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STUDY OF ALUMINUM ALLOYS CONTAINING
UP TO 45 w/o URANIUM

by

Norman E. Daniel
Ellis L. Foster
John A. DeMastry
Arthur A. Bauer
Ronald F. Dickerson

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BATTELLE MEMORIAL INSTITUTE
505 King Avenue
Columbus 1, Ohio

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Norman E. Daniel, Ellis L. Foster, John A. DeMastry,
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Casting techniques for the production of sound homogeneous extrusion blanks composed of aluminum-uranium alloys with 30 to 45 w/o uranium were investigated. Satisfactory extrusion blanks in the form of sleeve castings containing 30 w/o uranium were produced utilizing air-melting techniques. Sound castings 2 in. in diameter containing 45 w/o uranium were produced by conventional vacuum-melting techniques utilizing zirconia crucibles. An extrusion blank containing 45 w/o uranium with side walls 0.55 in. thick was vacuum melted and cast. This casting was essentially sound, porosity being found only in the area of the pour, and was attributed to problems in mold design.

As a supplement to the casting program, exploratory investigations were made of the fabricability and the corrosion resistance of the alloys. The diffusion rate of the uranium from the alloys into 2S aluminum cladding was also studied. All of the alloys, enclosed in heavy-walled-aluminum tubes, were press forged at 800 and 1000 F. The corrosion resistance of the high-uranium alloys is comparable to that exhibited by the aluminum-16 w/o uranium alloy, and diffusion of the uranium from the alloys into 2S aluminum after 30 days at 750 F could not be detected by metallographic techniques. The effects of a 0.5 w/o silicon addition on the melting characteristics of the aluminum-16 w/o uranium alloy were negligible when compared with the effects of small variations in the uranium content.

INTRODUCTION

Aluminum-uranium alloys clad with aluminum are particularly attractive as fuels for low-temperature water-cooled and water-moderated reactors. They are easily fabricated at temperatures below 1000 F, they possess relatively good corrosion resistance in water up to 300 F, and they have good thermal conductivity. Aluminum itself has a low capture cross section for thermal neutrons and is relatively inexpensive.⁽¹⁾

Although alloys in this system containing 16 to 20 w/o uranium have been proven as reactor fuels in relatively low-temperature reactors, two obvious advantages are to be gained by increasing the uranium content of these alloys. In the case of foreign reactors in which these alloys may be used as fuel, the 20 w/o enrichment limit for exported uranium alloys makes an increase in the total uranium content of the alloys imperative. In the case of domestic reactors, an increase in uranium content and the consequent decrease in aluminum would provide better neutron economy.

(1) References at end of report.

U. S. NUCLEAR ENERGY

An increase of uranium contents to amounts of over 30 w/o makes the production of sound homogeneous castings a problem. The porosity normally found in these ingots results from the evolution of hydrogen during ingot solidification, and the wide difference in densities of the alloy components is responsible for the inhomogeneity. Special melting and casting techniques are needed to overcome these difficulties. Although some improvement in quality has been obtained for relatively small flat billets which are subsequently fabricated into flat-plate-type elements⁽²⁾, the production of sound homogeneous ingots of more complicated shapes is a more difficult problem. An example of such an ingot is the extrusion billet required for the production of the tubular elements. With this in mind, a program of research was initiated for the purpose of developing a casting technique which would produce sound, homogeneous-extrusion billets of the required type.

Due to the scarcity of available information concerning the physical characteristics of the alloys, it was also desirable to examine their corrosion behavior in 300 F water, the extent that the increased uranium content might alter diffusion characteristics into 2S aluminum, and their relative extrudability.

It is the object of this report to discuss the results of these studies.

MELTING AND CASTING STUDIES

The first phase of the over-all program consisted of the investigations concerned with the development of a practical melting and casting technique for the production of extrusion billets containing high uranium contents. Experience with the aluminum-16 w/o uranium alloy⁽³⁾ pointed out that porosity in the cast alloy was due to hydrogen evolved from the melt during solidification, and that this problem would become more severe as the molten-metal temperature was increased. The prior work also indicated that homogeneity was a real problem in this system. With these points in mind, the melting and casting study was approached in four steps. The first of these consisted of a study of small experimental melts; the second was an attempt to apply techniques developed for the 16 w/o uranium alloy to the higher uranium alloys in order to determine the upper limit in uranium content that could be cast by these techniques; the third step was a melting and casting study concerned only with the 45 w/o uranium alloy; and the fourth step consisted of applying the results of that study to the casting of an extrusion billet.

Small Experimental Melts

Often many of the effects of the variables encountered in the melting and casting of an alloy can easily and quickly be evaluated on small-scale melts where process control is relatively easy. Therefore, before initiating the melting and casting of the full-size extrusion billets, three series of 1-in.-diameter by 6-in.-long ingots were prepared by techniques which differed from one another in one of the major variables.

Ingots in the first series to be prepared were made of virgin material utilizing center-cut biscuit uranium and high-purity aluminum. The aluminum was melted in a graphite crucible under a dynamic vacuum at pressures less than 2×10^{-3} mm of mercury. When the temperature of the molten aluminum reached 1800 F, the uranium was added in the form of sheet stock 1/4 in. square by 1/8 in. thick. The temperature of the melt was then increased to 2000 F and helium was introduced into the system to obtain a static helium pressure of approximately 1/2 atm. The temperature of the melt was further increased to the pouring temperature, and the melt was poured immediately into heavy-walled graphite molds. A temperature approximately 50 F above the liquidus temperature was selected as the pouring temperature. The ingots obtained appeared sound, but, upon examination by radiographic techniques, there was evidence of unalloyed uranium in the alloys containing 35, 40, and 45 w/o uranium.

A similar series of melts was prepared utilizing manual stirring and a holding period of 10 min at the pouring temperature. These melts were poured into heavy-walled copper molds to effect a rapid quench and thereby inhibit any tendency that the alloys might possess for gravity segregation. Radiographic examination of these ingots revealed extensive porosity of such a nature as to suggest cold shuts due to the rapid chill.

From the results obtained on the preceding series of castings, it was deemed necessary to agitate the melt prior to pouring, and to use a mold that possessed less tendency to rapidly chill the castings than the massive copper and graphite molds previously used. Therefore, a series of melts was made utilizing both a 10-min holding time at the pouring temperature and manual stirring prior to pouring. These melts were poured into thin-walled graphite molds.

The radiographic examination of the melts poured into thin-walled graphite molds revealed little or no porosity in any of the castings except at the top of the 40 and 45 w/o uranium ingots. However, the porosity was minor, extending only 1/2 in. or so from the top and could be attributed to shrinkage. Chemical analyses obtained on these melts appear in Table 1. An examination of the table shows that relatively homogeneous experimental ingots could be produced by the aforementioned technique.

TABLE 1. URANIUM CONTENT OF ALUMINUM-URANIUM ALLOYS CAST INTO SMALL-DIAMETER GRAPHITE MOLDS

Nominal	Uranium Content (Balance Nominally Aluminum), w/o	
	Analyzed	
	Top	Bottom
16	16.1	15.6
25	26.1	24.9
30	30.2	30.0
35	35.4	34.6
40	41.5	39.3
45	45.1	43.4

Initial Casting of Full-Size Extrusion Billets

From the literature surveyed^(1, 3, 4, 5), and from the results obtained on the small-diameter ingots, there appeared to be two major problems to be overcome if satisfactory production-size extrusion billets were to be cast. The first of these was segregation. Though this problem was not seriously encountered in the small melts, it was thought that it would manifest itself to a greater extent in the larger melts. As the uranium content of the alloys increases, controlled solidification of the alloy becomes increasingly difficult to obtain due to the rapid increase in the liquidus temperature and the correspondingly greater temperature difference between the solidus and liquidus temperatures. In the 45 w/o uranium alloy, this temperature difference is approximately 1000 F.

The second major problem connected with these alloys was porosity. This porosity is caused by the release of hydrogen from the aluminum alloy during solidification. As the uranium content of the alloy is increased, the alloying and pouring temperatures must be increased, thus increasing the amount of hydrogen taken into solution. Subsequently during cooling, more hydrogen is released, thereby increasing the porosity of the casting.

In an effort to ascertain the alloy of the highest uranium content that could be melted and cast by the best available air techniques, a series of alloys containing each of the compositions under consideration was prepared. These melts containing 30, 35, 40, and 45 w/o uranium were made utilizing high-purity aluminum and biscuit uranium. The melting and casting techniques used were those found most satisfactory for the production of the 16 w/o uranium alloy. This technique is described briefly as follows:

- (1) The aluminum was melted in a graphite crucible and superheated to 1800 F.
- (2) The uranium was added.
- (3) The melt was mechanically stirred for 10 min.
- (4) The melt was degassed by bubbling helium through for 10 min.
- (5) The temperature was increased to approximately the pouring temperature.
- (6) The melt was skimmed, degassed by bubbling helium for about 5 min, and stirred vigorously.
- (7) The heat was cast into a steel mold.

The pouring temperature for each of the compositions was approximately 50 F above the liquidus. The temperatures of the melts were determined by an optical pyrometer previously calibrated at 1865 F with a Chromel-Alumel thermocouple. All melts were poured into a steel mold which was heated to approximately 1000 F at the time of pouring. A copper insert was used to form the bottom of the mold. This insert provided a preferential heat path that imparted directional solidification to the melt. A

sectional view of the mold setup is shown in Figure 1. The mold was cooled by a flow of water through the inlet on the bottom of the tank. The water impinged on the copper insert in the bottom of the mold and was allowed to rise till it was just below the shoulder shown on the outside of the mold.

The evaluation of these melts consisted primarily of radiographic examination of sections cut from the castings. However, chemical analyses of selected billets were also obtained. These analyses indicated that the magnitude of the segregation from top to bottom of the castings was approximately 5 per cent of the included uranium. The radiographs of the ingots revealed extensive porosity present in all castings containing more than 30 w/o uranium. It is believed that the increased porosity noted in the higher uranium alloys is due to the increased melting and pouring temperatures used in casting the alloys. The increased temperatures permit the absorption of more hydrogen by the melt, thus more hydrogen is evolved upon solidification.

The results obtained on these melts clearly indicated that the major obstacle to the production of acceptable castings was the porosity. It was felt that this porosity was due to the ineffectiveness of bubbling helium through the molten material to remove the hydrogen present in the starting stock. Therefore, a series of melts was made utilizing degassed uranium. It was felt that perhaps some of the hydrogen present in the uranium could be removed prior to alloying and thus decrease the amount of gases available for incorporation into the alloy melt. The uranium was prepared by melting and casting center-cut biscuit under a dynamic vacuum of less than 4×10^{-3} mm of mercury. However, the results obtained on these melts were no better than those obtained on the melts made with virgin material.

Another series of melts was made by melting and casting under a static-helium atmosphere utilizing both as-received and vacuum-degassed melting stock. The helium atmosphere was used when it was observed that the aluminum vaporized to some extent when the temperature approached 2300 F in vacuum. The vacuum-degassed aluminum was prepared by melting the as-received material in a zirconia crucible under a dynamic vacuum of less than 5×10^{-3} mm of mercury and pouring at 1770 F into a graphite mold. These charges were melted in the following manner:

- (1) The aluminum was charged into a graphite crucible.
- (2) The melt chamber was evacuated to less than 5×10^{-3} mm of mercury.
- (3) The aluminum was melted and superheated to 1800 F (optical reading).
- (4) Reactor-grade uranium was added to molten aluminum and the charge was stirred for 10 min to insure its solution.
- (5) The pressure in the chamber was increased to 1/2 atm by the introduction of helium.
- (6) The melt was increased to pouring temperature (100 F above liquidus) and stirred for 10 min.

FIGURE 1. SECTIONAL VIEW OF THE MOLD AND COOLING TANK USED TO PRODUCE THE EXTRUSION BILLETS CAST IN AIR

The same mold minus the cooling apparatus was used for casting under a partial pressure.

FIGURE 2. PRINT OF RADIOGRAPH OF 30 w/o URANIUM EXTRUSION BLANK (TOP SECTION) MELTED IN VACUUM AND CAST IN A PROTECTIVE ATMOSPHERE

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FIGURE 3. PRINT OF RADIOGRAPH OF 30 w/o URANIUM EXTRUSION BLANK (WALL)
MELTED IN VACUUM AND CAST IN A PROTECTIVE ATMOSPHERE

FIGURE 4. PRINT OF RADIOGRAPH OF 30 w/o URANIUM EXTRUSION BLANK (TOP SECTION) MELTED IN VACUUM AND CAST IN A PROTECTIVE ATMOSPHERE

FIGURE 5. PRINT OF RADIOGRAPH OF 30 w/o URANIUM EXTRUSION BLANK (WALL) MELTED IN VACUUM AND CAST IN A PROTECTIVE ATMOSPHERE

- (7) The melt was bottom poured into steel mold with the copper insert at room temperature.

The mold and insert used were the same as those used to produce the series of castings poured in air. A sectional view of this mold is seen in Figure 1. The water tank also shown in this drawing was not used, since these melts were allowed to cool in the melt chamber without the use of a water quench. The pouring temperatures were approximately 100 F above the liquidus temperatures of the alloys.

After casting, the ingots were evaluated by various techniques, including visual and radiographic examination, and chemical analysis. The visual examination was often sufficient to eliminate further evaluation. This was particularly true of the 40 and 45 w/o uranium-alloy ingots. The radiographic examination of the 30 w/o uranium billets revealed no areas of porosity or inhomogeneities within the billets.

Since the primary purpose of these melts was to determine the improvement in ingot soundness, if any, that could be obtained through the use of materials which had been outgassed prior to alloying, the castings were examined with this objective in view. However, castings made from both the as-received and vacuum-outgassed material indicated erratic occurrence of porosity. Therefore, no appreciable improvement in ingot soundness was noted that could be attributed to the prior melting of the alloying materials. While some of the voids were attributed to turbulence in pouring, general areas of porosity at the top of ingot sections were attributed to gas. Prints of radiographs made of extrusion-blank walls and sections from the top of extrusion blanks are shown in Figures 2 through 5. These prints are indicative of the extremes in ingot soundness found in 30 w/o uranium-alloy extrusion blanks.

FIGURE 6. PRINT OF RADIOGRAPH OF LONGITUDINAL SECTION CUT FROM 45 w/o URANIUM EXTRUSION-BLANK WALL

Due to the lack of a suitable source of radiation, radiographs of the walls of the whole billets containing more than 35 w/o uranium could not be made. Therefore, thin sections of the walls were cut from these ingots and radiographed. A typical radiograph of a 1/4-in. longitudinal section of a 45 w/o uranium billet is shown in Figure 6. Extensive porosity is obvious in this casting. This type of porosity was encountered in all thick-walled billets containing 40 and 45 w/o uranium. The chemical analyses of samples obtained from these ingots revealed little segregation from the top to the bottom.

Two-Inch-Diameter Melts of Aluminum-45 w/o Uranium Alloys

Since satisfactory extrusion billets containing 40 and 45 w/o uranium were not produced by the techniques described in the preceding section, the problems involved in casting high-uranium alloys were re-evaluated. The principal problems encountered in the casting of these billets were the incomplete removal of hydrogen gas and the inhomogeneities of the alloy. These problems were aggravated by the relatively complex design of the extrusion billet. Therefore, it was decided to continue the casting study using a simple right-cylinder ingot and to concentrate the effort expended upon the 45 w/o uranium alloy. The main objectives of this phase of the program were the removal of hydrogen and the improvement of the homogeneity of the alloy. Once these problems were solved for simple castings, steps could be taken to apply the most successful technique to the production of extrusion-billet castings. Among the variables investigated were atmosphere, crucible material, outgassing procedure, and the mold temperature. Variations in techniques under which each ingot was poured are given in Table 2. All of the melts in this series were prepared with high-purity aluminum and reactor-grade vacuum-degassed uranium.

The first three ingots in the series were made using the following melting techniques:

- (1) The aluminum was melted under a dynamic vacuum in a graphite crucible. By doing this, the effects of vacuum degassing the aluminum were obtained without the addition of another melt.
- (2) When the temperature of the aluminum reached 1800 F, uranium was added and the melt stirred vigorously to insure complete solution.
- (3) The dynamic vacuum was interrupted and a static helium pressure of about 4 mm of mercury was established in the melt chamber.
- (4) The temperature was increased to 2500 F (platinum-platinum rhodium thermocouple reading).
- (5) The melt was degassed by bubbling helium through the melt while continuously pumping on the system in order to maintain pressure of approximately 4 mm of mercury.
- (6) The melt was tilt poured into an outgassed mold.

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TABLE 2. DESCRIPTION OF 2-IN. -DIAMETER CASTINGS OF ALUMINUM-45 w/o URANIUM ALLOYS

Ingot	Melting Procedure	Crucible	Mold	Pressure on Melt, mm of mercury	Pouring Temperature, F	Pouring Rate	Surface Condition of Casting	Radiographic Examination	Location of Sample	Chemical Analyses, w/o		
										Zr	U	C
1030	Explained fully in text	Outgassed graphite	Graphite 2-in. in diameter by 16 in. long; outgassed	4 of helium	2530	Medium	Poor	Very porous for one-half of longitudinal section	Top Bottom	-- --	44.3 43.7	0.17 0.17
1031	Same as above	Outgassed	Copper; with hot top	5 of helium	2530	Medium	Very poor	More porous than above casting		None taken		
1032	Same as above	Outgassed	Graphite at 1100 F	4 of helium	2500	Medium	Poor	Ingot cracked and porous		None taken		
1033	Melt aluminum in <u>vacuo</u> to 1800 F; add helium to increase chamber pressure to 5000 μ ; add uranium and stir; raise temperature to 2410 F; cool melt to 1650 F and decrease pressure in chamber to less than 50 μ ; raise temperature rapidly to 2500 F and pour	ZrO ₂	Graphite at 1100 F	0.05 of helium	2500	Medium	Fair	Good	Top Middle Bottom	0.008 0.006 0.005	47.5 44.3 42.7	-- -- --
1034	Aluminum melted in <u>vacuo</u> to 1800 F; uranium added	ZrO ₂	Graphite; top 4 in. of mold at 1100 F	0.001 of air	2500	Fast	Good	Sound except for top 4 in. of casting, where shrinkage occurred	Top inside Top outside Bottom inside Bottom outside	0.042 0.042 0.033 0.036	45.5 45.2 44.4 43.3	-- -- -- --
1035	Same as above	ZrO ₂	Graphite; warm, approximately 200 F	0.002 of air	2505	Fast	Good	Sound except for top 4 in. of casting, where shrinkage occurred	Top inside Top outside Bottom inside Bottom outside	0.033 0.034 0.034 0.036	46.2 43.6 44.4 42.5	-- -- -- --

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TABLE 2. (Continued)

Ingot	Melting Procedure	Crucible	Mold	Pressure on Melt, mm of mercury	Pouring	Pouring Rate	Surface	Radiographic Examination	Location	Chemical Analyses,		
					Temperature, F		Condition of Casting		of Sample	w/o		
										Zr	U	C
1036	Aluminum melted in <u>vacuo</u> to 1800 F; uranium added	ZrO ₂	Cold graphite	0.001 of air	2500	Slow	Good	Sound, except for top 4 in. of casting, where shrinkage occurred	Top inside Top outside Bottom inside Bottom outside	0.034 0.035 0.031 0.036	49.9 44.5 44.7 43.4	0.06 0.04 0.05 0.04
1037	Same as above	ZrO ₂	Cold graphite	0.001 of air	2400	Slow	Good	Sound, except for top 4 in. of casting, where shrinkage occurred	Top inside Top outside Bottom inside Bottom outside	0.046 0.044 0.040 0.043	48.6 48.3 43.7 42.1	0.08 0.12 0.10 0.10

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The molds into which these melts were poured were selected for their ability to impart distinctively different cooling rates to the ingots. The first ingot was cast into a cold graphite mold, the second was cast into a cold copper mold, and the third was cast into a graphite mold with an enlarged section for a hot top heated to 1100 F.

As can be seen in Table 2, all of the ingots produced by the above melting technique were porous. However, the over-all quality of the ingot cast into the cold graphite mold was superior to the quality of the other two ingots. Much of the porosity exhibited by this ingot was attributed to turbulence due to the fact that it was concentrated along the pour side of the ingot. The ingot cast into the chilled copper mold was of extremely poor quality. This ingot exhibited evidence of both cold shuts and gaseous porosity. The ingot poured into the heated graphite mold was less porous than the above melts; however, this porosity was concentrated midway along the length of the ingot. This caused the ingot to crack when it was removed from the mold. The surface of this ingot was extremely poor.

Since all of the above castings were porous and since it was evident that this porosity was due to gases evolved during solidification, a melt was made utilizing a melting cycle designed to decrease the gas content of the molten metal prior to pouring. This melting cycle is described briefly as follows:

- (1) The aluminum was melted in a zirconia crucible under a dynamic vacuum of less than 4×10^{-3} mm of mercury and superheated to 1800 F.
- (2) Vacuum-degassed uranium was introduced into the melt and stirred vigorously to insure its complete solution.
- (3) Helium was then introduced into chamber to maintain a static pressure of 5 mm of mercury over the melt.
- (4) The melt temperature was increased to approximately 10 F above the liquidus and held for 10 min.
- (5) The charge was allowed to cool in situ to 1650 F under a dynamic vacuum of less than 5×10^{-3} mm of mercury.
- (6) When the temperature reached 1650 F, the temperature of the melt was increased rapidly to 2500 F and tilt poured into a graphite mold.

The primary purpose of this melting cycle was to remove the gases evolved during solidification by partial solidification of the melt in the crucible before pouring. Therefore, this type of melting cycle should reduce the amount of hydrogen present in the melt immediately before pouring. The casting produced by this technique was sound.

Although some vaporization of the aluminum had been noted when a high vacuum was used in the temperature range above 2300 F, it was deemed desirable to investigate the possibility of casting the 45 w/o uranium alloy by conventional vacuum-melting techniques. Therefore, a series of melts was made and poured under a dynamic

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FIGURE 7. PHOTOGRAPH OF 2-IN.-DIAMETER 45 w/o URANIUM-ALLOY INGOT
CAST INTO A COLD GRAPHITE MOLD

The top 4 in. of this mold was heated to 1100 F. Notice surface irregularities over top 4 in. of ingot.

vacuum of less than 2×10^{-3} mm of mercury. A zirconia crucible was used for these melts to allow for a maximum of inductive stirring and thereby expose more of the metal to the vacuum in the chamber.

The first melt in this group was poured at 2505 F into the 2-in.-diameter graphite mold heated to 1100 F. The second and third castings were tilt poured at the same temperature into warm graphite molds. The only difference between the latter two castings was the rate of pour.

The surface condition of each of the 2-in.-diameter ingots produced is given in Table 2. Also in this table are the results of the radiographic examinations and chemical analyses.

The most satisfactory surface was obtained on the ingots cast into warm molds. In those melts where heat was applied to the complete length of the molds, the surface of the ingot was irregular and required the removal of 1/8 in. or more metal from the diameter to obtain smooth shapes. The ingots cast into warm molds were very smooth and the casting surface faithfully reproduced the internal surface of the mold. A photograph of such an ingot is shown in Figure 7.

The radiographic examination of the melts indicated that sound material could be produced by vacuum melting. However, approximately one-fourth of each of the vacuum-melted ingots contained areas of slight shrinkage porosity. This was most often found in the top of the ingot on the pour side. Shown in Figure 8 are photomacrographs of sections taken from the top and bottom of a typical 2-in.-diameter vacuum-melted ingot. The area of shrink porosity can be seen in the section from the top of this ingot. This type of porosity was common to all of the vacuum-melted ingots. The structure exhibited by the bottom section of the ingot is typical of the ingots produced.

FIGURE 8. PHOTOMACROGRAPH OF SECTIONS TAKEN FROM TOP AND BOTTOM OF INGOT SHOWN IN FIGURE 7

Note porosity in top.

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The chemical analyses obtained from the top and bottom of the ingots reveal longitudinal and transverse segregation. In all of the ingots produced, the uranium content increases from bottom to top and from outside to inside. The samples from two ingots made utilizing a rather high rate of pour deviated less from the nominal composition than samples usually obtained from the top of the ingots. The longitudinal segregation may be reduced a great deal by the proper hot topping of the mold and the proper pouring rate. However, further work would be necessary to establish the requirements necessary for the production of an ingot not exhibiting segregation on the order shown by these ingots.

The evaluation of these melts indicates that sound and essentially homogeneous aluminum-uranium alloys can be produced. The ingots produced in this study yielded 75 per cent or more of sound material which contained ± 5 w/o of the included uranium. While the effects of each of the variables investigated have not been conclusively ascertained, it seems apparent that the real difference in the procedures used for the production of sound castings was the presentation of new metal to the low-pressure surface of the melt. This is the only factor that emerges as being critical. When the graphite crucibles were used with the induction-melting techniques, the induction currents were shielded by the conducting wall of the crucible and no agitation of the melt resulted. With zirconia crucibles, the induced currents in the melt gave a vigorous vertical stirring action that exposed the metal to the low pressure of the vacuum and effectively outgassed the melt. A life expectancy of six to eight melts can be obtained from such a crucible when proper precautions are taken during the heating cycles to minimize the effects of thermal shock. It was found that graphite-crucible life was on the order of two or three melts on high-uranium-content melts made in vacuum.

Casting the Aluminum-45 w/o Uranium Alloy into an Extrusion Blank

At the conclusion of the study of melting variables made on the 2-in.-diameter ingots, it was desirable to determine the application of the improved techniques to the casting of an extrusion blank. Since the alloy castings containing 45 w/o uranium are to be fabricated to fuel elements with the same total fuel loading as the presently used 16 w/o uranium alloy, the cross-sectional area of the high-uranium-alloy castings was reduced. The proposed extrusion-blank casting retained the inside diameter and length measurements, but the outside diameter was decreased.

Therefore, a hollow cylindrical-graphite mold measuring 3.70 in. in ID by 4.70 in. in OD by 7.00 in. long was used to produce the final casting. These dimensions contrast with the 3.60 in. in ID by 6.125 in. in OD by 8.5-in.-long castings required for the production of the 16 w/o uranium-alloy extrusion blanks. The shorter length of the 45 w/o uranium blank was not dictated by any proposed shortening of the extrusion blank but rather by the limited capacity of the tilt-pour vacuum furnace available. The techniques utilized for the production of this casting are described briefly as follows:

- (1) The aluminum charge was melted in a ZrO_2 crucible under a dynamic vacuum of less than 2×10^{-3} mm of mercury. The charge was then superheated to 1800 F and held for 5 min.
- (2) The uranium was added slowly to the molten aluminum.

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- (3) The melt was mechanically stirred to insure complete solution of the uranium.
- (4) When it was thought that all of the uranium was in solution, the temperature of the melt was increased to 2500 F (platinum-platinum rhodium thermocouple reading).
- (5) The 2500 F temperature was maintained for 10 min to allow thorough outgassing of the melt.
- (6) The melt was tilt poured into the cylindrical graphite mold at room temperature.

During the holding period at 2500 F, there was sufficient evolution of material from the melt to impair the vision of the operator during pouring. It is believed that the material evolved was primarily aluminum. Due to the obscured vision of the operator during pouring, a determination of the relative pouring rate as compared with the pouring rates used on the 2-in.-diameter ingot was not possible.

Methods of evaluating this casting were the same as those used on previous castings. These included radiographic examinations and chemical analyses of samples from the top and bottom of the cast cylinder. A photograph of the casting is shown as Figure 9. As can be seen in the photograph, the surface of the billet is extremely good in all areas except for the area shown at the top-right side of the photograph. This area corresponds to the position where the molten alloy was poured into the mold. Samples taken from the top and bottom of the cylinder were found by chemical analyses to contain 44.9 and 46.8 w/o uranium, respectively.

From an examination of the radiographs, it was found that the porosity exhibited by the ingot was concentrated in the same area as the poor surface shown in the above photograph. The location of this porosity at the top in a position relative to the pouring stream indicates that it is a result of a molding fault rather than the effect of absorbed gas. It is felt that sound cylinders of this composition and dimensions can be produced. However, further work is required to correct limitations present in mold design and pouring method.

DIFFUSION STUDIES

The rate of diffusion of uranium from the aluminum-uranium fuel alloy into its protective cladding is of interest to those concerned with reactor design and development. Therefore, the rate of diffusion of uranium from the aluminum-uranium alloys into 2S aluminum was investigated as a supplement to the melting and casting program.

Alloys containing 16, 25, 30, 35, 40, and 45 w/o uranium were prepared for this study by induction melting high-purity aluminum and center-cut biscuit uranium. A zirconia crucible was used and the alloy cast into a 1-in.-diameter copper mold. The melting and casting was done in a vacuum-induction furnace under a pressure of less

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FIGURE 9. PHOTOGRAPH OF ALUMINUM-45 w/o URANIUM ALLOY
EXTRUSION-BLANK CASTING

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than 2×10^{-3} mm of mercury. These 1-in.-diameter ingots were then cut into 1/8-in.-thick wafers which were ground on 200-grit paper prior to bonding.

After grinding, the alloys were cleaned with acetone and sandwiched between degreased 2S aluminum sheet 1/16 in. thick to form the couples for diffusion-rate tests. These couples were pressure bonded at 9000 psi and 930 F for 15 min to insure contact between the core and the cladding.

The bonded diffusion couples were sealed in evacuated Vycor tubes and tested at 750 F for 30 days. Specimens of each composition were examined at the end of 1, 5, 10, 20, and 30 days. Figures 10 and 11 show typical microstructures of the zones between the cladding and the core after 30 days on test. There is no metallographic evidence of diffusion in any of these specimens.

CORROSION BEHAVIOR IN 300 F WATER

Experience with the aluminum-16 w/o uranium alloy has proven its reliability in boiling water. It would, therefore, be desirable for the alloys containing higher uranium contents to exhibit corrosion properties comparable to those exhibited by the 16 w/o uranium alloy. Therefore, an investigation of the corrosion behavior of the 16, 30, 35, 40, and 45 w/o uranium alloys was conducted in conjunction with the melting and casting program.

For this study, three series of similar specimens were prepared and subjected to the corrosive attack of deionized water at 300 F for a period of 30 days. These specimens were examined at the end of 1, 5, 10, 20, and 30 days and evaluated by noting the weight gained during exposure. Specimens in the first series which are listed in Table 3 were prepared by vacuum-induction melting the alloy charges and casting them into 1-in.-diameter copper molds. The 1/8-in.-thick test specimens were machined from the cast ingot.

Specimens in the second series, also listed in Table 3, were prepared from double-melted ingots. Initially, melting and casting were performed in air, with the melt being degassed by bubbling helium through the molten charge prior to pouring into graphite molds. These first-melt ingots were then remelted under a dynamic vacuum of less than 2×10^{-3} mm of mercury and poured into copper molds as soon as the charge reached pouring temperature. Corrosion-test coupons were then machined from these final ingots.

The third series of specimens, listed in Table 3, was prepared in the same manner as the first series except that the machined specimens were heat treated prior to corrosion testing. This treatment was intended to spheroidize the UAl_4 in the matrix and is described in detail in the section of this report dealing with fabrication.

The specimens of all three series were corrosion tested at the same time in an autoclave at 300 F. The results of these tests are shown in Table 3. It can be seen that in all cases except one the weight gains exhibited by the various compositions are of the same order of magnitude. The one specimen that exhibited an abnormal weight

FIGURE 10. PHOTOMICROGRAPH OF DIFFUSION COUPLE BETWEEN 2S ALUMINUM AND ALUMINUM-16 w/o URANIUM ALLOY

As can be seen, there is no evidence of diffusion of the uranium from the alloy into the 2S aluminum. Sample on test 30 days.

FIGURE 11. PHOTOMICROGRAPH OF DIFFUSION COUPLE BETWEEN 2S ALUMINUM AND ALUMINUM-40 w/o URANIUM ALLOY

No evidence of diffusion of the uranium from the alloy into the 2S aluminum can be found. Sample on test 30 days.

TABLE 3. CORROSION TEST RESULTS OF ALUMINUM-URANIUM ALLOYS IN 300 F H₂O

Series	Uranium Content (Balance Aluminum),	Melting Procedures	Heat Treatment	Total Weight Gain, mg per cm ² , In Indicated Time, days				
	w/o			1	5	10	20	30
1	16	Melted in ZrO ₂ crucibles under vacuum and cast into copper molds	None, as cast	0.35	0.37	0.43	0.52	0.52
	30			0.36	0.23	0.38	0.46	0.39
	35			0.36	0.38	0.40	0.52	0.44
	40			0.34	0.34	0.41	0.58	0.53
	45			0.34	0.40	0.45	0.31	0.63
2	30	Melted in air and poured in graphite; remelted in vacuum and poured in copper	None, as cast	0.30	0.33	0.36	0.49	0.49
	35			0.30	0.48	0.54	0.66	0.61
	40			0.38	0.41	0.48	0.62	0.63
	45 ^(a)			0.45	0.65	0.92	1.42	1.82
3	30	Same as Series 1	Sealed in evacuated Vycor and heat treated at 1022 F 24 hr	0.35	0.27	0.38	0.49	0.41
	35			0.34	0.29	0.39	0.34	0.49
	40			0.43	0.48	0.52	0.72	0.67
	45			0.40	0.38	0.42	0.65	0.44
2S aluminum				--	+0.20	+0.32	+0.32	--

(a) Sample very porous; therefore, not a good indication of the corrosion resistance of alloy.

gain was found to be porous. The visual examination of the other specimens indicated that the oxide film formed was typical of aluminum alloys, that is, dense and tenacious. This accounts for the relatively good corrosion resistance of the alloys and the fact that very little difference in the corrosion rates was noted for the various compositions.

The corrosion rate for a 2S aluminum control specimen is also listed in the table. The maximum weight gain for any of the sound alloy specimens is approximately twice that exhibited by the high-purity aluminum. A direct comparison by the weight gain of the alloys and pure uranium is not possible since the uranium oxide film is not adherent and is lost quickly to the corrosive medium.

FABRICATION STUDIES

In any study concerned with the application of an alloy as a fuel in a reactor, consideration must be given to the method or methods by which the material is to be formed. Since the primary concern of this program was the development of melting and casting techniques for the production of extrusion billets, and since some idea of the relative extrudability of the castings was desired, a cursory study was initiated to obtain the desired information.

Before fabrication of the alloys was begun, as-cast specimens of each composition were subjected to two different heat treatments designed to alter the makeup of the matrix phase of the alloys. Specimens for these heat treatments were obtained from ingots vacuum melted and cast into copper molds. The typical as-cast structures of these alloys are shown in Figures 12 through 15. These structures consist of massive UAl_4 particles in a matrix showing the pearlitic structures associated with the eutectic. The eutectic is composed of platelets of essentially pure aluminum and the UAl_4 compound.

Specimens obtained from the same 1-in.-diameter ingots were sealed in Vycor and heat treated at 750 F for 8 hr. The metallographic examination of these specimens failed to reveal any significant change in the microstructures. A similar series heat treated at 1025 F for 24 hr exhibited marked differences in the structure of the continuous phase. Figures 16 through 19 show the microstructures of these specimens after the latter heat treatment. A comparison of these photomicrographs with those of the as-cast material reveal that the typical pearlitic structure in the matrix has been altered by spheroidization of the lamellar UAl_4 by the prolonged heat treatment at the elevated temperature.

Hot-hardness values were obtained on specimens in both the as-cast and heat-treated conditions. These values were essentially the same, differing only with the composition and not with the prior heat treatment. The average values obtained on the specimens at the temperatures of interest were plotted against the temperature. This is illustrated in Figure 20. This method of presenting the data indicates that there is a decided decrease in the hardness values for all of the alloys within the temperature range between 570 and 665 F.

By replotting these data with the composition versus the hardness, as shown in Figure 21, it can be seen that the hardness is primarily a function of the temperature

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FIGURE 12. PHOTOMICROGRAPH OF AS-CAST ALUMINUM-30 w/o URANIUM ALLOY

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FIGURE 13. PHOTOMICROGRAPH OF AS-CAST ALUMINUM-35 w/o URANIUM ALLOY

500X

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FIGURE 14. PHOTOMICROGRAPH OF AS-CAST ALUMINUM-40 w/o URANIUM ALLOY

500X

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FIGURE 15. PHOTOMICROGRAPH OF AS-CAST ALUMINUM-45 w/o URANIUM ALLOY

250X

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FIGURE 16. PHOTOMICROGRAPH OF ALUMINUM-30 w/o URANIUM ALLOY HEAT TREATED FOR 24 HR AT 1025 F

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FIGURE 17. PHOTOMICROGRAPH OF ALUMINUM-35 w/o URANIUM ALLOY HEAT TREATED FOR 24 HR AT 1025 F

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FIGURE 18. PHOTOMICROGRAPH OF ALUMINUM-40 w/o URANIUM ALLOY HEAT TREATED FOR 24 HR AT 1025 F

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FIGURE 19. PHOTOMICROGRAPH OF ALUMINUM-45 w/o URANIUM ALLOY HEAT TREATED FOR 24 HR AT 1025 F

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FIGURE 20. PLOT OF HARDNESS VERSUS TEMPERATURE FOR SEVERAL ALUMINUM-URANIUM ALLOYS

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FIGURE 21. PLOT OF HARDNESS VERSUS COMPOSITION FOR SEVERAL ALUMINUM-URANIUM ALLOYS

and is almost independent of the composition at temperatures above 750 F. This would indicate that all of the alloys can be fabricated at or near the same temperature if reasonable precautions are taken to prevent edge cracking.

Due to the lack of suitable extrusion facilities, it was necessary to adopt techniques which would yield information relative to the extrudability of the alloys. To do this, it was desirable to fabricate in such a manner as to maintain a restraining force on all sides of the alloys to curtail any tendency for the alloys to fracture. Alloys in this system containing up to 35 w/o uranium have been hot rolled successfully by jacketing the material in aluminum prior to fabrication.⁽¹⁾ Therefore, the specimens to be fabricated were enclosed in heavy-walled 2S aluminum cylinders prior to fabrication. These cylinders were prepared by drilling and boring a 1-in.-diameter hole through a 2-in.-diameter piece of round stock 2 in. long.

The as-cast alloy specimens, 1 in. in diameter by 1 in. long, were then press fitted into the aluminum cylinders which were then sealed by welding 2S aluminum plugs into each end. This effectively surrounded each specimen by a tight-fitting envelope 1/2 in. thick of 2S aluminum. One series containing specimens of each composition was then press forged to a total reduction of 50 per cent from a furnace at 800 F. A second series was press forged in a like manner from a furnace at 1000 F. After forging the specimens were examined by radiographic and metallographic techniques. An examination of the radiographs obtained from these specimens failed to reveal any evidence of edge cracking or tearing in any of the alloys. The radiographed disks were then cut in half and macroetched. Photomacrographs of specimens from each series are shown in Figures 22 and 23. An examination of these macroetched specimens also failed to show any evidence of edge cracking or tearing in the alloys. However, a slight indication of edge roughening can be noted in the 45 w/o uranium alloy.

Although the information obtained from hot-hardness determinations and upset press forging of canned specimens can not be construed as an absolute measure of the extrudability of an alloy, the results of these tests indicate that the extrusion of these alloys is feasible.

EFFECT OF SILICON ON THE LIQUIDUS-SOLIDUS TEMPERATURES OF THE ALUMINUM-16 w/o URANIUM ALLOY

The effect of silicon on the liquid and solidus temperatures of the aluminum-16 w/o uranium alloy is of interest in the production of extrusion billets. In the production melting and casting operations of billets, approximately 60 per cent of the charge makeup is recycled material. Some of this material has been AlSi brazed and, therefore, contains silicon in quantities which might be great enough to affect the melting characteristics of the alloy and thereby influence the quality of the casting produced. Therefore, an investigation was undertaken to determine the effect of 0.5 w/o silicon or less on the solidus and liquidus temperature of the aluminum-16 w/o uranium alloy.

Alloys for this study were prepared by master alloying center-cut biscuit uranium and high-purity transistor-grade silicon in a small laboratory-type arc furnace. A charge consisting of master alloy containing 6 w/o silicon, biscuit uranium, and

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FIGURE 22. PHOTOMACROGRAPH OF ALUMINUM-URANIUM-ALLOY SPECIMENS CLAD WITH HEAVY-SECTION 2S ALUMINUM

Press forged at 800 F, 50 per cent reduction.

FIGURE 23. PHOTOMACROGRAPH OF ALUMINUM-URANIUM-ALLOY SPECIMENS CLAD WITH HEAVY-SECTION 2S ALUMINUM

Press forged at 1000 F, 50 per cent reduction.

high-purity aluminum was then vacuum-induction melted and cast into a 1/2-in.-diameter by 6-in.-long split copper mold. The chemical analyses of the alloys prepared in this manner are given in Table 4.

TABLE 4. CHEMICAL ANALYSES OF ALUMINUM-16 w/o URANIUM ALLOYS CONTAINING SILICON ADDITIONS

Alloy	Uranium, w/o	Silicon, w/o
1	16.1	0.05
2	16.2	0.13
3	16.0	0.20
4	16.5	0.31
5	16.1	0.50

A 3/4-in. length was removed from each end of each casting for differential thermal analyses. The remaining portions of the ingots were cold swaged to 50-mil wire for thermal analysis by electrical-resistance techniques.

Two holes were drilled to a depth of 1/2 in. into the 3/4-in. lengths of rods for insertion of temperature-measuring and differential thermocouples. A graphite block was employed as the neutral body for differential measurements. The complete assembly was placed in a Vycor tube to act as a container during melting. A record of the heating curve was obtained on a differential X-Y recorder. The specimens were heated under a vacuum of about 5×10^{-4} mm of mercury at a heating rate of 5 F per min.

The swaged wire was cut into 3-in. lengths for electrical-resistance measurements. The wire was clamped between copper blocks, which acted as current leads to the specimen. Voltage probes, in the form of tantalum wire, were spot welded to the alloy wire at points approximately 3/8 in. from the copper blocks. A 30-gage Chromel-Alumel wire was spot welded to the center of the specimen. The specimen was then coated with a ceramic cement to act as a container during melting.

A current was then applied to the specimen from a constant current source and the voltage drop across the specimen was recorded. The assembly was then inserted in a hot furnace and the temperature change and voltage-drop change across the specimen (which is proportional to the resistivity change) were recorded on a high-speed two-pen recorder.

The differential-thermal-analysis techniques provided an accurate determination of the beginning of melting in the alloy. Within the experimental accuracy of measurement, which is ± 5 F, all of the alloys melted at the eutectic temperature of 1185 F.

Thermal analysis by electrical-resistance measurements indicated that in all of the alloys investigated the end of melting or liquidus temperatures occurred between 1310 and 1335 F. However, as the variations were essentially random, no trend assignable to silicon content could be detected.

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It should be recognized that a 16 w/o uranium alloy melts at approximately 1150 F. A variation of ± 0.5 w/o uranium necessarily denotes a variation in liquidus temperature of about ± 32 F. Thus, the differences detected fall well within the experimental accuracy of alloy preparation. As a result, it may be concluded that differences in liquidus temperatures were a result of variations in uranium content and that such differences were probably greater than those produced by silicon additions of up to 0.5 w/o.

CONCLUSIONS AND RECOMMENDATIONS

The results of the melting and casting studies discussed in the preceding sections of this report show that acceptable castings of aluminum-uranium alloys containing high uranium contents can be produced by a single melting process; however, the specific techniques will vary with the uranium content. Relatively sound homogeneous extrusion-billet castings containing up to 30 w/o uranium were cast using air-melting techniques. Melting was accomplished in a graphite crucible and the heats were tilt poured into a steel mold at approximately 1000 F. Directional solidification of the ingot was produced by utilizing a cold copper insert in the bottom of the mold.

As a result of further study performed with 2-in.-diameter right-cylinder castings, it was found that the alloys containing up to 45 w/o uranium could be cast into right-cylinder ingots that were free of gas porosity and were relatively homogeneous. These alloys were successfully melted and cast by conventional tilt-pour vacuum-induction melting techniques using a zirconia crucible and warm graphite molds. The ceramic crucibles allowed full penetration of the inductive currents and thereby provided for thorough stirring of the melt. This stirring resulted in all of the molten metal being exposed to the vacuum during the hold period and, consequently, insured removal of hydrogen in a manner more efficient than the dry-gas flush. The thorough stirring also assured better homogeneity of uranium in the melt. It is interesting to note that the rammed ZrO_2 crucibles exhibited a longer life than did graphite crucibles when melting in the vicinity of 2500 F.

A relatively sound extrusion billet of aluminum-45 w/o uranium alloy was cast by this technique. This casting was free of gas porosity and had an excellent surface. Because of the mold design, there was considerable shrinkage at the top. It is felt that, with additional research on mold design, extrusion billets containing up to 50 w/o uranium can be cast with a minimum of inhomogeneity and shrinkage.

As a result of the other studies that were performed in conjunction with the casting research, the following conclusions are presented:

- (1) The corrosion resistance of aluminum-uranium alloys containing up to 45 w/o uranium in water at 300 F is comparable to that exhibited by the 16 w/o uranium alloy. No appreciable difference was noted between the corrosion resistance of as-cast and heat-treated material.

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- (2) Metallographic studies failed to indicate any diffusion of uranium from the 16 to 45 w/o uranium alloys into 2S aluminum after 30 days at 750 F.
- (3) Hot-hardness tests performed on alloys containing from 16 to 45 w/o uranium indicate that a fabrication temperature above 750 F would be satisfactory for all compositions.
- (4) Press-forging tests indicated that the alloys in all ranges of composition studies would be amenable to extrusion.
- (5) The addition of up to 0.5 w/o silicon to the 16 w/o uranium alloy made no significant difference in the liquidus-solidus temperatures.

From the point of view of future applications of aluminum-uranium alloys as reactor fuels, the results of these studies are quite encouraging. The fact that the high-uranium alloys can be cast into sound homogeneous shapes as complex as an extrusion billet by a single-melting technique should remove a major production problem. It is recommended, however, that additional development work be done in order to obtain the maximum efficiency from the process. In addition, it is suggested that the effects of heat treatment on the strength and fabricability of these alloys be investigated, as should be the effects of ternary alloying additions.

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