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METALLURGY AND CERAMICS

UNITED STATES ATOMIC ENERGY COMMISSION

QUARTERLY SUMMARY RESEARCH REPORT IN METALLURGY

For July, August, September, 1954

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For The Atomic Energy Commission

H. F. Canale
Chief, Declassification Branch *tc*

February 17, 1955

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QUARTERLY SUMMARY RESEARCH REPORT IN METALLURGY

For the period July, August, September, 1954

This report is prepared from material
submitted by the group leaders of
the Laboratory

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METALLURGY

Under the direction of F. H. Spedding, H. A. Wilhelm,
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D. Peterson, and J. F. Smith

1. Purification and Separation Studies

1.1 Niobite-Tantalite Ore Treatment (1)*

Two different test scale fusions were made in which a total of sixteen pounds of niobite-tantalite ore was treated. Both fusions were made in a continuous fusing apparatus. The first fusion involved sodium hydroxide and sodium peroxide with the ore. Only sodium hydroxide and ore were used in the second fusion. An excess of ore was used to facilitate the movement of the charge through the furnace.

The fused products were leached with a hot one-molar sodium hydroxide solution. The residual sodium compounds of niobium and tantalum were converted to their acid oxides by boiling them in a mixture of nitric acid and hydrogen peroxide. A minimum amount of aqueous hydrofluoric acid was then employed to dissolve the acid oxide residue giving an acid fluoride solution of niobium and tantalum and leaving the unreacted ore as a residue to be recycled.

Since the charge was stirred in contact with air as it passed through the furnace tube, the sodium peroxide addition did not seem to be essential. About 2,000 g of combined niobium and tantalum oxides were recovered from 4,500 g of ore after one pass through the furnace. This is roughly equivalent to yields from similar charges heated in a pot.

The continuous fusing apparatus consists of a stainless-steel tube through which the charge is moved by means of a stainless-steel auger. The auger is driven by a variable speed drive that can be controlled at from approximately 0 to 60 R.P.M. A charge is placed in a feeding hopper and fed by another variable speed motor-driven auger into a receiving hopper mounted on the furnace tube. A hinged-type furnace is mounted around the tube for heating and the temperature is measured by thermocouples.

*Numbers indicate group leaders in charge of work. See Appendix III.

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1.2 Separation of Niobium from Tantalum (1)

The tantalum and niobium from the ore treatments described were separated by extractions employing diethylketone on the hydrofluoric acid solution.

1.3 Studies on the Separation of Titanium from Niobium (1)

Previously, small amounts of niobium oxide (0.5 g) spectrographically free of titanium were obtained by extraction of the niobium from a concentrated hydrofluoric acid solution of niobium and titanium by isophorone, and subsequent washing of the isophorone solution with dilute hydrofluoric acid. The niobium was recovered from the organic phase by treatment with ammonium hydroxide solution. However, when larger amounts of materials were employed, the final precipitate of niobium contained spectrographic traces of titanium. It was possible, by employing considerable care, to obtain 2.5 g batches of niobium oxide spectrographically free of titanium by precipitating the niobium from 250 ml of isophorone. However, results are not yet consistently good, indicating the presence of a minor source of contamination yet to be eliminated.

1.4 Studies on the Separation of Hafnium from Zirconium (1)

Work was continued on the separation of hafnium and zirconium by liquid-liquid extraction. It had been noted that upon extracting an aqueous zirconium sulfate solution with 25% Primene JM-T and 10% paraldehyde in hexone there was an enrichment of hafnium in the organic phase. However, as it appeared that this material was very tightly held, experiments were made to see how much could be scrubbed back by acid solutions of various strengths. It was found that upon scrubbing 20 ml of the organic phase from an extraction with 5 ml of 2.0 molar hydrochloric acid, about one-fourth of the material originally in the organic phase was transferred to the scrub solution with a slight gain in purity of the hafnium remaining in the organic phase. Using 8 molar sulfuric acid in place of hydrochloric acid gave only a 3% transfer to the scrub solution.

The effects of replacing Primene JM-T with tertiary octylamine and with tertiary butylamine were investigated. These amines are similar to Primene, but are of lower molecular weight. Replacing Primene with an equivalent amount of tertiary octylamine gave less transfer of zirconium and hafnium to the organic phase and very little separation of the two. Tertiary butylamine dissolved exclusively in the aqueous phase and gave no transfer at all.

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Experiments were resumed with the zirconium sulfate, sulfuric acid, thiocyanate system. Various acid concentrations were tried with three different organic solvents used; hexone, cyclohexanone, and isophorone. The purity of the zirconium remaining in the aqueous phase after extractions with isophorone or cyclohexanone was much better than with hexone.

1.5 Separation of Magnesium from Calcium Metal by Sublimation (2)

An investigation of the preparation of high purity calcium was begun. Attempts have been made to remove magnesium from commercial calcium which contains approximately 3000 ppm Mg. This has been done by sublimation in a vacuum of about 0.1 micron and at temperatures at which magnesium may be preferentially removed because of its slightly higher vapor pressure. As shown in the accompanying table, the magnesium content is noticeably reduced as the sublimation temperature is increased. The sample which had been held at 600°C

Removal of Mg from Ca Metal by Sublimation

Sublimation temp. °C	Time in hrs.	Mg content ppm
425	2	2100
475	2	1700
500	1 1/2	1500
550	2	1200
600	2	700

was then distilled at 900°C and the calcium collected at a condenser temperature of 400-500°C. The magnesium content of this material was 200 ppm. It thus appears that by a combination of low temperature sublimation and maintaining the condenser at relatively high temperatures during distillation, the magnesium content can be reduced to less than 100 ppm.

1.6 Extraction of Zirconium and Molybdenum from Molten Uranium with Silver (2)

A series of experiments has been carried out on the extraction of zirconium and molybdenum from molten uranium with silver. Uranium

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alloys containing 0.01, 0.1 and 1.0 w/o zirconium, and 0.1, 0.5 and 1.0 w/o molybdenum were prepared by arc melting and then contacted with silver at 1200°C by self-induction heating. The immiscible liquids separated upon being heated in a resistance furnace at 1250°C for 3 hours and then were quenched with helium gas. The extractions were carried out in graphite crucibles. As can be seen in the accompanying table, practically none of the zirconium or molybdenum has been extracted into the silver layer. Molybdenum appears to carry some silver into the uranium layer, however. The zirconium has also disappeared from the uranium layer. Microscopic examination revealed that the zirconium had segregated at the interface between the two liquids, apparently having floated to the top of the uranium phase. This was confirmed by spectrographic analyses of the interface region which revealed a concentration of zirconium in that area. X-ray diffraction studies of crystals isolated from that region showed the segregating phase to be ZrC.

2. Preparation of Pure Compounds

2.1 Preparation of Thorium Tetrachloride (5)

The chlorination apparatus designed for continuous preparation of thorium tetrachloride was successfully operated. The exhaust system was capable of removing the exit gases and the heating element maintained the desired operating temperature.

The tube leading from the reaction zone to the product-collecting cyclone settler became plugged with condensed thorium tetrachloride. This material was a loose powder which was successfully removed by intermittent scraping. The discharge gas from the cyclone settler was found to be carrying a considerable amount of the product, probably in the form of extremely small particles. A Saran bag filter was used to collect this material with rather poor success. The recovery was incomplete and after several hours operation, the filter became plugged and prevented the discharge of the exit gases. In addition, since the temperature of the exit gas was about 140°C, the Saran filter cloth underwent rapid deterioration. A preliminary cooling of the exit gas and a more efficient bag filter might be a solution to this problem. The operating temperature in the chlorination zone was 1000-1100°C and after the sixth run the quartz reaction tube was found to be extensively devitrified and had to be replaced. Analyses of the thorium tetrachloride produced in these experiments indicated a product of about 94% purity. The principal impurities are thorium oxide and carbon carried over from the charge.

An investigation was started of the possibility of purifying thorium tetrachloride by sublimation or distillation under vacuum.

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Results of Ag Extraction of Zr and Mo from Uranium

Sample Number	Composition of U alloy, w/o		U phase, w/o			Ag phase, w/o			
	Zr	Mo	Ag	Zr	Mo	U	Ag	Zr	Mo
1-44-1	0.064	-----	0.052	0.01	----	7.32	92.35	0.02	----
1-46-2	0.064	-----	0.058	0.01	----	4.87	93.54	0.02	----
1-47-2	0.065	-----	0.074	0.01	----	7.89	91.58	0.01	----
1-48-3	1.0	-----	0.036	0.01	----	7.26	92.16	0.01	----
1-47-3	1.0	-----	0.110	0.01	----	6.46	93.33	0.02	----
1-47-3A	1.0	-----	0.08	1.0	----	----	----	----	----
1-48-2	1.0	-----	0.082	0.028	----	8.09	91.63	0.01	----
1-48-2A	1.0	-----	0.088	0.380	----	----	----	----	----
1-46-3	----	0.115	0.125	----	0.115	7.01	92.58	----	Not detected
1-50-1	----	0.35	0.095	----	0.40	8.53	91.34	----	Not detected
1-49-2	----	0.84	0.105	----	1.0	9.12	90.76	----	Not detected

A still consisting of a stainless steel tube and a cold finger condenser was assembled. Under a pressure of about 50 microns, a temperature of 700° was necessary to obtain an appreciable rate of vaporization. The starting material analyzed 94% thorium chloride and the product analyzed 99+%. The product was a dense white crystalline material which appeared to be a suitable form for use in the reduction operation. The purification obtained in vacuum distillation appears to be satisfactory and the optimum temperatures and time for the process will be investigated.

3. Metal Preparation Studies

3.1 Preparation of Vanadium Metal (5)

Vanadium powder has been prepared by reducing vanadium tetrachloride with magnesium at about 900°C. The product is a fine metallic powder. Efforts are now being made to improve the purity and increase the particle size of the vanadium.

A reduction was made in which an excess of vanadium tetrachloride was used. This was done in the hope that the particle size of the product could be increased. Some of the vanadium was in the form of needles about 40 by 500 microns in size. However, most of the product was in the particle range of 1-5 microns. The powder was separated by leaching, degassed under vacuum and arc-melted. The metal had a hardness of Rockwell A-48. The carbon content was 600 ppm, nitrogen content was 250 ppm, and the oxygen content was 900 ppm. An iron analysis showed between 0.1 and 0.5% iron. This must have been introduced from the iron crucible used in the reduction.

A reduction was made with a 20% excess of magnesium. A sample of the product treated by leaching had a hardness of A-48 and the same carbon and nitrogen content as the metal from the previous reduction. Samples of vanadium from which the excess magnesium and magnesium chloride were removed by heating under vacuum to 950-1000°C had a higher nitrogen content and a hardness of A-58. The source of this contamination in the vacuum distillation method has not been determined.

3.2 Studies on the Preparation of Tantalum and Niobium Metals (1)

Experimental work on the carbon reduction of tantalum pentoxide was started. No reduction occurs until a temperature of approximately 1700°C is reached in a vacuum. The reduction at this temperature is from the pentoxide to the tetraoxide. It appears that at 1900°C the tetraoxide is reduced to the metal which immediately reacts with any unreacted carbon present to form tantalum carbide. At some temperature between 1900°C and 2300°C tantalum carbide and tantalum oxide react to give the metal. The metal product thus

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far has been somewhat brittle. Similar studies on the carbon reduction of niobium oxide are being continued.

Niobium oxide is readily reduced by a mixture of calcium and aluminum metal to form a low-melting niobium alloy and a low-melting slag. The metal so far obtained by this combination has been quite brittle. Some reductions of niobium oxide by sodium metal have been attempted. The products appear to be lower oxides of niobium.

3.3 Preparation of Thorium Metal (5)

The investigation of the reduction of thorium tetrachloride as a method of producing thorium metal has been hindered by a lack of sufficient quantities of pure thorium tetrachloride. As soon as pure thorium tetrachloride is available, the reduction study will be continued.

4. Alloy Systems

4.1 Uranium-Zinc System (6)

Metallographic, x-ray, and thermal data thus far obtained indicate that the outlines of the U-Zn system correspond to the tentative phase diagram shown in Figure 1 of report ISC-532. The system contains one compound UZn_9 which melts at approximately 1050°C , and a eutectic at about 42 w/o uranium with a melting point of 945°C .

4.2 Thorium-Zirconium System (2)

Work has been resumed on the thorium-zirconium alloy system. High temperature x-ray studies are being started in an effort to clarify one of the questionable high temperature regions of the diagram. Alloys have been prepared and a high temperature x-ray camera is now being readied for operation. The camera is of the design developed by Chiotti of this Laboratory.

4.3 Thorium-Hafnium System (2)

Investigation of the thorium-hafnium phase diagram has continued. A thorium-10 w/o hafnium alloy was prepared by forming a ternary zinc alloy by the bomb co-reduction of HfF_4 and ThF_4 with calcium in the presence of zinc metal. The alloy was then dezincized and alloys containing up to ten per cent hafnium were prepared by arc-melting the required amounts of the 10% master alloy together with thorium. In preparing alloys of higher hafnium content than this, sponge hafnium and sponge thorium were arc-melted together.

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In order to determine the constitution of the phases below 1000°C differential cooling curves were run on alloys containing 5, 10, 20, 30, 50 w/o hafnium. No observable thermal arrests were detected. These alloys were then placed in a resistance furnace and annealed at 950°C for 250 hours and quenched. Microscopic examination of the alloys revealed a two phase structure resembling that of a eutectoid in all the alloys except for the 5 w/o alloy which appeared to be one phase.

Electrical resistivity vs temperature data were obtained on the thorium-50 w/o hafnium alloy. These data indicated some discontinuity in the resistivity at about 1280°C.

4.4 Thorium-Carbon Alloy Studies (6)

The effect of carbon on the $\alpha \rightarrow \beta$ transformation in thorium in the composition range of 0.02 to 0.18 w/o carbon has been fairly well established by electrical resistance measurements. Extrapolation of these data to 0% carbon indicates that pure thorium transforms at 1360 + 10°C. At 0.10 w/o carbon the transformation starts at 1475°C and is complete at 1545°C. At higher carbon content the transformation range is not as sharply defined. However, a sample containing 0.18 w/o carbon gave a fairly well defined break at 1495°C and another break, indicating completion of the transformation, at 1575°C.

4.5 Uranium-Niobium

For niobium contents exceeding 5%, the diagram of this system probably will require only a little refinement. For lower percentages, however, the situation is less satisfactory.

During the quarter just completed, x-ray studies have provided the necessary data for drawing the curve of lattice parameter vs composition for alloys in the solid solution area below the solidus. The curve sags considerably below a straight line between the parameters of the two elements. These studies have established also that the two-phase area of alloys cooled below the one-phase area has its maximum at about 900°C. At its greatest width near 645°C, this area extends approximately from 5 to 45% niobium. The bottom of the two-phase region is formed by the eutectoid horizontal line that extends from below 5% niobium to approximately 45% niobium where it encounters the boundary of the niobium-rich solid solution region.

The horizontal line at 645°C cannot be detected by dilatometric methods in a 2% niobium alloy but probably it does extend to an even lower niobium content. With the 2% alloy the dilatometric curves begin to show prominent transitions in the vicinity of 660°C.

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4.6 Tantalum-Zirconium System (3)

A new investigation of the solidus line was conducted. New alloys were prepared and greater care taken to ensure their homogeneity. The new data show that the solidus has a horizontal portion extending from 2% tantalum or less to more than 40%. From the tantalum end of this line, the solidus rises at an increasing rate until it reaches the melting point of tantalum. The temperature of the horizontal portion is 1830°C. Its existence indicates that the diagram of these two metals must show a eutectic at this temperature. The corresponding composition is in doubt, however. None of the alloys melted completely at the solidus temperature, but the liquidus appears never to be more than 20° or 25°C above the solidus for any niobium content below 50%.

Both dilatometric and resistance-temperature measurements show a eutectoidal horizontal that extends from a few per cent to more than 70% niobium, but its extremities have not been located accurately.

4.7 Zirconium-Zinc System (6)

Further thermal and vapor pressure data on Zr-Zn alloys have been obtained. However, not enough data have been obtained for critical evaluation. Work on this system is currently in progress.

5. Corrosion of Containers by Liquid Alloys

5.1 Corrosion of Refractory Metals (5)

Additions of one weight per cent of iron and of manganese were made to the uranium-chromium eutectic alloy. This was done to determine whether the liquidus temperature of the resulting alloy would be lowered to an extent which would reduce the rate of corrosion at a temperature just above the liquidus. Thermal analysis of the alloy with one per cent iron addition indicated that the solidus temperature was 776°C and the liquidus temperature was 841°C. This lowering of the liquidus was less than expected and necessitated corrosion testing these alloys at 850°C.

Under static conditions at 850°C, the time required for complete penetration of a 5 mil tantalum container by both the alloy with the iron addition and the alloy with the manganese addition was about 30 days. This is about the same rate of corrosion as observed for tantalum by pure uranium-chromium at 900°C. A 5 mil zirconium container was penetrated by both alloys, under the same conditions, in about 21 days. The additions of iron and manganese did not lower the liquidus of the resulting alloys sufficiently to reduce the rate of corrosion by permitting operation at a lower temperature.

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The corrosion of tantalum by uranium-chromium eutectic alloy was investigated in an agitated capsule test. A tantalum tube was partially filled with the alloy and rocked to cause the alloy to flow from one end to the other. One end of the tube was maintained at 900°C and the other end at 1000°C. The most rapid corrosion was observed on the end cap at the hot end of the tube. This cap, which was 10 mils thick, was penetrated in 14 days. This is a corrosion rate approximately twice that observed in static tests.

5.2 Case-Carburized Tantalum (3)

Because of the unsatisfactory performance of every tantalum alloy tried as a containing material for molten uranium-chromium eutectic alloy, attention is being given to coatings, particularly carbide. Small tantalum crucibles have been coated with a thin layer of golden-colored carbide. Microscopic examination of a crucible and its solidified charge after they had been held in a vacuum furnace at 1000°C for 100 hours showed very little evidence of attack. A test of 336 hours on another crucible charge under the same conditions also indicated very little attack but the evidence was less certain. A test of 720 hours was under way at the close of the quarter.

Tantalum crucibles with and without carbide coatings have been tested also for containing the thorium-aluminum eutectic. The results have been generally unsatisfactory but they are not sufficiently definite for conclusions to be stated.

5.3 Metal Coating (for further details and data see ISC-532) (6)

(a) Dip Coating Uranium with Zinc and Cu-9.7 w/o Mg Alloy

Standard four-inch Hanford uranium slugs have been successfully coated with zinc and Cu-9.7 w/o Mg eutectic by the hot-dip method following procedures previously worked out for small uranium specimens. To prevent tongs marks on the coat and to prevent uneven accumulation of coating material on the ends of the slugs, the slugs can be rolled down an inclined steel chute into a quenching bath.

(b) Dip Coating of Aluminum and Zinc with Lead

Attempts to obtain a bond between lead and aluminum which is resistant to the action of boiling water have been unsuccessful. Wetting and bonding can be obtained by dipping the aluminum in lead but the lead coat blisters and peels when exposed to 100°C distilled water.

The possibility that a lead outer coat might be of interest for unbonded fuel elements has led to a study of the application of a lead

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coat over zinc-coated and Cu-9.7 w/o Mg-coated uranium. Results show that the composite coat obtained by hot-dipping is inferior, corrosion-wise, to the initial coat prior to the lead dip. In the case of zinc-coated uranium the lead bath dissolves the zinc coat. Efforts are being made to overcome this difficulty.

5.4 Hot Press Bonding of Aluminum to Other Metals (for further details and data see ISC-532) (6)

Further work has been done on the hot-press bonding of aluminum to magnesium and zinc. All bonds obtained between aluminum and magnesium have been found to be extremely brittle and weak. However, a good bond can be obtained between aluminum and zinc. Hot-press dies are being constructed for preparing bonded couples which can be machined into tensile test specimens for quantitative evaluation of bond strength.

A three piece die for hot-press canning coated slugs in aluminum is also in the process of construction. Some preliminary studies have been made with a small single piece die.

5.5 Corrosion Resistance of Some Magnesium Alloys (for further details and data see ISC-532) (6)

The corrosion resistance of a number of magnesium alloys in 178°C water has been measured. Of the alloys tested 2Al-2Sn-96Mg and 5Al-5Sn-90Mg are the most promising. Whereas pure magnesium samples were completely disintegrated in less than 50 hours, similar samples of these alloys were not completely decomposed in 100 hours.

6. Solid State Investigations

6.1 Physical Constants of Metals from Ultrasonic Measurements (4)

A study of the amount of error to be expected in shear wave velocities as a function of misalignment of a y-cut piezoelectric quartz generator and a metal crystal has been conducted on a zinc single crystal. The direction of propagation was an $\langle hk0 \rangle$ direction perpendicular to the c-axis. The two ideal orientations are such that the atomic displacement vectors are (1) parallel to the c-axis and (2) perpendicular to the c-axis:

$$v_{s\parallel} = \sqrt{C_{44}/\rho}$$

$$v_{s\perp} = \sqrt{\frac{C_{11} - C_{12}}{2\rho}}$$

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where v_s is the shear wave velocity, c is the appropriate elastic constant, and ρ is the crystal density.

The results of the measurements show that the parallel shear wave velocity is far less sensitive to misalignment than the perpendicular shear velocity. The following results indicate this difference in sensitivity:

Vibration	Angle of misalignment	Difference in velocity from 0° misalignment
parallel to c-axis	5°	0.3%
	15°	1.0%
	30°	2.1%
perpendicular to c-axis	1.5°	0.3%
	5°	1.1%
	15°	2.2%
	30°	2.5%
	40°	2.5%

A design for low temperature apparatus to be used in conjunction with the ultrasonic equipment has been completed and has been submitted to the shop for construction.

Difficulty has been encountered in attempts to measure the ultrasonic wave velocities in sodium tungsten bronze, Na_xWO_3 , because of high attenuation of the amplitude of the propagated pulse. However, a value of the shear wave velocity has been measured reproducibly on a sample of sodium concentration, $x \sim 0.78$. Wave propagation was in the $[100]$ direction. The measured velocity was 3.329×10^5 cm/sec. Using the x-ray density this gives a value of $C_{44} \cong 8.1 \times 10^{11}$ dyne/cm². Measurement of longitudinal velocities in sodium tungsten bronze at room temperature has not yet been satisfactorily accomplished. A longitudinal pulse has been propagated through a bronze crystal at liquid nitrogen temperature but the scatter in the numerical values of the velocity is too high to be of use. The difficulty in measurement is due to the high attenuation which is presumably due to the large number of imperfections in the crystal.

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6.2 Structure of Al-V Intermetallic Phases (4)

Work is still in progress on the structures, Al_{11}V and Al_6V . Definite assignment of the α -phase, Al_{11}V , to the space group $\text{Fd}\bar{3}$ or $\text{Fd}3\text{m}$ has not as yet been made because of mechanical difficulties with the Weissenberg camera. However, on the basis of the zero, first, and second layer lines, the space group assignment is still $\text{Fd}3\text{m}$. The Patterson projection $P(x,y)$ for the α -phase indicates that all of the parameters are close to a multiple of one-sixteenth of the lattice parameter. This is consistent with the intensity of the $16,0,0$ and the 880 reflections which are both an order of magnitude larger than any of the other observed reflections. This would give a positive phase sign to the structure factors of these two reflections for the $P(x,y)$ projection of this structure. This conclusion is reached because the parameterless vanadium positions must be at integral multiples of one-sixteenth and there are enough Al-V vectors that peaks should appear in the Patterson projection if the Al atoms were at positions which were $n + 1/2$ multiples of one-sixteenth.

The atomic positions in the structure of Al_6V are still in doubt. However, it is clear that for the 6h and 12k positions in the space group $\text{C6}/\text{mmc}$, the values of the x parameter must be approximately $7/60$, $13/60$, or $27/60$. Interpretation of the vector maps indicates that there should be something at or near $z = 1.5/60$, $6.5/60$, and $15/60$. Then spatially an aluminum set in a 12k set with $x = 13/60$ and $z = 1.5/60$ would be the only 12k set possible with that z parameter. This would allow another 12k set for aluminums with either $x = 7/60$ or $27/60$ at $z = 6.5/60$ and very likely a 6h set at $z = 45/60$ with $x = 13/60$. One set of four vanadiums probably goes into a 4f set with undetermined z parameter. The other four vanadiums could go either into a 4e or a second 4f set. This still leaves eighteen aluminum atoms to place in the trial structure. One definite statement that can be made is that the structure has a layer arrangement in the $[110]$ direction. No other definite conclusions about the structure are possible at this stage of the investigation.

6.3 Calcium Allotropes (4)

A study of the allotropes of calcium has been undertaken. Published information on this subject differs, presumably due to the degree of purity of the calcium used by the investigator. Analysis of the calcium used for the first measurements at this Laboratory showed magnesium (0.43%) to be the major contaminant. Complete analytical results will be included in the next report.

X-ray diffraction patterns and electrical resistivity measurements were taken on this material at various temperatures. The electrical resistivity was not measured above 550°C . A plot of

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electrical resistivity against temperature shows two breaks below 550°C: one between 245° and 285°C and the second between 425° and 510°C. The following values were obtained for this calcium:

resistivity at 16°C 7.94 x 10⁻⁶ ohm cm

temperature coefficient of
resistivity 0.00613 per °C

The x-ray diffraction patterns were taken on a high temperature diffraction camera¹ using a helium atmosphere. The helium was purified by passage over a series of two liquid nitrogen cold traps and then over calcium granules heated to 600°C. The camera was first evacuated to 5 x 10⁻⁵ mm Hg and then the helium was bled into the system. This process was repeated several times before the sample was heated. This elaborate procedure was found to be necessary because of the formation of hydroxide and oxide films on the sample when the above precautions were neglected. However, when the described procedure was used the samples could be kept in the camera for a period of a week with daily thermal cycling without picking up an interfering film.

The room temperature phase was found to be face-centered cubic in agreement with all literature reports. The measured lattice parameter was 5.59 Å. Hull² found the lattice parameter to be 5.560 Å.

A second phase was found in patterns run at 319°C. The following fifteen peaks are characteristic of this phase:

2θ	d	sin ² θ
26.52	3.358	0.05244
27.50	3.241	0.05641
28.20	3.162	0.05934
30.80	2.901	0.07054
34.15	2.624	0.08644
45.60	1.988	0.1502
47.20	1.924	0.1603
52.30	1.748	0.1945
56.05	1.667	0.2209
67.00	1.396	0.3046

¹ P. Chiotti, Rev. Sci. Instr., 25, 683 (1954).

² A. W. Hull, Phys. Rev., (2), 17, 42 (1921).

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2 θ	d	$\sin^2 \theta$
73.50	1.287	0.3576
75.40	1.260	0.3739
90.70	1.083	0.5062
110.15	0.9395	0.6724
118.00	0.8986	0.7348

This phase is definitely non-cubic. Attempts to index the pattern by the analytical methods of Hesse,³ Stosick,⁴ and Lipson⁵ indicate that within the precision of the measurements the symmetry is probably monoclinic or triclinic.

A third phase was found in patterns run at 450°C. The peaks in this pattern could be indexed as a hexagonal close-packed structure with $c = 6.46 \text{ \AA}$ and $a = 3.94 \text{ \AA}$. This compares with $c = 6.54 \text{ \AA}$ and $a = 4.02 \text{ \AA}$ found by Graf.⁶

A fourth phase was found in a pattern at 626°C. Only four peaks were observed in this pattern, but they could be indexed satisfactorily as body-centered cubic with a lattice parameter of $4.47 \text{ \AA}$. Graf has also reported a body-centered structure with $a = 4.48 \text{ \AA}$. He attributed this phase to impurity contamination, as he observed this phase simultaneously with the hexagonal phase. This fourth phase appears at a higher temperature than was reached in the initial resistivity measurements. All four phases were reproducible whether cycling up or cycling down. However, the transformations were somewhat sluggish. It is planned to repeat the resistivity measurements using a larger temperature range and also to repeat all of the work using the purer calcium which is now in preparation.

6.4 Physical Properties of Hafnium Metal (2)

As has been reported previously, hafnium sponge was prepared by the bomb reduction of HfF_4 with calcium in the presence of zinc.

³R. Hesse, Acta Cryst., 1, 200 (1948).

⁴A. J. Stosick, Acta Cryst., 2, 271 (1949).

⁵H. Lipson, Acta Cryst., 2, 43 (1949).

⁶Von L. Graf, Physik., (2), 35, 551 (1934).

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About 65 pounds of the dezinc metal in the form of sponge have been sent to Foote Mineral Company for reprocessing by the iodide method. A pound of this sponge was retained and some of its properties were determined.

The lattice constants of the sponge were determined using a back reflection camera ($\text{CuK}\alpha$ source) and computing the values by the least squares method. The lattice constants are as follows: $a_0 = 3.1991 \text{ \AA}$, $c_0 = 5.072 \text{ \AA}$. The melting point of the arc-melted metal was found to be $2235^\circ\text{C} \pm 5^\circ\text{C}$ based upon two independent determinations. This metal has a hardness of Rockwell 69 A and a density of 13.19 g/cc at 25°C . The microstructure of the arc-melted sponge is shown in Figure 1.

7. Other Investigations

7.1 Reclamation of Zirconium Chips (2)

Zirconium and zircaloy-2 scrap consisting of millings and lathe chips varying from 0.002 in. to 0.004 in. in thickness, heavily coated with oil, was received from the Westinghouse Atomic Power Project. Prior to any cleaning treatment the scrap was washed with acetone to rid it of this oil, and then passed over an electromagnet to remove any magnetic impurities.

It has been assumed that the contaminant of the scrap material is primarily an oxide layer formed on the metal turnings during the machining of the metal, and as previously reported, a leaching treatment with dilute sulfuric acid at elevated temperatures apparently dissolves this oxide layer. A study of leaching time, temperature and concentration of the sulfuric acid used in this treatment was made in order to determine the optimum conditions for this oxide removal. The results of these tests indicate that a 33% (by volume) solution of sulfuric acid for 2 1/2 hours at 120°C provided the best leaching treatment.

This treatment resulted in a weight loss of no greater than 1%. The hardness of the melted scrap material has been used as a criterion for the quality of the metal. Following the leaching treatment, the scrap was pressed into wafers and either arc-melted in an inert atmosphere or vacuum-melted in an ATZ graphite crucible. No significant difference was observed between the hardness of arc-melted and graphite-melted samples. Metal obtained by melting the scrap in a graphite crucible had a hardness of Rockwell 43-45 A. Zircaloy-2 metal treated similarly has a hardness of 52-54 A. Supplementary tests also indicated the acid could be re-used at least four times without increasing the hardness of the final product.

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Figure 1 - Arc-melted Hafnium Sponge,
Etchant: CHNO_3 + a few drops of
HF. X250.

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Chemical analysis was made of the melts of zirconium scrap treated with sulfuric acid and is expressed in the following table.

Analysis of Zirconium Scrap Treated with 33% H_2SO_4

	Graphite Cast ppm	Arc Melted ppm
C	----	100
N	86	----
Si	90	170
Cr	30	30
Mg	5	50
Ni	16	15
Cu	140	16
Ti	20	18
Fe	175	400
Ca	5	15
Al	30	30
Hf	100	100
S*	20	20

*Lower limit of detection is 20 ppm.

In connection with the experimental work with the sulfuric acid leaching of zirconium and zircaloy-2, 148.6 lbs of the scrap material were treated with 33% sulfuric acid for 2 1/2 hours at 120°C, water- and acetone-washed, and air dried. This material is to be shipped to the Bureau of Mines at Albany, Oregon, for vacuum arc-melting and further evaluation.

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The possibility of removing the oxide layer from the zirconium scrap by using zinc as a "getter" material was investigated. The following procedures were employed: (1) alloying the scrap with zinc, then leaching with HCl to remove the zinc and any zinc oxide formed, (2) alloying the scrap with zinc and vacuum distilling the zinc away from the zirconium, (3) dipping the scrap in molten zinc followed by an HCl leach and (4) passing zinc vapors through the scrap at about 1000°C in an evacuated system.

None of these procedures has produced metal with acceptable qualities.

7.2 Correlation of Brinell & Rockwell Hardness Measurements on Zirconium (2)

Both Rockwell and Brinell hardness scales have commonly been used for expressing the hardness of zirconium and zirconium alloys. Recently some confusion has arisen over the conversion of hardness values from one scale to the other for zirconium and its alloys. Both Rockwell A and Brinell (3000 kg load) readings were made on many of the samples which have been prepared in the zirconium scrap reclamation work. Since these samples varied from brittle to quite ductile, it was felt that such a correlation of hardness might be obtained for this material. Measurements were made on samples which have been vacuum melted in an ATZ graphite crucible in a graphite resistor furnace. No additional heat treatment of the alloys was carried out. The tests were run on cylindrical samples 1 1/4" in diameter and 3/8" to 1/2" thick. These data have been plotted in Figure 2 and a curve has been constructed through these points. As can be seen these data agree quite well with a similar curve as determined by Westinghouse, particularly in the lower hardness region of the curve.

As part of this same investigation, a study was made of the reliability of hardness reported on the basis of an average of about seven individual readings as is normally done in measuring the hardness of a material. A uniform zirconium specimen two inches in diameter and one and a half inches high was prepared by melting in graphite. This was cut into three equal sections approximately a half inch thick. 336 Rockwell A readings and 39 Brinell readings were taken and have been recorded in the accompanying table. Readings were taken to the nearest whole number.

A plot of the Rockwell A hardness data is shown in Figure 3 and compared with a normal curve.⁷ The standard deviation was calculated from the equation

$$\frac{\sigma}{\sqrt{n}} \quad \sigma = \sqrt{\frac{\sum x^2}{n - 1}}$$

⁷I. W. Burr, "Analyses and Correlation of Test Data," Metals Progress, 64, 112 (1953).

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Fig. 2

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Rockwell A hardness	No. of readings n	Diff. from mean X	Brinell (3000 kg load)	No. of readings n	Diff. from mean X
40	2	-3.2	114	1	-12.4
41	6	-2.2	116	2	-10.4
42	48	-1.2	118	2	- 8.4
43	164	-0.2	121	5	- 5.4
44	92	0.8	123	9	- 3.4
45	20	1.8	126	13	- 0.4
46	3	2.8	128	6	1.6
47	1	3.8	131	1	4.6
Mean	43.2		126		
Limits of Accuracy	± 0.05		± 0.7		
Standard Deviation	± 0.93		± 4.5		

The 336 Rockwell readings were so grouped that 90 groups of seven each were obtained corresponding to 90 individual hardness measurements as they are usually determined in the zirconium recovery investigations.

The arithmetic mean was calculated for each of these groups of seven and the standard deviation from the true mean of 43.2 was calculated for these group averages. This standard deviation was ± 0.15 . Thus on the basis of this study it was concluded that use of the average of 7 Rockwell A readings was sufficiently accurate for the purposes intended.

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Fig. 3

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7.3 Vacuum Fusion Analysis of Metals (5)

Samples of three of the National Bureau of Standards cooperative steel samples were analyzed by vacuum fusion. The determination of the composition of the evolved gas by mass spectrometry was not completed due to instrument trouble in the mass spectrometer. However, the amount of gas evolved checked very closely with the reported analyses of all three samples. The precision of the results on each sample was excellent. This indicates that the furnace and collecting system is operating properly.

An investigation of the vacuum fusion analysis of the rare earth metals for oxygen, nitrogen and hydrogen was begun. A series of samples of lanthanum metal was prepared by melting lanthanum metal under vacuum by self induction in calcium fluoride crucibles. After the molten metal was outgassed, the furnace was closed and a known quantity of oxygen or nitrogen was introduced and allowed to react with the molten metal. When the reaction was complete, the sample was cooled and examined. The samples containing the larger amounts of oxygen and nitrogen were covered with a crust or scale, which indicates that the intended composition may have been above the liquid solubility limit. These samples will be analyzed to determine whether quantitative recovery of the added element is achieved.

7.4 The Preparation and Testing of Some Mold Dressings in Uranium Casting (1)

Steel molds for casting uranium rods were treated with different dressing materials. Uranium rods were cast in these molds and the quality of the rods compared. A water slip of alumina, a water slip of zirconia and benzene smoke received most study as coating materials for the steel mold walls.

It was found that the alumina and zirconia had to be calcined at a high temperature to give a slip deposit that would dry essentially free of water at 90°C. The quality of the outside surface of rods cast in the alundum-treated molds was not as good as for the zirconia and smoke treatments. The relative merits of the zirconia and smoke treatments of the steel molds for uranium rods is to be determined.

7.5 Melting Uranium in Oxide-Painted Graphite Crucibles (1)

Small charges of about one pound each of uranium metal were melted in graphite crucibles painted on the inside with water suspensions of various oxides.

The maximum temperature employed in the melting was just above that ordinarily required in production melting for casting of uranium.

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Comparisons of the various oxide treatments were made in a qualitative way by inspection of the crucible, the metal and its microstructure after the melting operation.

Oxides that appeared to give favorable protection of the metal from reaction with the container were oxides of thorium, zirconium, uranium and magnesium. On the basis of the qualitative information, melts of 325 pounds each were made under normal casting conditions with oxides of zirconium, uranium and magnesium being employed as paints on the inside surface of the graphite crucibles. The few such meltings made so far show zirconium oxide as best of these three with uranium oxide next and magnesium oxide a poor third as far as carbon pick-up is concerned. The thorium oxide has not yet been tested in the production casting set-up but on the basis of the small scale tests, it would be expected to rate quite favorably with the other oxides tested in this manner.

• 7.6 Low Melting Fuel Element Studies (1,4)

Combinations of uranium and thorium dissolved in aluminum and/or magnesium are being studied to determine if a low melting alloy containing both uranium and thorium in concentrations suitable for use as liquid atomic fuel material can be obtained. Quaternary alloys containing the mixtures of the binary eutectics of uranium-aluminum and of thorium-magnesium are being studied. The ternary alloys containing mixtures of uranium, thorium and aluminum as well as low melting quaternary alloys of the above four elements are to be studied.

8. Rare Earth Metallurgy

This work is reported in ISC-530.

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APPENDIX I: LIST OF REPORTS FROM THE AMES LABORATORY

1. Reports for Cooperating Laboratories

- ISC-437 (rev) G. H. Beyer, D. R. Spink, J. B. West and H. A. Wilhelm. Caustic Treatment of Zircon Sand. UNCLASSIFIED.
- ISC-443 Philip Phipps and D. J. Zaffarano. The Half-Lives of Some Short-Lived Low Z Nuclei Formed by Photonuclear Reactions. UNCLASSIFIED.
- ISC-468 A. L. Loeffler, Jr. and B. J. Ruth. Mechanism of Hindered Settling and Fluidization. UNCLASSIFIED.
- ISC-474 G. H. Murphy, D. F. Young, and R. J. Burian. Progress Report on Friction Loss of Slurries in Straight Tubes. UNCLASSIFIED.
- ISC-482 M. E. Dooley and D. F. Atkins. A New Electronic Vacuum Dilatometer. UNCLASSIFIED.
- ISC-484 Ames Laboratory Staff. Chemistry. Quarterly Summary Research Report. January, February, March, 1954. SECRET.
- ISC-485 Ames Laboratory Staff. Metallurgy. Quarterly Summary Research Report. January, February, March, 1954. SECRET.
- ISC-486 Compiled by P. Chiotti and O. N. Carlson. Hanford Slug Program. Quarterly Summary Research Report. January, February, March, 1954. SECRET.
- ISC-487 Ames Laboratory Staff. Physics. Quarterly Summary Research Report. January, February, March, 1954. UNCLASSIFIED.
- ISC-488 D. R. Wilder and E. S. Fitzsimmons. Further Study of Sintering Phenomena. UNCLASSIFIED.
- ISC-490 A. J. Bureau, H. A. Austrheim, and D. J. Zaffarano. An Electron Injector for a 70 Mev Synchrotron. UNCLASSIFIED.
- ISC-491 J. T. Williams. The Vanadium-Zirconium Alloy System. UNCLASSIFIED.
- ISC-492 K. W. Olson and G. C. Danielson. Variations of Electrophotoluminescence Phenomena with Frequency of Applied Field. UNCLASSIFIED.

- ISC-494 M. R. Nadler and E. S. Fitzsimmons. Preparation and Properties of Calcium Zirconate. UNCLASSIFIED.
- ISC-500 B. A. Rogers and D. F. Atkins. The Zirconium-Columbium Diagram. UNCLASSIFIED.
- ISC-501 M. Margoshes and V. A. Fassel. Infrared Functional Group Analysis of Aryl Silanes. UNCLASSIFIED.
- ISC-502 M. Margoshes and V. A. Fassel. The Infrared Spectra of Aromatic Compounds. I. The Out-of-Plane C-H Bending Vibrations in the Region $625-900\text{ cm}^{-1}$. UNCLASSIFIED.
- ISC-504 Ames Laboratory Staff. Engineering. Quarterly Summary Research Report. April, May, June, 1954. SECRET.
- ISC-505 Ames Laboratory Staff. Chemistry. Quarterly Summary Research Report. April, May, June, 1954. CONFIDENTIAL.
- ISC-508 Ames Laboratory Staff. Physics. Quarterly Summary Research Report. April, May, June, 1954. UNCLASSIFIED.
- ISC-514 D. R. Wilder and E. S. Fitzsimmons. Improved Calcium Fluoride Shapes. UNCLASSIFIED.
- ISC-516 V. A. Fassel, B. Quinney, L. C. Krotz and C. F. Lentz. Quantitative Spectrographic Analysis of the Rare Earth Elements. UNCLASSIFIED.

2. Papers Published in Scientific Journals

- Altshuler, S. and J. F. Carlson
Time Dependent Variational Principle. Phys. Rev. 95, 546-548 (1954).
- Chiotti, P.
Adaptation of a Geiger-Counter X-Ray Diffractometer for High Temperature Investigations. Rev. Sci. Instr. 25, 683-688 (1954).
- Dickinson, J. M., O. N. Carlson and H. A. Wilhelm
The Niobium-Vanadium Alloy System. J. of Metals 6, 915-918 (1954).
- Duke, F. R.
Qualitative Theory of Electron-Transfer Mechanism. Record of Chem. Prog. 15, 55-59 (1954).
- Duke, F. R. and V. C. Bulgryn
The Kinetics of the Periodate Oxidation of Ethylene Glycol and a Series of Methylated Ethylene Glycols. J. Am. Chem. Soc. 76, 3803-3806 (1954).

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Duke, F. R. and R. Laity
Transport Numbers in Pure Fused Salts. J. Am. Chem. Soc. 76, 4046 (1954).

Duke, F. R. and P. R. Quinney
The Kinetics of the Reduction of Perchlorate Ion by Ti(III) in Dilute Solution. J. Am. Chem. Soc. 76, 3800-3803 (1954).

Fassel, V. A. and R. H. Heidel
Fluorescence Spectrometric Determination of Terbium. Anal. Chem. 26, 1134-1137 (1954).

Fritz, J. S. and M. O. Fulda
Titrimetric Determination of Zirconium. Anal. Chem. 26, 1206-1208 (1954).

Herwig, L. O. and G. H. Miller
Relative Ionization Yields for Fission Fragments in Various Gases. Phys. Rev. 95, 413-417 (1954).

Horrocks, D. L. and A. F. Voigt
Half-Life of Thallium²⁰⁴. Phys. Rev. 95, 1205 (1954).

APPENDIX II: LIST OF SHIPMENTS

<u>Destination</u>	<u>Item</u>
Air Reduction Company, Inc. Murray Hill, New Jersey	10 lb. calcium metal (redistilled)
Aluminum Company of America Research Lab., Freeport Road New Kensington, Pennsylvania	1 g. thulium metal
Argonne National Laboratory P. O. Box 299 Lemont, Illinois	10 g. thulium metal
Atomic Energy of Canada, Ltd. USAEC, Liaison Officer Chalk River, Ontario, Canada	1/2 g. holmium metal 1 g. holmium oxide
Brookhaven National Laboratory Warehouse 100 Upton, Long Island, New York	5 g. samarium metal 25 g. yttrium oxide
Rowland Burns 102 Northview Avenue New Castle, Pennsylvania	4 g. cerium metal 2 g. lanthanum metal 1 g. neodymium metal

<u>Destination</u>	<u>Item</u>
Carbide and Carbon Chemical Company P. O. Box P Oak Ridge, Tennessee	10 g. thulium metal
Dow Chemical Company Midland, Michigan	50 mg. holmium oxide 50 mg. thulium oxide 50 mg. lutetium oxide 50 mg. terbium (sample of uranium alloy)
General Electric Company The Knolls Schenectady, New York	25 g. thorium oxide
Dr. Jules Gueron C. E. A. (DCC360) 69 rue de Varenne Paris 7, France	2 g. lutetium metal 2 g. ytterbium metal 2 g. thulium metal 2 g. erbium metal 2 g. holmium metal 2 g. dysprosium metal 2 g. yttrium metal 10 g. praseodymium metal
Johns Hopkins University Department of Physics Baltimore 18, Maryland	3 g. lutetium oxide
Mallinckrodt Chemical Works 3600 North 2nd Street St. Louis 7, Missouri	100 g. lanthanum metal
Montefiore Hospital Division of Neoplastic Diseases Green Hill Road near Jerome Avenue New York 67, New York	5 g. yttrium oxide
National Lead Company of Ohio Mt. Healthy Station Cincinnati, Ohio	1 g. cerium oxide 1 g. lanthanum oxide 1 g. praseodymium oxide 1 g. samarium oxide 1 g. gadolinium oxide 1 g. erbium oxide 1 g. ytterbium oxide 100 mg. dysprosium oxide 100 mg. holmium oxide 100 mg. thulium oxide 100 mg. lutetium oxide 1 g. yttrium oxide

<u>Destination</u>	<u>Item</u>
Pupin Physics Laboratories 538 West 120th Street New York 27, New York	1 cylinder of lanthanum metal, 3/4 in. dia. and 3 1/2 in. long
University of California Barrows Lane and Bancroft Way Berkeley 4, California	1 g. lanthanum oxide 1 g. cerium oxide 1 g. praseodymium oxide 1 g. neodymium oxide 50-100 mg. of the remaining rare earths (oxides) except terbium, europium, and promethium
University of California Los Alamos Scientific Laboratory Los Alamos, New Mexico	10 g. terbium oxide
University of Wisconsin Special Materials Department Madison 6, Wisconsin	1 g. cerium metal 1 g. lanthanum metal 1 g. praseodymium metal 1 g. neodymium metal 1 g. samarium metal 1 g. gadolinium metal 1 g. yttrium metal 1 g. erbium metal 100 mg. dysprosium metal 100 mg. holmium metal 100 mg. thulium metal 100 mg. lutetium metal 100 mg. ytterbium metal
U. S. Bureau of Mines Rare Materials Branch Raleigh, North Carolina	3 g. Thorin
Watertown Arsenal Ordnance Corps Watertown, Massachusetts	10 g. gadolinium metal

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APPENDIX III

- (1) H. A. Wilhelm
- (2) O. N. Carlson
- (3) B. A. Rogers
- (4) J. F. Smith
- (5) D. Peterson
- (6) P. Chiotti

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