, 1966).

TABLE LIV. SOLUBILITY OF THULIUM IN LIQUID PLUTONIUM

<u>Temperature, °C</u>	<u>Solubility, at. % Tm</u>
700	2.35 ± 0.06
750	3.08 ± 0.04
800	4.09 ± 0.18
850	5.04 ± 0.14
900	6.79 ± 0.10
950	7.25 ± 0.07
1000	8.48 ± 0.21



Pu-U □

Plutonium-Uranium

Complete phase diagrams for this system have been reported by [1-3]. All are in agreement as to the general form of the diagram but some of the phase boundaries determined by [3] are quite different from the corresponding boundaries determined by [1] and by [2]. The diagram shown in Fig. 42 is based on the thermal, dilatometric, x-ray, and microstructural studies of [1] and [2]. A major disagreement concerns the β -Pu field, which [3] found to extend from about 4 at.% U in the vicinity of 120°C to roughly 17 at.% U at about 300°C. The data of [1], on the other hand, show the β -Pu field extending from

essentially zero at.% U at 125°C to about 2 at.% U at 280°C. These latter results appear to be confirmed by the work of [2]. Also, [3] found the ζ field to extend to higher U contents from about 6 at.% U at 120°C, whereas [1] found the corresponding point to be at about 25 at.% U and 278°C. There is better agreement among the three groups of investigators concerning the U-rich portion of the diagram. Work by [4] places the α U/(α U + ζ) boundary at about 85 at.% U at 400°C, which agrees reasonably well with the results of [1] and [2] as well as with revised work of [3] as mentioned by [5]. Also, the solubility data of [6] between 410 and 546°C fall within ~ 1 at.% of the boundaries established by [1] and [2] for the ζ + α U field. Crystal structure data are given in Table LV.

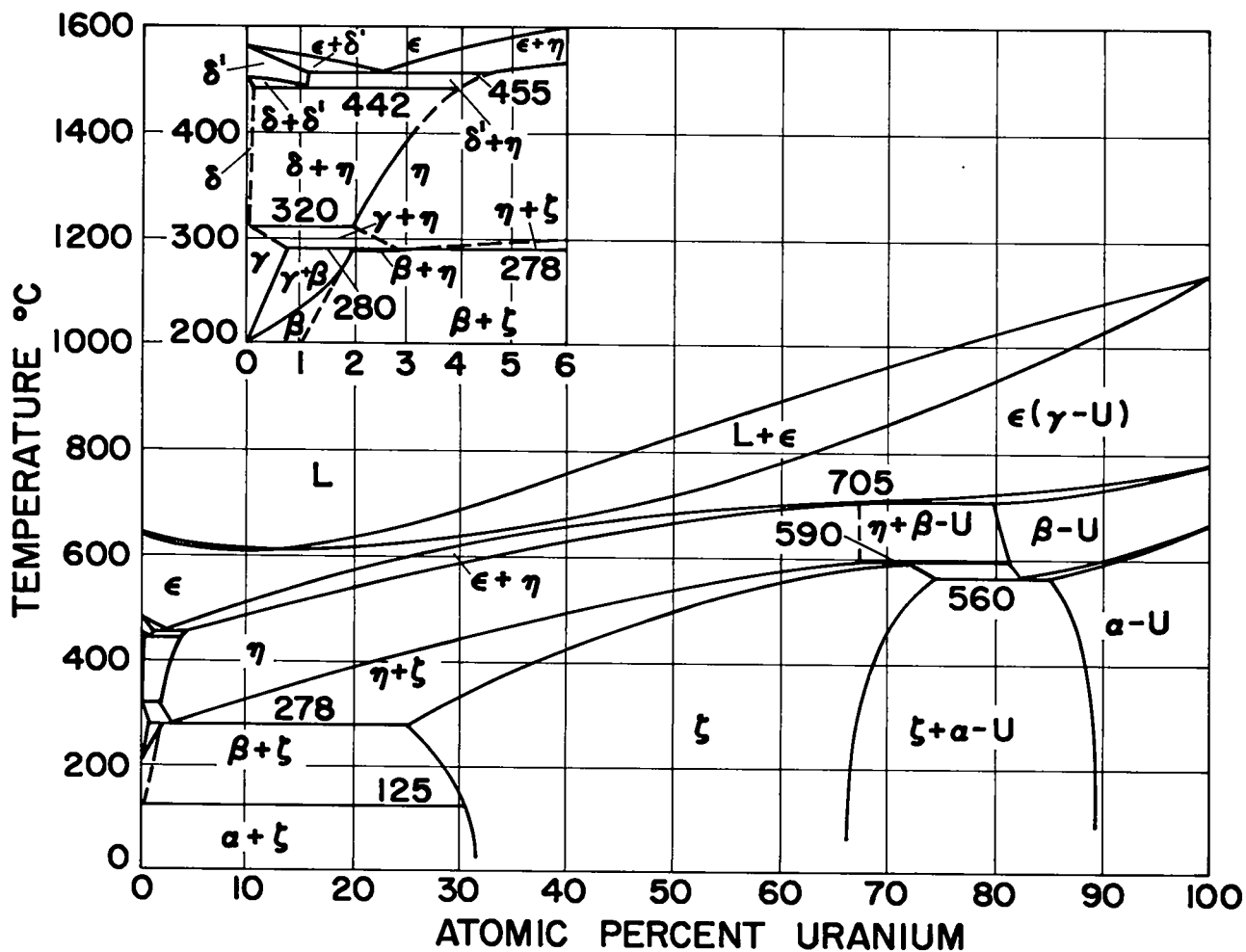


Fig. 42. Plutonium-uranium phase diagram.

- [1] F. H. Ellinger, R. O. Elliott, and E. M. Cramer, "The Plutonium-Uranium System." *J. Nucl. Mater.*, **1**: 233-243 (1959).
- [2] M. B. Waldron, "Phase Diagrams of Plutonium Alloys Studied at Harwell," Chap. XXI in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), pp. 233-234, The University of Chicago Press, Chicago (1961).
- [3] A. A. Bochvar, S. T. Konobeevsky, V. I. Kutaitsev, T. S. Menshikova, and N. T. Chebotarev, "Interaction Between Plutonium and Other Metals in Connection with Their Arrangement in Mendeleev's Periodic Table," *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958*, Vol. 6, pp. 191-193, United Nations, Geneva (1958).
- [4] A. F. Berndt, "Room Temperature Lattice Constants of Alloys of Plutonium in Alpha-Uranium," *J. Nucl. Mater.*, **9**(1): 53-58 (1963).
- [5] F. W. Schonfeld, "Plutonium Phase Diagrams Published by the Russians," Chap. XXIII in *The Metal Plutonium*, A. S. Coffinberry and W. N. Miner (Eds.), p. 263, The University of Chicago Press, Chicago (1961).
- [6] D. Calais, M. Dupuy, M. Mouchnino, A. Y. Portnoff, and A. Van Craeynest, "Diffusion of Plutonium in the Solid State," in *Plutonium 1965*, A. E. Kay and M. B. Waldron (Eds.), p. 379. Chapman and Hall, London (1967).

TABLE LV. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-URANIUM ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
Pu-U ξ		cubic (at room temp.)	P	10.692 (35 at.% U)			58 atoms	18.95	[1]
				10.651 (70 at.% U)					
Pu-U η		tetragonal (?)		10.57	10.76		52 atoms	17.2	[1]
				10.73	10.44		56 atoms		

Pu-V □

Plutonium-Vanadium

The phase diagram (see Fig. 43) is the simple eutectic type. According to [1], the ϵ Pu/V eutectic composition is less than 2 at.% V and the eutectic temperature is 625°C. Negligibly small solid solubility of V in the Pu allotropes is indicated by the absence of any effect of V additions on the transformation temperatures of the allotropes. The average solubility of V in liquid Pu at selected temperatures between 700 and 1000°C, as determined by [2], is given in Table LVI.

[1] S. T. Konobeevsky, "Equilibrium Diagrams of Certain Systems of Plutonium," Session on the Peaceful Uses of Atomic Energy. Section on Chemical Sciences, III: pp. 362-374, USSR Academy of Sciences. Moscow (1955).

[2] D. F. Bowersox and J. A. Leary, "The Solubilities of Selected Elements in Liquid Plutonium: IX. Vanadium," USAEC Report LA-3594, Los Alamos Scientific Laboratory, (Sept. 15, 1966); also *Am. Nucl. Soc. Trans.*, **10**(1): 106 (1967).

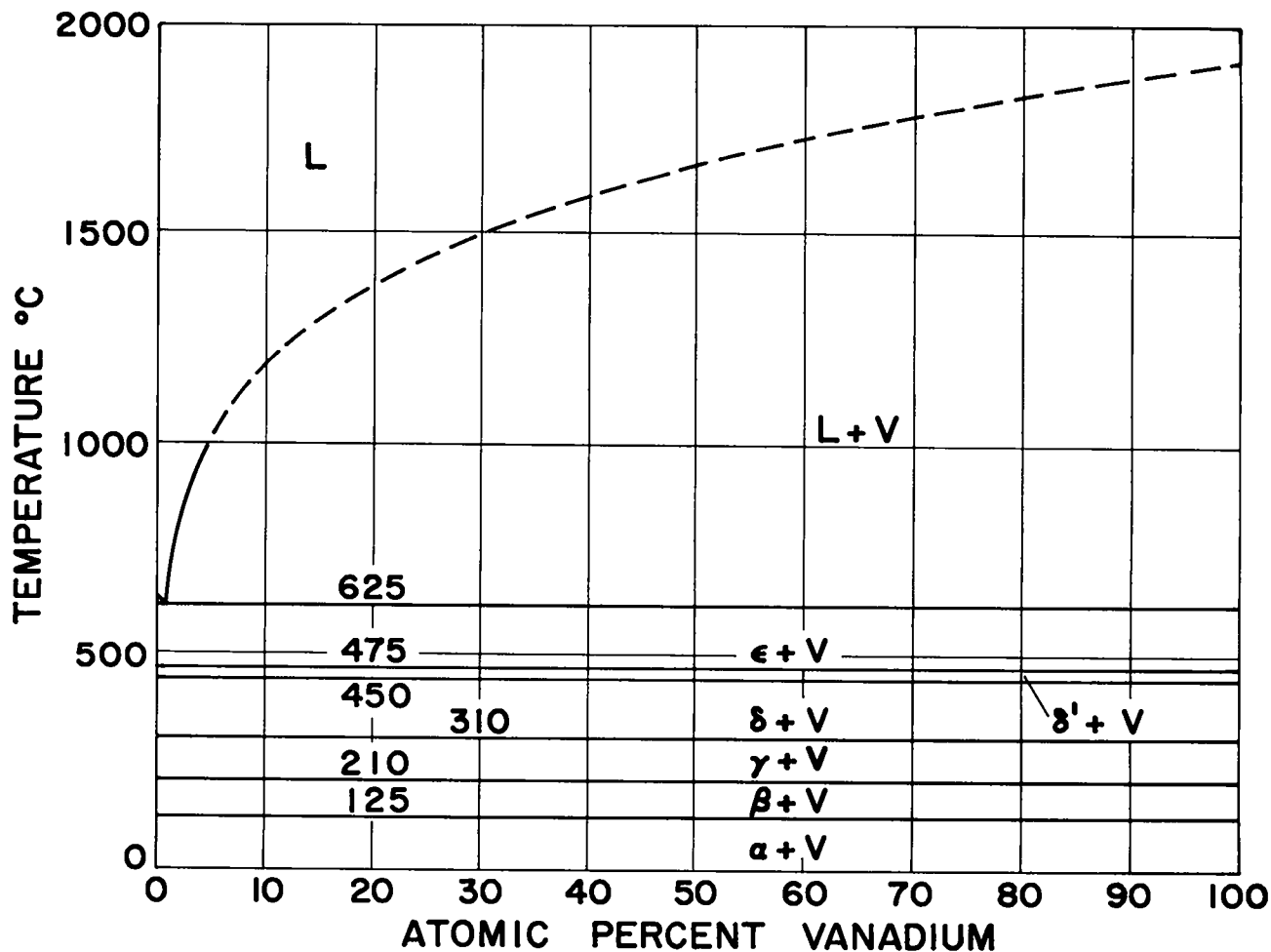


Fig. 43. Plutonium-vanadium phase diagram.

TABLE LVI. SOLUBILITY OF VANADIUM
IN LIQUID PLUTONIUM

<u>Temperature, °C</u>	<u>Solubility, at. % V</u>
700	1.02 ± 0.05
750	1.34 ± 0.09
800	1.67 ± 0.05
850	2.29 ± 0.05
900	2.96 ± 0.14
950	3.78 ± 0.09
1000	4.46 ± 0.09

Pu-W □

Plutonium-Tungsten

Plutonium that had been heated to 1750°C in the presence of an excess of W was found by [1] to contain primary dendrites of W in a matrix of α Pu. On the other hand, [2] have estimated, by lineal analysis, the minor phase in the microstructure of an alloy containing 1.3 wt% W to be Pu_2W_3 . The solubility of W in liquid Pu at several temperatures between 700 and 950°C, as determined by [3], is given in Table LVII. The phase diagram is shown in Fig. 44.

- [1] E. M. Cramer, Los Alamos Scientific Laboratory, unpublished work.
- [2] R. G. Cope, J. N. Lowe, and D. C. Miller, "Studies of Alloys of Plutonium with Transition Elements and Gold," UKAEA Report AWRE 0-36/61 (1961).
- [3] D. F. Bowersox and J. A. Leary, "The Solubilities of Carbon, Tantalum, Tungsten, and Rhenium in Liquid Plutonium," *J. Nucl. Mater.*, **21**(2): 219-244 (1967).

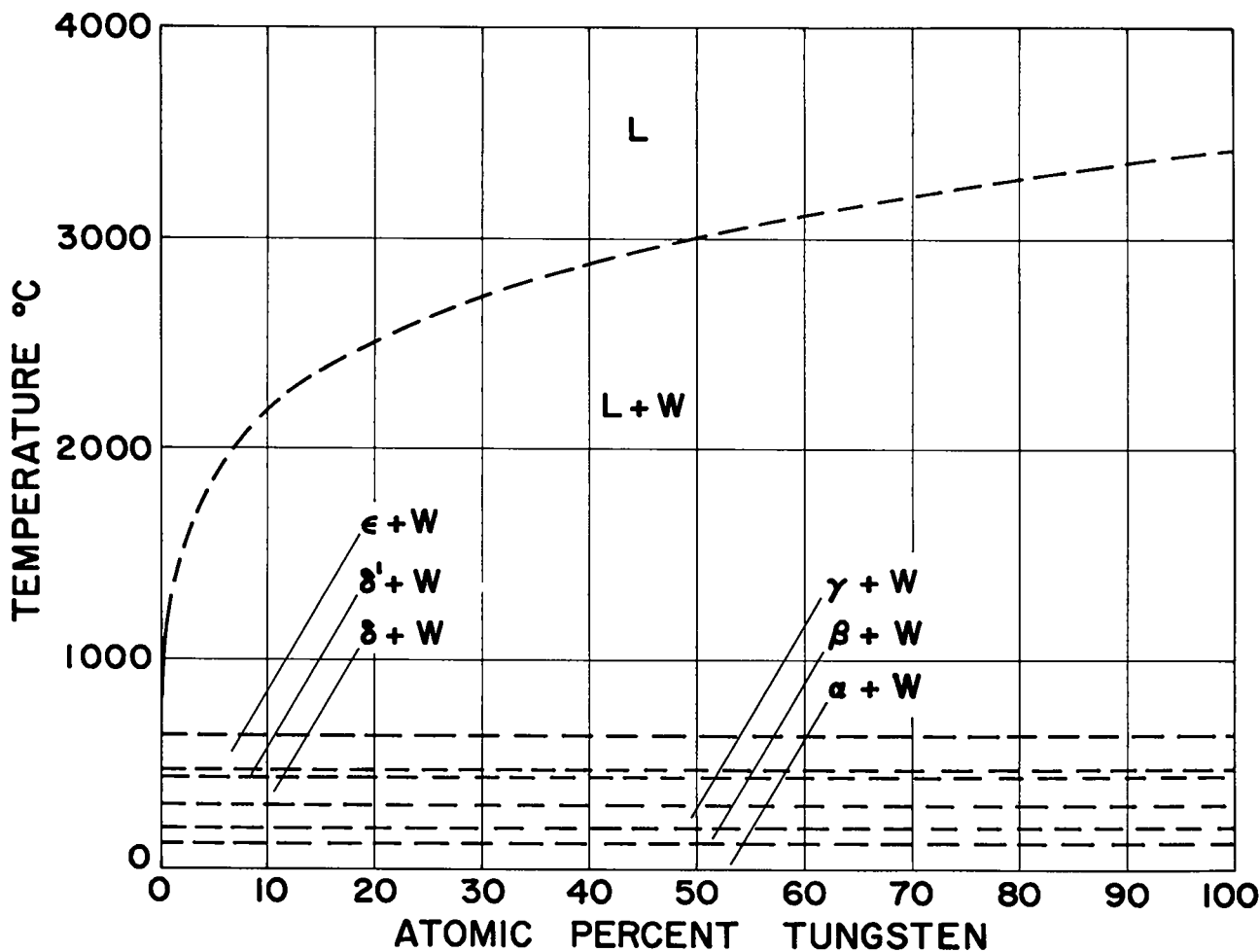


Fig. 44. Plutonium-tungsten phase diagram.

TABLE LVII. SOLUBILITY OF TUNGSTEN
IN LIQUID PLUTONIUM

<u>Temperature, °C</u>	<u>Solubility, at. % W</u>
700	0.0038 ± 0.0001
750	0.0072 ± 0.0003
800	0.0105 ± 0.0002
850	0.0159 ± 0.0005
900	0.0255 ± 0.0011
950	0.0379 ± 0.0002

Pu-Y □

Plutonium-Yttrium

The diagram of Fig. 45 as reported by [1] is based on thermal, dilatometric, and x-ray dif-

fraction data. The significant features of the diagram have been confirmed by [2] and [3]. The placing of the β Y field by [1] appears to be largely conjectural. The maximum solubility of Pu in α Y is given as 15 at.% at 820°C. 20 at.%

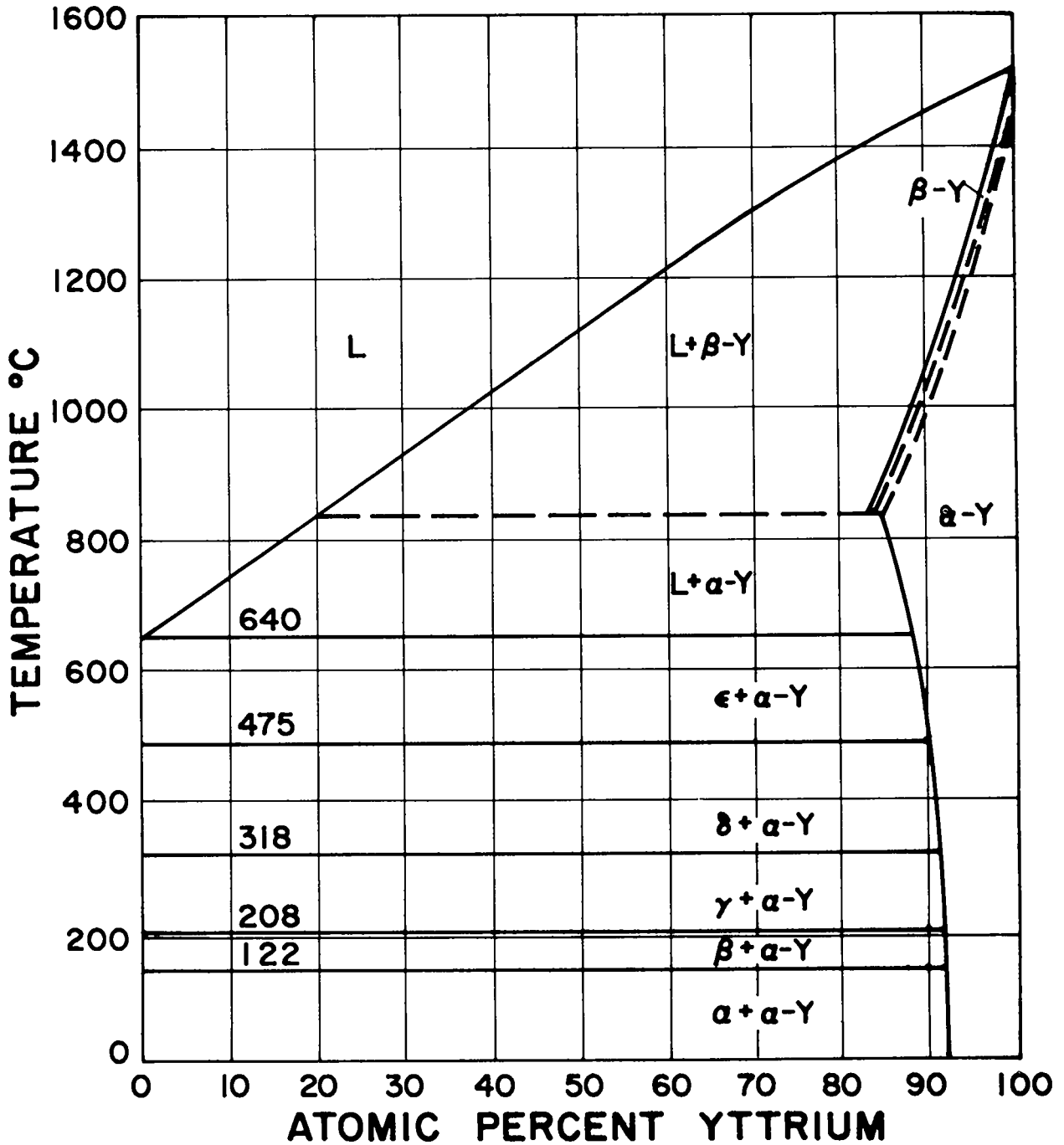


Fig. 45. Plutonium-yttrium phase diagram. (Redrawn from Ref. 1.)

at 640°C and 17 at.% at 635°C by [1], [2], and [3], respectively.

- [1] V. I. Kutaitsev, N. T. Chebotarev, I. G. Lebedev, M. A. Andrianov, V. N. Konev, and T. S. Menshikova. "Phase Diagrams of Plutonium with the Metals of Groups IIA, IVA, VIIB and IB," in *Plutonium 1965*. A. E. Kay and M. B. Waldron (Eds.), pp. 420-449, Chapman and Hall. London (1967).
- [2] L. J. Wittenberg and G. R. Grove, "Reactor Fuels and Materials Development Plutonium Research: 1963 Annual Report." USAEC Report MLM-1184. pp. 12-16. Mound Laboratory (Dec. 31. 1963).
- [3] F. H. Ellinger. Los Alamos Scientific Laboratory, unpublished work as of June 1967.

Pu-Yb **Plutonium-Ytterbium**

See Pu-Eu (Plutonium-Europium).

Pu-Zn **Plutonium-Zinc**

The phase diagram given in Fig. 46 is based on information drawn from several sources. The microstructural, x-ray, and thermal studies of [1] provide the data in the composition region from 0 to 65 at.% Zn; the high-temperature results of [2] are shown in the region from 65 to 78 at.% Zn; and the microstructural, x-ray, and thermal data of [3] and the solubility limits of Pu in liquid Zn between 450 and 735°C, as determined by [4], are shown in the region from 78 to 100

at.% Zn. All the known intermediate phases are Zn-rich (see Table LVIII). [1] and [2] reported the existence of PuZn_2 , Pu_2Zn_9 , PuZn_8 , and $\text{Pu}_2\text{Zn}_{17}$. In later work, however, [3] found the PuZn_8 phase to be $\text{Pu}_2\text{Zn}_{17}$, as proposed by [5], and reported the existence of a new phase, tentatively $\text{PuZn}_{7.7}$, but failed to find any evidence for the PuZn_{12} phase postulated by [5]. Subsequently, the crystal structure work of [6] showed $\text{PuZn}_{7.7}$ to be $\text{Pu}_3\text{Zn}_{22}$. [3] have also identified three additional phases structurally related to $\text{Pu}_2\text{Zn}_{17}$ ($\text{Th}_2\text{Zn}_{17}$, rhombohedral type). These are all hexagonal, space groups $P6_322$, $P6_3/mmc$,

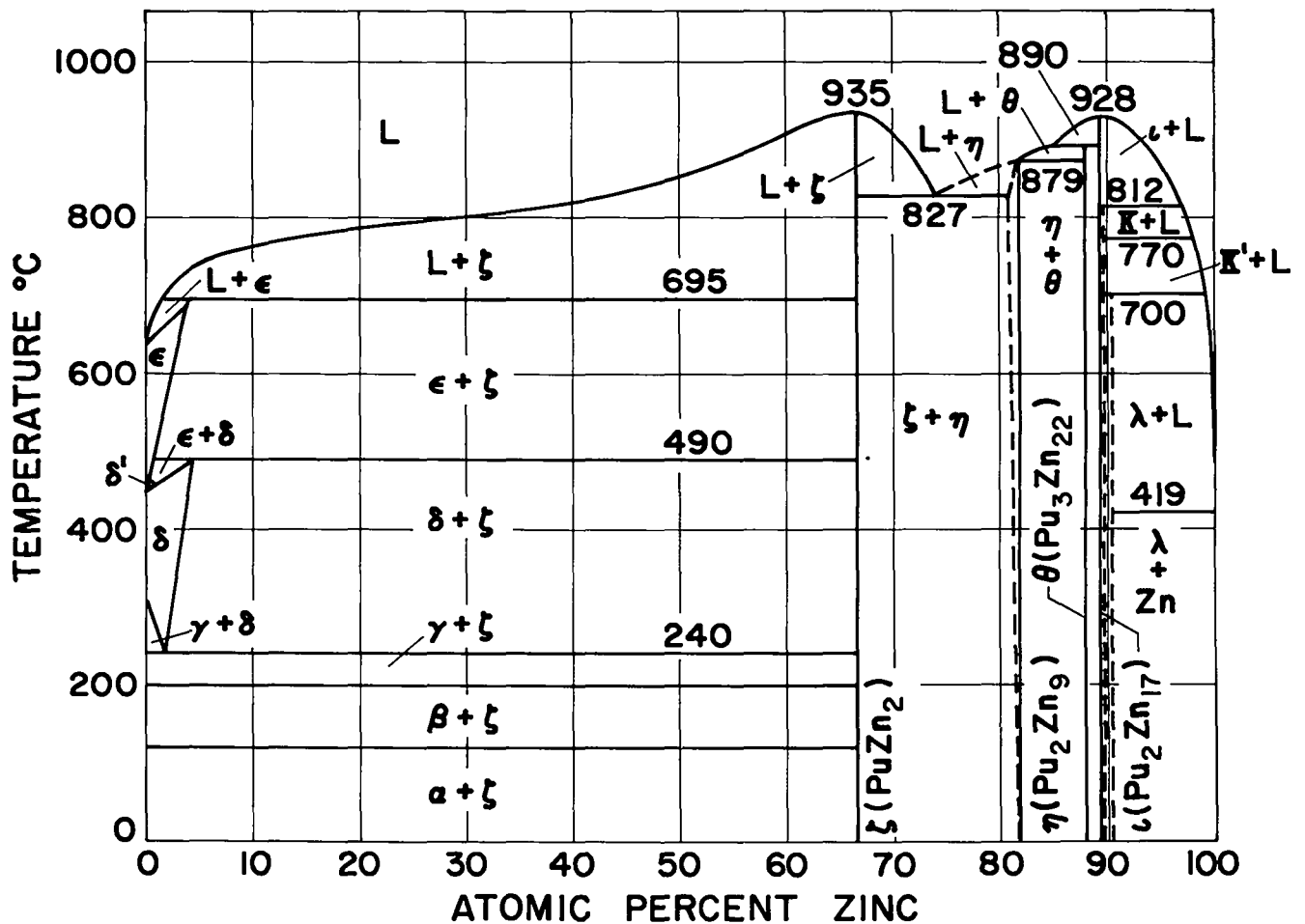


Fig. 46. Plutonium-zinc phase diagram.

and P6/mmm. and occur on the Zn-rich side of the rhombohedral $\text{Pu}_2\text{Zn}_{17}$. One of the three appears to be of the $\text{Th}_2\text{Ni}_{17}$ type, the other two are closely related to that type. Crystal structure data for Pu_2Zn_9 have been reported by [7].

- [1] E. M. Cramer, F. H. Ellinger, and C. C. Land, "Plutonium-Zinc Phase Diagram." in *Extractive and Physical Metallurgy of Plutonium and Its Alloys*, W. D. Wilkinson (Ed.), pp. 169-180. Interscience Publishers, New York (1960).
- [2] E. D. Albrecht, "The Plutonium-Zinc System." *J. Nucl. Mater.*, **12(2)**: 125-130 (1964).
- [3] E. M. Cramer and D. H. Wood, "Phase Relations in the Zinc-Rich Portion of the Plutonium-Zinc System." *J. Less-Common Metals*, **13(1)**: 112-121, (1967).
- [4] G. R. B. Elliott and R. U. Sweezer, "Study of the System Plutonium-Uranium-Zinc. Chemical Engineering Division Summary Report, July, August, and September, 1957." USAEC Report ANL-5789, pp. 89-95. Argonne National Laboratory (1957). (Classified).
- [5] I. Johnson and M. G. Chasanov, "Thermodynamics of the Plutonium-Zinc System," *J. Inorg. Nucl. Chem.*, **26**: 2059-2067 (1964).
- [6] Q. C. Johnson, D. H. Wood, and G. S. Smith, "The Crystal Structure of $\text{Pu}_3\text{Zn}_{22}$," USAEC Report UCRL-70500. Lawrence Radiation Laboratory, Livermore, California (1967).
- [7] A. C. Larson and D. T. Cromer, "The Crystal Structure of Pu_2Zn_9 ," *Acta Cryst.*, **23(1)**: 70-77 (1967).

TABLE LVIII. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-ZINC ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
PuZn_2 ζ	Cu_2Mg	face-centered cubic	Fd3m	7.760 (Pu-rich) 7.747 (Zn-rich)			8	10.5	[1]
$\text{Pu}_2\text{Zn}_{10}$ η		hexagonal	probably $P6_3/mmc$	28.86	14.14			9.05	[7]
$\text{Pu}_3\text{Zn}_{22}$ θ		body-centered tetragonal	$I4_1/amd$	8.85	21.18		4	8.71	[6]
$\text{Pu}_2\text{Zn}_{17}$ ι	$\text{Th}_2\text{Zn}_{17}$	rhombohedral (in hexagonal system)	R3m	8.95		13.1			[3]
" $\text{Pu}_2\text{Zn}_{17}$ " κ		hexagonal	P6/mmm	8.9	17.7				[3]
" $\text{Pu}_2\text{Zn}_{17}$ " " $\text{Th}_2\text{Ni}_{17}$ " κ'		hexagonal	$P6_3/mmc$	8.98	8.85				[3]
" $\text{Pu}_2\text{Zn}_{17}$ " λ		hexagonal	$P6_322$	8.98	8.85				[3]

Pu-Zr □
Plutonium-Zirconium

Complete diagrams, which differ in several details, have been published by [1] and [2], additional work on Pu-rich alloys has been reported by [3] and [4]. Alloys containing between 30 and 60 at.% Pu have been studied by [5]. and solubility data for Zr in liquid Pu between 700 and 950°C have been obtained by [6] (see Table LIX). The diagram given in Fig. 47 is largely that of [2] but has been modified to show the ζ phase ($\sim\text{Pu}_{10}\text{Zr}$) as found by [3] and the solubility data of [6]. The solidus curve determined by [1] is consistently higher than that of [2], being as much as 100°C higher at 50 at.% Zr. There is better agreement between their

liquidus curves. ζ , which has a complex crystal structure, was found by [4] to exist between ~ 3 and 5 at.% Zr and to decompose to β and θ at $\sim 220^\circ\text{C}$, although under conventional cooling they state that it is readily supercooled and may exist as a metastable phase at room temperature. [1] found θ to be formed at about 14 at.% Zr (Pu_6Zr) and 350°C, and to have a homogeneity range from 12.1 to 20.6 at.% Zr. [7] has indexed the x-ray powder pattern for the θ phase at 21 at.% Zr on the basis of a tetragonal unit cell. Although κ is believed to correspond stoichiometrically to about PuZr_3 , its structure is based on the hexagonal AlB_2 type. The maximum solubility of Pu in αZr was found by [1] to be about 16 at.% Pu at 618°C and 11 at.% at 150°C. Crystal structure data are given in Table LX.

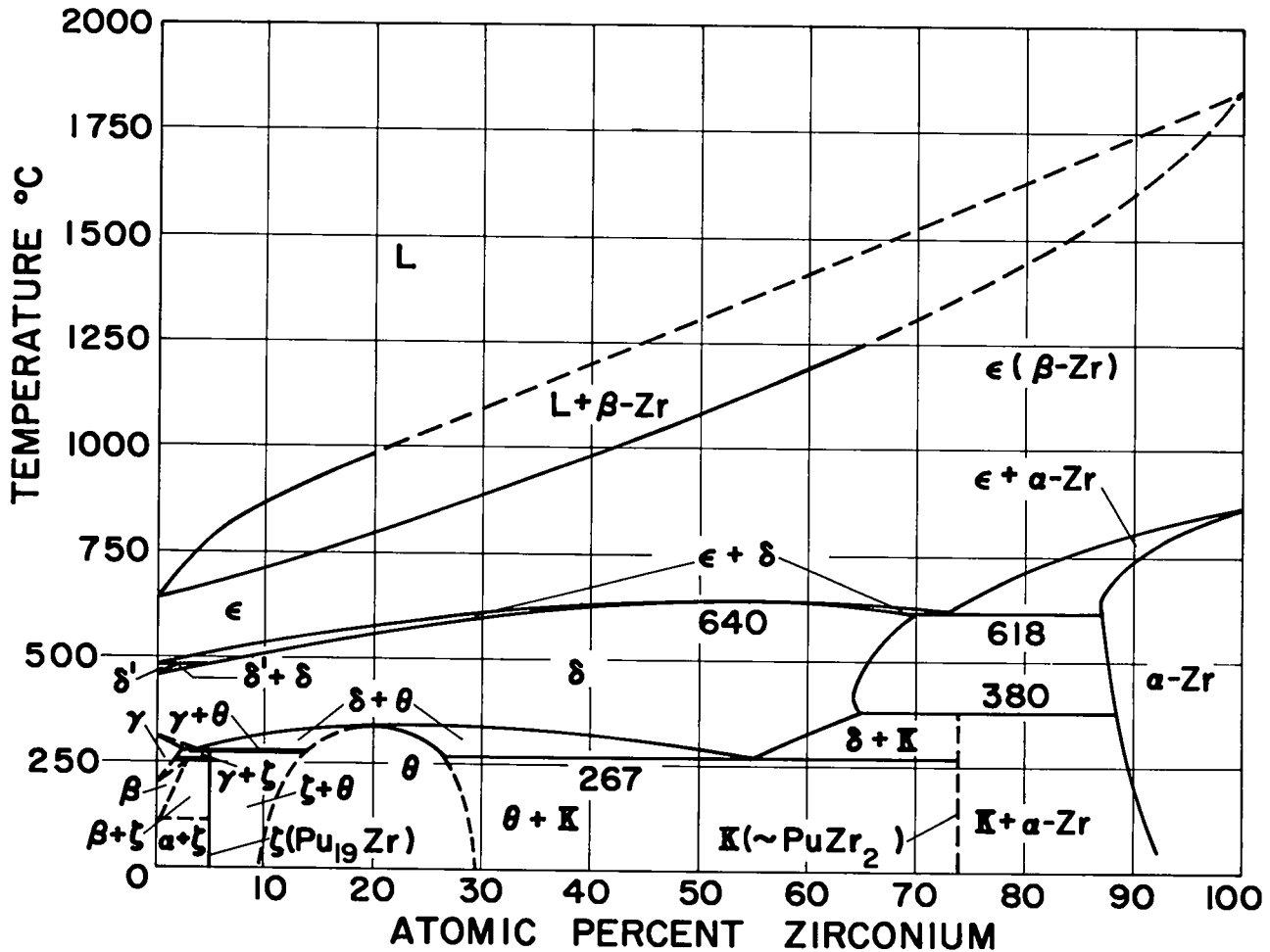


Fig. 47. Plutonium-zirconium phase diagram.

- [1] A. A. Bochvar, S. T. Konobeevsky, V. I. Kutaitsev, T. S. Menshikova, and N. T. Chebotarev, "Interaction Between Plutonium and Other Metals in Connection With Their Arrangement in Mendeleev's Periodic Table," *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958*, Vol. 6, p. 189-190, United Nations, Geneva (1958).
- [2] J. A. C. Marples, "The Plutonium-Zirconium Phase Diagram," *J. Less-Common Metals*, **2**: 331-351 (1960).
- [3] F. H. Ellinger, "Discussion of Plutonium-Zirconium Alloys," in *Plutonium 1960*, E. Grison, W. B. H. Lord, and R. D. Fowler (Eds.), pp. 318-319, Cleaver-Hume Press, Ltd., London (1961).
- [4] V. I. Kutaitsev, N. T. Chebotarev, I. G. Lebedev, M. A. Andrianov, V. N. Konev, and T. S. Menshikova, "Phase Diagrams of Plutonium with the Metals of Groups IIA, IVA, VIIIA and IB," in *Plutonium 1965*, A. E. Kay and M. B. Waldron (Eds.), pp. 420-449, Chapman and Hall, London (1967).
- [5] A. Robillard, "Discussion of Plutonium-Zirconium Alloys," in *Plutonium 1960*, E. Grison, W. B. H. Lord, and R. D. Fowler (Eds.), pp. 320-327, Cleaver-Hume Press, Ltd., London (1961).
- [6] D. F. Bowersox and J. A. Leary, "The Solubilities of Selected Elements in Liquid Plutonium: XI. Zirconium," USAEC Report LA-3742, Los Alamos Scientific Laboratory (July 5, 1967).
- [7] A. F. Berndt, "The Theta Phase in the Plutonium-Zirconium System," *J. Less Common Metals*, **12**(1): 82-83 (1967).

TABLE LIX. SOLUBILITY OF ZIRCONIUM IN LIQUID PLUTONIUM

Temperature, °C	Average solubility, at.% Zr
700	2.10
750	3.44
800	5.42
850	8.03
900	12.4
950	18.0

TABLE LX. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-ZIRCONIUM ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
Pu _x Zr ^a ζ		unknown							[3, 4]
Pu ₄ Zr θ		tetragonal	P4/ncc	10.893		16.889	16	15.76	[7]
PuZr ₂ - PuZr ₃ κ	AlB ₂	hexagonal	P6/mmm	5.060 5.055		3.119 3.123	1		[1] [2]

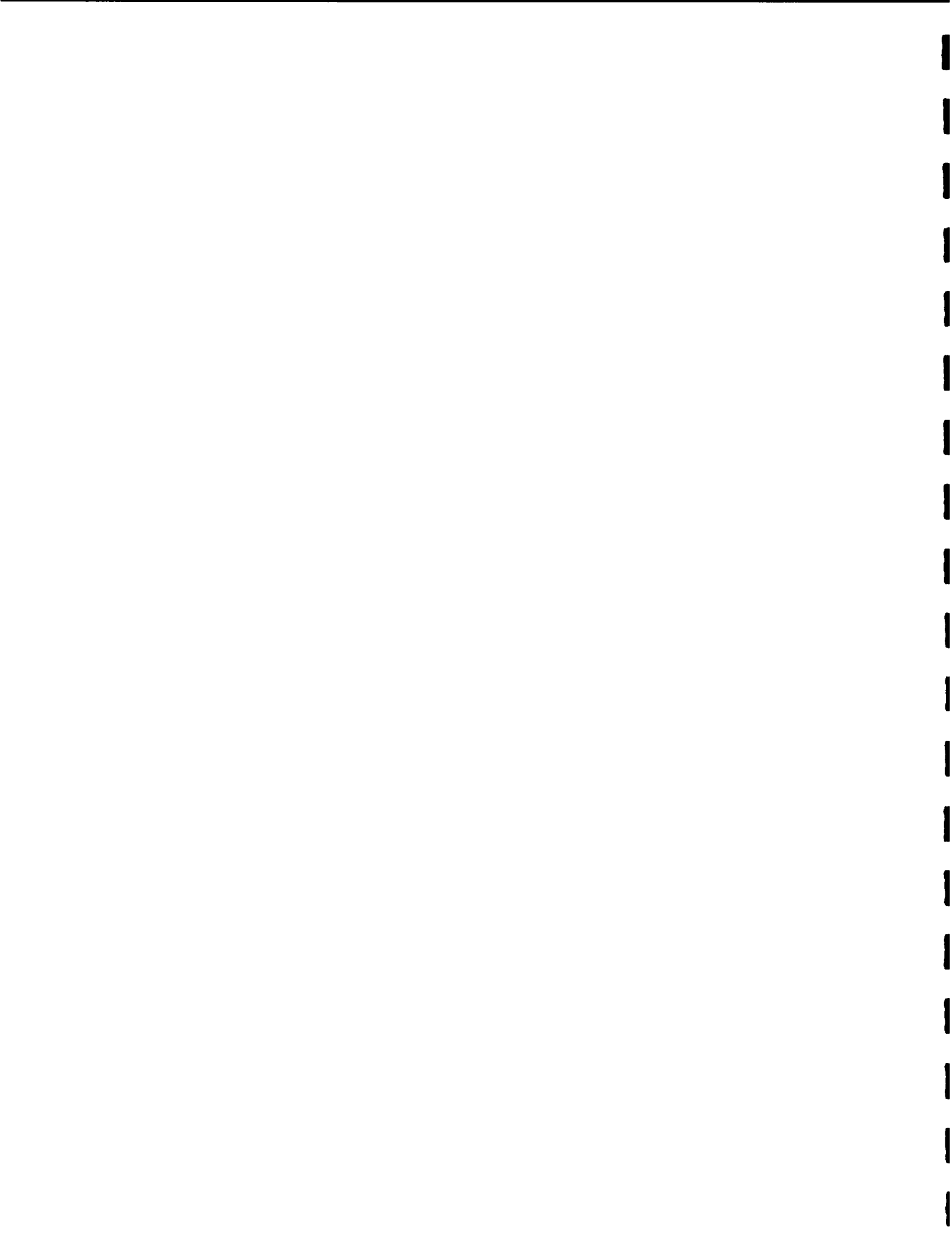
^aWhere x equals approximately 30.

TERNARY AND HIGHER SYSTEMS

Pu-Al-Ga □ **Plutonium-Aluminum-Gallium**

On the basis of three alloys examined. [1] concludes that the $(\alpha + \delta)/\delta$ phase boundary appears to be concave toward the Pu-rich corner.

- [1] J. E. Fuller, "Delta Stabilized Ternary Alloys of Plutonium," USAEC Report RFP-506. Rocky Flats Division, Dow Chemical Co. (April 22, 1965).



Pu-Ce-Co □
Plutonium-Cerium-Cobalt

[1] have determined the phase equilibria of the Pu-Ce-(Pu, Ce) Co₂ system by thermal, micrographic, and x-ray diffraction methods. The liquidus surface (Fig. 48) reveals a "eutectic" valley across the low-Co side of the system between the Pu-Co eutectic at 408°C and the Ce-Co eutectic at 422°C. The maximum temperature in the valley is 443°C. The liquidus boundaries define 11 four-phase equilibria and two pseudobinary sections. The 400°C isotherm (Fig. 49), where all alloys are solid, shows the compositions of the three

ternary intermediate phases and the extent of solid solubility of the third component in the binary intermediate phases. Crystal structure data are given in Table LXI.

- [1] F. H. Ellinger, C. C. Land, K. A. Johnson, and V. O. Struebing, "The Ternary System Plutonium-Cerium-Cobalt." *Trans. Met. Soc. AIME*, **236(11)**: 1577 (1966).
- [2] A. C. Larson, R. B. Roof, Jr., and D. T. Cromer, "The Crystal Structure of Phase C in the Ternary System Cerium-Plutonium-Cobalt," *Acta Cryst.*, **17**: 1382 (1964).

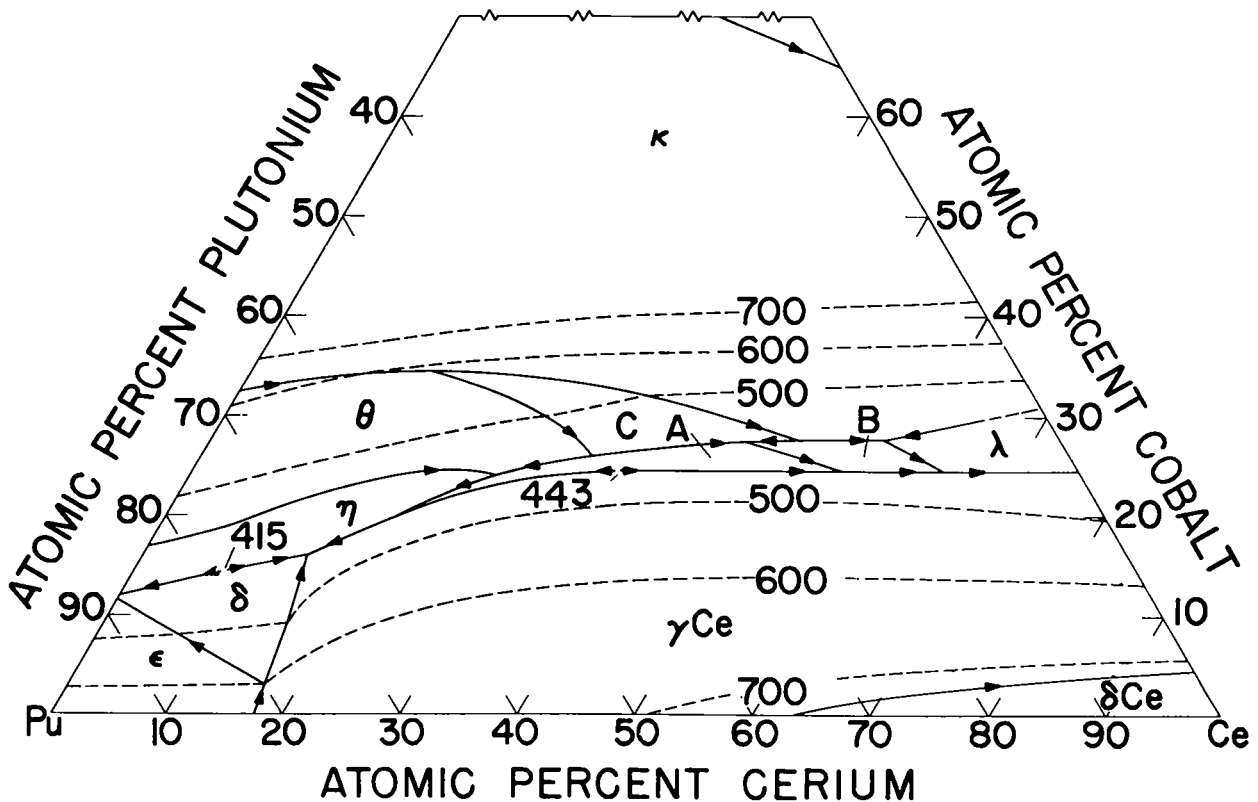


Fig. 48. Plutonium-cerium-cobalt system liquidus surface. (Redrawn from Ref. 1.)

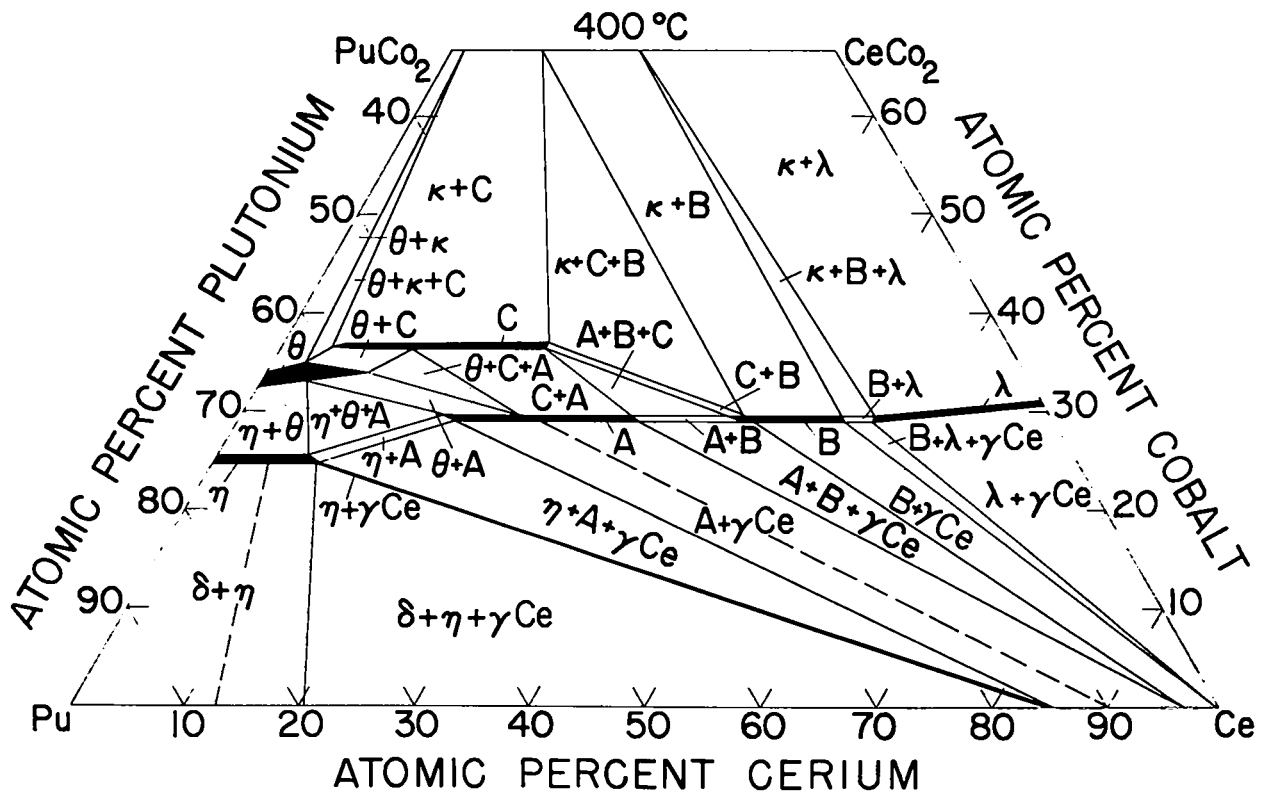


Fig. 49. Plutonium-cerium-cobalt system 400°C isotherm. (Redrawn from Ref. 1.)

TABLE LXI. CRYSTAL STRUCTURE DATA FOR PLUTONIUM-CERIUM-COBALT ALLOYS

Phase	Structure type	Symmetry	Space group	Unit cell dimensions, Å			Formula units per unit cell	X-ray density, g/cm ³	Ref.
				a	b	c			
A	Th ₇ Fe ₃	hexagonal	P6 ₃ mc				2		[1]
	Pu _{5.2} Ce _{1.8} Co ₃			9.298		5.887		12.60	
	Pu _{3.5} Ce _{3.5} Co ₃			9.358		5.986		11.00	
B	?	cubic	F	13.46 (Pu-rich)			10		[1]
				13.49 (Ce-rich)					
C	W ₅ Si ₃	tetragonal	I4/mcm	10.72 (4 at.% Ce)					
						5.346	4		[1]
				10.73 (10 at.% Ce)					
						5.359			[1]
				10.730 (12.5 at.% Ce)					
						5.383		13.64	[2]
				10.743 (22 at.% Ce)					
						5.367			[2]

Pu-Ce-Co-Mn □
Plutonium-Cerium-Cobalt-Manganese

In connection with work on Pu-Ce-Co alloys containing from 5.0 to 8.0 g/cm³ Pu, [1] states that differential thermal analysis data indicate that as much as one-half of the Ce of any particular ternary Pu-Ce-Co alloy may be replaced by Mn and yet maintain the liquidus temperature below 450°C.

- [1] "Quarterly Status Report on Plutonium Reactor Fuel Development for Period Ending November 20, 1964." USAEC Report LA-3208-MS, Los Alamos Scientific Laboratory (December 1964).



Pu-Ce-Cu □
Plutonium-Cerium-Copper

The liquidus surface of this system (Fig. 50) has been determined by [1] from thermal, micrographic, and electron microprobe data.

No ternary intermediate phases were found. A continuous solid solution and quasibinary section occur between PuCu_2 and CeCu_2 . The

liquidus boundaries of the primary phase fields in the Pu-Ce-(Pu, Ce) Cu_2 subsystem define four invariant reactions, and four invariant reactions were also found in the higher melting PuCu_2 - CeCu_2 -Cu subsystem.

- [1] L. J. Wittenberg, D. E. Etter, J. E. Selle, and P. A. Tucker. "Phase Equilibria in the Plutonium-Cerium-Copper System." *Nucl. Sci. Eng.*, **23**(1): 1 (1965).

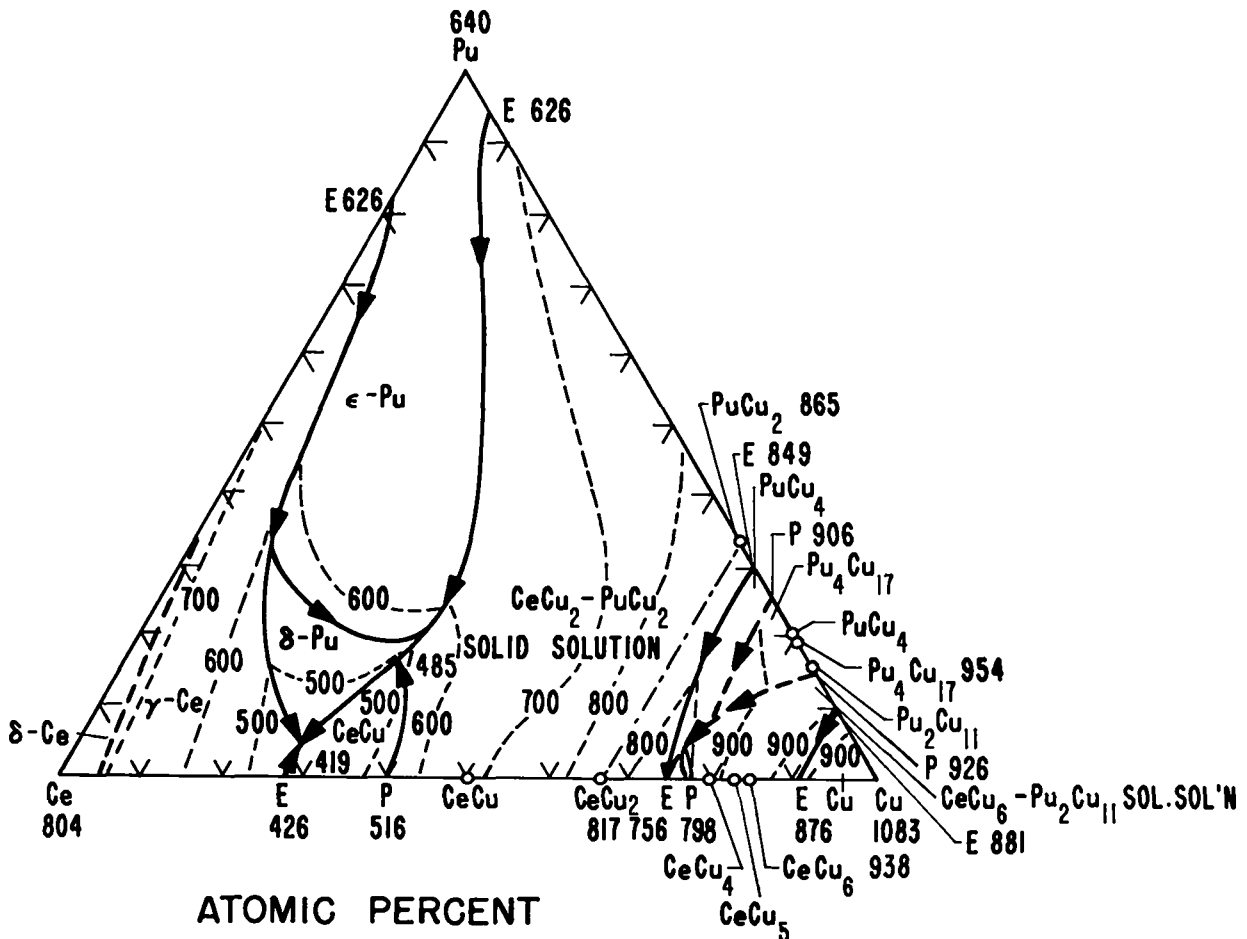
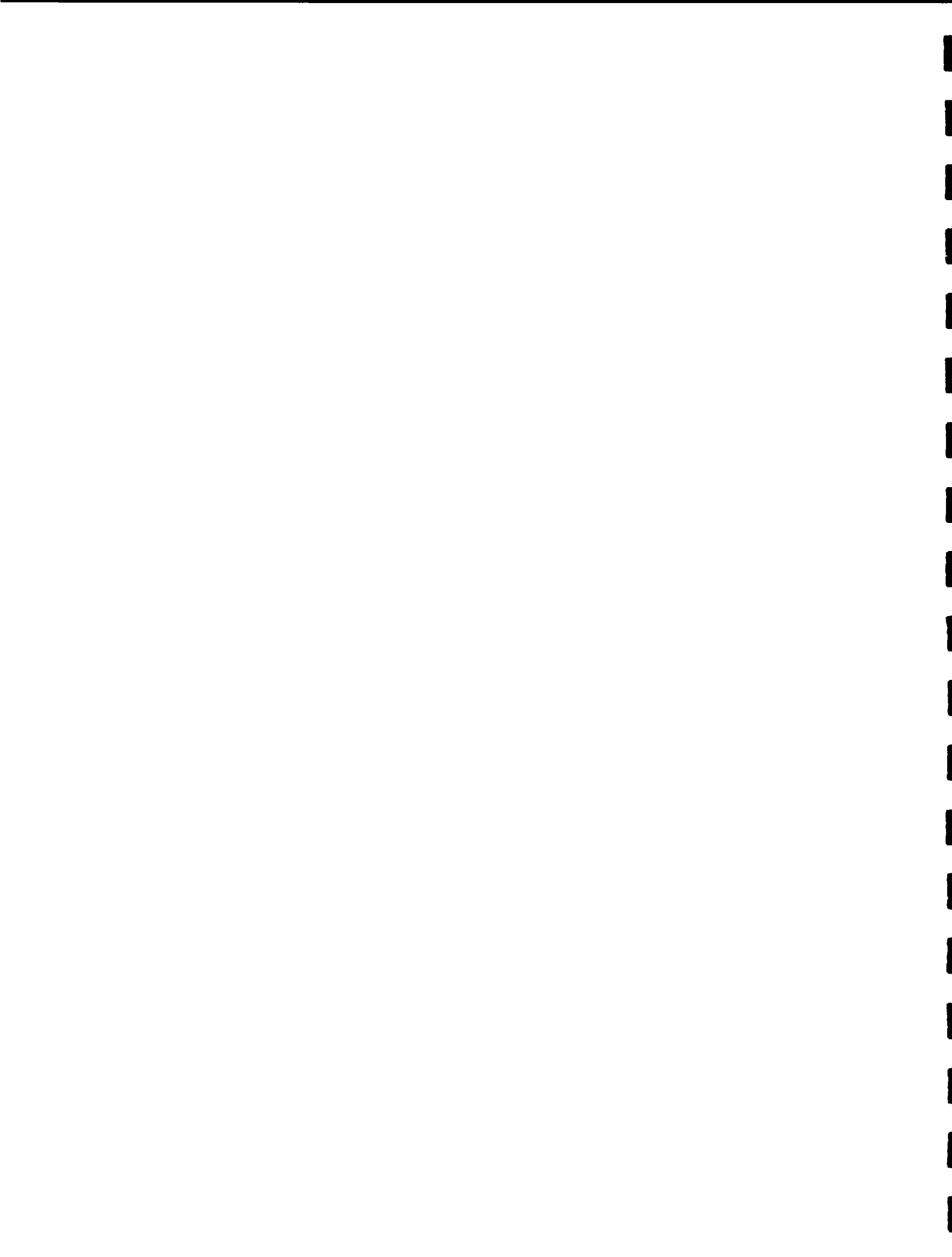


Fig. 50. Plutonium-cerium-copper system liquidus surface. (Redrawn from Ref. 1.)



Pu-Ce-Fe □
Plutonium-Cerium-Iron

The liquidus surface of this system, shown in Fig. 51, has been constructed by [1] from information obtained from thermal, micrographic, x-ray diffraction, and electron microprobe analyses.

No ternary intermediate phases are formed; CeFe_2 and PuFe_2 are mutually soluble in all proportions, and [1] give the lattice parameter *vs.*

composition relationships. An important feature of this system is the "eutectic" valley that extends from the Pu- Pu_6Fe eutectic at 411°C to a maximum at 660°C near the Ce- CeFe_2 eutectic point.

- [1] P. A. Tucker, D. E. Etter, and J. M. Gebhart III. "Phase Equilibria in the Ternary System Plutonium-Cerium-Iron," in *Plutonium 1965*. A. E. Kay and M. B. Waldron (Eds.), pp. 392-404. Chapman and Hall, London (1967).

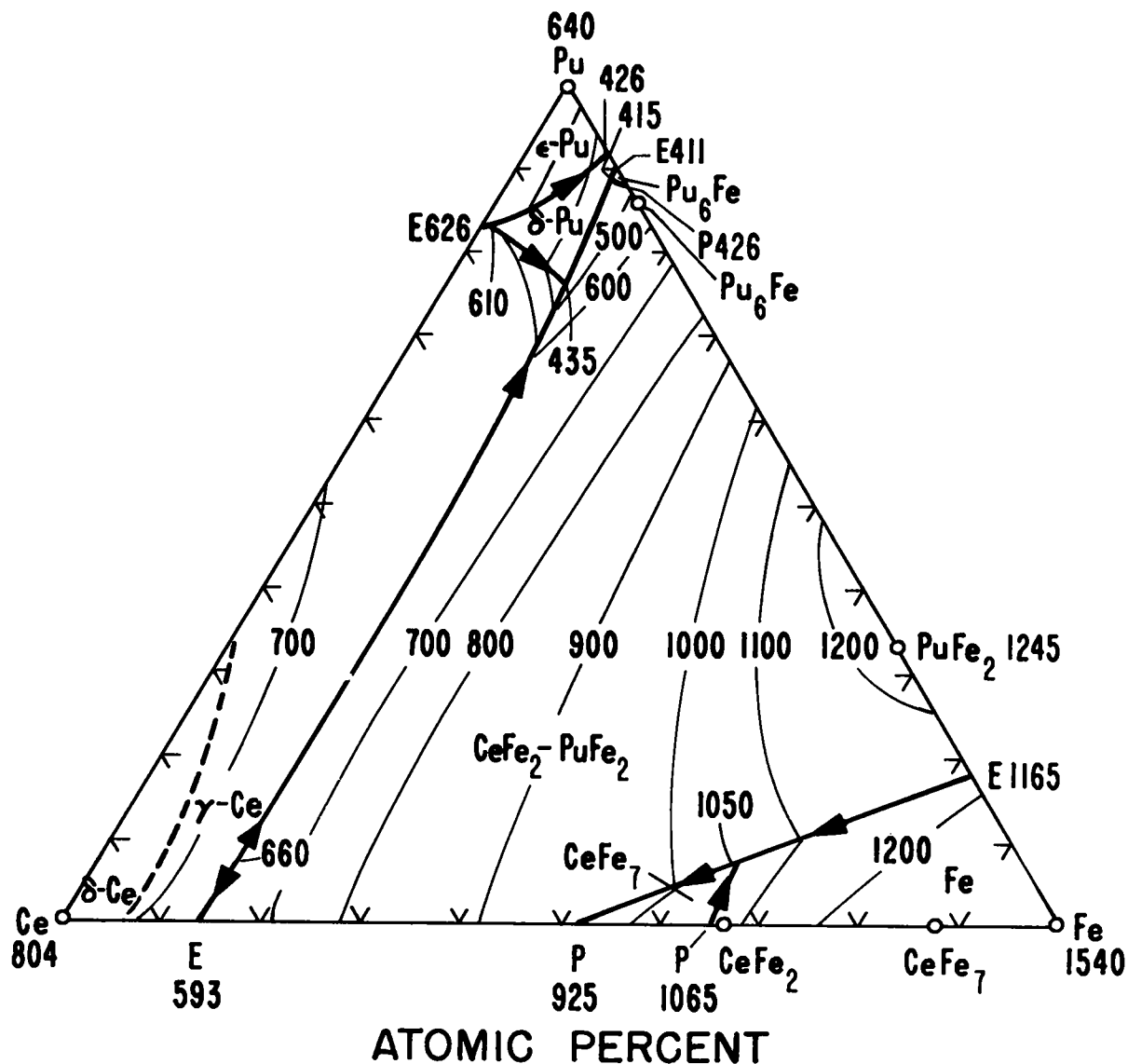
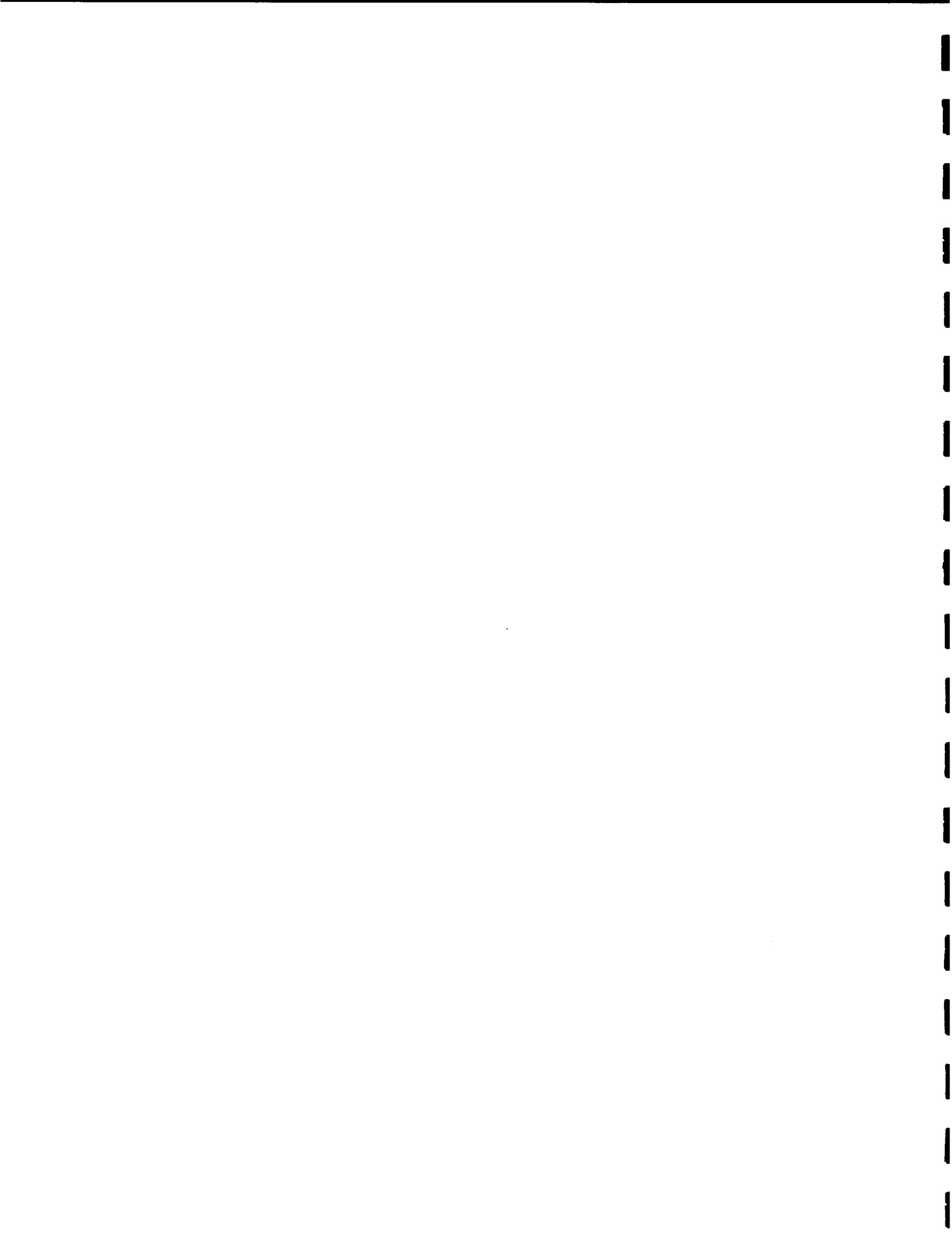


Fig. 51. Plutonium-cerium-iron system liquidus surface. (Redrawn from Ref. 1.)



Pu-Ce-Ga □
Plutonium-Cerium-Gallium

[1] concludes. on the basis of limited information. that the $(\alpha + \delta)/\delta$ phase boundary is concave toward the Pu-rich corner of the ternary diagram.

- [1] J. E. Fuller. "Delta Stabilized Ternary Alloys of Plutonium," USAEC Report RFP-506. Rocky Flats Division. Dow Chemical Co. (April 22. 1965).



Pu-Ce-Mn □
Plutonium-Cerium-Manganese

Differential thermal-analysis data reported by [1] for five alloys of this system indicate that there is a valley at 638°C in the vicinity of 25 at.% Pu-12 at.% Mn-63 at.% Ce.

- [1] "Quarterly Status Report on LAMPRE Programs for Period Ending November 20, 1963," USAEC Report LAMS-3021. p. 8. Los Alamos Scientific Laboratory (Dec. 23, 1963).



Pu-Ce-Ni □
Plutonium-Cerium-Nickel

The liquidus surface of this system as determined by [1] is shown in Fig. 52. The high-Ni corner is incomplete. "Eutectic" valleys extend across the system between the low-Ni. Pu-Ni, and Ce-Ni eutectics. These valleys define two ternary eutectics, both having melting points of 440°C.

The 430°C partial isotherm (see Fig. 53) determined by [1] shows the following main fea-

tures: (1) two three-phase triangles defining the compositions of the three phases solidifying at each ternary eutectic, (2) the extensive solubility of Pu in Ce_5Ni_2 (Ce_7Ni_3 according to [2]). and (3) the continuous solid solution between PuNi and CeNi.

The liquidus surface (Fig. 54) as given by [3] differs considerably from that of [1]. For example, it shows the $\delta Pu-\gamma Ce-(Ce, Pu)_7Ni_3$ eutectic point at 17 at.% Pu, 22 at.% Ni, 61 at.% Ce and 445°C as compared to 53 at.% Pu,

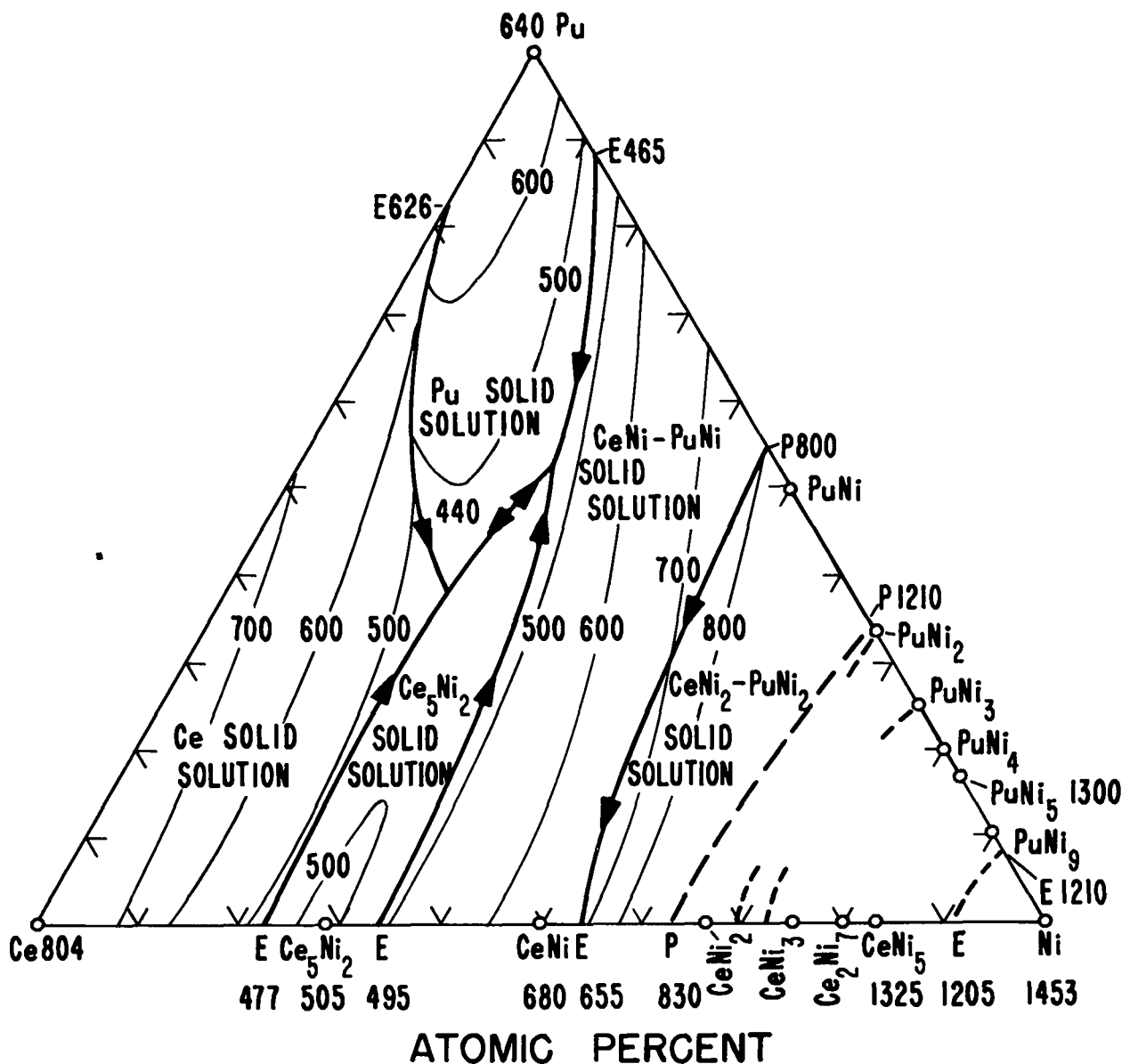
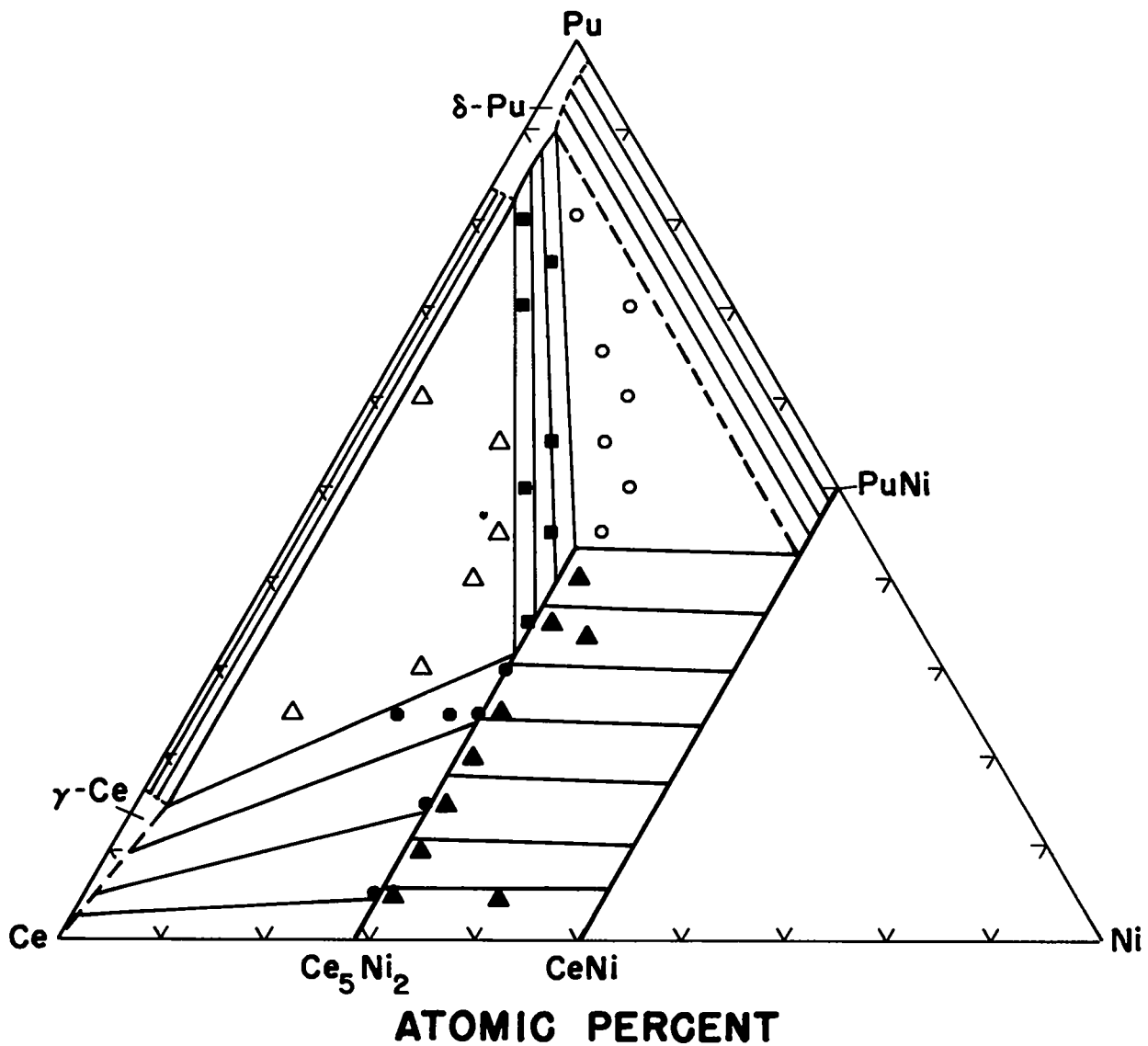


Fig. 52. Plutonium-cerium-nickel system liquidus surface. (Redrawn from Ref. 1.)



- $\delta\text{-Pu} + \text{Ce}_5\text{Ni}_2$ SOLID SOLUTION
- $\gamma\text{-Ce} + \text{Ce}_5\text{Ni}_2$ SOLID SOLUTION
- ▲ CeNi-PuNi SOLID SOLUTION + Ce_5Ni_2 SOLID SOLUTION
- △ $\delta\text{-Pu}, \gamma\text{-Ce} + \text{Ce}_5\text{Ni}_2$ SOLID SOLUTION
- $\delta\text{-Pu}, \text{CeNi-PuNi}$ SOLID SOLUTION + Ce_5Ni_2 SOLID SOLUTION

Fig. 53. Plutonium-cerium-nickel system 430°C isotherm. (Redrawn from Ref. 1.)

25 at.% Ni, 22 at.% Ce and 440°C found by [1]. Also, [3] show only about 12 at.% Pu soluble in Ce_7Ni_3 in contrast to about 41 at.% Pu found by [1].

[1] L. J. Wittenberg and G. R. Grove, "Reactor Fuels and Materials Development Plutonium Research: 1965 Annual Report, Mound Laboratory," USAEC Report MLM-1328. pp. 9-13 (Jan. 20, 1967).

[2] R. B. Roof, Jr., A. C. Larson, and D. T.

Cromer, "The Crystal Structure of Ce_7Ni_3 ," *Acta Cryst.*, **14**: 1084 (1961).

[3] V. I. Kutaitsev, N. T. Chebotarev, I. G. Lebedev, M. A. Andrianov, V. N. Konev, and T. S. Menshikova, "Phase Diagrams of Plutonium with the Metals of Groups IIA, IVA, VIIIA and IB," in *Plutonium 1965*. A. E. Kay and M. B. Waldron (Eds.), pp. 420-449, Chapman and Hall, London (1967).

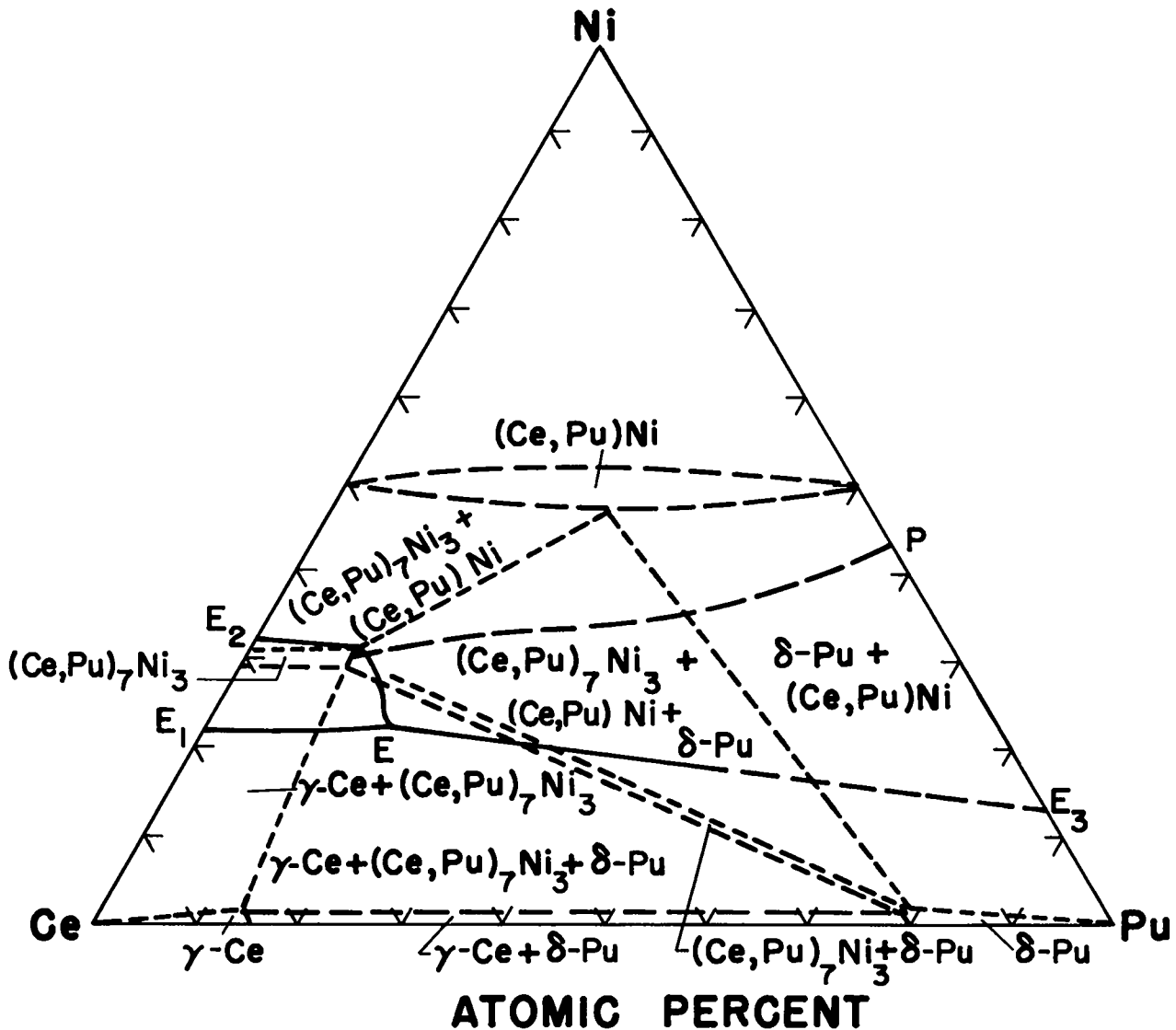
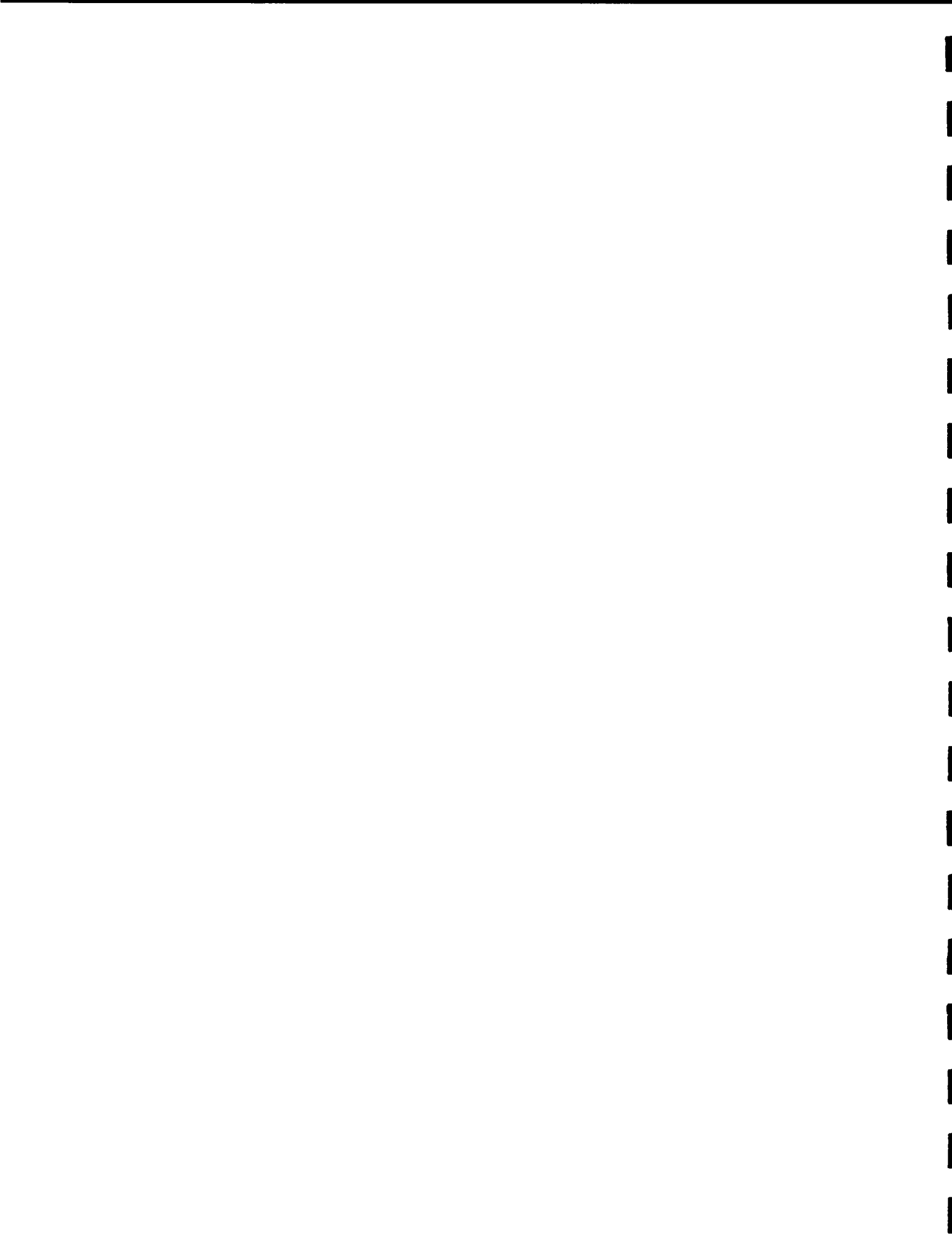


Fig. 54. Plutonium-cerium-nickel system liquidus surface and 400°C isotherm. (Redrawn from Ref. 3.)



Pu-Cu-Sn □
Plutonium-Copper-Tin

[1] reports that a Cu-7.6 at.% Sn-1 at.% Pu alloy consisted of a single phase after homogenization at 700°C.

- [1] S. T. Konobeevski, B. M. Levitski, L. D. Panteleev, and K. P. Dubrovin, "Effect of Irradiation on Phase Transformations in Cu-Sn and Cu-Sn-Pu Alloys." *J. Nucl. Mater.*, **5(3)**: 317 (1962).



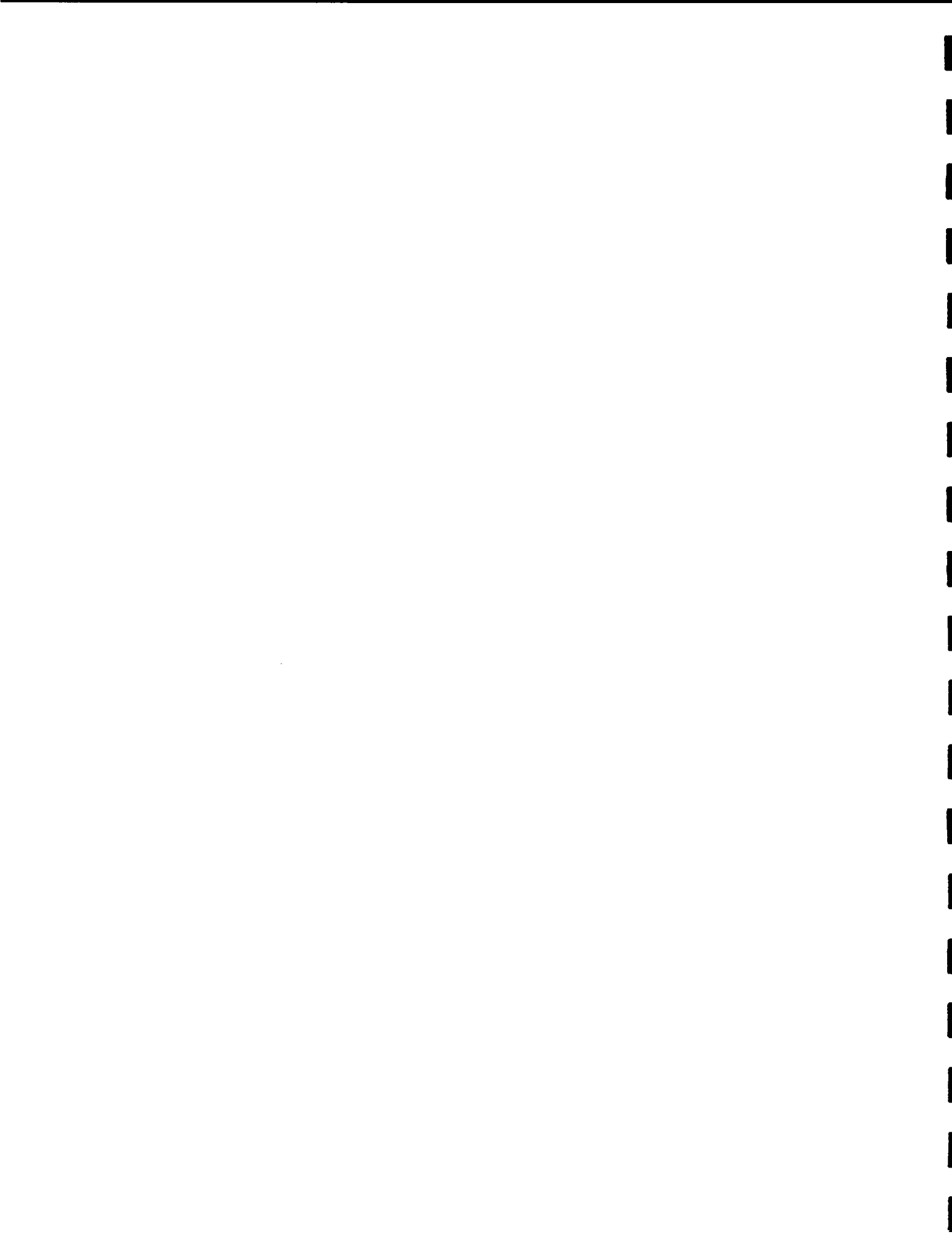
Pu-Fe-U □
Plutonium-Iron-Uranium

[1] have studied mainly two compositions. 74 U-25 Pu-1 Fe and 73.5 U-25 Pu-1.5 Fe (in wt%). The cast alloys were composed predominantly of the Pu-U ζ phase together with U_6Fe and Pu_6Fe . The latter compounds tended to form $(U, Pu)_6Fe$ on homogenization of the alloys. Dilatometric examination of these alloys revealed, respectively, transformations at $595 \pm 3^\circ C$ and $590 \pm 3^\circ C$ and fusion points at $705 \pm 5^\circ C$ and $680 \pm 5^\circ C$. Treating the alloys above the transformation temperatures resulted in the formation and retention of some Pu-U η phase. Treating the alloys below the transformation temperatures resulted in the formation of some αU .

[2] reports that continuous solid solutions exist between Pu_6Fe and U_6Fe and between $PuFe_2$ and UFe_2 . Also, he gives lattice parameter *vs.* composition data.

[1] R. Boucher, P. Barthelemy, and C. Milet. "A Study of Plutonium-Based Alloys Carried Out at Fontenay-aux-Roses," in *Plutonium 1965*. A. E. Kay and M. B. Waldron (Eds.). pp. 485-509, Chapman and Hall, London (1967).

[2] F. H. Ellinger. "A Review of the Intermetallic Compounds of Plutonium," Chap. XXV in *The Metal Plutonium*. A. S. Coffinberry and W. N. Miner (Eds.). pp. 265-280, The University of Chicago Press, Chicago (1961).



Pu-Fs(Fz)-U Plutonium-Fissium(Fizzium)-Uranium

Phase studies in the Pu-Fs-U and Pu-Fz-U quasi-ternary alloy systems by means of metallography and x-ray diffraction have been reported by [1] and [2]. Alloys investigated contained 20 wt% Pu and either 5, 10, or 15 wt% Fs or Fz.*

Solidus temperatures as a function of Fs content for U-20 wt% Pu-Fs alloys are shown in Fig. 55, increasing either the Pu or Fs content decreases the solidus temperature as shown in Fig. 56.

The compositions of alloys investigated by [1] and [2] and the phases found in equilibrium after isothermal annealing are shown in the isopleth presented in Fig. 57. The nomenclature used for the α U, β U, γ U, ζ (U-Pu), and η (U-Pu) phases is the same as that adopted by [3] for phases in the U-Pu system. Six other phases occur in the system:

Alpha-prime (α'): A metastable modification of α U that is also referred to as "distorted alpha".

Gamma-naught (γ^0): A tetragonal modification of the γ phase having a c/a ratio of approximately 0.98.

Gamma-prime (γ'): A tetragonal phase based on the binary U_2Mo phase but which may take Pu in solution.

Theta (θ): A phase containing U, Pu, and Pd that is present between room temperature and the solidus temperature in all alloys studied. Crystal structure type not identified.

U_2Ru : A monoclinic phase found in the Fs alloys.

ZrRu: A CsCl-type phase found only in the Fz alloys.

*The term fissium (Fs) denotes the equilibrium concentration of second long-period elements (Mo, Tc, Ru, Rh, Pd) that build up as fission products in the fuel during irradiation and which are not removed during normal pyrometallurgical fuel reprocessing. Fizzium (Fz) denotes the same group of solid fission products with the addition of Zr in approximately the same amount as Ru. The nominal composition of U-20 wt% Pu-5 wt% Fs is: 20 wt% Pu, 1.90 wt% Mo, 2.02 wt% Ru, 0.72 wt% Pd, and 0.28 wt% Rh, balance U (six components) and the nominal composition of U-20 wt% Pu-5 wt% Fz is: 20 wt% Pu, 1.31 wt% Mo, 1.38 wt% Ru, 1.45 wt% Zr, 0.53 wt% Pd, and 0.26 wt% Rh, balance U (seven components).

In addition to the phases shown in Fig. 57, all alloys contained the θ phase at all temperatures below the solidus. U_2Ru was found in all Fs alloys and increased in stability with an increase in Fs content. (Stable below 625°C for 5 wt% Fs, stable below 700°C for 10 wt% Fs, and stable below 775°C for 15 wt% Fs). In all Fz alloys, ZrRu was stable between room temperature and the solidus. In the construction of the isopleth shown in Fig. 57 and in the formulation of the four-phase invariant reactions given below, [1] and [2] assumed for simplicity that the θ , U_2Ru , and ZrRu phases do not take part in the transformations. The isothermal transformations indicated in Fig. 57 correspond to the following four-phase invariant reactions.

$\alpha + \beta + \eta \rightarrow \zeta$ (Peritectoid formation of ζ at 610°C)

$\beta + \gamma \rightarrow \zeta + \gamma'$ (Class II reaction at 555°C)

$\beta \rightarrow \alpha + \zeta + \gamma'$ (Eutectoid decomposition of β at 500°C).

Isothermal sections through the U-rich corner of the Pu-Fs-U pseudo-ternary are shown in Fig. 58. The phase regions in the isothermal sections have not been accurately determined but were drawn in agreement with the proposed four-phase reaction scheme and the established binary phase boundaries. Three-phase regions in equilibrium 10°C above and 10°C below the first invariant reaction are shown in Figs. 58a and 58b. Fig. 58c shows the location of the two three-phase regions ($\beta + \gamma + \zeta$ and $\beta + \gamma + \gamma'$) that take part in the invariant reaction at 555°C (Fig. 58d). Below the β eutectoidal decomposition temperature (500°C), the $\alpha + \zeta + \gamma'$ three-phase region is stable down to room temperature (Fig. 58f). The same three phases are in equilibrium in the U corner of the Pu-Mo-U system below the lowest temperature-invariant reaction at 525°C.

Lattice constants of the γ^0 and γ phases quenched from various temperatures between 625° and 775°C are given in Table LXII. The c/a ratio of the tetragonal γ^0 phase in U-20 wt% Pu-5 wt% Fs decreases from 0.99 to 0.98 with decrease in the annealing temperature. The lattice constant of γ decreases from 3.4196 Å to 3.3956 Å with increase in Fs content from 10 wt% to 15 wt%. The lattice constants for ZrRu quenched from temperatures between 625 and 775°C are listed in Table LXIII. All values of a lie between the value of 3.27 Å found by [4] in U-Fs alloys, and the value of 3.253 Å obtained by [5] on the pure ZrRu compound.

- [1] O. L. Kruger, "Phase Studies on U-20 wt.% Pu-Fs Alloys," USAEC Report ANL-6974, Argonne National Laboratory, (May 1965).
- [2] O. L. Kruger, "Phase Relations and Structures in Uranium-Plutonium-Fissium Alloys." *J. Nucl. Mater.*, **19(1)**: 29-41 (1966).
- [3] F. H. Ellinger, R. O. Elliott, and E. M. Cramer, "The Plutonium-Uranium System," *J. Nucl. Mater.*, **1(3)**: 233-243 (1959).
- [4] M. V. Nevitt and S. T. Zegler, "Transformation Temperatures and Structures in Uranium-Fissium Alloys," *J. Nucl. Mater.*, **1(1)**: 6-12 (1959).
- [5] A. E. Dwight, "CsCl-Type Equiatomic Phases in Binary Alloys of Transition Elements." *Trans. Met. Soc. AIME*, **215(2)**: 283-286 (1959).

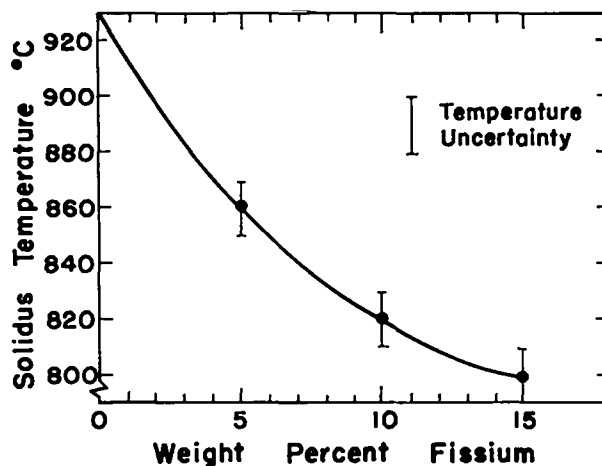


Fig. 55. Effect of fissium on the solidus temperature of uranium-20 wt% plutonium alloys. (Redrawn from Ref. 1.)

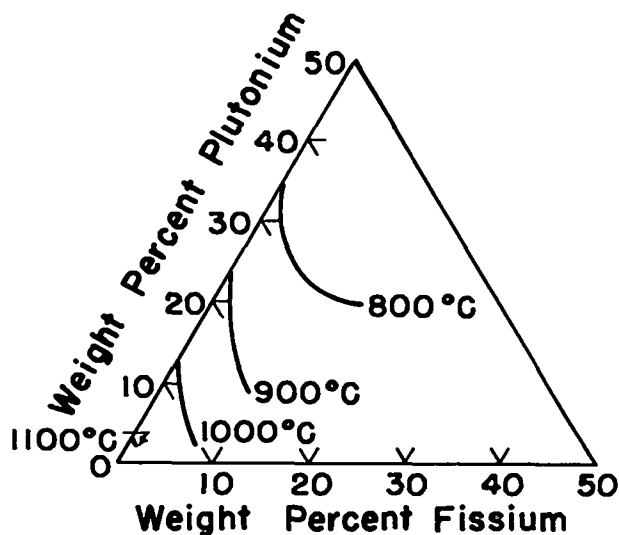


Fig. 56. Solidus temperatures in the uranium corner of the plutonium-fissium-uranium system. (Redrawn from Ref. 1.)

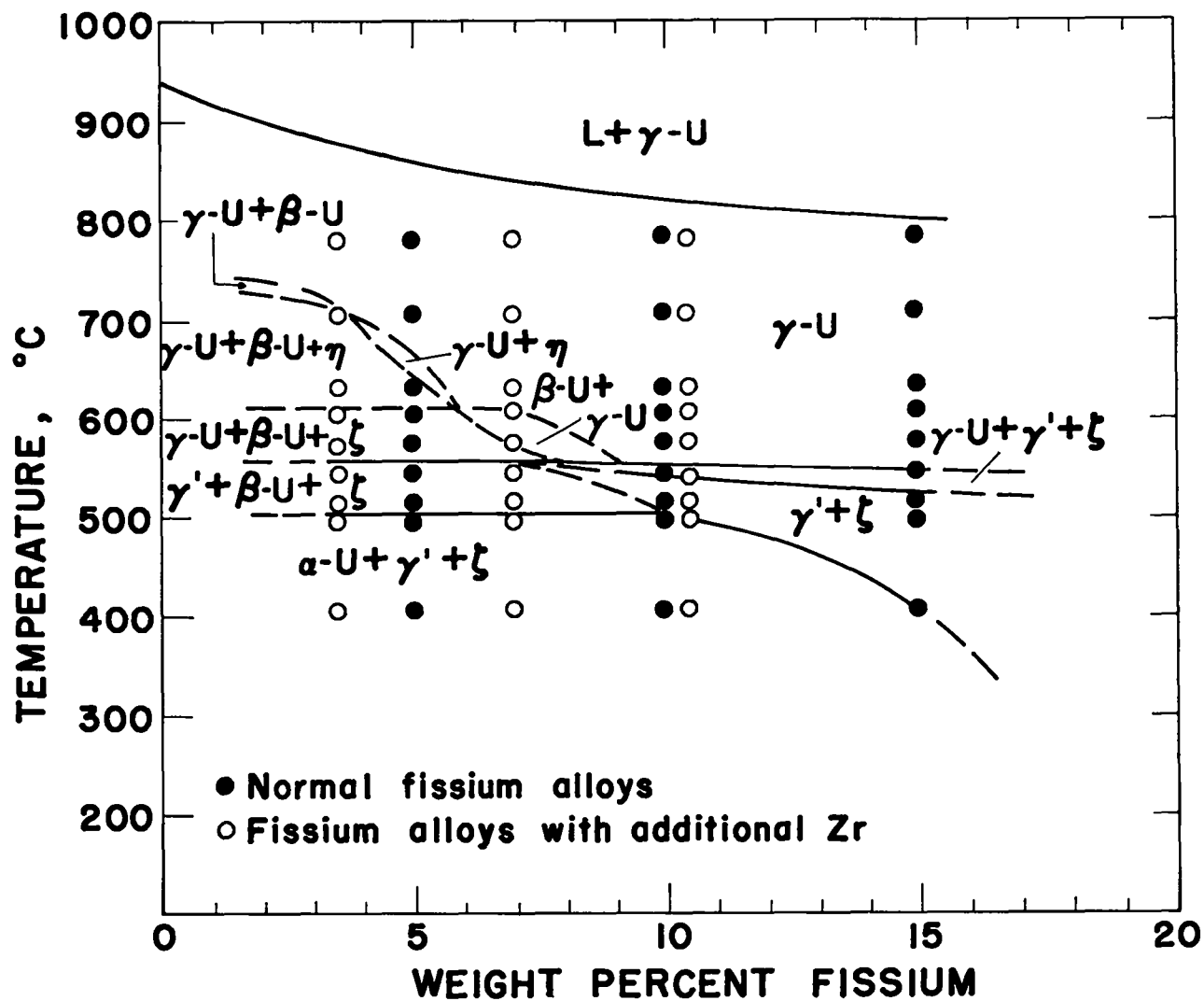
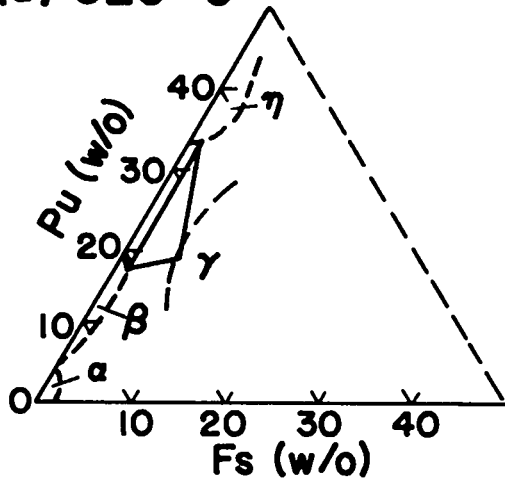
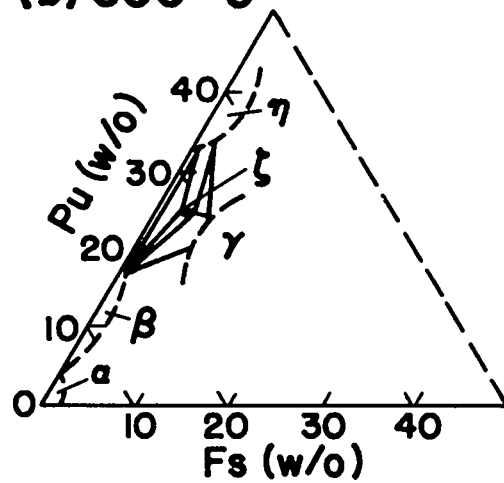


Fig. 57. Isopleth showing equilibrium phases and transformations in the pseudo-ternary plutonium-fissium-uranium system at 20 wt% plutonium. (Redrawn from Ref. 1.)

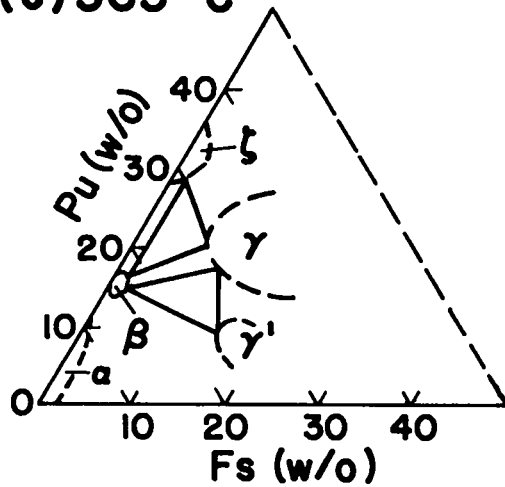
(a) 620 °C



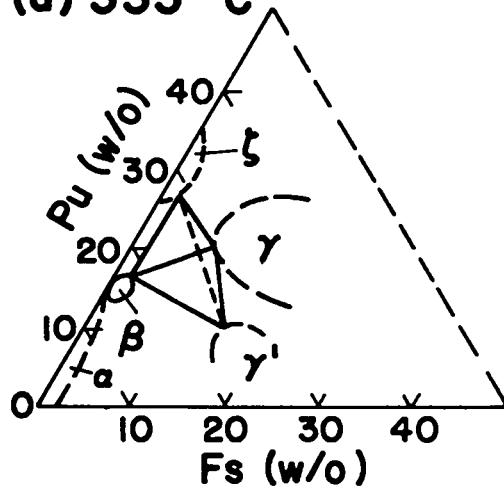
(b) 600 °C



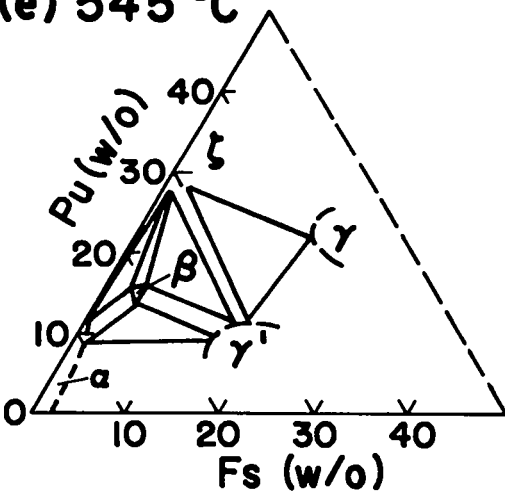
(c) 565 °C



(d) 555 °C



(e) 545 °C



(f) 475 °C

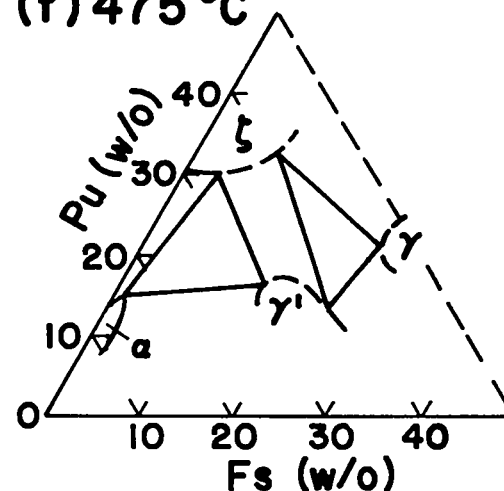


Fig. 58. Plutonium-fissium-uranium isothermal sections showing matrix phase equilibria and invariant reactions between 620 and 475°C. (Redrawn from Ref. 1.)

TABLE LXII. LATTICE CONSTANTS OF GAMMA-NAUGHT AND GAMMA PHASES IN URANIUM-20 WT% PLUTONIUM-5, 10, and 15 WT% FISSIUM ALLOYS QUENCHED FROM VARIOUS TEMPERATURES [1].

Annealing Temperature (°C)	5 wt% Fs (γ^0) (Å)	10 wt% Fs (γ) (Å)	15 wt% Fs (γ) (Å)
775	a = 3.46 c = 3.43 c/a = 0.99	a = 3.4196 ± 0.0001	a = 3.3956 ± 0.0001
700	a = 3.56 c = 3.41 c/a = 0.99	a = 3.430 ± 0.001	a = 3.4005 ± 0.0001
625	a = 3.46 c = 3.40 c/a = 0.98	a = 3.4284 ± 0.0002	

TABLE LXIII. LATTICE CONSTANTS OF ZrRu PHASE IN PLUTONIUM-FISSIUM-URANIUM ALLOYS QUENCHED FROM VARIOUS TEMPERATURES [1].

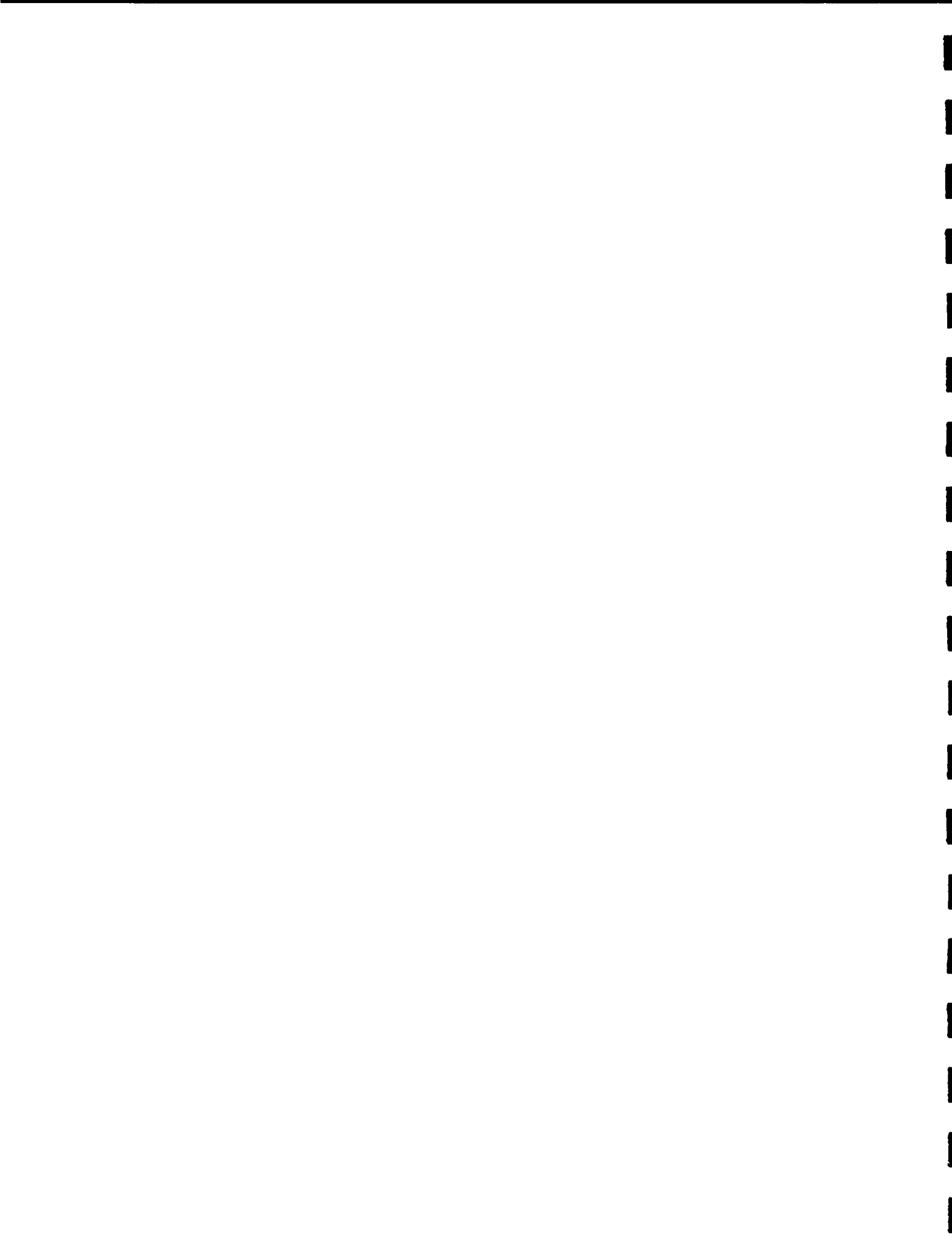
Composition	Annealing Temperature (°C)	Lattice Constant (Å)
U-20 wt% Pu- 3.5 wt% Fs-1.5 wt% Zr	700	3.260 ± 0.001
U-20 wt% Pu- 7.0 wt% Fs-3.0 wt% Zr	775	3.2624 ± 0.0001
U-20 wt% Pu- 7.0 wt% Fs-3.0 wt% Zr	700	3.2623 ± 0.0001
U-20 wt% Pu- 7.0 wt% Fs-3.0 wt% Zr	625	3.263 ± 0.001
U-20 wt% Pu-10.5 wt% Fs-4.5 wt% Zr	775	3.2611 ± 0.0001
U-20 wt% Pu-10.5 wt% Fs-4.5 wt% Zr	700	3.2612 ± 0.0001
U-20 wt% Pu-10.5 wt% Fs-4.5 wt% Zr	625	3.2641 ± 0.0001



Pu-Mn-U □
Plutonium-Manganese-Uranium

The microstructure characteristic of the U-Mn eutectic was found by [1] to be obliterated by additions of less than 10 at.% Pu. The alloys consistently showed three phases after heat treatments up to 700°C.

- [1] "Quarterly Status Report on LAMPRE Program for Period Ending August 20, 1961," USAEC Report LAMS-2620, Los Alamos Scientific Laboratory (September 1961).



Pu-Mo-U □ Plutonium-Molybdenum-Uranium

Constitutional studies of this ternary alloy system, mainly by [1], have been directed toward determining the boundaries of the γ U solid solution region and the phase equilibria associated with its decomposition. The liquidus projection covering the composition range 35-100 at.% U is shown in Fig. 59. In this composition range the alloys solidify either as the γ U solid solution or as a mixture of γ U and Mo. The solidus projection is shown in Fig. 60. With decreasing temperatures, the γ U region expands toward increasing Pu contents as shown by the isothermal sections at 1000, 900, 800, and 700°C (Figs. 61, 62, 63, and 64, respectively). Below 900°C, the γ U/(γ U + Mo) boundary develops a kink that foreshadows the splitting, at about 630°C, of the γ U solid solution into a U-rich region and a Pu-rich region, termed γ_1 and γ_2 , respectively. Below about 700°C, where the Pu-U phases, η and ζ , first appear, the ternary equilibria become complex, and have not been fully worked out. Six four-phase invariant reactions occur between 610 and 525°C. The γ_1 phase decomposes eutectoidally, $\gamma_1 \rightarrow \alpha\text{U} + \gamma' + \zeta$, with the eutectoid point lying in the neighborhood of 10-14 at.% Mo and 17-20 at.% Pu. [2] has shown that this reaction is very sluggish and proceeds in two stages. The isothermal section at 450°C (Fig. 65) shows the phase equilibria when γ U (γ_1 and γ_2) is no longer present.

Fig. 66 shows the phases to be expected on quenching alloys from the γ phase region. Besides the retained γ_1 and γ_2 phases, it shows the metastable phases designated γ_0 and α_1 . [2] has determined the lattice parameter-composition relationships of the γ phase preserved to room temperature by quenching from 900°C. He also reported that the γ_0 phase has a tetragonally-deformed γ U structure, but the identity of the true unit cell is uncertain. The four-atom, face-centered unit cell has the dimensions:

$$a = 4.91, c = 3.415\text{\AA}, c/a = 0.695.$$

The phase labelled α_1 has a distorted α U type of structure.

Transformations in homogenized and as-cast alloys containing 20 wt% Pu and 8, 10, and 12 wt% Mo, and 30 wt% Pu and 10 wt% Mo were studied by [3]. Physical property measurements on Pu-Mo-U alloys of interest as fast reactor fuels

have been reported by [4, 5, and 6] and have been summarized by [7].

- [1] P. G. Mardon, J. P. Evans, D. J. Hodkin, J. M. North, and J. H. Pearce, "The Constitution and Fabrication of Uranium-Molybdenum-Plutonium Fuels," in *Plutonium 1960*, E. Grison, W. B. H. Lord, and R. D. Fowler (Eds.), pp. 329-352, Cleaver-Hume Press, Ltd., London (1961).
- [2] F. Anselin, *Ibid.*, pp. 367-394.
- [3] R. Boucher, "Study of Uranium-Plutonium-Molybdenum Alloys," *J. Nucl. Mater.*, **6(1)**: 84-95 (1962).
- [4] R. Boucher and P. Barthelemy, "Comparison of the Alloys U-Pu-Mo, U-Pu-Nb, U-Pu-Ti, U-Pu-Zr" (in French), French Report CEA-R-2531 (July 1964), translation available as ANL-Trans.-138 (1964).
- [5] L. R. Kelman, "Fast Reactor Fuel Development at Argonne National Laboratory," USAEC Report TID-7546, presented at Fuel Element Conference, Paris, Nov. 18-23, 1957.
- [6] D. M. Poole, J. K. Critchley, J. A. C. Davidson, P. M. French, E. N. Hodkin, and M. J. F. Notley, "Properties of Plutonium Based Fuels," in *Plutonium 1960*, E. Grison, W. B. H. Lord, and R. D. Fowler (Eds.), pp. 627-649, Cleaver-Hume Press, Ltd., London (1961).
- [7] R. Boucher, P. Barthelemy, and C. Milet, "A Study of Plutonium-Based Alloys Carried Out at Fontenay-aux-Roses," in *Plutonium 1965*, A. E. Kay and M. B. Waldron (Eds.), p. 485, Chapman and Hall, London (1967).

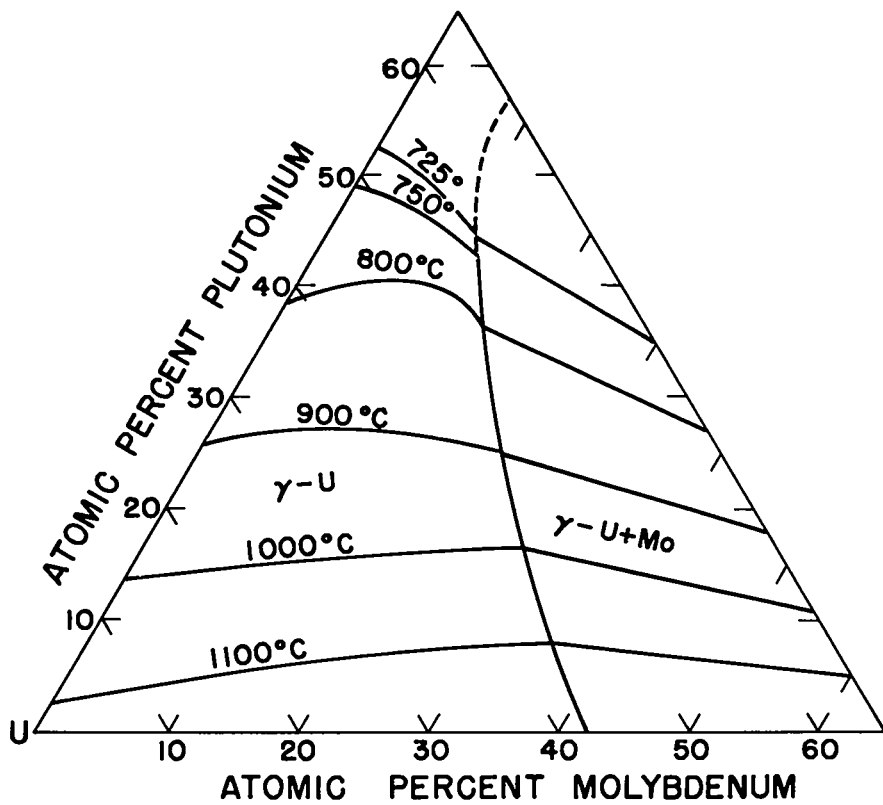


Fig. 59. Plutonium-molybdenum-uranium partial liquidus projection and fields of primary crystallization. (Redrawn from Ref. 1.)

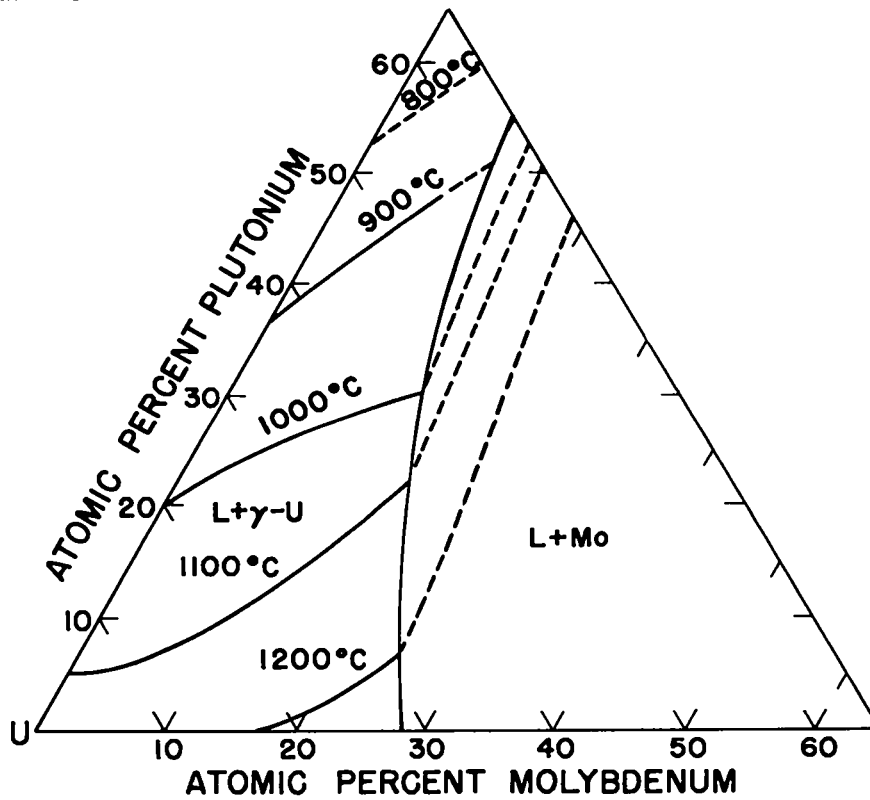


Fig. 60. Plutonium-molybdenum-uranium partial solidus projection. (Redrawn from Ref. 1.)

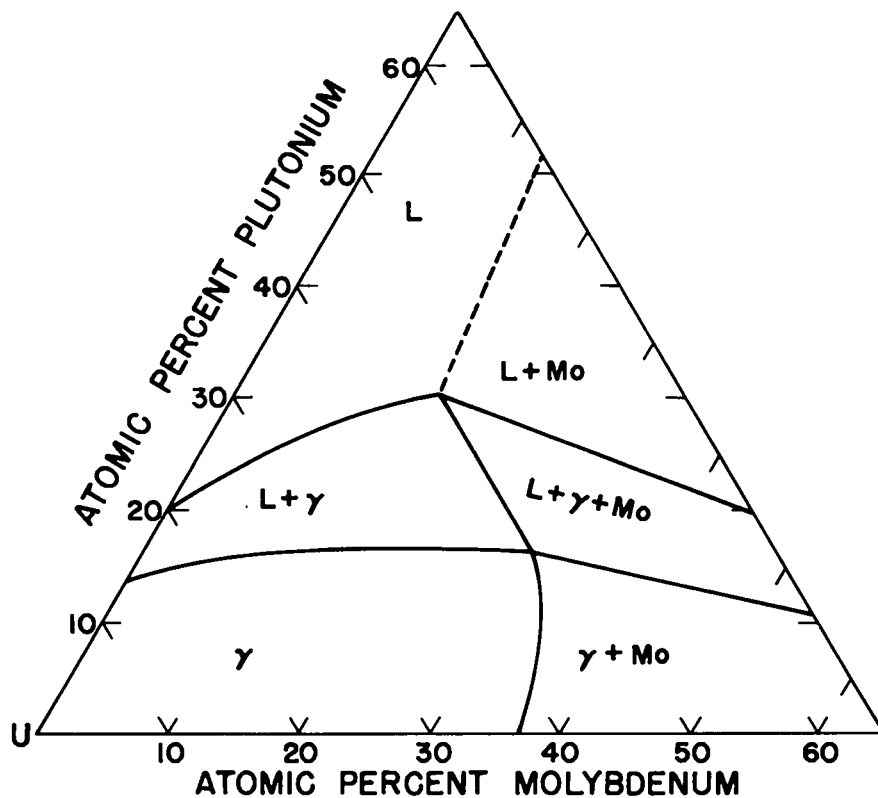


Fig. 61. Plutonium-molybdenum-uranium 1000°C partial isothermal section. (Redrawn from Ref. 1.)

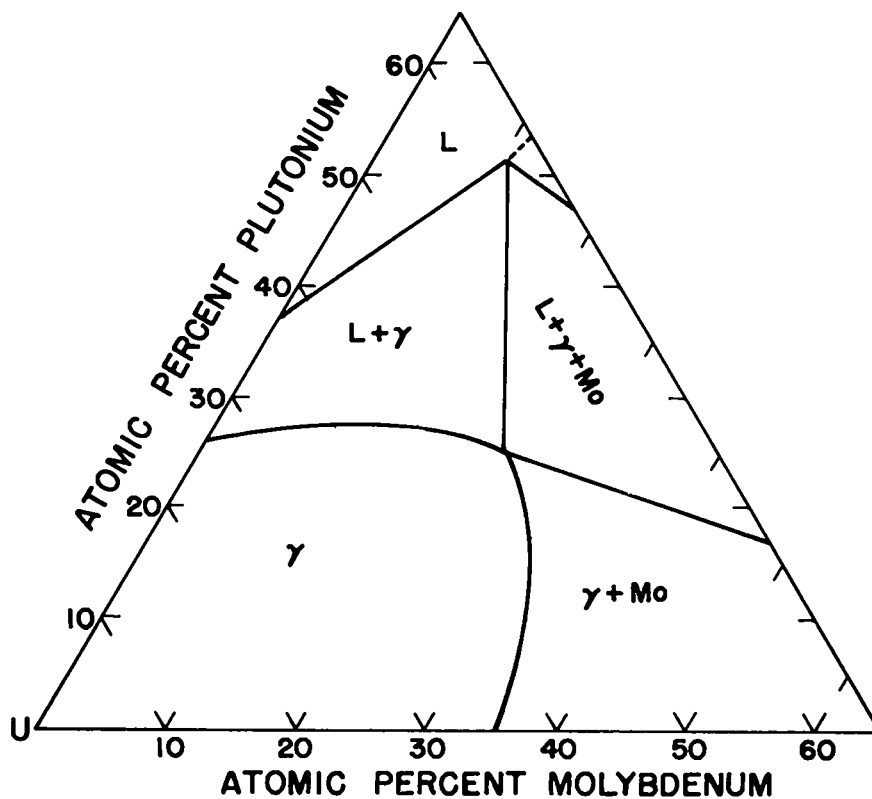


Fig. 62. Plutonium-molybdenum-uranium 900°C partial isothermal section. (Redrawn from Ref. 1.)

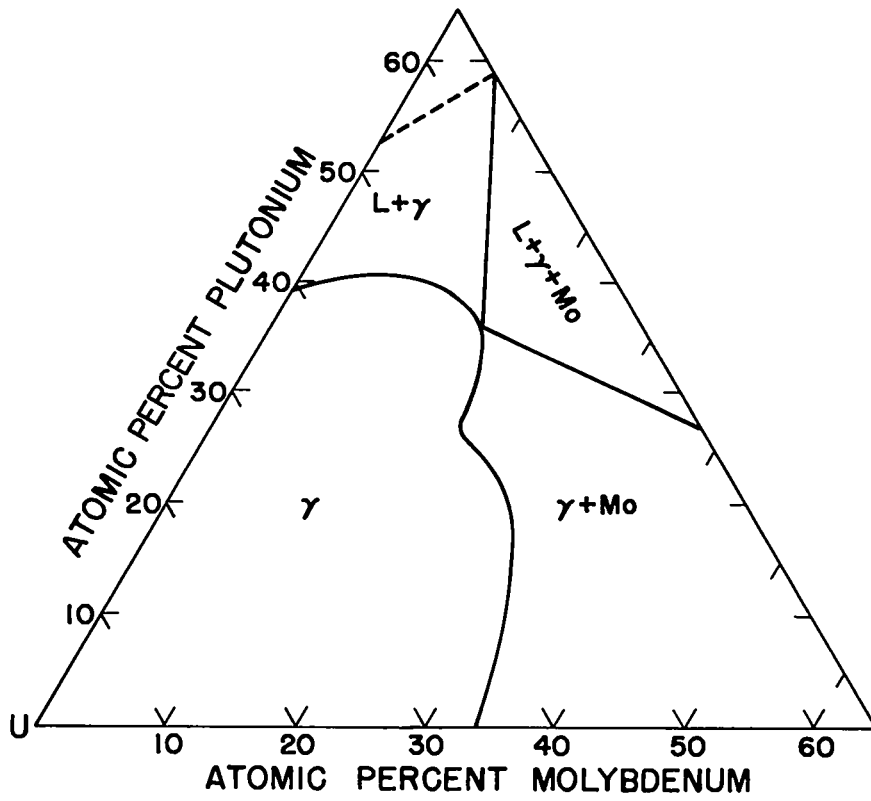


Fig. 63. Plutonium-molybdenum-uranium 800°C partial isothermal section. (Redrawn from Ref. 1.)

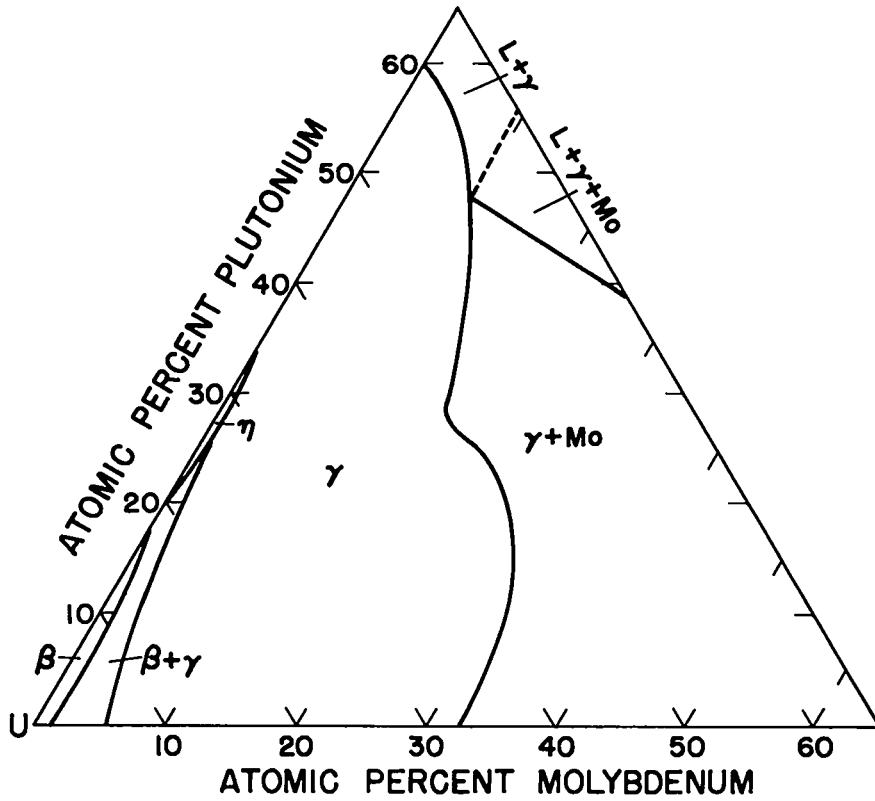


Fig. 64. Plutonium-molybdenum-uranium 700°C partial isothermal section. (Redrawn from Ref. 1.)

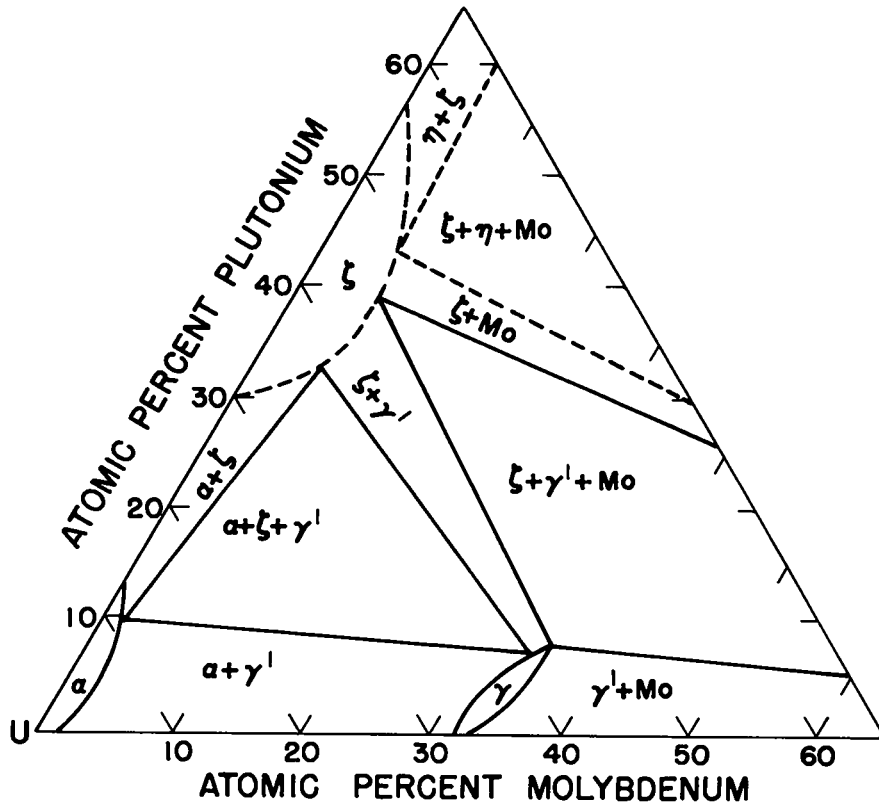


Fig. 65. Plutonium-molybdenum-uranium 450°C partial isothermal section. (Redrawn from Ref. 1.)

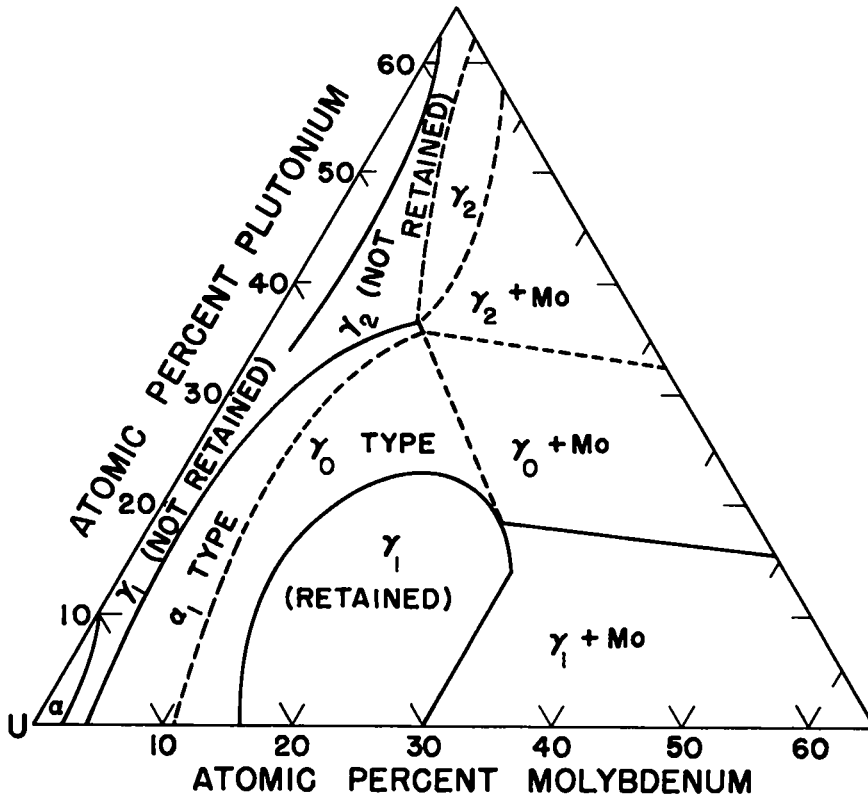


Fig. 66. Plutonium-molybdenum-uranium phases retained at room temperature by quenching from the gamma-phase region. (Redrawn from Ref. 1.)



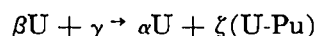
Pu-Nb-U □

Plutonium-Niobium-Uranium

Phase studies on a series of Pu-Nb-U alloys containing 10 wt% Nb and 5, 10, 15, and 20 wt% Pu have been reported by [1], who used x-ray diffraction and metallography. More recently, [2] have investigated the constitution of a series of alloys containing 20 wt% Pu and 10, 20, 30, 40, 50, and 60 wt% Nb by means of differential thermal analysis (DTA), dilatometry, x-ray diffraction, and optical metallography. Two solid-state transformations, one at about 590°C and the other about 630°C, in alloys containing 10, 20, and 30 wt% Nb were reported by [2]. Based on the established reactions in the U-Pu and U-Nb binary systems, the interpretation of the two solid-state transformations is as follows:



630°C Peritectoid Reaction



590°C Class II Reaction

The temperatures chosen for these reactions are the peak temperatures indicated by DTA. Slightly lower transformation temperatures were indicated by the dilatometric data. [1] have established the solidus temperature of U-20 wt% Pu-10 wt% Nb as $1075 \pm 5^\circ\text{C}$ by DTA. No other solidus temperatures have been reported. Tentative isothermal sections at 900°C (Fig. 67), 630°C (Fig. 68), 600°C (Fig. 69), 590°C (Fig. 70), and 350°C (Fig. 71) have been constructed on the basis of the phases reported by [1], and the two four-phase invariant reactions suggested above.

At 900°C (Fig. 67) the $\gamma_1 + \gamma_2$ miscibility gap extends into the ternary system to at least 17.2 at.% Pu. A two-phase alloy containing 17.2 at.% Pu-22.2 at.% Nb-60.6 at.% U consisted of approximately 90% γ_1 and 10% γ_2 as indicated metallographically. On the Nb-rich side of the miscibility gap, a 34.0 at.% U-13.6 at.% Pu-52.4 at.% Nb alloy consisted of approximately 70% γ_2 and 30% γ_1 . The limits of the single-phase regions along the binary sides of the isothermal sections have been taken from the binary diagrams summarized by [3]. The γ_1 and γ_2 lattice parameters for 45.7 at.% U-15.2 at.% Pu-39.1 at.% Nb quenched from 900°C, as determined by [1], are the following:

$$\gamma_1 \quad a_0 = 3.45 \text{ \AA} \text{ (bcc)}$$

$$\gamma_2 \quad a_0 = 3.34 \text{ \AA} \text{ (bcc)}$$

The tentative isothermal section at 630°C shows the peritectoidal formation of $\zeta(\text{U-Pu})$. The formation of the $\zeta(\text{U-Pu})$ phase by a Class III reaction in the Pu-Nb-U system is similar to the peritectoidal formation of ζ in the U-Pu-Mo, U-Pu-Th, and U-Pu-Zr alloy systems. At 600°C (Fig. 69) alloys with less than 63 at.% Nb contained the $\zeta + \gamma_2$ phase. The lattice parameters of the two phases in 17.2 at.% Pu-22.2 at.% Nb-60.6 at.% U alloy annealed at 600°C were determined by [1] as follows:

$$\zeta \quad a_0 = 10.7 \text{ \AA} \text{ (simple cubic)}$$

$$\gamma_2 \quad a_0 = 3.34 \text{ \AA} \text{ (bcc)}$$

The isothermal section at 590°C (Fig. 70) shows the Class II reaction $\beta\text{U} + \gamma\text{U} \rightarrow \alpha\text{U} + \zeta$. With decreasing temperature, the $\alpha\text{U} + \beta\text{U} + \zeta$ region presumably moves toward the U-Pu binary and exits from the ternary system as the eutectoid reaction $\beta\text{U} \rightarrow \alpha\text{U} + \zeta$ at 560°C. Figure 71 shows the boundary between the $\alpha + \gamma_2 + \zeta$ region and the $\gamma_2 + \zeta$ region as established by [1] by metallography and by x-ray diffraction. No ζ phase is found in alloys containing less than approximately 10 at.% Pu. At higher Pu contents, however, ζ appears and increases with increasing Pu content. Physical properties of Pu-Nb-U and other ternary Pu-U-X alloys are given by [4] and [5].

- [1] V. W. Storhok, *et. al.*, in *Progress Relating to Civilian Application*, Battelle Memorial Institute, USAEC Reports BMI-1581 (May 1962), BMI-1589 (July 1962), BMI-1596 (September 1962), BMI-1603 (November 1962) and BMI-1639 (June 1963).
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- [3] R. P. Elliott, *Constitution of Binary Alloys*, First Supplement, McGraw-Hill Book Company, Inc., New York, 1965.
- [4] J. Abgerall, P. Barthelemy, and R. Boucher, "Investigation of the U-Pu-Cb Alloys with 20 wt. % Pu" (in French), French Report CEA-2300, translation available as ANL-Trans.-89 (1964).

- [5] R. Boucher, P. Barthelemy, and C. Milel. "A Study of Plutonium Based Alloys Carried Out at Fontenay-aux-Roses." in *Plutonium 1965*, A. E. Kay and M. B. Waldron (Eds.). p. 489. Chapman and Hall. London (1967).

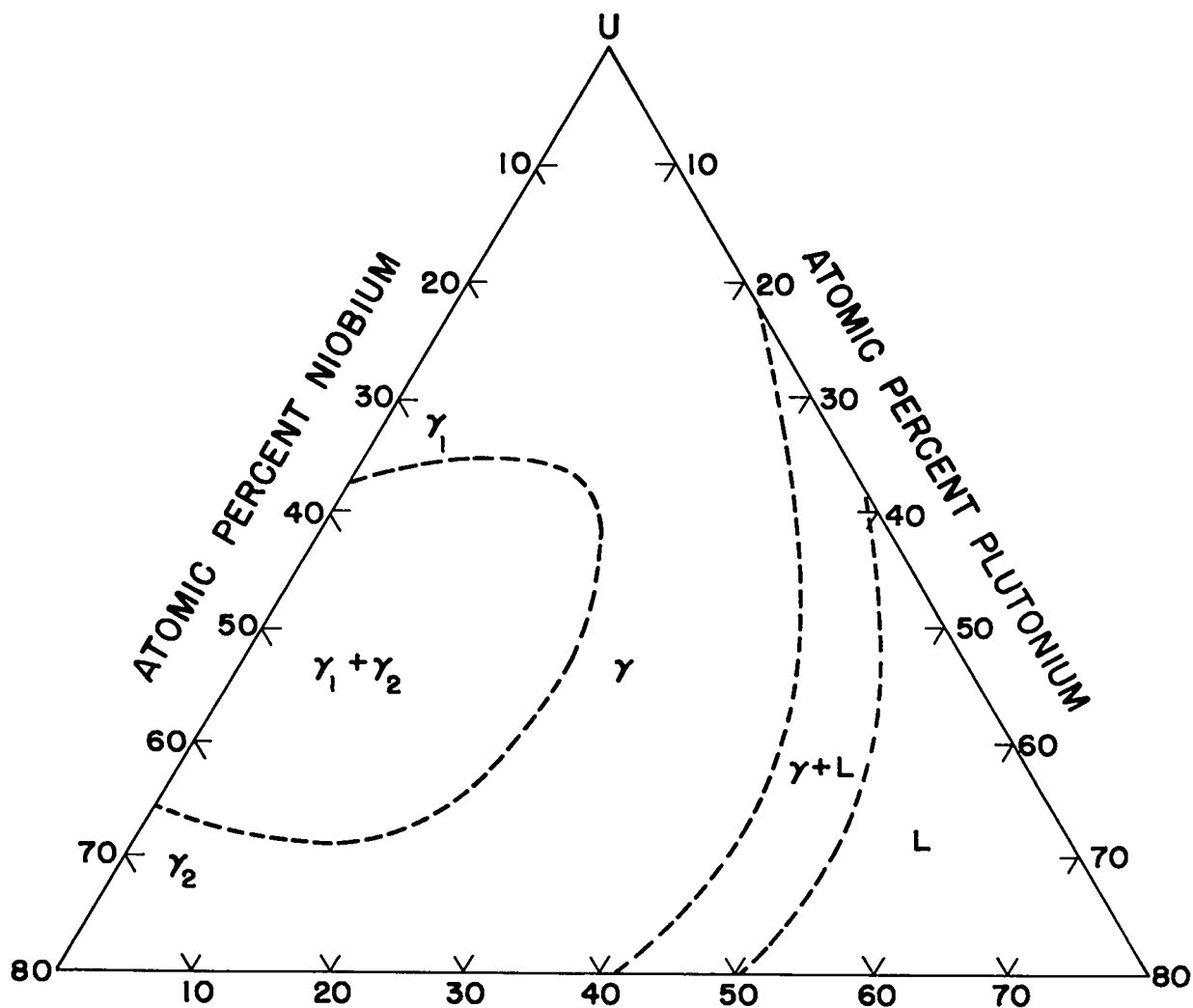


Fig. 67. Plutonium-niobium-uranium 900°C isothermal section (tentative).

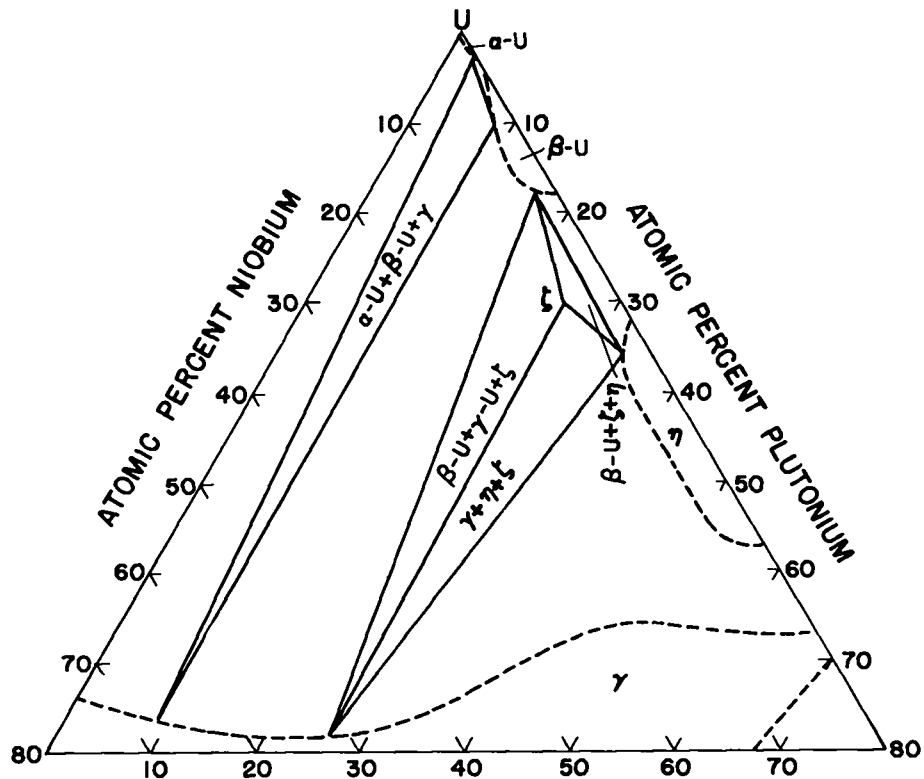


Fig. 68. Plutonium-niobium-uranium 630°C isothermal section showing peritectoidal formation of ζ by the reaction $\beta U + \eta + \gamma \rightarrow \zeta$ (tentative).

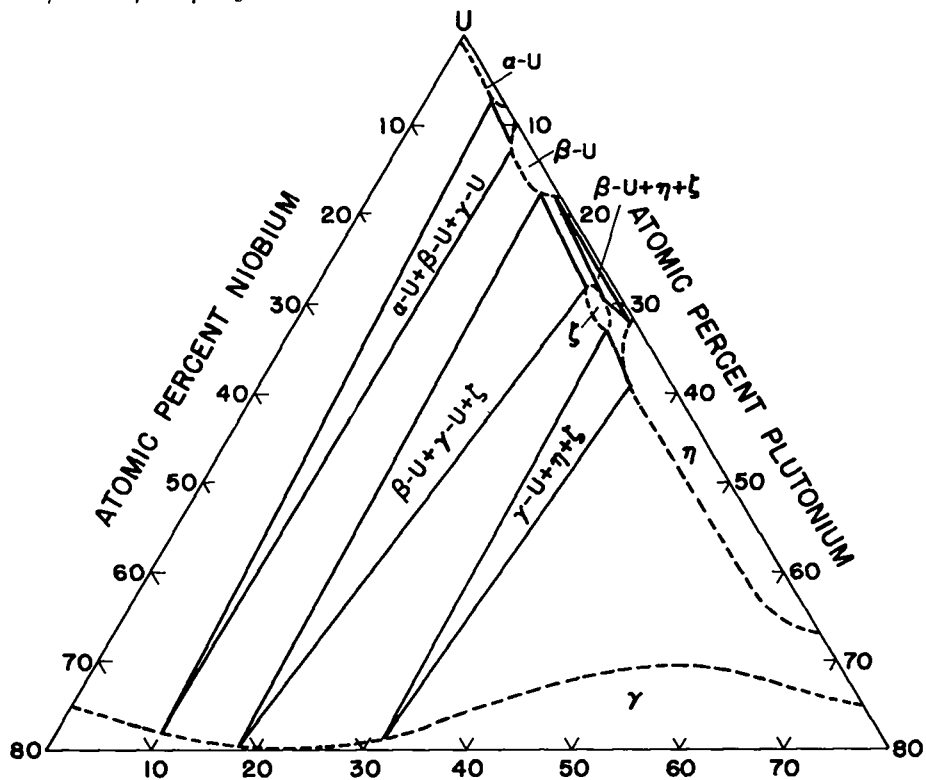


Fig. 69. Plutonium-niobium-uranium 600°C isothermal section (tentative).

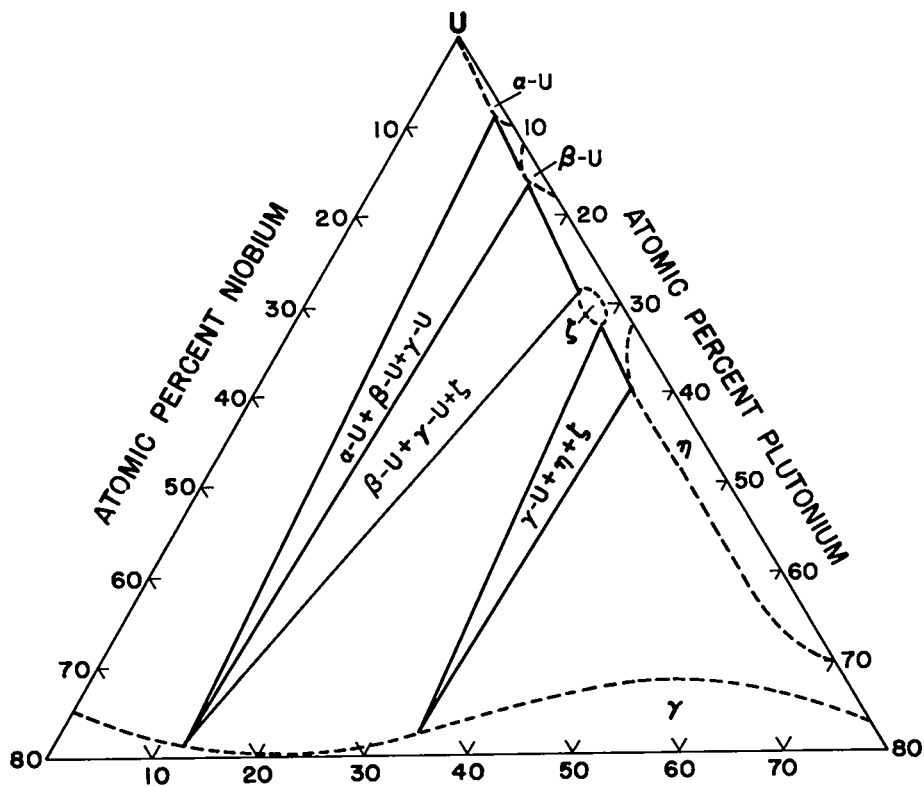


Fig. 70. Plutonium-niobium-uranium 590°C isothermal section (tentative), showing the Class II solid state reaction $\beta + \gamma \rightarrow \alpha + \zeta$.

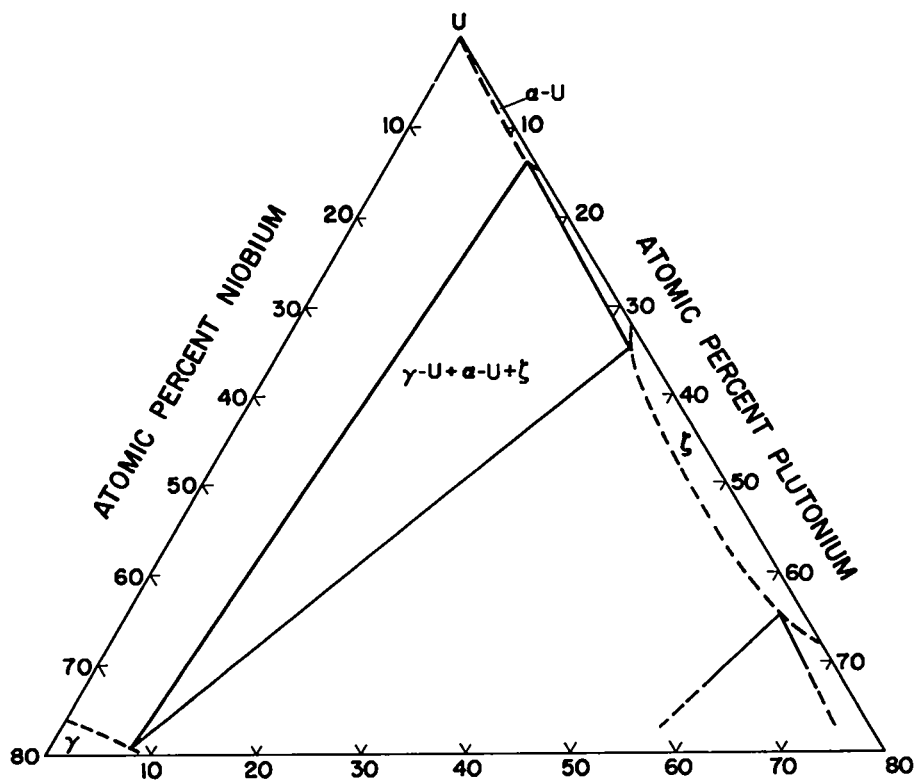


Fig. 71. Plutonium-niobium-uranium 350°C isothermal section (tentative).

Pu-Th-U \square Plutonium-Thorium-Uranium

As part of the USAEC program to develop Th-base fast reactor fuel alloys, phase studies in the Pu-Th-U system have been reported by [1]. Metallography, differential thermal analysis, and x-ray diffraction were used to establish solid-state phase transformations and to outline phase regions principally in the Th corner of the system. The liquidus projection in the Th corner of the system and regions of primary crystallization of β Th, α Th, γ U, and Pu_2Th are shown in Fig. 72. The

$L_1 + L_2$ miscibility gap is believed to be restricted along the Th-U side of the ternary system below which β Th is the first phase to crystallize. In the central region of the system, α Th is the first phase to crystallize from the melt. γ U (ϵ Pu) is the first phase to form from the liquid along the U-Pu side of the system, except for a small region near the Pu corner below 630°C where Pu_2Th is the first phase to freeze. In constructing the liquidus projection, [1] assumed that α Th forms peritectically in the Th-Pu system ($\beta\text{Th} + L \rightarrow \alpha\text{Th}$ at approximately 1400°C) and that the $\alpha\text{Th} + \beta\text{Th} + L$ three-phase region crosses the Th corner of the system with decreasing temperature to exit along the Th-U side at 1265°C.

Below 1086°C. the $\alpha\text{Th} + \gamma\text{U} + L$ three-phase region enters the ternary system from the Th-U binary and moves toward the Th-Pu side with decreasing temperature. The liquid corner of this three-phase region moves along the eutectic trough shown in Fig. 72 toward point P, and the position of the liquid region at 1000°C is shown in Fig. 73. At 900°C, Fig. 74, the $\alpha\text{Th} + \gamma\text{U} + L$ region has moved closer to the Pu corner. Below 760°C, the $\alpha\text{Th} + \beta\text{U} + \gamma\text{U}$ three-phase region moves out from the Th-U-binary and peritectoidally forms η (U-Pu) at 714°C by the invariant reaction $\alpha\text{Th} + \beta\text{U} + \gamma\text{U} \rightarrow \eta$ (U-Pu), as shown in Fig. 75.

Of the three three-phase regions formed at 714°C ($\alpha\text{Th} + \beta\text{U} + \eta$, $\alpha\text{Th} + \eta + \gamma\text{U}$, $\beta\text{U} + \gamma\text{U} + \eta$). the $\beta\text{U} + \gamma\text{U} + \eta$ regions exits along

the U-Pu side as the peritectoid reaction $\beta\text{U} + \gamma\text{U} \rightarrow \eta$ at 710°C and the other two three-phase regions remain in the ternary as shown in the isotherm at 700°C, Fig. 76. The ($\alpha\text{Th} + \gamma\text{U} + L$) three-phase region continues moving toward the Pu corner as the temperature decreases.

The ζ phase is formed by a peritectoid invariant reaction at 610°C as shown in Fig. 77. The phase relationships in the Pu corner of the system are complex and have not been experimentally defined. The 575°C isotherm, Fig. 78. shows the phases along the Th-U side that are stable to room temperature, i.e., alloys with less than 10 at.% plutonium contain $\alpha\text{Th} + \alpha\text{U}$ whereas an increase in Pu content above about 10 at.% results in an increase in the amount of ζ (U-Pu). Below 575°C. the $\alpha\text{Th} + \zeta + \eta$ region moves toward the Pu corner where additional invariant reactions must occur.

The isopleth given as Fig. 79 is a section through the Pu-Th-U system at 10 at.% Pu and shows the effect of U in decreasing the liquidus and solidus temperatures. As shown in the figure. an increase in U from 0 to 2 at.% reduces the solidus temperature from about 1300 to 905°C. Transformation temperatures indicated in the figure were determined by DTA and dilatometric analysis. Density, thermal expansion, thermal conductivity, thermal cycling behavior, compatibility with jacket alloys, and fabricability of Th-rich alloys have been reported by [2, 3, and 4].

- [1] B. Blumenthal, J. E. Sanecki, and D. E. Busch. "Thorium-Uranium-Plutonium Alloys As Potential Fast Power Reactor Fuels: Part I. Th-U-Pu Phase Diagram." USAEC Report ANL-7258 Argonne National Laboratory (in preparation).
- [2] B. Blumenthal *et al.*, Argonne National Laboratory. unpublished work. 1964.
- [3] B. Blumenthal *et al.*, Argonne National Laboratory. unpublished work. 1965.
- [4] B. Blumenthal *et al.*, Argonne National Laboratory. unpublished work, 1966.

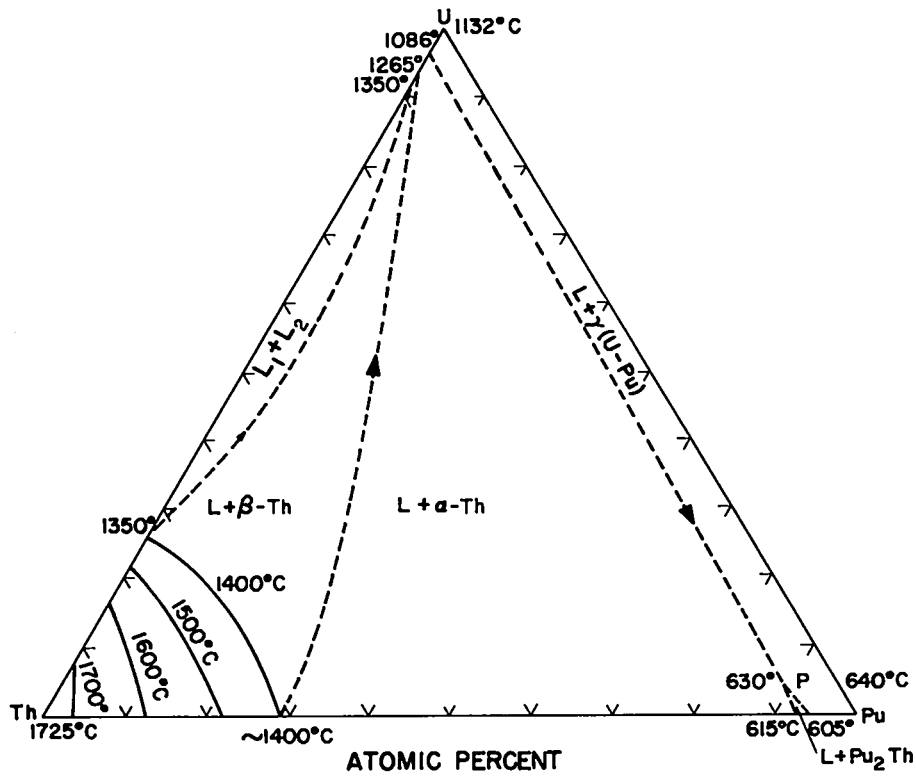


Fig. 72. Plutonium-thorium-uranium liquidus projection and fields of primary crystallization. (Redrawn from Ref. 1.)

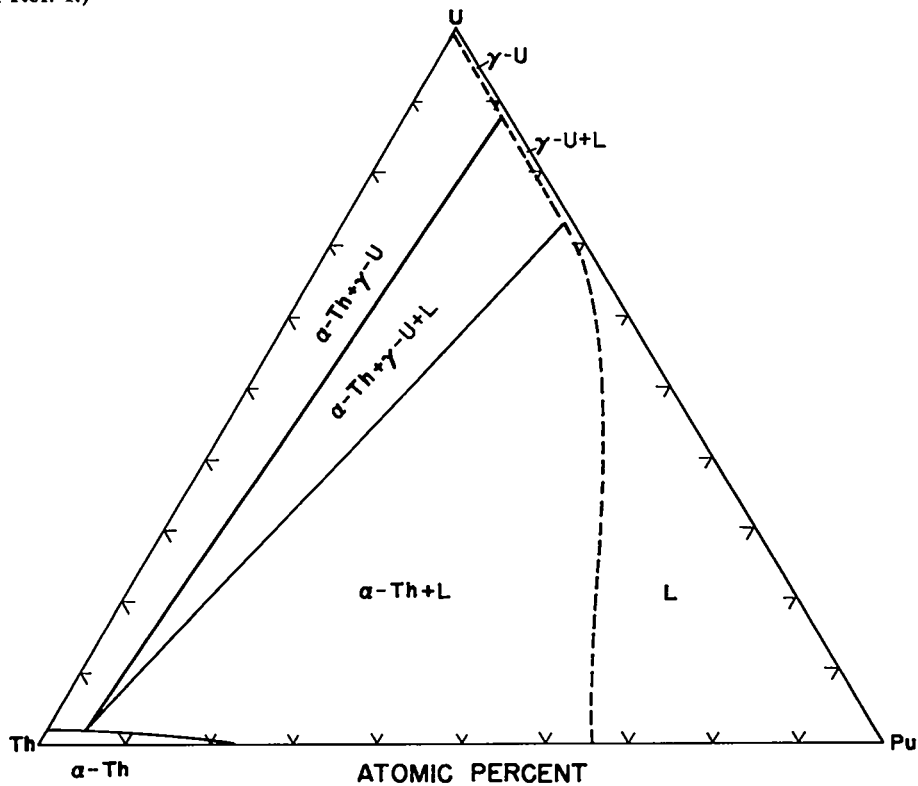


Fig. 73. Plutonium-thorium-uranium 1000°C isothermal section. (Redrawn from Ref. 1.)

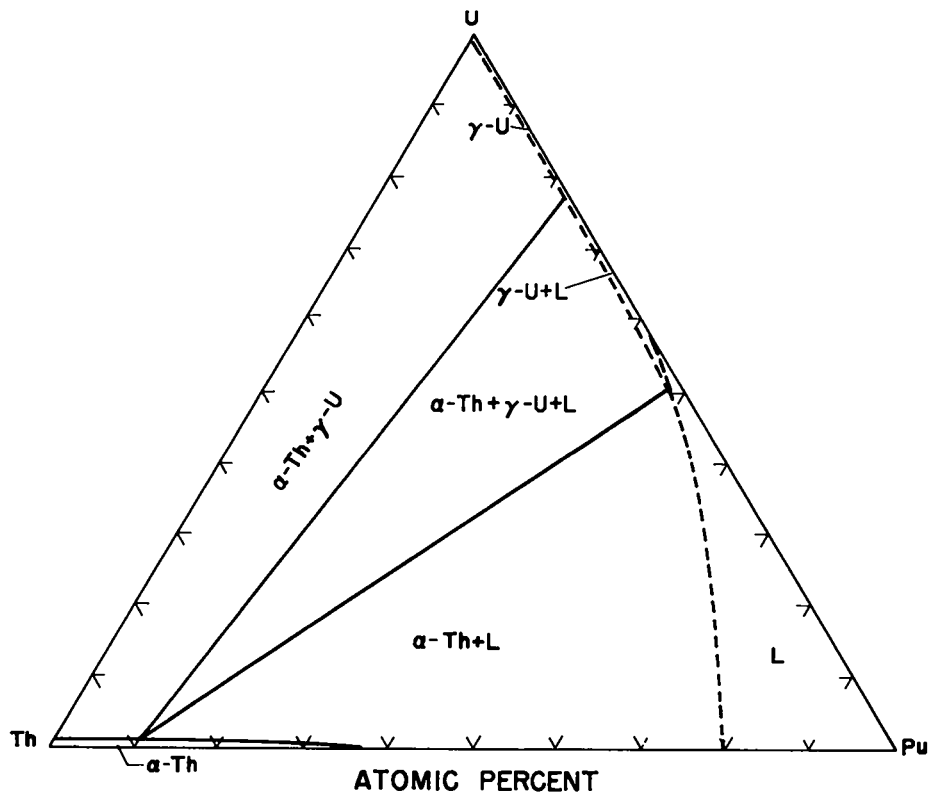


Fig. 74. Plutonium-thorium-uranium 900°C isothermal section. (Redrawn from Ref. 1.)

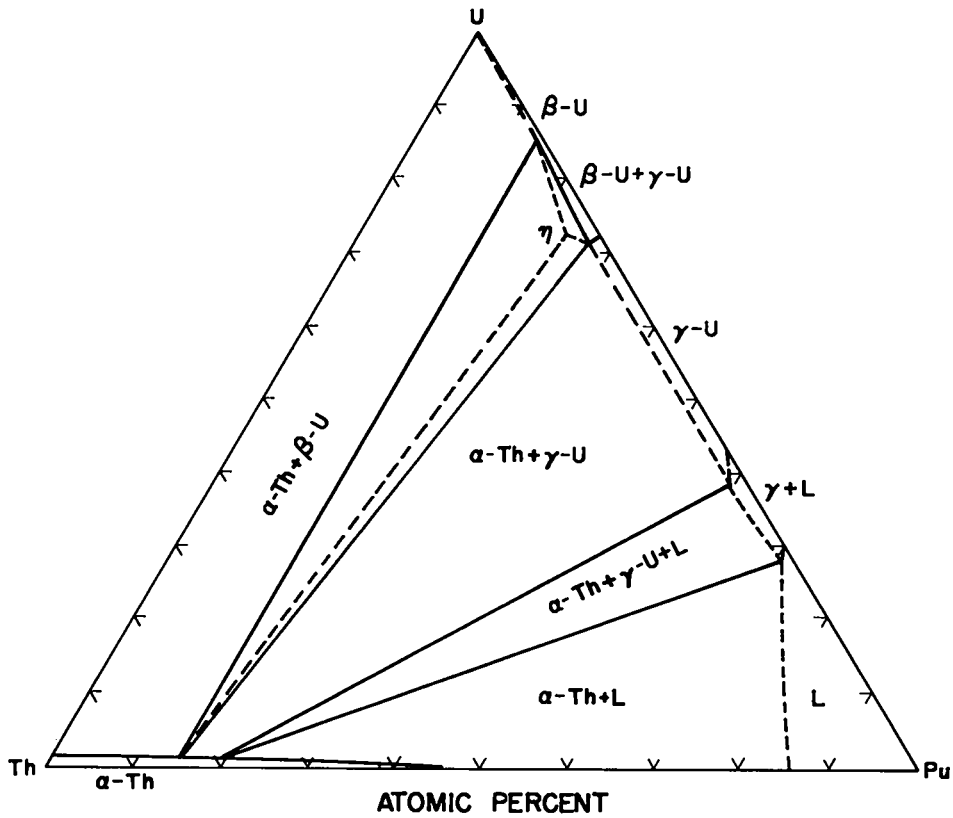


Fig. 75. Plutonium-thorium-uranium 715°C isothermal section, showing peritectoidal formation of η (U-Pu) phase by the reaction $\alpha\text{Th} + \beta\text{U} + \gamma\text{U} \rightarrow \eta$. (Redrawn from Ref. 1.)

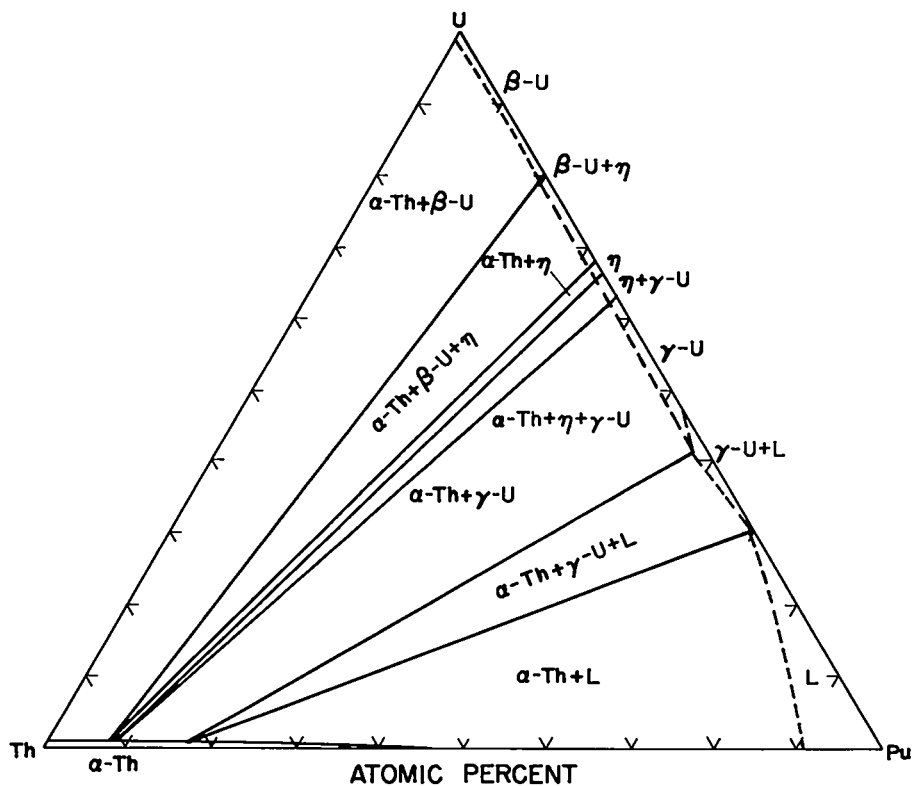


Fig. 76. Plutonium-thorium-uranium 700°C isothermal section. (Redrawn from Ref. 1.)

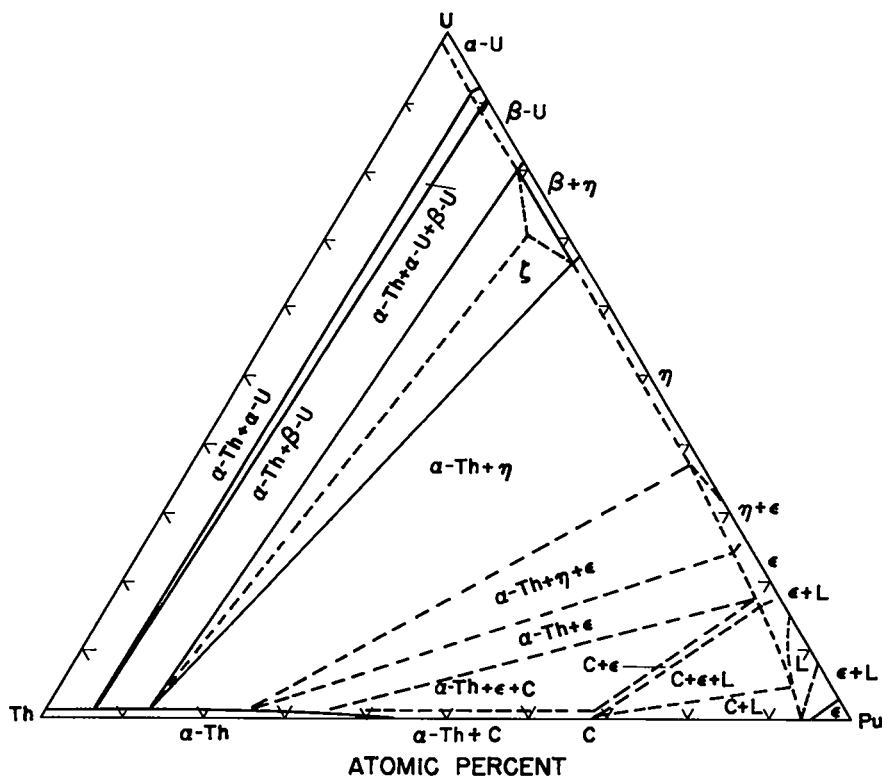


Fig. 77. Plutonium-thorium-uranium 610°C isothermal section, showing peritectoidal formation of ζ (U-Pu) phase by the reaction $\alpha\text{Th} + \beta\text{U} + \eta \rightarrow \zeta$. (Redrawn from Ref. 1.)

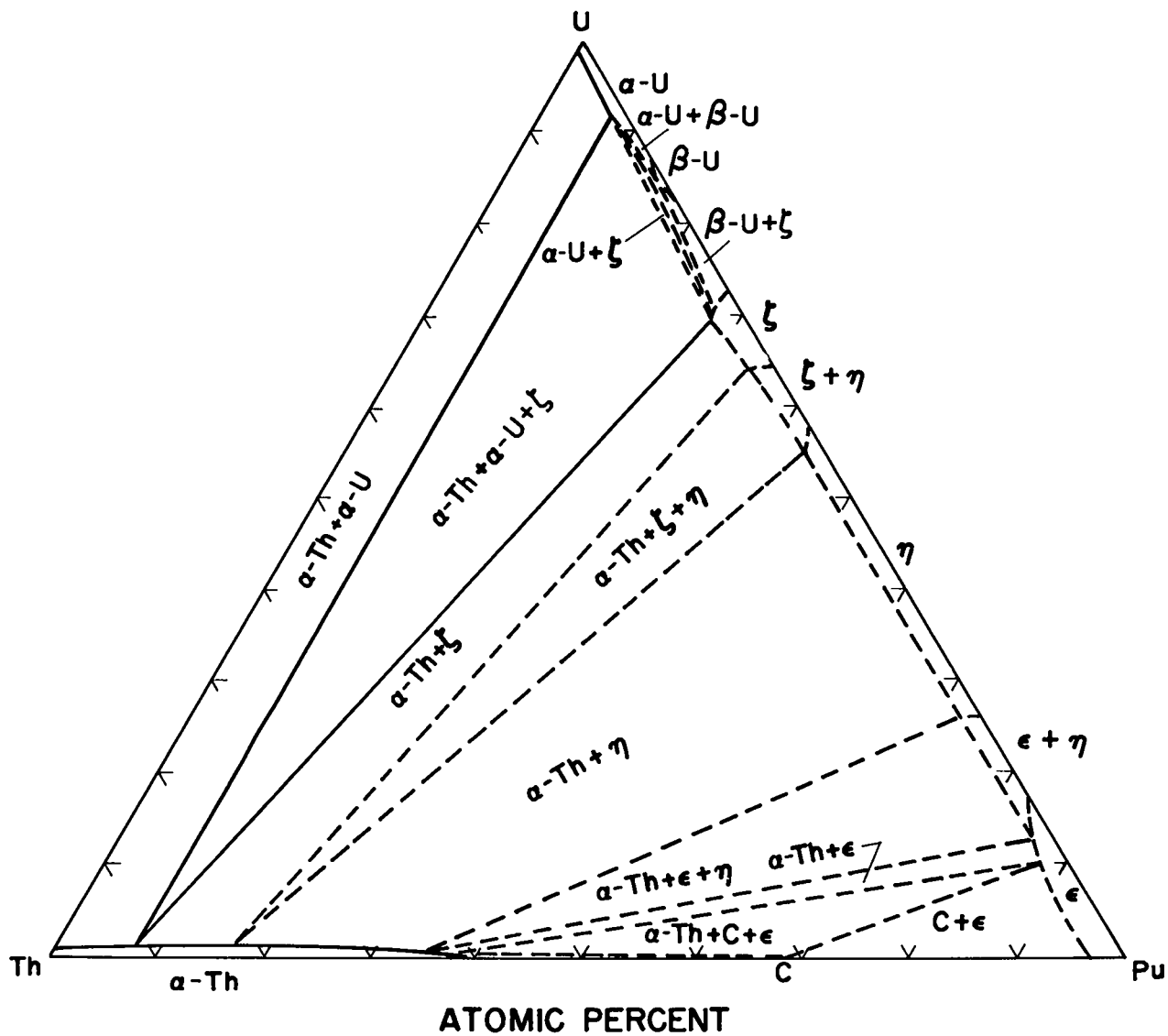


Fig. 78. Plutonium-thorium-uranium 575°C isothermal section. (Redrawn from Ref. 1.)

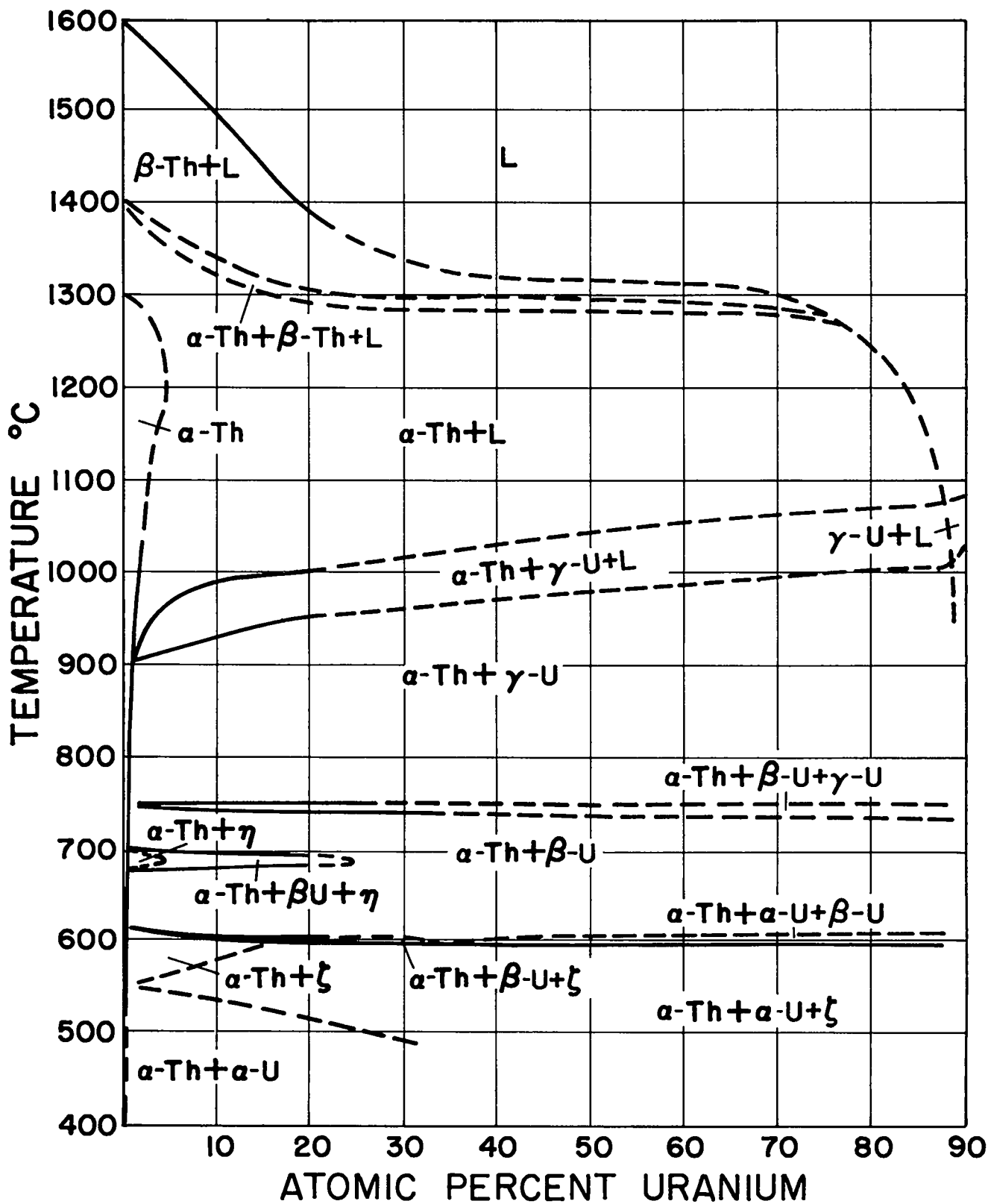


Fig. 79. Plutonium-thorium-uranium isopleth at 10 at.% plutonium. (Redrawn from Ref. 1.)

Pu-Ti-U \square Plutonium-Titanium-Uranium

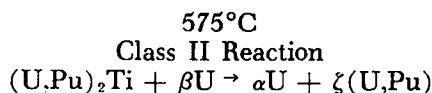
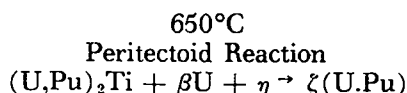
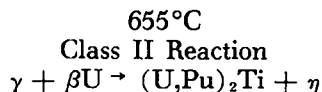
Alloys of interest as fast reactor fuels contain from 10 to 20 wt% Pu and from 7 to 14 wt% Ti, remainder U. Most phase studies in the ternary system have been restricted to alloys which contain more than 40 at.% U. The bcc high-temperature allotropes of both Ti (β Ti) and Pu (ϵ Pu) form a complete series of solid solutions with bcc γ U. Similarly, in the U corner of the ternary system the bcc γ phase is the first phase to freeze from the melt over a wide composition range. In the binary U-Ti system the hexagonal intermediate phase U_2Ti forms by a congruent solid state reaction at 898°C ($\gamma \rightarrow U_2Ti$), according to [1]. U_2Ti is capable of dissolving at least 17 at.% Pu in solid solution and extends sufficiently far into the ternary system as $(U, Pu)_2Ti$ to exert a strong influence on the properties and transformations of U-rich alloys.

Solidus temperatures for alloys containing more than 40 at.% U, as based on binary-solidus data and the metallographic and differential thermal analysis results of [2] for a limited number of ternary alloys, are given in Fig. 80. An increase in Ti at a constant Pu:U ratio causes a moderate increase in the solidus temperature, whereas the solidus temperature of binary U-Ti alloys is strongly reduced by the addition of Pu.

Pu stabilizes the γ phase of U-Ti alloys as shown in Fig. 81, based on the DTA and dilatometric results of [2] and [3]. Below the γ solid solution decomposition temperatures shown in Fig. 81, $(U, Pu)_2Ti$ is the first phase to form over the entire range of compositions. An increase in Pu from 0 to 30 at.% at 33.3 at.% Ti stabilizes the γ phase from 898 to 745°C.

The partial isopleth at 33.3 at.% Ti given in Fig. 82 shows the solvus boundaries between the $(U, Pu)_2Ti$ and γ single-phase regions and the $(U, Pu)_2Ti + \gamma$ two-phase region. Lattice parameter measurements by [4, 5, and 6] indicate that the cell volume of U_2Ti increases with the addition of Pu in solid solution. [4] has reported the lattice constants as $a = 4.832$, $c = 3.8586$ Å for 16.7 at.% Pu-33.3 at.% Ti-50.0 at.% U compared with $a = 4.828$ and $c = 2.847$ Å reported by [1] for the binary U_2Ti compound. Over the composition range shown in Fig. 82, the γ phase is not retained by quenching. The limited metallographic and x-ray evidence of [5] suggests that in the ternary system the single-phase region corresponds to the hyperstoichiometric compound $(U, Pu)_2Ti_{1+x}$.

Invariant four-phase reactions in U-rich alloys have been observed by [3 and 4] at 655, 650, and 575°C. Based on the observed isothermal reactions and on the established binary reactions, the following invariant reactions have been suggested by [7]:



Solid-state transformations along the Pu-Ti side of the ternary system are complex and have not been established. In addition to the equilibrium phases that occur in the system, a commonly observed white extraneous phase has been identified as O-stabilized α Ti by [8]. Data on mechanical properties have been given by [9], and the thermal conductivity, thermal expansion, density, compatibility, and fabricability of various Pu-Ti-U alloys have been reported by [6, 10, 11, and 12].

- [1] A. E. Knapton. "The System U-Ti." *J. Inst. Metals*, **83**: 497-504 (1954/55).
- [2] B. Blumenthal, Argonne National Laboratory, unpublished work, 1966.
- [3] H. V. Rhude, Argonne National Laboratory, unpublished work, 1966.
- [4] B. Blumenthal, Argonne National Laboratory, unpublished work, May 1965.
- [5] "Quarterly Status Report. Advanced Reactor Technology (ART) for the Period Ending January 31, 1965," USAEC Report LA-3244, Los Alamos Scientific Laboratory (February 1965).
- [6] R. Boucher and P. Barthelemy, "Comparison of the Alloys of U-Pu-Mo, U-Pu-Nb, U-Pu-Ti" (in French), French Report CEA-R-2531, translation available as ANL-Trans-138 (1964).
- [7] D. R. O'Boyle, H. V. Rhude, and B. Blumenthal, "Progress in the Development of Fast-Reactor Metal Fuels," *Trans. Am. Nucl. Soc.*, **9(1)**: 1 (1966).

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- [10] L. R. Kelman, H. Savage, C. M. Walter, B. Blumenthal, R. J. Dunworth, and H. V. Rhude, "Status of Metallic Plutonium Fast Power-Breeder Fuels," in *Plutonium 1965*, A. E. Kay and M. B. Waldron (Eds.). pp. 458-484. Chapman and Hall. London (1967).
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- [12] Argonne National Laboratory. unpublished work. 1966.

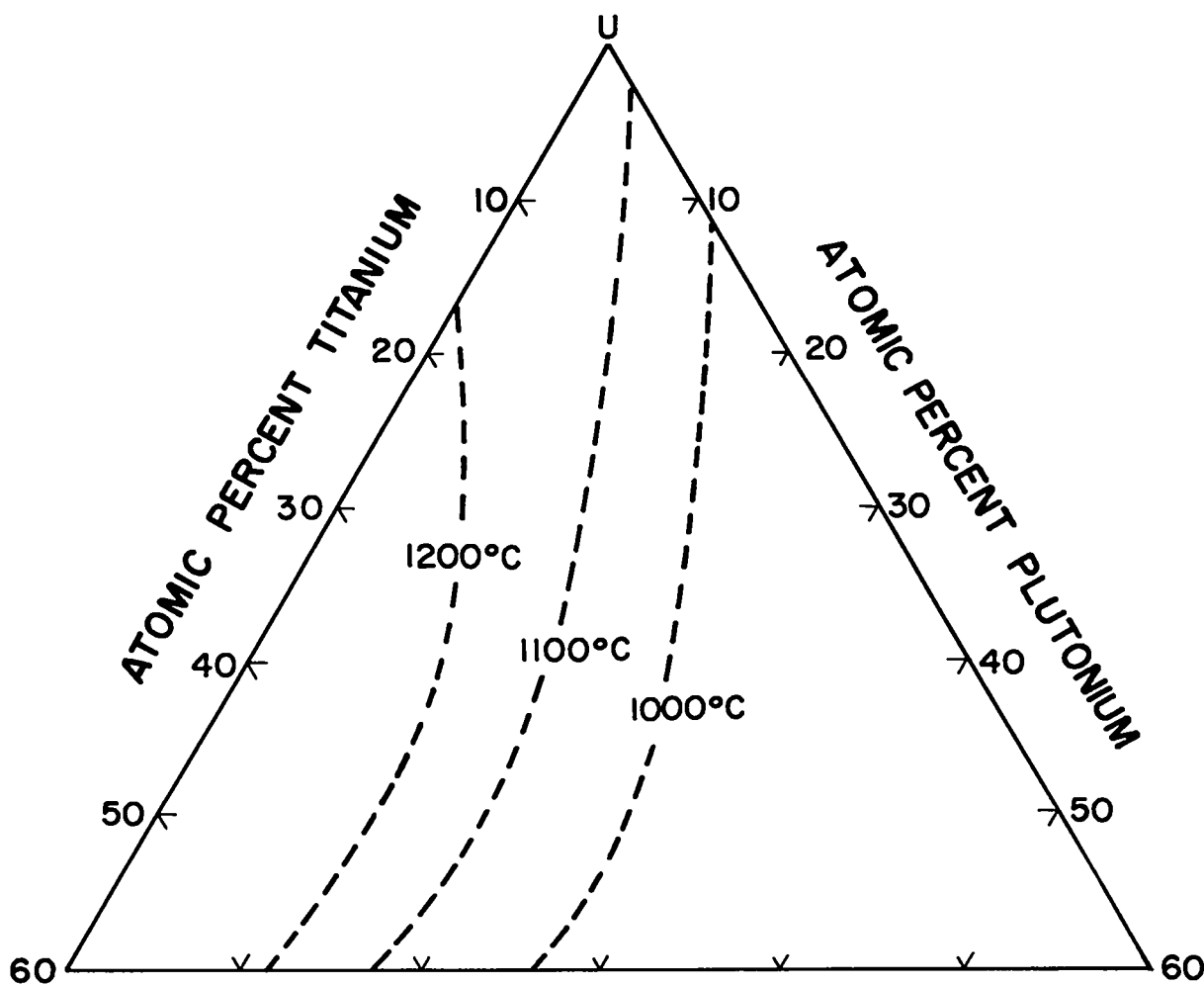


Fig. 80. Plutonium-titanium-uranium solidus projection in the uranium corner of the system.

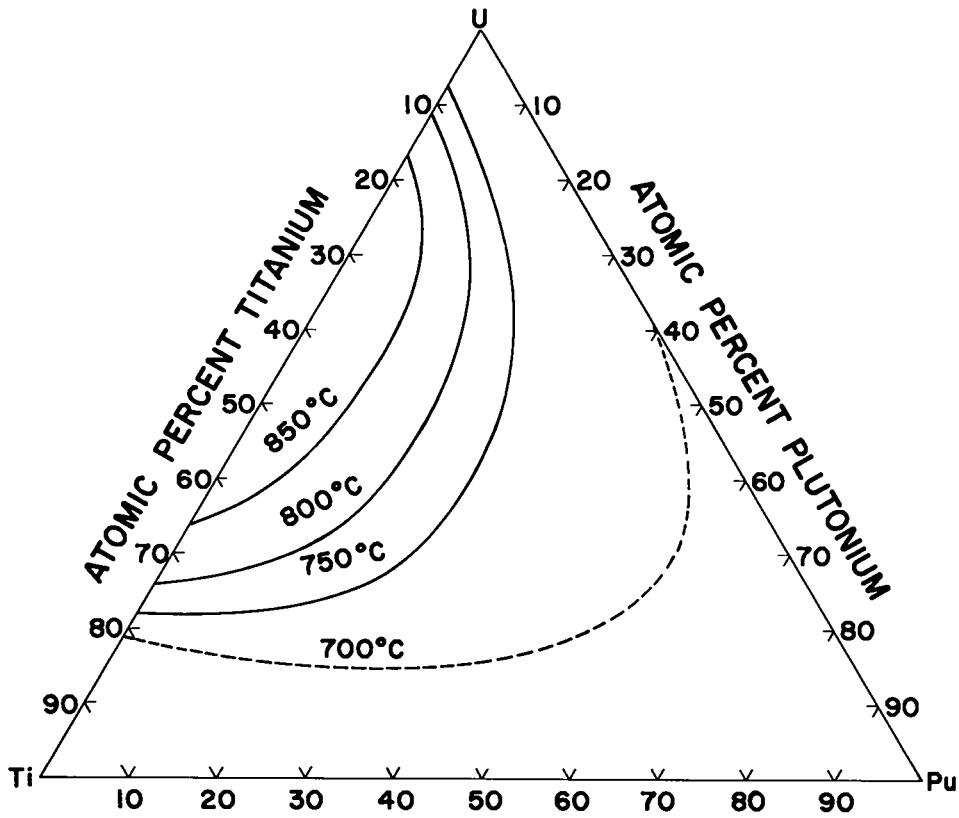


Fig. 81. Plutonium-titanium-uranium gamma solid solution decomposition temperatures.

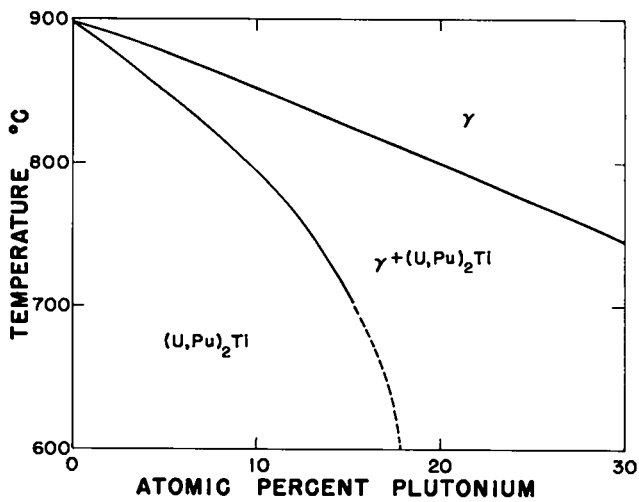


Fig. 82. Plutonium-titanium-uranium partial isopleth at 33.3 at.% titanium.



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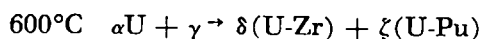
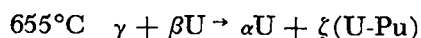
Pu-U-Zr □ Plutonium-Uranium-Zirconium

Early work on Pu-U-Zr alloys of interest as fast reactor fuels, containing 10 to 20 wt% Pu and 10 to 15 wt% Zr, was done by [1] and [2]. Zr increases the solidus temperatures of alloys in the U corner of the system as shown in Fig. 83, based on the work of [3] and [4]. Solidus and liquidus temperatures in the Pu corner of the system have been determined by [5] and are shown in Fig. 84. Both the solidus and the liquidus temperatures of binary U-Pu alloys are increased with the addition of Zr. As shown in Fig. 84, the liquidus temperature increases more rapidly with an increase in Zr than does the solidus. The effect of the minimum point in the solidus and liquidus of binary U-Pu alloys at about 15 at.% U is shown by the curvature of the ternary isotherms in the ternary system, Fig. 84.

The three binary systems show complete solid miscibility of the bcc high-temperature allotropes (γ U, ϵ Pu, β Zr). Similarly, in the ternary system the first phase to freeze over the entire range of compositions is the bcc γ phase. Isotherms showing the γ solvus are presented in Fig. 85 and are based on the work of [4] and [6].

In the U corner of the system both Pu and Zr are effective γ stabilizers. As a result of the increase in solidus temperature in ternary alloys, the bcc γ phase is stable over a wide range of temperatures (300 to 400°C). As temperature decreases, the γ single-phase field moves toward the Zr-Pu binary and then pulls rapidly toward the Pu corner.

In the U corner of the system, invariant reactions at about 655 and 600°C have been observed by [1] and [3]. [7] have suggested that these reactions are Class II-type phase transformations:



In alloys annealed below the invariant reaction at 600°C, [1] have identified the α U + ζ (U-Pu) + δ (U-Zr) phases by x-ray diffraction. In addition to the ternary phases, a light-etching Zr-rich impurity phase has been identified as oxygen-stabilized α Zr by [8].

- [1] R. Boucher and P. Barthelemy, "Comparison of the Alloys U-Pu-Mo, U-Pu-Nb, U-Pu-Ti, U-Pu-Zr" (in French), French Report CEA-R-2531, translation available as ANL-Trans.-138 (1964).
- [2] Argonne National Laboratory, unpublished work. 1965.
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- [8] D. R. O'Boyle, "Discussion on Alloy Systems," in *Plutonium 1965*, A. E. Kay and M. B. Waldron (Eds.), pp. 532-535. Chapman and Hall, London (1967).

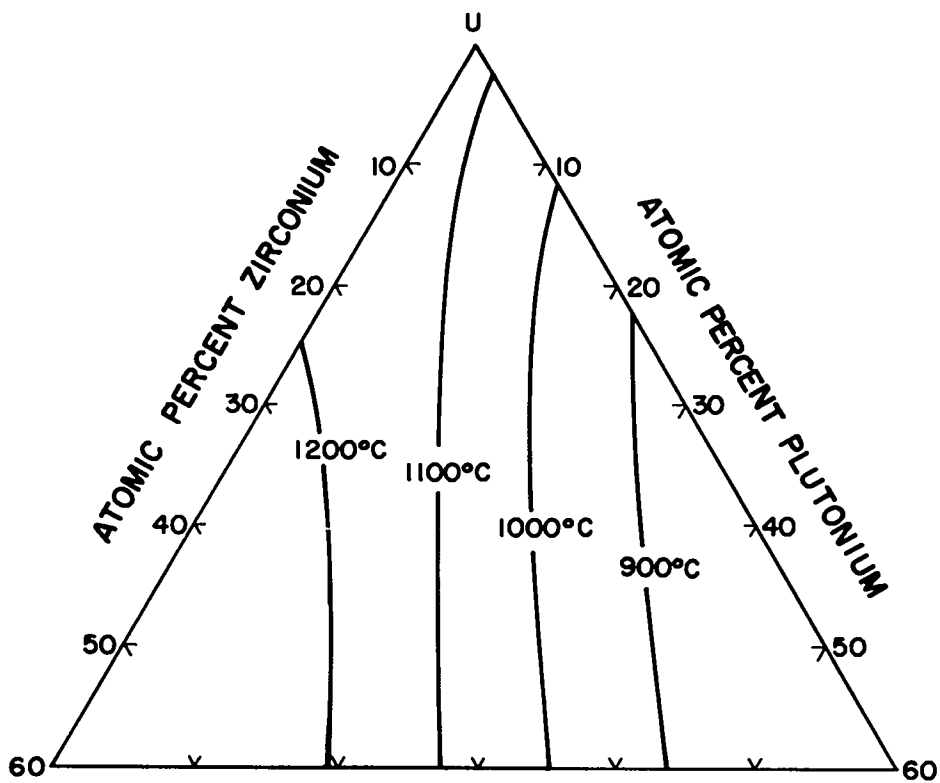


Fig. 83. Plutonium-uranium-zirconium solidus projection in the uranium corner of the system.

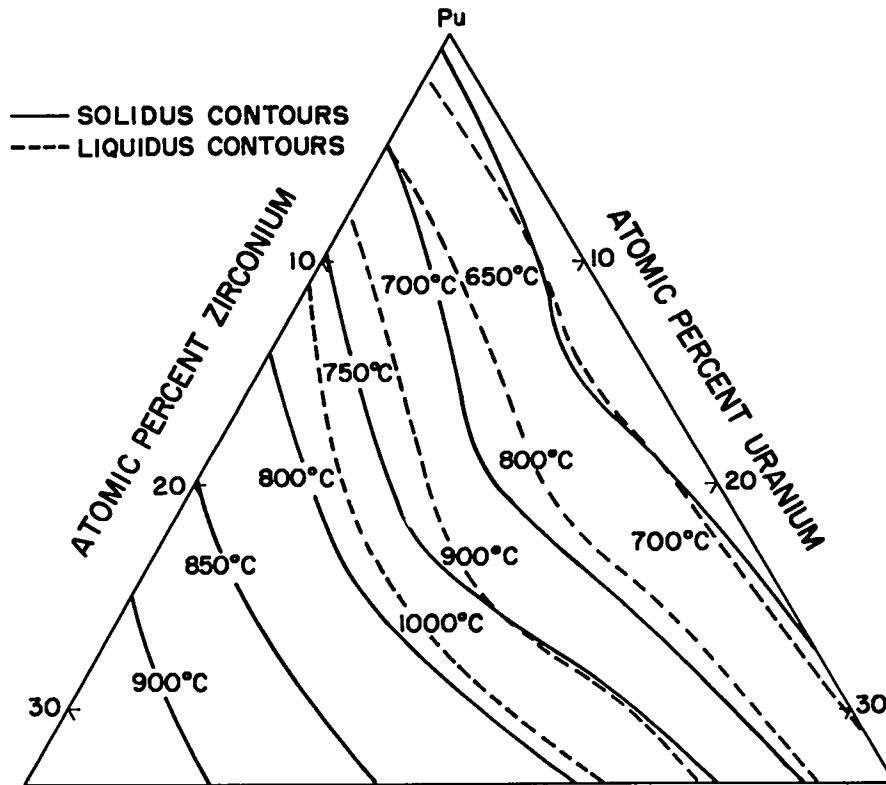


Fig. 84. Plutonium-uranium-zirconium solidus and liquidus projections in the plutonium corner of the system. (Redrawn from Ref. 5.)

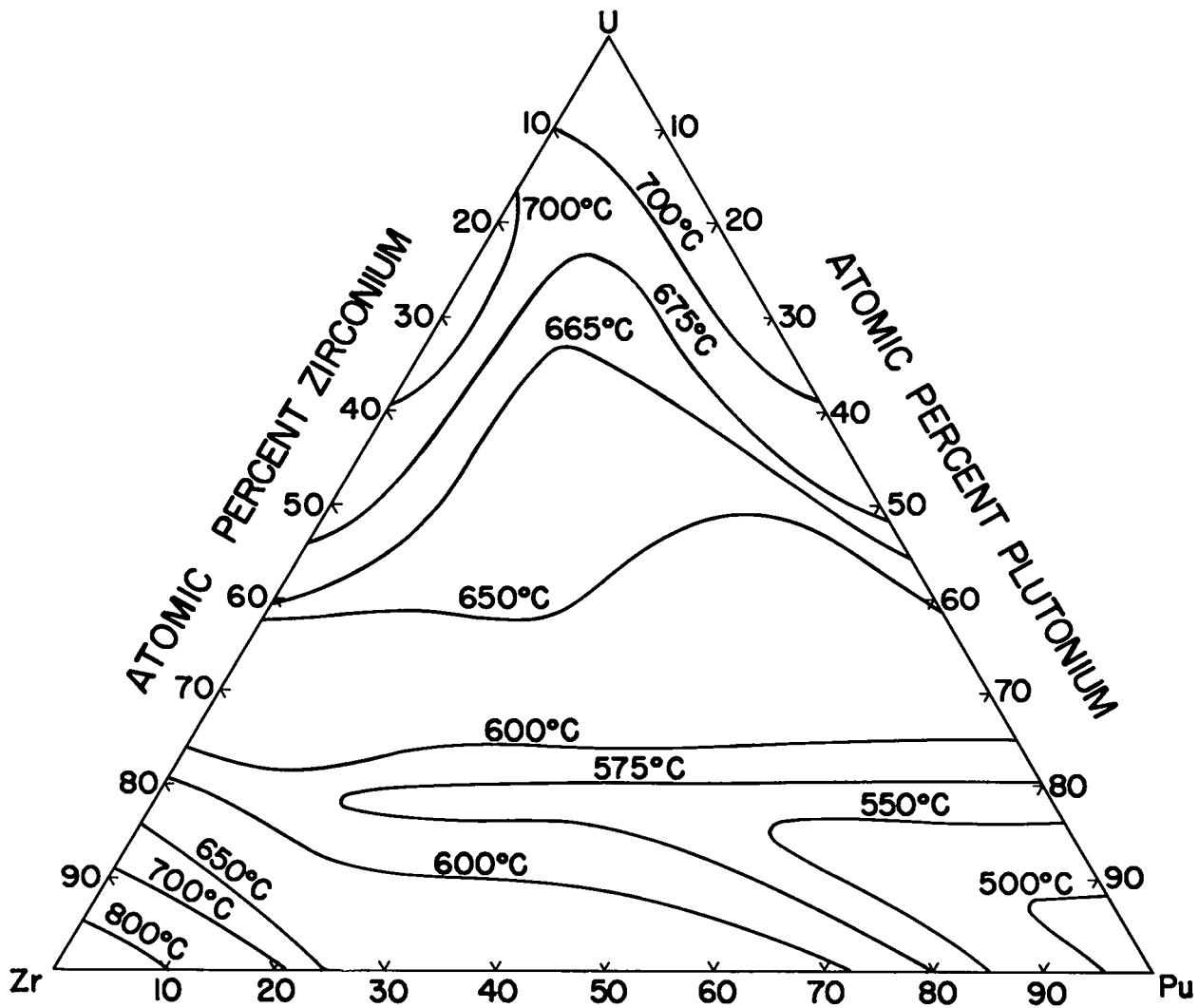


Fig. 85. Plutonium-uranium-zirconium gamma solid solution decomposition temperatures.

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