Contract No. W-7405-eng-92

A STUDY OF THE LIQUIDUS IN
ALUMINUM-URANIUM ALLOYS

by

Victor W. Storhok
Arthur A. Bauer
Ronald F. Dickerson

May 1, 1958

BATTELLE MEMORIAL INSTITUTE
505 King Avenue
Columbus 1, Ohio

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

TABLE OF CONTENTS

	<u>Page</u>
ABSTRACT	1
INTRODUCTION	1
EXPERIMENTAL PROCEDURES	1
Alloy Preparation	2
Thermal-Analysis Techniques	2
Metallographic Studies	3
EXPERIMENTAL RESULTS	3
Thermal-Analysis Results	5
Metallographic Data	9
SUMMARY AND CONCLUSIONS	9
REFERENCES	10

A STUDY OF THE LIQUIDUS IN ALUMINUM-URANIUM ALLOYS

Victor W. Storhok, Arthur A. Bauer, and Ronald F. Dickerson

Liquidus temperatures in the aluminum-uranium system have been redetermined for alloys containing 13 to 50 w/o uranium. Thermal analysis, electrical-resistance measurements, and metallography were employed in the investigation.

The binary aluminum-uranium alloys studied and, their corresponding liquidus temperatures are as follows: 17 w/o uranium, 755 C; 17.5 w/o uranium, 780 C; 24.5 w/o uranium, 954 C; 30.9 w/o uranium, 1068 C; 41.6 w/o uranium, 1190 C; 51.2 w/o uranium, 1265 C. Eutectic and peritectic temperatures were determined to be at 640 and 732 C, respectively.

Binary alloys containing ternary additions of up to 1 w/o silicon and up to 0.5 w/o magnesium were studied also. Neither silicon nor magnesium were found to have any effect on the liquidus temperatures. However, silicon lowered the peritectic temperature from 732 C to 700 C and among alloys containing less than 30 w/o uranium had a refining effect on eutectic UAl_4 particles. The only effect that could be attributed to the magnesium additions was a tendency toward coarsening the UAl_4 particles in alloys containing less than 30 w/o uranium.

INTRODUCTION

Aluminum-uranium alloys have been of interest as low-temperature-reactor fuel alloys for some time. This interest has been extended to alloys with uranium contents as high as 50 w/o. One of the problems associated with developing sound castings of these higher uranium alloys is the tendency for segregation within the casting as a result of the marked differences in density of phases in the system. The constitution of the system has been reported by Gordon and Kaufmann⁽¹⁾. However, it was felt that a redetermination of liquidus temperatures was desirable because of their importance in casting these alloys.

EXPERIMENTAL PROCEDURES

Alloys studied ranged in composition from 9 to 50 w/o uranium, the alloys in the lower ranges being investigated in part to establish the eutectic composition and peritectic composition limit. Besides the binary alloys, ternary alloys with 0.5 and 1.0 w/o of silicon and 0.25 and 0.5 w/o of magnesium were studied to determine the effect of these elements on the system.

A variety of thermal-analysis techniques, including measurement of electrical resistance during heating, and metallography were employed in this investigation.

(1) References appear at end of report.

Alloy Preparation

All the alloys studied in this program were prepared by induction melting of biscuit uranium and high-purity aluminum. The alloys with uranium contents up through 30 w/o were cast into cold copper molds, while the 40 and 50 w/o uranium alloys were cast into graphite molds. Pouring temperatures were 50 to 100 C above the liquidus determined by Gordon and Kaufmann. Where the pouring temperatures were above 1230 C, the alloys were melted under a helium atmosphere to reduce vaporization of aluminum. The lower melting alloys were melted under a vacuum of 15 to 20 μ .

Silicon was added in the form of a master alloy containing 50 w/o silicon and uranium and aluminum in the proportions of the base binary alloy. Magnesium was added directly to the charge. Generally, chemical analysis showed the silicon content to be near that desired. However, some difficulty was experienced in retaining magnesium, particularly in the higher uranium alloys, as it apparently vaporized during melting.

Compositions as used throughout this report are those obtained by chemical analysis.

The castings appeared generally sound and homogeneous. With the exception of some swaged and annealed specimens used for electrical-resistance measurements, all alloys were studied in the as-cast condition.

Thermal-Analysis Techniques

Liquidus temperatures varied widely in the extensive compositional range investigated. Because of this, different thermal-analysis techniques were employed for specific composition ranges. However, where overlap in the range of application occurred, agreement was good among results obtained by different techniques. Data obtained during heating of specimens form the basis for the results reported.

For alloys close to the eutectic composition, such as the 16 w/o uranium alloy, electrical-resistance measurements obtained on heating yielded the best data. Attempts to detect the end of melting by straight thermal-analysis techniques were unsuccessful since the large heat effect associated with eutectic melting tended to obscure any additional effect at slightly higher temperatures. Specimens for electrical-resistance measurements were cold swaged to 100-mil-diameter rod after casting and were held in copper blocks to act as current leads for potentiometric measurement of resistance changes. Tantalum wires spot welded to the specimen served as voltage probes, and a 30-gage Chromel-Alumel thermocouple was spot welded to the specimen center. The specimen was coated with a ceramic material to act as a container during melting and was heated by inserting the specimen assembly into a hot furnace. Resistance changes and temperature changes were recorded as functions of time on high-speed recorders, and the separate records were then combined to yield a plot of temperature versus resistance.

Differential thermal analysis (graphite was the neutral specimen) at a heating rate of 3 C per minute yielded satisfactory results with alloys containing 20 to 30 w/o uranium. A record of temperature versus differential emf was obtained on an X-Y recorder. The specimens were contained by quartz tubes during melting.

A variation of this technique was employed also for alloys containing 20 to 30 w/o uranium. In the variation, differential emf and temperature were recorded simultaneously on a two-pen recorder. This method was more sensitive than that in which the X-Y recorder was used. By plotting temperature versus differential emf a derived differential curve was obtained. The advantage of such a curve was the more greatly emphasized transition points compared with those obtained by differential analysis. However, due to the greater sensitivity of measurements, a number of spurious effects were noted, necessitating a number of runs to separate the spurious effects from those associated with melting of the alloys.

Straight time-temperature thermal curves generally were found to yield satisfactory data for all alloy compositions above 20 w/o uranium. Heating curves were recorded on a Speedomax recorder, with the liquidus being indicated by a change in slope as the solid phases disappeared. For alloys containing 30 w/o uranium or less, heating rates comparable to those employed with differential-thermal-analysis techniques were used. Alloys containing 40 w/o uranium or more were heated in an induction furnace, as the expected liquidus temperatures were above the limit of the vacuum resistance furnace employed for the alloys containing less uranium. In addition, the higher temperatures of melting necessitated the substitution of platinum-platinum plus 10 w/o rhodium thermocouples for the Chromel-Alumel thermocouples used at the lower temperatures. The specimens employed were 3/8 in. in diameter and 1/4 in. long. A 1/16-in. hole was drilled in the specimen for insertion of the thermocouple, which was coated with a ceramic to prevent reaction between it and the alloy. The specimens were heated at a rate of 80 C per minute; however, heating rates were varied from 40 to 120 C per minute with no effect on the recorded liquidus temperatures being observed. Although the specimens were set in open graphite crucibles during melting, very little reaction between the alloy and surrounding air was observed.

Metallographic Studies

All alloys were prepared for metallographic examination in the as-cast condition. The lower uranium alloys, (9 to 19 w/o uranium) were examined to aid in determination of the eutectic composition and the composition limit of the peritectic horizontal. At the higher uranium contents, metallography was employed mainly to determine the effects of magnesium and silicon ternary additions on the microstructure.

Specimens cut from cast ingots were ground through 600-grit paper and polished first on a wheel covered with Forstmann's cloth impregnated with diamond paste and then polished on microcloth impregnated with Linde B abrasive. The specimens were etched with a 1/2 per cent HF solution.

EXPERIMENTAL RESULTS

On the basis of data obtained by the techniques described, the liquidus was determined for alloys in the range 13 to 50 w/o uranium as shown in Figure 1. For comparison, the liquidus determined by Gordon and Kaufmann⁽¹⁾ is also shown.

FIGURE 1. PARTIAL ALUMINUM-URANIUM CONSTITUTIONAL DIAGRAM

Thermal-Analysis Results

Thermal-analysis data are given in Table 1 and are shown plotted in Figure 1. The individual temperatures given can be considered accurate to ± 20 C if the maximum variation in liquidus temperature obtained at any single composition is used as a criteria.

TABLE 1. LIQUIDUS TEMPERATURES FOR ALUMINUM-URANIUM BINARY ALLOYS AND TERNARY ALLOYS CONTAINING SILICON OR MAGNESIUM

Alloy Composition (Balance Aluminum), w/o	Liquidus Temperature, C	Technique of Determination ^(a)
15.7 uranium	713	B
17 uranium	755	D
17.5 uranium	780	C
24.5 uranium	954	D
30.9 uranium	1068	C, D
41.6 uranium	1190	D
51.2 uranium	1265	D
20.0 uranium-0.5 silicon	854	A
21.3 uranium-1.17 silicon	880	A
27.4 uranium-0.54 silicon	1015	A
41.9 uranium-0.58 silicon	1186	D
39.0 uranium-1.07 silicon	1171	D
48.5 uranium-0.99 silicon	1248	D
20.0 uranium-0.5 magnesium	855	A
49.6 uranium-0.54 magnesium	1254	D

- (a) A. Differential Thermal Analysis.
 B. Resistance-Temperature Analysis.
 C. Derived - Differential Thermal Analysis.
 D. Time-Temperature Analysis.

Typical curves obtained by the various techniques of analysis are shown in Figure 2. The liquidus for the 15.7 w/o uranium alloy is clearly indicated by the almost constant final resistance. Although not fully understood, the rather sharp breaks below the liquidus are felt to be related to the end of eutectic melting. The differential-thermal-analysis and the derived-differential-thermal-analysis curves are nearly identical, although the transformation temperatures appear to be somewhat more emphatically indicated on the derived-differential curve.

In addition to the liquidus temperatures, the eutectic and peritectic temperatures were established at 640 and 732 C, respectively. The data also indicated that the composition limit of the peritectic horizontal is between 15 and 17 w/o uranium. While the 17 w/o uranium alloy showed a very slight evidence of the peritectic reaction, the

a. Change of Resistance With Temperature in Aluminum-15.7 w/o Uranium Alloy

b. Differential-Thermal-Analysis Curve for Aluminum-20 w/o Uranium - 0.25 w/o Magnesium Alloy

c. Derived-Differential-Thermal-Analysis Curve for Aluminum 30.9 w/o Uranium Alloy

d. Time-Temperature Thermal-Analysis Curve for Aluminum 41.6 w/o Uranium Alloy

FIGURE 2. REPRESENTATIVE CURVES OBTAINED BY VARIOUS THERMAL-ANALYSIS TECHNIQUES

250X

RM2934

a. Aluminum-13.0 w/o Uranium

500X

RM2295

b. Aluminum-14.3 w/o Uranium

FIGURE 3. CAST STRUCTURE OF ALUMINUM-URANIUM ALLOYS OF APPROXIMATE EUTECTIC COMPOSITION

250X

RM9028

a. Aluminum-51.2 w/o
Uranium

250X

RM9122

b. Aluminum-48.5 w/o
Uranium-0.99 w/o
Silicon

250X

RM9027

c. Aluminum-49.6 w/o
Uranium-0.54 w/o
Magnesium

FIGURE 4. STRUCTURES OF ALUMINUM-50 w/o URANIUM ALLOY AND WITHOUT ADDITIONS OF SILICON AND MAGNESIUM

15 w/o alloy showed none. By extrapolation of the liquidus curve, it would appear that the peritectic composition limit is 16.2 w/o uranium.

Neither silicon nor magnesium additions in the amounts investigated had any detectable effects on the liquidus. However, silicon did lower the peritectic temperature from 732 to 700 C. This might be expected, as a 3 w/o silicon addition to an aluminum-48 w/o uranium alloy is known to suppress the peritectic reaction completely⁽²⁾. There appeared to be little difference in this effect observed between 0.5 and 1.0 w/o additions of silicon. No effect on eutectic temperature was observed for either ternary addition.

Metallographic Data

Examination of alloys containing 9 to 19 w/o uranium indicated a eutectic composition of 13.2 ± 0.2 w/o uranium. This value is based upon the observation of massive, proeutectic UAl_4 (see Figure 3) in all alloys richer in uranium than this composition, while only a trace of proeutectic compound is found in the 13 w/o uranium alloy and then only in scattered regions, probably as a result of local inhomogeneities. The change in amount of UAl_4 with composition and the relatively large amount in the 14.3 w/o alloy also indicated that the eutectic was very close to the 13.2 w/o composition, the limits of accuracy being estimated on this basis.

Silicon additions in the ranges studied appeared to reduce the amount of eutectic and increase the amount of free aluminum in the 40 and 50 w/o uranium alloys, as shown in Figure 4. This effect was not noted in the lower uranium alloys, although there seemed to be a tendency toward refinement of eutectic UAl_4 particle size.

Except for what is probably an insignificant change in peritectic UAl_4 shape, magnesium additions had no effect on microstructure of the higher uranium alloys. Below 30 w/o uranium, there appeared to be some tendency toward coarser eutectic UAl_4 particles among alloys with magnesium additions.

SUMMARY AND CONCLUSIONS

The liquidus line of the aluminum-uranium system has been determined from 13 to 50 w/o uranium. The data produced indicate that the liquidus is higher than originally shown by Gordon and Kaufmann. A need for higher casting temperatures is therefore indicated.

Neither silicon additions up to 1 w/o or magnesium additions up to 0.5 w/o were found to produce any change in liquidus temperature. However, the silicon additions did lower the peritectic temperature from 732 to 700 C.

REFERENCES

- (1) Gordon, P. , and Kaufmann, A. R. , "The Alloy Systems Uranium-Aluminum and Uranium Iron", Journal of Metals, 188, 182-194 (January, 1950).
- (2) "Metallurgy Division Annual Progress Report for Period Ending October 10, 1957", ORNL-2422.

VWS:AAB:RFD /mar