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WAPD- PWR-FEB-106

MASTER

PREPARATION OF URANIUM ALLOYS BY MELTING

65455

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For The Atomic Energy Commission

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Chief, Declassification Branch

A. General Considerations

Because uranium is so reactive with the elements of the atmosphere, precautions must be taken to isolate it from the atmosphere during melting. There are three methods of accomplishing this: by vacuum melting in which the melting chamber is evacuated to 10 microns or less, melting in an inert gas atmosphere such as helium or argon, and melting under a slag. By far the most popular method of alloy preparation is vacuum-induction melting although many alloys, especially those prepared by arc melting, are melted under inert gas atmospheres. The use of slags has been limited exclusively to melting uranium and has not been adopted for use in alloy preparation.

In addition to its reactivity with the atmospheric elements, uranium reacts with practically all refractory materials; hence, care must be taken in the selection of the proper refractory for melting to prevent excess contamination of the melt by the crucible.

1. Vacuum Induction Melting

a) Equipment

Vacuum induction furnaces are of two basic designs which are the can type in which the induction coil is within the vacuum chamber and the tube type in which the induction coil is on the outside of the furnace. The advantages of the can-type furnace are:

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- 1) Tilt pouring can be utilized.
- 2) Buckets for making alloy additions are readily installed and manipulated.
- 3) Mechanical agitators can be utilized for stirring the melt.

The two advantages of the tube type furnace are simple construction and smaller space requirements.

Power supply is a function of furnace size, construction, frequency of power source, and efficiency desired. Standard motor-generator sets are utilized as the power source for vacuum induction melting.

Vacuum equipment usually consists of a mechanical roughing pump which is used to reduce the furnace pressure to less than 100 microns, and a diffusion pump backed in series by another mechanical booster pump to reduce the pressure to 1 micron or less.

Vacuum induction melting has the following advantages over arc melting or consumable arc melting.

- 1) Preparation of larger ingots in less time.
- 2) Better chemical homogeneity unless arc melted ingots are remelted.
- 3) Shapes may be cast more easily.
- 4) Control over the cooling rate can be exercised.

A good practice to follow when induction melting uranium alloys is to place the alloy addition on the bottom of the crucible with the uranium on top. In all of the commonly used alloys with the exception of the uranium-aluminum alloys the uranium is the largest constituent by volume as well as weight and it is also the low melting element; hence, the uranium melts first and the alloy addition floats up through the molten uranium affording an excellent opportunity for uniform alloying before the liquidus is reached.

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b) Refractories for Containing Molten Uranium and Its Alloys

Several refractories are available for vacuum induction melting uranium alloys although the selection of the proper refractory depends upon several factors. Among these are:

- 1) Chemical stability in vacuo.
- 2) Reaction between the refractory and the alloy.
- 3) Strength at high temperature
- 4) Resistance to thermal shock.
- 5) Outgassing.
- 6) Availability.
- 7) Cost.

Graphite

Graphite is probably the most commonly used material for high temperature operations being used extensively for crucibles, molds, and structural furnace parts. Moreover, graphite crucibles washed with a slurry of some refractory oxide are often utilized to reduce carbon contamination in the alloy.

Graphite has many advantages over oxide refractories in that it has excellent high temperature properties, is stable in vacuo, is readily available at low cost, and is easily machined. The one disadvantage in the use of a graphite crucible is that it chemically reacts with uranium at temperatures over 1000°C and, in addition, some carbon particles are always picked up in the melt by mechanical action in pouring. The amount of carbon contamination in alloys melted in graphite varies widely depending upon the alloy, the temperatures of alloying, surface to volume ratio of the melt, and the holding time before pouring. Aluminum-uranium alloys heated to about 1000°C for one hour pick up about 100-200 ppm of carbon while uranium-molybdenum alloys heated to 1500°C for 15-min

pick up 0.4-0.5 w/o of carbon.

Carbon in amounts exceeding 500 ppm is known to be deleterious in some uranium alloys in that corrosion and radiation characteristics are adversely affected. In U-3.8 w/o Si alloy, carbon lowers the temperature of the peritectoid reaction by which the intermetallic compound U_3Si is formed.

Several different grades of graphite are available. AUC graphite is a fine grained, dense material having about 20% porosity and is the most chemically pure. It is the best grade for crucibles and stopper rods. CS or ATC graphite contains about 25% porosity and is probably the most extensively used crucible material. Grades AGR and AGX contain about 32% porosity and are generally used for structural furnace parts. These grades should not be used for crucibles or molds without an oxide wash.

Oxides

Alumina- Al_2O_3 ^{3,16/}

Alumina crucibles have not been used extensively in vacuum melting uranium alloys although alumina molds have been used for casting uranium slugs. In air aluminum melts at $2050^{\circ}C$ and has relatively good resistance to thermal shock; however, its use for melting crucibles at temperatures above $1300^{\circ}C$ has not been extensively investigated. Limited attempts to melt uranium-molybdenum alloys at $1475^{\circ}C$ have shown severe outgassing of the alumina and a reaction between the crucible and the melt. Alumina is reduced by graphite at about $1500^{\circ}C$ so care should be taken in its contact with furnace parts.

Beryllia - (BeO) ^{3,12/}

Beryllia crucibles are used extensively for alloying uranium alloys above $1500^{\circ}C$ and can be used close to their melting point of $2500^{\circ}C$ if properly fired. Beryllia is very stable in vacuo and reacts very slowly with uranium

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Alloys -- less than 100 ppm of beryllia are picked up during use as a slurry for washing graphite crucibles has proved effective in preventing carbon contamination in several uranium alloys. See table below.

| <u>Alloy</u> | <u>Temperature °C</u> | <u>Carbon Contamination (ppm)</u> |
|---------------|-----------------------|-----------------------------------|
| *U-1.8 w/o Si | 1650 | 100-400 |
| U-10 w/o Nb | 1700-1750 | 100-300 |
| U-12 w/o Nb | 1775 | 300-350 |
| U-13.5 w/o Mo | 1650 | 100-200 |
| U-15 w/o Mo | 1775 | 300-500 |

*Holding time at temperature about 15 minutes. Extreme care must be used in crucible preparation for this alloy.

Beryllia, however, has several disadvantages that preclude its use as a refractory material for temperatures below 1500°C where other oxide refractories can be used. Among the chief disadvantages of beryllia crucibles are their poor resistance to thermal shock and their retention of residual stresses even after high firing. In addition, it is difficult to seat an oxide stopper rod in a beryllia crucible. These disadvantages may be overcome somewhat by packing the crucible in beryllia sand and using a tilt pour arrangement. Beryllia is a toxic material and special handling precautions must be taken to avoid health hazards. Beryllia crucibles are expensive and not readily available; approval must be obtained from the AEC prior to their procurement.

Lime (CaO)^{5/}

Lime crucibles are not used in the preparation of uranium alloys; however, lime rammed steel bombs are used for the reduction of uranium and co-reduced uranium alloys. Lime washes for graphite crucibles have received a limited use for melting a ternary alloy of U-5 w/o Zr-1.5 w/o Nb below 1500°C.

Magnesia (MgO)^{11/}

Magnesia crucibles may be used for alloying uranium alloys at

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temperatures below about 1500°C. Above this temperature severe out-gassing and spalling occur if melting is done in vacuo although slightly higher temperatures may be utilized by using an inert gas atmosphere. Strongly oxidizing atmospheres are needed to prevent volatilization of the magnesia at temperatures over 1700°C.

Magnesia has relatively good thermal shock resistance and good strength up to 1550°C; however, above this temperature there is a slight decrease in its mechanical properties. Magnesia and magnesium-zirconate ($MgZrO_3$) washes for graphite crucibles have been used successfully for uranium alloy melting temperatures up to 1500°C maintaining carbon contamination between 200-600 ppm while magnesium contamination is less than 100 ppm.

Silica (SiO_2)

Silica crucibles are not usually used for melting uranium alloys since they soften about 1400°C; however, some recent results indicate that fused quartz crucibles packed in refractory sand may be used for short periods up to about 1650°C.

Thoria (ThO_2)^{16/}

Thoria has the highest melting point of any available oxide — above 3000°C, and it is stable in vacuo up to 2250°C. The use of thoria crucibles for melting uranium alloys has been limited and not many data are available although thoria stopper rods have been used successfully for many alloys up to 1750°C. Some investigations have shown that U-Si alloys react with thoria about 1600°C. Thoria has poor high temperature strength and resistance to thermal shock. Care must be taken to isolate the furnace from external vibrations if thoria crucibles or stopper rods are used. Thoria is radioactive and toxic; consequently, handling precautions must be taken to prevent health hazards.

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Urania (UO₂)^{10/}

The use of urania crucibles has been limited to the preparation of high purity uranium. The reaction between uranium and urania occurs slowly at temperatures up to 1700°C; consequently, urania is an excellent material for the preparation of high purity uranium alloys since the only impurity introduced during melting is less than 10 ppm of oxygen.

Urania has poor thermal shock resistance and care must be taken in introducing power into the furnace to prevent breaking the crucible. Urania crucibles are hard to fire, requiring temperatures around 2000°C. Their manufacture is expensive and AEC approval must be obtained for their use.

Zirconia (ZrO₂)^{3,11,12,13/}

Zirconia crucibles are used for melting many uranium alloys whose alloying temperatures do not exceed 1500°C. Above 1500°C, in vacuo, zirconia forms carbides when in contact with graphite and the refractory itself begins to decompose about 1600°C. It should be noted that zirconia is a fairly good electrical conductor above 1600°C and consequently should not be used in contact with resistance windings above this temperature.

Zirconia crucibles are relatively inexpensive and available in a large range of sizes. They have good high temperature mechanical properties and fairly good resistance to thermal shock. Zirconia reacts with uranium to cause zirconium contamination in the alloy. The amount of contamination is usually less than 500 ppm if alloying temperatures do not exceed 1500°C. At 1600°C the reaction is rapid and zirconia crucibles are "wet" by the melt.

Zirconia slurries have been used for washes on graphite crucibles for U-Mo and U-Nb alloys with excellent results up to 1500°C. Carbon contamination is in the range of 200-600 ppm.

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Below is a table summarizing pertinent information about oxide refractories.

| Chemical Composition | Melting Point °C | Stable In Vacuo °C | Thermal Shock | Remarks |
|--------------------------------|------------------|--------------------|---------------|--------------------------------------|
| Al ₂ O ₃ | 2050 | 1450 | Good | Reduced by graphite at 1500°C |
| BeO | 2570 | 2200 | Poor | Toxic, AEC approval required, Stable |
| CaO | 2572 | 1500 | --- | Data limited |
| MgO | 2300 | 1500 | Good | Outgasses and spalls about 1500°C |
| *SiO ₂ | 1585 | M.P. | Good | Softens about 150°C below M.P. |
| ThO ₂ | 3000 | 2250 | Poor | Toxic, radioactive |
| UO ₂ | 2176 | 1900 | Poor | Radioactive, AEC approval required |
| ZrO ₂ | 2700 | 1585 | Good | Reacts with uranium |

*Cristobalite; quartz 1475°C, tridymite 1670°C.

Special Refractories

Several new classes of refractories have recently been developed that may prove useful in melting uranium alloys.

Carbides^{3/}

Carbide crucibles have not been used for the preparation of uranium alloys because of the possibility of contamination of the alloy with carbon, silica, boron, and sodium. Crucibles of boron carbide are being developed at present but have not been evaluated.

Nitrides^{3/}

Some nitride refractories are obtainable but no data are available on their use as melting crucibles for uranium alloys.

Sulfides^{1,2/}

Sulfide refractories are a recent development and are not generally available at present. Investigations have indicated that some sulfide refractories may be useful in the preparation of uranium alloys. Below is a table listing pertinent facts about sulfide refractories.

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| Chemical Composition | Common Name | Melting Point °C | Stable In Vacuo °C | Thermal Shock | Remarks |
|--------------------------------|----------------|------------------|--------------------|---------------|--|
| CeS | Cerium brass | 2450 | 1900 | Excellent | Slight sulfur contamination |
| Ce ₂ S ₃ | _____ | 1980 | 1500 | Fair | Sulfur given off about 1475°C |
| Ce ₃ S ₄ | Cerium Black | 2050 | 1840 | Fair | More S contamination than CeS |
| ThS | Thorium Silver | 2100 | 1900 | Poor | Must be kept from oxygen at high temp. |
| ThS ₂ | _____ | | 1500 | Poor | Decomposes at 1500°C |
| Th ₂ S ₃ | _____ | 2000 | 1800 | Poor | Some S contamination |
| Th ₄ S ₇ | _____ | 2100 | 1800 | Poor | Some S contamination |
| BaS | _____ | 2000 | 1600 | Fair | |

It should be noted that the noble metals should not be melted in cerium sulfide crucibles because they displace the cerium.

c) Molds

The type of mold material utilized for uranium alloys depends upon:

- 1) Properties desired in the as-cast material.
- 2) Weight of melt.
- 3) Casting temperature.
- 4) Shape of the cast piece.

Chill castings have several advantages over castings poured in hot molds. These advantages are:

- 1) Minimum of transformation products, often of importance in uranium alloy castings.
- 2) Reduction of coring.
- 3) Fine grain size.

The advantages of pouring into refractory molds are:

- 1) Control of cooling rate.
- 2) Sounder casting.
- 3) Reduction of primary pipe.

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4) Improvement of surface finish.

Water cooled copper and water cooled steel molds are used for casting ingots of many uranium alloys. Usually the copper molds are used with no coating while coatings such as colloidal graphite, silica, or zirconia are used on steel molds to prevent the alloy from attacking the steel. The weight of the melt and the temperature at which it is to be poured determine the advisability of using water cooled molds.

Graphite molds are probably the most extensively used in casting uranium alloys. AGR or AGX graphite is an excellent mold material being inexpensive, easily machinable, and it can be heated to any desired temperature by resistance, induction, or radiation. Molten uranium reacts chemically as well as mechanically with a graphite mold; however, this reaction is usually slight and carbon contamination is limited to the very surface of the casting. This contamination may be removed by mechanical cleaning or by pickling in nitric acid.

The use of an oxide wash on graphite molds not only minimizes carbon contamination but greatly increases mold life by preventing mold erosion. Pouring onto a thin disc of some refractory metal, such as molybdenum or tantalum, prevents mold erosion at the mold bottom, which can be quite a problem for the dense uranium alloys particularly in large size castings.

Investment type molds of silica and zircon flour have been used successfully for the production of uranium alloy castings. These can be heated to about 1300° - 1500° C without severe outgassing. In general, castings made in investment molds have a superior surface finish to castings made in graphite or water cooled molds.

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2. Non-Consumable Electrode Arc Melting^{12,13,14/}

Arc melting has two outstanding advantages over any other type of melting. These are:

- a) Preparation of alloys whose melting point is beyond the temperature range of induction or resistance equipment.
- b) Melting of alloys which react with all refractory crucibles. In addition, it is usually easier to prepare small buttons of experimental alloys by arc melting than by any other means.

The disadvantages of arc melting are:

- a) Alloy homogeneity is not as good as induction melted alloys when the melting constituents have widely divergent melting points.
- b) Preparation of large ingots is time consuming in the non-consumable electrode arc melting process.

Uranium alloys are usually arc melted in a partial atmosphere of 15-20 in. of mercury. This partial pressure is necessary to reduce the corona effect of the arc which occurs at low pressure. The atmospheres most commonly used are helium and argon or mixtures of these inert gases.

Thoriated tungsten tipped water cooled electrodes are usually used for arc melting. Tungsten contamination is negligible in most alloys although tips must be replaced regularly due to erosion.

3. Consumable Electrode Arc Melting^{9,12,13,14/}

The advantages of consumable electrode arc melting are:

- a) Production of sound ingots from porous and pipe filled induction castings.
- b) Production of a sound ingot from composite electrodes.

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c) Production of a semi-chill microstructure providing the diameter of the ingot being melted is not appreciably larger than the consumable electrode.

d) Improvement in homogeneity of previously non-consumable electrode arc melted or induction melted material whose constituents have great differences in density. An outstanding example of this type of alloy is Zr-U alloy containing about 0.03 w/o boron.

Melting conditions for consumable electrode arc melting are practically the same as direct arc melting. A precaution that should be taken prior to striking the arc is to place machine chips of the alloy being melted in the bottom of the crucible to prevent the arc from burning through. This also reduces copper contamination in the alloy.

B. Preparation of Specific Alloys

1. Uranium-Aluminum Alloys

Alloys of aluminum and uranium having from 5 to 40 w/o uranium have been prepared by induction melting in air, helium, and vacuo. Because of the wide differences in densities between these elements, the atomic percent of uranium in aluminum ranges from 0.59 w/o to 7.02 w/o for these alloys; consequently, the melting characteristics of these alloys are practically identical.

Al-U alloys are melted in graphite crucibles, although any oxide crucible would be satisfactory for these alloys. Alloy temperatures vary between 1000°C and 1400°C depending on the holding time desired. Soundest ingots are produced if the melt is poured slowly at about 675°-700°C. Skulls form if the pouring temperature is below 665°C. Carbon contamination from graphite crucibles is about 100 ppm if the alloy is superheated below 1000°C.

Al-U alloys must be poured into water cooled molds to prevent alloy

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segregation. Arc melting the top of ingots to reduce primary pipe is not advisable since segregation is promoted. Consumably arc melted ingots have fair alloy homogeneity but tend to have porosity near the edges.

2. Uranium-Chromium Alloys^{8/}

The alloy of the uranium-chromium system receiving the most study is a eutectic occurring at about 19.4 a/o Cr (4.99 w/o Cr) at about 860°C. Alloys are made with an initial charge weight of 5.2 w/o chromium to allow for chromium losses during melting. Alloys are melted in graphite crucibles and superheated about 550°C to 1400°C for 5 or 6 minutes. Probably a partial helium atmosphere is best for melting this alloy to reduce chromium losses. The melt is cooled to 1000°C prior to pouring and cast into molds heated to 600°C to form sound castings.

This alloy is used for the production of radiator type castings which are cast into a graphite washed steel mold. This mold has a cross-sectional configuration similar in design to a honey comb which produces water channels in the casting. Heating of the mold is necessary to prevent the alloy from freezing and forming pipe before the mold is completely filled. A heated mold has the disadvantage of producing a large grained casting. Attempts to centrifugally cast the U-5 w/o Cr alloy into cold graphite coated steel molds to produce a fine grained structure have not been successful.

Uranium-Molybdenum^{11/}

Uranium-molybdenum alloys of 6-13.5 w/o molybdenum have been extensively investigated. Alloys up to 12 w/o Mo are induction melted in zirconia washed graphite crucibles. Alloys melted in this manner show about 250-450 ppm carbon contamination and less than 500 ppm zirconium contamination. The U-13.5 w/o Mo

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alloy is induction melted in beryllia washed graphite because of its high alloying temperature of 1650°C. Carbon contamination is about 200-350 ppm while beryllium contamination is less than 100 ppm.

Magnesia and magnesium zirconate washes have been used on graphite molds for alloys up to 1500°C. In general, they have not been as successful in preventing carbon contamination as zirconia since carbon contamination in alloys prepared in this manner exhibit about 0.1 w/o C contamination. Magnesia crucibles outgas severely at about 1475°C when used for melting the U-12 w/o Mo alloy. Zirconia makes an excellent crucible for induction melting uranium-molybdenum alloys up to 12 w/o Mo in that carbon contamination is eliminated and zirconium contamination is less than 500 ppm.

Good castings up to 48 in. long, exhibiting small amounts of pin point porosity along their longitudinal axes, can be produced by casting into 2-in. diam cold graphite molds. This pin point porosity can be entirely eliminated by consumable arc remelting. If water cooled copper molds are utilized, it is mandatory to consumably arc remelt to obtain a sound casting.

Uranium-molybdenum alloys exhibit micro-coring after solidification. This effect can be overcome by homogenizing the castings for 24 hr at 900°C in inert gas or vacuo.

Uranium-Niobium^{12,13/}

Uranium-niobium alloys up to 12 w/o niobium have been prepared by the duplex melting technique of induction melting then consumable arc remelting. Because of the high melting point of these alloys, it is necessary to use beryllia or beryllia washed crucibles for induction melting uranium-niobium alloys over 4 w/o niobium to maintain carbon contamination below 500 ppm and

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prevent a reaction between the melt and the crucible.

Alloys of U-10 w/o Nb superheated to 1750°C in beryllia washed graphite crucibles show less than 100 ppm of beryllium (lower limit of detection) and 250-400 ppm of carbon.

Casting U-10 w/o Nb into cold graphite molds produces about 30% of a second phase which is probably transformed alpha. Casting into water cooled copper molds reduces this transformation product to about 5-10 percent which is the amount observed in consumable arc remelted ingots.

As in the uranium-molybdenum alloys, the uranium-niobium alloys exhibit micro-coring. This can be reduced by heat treatments on the order of 1000°C for 48 hr in inert atmospheres or vacuo.

Uranium-Silicon^{12,15/}

The only uranium-silicon alloy which has been considered for reactor use is U-3.8 w/o Si which is heat treated to produce the intermetallic compound U_3Si . This alloy is produced by melting lump or granular silicon with uranium at 1600°C - 1650°C in vacuo. The alloy can be cast in graphite or investment molds to form sound rods as small as 3/8-in. diam. If cast into water cooled molds, the ingot must be consumably arc remelted to produce a sound casting.

The compound, U_3Si , is formed by a peritectoid reaction at 930°C which is drastically lowered by carbon contamination (1000 ppm lowers the temperature 100°C). To produce this reaction a heat treatment of 800°C for 168 hr is required. Carbon also has a deleterious effect on the corrosion resistance of this alloy; so it is imperative to keep it to an absolute minimum, preferably less than 200 ppm. Beryllia washed graphite crucibles have produced alloys with less than 200 ppm of carbon and less than 100 ppm of beryllium contamination.

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fairly consistently. Zirconia, thorium, and magnesia crucibles have shown severe spalling, outgassing, and some reaction with the melt at 1600°C. Some results indicate that fused quartz crucibles packed in refractory sand may be useful in the preparation of this alloy. Analytical results indicate that 3.7 w/o silicon should be charged in the melt to obtain an alloy of 3.8 w/o.

Arc melted ingots of the U-3.8 w/o Si alloy, unless remelted several times, exhibit segregation to an extent which prevents the satisfactory formation of the intermetallic compound. Because of its poor fabrication characteristics and the fact that fabrication reduces the corrosion resistance of the alloy, it is believed that the alloy should be used in the cast and heat treated condition. Many different ternary alloys of U-3.8 w/o Si, with the addition varying from 0.1 w/o to 0.5 w/o, have been prepared in an effort to reduce the effects of carbon contamination and to improve the characteristics in general.

Uranium-Zirconium^{4,7/}

Uranium-zirconium alloys have been studied in all compositions. Because of the tremendous reducing power of zirconium, no known oxide or sulfide crucible is suitable for the preparation of alloy containing over 4 or 5 w/o zirconium. Induction melting uranium-zirconium alloys in graphite is possible although about 1 w/o of carbon is picked up.

Low zirconium-uranium alloys are melted in CaO rammed graphite crucibles and magnesia crucibles. Not many data are available on the refractory pick up of these alloys, but it is probably high.

The most commonly used uranium-zirconium alloys are Zr-50 w/o U,

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U-2 w/o U, and Zr-6 w/o U. These alloys are usually prepared by the following techniques.

- 1) Non-consumable arc melt.
- 2) Roll out ingot and chop into small pieces.
- 3) Non-consumable arc melt.
- 4) Consumable arc melt.

The consumable arc melting is necessary to produce a sound casting. In alloys used for STR fuel which contain boron additions on the order of 0.03 w/o it is necessary to consumably arc melt to obtain the chemical homogeneity desired.

Ternaries

No special techniques are required for the preparation of most ternary alloys providing the ternary addition is small. Practically every alloy discussed has been prepared with many different ternary additions.

One of the most commonly prepared ternary alloys is U-5 w/o Zr-1.5 w/o Nb which is induction melted in lime washed graphite molds under an argon atmosphere.^{5/}

Special Techniques

a) Vibration During Freezing^{5/}

Several methods of vibrating the alloy during freezing have been employed to produce a fine grained casting. These methods of agitation include:

- 1) Inductive stirring.
- 2) Electromagnetic.
- 3) Piezoelectric.
- 4) Magnetostrictive.

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Castings produced by such methods show slightly higher densities. Varying the vibration rate from 20 to 2000 cycles per second has not produced finer grained castings, but has broken the inclusion into smaller pieces.

b) Centrifugal Casting^{17/}

Recently, much effort has been expended on the development of centrifugal casting of uranium alloys. The advantages of centrifugal casting are:

- 1) Casting of intricate shapes.
- 2) Sounder castings.
- 3) Fine grained, chill cast castings.

This process is not widely used at this time.

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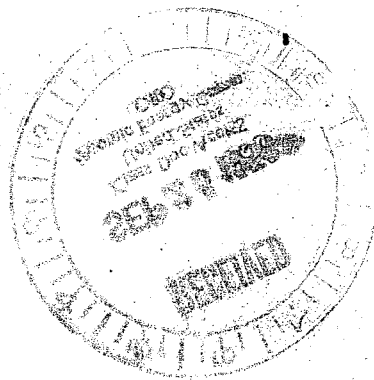
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