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LUCAS HEIGHTS

PREPARATION OF THE BERYLLIDES OF URANIUM AND THORIUM

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ABSTRACT

Uranium and thorium beryllides and their solid solutions were prepared by reaction-sintering, reaction-pressing and arc-melting of mixed metal powders. The major difficulties of preparation were the large swellings accompanying reaction without pressure and the loss of beryllium at temperatures above 1200 - 1300 °C. The reaction-pressing method suffered least from these difficulties and yielded the purest and most dense products.

CONTENTS

1. INTRODUCTION

One of the fuel systems proposed for a high–temperature gas–cooled reactor is a dispersion of a mixed uranium – thorium beryllide in a matrix of beryllium (Alder 1961). Both uranium and thorium form beryllides of the general formula MBe_{13}, and the intermetallic compound formed in the ternary system uranium – thorium – beryllium is a solid solution of the binary compounds UBe_{13} and $ThBe_{13}$; this may be conveniently denoted by the formula $(U,Th)Be_{13}$.

Fuel systems have been proposed in which the uranium to thorium ratio is between 1 : 30 and 1 : 50. For irradiation testing however, this ratio must be considerably higher in order to accelerate the rate of fission–product damage in the beryllide particles and the matrix immediatel surrounding them. Interest therefore lies in the preparation of beryllide solid solutions covering th full range of composition.

This report describes work done to find methods for producing uranium and thorium beryllid and their solid solutions in large homogeneous dense pieces which would be suitable for property evaluation and, after crushing to powder, for use in fuel element specimens. The main methods examined were reaction–sintering (that is, simultaneous reaction and sintering) and reaction–pres (that is, simultaneous reaction and hot–pressing); however some preliminary experiments were als made with the arc–melting technique. Metal powders were used as the starting materials in each c and chemical reduction methods were not considered.

2. PREVIOUS WORK

Several workers have attempted to prepare beryllides in the past but from the published acc it is difficult to evaluate the quality of the products, particularly with regard to true composition a density.

Reaction–sintering at temperatures between 1000 °C and 1250 °C has been used to produce UBe_{13} but it is clearly difficult to produce a dense product in this way. Angier and Hausner (1948 found that compacts of uranium and beryllium powders expanded by about 36 per cent. in volume wh reacted at 1235 °C in vacuum. Crushing, compacting, and re–sintering the products resulted in fur but slight increases in volume. Doll (1949) observed similar expansion when sintering under hydro at 1100 °C and attempted to overcome it by reacting at 400 °C and 1050 °C under loads of 80 p.s.i.; although densities were improved, the compacts swelled during subsequent sintering treatments. Williams and Jones (1956) found swelling to be a maximum in stoichiometric mixtures of uranium ar beryllium and attributed the increase in volume to diffusional porosity resulting from a Kirkendal Effect associated with the relatively rapid rate of diffusion of the beryllium atom.

Induction melting was used by Angier and Hausner (1948) and Heumann and Cashin (1958) t react uranium and beryllium. Angier and Hausner observed "flashing" at 1300 °C and obtained butt which fractured easily but were more difficult to crush than their sintered product. Heumann and C found that reaction proceeded rapidly at temperatures as low as 1150 °C but did not describe the qu of the product. Chubb and Dickerson (1959) prepared the beryllides of zirconium, yttrium, and niob by melting metal powder compacts in a helium arc furnace but sound buttons could be obtained only after repeated melting; this produced considerable volatilisation of beryllium.

Pemsler (1960) employed hot pressing at 1500 °C of pre–formed $ZrBe_{13}$ and UBe_{13} powders and achieved densities of 95 and 76 per cent. of theoretical respectively. Stonehouse and co–work (1959) used the same technique on a number of transition metal beryllides and claimed densities of 97 to 100 per cent. of theoretical.

Runnals (1959) prepared the beryllides of several metals by reduction of their fluorides with beryllium metal. The nature of the product was not described but it is likely that such a method wo produce finely divided material unless very high temperatures were achieved. Bomb conditions wou probably be necessary to prevent excessive evaporation of beryllium.

In view of the lack of detail in all reports mentioned above, the densities claimed must be accepted with caution. Significant loss of beryllium must be expected during reaction or consolida and if free uranium is present in the product, bulk densities close to, or greater than, theoretical density can be obtained even when porosity is present.

3. MATERIALS USED IN EXPERIMENTAL WORK

The materials used in the experiments were as follows:

Beryllium: French electrolytic powder screened to minus 100 mesh B.S.S. The nominal conten of the major impurities were oxygen 5000 p.p.m., aluminium 1000 p.p.m., silicon 100 p.p iron 750 p.p.m., and halogens 300 p.p.m.

Uranium: Calcium reduced powder, screened to minus 300 mesh B.S.S. The nominal contents o the major impurities were iron 65 p.p.m., silicon 30 p.p.m., aluminium 130 p.p.m., and nitrogen 200 p.p.m.

Thorium: Calcium reduced powder, screened to minus 300 mesh B.S.S. The nominal contents o the major impurities were aluminium 800 p.p.m., iron 350 p.p.m., and silicon 120 p.p.m.

Reaction mixtures were prepared by carefully weighing the powders under an argon atmo and mixing for one hour, either on mixing rolls or in a flask shaker.

4. REACTION – SINTERING

4.1 Experimental Procedure

Two series of stoichiometric mixtures of metal powders were compacted to 80 per cent. theoretical density under a pressure of 50 t.s.i. The compacts were reacted at the desired temp atures for three hours under a vacuum of 10^{-3} mm Hg.

The uranium to thorium ratio in the first series of compacts was varied in nine stages fr pure UBe_{13} to pure $ThBe_{13}$. Compacts were reacted in quadruplicate at 1200 °C.

The uranium to thorium ratio in the second series of compacts was kept constant at 1:2 they were reacted in quadruplicate at nominal temperatures of 1000, 1100, 1200, 1300, and 1400

In all cases the dimensions of the compacts were measured before and after reaction an volume changes calculated. The compacts were then crushed and the powders examined metall ically.

4.2 Results and Discussion

The behaviour of reaction–sintered compacts is summarised in Tables 1 and 2. Large increases in volume occurred in all cases. A mean swelling of 32 per cent. occurred in U–Be compacts reacted at 1200 °C and the swelling was greater in those of high thorium content; equ mixtures swelled by 90 per cent. and thorium rich mixtures by more than 100 per cent. The resu indicate that the swelling reached a maximum at a uranium to thorium ratio of about 1 : 4 but no experiments were done to confirm this.

Compacts of constant composition (U:Th = 1 : 2) reacted in the range 1000 to 1400 °C a increased in volume at least at temperatures up to 1300 °C. The swelling decreased, with incre ing temperature, from 290 per cent. at 1000 °C to 42 per cent. at 1300 °C; at 1400 °C, the highes temperature used, the compacts actually shrunk. Metallographic examination of the compacts reacted at 1400 °C revealed large amounts of free uranium, and chemical analysis showed that t beryllium content had decreased to a mean value of 15.8 per cent., that is, $(U,Th)Be_{4.9}$. Beryll contents determined by chemical analysis of the compacts reacted at the lower temperatures we unaccountably higher than the original contents; however the results did indicate that beryllium losses increased as the reaction temperature increased. Thus, it is not clear whether sintering loss of beryllium is the more important factor contributing to the decrease in swelling with incr ing temperature.

The results of these experiments, although limited, clearly show that significant losses beryllium occurred at the higher temperatures and that marked swelling occurred at low tempera where volatilisation was less. Thus, reaction–sintering is not a practical method of producing $(U,Th)Be_{13}$ and no further work with this technique is contemplated.

The reasoning of Williams and Jones (1957) that swelling of uranium and beryllium mixtures is attributable to diffusional porosity associated with a Kirkendal Effect can also be applied to the thorium–beryllium system and to ternary systems. A comparison of atomic radii (which can be use to assess roughly the relative diffusion rates, the rates being inversely proportional to the radii,) supports this suggestion and indicates that the effect should be more pronounced in thorium–beryll mixtures than in uranium–beryllium mixtures. This agrees with the results described above but th extent of the swellings encountered in these experiments was so great that the Kirkendal Effect do not seem to be the only factor involved.

It is difficult to envisage the actual changes which occur within a compact if the swelling solely due to the Kirkendal Effect. It may be supposed, perhaps, that all the beryllium diffuses ou of the space it originally occupied and that this space then appears as voids within the product. The volume of the reacted compact would then be the sum of the volumes of the reaction products, the diffusional porosity, and the original porosity. If this were the case the calculated swelling accompanying reaction in a fully dense powder compact would be 94 per cent. for uranium–berylliu mixtures and 74 per cent. for thorium–beryllium mixtures. If the compact was of less than theoreti density, volume changes expressed as percentages of the original volume would be proportionately less. The only way in which the swelling can become greater than these postulated values is for existing pores to swell as reaction proceeds. This could conceivably occur if partially reacted particles, for example uranium surrounded by shells of UBe_{13}, were in contact and pushed each other apart as they, themselves, increased in volume.

It seems doubtful that such a picture would be entirely true and it is probable that some swelling would occur into part of the volume originally occupied by the beryllium. The above consideration also neglects sintering which would be expected to occur to some extent in the temperat ranges used.

A further factor which must contribute to the swelling is radial fracture of beryllide shells surrounding partially reacted particles of uranium or thorium. As beryllium diffuses across these shells the underlying heavy metal will transform to the beryllide with an increase in volume. This will place the shells in tension and eventually cause fracture. The fissures so formed will widen as reaction proceeds and lead to a significant increase in porosity. Unfortunately, this contributio is difficult to evaluate.

The total swelling due to the Kirkendal Effect may conceivably approach 100 per cent. However, the swellings of nearly 300 per cent. encountered at 1000 °C suggest that another unknow factor was operative. If this is true, the magnitude of the effect suggests that it will also operate other temperatures.

5. REACTION–PRESSING

The term reaction–pressing is used to describe the simultaneous reaction and consolidation under pressure of stoichiometric mixtures of the constituent metal powders.

5.1 Experimental Procedure

5.1.1 Fabrication

The reaction–pressing assembly is illustrated in Figure 1. Mixtures were pressed in an induction heated graphite die at temperatures up to 1650 °C and a pressure of 1 t.s.i. By containin the die inside a graphite pot it was possible to maintain a protective atmosphere around the die during reaction. The lid of the graphite pot had a clearance hole for the plunger and supported inle and outlet tubes for the protective gas. The graphite pot was placed on refractory bricks and surrounded with alumina–grains for thermal insulation. The alumina grain was contained within a silica tube about which the induction coil was placed.

To prevent contamination of the reaction mass with graphite from the die, those parts of the die which contacted the reaction mass were coated with a beryllium oxide crucible wash. To simpl the application of this wash the die was fitted with a tight fitting liner, half as high as the die itsel This was readily coated with the wash and filled with reactants before insertion into the die body.

The die was loaded under argon, evacuated, and flushed with argon to remove macroscopic pockets of air, and then set up for reaction–pressing. The temperature was raised as rapidly as possible (about 30 minutes to 1550 °C) and light pressure was applied at about 800 °C and increas to the full load of 1 t.s.i. at about 1000° C. Full pressure was maintained until the end of the hea cycle when both pressure and heating power were removed together. When cool, the compact was removed from the die, cleaned and examined. During a run the temperature was measured with an optical pyrometer, sighting onto the die through the argon outlet tube.

For convenience of reference, runs were divided into three series. Series I (runs HP 2 to HP 20) consisted of 30 gram lots pressed in the temperature range 1300 °C to 1650 °C. At the tim these were done, there was little time to find a method capable of producing beryllides of the com positions needed for irradiation test specimens. Consequently, many of the reaction mixtures hac uranium : thorium ratios in the range 3 : 1 to 1 : 2 (atomic) and it was hoped that variations withi these limits would not greatly affect the compacting characteristics. Details of the reaction mixt are given in Table 3.

The densities of compacts were determined by displacement weighings and their microstru were examined. Where compacts were not recovered whole, densities of the larger fragments were determined. Several compacts were crushed to powder and submitted for chemical analysis.

In the Series II compacts (runs HP 23 to 27), a nine per cent. excess of beryllium was use reaction mixtures of uranium and beryllium. The optimum temperature was selected from previous as 1550 °C and this temperature was held for 1, 10, 20, 40, and 60 minutes. Compacts were weig after removal from the die and the weight loss during reaction was estimated. Samples of each co were examined metallographically for porosity and free uranium, and others were submitted for ch analysis for uranium and beryllium.

Series III consisted of two runs, one to prepare UBe_{13} (Run HP 21) and one to prepare Th (Run HP 22), in which a two inch diameter die and reaction lots of over 300 grams were used. To accommodate the larger die in the pressing assembly it was necessary to dispense with the argon atmosphere and its associated chamber. However, powder mixing and die loading were done unde argon and the loaded die was evacuated and flushed with argon before being placed in the reactio assembly. Light pressure (about 500 p.s.i.) was applied at 1000 °C and was gradually increased 1 t.s.i. as the temperature increased to about 1300 °C. In both cases the die fractured when the f load was applied and the runs were terminated and the assembly allowed to cool. Samples from t recovered compacts were taken for density determination and metallographic examination.

5.1.2 Homogeneity of solid solutions

Two methods were used to estimate the degree of solid solution in beryllides containing uranium and thorium. Micropolarography was used to determine the uranium to thorium ratios of individual particles of a size approaching that of the proposed fuel particles (150 μ diameter), ar X-ray diffraction was used on powder samples weighing 100 to 200 milligrams.

Material from reaction-pressing run HP 8 was crushed and screened to obtain particles ir ranges 200 - 250 microns and 250 - 300 microns. Six particles from each range were picked out each analysed separately by micropolarography for its uranium and thorium content.

X-ray diffraction patterns were obtained from several powder samples using a Philips diffractometer and copper Kα radiation. Particular attention was given to the position of high an reflections, for example 10 4 0 to 10 8 2 where the Bragg angles from UBe_{13} and $ThBe_{13}$ differ b 1 to 2½ degrees (that is, 2θ differs by 2 to 5 degrees). Complete solid solution was indicated by absence of reflections from the two pure compounds, and the lack of perfection of solution was indicated by the broadening of the reflections from the solid solution.

In Table 7 the Bragg angles for the 10 8 0 reflection of the solid solution obtained from I HP 8 are compared with those of UBe_{13} and $ThBe_{13}$ prepared by the same method. The uranium thorium ratio of the solid solution was determined by interpolation assuming that a linear relation existed between lattice parameter and molar composition.

5.2 Results and Discussion

Details of reaction-pressing runs are given in Table 3. Compacts showed a marked tend to fracture during cooling. Fracture faces were generally blue to purple in colour, indicating tha fracture occurred while the compact was still hot and not during recovery from the die. The tend to fracture was noticeably reduced by cooling without pressure on the compact.

No reaction of the beryllium oxide die wash with the graphite die was encountered and in cases the oxide came away with the compact. At 1600 °C the reaction mass penetrated the oxide appreciably and at 1650 °C this was severe enough to make removal of the oxide skin very difficu Generally, the oxide was easily removed from compacts reacted at 1550 °C.

There was a wide scatter – from 75 to 110 per cent. of theoretical – in the densities of Series I compacts. There was no consistent correlation between density and reaction temperature but of the five compacts prepared below 1550 °C the densities of three were below 90 per cent. of theoretical whereas most of the compacts prepared at 1550 °C or above were greater than 90 per c of theoretical density. This, and the observation that reaction with the die wash became troubles at 1600 °C, indicated that the optimum temperature was about 1550 °C.

The preparation of uranium beryllide in the Series II runs (HP 23 to 27) involved losses of weight which increased with time at the maximum temperature of 1550 °C. Longer time at tempera also caused a decrease in the weight ratio of beryllium to uranium from 0.54 to 0.51 indicating tha even the compact reacted for 60 minutes had not lost all its excess beryllium. The densities of th compacts from these runs were all 98 per cent. or more of the theoretical densities calculated from chemical compositions.

The densities of the two Series III compacts (runs HP 21 and 22), where the maximum obse temperatures were as low as 1300 °C and 1350 °C respectively, were 4.05 and 3.7 g cm^{-3}. The mi structures (see Figures 3 and 4) showed little porosity which, in view of the apparent densities of per cent., suggests that other regions of the compacts may have been more porous. However, thes densities are high for an observed reaction temperature of 1300 °C. This could be due to the fact the temperature developed within the reaction mass was considerably higher than the optically me ured temperatures. The specific surface area of the two-inch diameter compacts is about one qua of that of the one-inch compacts so that the heat of reaction would be more readily retained and w lead to a larger temperature rise.

Metallographic examination showed that even high density compacts generally contained p (see Figures 2 to 6) and, sometimes, small amounts of free heavy metal at grain boundaries. The metal in solid solution beryllides always appeared to be uranium, suggesting that the free energy formation of $ThBe_{13}$ is greater than that of UBe_{13}. This supports the observation of Farrell (196 that $ThBe_{13}$ is more resistant to chemical attack than UBe_{13}.

The microstructures of individual Series II compacts were similar and showed limited poro and some oxide inclusions. None contained free uranium or beryllium. The cleanest microstructu were exhibited by the compacts reacted for one and ten minutes (Figures 5 and 6) and these compa had densities of 98 and 97 per cent. of theoretical respectively.

The microstructures or uranium beryllide compacts always showed dark, randomly disperse rings, such as those illustrated in Figure 3. It is thought that these were rings of BeO particles derived from oxide layers on the surface of the uranium particles used as the reactant.

The composition of Series I compacts determined by chemical analysis (see Table 4) usual varied considerably from that of the reaction mixtures. The uranium content varied randomly from t intended value and the thorium and beryllium contents generally appeared to be lowered during rea It is unlikely that true changes of uranium and thorium content of the size indicated in Table 4 wo occur during reaction-pressing. The apparent changes were most probably due to inhomogeneity a inadequate sampling procedure. As all samples contained some insoluble impurities the beryllium contents, though generally lower than the stoichiometric value, do not necessarily reflect loss of beryllium by evaporation alone, but may also indicate that oxidation occurred. When compositions compared on a basis of weight ratio rather than weight per cent. of total compact (that is, insolubl impurities are neglected) the changes in thorium content assume a random variation similar to that of the uranium figures, that is, some increase on reaction whereas others decrease.

The uranium : thorium ratios of compact HP 8 determined by micropolarography (Table 6) we in the range 0.5 to 0.9. The mean of the twelve values was 0.71 which represents a uranium conte of 28.2 weight per cent. if the compound is assumed to be stoichiometric. The ratio determined by X-ray analysis (see Table 7) was 0.60 corresponding to a uranium content of 25.5 weight per cent. which agrees well with the value of 26.0 per cent. for the ''as-weighed'' reaction mixture.

It is worth noting that line broadening was very slight in the diffraction pattern from comp HP 8. Patterns from other compacts have shown broader reflections but in all cases the pattern been free from reflections from the binary compounds.

The micropolarographic method is capable of an accuracy of ± 2 per cent. but the sample be weighed to this accuracy unless it weighs at least 100 micrograms (that is, 350 microns diam However, accurate uranium : thorium ratios can be determined using particles weighing as little 30 micrograms (that is, 200 to 250 microns diameter) and such determinations are useful in indic the point-to-point variations in inhomogeneity within a given sample. The X-ray technique, ho is much more rapid and convenient, particularly as the modern diffractometer permits the selectio only those reflections of interest.

6. ARC-MELTING

6.1 Experimental Procedure

An exploratory examination was made of the preparation of uranium beryllide by arc melti an Edwards tungsten electrode argon-arc furnace. The following specimens were melted:

(i) Cold compacted UBe_{13} pre-formed by reaction sintering at 1200 °C.

(ii) The button obtained from (i).

(iii) The button obtained from (ii).

(iv) Cold compacted UBe_{13} as used in (i), the button being turned half way through the melting cycle.

(v) A cold compacted, stoichiometric mixture of uranium and beryllium metal powders, the button being turned half way through the cycle.

In all cases the melting time was kept as short as possible, the arc being quenched as s the specimens appeared to be fully molten. All the ingots were weighed before and after melting the densities of the buttons were determined by weighing in air and in water. After sectioning, t buttons were examined metallographically.

6.2 Results and Discussion

The behaviour of compacts subjected to arc melting is detailed in Table 8. The outstan feature of the experiments was the loss of weight which accompanied all the meltings, the losse ranging from 4.3 to 13 per cent. The bulk densities of all buttons were higher than the theoretic density of UBe_{13} and metallography revealed considerable porosity in them all. Free uranium w also visible in the microstructures and this would account for the high bulk densities even when porosity is not taken into account.

7. CONCLUSIONS AND RECOMMENDATIONS

Reaction-pressing has proved to be the most suitable method for preparing beryllide com from the constituent metal powders as it suffers least from swelling and volatilisation problems iated with reaction at high temperatures. It appears that good compacts can be obtained by reac pressing at 1550 °C (or below) and 1 t.s.i. without maintaining the maximum temperature for more one minute. On a laboratory scale however, arc-melting may possibly be developed to yield a p product if beryllium volatilisation can be tolerated and safely controlled. Care would be necess however, in controlling the melting cycle so that a stoichiometric compound could be guaranteed higher temperatures attainable in the arc furnace would probably assist the rapid homogenisation ternary compounds containing both uranium and thorium. Therefore further investigation of the a melting method is recommended.

The quality of the product so far obtained by reaction-pressing has been variable; furthe development is required so that high density and stoichiometric composition will be assured. P attention should be given to the composition changes which occur at high temperatures and to th for which the maximum temperature should be maintained. Although there are indications that sh reaction cycles are desirable this would need rationalising with product homogeneity where soli solutions of compounds are being prepared.

The cause of swelling of the reaction mass is not yet understood and requires investigatio If for example, it is due to impurities, pre-treatment of the metal powders may eliminate the proble and make full density more easily obtainable.

Until now, lattice parameters and densities of solid solutions have been assumed to vary linearly with the molar composition. In order that the theoretical densities of solution may be kno more accurately it is desirable that the variation of lattice parameter with composition be studied. Such a study should be preceded by an examination of the variation of lattice parameter with stoichiometry (that is, heavy metal : beryllium ratio) and, if this is appreciable, the composition of the so solutions should be kept to the beryllium rich limit.

8. ACKNOWLEDGMENTS

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TABLE 1

EXPANSIONS OF COMPACTS SINTERED AT 1200 °C

Run Number	U : Th ratio	Observed Expansions % of Original Volume				Mean Expansion % of Original Volume
1	1 : 0	30	40	27	30	32
2	9 : 1	52	52	45	48	49
3	8 : 2	70	69	70	65	68
4	6 : 4	85	81	81	85	83
5	1 : 1	88	88	92	90	90
6	4 : 6	95	102	106	104	102
7	2 : 8	120	129	116	110	119
8	1 : 9	111	110	107	111	110
9	0 : 1	108	106	111	–	108

TABLE 2

EXPANSIONS OF COMPACTS SINTERED AT 1000 °C TO 1400 °C

(U : Th = 1 : 2)

Run Number	Sintering Temperature °C	Expansion % of Original Volume	Original Be Content %	Final Be Content %
T1	1000	290	33	39.9
T4	1120	117	33	38.8
T2	1200	114	33	35.4
T5	1300	49	33	35.0
T3	1400	–90*	33	15.8

* Specimen T3 had shrunk severely and volume measurement was very approximate.

TABLE 3

SUMMARY OF REACTION–PRESSING RUNS

Run Series	Run Number (HP.)	U : Th Ratio	Weight of Reactants g	Maximum Temperature °C	Time at Temperature Minutes	Per Cent. o Theoretical Density
I	2	1 : 3	30	1375	30	88.5
	4, 6	12 : 19	30	1550	30	97, 83
	8, 11, 14	12 : 19	30	1550	15	95, 91, 105
	10	12 : 19	30	1600	15	90
	9	12 : 19	30	1650	15	110
	17	UBe_{13}	30	1500	15	97
	18, 19	UBe_{13}	30	1500	No Dwell	75, 83
	20	UBe_{13}	30	1550	20	98
II	23	UBe_{13}	30	1550	1	98
	24	UBe_{13}	30	1550	10	97
	25	UBe_{13}	30	1550	20	103
	26	UBe_{13}	30	1550	40	99
	27	UBe_{13}	30	1550	60	97
III	21	UBe_{13}	338	1300	Die failure	93
	22	$ThBe_{13}$	328	1350	Die failure	90

TABLE 4

COMPOSITIONS OF REACTION MIXTURES AND REACTION-PRESSED COMPACTS

Run Number	Reaction Mixture			Reacted Compact (Chemical Analysis)		
	w/o U	w/o Th	w/o Be	w/o U	w/o Th	w/o
HP.2	17	49.7	33.3	15	41	38
HP.4	26.2	40.5	33.3	25.1	40.0	32.
HP.5	22.6	44.1	33.3	22.2	43.8	31.
HP.6	26.2	40.5	33.3	26.4	40.1	32.
HP.7	52.1	14.8	33.1	50.5	11.5	31.
HP.10	26.2	40.5	33.3	29.2	37.7	30.
HP.11	26.2	40.5	33.3	28.6	38.2	30.

TABLE 5

COMPOSITION CHANGES DURING REACTION-PRESSING AT 1550 °C

(Series II Compacts)

Run Number	Time at 1550 °C Minutes	Weight Loss During Reaction g	Weight Ratio Be:U	Atom Rat Be:U
Reaction Mixtures	No reaction	–	0.54	14.3
HP 23	1	nil	0.54	14.3
HP 24	10	1.3	0.54	14.3
HP 25	20	0.9	0.52	13.8
HP 26	40	1.4	0.53	14.0
HP 27	60	4.5	0.51	13.5

TABLE 6

MICROPOLAROGRAPHY RESULTS, RUN HP 8

Sample Weight μg	% U	% Th	U : Th Ratio
34	36	40	0.9
34	22	47	0.5
35	34	47	0.7
39	33	44	0.8
45	31	42	0.8
46	31	44	0.7
51	32	39	0.8
61	29	43	0.7
66	29	46	0.63
74	27	39	0.69
137	29	40	0.73
140	30	41	0.73
Mean	30.25	42.70	0.71

TABLE 7

X-RAY DIFFRACTION RESULTS, RUN HP 8

Sample	Reflection	θ	Lattice Parameter Å	Calculated U : Th Ratio	U : Th Ratio in Reaction Mixture
UBe_{13}	10 8 0	73.8	10.272	–	–
$ThBe_{13}$	10 8 0	71.55	10.399	–	–
HP 8	10 8 0	72.4	10.348	0.60	0.63

TABLE 8

SUMMARY OF ARC-MELTING EXPERIMENTS

Run Number	Starting Material	Density Prior to Melting g cm^{-3}	Density After Melting g cm^{-3}	% Weight Loss on Melting	Remarks
M1	UBe_{13} Powder Compact	–	2.7	–	Porosity increase from top to bottom of button
M2	Button from Run M1	2.7	5.6	7	Large amounts of U in microstructure
M3	Button from Run M2	5.6	5.9	4.3	Large amounts of U in microstructure
M4	UBe_{13} powder compact. Turned during melt.	–	5.6	13	Large areas of U in microstructure
M5	UBe powder compact. Turned during melt.	–	4.17	5.6	Areas of U in microstructure

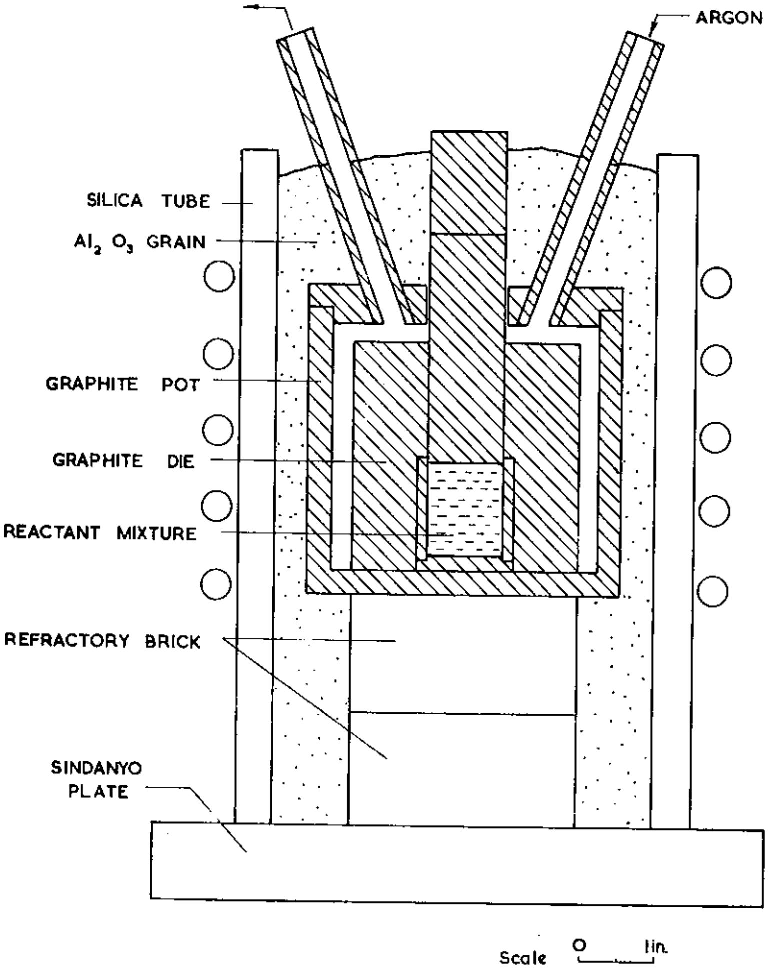


FIGURE 1 REACTION-PRESSING ASSEMBLY USED FOR THE PREPARATION OF INTERMETALLIC COMPOUNDS

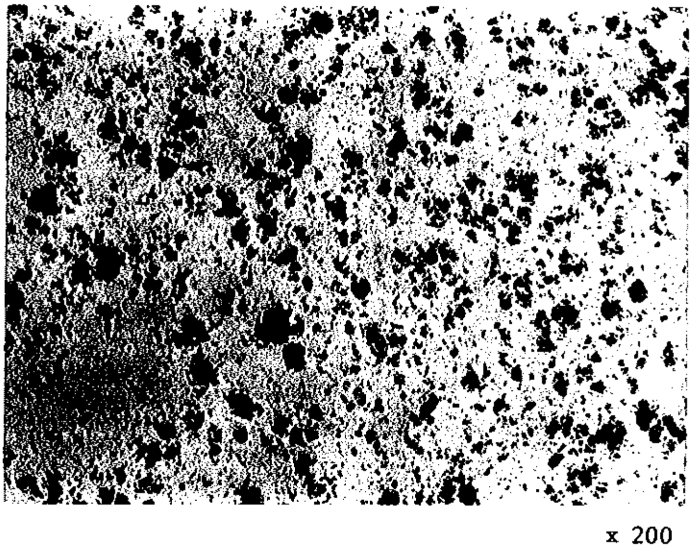
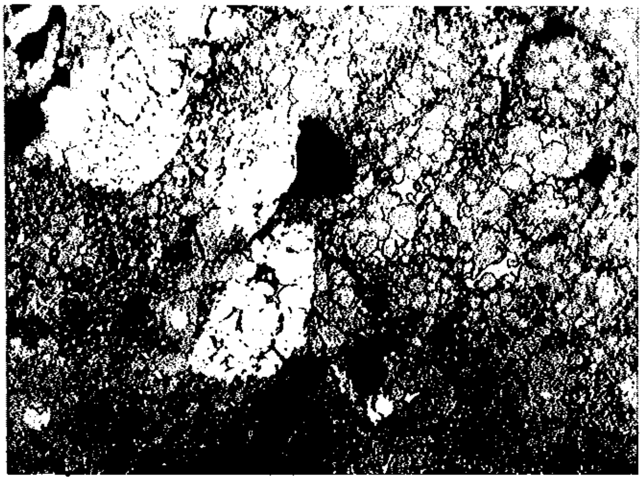


FIGURE 2 MICROSTRUCTURE OF REACTION-PRESSED COMPACT FROM RUN HP. 2



x 500

FIGURE 3 MICROSTRUCTURE OF UBe_{13} COMPACT FROM RUN HP. 21

(Large white areas are unexplained etching phenomenon)

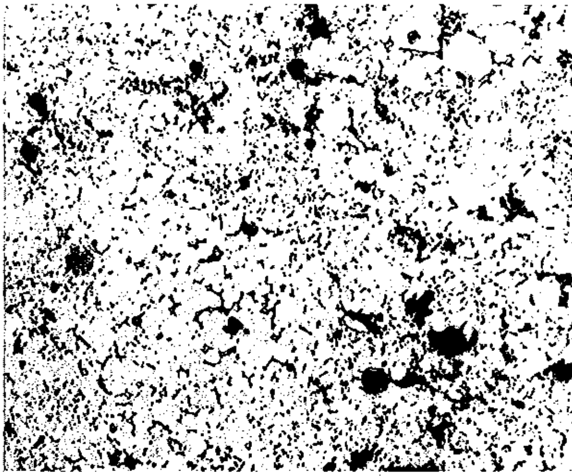
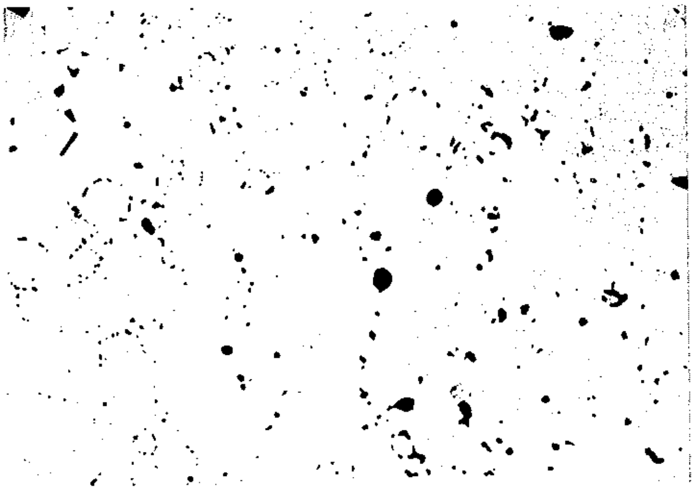


FIGURE 4 MICROSTRUCTURE OF Th
COMPACT FROM RUN HP. 22



x 250

FIGURE 5 MICROSTRUCTURE OF UBe_{13} COMPACT FROM RUN HP. 23
(1 minute at 1550 °C)

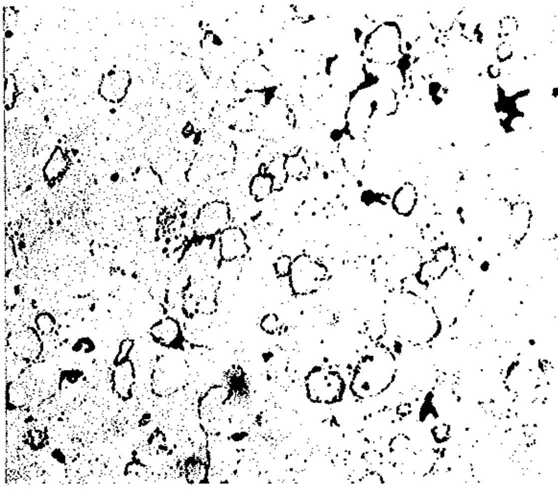


FIGURE 6 MICROSTRUCTURE OF U
COMPACT FROM RUN HP. 24
(10 minutes at 1550 °C)