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LOS ALAMOS SCIENTIFIC LABORATORY
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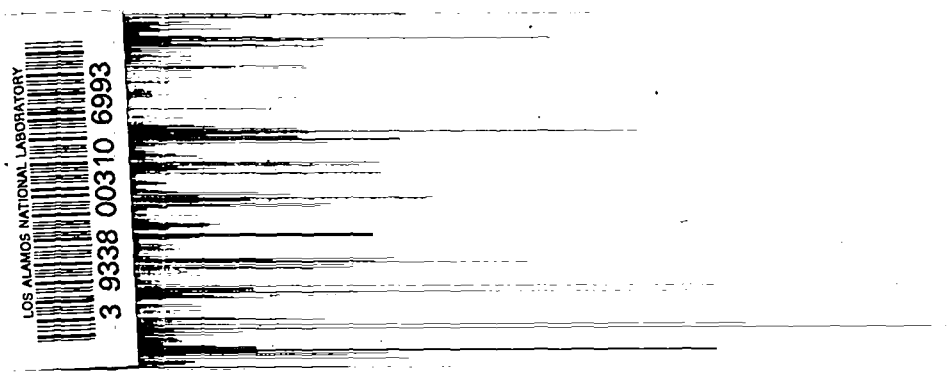
THE SOLUBILITY OF SELECTED ELEMENTS IN LIQUID PLUTONIUM
II. TANTALUM



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THE SOLUBILITY OF SELECTED ELEMENTS IN LIQUID PLUTONIUM II. TANTALUM

by

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Abstract

The solubility of tantalum in liquid plutonium has been measured over the temperature range 700° to 1000°C. The solubility increases from 45 ppm (by weight) Ta at 750° to 1070 ppm Ta at 1000°C. The data fit the emperical equation

$$\log N_{Ta} = 3.06 - 7.50 \times 10^3 T^{-1},$$

where N_{Ta} is the solubility of Ta expressed as the mole fraction and T is the temperature in degrees Kelvin.

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INTRODUCTION

The solubilities of the slightly soluble elements, up to approximately 1 per cent by weight, in liquid plutonium are being studied in the laboratory.[1] The availability of high purity plutonium has made possible measurements that are not affected by interaction with impurity elements.

This report summarizes the investigation of the solubility of tantalum over the temperature range 700° to 1000°C. Jones, Ofte, Rohr and Wittenberg[2] reported no evidence of Ta solubility over the temperature range 645° to 950°C. during a study of the viscosity and density of Pu in this temperature range. Schonfeld[3], however, has predicted that Ta should be appreciably soluble in this range. This study demostrated that Ta is, indeed, sparingly soluble in liquid Pu.

EXPERIMENTAL

Electrorefined plutonium [4] was used throughout this investigation. Analysis of the plutonium is shown in Table 1. The analysis of the tantalum that was employed for this investigation is given in Table 2. In both cases the impurities are minor or not detectable; hence the metals are essentially pure.

Table 1

Analysis of Electrorefined Pu

Spectrochemical		Chemical	
Element	ppm	Element	ppm
Li	< 0.2	Am	15
Be	< 0.1	Fe	< 20
Na	< 10	Pu	99.91%
Mg	< 5		
Ca	< 10	B	< 0.5
Al	< 5	Ni	< 20
La	< 10	U	< 30
Si	< 10	Th	< 15
Pb	< 2	Cl^-	< 10
Cu	< 10	Ta	< 30
Cr	< 10		
Mn	< 2	W	15 ± 5
Sn	< 1	Mo	< 5
Bi	< 1	C[a]	< 10
CO	< 10	N[a]	5 ± 3
Y	< 5	H[a]	5 ± 5
		O[a]	5-20

[a]Typical analysis of electrorefined Pu. This sample was not analyzed for these elements.

Table 2

Analysis of Ta

Element	ppm	Element	ppm	Element	ppm
Li	ND[a]	Cr	< 10	Mo	<100
Be	ND	Mn	< 10	Ag	ND
Na	ND	Fe	<100	Cd	ND
Mg	< 10	Ce	ND	Sn	ND
Al	ND	Ni	<100	Ba	ND
Si	< 10	Cu	<100	Hf	ND
K	ND	Zn	ND	Ta	Major
Ca	ND	Sr	ND	W	ND
Ti	ND	Zr	<100	Pb	ND
V	ND	Cb	<100	Bi	ND

[a]ND is not detected.

The reaction vessels and enclosing furnace tubes were similar to those commonly employed in quenching studies[5]. In several experiments a stainless steel furnace tube with either a quartz or tantalum liner was used. Pu was contacted at a selected temperature for a given time interval with a coupon of Ta in a Mg casting crucible, and then quenched by pouring the melt into a cold casting block. In some experiments, the liquid Pu was contacted with a Ta container crucible and then rapidly cooled to room temperature.

Typical experiments were made by the following procedure. Initially the apparatus, complete except that no Pu was in the reaction crucible, was outgassed under vacuum at 900°C. until the pressure was less than 4×10^{-5} Torr. Then, after cooling, approximately 50 *g.* of Pu was placed in the reaction crucible and the apparatus was evacuated and heated to the selected temperature. During the timed interval at this temperature the entire furnace tube was shaken by means of an external vibrator in order to agitate the liquid Pu.

Samples were broken from the cooled castings (or melts) and submitted for analysis. These were homogeneous, and the two methods appeared to give equivalent results. The castings (or melts) were reused in subsequent solubility measurements.

RESULTS AND CONCLUSIONS

Experiments were made to determine the time required to reach equilibrium between the solid (Ta) phase and the liquid (Pu) phase at each temperature. It was found that Ta was not detected in the castings for intervals of up to 20 hours, and that this time interval decreased with increased temperature. The time dependence experiments at 850°C., which were typical of the study, are summarized in Table 3. In this case no Ta was detected at 22.0 hr. or less at temperature. The solubility was independent of time after 44.0 hr. at temperature.

Table 3

The Solubility of Ta in Pu as a Function of Time at 850°C.

Total Time at 850°, hours	Solubility, ppm Ta
1.0	< 50
3.0	< 35
6.0	< 35
16.0	< 35
22.0	< 50
44.0	220
66.0	210
88.0	215
112.0	195
158.0	205
187.0	215

Solubility measurements were made by several approaches in order to check that equilibrium criteria had been met. In addition to the direct approach from lower temperatures, castings were reused after quenching from a higher temperature. In other cases a melt was held up to 24 hr. at a higher temperature and then cooled to the temperature selected for study. These approaches caused no significant changes in the measured solubility values at these temperatures.

The values obtained from these solubility measurements are shown in tabular form in Table 4. These measurements were made with several different castings of Pu, but each casting was from the same lot that had been analyzed (see Table 1). The average solubility value at each temperature is well within the range that would be expected due to the analytical and experimental errors inherent in this system.

The average solubility data from Table 4 are plotted, as the mole fraction, as a function of 1/T in Fig. 1. The data fit the emperical equation

$$\log N_{Ta} = B + AT^{-1}, \qquad (1)$$

where N_{Ta} is the solubility of Ta expressed as the mole fraction and T is the temperature in degrees Kelvin. From least squares treatment, this equation can be expressed as

$$\log N_{Ta} = 3.06 - 7.50 \times 10^{3}\, T^{-1}.$$

These data fit this equation quite well.

Table 4

The Solubility of Ta in Pu as a Function of Temperature

Temperature, °C.	Hours at Temperature		Solubility, ppm Ta[a]	Mole Fraction Ta, $N_{Ta} \times 10^4$
700	97.0		< 35	----
750	24.0		45	0.59
800	15.5		85	1.12
	18.0		85	1.12
	65.5		80	1.06
		Avg.	83	1.10
850	44.0		220	2.90
	66.0		210	2.77
	88.0		215	2.84
	112.0		195	2.57
	158.0		205	2.70
	215.0		215	2.84
		Avg.	210	2.77
900	16.0		375	4.95
	40.0		385	5.08
	45.0		385	5.08
	88.0		380	5.01
		Avg.	381	5.03
950	16.0		520	6.86
	40.0		620	8.19
	72.0		520	6.86
		Avg.	550	7.26
1000	68.0		1070	14.11

[a] Parts per million parts Pu, by weight.

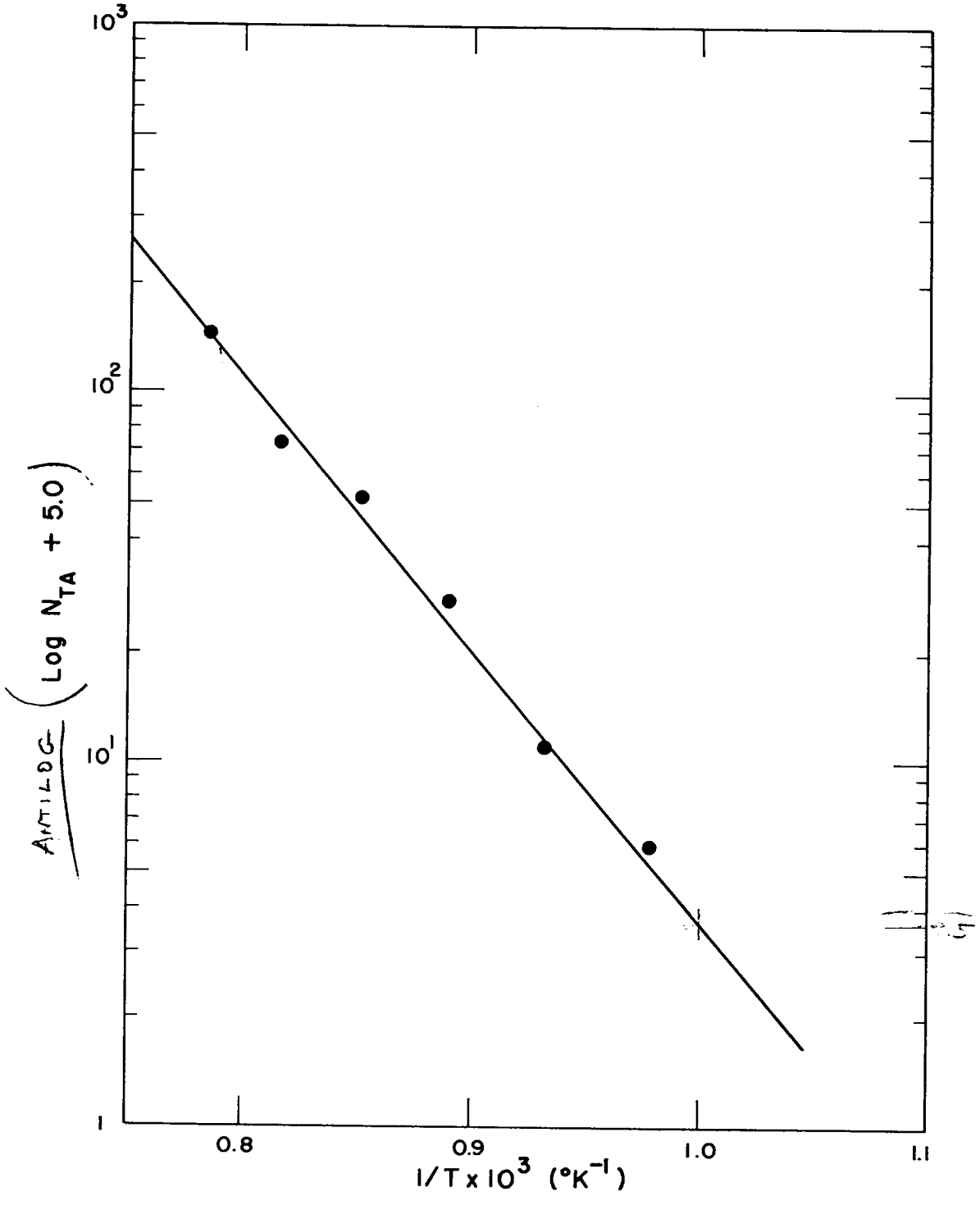

Fig. 1. Plot of log N_{TA} vs. 1/T.

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