

Beryllium-Uranium System¹

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The phase diagram of the system beryllium-uranium was constructed from data obtained by thermal analysis, metallographic examination and X-ray diffraction. The system is characterized by one intermetallic compound, UBe_{13} , melting at about $2,000^{\circ}\text{C}$, a eutectic having a composition of approximately 0.12 weight percent of beryllium, occurring between uranium and UBe_{13} , at $1,090^{\circ}\text{C}$, and a monotectic occurring at $1,550^{\circ}\text{C}$ and 60-weight-percent of beryllium. The solid solubility of beryllium in uranium is slight; no solid solubility of uranium in beryllium was detected. The transformation temperatures of uranium are not affected by additions of beryllium.

1. Introduction

The literature contains very little data on the beryllium-uranium system. This lack of information can be ascribed to the technical difficulties resulting from the high vapor pressure of beryllium and the large difference in the densities of the two components. A tentative phase diagram of the beryllium-uranium system was presented in the literature review by Buzzard and Cleaves.² The diagram shows one compound, approximating UBe_9 in composition and two eutectics, one at $1,060^{\circ}\text{C}$ between gamma uranium and UBe_9 , and the other at $1,200^{\circ}$ to $1,250^{\circ}\text{C}$ between beryllium and UBe_9 .

In order to have available a more accurate diagram of the beryllium-uranium system, work was initiated at the National Bureau of Standards at the request of the Atomic Energy Commission. A series of beryllium-uranium alloys was prepared with compositions ranging from 0- to 100-percent beryllium and analyzed metallographically, by X-ray diffraction, and by thermal analysis.

2. Preparation of Alloys

2.1. Stock materials, apparatus, and procedures

The stock materials consisting of extruded beryllium and uranium were obtained from the Atomic Energy Commission. The alloys were prepared in a specially constructed, high-frequency induction furnace, in which the charges could be melted and the ingots poured in an atmosphere of purified helium. This was accomplished by enclosing the beryllia crucible and mold assemblies in a silica tube and by using bottom-pour crucibles. Details of the construction are shown schematically in figure 1. It is important that the beryllia be fired at approximately $1,900^{\circ}\text{C}$ and be of high purity. The pouring temperatures were determined by sighting through the "sight" tube on the bottom of the stopper with an optical pyrometer. The stopper was essentially a closed end beryllia tube.

¹ Investigation sponsored by the Atomic Energy Commission at the National Bureau of Standards.

² R. W. Buzzard and H. E. Cleaves; Issue No. 1, p. 26-27, *Journal, Metallurgy and Ceramics* (TID 65), July 1948.

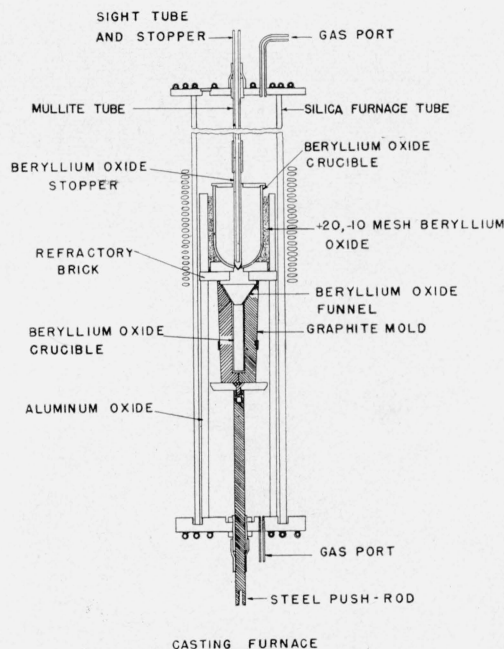


FIGURE 1. Controlled atmosphere, high frequency induction furnace.

2.2. Segregation

The density of the beryllium-uranium compound was considerably greater than the beryllium-rich liquid phase, thereby creating a tendency for the compound to collect at the bottom of the crucible or mold as it precipitated from the cooling liquid. This settling-out tendency was not so pronounced in the uranium-rich phase. It was possible to prepare beryllium-rich alloys reasonably free of segregation by pouring the ingots with the power on full into a cold mold of small cross section. The large supply of power, immediately before pouring, caused a vigorous stirring action, which kept the compound uniformly distributed.

The distribution of uranium in the ingots in the "as cast condition" was checked both by chemical analysis (table 1) and microradiographic examination (figs. 2 and 3). Chemical analysis of the ingots

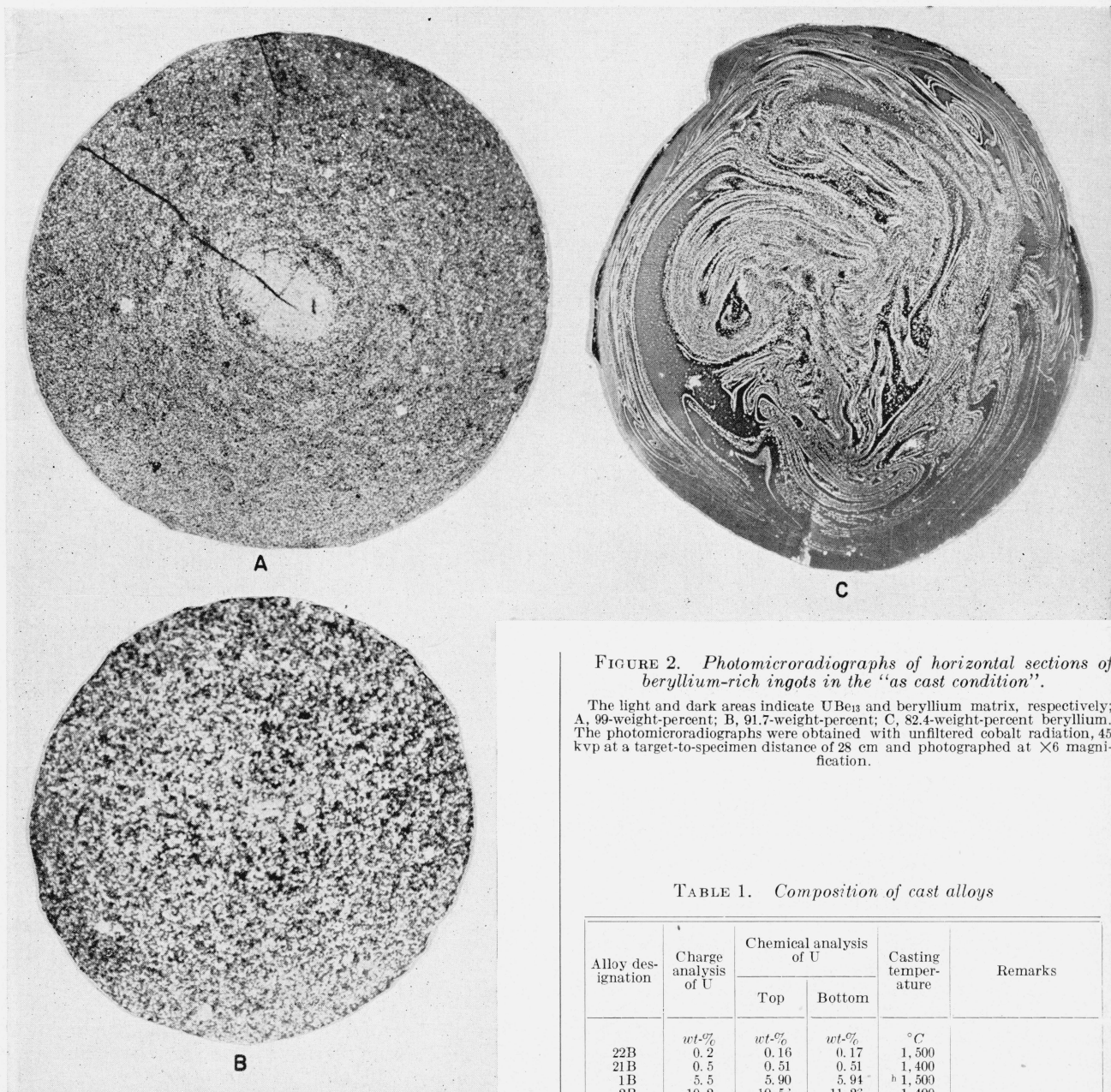


FIGURE 2. Photomicroradiographs of horizontal sections of beryllium-rich ingots in the "as cast condition". The light and dark areas indicate UBes and beryllium matrix, respectively; A, 99-weight-percent; B, 91.7-weight-percent; C, 82.4-weight-percent beryllium. The photomicroradiographs were obtained with unfiltered cobalt radiation, 45 kvp at a target-to-specimen distance of 28 cm and photographed at $\times 6$ magnification.

TABLE 1. Composition of cast alloys

Alloy designation	Charge analysis of U	Chemical analysis of U		Casting temperature	Remarks
		Top	Bottom		
	wt-%	wt-%	wt-%	$^{\circ}\text{C}$	
22B	0.2	0.16	0.17	1,500	
21B	0.5	0.51	0.51	1,400	
1B	5.5	5.90	5.94	^b 1,500	
2B	10.2	10.53	11.25	1,400	
3B	14.8	14.94	15.34	1,400	
16B	19.0	19.45	20.25	^b 1,425	
5B	30.0	30.50	30.17	^b 1,900	
6B	40.0	43.05	42.29	^b 1,850	
7B	50.0	44.02	49.68	^b 1,910	
8B	65.0	64.56	65.21	^b 2,020	
17B	73.5	73.06	73.86	^b 2,000	
9B	75.6	76.43	75.9	^b 1,900	Did not pour at 1,950 $^{\circ}$ C.
18B	77.0	77.54	76.70	^b 2,030	
15	85.0	85.22	85.32	^b 1,920	1,900 $^{\circ}$ C too low for good casting.
10B	95.0	^a 94.85 ^b 94.07	^a 94.57 ^b 93.70	^b 1,925	
11B	99.0	^a 98.72 ^b 98.70	^a 99.22 ^b 98.60	^b 1,650	
12B	99.8	^a 99.80 ^b 99.84	^a 99.63 ^b 99.82	^b 1,400	
14B	99.95	^a 99.85 ^b 99.91	^a 99.85 ^b 99.91	^b 1,400	
13B	99.6	^a 99.89 ^b 99.94	^a 99.85 ^b 99.93	^b 1,410	

^a Wet chemical analysis for U.

^b Graphite heater around crucible.

^b Spectrographic analysis of Be, U by difference.

showed satisfactory uniformity of composition from top to bottom, with the exception that the ingot in the 50-weight-percent range had a deviation of 5 percent over its length. The variation from top to bottom in the other ingots was within 1 percent, and the ingots having compositions in the high-uranium and high-beryllium fields showed little deviation in composition from top to bottom. The microradiographs showed good distribution of the compound in the beryllium-rich ingots, with the exception of the 99-weight-percent beryllium ingot. The swirl distribution of the compound shown in the photomicro-radiograph (fig. 2, A) is caused by the pouring action and is apparent in this ingot because of the low concentration of the compound.

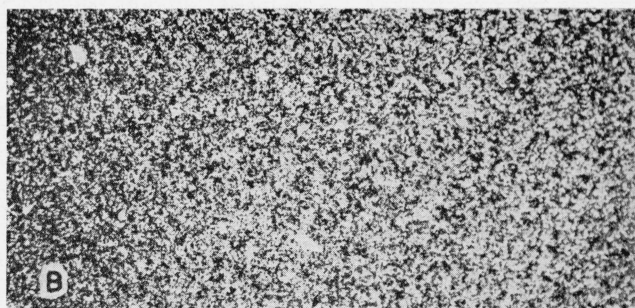
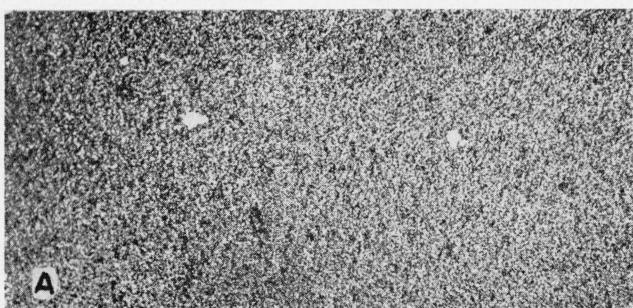


FIGURE 3. Photomicroradiographs of vertical sections of beryllium-rich ingots in the "as cast condition".

A, 82.4-weight-percent and B, 91.7-weight-percent beryllium $\times 6$ magnification.

3. Thermal-Analysis Apparatus and Procedure

The solid-phase arrests were determined by time-temperature analysis, using a modification of the apparatus described by Teitel and Cohen.³ The accessory apparatus included a Brown Air-o-line program controller to maintain uniform heating and cooling rates. A furnace atmosphere of purified argon was used in order to minimize the evaporation of beryllium. Arrests with this equipment could be determined with an accuracy of ± 2 deg C.

For regions involving a liquid phase, the high-frequency induction furnace used to prepare the cast alloys was modified for thermal analysis by substituting a tapered crucible for the bottom-pour-casting crucible. A beryllia plug was inserted in the stopper tube to a height such that temperature observations were made at the center of the melt, using a high precision optical pyrometer. The cooling rates were controlled by manually reducing the power of the convertor at a uniform rate in a manner that cooling rates of the order 0.5 to 2.0 deg/min were obtained through the thermal arrests. The solidus and solid-phase arrests of the beryllium-rich alloys were determined in this furnace, using platinum-rhodium thermocouples enclosed in high-fired-alumina protection tubes and inserted through the sight tubes. The tip of the couple was seated on the top of the beryllia plug. The convertor control was modified so that the power could be reduced mechanically with an electric motor operating through a gear box. The time-temperature curves were recorded on a Brown electronic recorder.

4. Results

4.1. Beryllium-uranium compound

Metallographic examination showed all of the cast alloys (table 1) to consist of two phases. One phase increased in amount from either the uranium or beryllium end of the system until at approximately 35-weight-percent beryllium, the alloy appeared to be composed almost entirely of this phase (fig. 4, E, F). Alloys in this range were hard and very brittle.

It was found that this phase could be readily isolated from a uranium matrix by electrolytically dissolving the uranium in a 10-percent chromic-acid solution. The separated phase was identified as the intermetallic compound UBe_{13} having a cubic structure, as reported by Baenziger and Rundle.⁴ Precision determination of the lattice constant gave a value of $a = 2 \times 5.1849 \pm 0.0001$ Å at 27° C.

In order to check the stoichiometric formula, UBe_{13} , a density determination and chemical analysis were made. The values obtained were 4.420 ± 0.002 g/cm³ at 26° C and 32.15 ± 0.05 -weight-percent beryllium as compared to the computed values of 4.373 g/cm³ and 33.0-weight-percent beryllium. The actual and computed values are in sufficient agreement to conclude that the formula is UBe_{13} and not UBe_9 as previously reported. The thermal analysis results indicate that the melting point of UBe_{13} was approximately 2,000° C.

4.2. Beryllium-rich alloys

Data (table 2) from the time-temperature cooling curves obtained for the beryllium-rich alloys show a monotectic at 60-weight-percent beryllium and 1,550° C (fig. 5). The liquidus and liquid solubility temperatures above 1,600° C were influenced by the vapor phase of the beryllium, and the accuracy is not greater than ± 25 deg C. The maximum temperature indicated for the UBe_{13} composition is a selected value based on an evaluation of the thermal data, casting temperatures, and extrapolation of the liquidus curve. The solidus ($1,280^\circ \text{C} \pm 3^\circ$) coincided with the melting point of beryllium at $1,283^\circ \text{C}$. A pronounced arrest occurred below the solidus at $1,250^\circ \text{C} \pm 10^\circ$, which can be ascribed either to impurities or an allotropic transformation in the beryllium phase, although the strong arrests observed at this point indicate a possibility of the latter. Previous investigators^{2, 5, 6, 7} have observed similar double points in the melting-point determinations of beryllium and have concluded impurities to be the cause, but lacking conclusive proof as to the cause of the observed double arrest, the line is dotted on the diagram.

⁴ N. C. Baenziger and R. E. Rundle, United States Atomic Energy Commission, AEC-D-2506 (April 8, 1949).

⁵ G. Oosterheld, Z. anorg. u. allgem. Chem. **97** 1-6 (1916).

⁶ L. Losano, Alluminio **8** 67 (1939).

⁷ H. A. Sloman, J. Inst. Metals **49** 365 (1932).

³ R. J. Teitel and [Morris] Cohen, Trans. Am. Inst. Mining Met. Engrs. **155** 285-296 (1949).

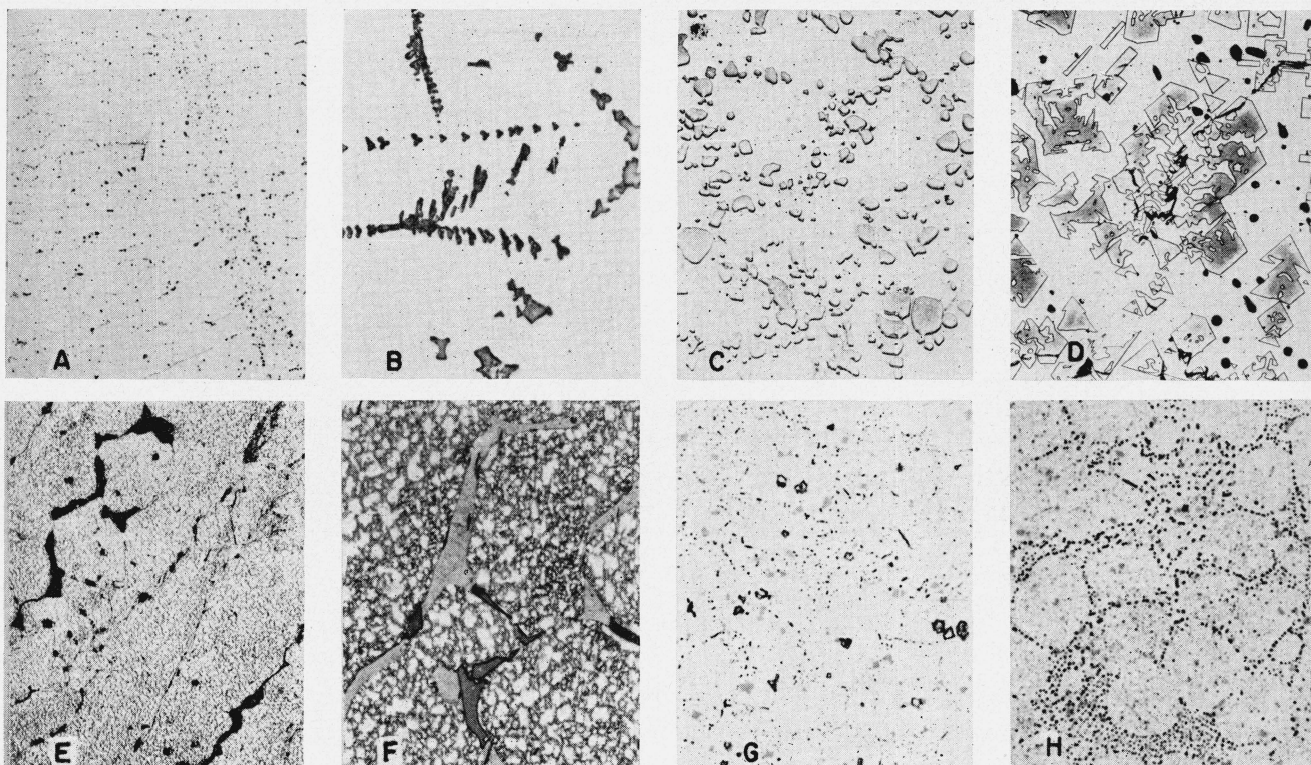


FIGURE 4. Typical microstructures of beryllium-uranium alloys in the "as cast condition".

Etchant 5 percent HF, A, B, C, D, E, G, and H at $\times 100$, F at $\times 500$. A, 99.49-weight-percent beryllium; B, 98.86-weight-percent beryllium; C, 80.15-weight-percent beryllium; D, 69.7-weight-percent beryllium, dark constituent, UBe_{13} , in light matrix of beryllium; E and F, 35.1-weight-percent beryllium, dark areas at low magnification are shown at high magnification to be beryllium (pale grey) and the mottled background is UBe_{13} ; G, 0.17-weight-percent beryllium, small angular areas, sharply outlined, are UBe_{13} particles; H, 0.09-weight-percent beryllium. The uranium- UBe_{13} eutectic is shown in a uranium matrix.

TABLE 2. Summary of thermal data obtained for beryllium-rich alloys

Be	Thermal arrests ^a				
	Miscibility of L_1+L_2	Liquidus	$L_1+\delta \rightarrow L_2+\delta$	Solidus	$\epsilon+\delta \rightarrow \eta+\delta$
wt-%	$^{\circ}C$	$^{\circ}C$	$^{\circ}C$	$^{\circ}C$	$^{\circ}C$
100	-----	$\begin{cases} b \ 1,282 \\ 1,283 \\ b \ 1,283 \\ 1,286 \\ c \ 1,284 \end{cases}$	-----	-----	$b \ 1,263$
99.5	-----	-----	-----	-----	$b \ 1,264$
99.0	-----	-----	-----	-----	$c \ 1,233$
98.5	-----	1,289	-----	-----	-----
98.0	-----	1,286	-----	1,282	1,223
96.5	-----	1,294	-----	1,283	1,234
95.0	-----	1,289	-----	$\begin{cases} c \ 1,281 \\ 1,283 \end{cases}$	$c \ 1,241$
93.5	-----	1,285	-----	-----	1,228
92.5	-----	1,299	-----	1,282	-----
91.0	-----	1,297	-----	1,271	-----
90.0	-----	1,303	-----	$\begin{cases} c \ 1,280 \\ 1,284 \end{cases}$	$c \ 1,238$
87.5	-----	1,399	-----	-----	1,239
85.0	-----	1,399	-----	$c \ 1,286$	$c \ 1,240$
82.5	-----	1,548	-----	1,280	-----
80.0	1,588	1,548	-----	-----	-----
77.5	1,682	1,551	-----	-----	-----
75.0	1,832	1,550	-----	-----	-----
72.5	1,808	1,550	-----	-----	$c \ 1,239$
70.0	1,874	1,562	-----	-----	-----
67.5	1,808	-----	-----	-----	-----
65.0	1,748	1,550	-----	-----	-----
64.0	-----	-----	-----	$b \ 1,284$	$b \ 1,267$
61.0	1,642	-----	1,547	-----	-----
60.0	1,551	-----	1,550	$c \ 1,282$	$c \ 1,253$
57.5	-----	1,757	1,548	-----	-----
55.0	-----	1,820	1,556	-----	-----
50.0	-----	1,885	1,554	-----	-----
45.0	-----	1,882	1,552	$c \ 1,270$	$c \ 1,237$
40.0	-----	1,843	-----	-----	-----
35.0	-----	1,858	-----	-----	-----

^a All points not otherwise indicated were determined in high-frequency induction furnace, using an optical pyrometer.

^b Points determined in resistance furnace.

^c Points determined in high-frequency furnace with thermocouple.

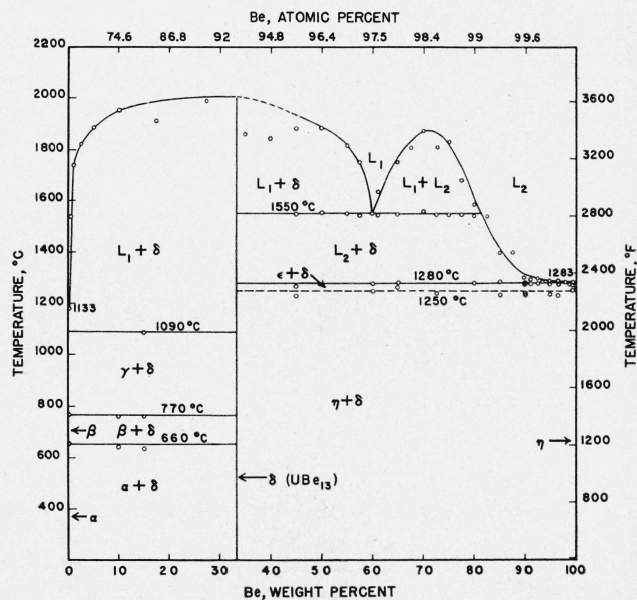


FIGURE 5. Beryllium-uranium phase diagram.

Metallographic examination showed that the cast structures of the alloys in the composition range 35- to 100-weight-percent beryllium consisted of particles of UBe_{13} embedded in a beryllium matrix. Homogenizing treatments up to $1,225^{\circ}\text{C}$ produced no change in the structures. The compound appeared in an angular form (figs. 4, A, B, D) over the entire composition range, with the exception of a narrow range between 92- to 80-weight-percent beryllium, in which the compound was observed to have a globular form (fig. 4, C). All evidence indicated the solubility of uranium in beryllium was very limited and was not detected in this investigation.

4.3. Uranium-rich alloys

The cooling curves (table 3) obtained for the uranium-rich alloys indicate a eutectic at approximately 0.12-weight-percent beryllium and $1,090^{\circ}\text{C}$. Arrests were obtained at 770° and 660°C that correspond to the two allotropic transformation temperatures, indicating little or no solid solution of beryllium in alpha or beta uranium (fig. 6).

TABLE 3. Summary of thermal data obtained for the uranium-rich alloys

Be	Thermal arrests ^a			
	Liquidus	Solidus	$\gamma-\beta$	$\beta\rightarrow\alpha$
wt-%	$^{\circ}\text{C}$	$^{\circ}\text{C}$	$^{\circ}\text{C}$	$^{\circ}\text{C}$
0.000 ^b	b 1,134	-----	b 768	b 657
.027	b 1,128	b 1,116	772	b 660
.043	b 1,117	b 1,108	b 770	b 657
.079	b 1,100	b 1,090	b 770	b 658
.0955	1,095	-----	-----	-----
.107	b 1,119	b 1,085	b 766	b 652
.113	b 1,109	b 1,086	b 768	b 655
.15	-----	-----	b 769	b 655
.177	-----	b 1,091	b 766	b 654
.2	1,183	-----	-----	-----
.5	1,543	-----	-----	-----
1.0	1,743	-----	-----	-----
2.5	1,827	-----	-----	-----
5.0	1,887	-----	-----	-----
10.0	1,952	-----	b 765	-----
15.0	1,910	b 1,090	b 761	-----
20.0	1,910	-----	-----	-----
27.5	1,993	-----	-----	-----

^a All points not otherwise indicated were determined in high-frequency induction furnace, using an optical pyrometer.

^b Points determined in resistance furnace.

Metallographic examination showed that alloys in the vicinity of the uranium- UBe_{13} eutectic, as indicated by thermal analysis, contained a definite eutectic structure (figs. 4, G, H). Homogenization near the solidus spheroidized the UBe_{13} . X-ray analysis of alloys quenched from $1,075^{\circ}\text{C}$ in the range 0.027 to 0.079-weight-percent beryllium, placed the solid solubility of beryllium in gamma uranium between 0.042 to 0.079-weight-percent at $1,075^{\circ}\text{C}$.

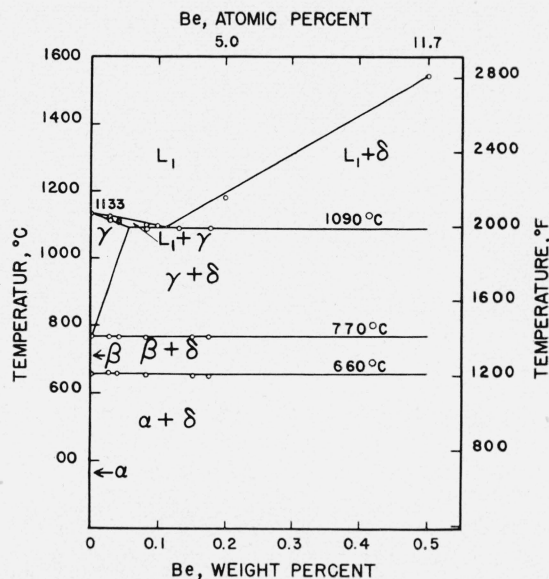


FIGURE 6. Uranium-rich section of beryllium-uranium phase diagram.

5. Summary

The phase diagram of beryllium and uranium was constructed from data obtained by thermal analysis, metallographic examination, and X-ray diffraction (figs. 5 and 6). The beryllium-uranium system is characterized by one intermetallic compound, UBe_{13} , having a melting point of approximately $2,000^{\circ}\text{C}$. A eutectic, having a composition of approximately 0.12-weight-percent beryllium, occurs between uranium and UBe_{13} at $1,090^{\circ}\text{C}$. A monotectic occurs at $1,550^{\circ}\text{C}$ and 60-weight-percent beryllium.

A slight solubility of beryllium in gamma-uranium is indicated, but no solubility of uranium in beryllium was detected. The transformation temperatures of uranium were unaffected by additions of beryllium.

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