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Frank G. Foote, Director
James F. Schumar, Associate Director
Haim H. Chiswik, Associate Director

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*Armour Research Foundation, Chicago, Illinois

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ABSTRACT

Advanced Water Reactor Program - Activity 4102

Firing tests have been carried out on thorium-uranium pellets for the purpose of producing bodies of controlled closed porosity. Using a binder of Carbowax and pore-forming burnout grains of Ethocel and shellac, various pressures and firing exposures were employed to determine optimum conditions for producing 4 to 20 v/o closed porosity with negligible open porosity.

A firing cycle of 1750°C for 9 hours, followed by a 19-hour soak at 1400° - 1450°C, produced sound pellets ranging 7 - 16% closed and 2 - 2½% open porosity. It was found that pores formed by shellac were spherical; those formed by Ethocel tended to flatten out under pressure.

Fast Power Breeder Program - Activity 4104

Most items of process equipment for the new Plutonium Fabrication Facility, except some auxiliary devices, have been procured or constructed. Installation of equipment has been bid at a total of \$1,912,000 as a result of subdivision of the contract into seven parts, compared to a general contract bid of \$5,282,000 previously, for a savings of \$3,370,000.

A production run of about 2000 injection castings is being prepared for checking on full-scale operation of the prototype casting machine in the production of EBR-II fuel elements. Mold coatings fired directly onto the inner surface of the glass tube molds by the manufacturer are in development for the injection casting process.

Dilute plutonium alloys are of interest as thermal fuels which would be independent of U²³⁵ sources, as in certain foreign countries. Fabrication tests on a Zr-5.9 w/o Pu alloy showed a 6.4 w/o loss of plutonium on arc melting. By a combination of hot pressing and cold rolling, a button, about 0.2" thick, was reduced to 3-mil foil with a hardness change of from 43 to 53 R_A.

Rod stock production for the EBR-I, Mark III reactor has been completed according to the procedure described in ANL-5790. Yield for fuel and blanket rod stock ranged from 67 to 76%; the principal causes for rejection were the extrusion defect and dimensions.

Irradiation tests at MTR on 13 specimens of coextruded U-2 w/o Zr at temperatures 200° - 1250°C and burnups of 0.051 to 0.16 a/o showed little dependence on heat treatment. Melting damage and swelling were evident at the higher temperatures. One specimen, induction cast into a 22-mil cladding, showed much higher resistance to swelling than the coextruded samples.

Graphite-1 w/o U_3O_8 specimens jacketed in cans of Zircaloy-3 and heated to 800°C in vacuum for two weeks showed upon visual examination no reaction at the inside can interface.

Advanced Engineering and Development - Activity 4203

An ultrasonic inspection of the EBR-I, Mark III fuel elements, using a 5-Mc transmission technique sensitive to a 1/16" diameter nonbonded area, has been used on 590 blanket fuel elements. Only three of the 590 elements were rejected.

An eddy current inspection system is being prepared for checking variations in cladding thickness of EBR-I, Mark III rods. The development consists of improving stability and sensitivity of existing instruments.

Corrosion work is being accomplished on alloys of uranium, magnesium and aluminum as below.

Uranium with 1 - 5 w/o Ru tested in distilled water for an additional four days over the 2.9 days reported in ANL-5790 displayed an acceleration of corrosion rate.

Dilute fluoride solutions have been found to be effective corrosion inhibitors for commercial magnesium alloys.

Solutions of 0.5 ppm Ni^{++} as a sulphate have been shown to reduce materially the corrosion rate of M-388 aluminum alloy at 260°C both at low and high velocities of circulating water.

Production, Treatment, and Properties of Materials - Activity 5410

The program for preparing high-purity plutonium by fused salt electrolysis has progressed to the point of constructing a prototype cell and running it on uranium to study its operability in the helium gloveboxes. A liquation furnace for consolidating the electrolytic crystals has been built. In cooperation with the Chemistry Division analytical procedures have been set up for carbon and hydrogen. One liquation melt at 700°C in a thoria crucible pointed up the severity of the crucible material and segregation problems.

Measurements have been made of the ultrasonic wave velocities in directions suitable for determining the elastic constants C_{12} , C_{13} and C_{23} . This completes the measurements of wave velocity required to determine all nine elastic constants of alpha-uranium single crystals. From these, there have been calculated the following elastic parameters: Young's Moduli, Poisson's Ratios, Linear and Volume Compressibilities, Bulk Modulus, and Moduli of Rigidity.

Dilatation curves of single crystals of alpha uranium in the [100] and [001] directions in correlation with calculated values from X-ray parameter data indicate that in polycrystalline uranium a measurable restraint of the contraction in the [010] direction results from the expansion in the [001] direction.

In the study of the recrystallization of cold-rolled uranium sheet, efforts to explain discrepancies reported in ANL-5643, ANL-5709, and ANL-5790 have shown that preferential grain growth following recrystallization probably is responsible for the change of expansion coefficient at the 400°C anneal and its absence in a 360°C anneal.

Self-diffusion runs in polycrystalline uranium in the gamma phase, using the sputtering technique, have yielded a tentative value for the diffusion coefficient between 10^{-8} and 10^{-7} cm²/second. This value is of the same order of magnitude as the self-diffusion coefficient at the same fraction of the melting temperature ($0.873 T_m$) for the body-centered cubic metals thorium, iron, and sodium; it is less than that for tantalum. It is much greater than for face-centered cubic metals.

In uranium-base alloy studies, work has been done on plutonium, carbon and fissionium additions, as below. Specimens of high-purity uranium-base plutonium alloys at intervals of 5 a/o up to 50 a/o Pu have been successfully prepared by repeated arc melting. The alloys were readily deformed at 630°C up to about 50% reduction in thickness and have shown a considerable degree of resistance to disintegration, which was observed earlier in alloys prepared by other methods. Improvement in the melting process has reduced weight loss of the melt to 1.5% which can be attributed almost entirely to plutonium vaporization.

Microstructure, density and thermal arrest data have been obtained for U-5 w/o fissionium and U-8 w/o fissionium alloys. The phase diagrams of the uranium binaries with fissionium elements of molybdenum, ruthenium and rhodium have received further study. The uranium-molybdenum transformation is shown to be quite sluggish.

Measurements of emf and resistance of corrosion films on zirconium-base low titanium alloys exposed 16 to 272 hours at 290°C have shown resistances of the order of 10^{10} ohms.

Alloy Theory and the Nature of Solids - Activity 5420

New phases have been shown to exist in various systems of the following types: (1) Ti_2Ni , (2) TiNi_3 , (3) $\alpha\text{-Mn}$, (4) Laves, and (5) CuAu . Possible CsCl-type phases were found to exist in the Y-Cu, Y-Ag and Gd-Ag alloy systems.

Preliminary testing of a new cryostat for measurements of magnetic moment is essentially complete. This apparatus is being developed primarily for studying the magnetic properties of Cr_3O and sigma-type phases.

Effects of Irradiation on Materials - Activity 5430

The experimental and analytical work on the axial and radial distributions of thermal neutrons in a cylindrical-shaped fuel specimen is complete. Preliminary experimental data were presented in ANL-5709, pp. 59-65. Analytical expressions have now been developed for (1) radial distribution of neutrons, (2) average-to-surface ratio of neutron density, (3) axis-to-surface ratio of neutron density, and (4) absorption parameter.

I. ADVANCED WATER REACTOR PROGRAM - ACTIVITY 4102

1. Thoria-Urania Ceramic Fuel Bodies (P. D. Shalek, J. H. Handwerk)

Three firings were made of initial closed-porosity fuel pellet bodies. Each firing contained pellets of composition 90 w/o ThO_2 -10 w/o UO_2 with added 0.5 w/o CaF_2 having different combinations of the following variables:

1. Forming and body-opening binder: Carbowax; 3 or 6 w/o.
2. Pore-forming burnout grain: Ethocel or shellac; 0, 10 or 20 v/o of -60 +100 M or -100 + 170 M grain.
3. Forming pressure: 10, 20, 30, 35, or 40,000 psi.
4. Firing exposure: Pellets placed in open or closed boats.

The final variable was the severity of the firing cycle. The firings were made in the Remmey gas kiln (1) to 1680°C for 4 hours, (2) to 1800°C for 2 hours, and (3) to 1750°C for 9 hours with a 19-hour soak at 1400° - 1450°C.

Only the last of the above cycles gave promising results. In the first firing, the highest bulk density achieved by the control pellets (ones containing no pore-forming grain) was only 8.00 g/cc, as contrasted with the theoretical body density of 10.09 g/cc. Total porosities varied from 21 to 40 v/o, with closed porosities ranging from only 1 to 5 v/o.

The second firing was considerably more severe and gave control pellets with bulk densities up to 8.78 g/cc. Total porosities were lower at 13 to 34 v/o, while the closed porosities increased to a range of 1 to 11 v/o.

The third cycle gave considerably more opportunity for sintering to take place and the control pellets ranged up to 9.15 g/cc. Total porosity again decreased, varying from 9 to 30 v/o, while the closed porosities increased to the range of 7 to 18 v/o. In this group of specimens the lowest open-porosity value was 0.5%, shown by some of the control pellets. However, the majority of suitable pellets in this closed-porosity range had open-porosity values of 2 - 4%. The best pellet on the low-porosity end was a control pellet with 7.3% closed porosity and 2.0% open porosity. The best on the high-porosity end was a pellet containing 10 v/o of -100 + MOM Ethocel which had 16.4% closed pores and 2.4% open pores. Promise was also shown by a pellet containing 10 v/o of -60 +100 M shellac with 17.8 v/o closed pores and 4.1 v/o open pores. Other pellets had porosity values intermediate to these extremes.

It is apparent that denser basic bodies, as shown by the control pellets, will have to be obtained before the somewhat revised goal of from 4 to 20 v/o closed porosity with negligible open porosity can be more closely attained.

Various effects were shown by the other parameters:

1. Varying the amount of carbowax had negligible effect on the properties of the fired pellets. However, the shellac pellets containing 6% carbowax cracked considerably during bakeout whereas those containing 3% did not.
2. All pellets containing 20 v/o grain additions had a high per cent of open pores, whereas those containing 10 v/o showed a low per cent open pores. The finer grain, in general, gave slightly better results, as did the Ethocel grain compared to the shellac.
3. Not enough pellets were available to study the effect of forming pressure on the fired structure of the pellets. Increasing the forming pressure did improve the pellet burnout strength markedly.
4. The pellets fired in closed boats showed considerably more skin densification effect than did those from the open boats.

The effect of pressure on Ethocel and shellac was checked and it was found that under pressure the Ethocel grains completely flatten out in a plane perpendicular to the directed pressure, whereas the shellac grains deform elastically in a similar manner but to a much lesser degree, and return somewhat to their original shape when the pressure is released. This explains the lateral cracking that was so prevalent in the pellets containing the larger -60 +100 M shellac grains. Since spherical pores are desired, only shellac grains will be used in further work. Smaller grains will be used to minimize the elastic deformation in pressing. Microscopic examination of sized fractions shows that under 140 M shellac particles lose their irregular shapes and more closely approximate spheres.

Using the information gained thus far, one more group of bodies is being prepared before irradiation samples are attempted. Closer body particle size control and larger additions of densifying materials are being employed to promote sintering action. This composition of the bodies will be as follows:

Body: 90 w/o ThO_2 -10 w/o UO_2 (as U_3O_8)
 Densifier: (a) 2 w/o CaF_2 , (b) 2 w/o TiO_2
 Binder: 4 w/o Carbowax 4000

- Grain: (a) 0, 6, 12, 18, v/o -140 +170 M shellac; 10 v/o each
of -60 +100, -100 +120, -120 +140, -170 +200, -200 +325,
-325 M shellac
(b) 0, 6, 12, 18, 24 v/o -170 +200 M Shellac; 10 v/o -140
+170 M shellac

It is believed the above work will yield a satisfactory range of
closed-porosity fuel pellet specimens.

II. FAST POWER BREEDER REACTOR PROGRAM - ACTIVITY 4104/4253

1. Fabrication of Plutonium Fuel Elements (A. B. Shuck, R. M. Mayfield, J. E. Ayer, F. L. Yaggee, H. F. Jelinek)

a. Process Equipment for Facility 350

All items of equipment except the auxiliary devices - manipulators, tensioners, vacuum and inert atmosphere furnaces, automatic induction heating controllers, etc., for the two rolling mill lines, the injection casting machine and a few miscellaneous items of equipment which, of necessity, must be adapted to certain contractor installed items - have been procured or constructed by ANL Central Shops.

b. Installation and Hooding of Process Equipment - Facility 350

After the unsatisfactory bidding for the fabrication of hoods and complete installation of hoods, equipment and services as a lump sum contract, the specifications were rewritten in the six divisions shown below. Bids were taken on these and the amount of low bid is as shown.

<u>Part</u>	<u>Work</u>	<u>Low Bidder</u>	<u>Bid Amount</u>
II-A	Hood Fabrication	Nucledyne Corp.	\$ 595,000
II-B	Erection Hoods and Equipment	Midwest Machinery Movers, Inc.	265,070
II-C	Process Piping	Northwest Heating and Plumbing Co.	315,833
II-D	Ventilation and	Bloomer Heating	
II-F	Controls	and Ventilating Co.	267,843
II-E	Electrical	Hyre Electric Co.	81,859
II-G	Painting	W-R Simmer Co.	38,517
Total of Contracts			1,564,122
Estimated Contingencies			194,878
Estimated Field Engineering and Supervisory Costs			150,000
Building Occupancy Costs			3,000
TOTAL			\$ 1,912,000

These low bids plus estimated contingencies, supervision, field engineering, and occupancy items compare with the rejected lump sum bid of \$5,282,000 for a savings of \$3,370,000.

Approval was received for awarding contracts for Part II-A and Part II-B but has not been received yet for Parts II-C through II-G. In

anticipation that such authorization will be received, the drawings have been reviewed with each of the low bidders and the required drawing revisions and clarifications are under way so that there should be no delay in start of the work once the approval is received and contracts are let for all other parts.

c. Fabrication of EBR-II Type Fuel Elements

The injection casting machine prototype has been set up to produce a run of 2000 to 3000 castings to be processed by the Chemical Engineering Division. It is hoped in this run to gain more information concerning process losses and the equilibrium alloy composition. The first runs produced faulty surfaces which were traced to a change in the mold coating drying techniques. A new drying oven was procured for predrying and the molds were further predried by heating to approximately 600°C by torch prior to use in the furnace.

Additional mold coatings were tested. An alcohol suspension of -5 micron thoria has shown promise in producing longer castings for a given temperature-pressure-pressurizing time relationship. Arrangements have been made with the glass manufacturer to fire the coating onto the mold bore surface prior to precision forming. The first molds so coated were not entirely satisfactory. Further work is in progress on this in the hope of removing one source of dimensional variation. Castings made at lower pressures and superheat temperatures appear to be more internally sound. This relationship is being further investigated.

Bench tests have been in progress on components for the mold removal, length shearing, inspection and weighing equipment. A rotary transfer table, which has now completed approximately 15,000 cycles of operation, was built. Crane ways and a mock shielding window (oil filled) have been erected for the final remote control testing of the equipment for refabrication.

2. Fabricability Tests on a Zirconium-5 w/o Plutonium Alloy (R. J. Dunworth, H. V. Rhude)

To determine the potential usefulness of a Zr-5 w/o Pu alloy, the fabricability of the alloy was tested. About 8 grams of the alloy were arc melted into buttons. One button that had a nominal composition of 5.9 w/o Pu was analyzed by radiochemical methods, and the plutonium content was reported as 5.7 w/o $\pm$ 0.4. The amount of plutonium lost in arc melting is within the analytical error, but a calculation of material balance showed that as much as 6.4 w/o of the plutonium was not recovered. The calculated density of the alloy agreed within 1% of the measured density, 6.70 g/cc. The button was reduced 62% in thickness by hot pressing, according to the data listed in Table I. The button, which was then 0.120" thick, was cold rolled to a thickness of 0.003". This cold reduction of 97.5% is about the

TABLE I

Pressing Data for Zirconium-5 w/o Plutonium Alloy

Temp. of Pressing (°C)	Dimensions Before Pressing		Dimensions After Pressing		Reduction(1) (%)	Effective Pressing Area of Spec. (in. ²)(2)	Pressure (psi)(3)
	Diam. (in.)	Thick. (in.)	Diam. (in.)	Thick. (in.)			
25	0.646	0.315	0.652	0.279	11.4	0.512	23,400
528	0.620	0.318	0.725	0.202	36.5	0.570	21,000
730	0.725	0.202	0.938	0.120	62.2	0.737	16,200

(1) Total reduction in thickness.

(2) Calculated from diameter after pressing.

(3) Total load was 60,000 pounds, limit of Riehle testing machine.

same as the maximum cold reduction of unalloyed zirconium. The hardness of the cast button was 43 R_A, which increased to 53 R_A after a 50% reduction by cold rolling. Under polarized light, metallographic examination of the casting showed a single-phase, anisotropic structure - presumably h.c.p. alpha zirconium.

3. Manufacture of Rod for Fuel and Blanket Elements, EBR-I, Mark III (R. E. Macherey, N. J. Carson, Jr., R. L. Salley, R. A. Beatty)

The manufacture of rod stock from which fuel and blanket elements for the Mark III loading of EBR-I were produced has been completed. The basic manufacturing procedure employed in the production of this rod stock was described in ANL-5790. It is recalled that three types of rod were required: natural uranium blanket rod, natural uranium fuel blanket rod, and enriched uranium fuel rod. The blanket rod was coextruded from 2 $\frac{3}{4}$ " diameter composite billets; both the fuel blanket and fuel rod were extruded from 2" diameter billets.

A total of 64 billets, 2 $\frac{3}{4}$ " diameter, were obtained from nine heats. From these, 64 Zircaloy-2 clad, U-2 w/o Zr alloy rods, acceptable stock for 608 blanket elements, were obtained. The approximate recovery of core alloy was 67%. The stock required for 260 blanket slugs for the fuel rods was obtained from 29 extrusion billets, 2" diameter. The recovery from this processing was approximately 69%. The enriched fuel slugs required for the fuel elements were obtained from 19 composite billets. The yield here was approximately 76%. The principal causes of rod rejection were the extrusion defect and dimensions.

Because of difficulties encountered in producing acceptable stock for the fuel and fuel blanket slugs from the 2" extrusions, modification of the manufacturing procedure discussed in ANL-5790 was required. Dimensional changes caused by the final dimensional stability heat treatment of fuel and blanket elements required a preliminary heat treatment to stabilize these dimensions before machining the elements to length for assembly. In the case of the blanket rod stock, a single heat treatment performed after the swaging operation was sufficient to stabilize the dimensions. This heat treatment after swaging was satisfactory because the resulting dimensional changes were small and more or less uniform. The diameters increased in the order of 1 mil and the length decreased in the order of 5 mils/inch. This procedure, however, was unsatisfactory for both the fuel blanket and fuel rods, because the preliminary heat treatment produced large and varying changes in dimensions. These variations in dimensional changes were so large as to cause excessive stock rejection. In addition to these dimensional changes resulting from heat treatment, great difficulty was experienced in producing the required diametral tolerances by a swaging operation. Apparent physical structural variation of the core alloy along the length of these rods made reduction during swaging vary. These difficulties were eliminated by subjecting the as-extruded and stripped rod to a preliminary heat treatment either before or during the swaging operation. This heat treatment (800°C, air-cool) produced the necessary uniformity of structure in the core alloy so that swaging to dimensional tolerances was made easier. Because of the cold work induced during swaging, however, a second preliminary heat treatment was necessary to stabilize dimensions before the final blanket or fuel elements were machined to length. Dimensional changes produced during this second preliminary heat treatment were of the same order as those experienced with the blanket rod. Based on metallographic examination, the structure of the core alloy produced by this double preliminary heat treatment procedure was essentially the same as that resulting from the single preliminary heat treatment procedure.

In ANL-5790 two methods were proposed for finishing the assembled fuel and blanket rods to dimensions and surface finish necessary for rib welding. One was blasting and pickling and the other was grinding. Since it was found that grinding would produce dimensions falling within specifications, particularly cladding thickness, grinding was selected. Because of the small excess stock on the blanket rods, wet abrasive blasting to remove the contamination resulting from the final heat treatment was found necessary to promote uniform bite by the grinding wheel. The excess stock on the fuel rods, however, made this blasting operation unnecessary on these elements. In these rods, the depth of grinding was sufficient so that penetration beneath the oxide was possible.

4. Effect of Irradiation on Extruded and Heat-Treated U-2 w/o Zr Alloy (J. H. Kittel)

The Mark III core and blanket for EBR-I is U-2 w/o Zr alloy, co-extruded with Zircaloy-2 cladding into rods with a final diameter of

approximately 0.404". It was anticipated by all concerned that as-extruded material would be dimensionally unstable when subjected to irradiation; accordingly, irradiation tests were scheduled to evaluate the most promising heat treatments.

Based on earlier irradiation experiments on swaged U-2 w/o Zr, three heat treatments were selected for the irradiation specimens. They were as follows:

- A. 800°C - 15 minutes, 500°C - 1 hour, air-cooled.
- B. 800°C - 15 minutes 690°C - 1 hour, air-cooled.
- C. 800°C - 15 minutes, water quenched, 690°C - 1 hour, water quenched.

In addition, material was supplied which had been prepared by passing an induction coil along an extruded clad rod so that the alloy was briefly melted and subsequently solidified within the cladding. Microstructures of the above four specimen materials are shown in Figures 1 through 4.

Figure 1. Microstructure before Irradiation of Extruded U-2 w/o Zr Alloy, Heat Treated as Follows:
800°C - 15 Minutes; 500°C - 1 Hour, Air-Cooled.

Micro 21429

100-X

Figure 2. Microstructure before Irradiation of Extruded U-2 w/o Zr Alloy, Heat Treated as Follows:
800°C - 15 Minutes; 690°C - 1 Hour, Air-Cooled.

Micro 21430

100-X

Figure 3. Microstructure before Irradiation of Extruded U-2 w/o Zr Alloy, Heat Treated as Follows: 800°C - 15 Minutes, Water Quenched, 690°C - 1 Hour, Water Quenched.

Micro 21431

100-X

Figure 4. Microstructure before Irradiation of Extruded U-2 w/o Zr Alloy Induction Cast in its Cladding.

Micro 21432

100-X

Five NaK capsules, each containing one of the heat-treated specimens, were sent to the MTR for irradiation. Prior to irradiation, the co-extruded cladding was removed from two sets of the specimens. One of the capsules also contained one of the induction-cast specimens. Because of space problems at the MTR, only four of the capsules have been irradiated to date, and the results obtained with these capsules are summarized in Table II. Photographs of these specimens after irradiation are shown in Figures 5 through 8.

TABLE II
Summary of Irradiation Effects on Specimens of Extruded and Heat Treated U-2 w/o Zr Alloy

Original Specimen No.	Irradiation Specimen No.	Heat Treatment	Cladding Thickness (in.)	Estimated Irradiation Temp (°C)	Burnup (a/o)	Length Change (%)	Growth Rate (G_1)*	Diameter Change (%)	Density Decrease (%)	Weight Change (mg)	Hardness Change (R_A)	Remarks
26-4	CG-22-1	800°C - 15 min, 500°C - 1 hr, A.C.	None	200	0.051	0.26	5.2	0.61	0.735	-1.5	5.7	--
26-4	CG-18-1	Same	None	1150	0.13	1.94	15	2.70	5.614	-1.8	0.4	Some melting damage evident.
26-4	CG-24-1	Same	0.022	400	0.13	0.66	5.1	0.57	1.265	-4.2	--	--
26-4	CG-19-1	Same	0.022	700	0.16	1.72	11	0.70	5.188	-4.1	--	Cladding bulged.
	Average						9.1					
26-6	CG-22-2	800°C - 15 min, 690°C - 1 hr, A.C.	None	200	0.051	1.84	36	-0.17	0.258	-6.3	0.7	--
26-6	CG-18-2		None	1150	0.13	0.25	1.9	2.68	5.673	-25.3	-7.9	Some melting damage evident.
26-6	CG-24-2		0.022	400	0.13	0.90	6.9	0.37	1.456	-4.2	--	--
26-6	CG-19-2		0.022	700	0.16	1.63	10	0.80	5.899	-4.2	--	Cladding bulged.
	Average						13.7					
26-8	CG-22-3	800°C - 15 min, W.Q. 690°C - 1 hr, W.Q.	None	200	0.051	2.07	42	0.06	0.237	-1.4	9.9	--
26-8	CG-18-3		None	1150	0.13	0.41	3.2	3.63	5.794	-6.9	-5.9	Some melting damage evident.
26-8	CG-24-3		0.022	400	0.13	1.41	11	-0.12	1.886	-4.3	--	--
26-8	CG-19-3		0.022	700	0.16	1.13	7.1	0.74	7.037	-3.6	--	Cladding bulged.
	Average						15.8					
A-60	CG-18-4	Induction cast in cladding.	0.022	1250	0.13	-0.31	-2.4	0.80	2.142	14.6	--	Cladding slightly bulged.

* G_1 = microin/in/fission/ 10^6 total atoms

It can be noted from the data that the four groups of specimens were subjected to burnups ranging from 0.051 to 0.16 a/o, at temperatures from 200° to 1250°C.

With regard to the heat-treated specimens, it can be noted that the clad specimens, as expected, showed generally less growth than the bare specimens. There appears to be little to choose from in determining the best of the three heat treatments, since all appeared to be quite effective in stabilizing the material. For operation at elevated temperatures, heat treatments B and C appear to be somewhat better.

The data show that the effect of irradiation temperature outweighed the lesser differences resulting from differences in heat treatment, with higher temperatures generally tending to lower growth rates. Swelling, however, was much greater in those specimens irradiated at elevated temperatures.

Figure 5. Effect of Irradiation on Extruded U-2 w/o Zr Alloy, Heat Treated as Follows:
800°C - 15 Minutes, 500°C - 1 Hour, Air-Cooled. Magnification 2-X.

Macro No.	21939	21947	21936	21942

Specimen No.	CG-22-1	CG-18-1	CG-24-1	CG-19-1
Cladding Thickness, in.	None	None	0.022	0.022
Estimated Irradiation Temperature, °C	200	1150	400	700
Burnup, a/o	0.051	0.13	0.13	0.16

Figure 6. Effect of Irradiation Temperature on Extruded U-Zr Alloy, Heat Treated as Follows: 800°C - 15 Minutes, 690°C - 1 Hour, Air-Cooled. Magnification 2-X.

Macro No.

21938

21946

21935

21941

Specimen No.

CG-22-2

CG-18-2

CG-24-2

CG-19-2

Cladding Thickness, in.

None

None

0.022

0.022

Estimated Irradiation
Temperature, °C

200

1150

400

700

Burnup, a/o

0.051

0.13

0.13

0.16

Figure 7. Effect of Irradiation on Extruded U-Zr Alloy, Heat Treated as Follows: 800°C - 15 Minutes, Water Quenched, 690°C - 1 Hour, Water Quenched. Magnification 2-X.

Macro No.

21937

21945

21934

21940

Specimen No.

CG-22-3

CG-18-3

CG-24-3

CG-19-3

Cladding Thickness, in.

None

None

0.022

0.022

Estimated Irradiation
Temperature, °C

200

1150

400

700

Burnup, a/o

0.051

0.13

0.13

0.16

Figure 8. Effect of Irradiation on Extruded U-Zr Alloy,
Induction Cast in its Cladding.

Specimen No.	CG-18-4
Cladding Thickness, in.	0.022
Est. Irradiation Temp., °C	1250
Burnup, a/o	0.13

The induction-cast specimen shortened slightly, in contrast to the heat-treated group. Also, it showed a much higher resistance to swelling than the heat-treated specimens. The rate at which it increased in volume when irradiated at 1250°C was less than half that displayed by comparable heat-treated specimens irradiated at only 700°C.

5. Graphite-Urania Fuel Elements for the TREAT Reactor (J. H. Handwerk, R. C. Lied)

Fabrication procedures for making graphite-urania test specimens were investigated. Graphite flour, 1 w/o normal U_3O_8 and petroleum pitch were hot mixed and then frozen and crushed in a disc pulverizer. This material was hot pressed at 100°C in steel molds using a pressure of 5000 psi. The pressed ware was found to have an average density of 1.80 g/cc. These samples were baked at 980°C, and the baked ware was found to have an average density of 1.40 g/cc.

Additional specimens measuring 1" in diameter and 1" long were pressed from a graphite mixture used by Great Lakes Carbon Corporation. The graphite binder mixture was frozen and passed through a disc pulverizer. One weight per cent U_3O_8 (based on total carbon content) was added and the resultant batch was mixed in a twin shell blender. Samples were pressed at 100°C in steel molds using a pressure of 5000 psi. The pressed ware was found to have an average density of 1.82 g/cc. These samples are now being baked.

Graphite-1 w/o U_3O_8 specimens were jacketed in Zircaloy-3 cans. These cans were placed in a stainless steel cylinder which was purged with helium and then vacuum sealed at a pressure of 0.5 micron. Eight of these assemblies were fabricated, and one assembly was heated at each of the 100°C temperature increments from 100° to 800°C. These temperatures were maintained for a two-week period. Visual examination of the 600°, 700° and 800°C specimens disclosed no reaction between the graphite and the Zircaloy-3 interface.

Attempts were made to evaluate the thermal shock and gas evolution of these specimens. The thermal shock test consisted of heating the specimens to 1000°C in helium and then chilling them with a blast of cold helium. This test fractured most of the specimens; however, it appeared that the specimens were rather severely oxidized because of the oxygen content of the helium. Specimens were encased in stainless steel tubes which were evacuated and then heated to 800°C. The gas evolved from the sample was then measured, but the data appear to be unreliable as the system was found to leak at a high rate.

TABLE III

Tests on Graphite- U_3O_8 Samples from Outside Suppliers

Supplier	Bulk Density (g/cc)	Apparent Density (g/cc)	Apparent Porosity (%)
Los Alamos	1.86	1.93	5.27
Great Lakes	1.70	2.03	15.90
United Carbon	1.72	1.90	7.80

III. ADVANCED ENGINEERING AND DEVELOPMENT - ACTIVITY 4203

1. Ultrasonic Tests on EBR-I, Core III Fuel Elements

(W. J. McGonnagle, W. N. Beck, C. J. Renken, R. G. Myers)

a. Bond Tests

Ultrasonic bond tests have been conducted on 590 EBR-I, Mark III blanket fuel elements. Three elements with indicated inhomogeneities in the bond layer were withheld. In order to facilitate this test program, an offset roller lead assembly was designed and fabricated. This conveyor constitutes two diametrically opposed rows of rubber-tired, ball bearing wheels. One row of wheels is ganged to a common drive system. The others act as idlers. The swivel platforms supporting each opposed pair of wheels are connected to a common guide bar, the actuation of which will change the "feedpitch" of the conveyor. For these particular tests, the speed and pitch of the conveyor were fixed so that one fuel element was processed every minute.

The ultrasonic inspection is performed at a frequency of 5 megacycles utilizing a transmission technique. An automatic alarm system is preset to detect a minimum nonbonded area of $1/16$ " diameter.

Approximately 100 feet of Zircaloy rod which were to be used in the fabrication of EBR-I, Mark III fuel rods were inspected ultrasonically. The purpose of the inspection was to detect stringers and small cracks in the Zircaloy. A permanent recording was not made of the defects detected; instead an alarm system was improvised which provided an audible signal whenever a change in acoustic transmission characteristic of the Zircaloy was noted. These areas were marked, sectioned and inspected. It was observed that the techniques isolated areas containing "star cracks," transverse cracks and stringers.

The section bonding in the EBR-I, Mark III elements was tested ultrasonically. The section bond is the juncture between the separate rod sections which constitute the fuel element. The bonding was accomplished by an induction-melting process which fuses the juncture. For reasons of mechanical strength, a test which would determine this bond area was requested. A two-transducer technique was used, one transducer being located on one side of the rod, the other on the opposite side. The ultrasonic beam was made to impinge at an angle on the section bond. The energy reflected from the bond impinged on the receiving transducer. An alarm system was used to select junctures not adequately bonded. These crystals were also mounted on the roller feed assembly so that the section bond, as well as the core-to-clad bond, of the fuel rod could be determined in one operation.

b. Tests of Cladding Thickness

The blanket rods and part of the fuel rods for the EBR-I, Mark III loading are being checked for cladding thickness. The development work on the eddy current system used to make these measurements is not complete, but the instrument was judged to be useful in its present stage of development. As with most nondestructive testing instruments producing a quantitative answer, known standards must be used in the calibration. This system is normally accurate to ± 0.0002 " of clad on the EBR-I, Mark III blanket and fuel rods. An additional uncertainty is due to the difficulty involved in finding a standard rod of sufficiently uniform clad.

Development is continuing on a family of instruments similar to the one just mentioned, but of greater stability and sensitivity. These instruments will be useful in much wider fields of application.

In response to a need for a quick method of checking for cracks and laminations in the zirconium wire used as spacers on the EBR-I, Mark III rods, an existing eddy current instrument was modified to make it useful for inspection of the wire. Sectioning the wire over suspected points proved the utility of the instrument. To date a large amount of this wire has been inspected by this method.

A cladding measuring instrument which uses pulsed eddy currents was built. There was considerable instability in the instrument and work was carried on to eliminate this problem. More efficient probes were designed to increase the amount of change in the output voltage for a given change in cladding. The instrument will be suitable for production line applications, since it reduces the effects of probe-to-metal spacing.

Experiments were continued to determine the optimum geometry and design of probe coils.

2. Corrosion-Resistant Alloys

a. Corrosion-Resistant Uranium Alloys (S. Greenberg, G. M. Dragel)

Zirconium-Niobium: Electron microscopic study of the U-5 w/o Zr-1.5 w/o Nb alloy continued with examination of more severely aged material, i.e., 6 hours at 400°C and various times at 475°C. Results confirmed previous ideas concerning precipitation and agglomeration.

A cathodic etching apparatus has been assembled and is being tested for use with this study. It is hoped that cathodic etching technique will produce structures which lead to clearer interpretation than those produced by electrochemical means.

Ruthenium: The material discussed last quarter was tested for an additional 4-day period in boiling distilled water. Results are:

<u>Ruthenium (w/o)</u>	<u>Heat Treatment</u>	<u>Average Rate of Weight Loss (mcd)</u>
1	Gamma Quench	Cracked into several pieces and powder
2	Gamma Quench	36
5	Gamma Quench	260
1	Beta Quench	89
5	Beta Quench	309

Only the gamma-quenched alloy containing 2 w/o Ru showed no acceleration of rate as compared with the initial 3-day test.

b. Development of Corrosion-Resistant Magnesium Alloys
(S. Greenberg, G. M. Dragel)

Corrosion of magnesium alloys is being studied in support of the Mighty Mouse reactor program. It is desired to use magnesium, or a suitable alloy thereof, as a cladding material. The fuel elements will remain in the reactor for 5 days. At a maximum estimated temperature of 150°C, the allowable corrosion rate is 1.5 mils/day.

In pure water at 150°C neither the commercially available alloys having acceptable cross section nor several special alloys produced here met the above specifications. The commercial alloys tested include: high-purity magnesium, AZ 31B(FS-1), photoengraving grade, AZ 91, Dow M, ZK 60A, EK 30, and HZ 32 (ZT 1). In addition, alloys containing 1, 5, 10, and 20% lead, 10% tin, and 7% zinc failed at 150°C.

Dilute fluoride solutions (100 ppm fluoride or less) have been found to be effective inhibitors under the conditions of interest. There are many fluorides, including those which are acceptable on the basis of nuclear considerations, which reduce the corrosion rate of commercial alloys below 1.5 mils/day at 150°C. For example, the rate for AZ 31B is about 0.5 mil/day at 50 - 100 ppm fluoride, added as PbF₂. One hundred ppm fluoride (PbF₂) reduces the rate of HZ 32 to 0.4 mil/day. Corresponding rates in pure water are 3.2 and 1.4 mils/day, respectively.

Present work is concentrated on understanding the nature of the inhibiting mechanism and testing noncommercial alloys. It appears definite that fluoride is used up as corrosion proceeds. It may thus be possible to obtain protection with much lower concentrations, continuously maintained in flow systems, than we have used in static nonreplenished systems. The pH may be an important variable. In general, greatest protection is obtained with the fluorides of sodium and potassium.

c. Corrosion-Resistant Aluminum Alloys (W. E. Ruther, D. J. Dorman)

Dynamic Testing: Two new experiments have been completed in this phase of the program. These experiments are attempts to explain the unusual effects of velocity and surface-to-volume ratios in the dynamic testing of aluminum alloys. In this group the hypothesis being tested is that ions of metals cathodic to aluminum (Ni^{++} , Fe^{++} , etc.), obtained from the corrosion of the aluminum alloy and the loop, are responsible for these effects.

In ANL-5790 the results of the first test with distilled water were described. The first new experiment duplicated this test as nearly as possible, except that a 0.5-ppm Ni^{++} solution (NiSO_4) was fed into the loop instead of distilled water. Unfortunately, the solution coming from the loop contained very little nickel (<0.1 ppm). Subsequent testing has indicated that both reduction of the nickel by the hydrogen gas and precipitation of an insoluble compound occurred. In spite of the poor solution control, the test was continued because of the marked improvement in alloy corrosion behavior, as shown in Figure 9. Each line shows the corrosion behavior of an individual sample. Reproducibility was good so that duplicate samples formed nearly identical curves.

Figure 9. Dynamic Corrosion of M388 Aluminum Alloy at 260°C

The originally loaded samples in the fast (22 feet/second) channel corroded about the same amount initially as in distilled water, but the corrosion rate did not break upward after 12 days. The replacement

samples (not shown in the figure) corroded only about 15% more during the initial period than the originals and at only a slightly higher rate throughout the corrosion test. This is in contrast to the rapid corrosion for replacement samples in distilled water.

In the slower channel the corrosion rates for both the original and replacement samples were lower than for the distilled water case.

A second short experiment performed during this quarter evaluated the corrosion behavior of certain selected aluminum-nickel alloys. Conditions of 260°C, distilled water, fast flow (22 feet/second) and low alloy surface-to-water volume ratio were chosen to provide a rapid rate of corrosion.

Alloys of aluminum-1/2 and 4 w/o nickel, Al-1 w/o Ni-0.1 w/o Ti, and Al-5.5 w/o Ni-0.3 w/o Fe-0.2 w/o Ti were compared with M-388 alloy as a standard. The titanium-bearing alloys had previously shown somewhat improved corrosion behavior in a 290°C static test. In this dynamic test (13 days total) there was no difference in corrosion for any of the alloys tested. All exhibited the high corrosion rate obtained for replacement samples in a previous dynamic test.

New Wrought Alloys: Alloys containing 1 w/o nickel and small additions of molybdenum and molybdenum-titanium were investigated in water at 290°C (refreshed autoclaves). Except for small upward displacement of the curves, their corrosion behavior was similar to M-388 in a 40-day test.

Powder Metallurgy: The corrosion coating on most high-temperature aluminum specimens is duplex in nature. A relatively porous outer coating covers a more compact inner film. It is possible that the corrosion rate of the alloy is determined by the rate of transformation of the compact layer into porous coating.

It has been shown that certain materials, notably dilute phosphoric acid, are capable of reducing the corrosion rate. Presumably these materials act after diffusing through the porous part of the coating. If it were possible to supply these materials from the metal side they might be in a favorable spot for inhibition. At the same time they might have a low enough diffusion rate through the porous layer of oxide to avoid seriously contaminating the pure water corrodant.

Attempts have been made, with no success, to achieve inhibition by incorporating small amounts of phosphorus, antimony and bismuth into cast alloys.

A new technique involving the coextrusion of oxides or phosphates with atomized M-388 powder is now being used. Two extrusions,

containing 4 w/o AlPO_4 and 4 w/o SiO_2 , respectively, are being tested at 290°C in replenished water autoclave systems. Preliminary results indicate the same order of corrosion rate as the wrought M-388 alloy. In small static autoclaves at 350°C the extrusions appear slightly better than M-388 (based on weight gain and appearance).

Combinations of AlPO_4 and SiO_2 in M-388 powder will also be extruded and other possible inhibitors will be tried.

EBWR Dummy Assembly: During August, 1957, two M-388 aluminum alloy dummy fuel assemblies were removed from EBWR. The assemblies had been located in core positions D-13 and K-2. The complete assemblies were visually examined in a cave for evidence of unusual corrosion damage. None was noted. The adherent speckled brown coating was assumed to derive its unusual color from iron rust.

A hole saw was used to sample each assembly in six places: top, center, and bottom of each of two sides. Some of these samples were defilmed using a boric acid - AC current technique which does not attack the metal. Superficial pitting about 0.3 mm diameter by 0.1 mm deep was noted on most coupons. Pitting has not been a characteristic of this aluminum alloy in previous high temperature testing.

The outer surface of the dummy assembly was generally slightly more corroded than the inner surface. However, the best appearing coupon surface of the lot was from the outside-top of K-2.

A small piece of D-13 was polished and the corrosion coating thickness estimated at 0.0009". Original dimensions of the plate were not accurately known so a penetration estimate could not be made.

IV. PRODUCTION, TREATMENT, AND PROPERTIES OF MATERIALS ACTIVITY 5410

1. Preparation of High-Purity Plutonium (B. Blumenthal)

The progress of the program of preparing high-purity plutonium by fused salt electrolysis may be summarized as follows.

The electrolytic cell has been built and is now running on uranium to show that the evolution of chlorine and hydrogen chlorine gas from the electrolyte can be controlled and does not interfere with the operation of the glovebox and system for helium purification.

The air box which will house the electrolytic cell is being converted for use with helium.

The liquation furnace for the conversion of the electrolytic crystals to ingots is built and ready for mock-up.

The sampling procedure for preparing analytical and metallographic specimens has been established. The method uses a lathe; the solid specimens are cut with a dry silicon-carbide disc at a rate of 1300 rps for the disc and 100 rpm for the work piece. If turnings are desired a stationary carbide tool is used. Turnings and grindings are collected in a plastic box surrounding the work piece.

To develop a metallographic polishing procedure by which the inclusions are retained, we are experimenting with the Syntron vibrator. The results are promising. We have satisfactorily prepared uranium specimens containing many carbide inclusions.

Through the cooperation of the Chemistry Division we have obtained carbon and hydrogen analyses of impure plutonium of a precision comparable to that of uranium.

Spectrochemical analyses by the carrier distillation method have been obtained through the courtesy of the Los Alamos Scientific Laboratory.

One plutonium biscuit was analyzed (Table IV). Its impurity level is very high, but the composition is remarkably uniform when compared with uranium, where considerable differences occur between bottom, top, and edge of derbies. Since the plutonium biscuit weighed only about 1200 grams it probably had solidified rather quickly, allowing no time for segregation to occur.

TABLE IV

Composition of Plutonium Biscuit
(Biscuit No. 3-424)

Sample Location	Spectrochemical Analysis ⁽¹⁾ (ppm)										Chemical Analysis ⁽²⁾ (ppm)		Radiochem. Am (ppm)
	Al	Cr	Cu	Fe	Mg	Mn	Na	Ni	Pb	Si	C	H	
Top	200	230	20	100	90	40	10	110	7	45	95	7.5	220
Edge	200	225	20	100	90	20	10	120	4	45	92	10.8	230
Bottom	175	250	20	125	90	45	10	120	9	45	104	8.9	230

(1) All other elements below limits of spectrochemical detection.

(2) Carbon and hydrogen data are mean values of 3 to 4 determinations. The range of individual carbon analyses may vary considerably, e.g., for the edge sample: C = 130, 92, 54 ppm. Nitrogen and oxygen analyses have been requested.

The data of a single stationary liquation experiment carried out in our thermal analysis furnace are listed in Table V. To assure a uniform starting material, the metal was premelted by induction from available stock and cast in a graphite mold. The charge was heated in a thoria crucible at a rate of 5° C/minute, held for 1 hour at 700° C, and cooled to room temperature at a rate of 3° C/minute. The melt picked up about 100 ppm of carbon, probably from the oil of the vacuum system. Apparently plutonium will require similar care in furnace design and handling as uranium to obtain metal of low carbon content. During cooling the top solidified first, as shown by the raised ingot top (Figure 10). Consequently, the impurity-rich mother liquor was concentrated near the bottom of the ingot, and spectrochemical and chemical analyses showed a much increased concentration of chromium, iron, nickel, silicon, and hydrogen near the bottom. On the other hand, the Am²⁴¹ content produced by beta decay of Pu²⁴¹ was uniform throughout the ingot. The ingot of 91.398 grams weight had wet part of the crucible, indicating the seriousness of the crucible problem, while the temperature was only 700° C. The melting loss was 2.8%.

TABLE V

Liquation of Plutonium

Name	Number	Sample Location	Spectrochemical Analysis ⁽¹⁾ (ppm)												Chemical Analysis ⁽²⁾ (ppm)		Radiochem. Am (ppm)
			Al	B	Cr	Cu	Fe	Mg	Mn	Na	Ni	Pb	Si	Sn	C	H	
Premelt	3-423/4 3-423/5	Top													232, 50, 137	2.5, 5.5, 5.3	-
		Bottom					Not Analyzed								62, 57	8.2, 6.1	
Liquation	3-423/17 3-423/18	Top	175	2	150	40	375	L 5	40	20	270	1	680	1	119, 191, 200	4.4, 2.2, 3.1	340
		Bottom	200	2	280	40	775	L 5	45	10	520	1	1000	1	137, 197, 196	13.5, 5.5, 8.5	340

(1) All other elements below limits of spectrochemical detection.

(2) Nitrogen and oxygen not yet reported.

Figure 10. Liquefied Plutonium Ingot with Portion of Thoria Crucible Showing Wetted Area.

PU-185

1-X

2. Physical Metallurgy of Uranium

a. Elastic Moduli of Single Crystals of Alpha Uranium (E. S. Fisher, L. T. Lloyd)

Velocities of ultrasonic waves propagated in directions suitable for determining the room-temperature elastic moduli C_{12} , C_{13} and C_{23} of alpha uranium have been determined from measurements obtained at Bell Telephone Laboratories by H. J. McSkimin on four single-crystal specimens.

Experimental Data: The procedures for preparing the two parallel, flat faces on the crystals were described in ANL-5709. Four specimens were prepared, with thicknesses ranging from 1.7 to 3 mm and with lateral dimensions of 4 to 6 mm. The directions of propagation for each crystal as indicated by Laue photographs are given in Table VI; the considerations and calculations for their selection are described in ANL-5790. Crystal G is a duplicate of Crystal F in the sense that it was originally sectioned to provide a face containing the [001] direction; however, misalignments in the lapping process resulted in a 1° error.

The three limiting wave velocities corresponding to the three modes of vibration were separately determined for each crystal by the procedure described in ANL-5709. These limiting velocities are listed in Table VI.

TABLE VI

Limiting Velocities Measured from Crystals D, E, F, and G

Crystal	Direction of Wave Propagation	Direction of Particle Motion	Mode Type	Limiting Velocity (cm/sec)
D	90° from [010]; approx. 45½° from [001]	1) Near propagation direction in (010) plane	quasi-longitudinal	V ₁ = 328,850
		2) Perpendicular to 1 and 3	quasi-shear	V ₂ = 238,085
		3) [010]	shear	V ₃ = 227,980
E	90° from [100]; approx. 36° from [001]	4) Near propagation direction in (100) plane	quasi-longitudinal	V ₄ = 396,630
		5) Perpendicular to 4 and 6	quasi-shear	V ₅ = 189,565
		6) [100]	shear	V ₆ = 196,810
F	90° from [001]; approx. 44½° from [100]	7) Near propagation direction in (001) plane	quasi-longitudinal	V ₇ = 324,980
		8) Perpendicular to 7 and 9	quasi-shear	V ₈ = 204,890
		9) [001]	shear	V ₉ = 227,535
G	Approx. 89° from [001]; approx. 45½° from [100]	10) Near propagation direction in (001) plane	quasi-longitudinal	V ₁₀ = 324,920
		11) Perpendicular to 10 and 12	quasi-shear	V ₁₁ = 205,040
		12) ~[001]	shear	V ₁₂ = 229,325

Calculation of Cross Coupling Moduli: From the solutions to the secular equations of Christoffel for elastic wave propagation 90° to [010] and at an angle θ to [001], the following relations are obtained:

$$C_{13} = \pm \left[\frac{1}{sc} \sqrt{(\lambda_{11} - \rho V^2)(\lambda_{33} - \rho V^2)} \right] - C_{55} \quad (1)$$

where

$$\begin{aligned} s &= \sin \theta \\ c &= \cos \theta \\ \lambda_{11} &= S^2 c_{11} + C^2 c_{55} \\ \lambda_{33} &= S^2 c_{55} + C^2 c_{33} \\ \rho &= \text{density} \\ V &= \text{either the quasi-longitudinal or quasi-shear mode velocity} \end{aligned}$$

Analogous equations are obtained for computing C_{12} and C_{23} .

There are two problems connected with the computation, one pertaining to the choice of algebraic sign of the square root and the other concerning the uncertainty of the values used for computation. Regarding the first problem, the negative sign always gives a negative C_{ij} and, in the present case, the positive sign gives a positive C_{ij} . Elastic theory alone does not offer a basis for choosing the correct sign; however, it can be shown that in the present case the positive C_{ij} are necessary to be consistent with experimental observations.

The uncertainties involved in the positive value of C_{ij} will, of course, depend on the uncertainties in (1) the angles θ , (2) the diagonal moduli $C_{11} \dots C_{66}$, (3) the measured velocities, and (4) the density. In regard to variables 2, 3, and 4 we have little choice, i.e., the best values for $C_{11} \dots C_{66}$ determined from crystals A, B, and C (ANL-5717) were used and the density used was 19.04 grams/cc, determined by lattice parameter measurements in high-purity uranium.¹ It is found, however, that in the present case the uncertainty in the rotation angle is most important. For an uncertainty of $\pm 1/2^\circ$, which is assumed to be that obtained from the back-reflection Laue films, the probable error in the computed C_{ij} can be as high as 5.5%. To obtain a more accurate estimate of the angle θ one can use the pure shear mode velocities noted in Table VI, since these velocities are functions of the shear moduli C_{44} , C_{55} and C_{66} (according to the solutions of the secular equation) which were directly determined previously. For the rotation of Crystal D, for example:

$$\cos^2 \theta_D = \frac{\rho V_3^2 - C_{66}}{C_{44} - C_{66}}, \quad (2)$$

which gives a θ_D value of $45^\circ 29'$ from [001], compared to $45^\circ 50'$ indicated by the Laue film. Furthermore, if one assumes each velocity measurement to have a probable error of 0.07%,² the calculated probable error in θ_D derived from equation 2 is $\pm 11'$. Analogous calculations for Crystals E, F and G gave the following:

$$\begin{aligned} \theta_E &= 39^\circ \pm 6^\circ \text{ from [001]} && ; \\ \theta_F &= 44^\circ 36' \pm 9' \text{ from [100]} && ; \\ \theta_G &= 46^\circ 17' \pm 9' \text{ from [100]} && . \end{aligned}$$

¹M. H. Mueller, "Lattice Parameters of Alpha Uranium," ANL-5367, Appendix I, pp. 33-34 (December 1954).

²H. J. McSkimin and W. L. Bond, "Elastic Moduli of Diamond," Phys. Rev. 105 116(1957)

For computing C_{12} and C_{13} the calculated rotation angles for Crystals D, F and G were used. The modulus C_{23} was calculated from Crystal E using the rotation angle indicated by the Laue pattern, since the calculated θ_E was subject to a very large error, as indicated above. The computed values and the probable errors are given in Table VII. The probable errors in the C_{ij} were calculated assuming a 0.07% probable error in each velocity, or a 0.14% error in each ρV^2 (disregarding any error in density), and the probable errors in θ as given above. Because of rather large uncertainty in the thickness dimension of Crystal G, no probable error is given for the values computed from the data of this crystal.

TABLE VII

Cross Coupling Moduli Derived from Measured
Velocities of Propagation

C_{ij}	Velocity* Used for Computing	Stiffness Moduli	
		Value (dynes/cm ² x 10 ¹²)	Probable Error (%)
C_{12}	V_7	0.4639	± 1.36
	V_8	0.4651	± 0.71
	V_{10}	0.4715	± 1.36
	V_{11}	0.4630	± 0.71
C_{13}	V_1	0.2174	± 3.76
	V_2	0.2181	± 1.82
C_{23}	V_4	1.0797	± 0.95
	V_5	1.0779	± 1.22

*Subscript refers to velocity values given in
Table VI.

Summary of Data: The weighted mean values of all nine stiffness moduli are given in Table VIII, along with the compliance moduli derived from the former. Table IX gives various elastic parameters derived from the fundamental moduli.

TABLE VIII

Elastic Moduli of Uranium

Stiffness Moduli (dynes/cm ² x 10 ¹²)	Compliance Moduli (cm ² /dynes x 10 ⁻¹²)	Stiffness Moduli (dynes/cm ² x 10 ¹²)	Compliance Moduli (cm ² /dynes x 10 ⁻¹²)
$C_{11} = 2.1474 \pm 0.14\%$	$S_{11} = 0.4906$	$C_{33} = 2.6711 \pm 0.14\%$	$S_{33} = 0.4798$
$C_{12} = 0.4649 \pm 0.58\%$	$S_{12} = -0.1191$	$C_{44} = 1.2444 \pm 0.10\%$	$S_{44} = 0.8036$
$C_{13} = 0.2177 \pm 1.47\%$	$S_{13} = 0.0079$	$C_{55} = 0.7342 \pm 0.10\%$	$S_{55} = 1.3620$
$C_{22} = 1.9857 \pm 0.14\%$	$S_{22} = 0.6741$	$C_{66} = 0.7433 \pm 0.10\%$	$S_{66} = 1.3453$
$C_{23} = 1.0791 \pm 0.71\%$	$S_{23} = -0.2625$		

TABLE IX

Elastic Parameters Derived from Fundamental
Moduli for Alpha Uranium

Parameter	Value	Definition
$E_{[100]}$	2.038×10^{12} dynes/cm ²	Young's modulus in [100] direction
$E_{[010]}$	1.484×10^{12} dynes/cm ²	Young's modulus in [010] direction
$E_{[001]}$	2.084×10^{12} dynes/cm ²	Young's modulus in [001] direction
C_{44}	1.2444×10^{12} dynes/cm ²	Shear modulus for (001)[010] or (010)[001]
C_{55}	0.7342×10^{12} dynes/cm ²	Shear modulus for (001)[100] or (100)[001]
C_{66}	0.7433×10^{12} dynes/cm ²	Shear modulus for (010)[100] or (100)[010]
$\beta_{[100]}$	0.380×10^{-12} cm ² /dyne	Linear compressibility in [100]
$\beta_{[010]}$	0.292×10^{-12} cm ² /dyne	Linear compressibility in [010]
$\beta_{[001]}$	0.225×10^{-12} cm ² /dyne	Linear compressibility in [001]
β_V	0.897×10^{-12} cm ² /dyne	Volume compressibility
K	1.115×10^{12} dynes/cm ²	Bulk modulus
σ_{12}	+0.243	Poisson's ratio: $\sigma_{ij} = \frac{\text{compressive strain in } j \text{ direction}}{\text{tensile strain in } i \text{ direction}},$ for stress in i direction; 1, 2 and 3 refer to [100], [010] and [001], respectively
σ_{21}	+0.177	
σ_{13}	-0.017	
σ_{31}	-0.017	
σ_{23}	+0.390	
σ_{32}	+0.548	

The bulk modulus derived from these data is approximately 11% greater than that determined by Bridgman by hydrostatic pressure in 1931.³ Other values for the dynamically determined bulk modulus of polycrystalline uranium are 1.08 and 1.26×10^{12} dynes/cm².⁴

b. Thermal Expansion of Single Crystals of Alpha Uranium
(L. T. Lloyd)

Dilatation curves in the [100] and [001] directions have been obtained from a pseudocubic single crystal of alpha uranium. The techniques of specimen preparation and testing were described in ANL-5790.

³P. W. Bridgman, "The Physics of High Pressures," MacMillan Co. New York, p. 161 (1931).

⁴H. L. Laquer, W. E. McGee and M. F. Kilpatrick, "Elastic Constants of Uranium," Trans. ASM 42 771-782 (1950).

The same specimen (T_D) was tested first in the [100] direction and then in the [001] direction. The directions were accurate within 0.5° , as determined by Laue back-reflection photograms. The original lengths at 25°C were 0.1299" and 0.1156" for the [100] and [001] directions, respectively. Four curves were obtained for each direction; the data reported in Table X and shown graphically in Figures 11 and 12 are from reproducible curves. Data calculated from the equations fitted to the lattice parameter measurements reported by Bridge, Schwartz and Vaughan⁵ are included for purposes of comparison. Microscopic observations of the crystal's faces after both sets of tests indicated that no appreciable changes had occurred in the structure.

TABLE X
Expansion of Alpha Uranium in the [100] and
[001] Directions.*

Temperature ($^\circ\text{C}$)	Expansion in the [100] Direction (in./in. $\times 10^4$)		Expansion in the [001] Direction (in./in. $\times 10^4$)	
	Measured from Single Crystal	Calculated from Equation**	Measured from Single Crystal	Calculated from Equation**
50	4.61	5.87	5.16	4.97
75	10.22	11.91	10.34	10.08
100	15.84	18.14	15.51	15.29
125	22.47	24.59	22.38	20.70
150	29.10	31.24	28.70	26.24
175	37.24	38.13	35.01	32.01
200	44.38	45.26	42.45	38.00
225	52.53	52.66	49.34	44.24
250	59.67	60.34	56.22	50.74
275	66.82	68.30	63.68	57.54
300	74.97	76.55	72.26	64.65
325	84.13	85.13	81.67	72.10
350	93.29	94.03	90.56	79.88
375	102.95	103.28	100.83	88.05
400	113.11	112.88	112.34	96.62
425	123.28	122.86	120.82	105.61
450	133.94	133.22	129.97	115.04
475	144.41	143.99	141.37	124.93
500	155.77	155.15	152.21	135.32
525	167.94	166.75	163.05	146.22
550	179.10	178.78	174.45	157.62
575	190.26	191.27	184.73	169.59
600	203.43	204.23	196.12	182.14
625	221.10	217.66	208.08	195.27
650	235.26	231.60	219.48	209.02

*Based upon 1" specimen length at 25°C

**The equations for expansion in the [100] and [001] directions, as derived from the data of Bridge, Schwartz and Vaughan, are:

$$[100] \quad L_T = L_0^\circ\text{C} (1 + 22.50 \times 10^{-6}T + 11.97 \times 10^{-9}T^2 + 14.74 \times 10^{-12}T^3)$$

$$[001] \quad L_T = L_0^\circ\text{C} (1 + 19.37 \times 10^{-6}T + 5.71 \times 10^{-9}T^2 + 23.26 \times 10^{-12}T^3)$$

where L is length and T is expressed in $^\circ\text{C}$.

⁵J. R. Bridge, C. M. Schwartz, and D. A. Vaughan, "X-Ray Diffraction Determination of the Coefficients of Expansion of Alpha Uranium," Trans. AIME 206 1282 (1956).

Figure 11. Expansion of Alpha Uranium in the [100] Direction as a Function of Temperature.

Macro 22345

Figure 12. Expansion of Alpha Uranium in the [001] Direction as a Function of Temperature.

Macro 22346

The measured and calculated curves for expansion in the [100] direction agree quite well; the experimental curve for the [001] direction falls above the calculated curve. This is in agreement with the results reported in ANL-5790 where the measured [010] curve fell below the calculated [010] curve. Apparently, in the polycrystalline material studied by Bridge, Schwartz and Vaughan, the contraction of the [010] direction was restrained by the [001] expansion. In the case of single crystals this situation does not arise. The volume expansions calculated from lattice parameter data and from single crystal measurements should agree well. Such comparisons are pending the procurement of check data for expansions of single crystals in the three principal directions.

c. Recrystallization of Heavily Cold-Rolled Uranium Sheet
(W. R. Yario, L. T. Lloyd)

The investigation of recrystallization in heavily cold-rolled alpha-uranium sheet without a texture change, reported by Cabane and Petit,⁶ has been essentially concluded during this quarter. The preparation of the high-purity sheet, reduced 80% in thickness, along with a metallographic study of annealed samples, was reported in ANL-5623. This study showed that various bands of the deformed material behaved quite differently during the course of annealing. A metallographic investigation of specimens annealed for short times at 400°C, and long times at 320° and 360°C, was reported in ANL-5717. During the last stages of recrystallization, relatively large grains were observed in the process of absorbing extremely fine recrystallized grains. Dilatation studies reported in ANL-5643, -5709, and -5790 showed that the changes in mean expansion coefficient between 200° and 300°C for annealed specimens over that measured for the as-cold-rolled specimens were not a simple function of the measured volume percent recrystallization. For samples annealed at 400°C there is essentially no change in texture up to 80% recrystallization, as measured by the change in mean expansion coefficient; however, the texture changes rapidly upon further recrystallization. The specimens annealed for long times at 320° and 360°C had only a small change in mean expansion coefficient up to 98% recrystallization. Two specimens annealed together for 400 hours at 320°C to 85% recrystallization had anomalous and appreciable increases in mean expansion coefficients.

In this quarter an attempt was made to resolve the discrepancy caused by the specimens annealed for long times at 320°C and to completely recrystallize a specimen at 360°C. Two specimens were again annealed for 400 hours at 320°C. The mean expansion coefficients were determined and one of the specimens was metallographically examined to measure the amount of recrystallization.

⁶G. Cabane and J. Petit, "Study of the Annealing of Rolled Uranium,"
Revue de Metallurgie 51 603-613 (1954).

As shown by Table XI the mean expansion coefficients for the 320°C annealed specimens did not increase over the as-rolled values, but showed a noticeable decrease. This decrease in coefficient is not considered significant and is probably due to the severe inhomogeneity in the sheet (ANL-5623). In addition, these latest 320°C annealed specimens have recrystallized less than the specimens reported in ANL-5790. For those reasons it is believed that during the course of the 400-hour anneal of the earlier samples (reported in ANL-5790) the furnace temperature probably rose to a value considerably higher than 320°C.

TABLE XI

Volume Percent Recrystallization and Change in Mean Expansion Coefficient of Heavily Cold Rolled High Purity Uranium Sheet after Various Annealing Treatments

Annealing Treatment		Volume Percent Recrystallization	Change of Mean Expansion Coefficient* (in./in. - °C x 10 ⁶)
Temperature (°C)	Time (hr)		
320	400	75	-1.04
320	400	-	-0.21
360	400	94	+1.29
360	400	-	+1.88

*Change in mean expansion coefficient between 200° and 300°C over the average of that obtained from three cold-rolled specimens. Dilatation tests were made in a direction perpendicular to the rolling direction and lying in the rolling plane. During this quarter an additional as-rolled measurement of mean expansion coefficient has been made which somewhat lowered the average value.

After annealing two specimens for 400 hours at 360°C the mean expansion coefficients have increased only slightly over the as-rolled values to about the same values reported for the specimens annealed 200 hours at 360°C (ANL-5790). The specimen examined metallographically was 94% recrystallized. There were very few fine-grained areas remaining and the contrast between recrystallized grains was fairly sharp (Figure 13).

This entire study has shown that after severe deformation of uranium at room temperature the number of nucleation sites, as expected, is very large. This is indicated by the extremely small size of the grains formed in the initial stages of recrystallization. These as-recrystallized grains appear to have the same average orientation as in the as-rolled sheet, or at least one very similar to it. This is indicated by the fact that

Figure 13

High-Purity Uranium Rolled
80% at Room Temperature and
Annealed at 360°C for 400 hours
(94% Recrystallization).

Micro 22249

1000-X

Polarized Light

there is essentially no change in mean expansion coefficient up to 80% recrystallization for the 400°C annealed specimens, and no great change in mean expansion coefficient up to 96% recrystallization for the 360°C annealed specimens. For longer periods of time at 400°C, the temperature is high enough for grains of a particular range of orientations to grow at the expense of others and to effect an orientation change. This viewpoint has been substantiated by the pronounced change in mean expansion coefficient as the amount of recrystallization increases from 80 to 100%. In addition, during these last stages of recrystallization relatively large grains have been observed to absorb the fine grains. This absorption of small grains by larger ones has not been found to a great extent in specimens annealed for short times at 400°C, or in specimens annealed for times up to 400 hours at 320° and 360°C. For these reasons, it is believed that the change in mean expansion coefficient occurring at 400°C and not observed at 360°C is caused by preferential grain growth.

Therefore it is concluded that heavily cold-rolled uranium sheet can be almost completely recrystallized at temperatures from 320°C on up to somewhat below 400°C without a texture change. This texture may then be changed by annealing at temperatures of 400°C or higher.

d. Self-Diffusion in Uranium (S. J. Rothman, L. T. Lloyd)

Refining of Highly Depleted Uranium: Twenty-four kilograms of highly depleted uranium anodes for fused salt purification⁷ were produced from the 42 kilograms of reduction buttons sent to us by Mallinckrodt. Eighty-eight kilograms of uranium as UF₄ had been sent to them; 46 kilograms were shotted through the slag. Attempts to recover this metal were

⁷B. Blumenthal and R. A. Noland, "High Purity Uranium," Progress in Nuclear Energy, Vol. 1, Series V, p. 62 (1956).

made by melting the slag under vacuum. Unfortunately, the metal did not coalesce; the particles of metal were separated by an oxide skin. This unusually low yield was due to the poor quality of the UF_4 .

The electrolytic purification of this metal is in progress. The first fused salt electrolytic bath was made up using UO_3 instead of UF_4 as the carrier salt. Crystals were obtained from this bath and an ingot weighing 1.28 kilograms has been cast from these crystals. No analyses of this metal are available yet. The Vycor cell in which the electrolysis was being carried out broke after 3 weeks of use; a new bath, made up with UF_4 , is now in operation.

About 38 kilograms of highly depleted uranium as UO_3 had been obtained from Oak Ridge along with the UF_4 that was sent to Mallinckrodt for reduction. This UO_3 was converted to UF_4 , which in turn was bomb-reduced to uranium metal⁷ with a yield of 91%. This metal and the anode stubs will be melted together and cast into more anodes.

Some of the UF_4 from the batch that was sent to Mallinckrodt was also bomb-reduced by the same techniques. No slag metal separation was obtained, substantiating the poor quality of the UF_4 .

Experiments on Self-Diffusion in the Gamma Phase: A sample for gamma-phase diffusion had been prepared before the metallographic studies described in ANL-5790 were carried out. This sample was annealed in argon at $952^\circ \pm 2^\circ \text{C}$ for 499 hours and sectioned. The sections were analyzed mass spectrographically. Penetration of U^{235} into the base metal was found, with a diffusion coefficient between 10^{-7} and $10^{-8} \text{ cm}^2/\text{second}$.

Although the sample had been heated and cooled quite slowly, the interface on which the isotope had been deposited had warped badly. Thus, no accurate values of the diffusion coefficient could be obtained. Therefore the sample was sectioned on a lathe without taking particular care to align it after each cut.

Table XII shows the U^{235} concentration as a function of the distance from the interface. The distances were calculated from the weight of the specimen before and after each lathe cut. Before sectioning, 0.15" was removed from the diameter in an attempt to eliminate the effects of diffusion from the cylindrical surface. The initial concentration of U^{235} in the base metal was 0.7% according to Special Materials records. The lowest concentration in the diffused sample was 0.770%. If one then takes the characteristic distance of the diffusion $x = 2 \sqrt{Dt} \approx 0.5 \text{ cm}$, one obtains a D value between 10^{-8} and $10^{-7} \text{ cm}^2/\text{second}$.

TABLE XII

U^{235} Concentration as a Function of
Distance from the Interface

Concentration of U^{235} (%)	x - Distance from Center of Section to Interface (10^{-3} cm)
0.908	3.23
0.869	9.21
0.877	13.93
0.860	18.84
0.870	24.31
0.853	30.50
0.866	36.06
0.868	40.39
0.860	46.47
0.853	53.12

*The U^{235} concentrations are accurate to $\pm 1\%$ of the indicated values.

This value is of the same order of magnitude as the self-diffusion coefficient at the same fraction of the melting temperature ($0.873 T_m$) for thorium,⁸ iron,⁹ and sodium.¹⁰ It is less than that for tantalum.¹¹ These are the only body-centered cubic metals for which self-diffusion has been measured. The value is much greater than the values found for face-centered cubic metals. It is also greater than the chemical diffusion coefficients reported for the uranium-zirconium system.¹² Since more uranium self-diffusion experiments are now under way, these semi-quantitative results will not be analyzed further.

Development Work: The unsatisfactory diffusion interfaces and sputtered layers obtained in the present apparatus were described in ANL-5790. It was felt that cleaner interfaces and sputtered layers might be obtained in a smaller, all-glass system, using commercially obtainable, mass spectrographically pure gas, or in a sealed off, all-glass apparatus,

⁸ C. J. Meehan, NAA-SR-1714 (1957).

⁹ C. E. Birchenall and R. F. Mehl, J. App. Phys. 19 217 (1948).
C. E. Birchenall and R. F. Mehl, Trans. AIME, 188 144 (1950).
F. S. Buffington and M. Cohen, Trans. AIME, 194 859 (1952).

¹⁰ N. H. Nachtrieb, E. Catalano and J. A. Weil, J. Chem. Phys. 20 1185 (1952).

¹¹ P. L. Gruzin, "Problems of Metallography and the Physics of Metals," p. 570, Moscow (1955).

¹² Y. Adda and J. Phillibert, Comptes Rendus 242 3081 (1956).

where the sputtered uranium could act as a getter.¹³ The latter experiment was carried out. It failed because the specimen was too close to the glass. Sputtered metal deposited on the glass, arcing to the deposit occurred, and the heat of the arc cracked the glass.

Two other sputtering experiments were performed using the bell jar. In the first, uranium was deposited onto uranium that had not been cleaned by sputtering. In the second, the usual sputtering techniques were used, but with tank argon. The uranium metal layers deposited in both cases were nonadherent.

The gas-purification train components were changed. Although the uranium turnings removed from the gas train looked quite clean, the O₂ concentration was lowered from 0.07% to 0.02% when they were replaced with zirconium turnings.

Experiments on making uranium self-diffusion couples by pressure welding were tried. Neither pressure welding in a vacuum, nor the picture frame technique,¹⁴ worked. Some success was achieved using the method described by Hum.¹⁵ In view of the success of the sputtering method, this line of investigation has been temporarily abandoned.

3. Uranium-Base Alloys

a. Uranium-Plutonium Alloys (C. J. Biver)

Preparation of Alloys: A series of 10 alloys, at intervals of 5 a/o, has been prepared by arc melting with nonconsumable electrodes. The buttons made in this manner were approximately 1/4" thick and 1/2" in diameter, and had an average weight of 15 grams. The buttons were melted three times, for a total melting time of 2 minutes, and were turned over after each melting to insure uniformity of composition. The charge consisted of high-purity plutonium obtained from the Los Alamos Scientific Laboratory and ANL high-purity uranium. The compositions of these materials are listed in Table XIII.

Both metals were electrocleaned. The uranium cleaned in a standard phosphoric acid bath at a current density of 0.075 amp/cm² had a stain-free surface which remained bright in the glovebox atmosphere for several hours. Plutonium showed the cleanest surfaces when a saturated solution of potassium carbonate in water was used at room temperatures.

¹³This idea was suggested by Dr. M. Neumann, Chicago-Midway Laboratories.

¹⁴J. H. Kittel, "Layer Formation by Diffusion Between Some Reactor Construction Metals," ANL-4937 (1949).

¹⁵J. K. Y. Hum, UCRL-4488.

TABLE XIII

Composition of High-Purity Uranium and High-Purity Plutonium

Element	ANL Uranium (ppm)	LA Plutonium ¹ (ppm)	Element	ANL Uranium (ppm)	LA Plutonium ¹ (ppm)
CHEMICAL			SPECTROCHEMICAL (Cont'd.)		
C	60	25	Cu	1	3
O	6-7	40	Fe	10	L 25
N	L 10	-	K	L 50	-
SPECTROCHEMICAL			La	-	L 10
			Li	L 1	-
Ag	L 1	-	Mg	1	L 5
Al	5	5	Mn	L 1	10
As	L 10	-	Mo	L 20	L 30
B	L 0.1	-	Na	L 10	L 10
Be	L 0.5	-	Ni	L 5	L 20
Bi	L 1	-	P	L 50	-
Ca	L 20	L 5	Pb	L 1	-
Co	L 5	-	Sb	L 1	-
Cr	L 1	L 20	Si	20	115
			Sn	L 5	-
			Ti	L 50	-
			Zn	L 50	L 10

¹Elements spectrochemically present in trace quantities are not reported.

A current density of 0.40 amp/cm² at a closed circuit voltage of 10 volts produced a lustrous surface. Both the plutonium and uranium were rinsed and dried in absolute ethyl alcohol.

The components of the charge were placed on the hearth so that the higher melting metal, uranium, was on top. This procedure has been found to reduce greatly the plutonium losses by evaporation. The buttons prepared in this manner had bright, clean surfaces which required no cleaning other than an alcohol wash. The average weight loss during melting was 1.5%, which may be attributed almost entirely to plutonium vaporization, since repeated arc melting of two 20-gram samples of pure uranium under the same conditions has shown an average loss of only 0.05% per cycle.

The 10 and 15 a/o plutonium alloys were homogenized for 5 days at 630°C, without prior deformation, and then furnace cooled to room temperature at an average rate of 1°C/minute. These alloys were subsequently sectioned to obtain specimens for metallography, X-ray, chemical and spectrochemical analyses, and for heat-treatment studies.

The remaining alloys of this initial series have been deformed by hot pressing at 630°C. The reduction in thickness was approximately 50% in all cases, the entire reduction having been made in one operation. The apparatus used was a tensile testing machine with a small resistance furnace built around the pressing die assembly. The buttons were placed

between molybdenum sheets during pressing. The average pressure at 50% deformation was approximately 8400 lb/in.², the load being applied at a rate of 800 lb/minute to a maximum of 4000 pounds. The specimens were given an initial preheat of 30 minutes in all but two cases, before the load was applied. The two buttons which were not preheated for 30 minutes but only for 10 minutes exhibited small edge cracks, which were apparently the result of cold pressing. The buttons which were successfully pressed were free of defects and had clean, smooth surfaces. The deformation of these buttons is expected to reduce the time required for homogenization.

The uranium-plutonium alloys of this preliminary series have thus far shown a considerable degree of resistance to the disintegration to powder which is observed in alloys prepared by other methods. After a period of 3 weeks, alloys prepared in the arc furnace have shown only a slight dulling of the originally bright surface and a negligible weight increase.

b. Transformation Temperatures in the Uranium-Carbon System
(B. Blumenthal)

The data for the alpha-to-beta transformation temperature in high-purity uranium (ANL-5717) and uranium-carbon alloys (ANL-5790) have been confirmed by experiments on new specimens.

The method of extrapolation to the equilibrium transformation temperature was given in ANL-5717. The pertinent equations are of the form:

$$T_H = a_1 + b_1 \log r \quad ;$$

$$T_C = a_2 + b_2 \log r \quad ,$$

where T_H and T_C are the transformation temperatures ($^{\circ}\text{C}$) on heating and cooling, respectively, r is the rate of heating or cooling, and a_1 , a_2 , b_1 and b_2 are constants. The equilibrium transformation temperature T is given by the relationship:

$$T = T_H = T_C \text{ at suitably low values of "r".}$$

Alpha-Beta Transformation of High-Purity Uranium: The data for specimen B724, cycled in a UO_2 crucible, were given in ANL-5717. The experiments yielded the equations:

$$T_H = 671.79 + 0.064 \log r$$

$$T_C = 662.2 - 0.151 \log r$$

and

$$T = 669.0^{\circ}\text{C at } r = 2.4 \times 10^{-45} \text{ }^{\circ}\text{C/minute.}$$

A second sample, B-728-T, was cycled bare. The data yielded the following results:

$$T_H = 673.96 + 4.19 \log r$$

$$T_C = 659.61 - 3.76 \log r$$

and

$$T = 666.4^\circ \text{C at } r = 0.016^\circ \text{C/minute.}$$

The mean equilibrium alpha-to-beta transformation temperature in high-purity uranium, as determined from the above data, is $667.7^\circ \text{C} \pm 1.3^\circ \text{C}$.

Alpha-Beta Transformation of Low Carbon Uranium-Carbon Alloys: Data for sample B-727-B, cycled bare, were reported in ANL-5790. The equations and equilibrium transformation temperature are as follows:

$$T_H = 671.89 + 2.82 \log r$$

$$T_C = 661.27 - 1.86 \log r$$

and

$$T = 664.9 \text{ at } r = 0.0033^\circ \text{C/minute.}$$

A second sample, B-727-T, also cycled bare, yielded the following results:

$$T_H = 676.55 + 6.29 \log r$$

$$T_C = 662.19 - 3.09 \log r$$

and

$$T = 666.9 \text{ at } r = 0.029^\circ \text{C/minute.}$$

The mean equilibrium alpha-to-beta transformation temperature in low carbon uranium-carbon alloys, as determined from the above data, is $665.9^\circ \text{C} \pm 1^\circ \text{C}$.

The mean alpha-to-beta equilibrium transformation temperature of uranium-carbon alloys occurs at a value of 1.8°C lower than that temperature in high-purity uranium. These data show the reaction to be of an eutectoid type:

- c. Properties of Uranium-Fissium Alloys (S. T. Zegler, M. V. Nevitt)

Microstructure, density and thermal arrest data have been obtained for several injection cast, nominal U-5 w/o Fs and U-8 w/o Fs alloys.

Materials: One nominal U-5 w/o Fs alloy (Casting No. 5-1) and one nominal U-8 w/o Fs alloy (Casting No. 8-1) were prepared from scrap uranium containing from 300 to 800 ppm carbon. The castings, 0.145" diameter x 15" long, were part of a group of 15 castings simultaneously injection cast from the same melts. Table XIV details the chemical analyses of two castings, Nos. 3A4 and 4A2, made from the same melts, respectively, as Nos. 5-1 and 8-1. The analyses show no gross chemical segregation of the fission elements except for zirconium in the U-8 w/o Fs alloy. The total fission contents of both alloys, particularly that of the U-8 w/o Fs alloy, were somewhat lower than the intended nominal contents.

TABLE XIV

Chemical Analyses of Uranium-Fission Alloy Castings

Fission Element	Nominal 5 w/o Fs		Nominal 8 w/o Fs	
	Cast 3A4-Top (w/o)	Cast 3A4-Bottom (w/o)	Cast 4A2-Top (w/o)	Cast 4A2-Bottom (w/o)
Mo	2.59	2.53	2.99	2.97
Ru	1.72	1.74	2.01	2.03
Rh*	0.297	0.297	0.475	0.475
Pd	0.161	0.188	0.223	0.217
Zr	0.029	0.022	0.029	0.053
Nb*	0.006	0.006	0.01	0.01
Total w/o Fs	4.803	4.783	5.737	5.755

*No analyses were made for rhodium and niobium. The given values of w/o are based on charged amounts and assume no losses on melting.

One nominal U-5 w/o Fs alloy casting, 0.145" diameter x 6" long, prepared from higher purity uranium (less than 100 ppm carbon) was also studied.

Microstructures: The microstructures of the cross sections of the tops and bottoms of the three castings are shown in Figures 14 and 15. Castings No. 5-1 and 8-1 showed fine grained matrices with a two-phase product of a solid state transformation, presumably an eutectoid, in grain boundaries (Figure 14). The matrix grains responded to polarized light in the manner of isotropic material, suggesting the grains were a retained high-temperature phase, most probably the gamma phase. The transformation product in grain boundaries varied markedly in amount from top to bottom in both alloys, greater in the bottoms. The U-8 w/o Fs alloy, in contrast to the U-5 w/o Fs alloy, evidenced greater amounts of transformation product at both top and bottom.

Figure 14. Microstructures of As-Cast Uranium-Fissium Alloys.

14-a. U-5 w/o Fs Alloy - Top

Micro 21925 Bright Field 500-X

Casting No. 5-1

14-b. U-5 w/o Fs Alloy - Bottom

Micro 21926 Bright Field 500-X

Casting No. 5-1

14-c. U-8 w/o Fs Alloy - Top

Micro 21927 Bright Field 500-X

Casting No. 8-1

14-d. U-8 w/o Fs Alloy - Bottom

Micro 21928 Bright Field 500-X

Casting No. 8-1

Figure 15. Microstructures of As-Cast U-5 w/o Fs Alloy
Made from High-Purity Uranium.

15-a. Top

15-b. Bottom

Micro 21959 Bright Field 500-X

Casting No. 5-4

Micro 21960 Bright Field 500-X

Casting No. 5-4

The U-5 w/o Fs alloy casting (No. 5-4) prepared from high-purity uranium showed the same microstructures at both top and bottom (Figure 15). The alloy was fine grained and essentially single phase except for the presence of numerous, uniformly distributed, small angular inclusions. The matrix grains again appeared to be retained gamma phase. The fact that no decomposition product was in evidence suggests that the alloy was either cooled more rapidly from the melt than castings No. 5-1 and 8-1 or that with lower carbon the eutectoid transformation is suppressed to lower temperatures.

Densities: Density measurements were made of the tops and bottoms of each of the castings by the carbon tetrachloride displacement method. Five determinations were made of each specimen, approximately 1" long, following an initial electropolish in H_3PO_4 solution to remove oxidation products. The five determinations and calculated standard deviations of the averages for a 95% confidence level are given in Table XV.

Thermal Arrests: A 20-gram arc-melted and cast button prepared from the top of casting No. 5-1 (U-5 w/o Fs) was used for determining the temperature of the solid state transformation indicated from the microstructural studies. The button was heated to $800^\circ C$, held for approximately 15 minutes, then slow cooled to $492^\circ C$. A Chromel-Alumel thermocouple inserted in the button was used to determine thermal arrest temperatures. A single arrest was observed at approximately $628^\circ C$ on cooling at a controlled rate of $1.35^\circ C/\text{minute}$.

TABLE XV

Densities of Uranium-Fissium Alloy Castings

	U-5 w/o Fs Cast No. 5-1		U-8 w/o Fs Cast No. 8-1		U-5 w/o Fs Cast No. 5-4	
	Top	Bottom	Top	Bottom	Top	Bottom
Densities, grams/cc	18.2680	18.1486	17.9565	17.8475	17.9713	18.0155
	18.2680	18.1486	17.9500	17.8537	17.9774	18.0233
	18.2741	18.1464	17.9583	17.8544	17.9733	18.0100
	18.2774	18.1443	17.9543	17.8496	17.9794	18.0221
	18.2680	18.1443	17.9543	17.8516	17.9794	18.0163
Average	18.271	18.146	17.954	17.851	17.976	18.017
Standard Deviation	0.004	0.003	0.003	0.003	0.004	0.005
Standard Deviation of the Average (95% confidence)	0.005	0.003	0.004	0.004	0.004	0.007

d. Phase Diagrams of the Uranium-Fissium Elements
(A. E. Dwight)

Uranium-Molybdenum System: The β -to- α transformation has been found to be relatively sluggish at temperatures close to the equilibrium temperature. An alloy of 0.56 w/o Mo annealed 6 days at 635°C had a microstructure of eutectoidal $\alpha + \gamma$ in a matrix of untransformed β . This, together with the observation that there was no beta transformation after 11 days at 640°C, indicates the equilibrium β -to- α temperature is $637^\circ \pm 3^\circ\text{C}$. Figure 16 shows the uranium-rich portion of the phase diagram as determined in the past quarter.

Uranium-Ruthenium System: The limits of the β field have been investigated during the past quarter by metallographic examination. The extent of the β field and surrounding fields as defined by current data is as shown in Figure 17.

Uranium-Rhodium System: The compound URh_3 has been identified as the L12 structure, $a_0 = 3.991\text{\AA}$, isomorphous with URu_3 .

4. Mechanism of Zirconium Corrosion (R. D. Misch, J. E. Draley)

Another set of resistance measurements was made on corrosion films of zirconium-base, low titanium alloys. Previously, these alloys were examined at 185°C after being corroded at 290°C. The specimens were again exposed at 290°C for times from 16 to 272 hours and film resistance was measured at the same temperature in vacuum.

Figure 16. Uranium-Rich Portion of the Uranium-Molybdenum Phase Diagram

Macro 22308

Macro 22307

Figure 17

Beta Region of the Uranium-Ruthenium Phase Diagram.

The specimens were next aluminized and the emf and resistance across the films were measured at room temperature and at 290°C. The electrical properties were checked in air and vacuum before and after heating to verify reversibility. The emf of the oxide, which forms a corrosion cell, was found to be fairly regular, and sufficiently large and steady to permit a resistance determination without an external voltage source.

The zirconium was negative to the aluminum in the case of emf's measured in vacuum before and after heating. This polarity was expected from the nature of the corrosion model because the effective anode and cathode are the inner and outer surfaces, respectively, of the oxide. However, there were two cases where the opposite polarity was noted. The first case occurred when air was admitted to the specimen under vacuum. This always produced a change to positive polarity, but usually only temporarily. The second case was noted at 290°C when the four specimens with the thickest corrosion products exhibited positive polarities.

The highest emf was -0.50 V, which was much larger than the emf at 290°C observed by Thomas¹⁶ who studied the oxide formed in oxygen. This is perhaps an indication that the electronic transference number of ZrO₂ films is much lower in water (in which the present films were produced) than in dry oxygen. Thomas was not able to obtain reproducible data for the case in which water vapor was present with the oxygen.

TABLE XVI

Resistivity of the Corrosion Films
on Titanium-Zirconium Alloys

Specimen No.	Composition (w/o Ti)	Initial Resistivity at 290°C (ohm-cm)
19	0.05	2.5×10^{10}
20	0.05	2.9×10^{10}
21	0.10	4.6×10^9
22	0.10	5.6×10^9
23	0.25	1.84×10^{10}
24	0.25	8.5×10^9
25	0.50	1.26×10^{10}
26	0.50	4.37×10^{11}

At room temperature the resistances measured were approximately 10^{10} ohms in all cases. At 290°C the resistance tended to increase slightly with time in nearly all cases. The resistances were of the same order of magnitude except for specimen 26. The resistivities are given in Table XVI. Based on similar film thicknesses the best comparison can be made between specimens 19, 20 and 21, 22. The average resistivities are 2.7×10^{10} ohm-cm for the former specimens and 5.1×10^9 ohm-cm for the latter.

This difference is considered to be the reason for the higher corrosion rate of specimens 21 and 22.

¹⁶L. D. Kirkbride and D. E. Thomas, "Ionic Transport in Zirconium Oxide," WAPD-T-309, (2 February 1956)

In the case of specimens 23, 24, 25, 26, the film thicknesses are much larger than for specimens 19 through 22 and the surface was becoming white. Therefore, comparisons are subject to the danger that these films are not as compact as the interference color oxide. This was previously demonstrated by measurements of the capacitance of the corrosion oxide in aqueous electrolytes. An increase in capacitance was found to accompany the appearance of white oxide. This was attributed to cracking of the outer layer of the oxide. The electrical resistance of such a cracked layer would not be significant as a measure of diffusion rates controlling corrosion.

V. ALLOY THEORY AND THE NATURE OF SOLIDS - ACTIVITY 5420

1. Transition Metal Intermediate Phases (A. E. Dwight, M. V. Nevitt)a. Ti₂Ni-Type Phases

Four new isomorphs of Ti₂Ni, having the structure E9₃, have been discovered during this quarter, none of which has been previously reported. The tentative compositions of these, together with lattice parameter values, are given in Table XVII. A careful search has shown that the phase does not exist in the Zr-Pd system. There is some evidence that the stability of the phase in the Hf-Pd system is dependent upon the presence of oxygen. The relation of oxygen content to the stability of all the Ti₂Ni-type phases containing zirconium or hafnium is under study, and work is in progress to refine the lattice parameter values given in Table XVII.

TABLE XVII

Additional Ti₂Ni-Type Phases

Tentative Composition	a ₀ (Å)
Hf - 25 Pd	12.36
Zr - 20 Rh	12.45
Zr - 26 Ir	12.46
Zr - 25 Pt	12.57

b. TiNi₃-Type Phases

Three new examples of the DO₂₄ structure were discovered and are listed in Table XVIII. All three are stable at 900°C and can be retained at room temperature. The absence of the DO₂₄ structure in the Zr-Ni and Hf-Ni systems has been shown. In place of a DO₂₄ ZrNi₃ or HfNi₃, both a face-centered cubic phase and a close-packed hexagonal phase have been found in both systems. A more detailed determination of the phases present from 70 to 80 a/o nickel is in progress.

TABLE XVIII

Lattice Parameters and Structure Types of Intermetallic Compounds

Phase	Structure	a ₀ (Å)	c ₀ (Å)	c/a
ZrPd ₃	DO ₂₄	5.6138	9.2173	1.64
HfPd ₃	DO ₂₄	5.5947	9.1923	1.64
HfPt ₃	DO ₂₄	5.6360	9.2076	1.63
NbOs	A12	9.77		
ThOs ₂	C15	7.7050		
ThIr ₂	C15	7.6615		
NbIr	L10	4.0272	3.8586	0.9581
HfCo ₂	C15	6.9176		

c. Alpha Mn-Type Phases

An alloy of 50 a/o Nb + Os exhibits a diffraction pattern which can be indexed as an A12 structure. The a_0 is given in Table XVIII.

d. Laves-Type Phases

Two new $MgCu_2$ or C15 structure phases were found in the thorium-osmium and thorium-iridium systems. An attempt will be made to find similar phases in the thorium-rhenium and thorium-platinum systems. The existence of a Laves-type phase in the hafnium-cobalt system, previously reported by Elliott,¹⁷ was confirmed. Parameters are given in Table XVIII.

e. CuAu-Type Phases

A new L10 structure phase was found in the niobium-iridium system with parameters as shown in Table XVIII. A similar phase probably exists in the niobium-rhodium system, but the composition has not yet been established.

f. CsCl-Phases

Body-centered cubic (b.c.c.) equiatomic phases were found in the yttrium-copper, yttrium-silver, and gadolinium-silver systems. Further work is necessary to determine whether the structure is the B_2 or CsCl. The absence of a b.c.c. equiatomic phase was established in the yttrium-cobalt, yttrium-ruthenium, yttrium-osmium and yttrium-gold systems.

2. Occurrence and Magnetic Behavior of Cr_3O -Type and Sigma Phases
(M. V. Nevitt)

Preliminary testing of the cryostat for magnetic moment measurements at low temperature is substantially complete. A successful transfer of liquid helium into the cryostat has been made and the liquid helium loss rate was found to be 60 cc of liquid per hour. This rate is in good agreement with the calculated value. Copper-Constantan thermocouples for temperature measurement have been calibrated and these, together with the other measuring circuits, have been installed in the cryostat. Calibration of the induction coil-ballistic galvanometer circuit is in progress, using samples of carbonyl iron and carbonyl nickel.

The scope of the investigation has been widened to include the study of the CsCl phases FeTi, CoTi and NiTi. Samples of these alloys, as well as samples of the sigma phase, have been prepared.

¹⁷R. P. Elliott as quoted by F. Laves, "Crystal Structure and Atomic Size," Trans. ASM 48A 156 (1956).

VI. EFFECTS OF IRRADIATION ON MATERIALS - ACTIVITY 5430

1. Neutron Flux Distribution and Wilkins Effect in Irradiation Specimens (F. R. Taraba, S. H. Paine)

The experimental and analytical work on the radial distribution of thermal neutrons within cylindrically shaped fuel specimens during irradiation has been completed. The results of the experimental work are conveniently summarized in the following set of empirical equations.

- (1) Radial distribution of neutrons, normalized to unity at the surface.

$$n(r, \alpha) = A(\alpha) \left[1 + 0.69713 \alpha \left(\frac{r}{a} \right)^2 + 0.48598 \alpha^2 \left(\frac{r}{a} \right)^4 + 0.33879 \alpha^3 \left(\frac{r}{a} \right)^6 \right]$$

where $0 \leq r \leq a$, and $0 \leq \alpha \leq 2$;

- (2) Average-to-surface ratio of neutron density, taken over the cross section.

$$\bar{n}(\alpha) = A(\alpha) [1 + 0.34856 \alpha + 0.16199 \alpha^2 + 0.084698 \alpha^3]$$

where $0 \leq \alpha \leq 2$;

- (3) Axis-to-surface ratio of neutron density.

$$A(\alpha) = \frac{1}{1 + 0.69713 \alpha + 0.48598 \alpha^2 + 0.33879 \alpha^3} \quad \text{and}$$

- (4) Geometric and absorption parameter.

$$\alpha = a \cdot \Sigma_{\text{abs}}$$

where Σ_{abs} = macroscopic absorption cross section of the material, cm^{-1} , and

a = radius of the rod, cm.

Equation (1) gives the thermal neutron densities at positions, r , along the radius of the rod. Equation (2) gives the average thermal neutron density on the cross section of the rod. The neutron density is normalized to unit neutron density at the surface of the rod, i.e., $n(a, \alpha) = 1$. The parameter, α , is a dimensionless quantity which ties together the physical and geometrical properties of the specimen. The above empirical equations are strictly valid only in the range $0 \leq \alpha \leq 2$. The range of α is sufficiently large so as to include practically all cases of interest.

Figure 18 shows graphs of the functions $n(o,\alpha)$ and $\bar{n}(\alpha)$. Figure 19 is a graph of neutron density versus r/a for various values of the parameter, α .

Figure 18. Axial and Average Cross-Sectional Thermal Neutron Densities in Cylindrical Rods Versus $\alpha (= \alpha \Sigma_{abs})$

Figure 19. Radial Distributions of Thermal Neutron Densities Versus r/a for Several Values of $\alpha (= \alpha \Sigma_{abs})$