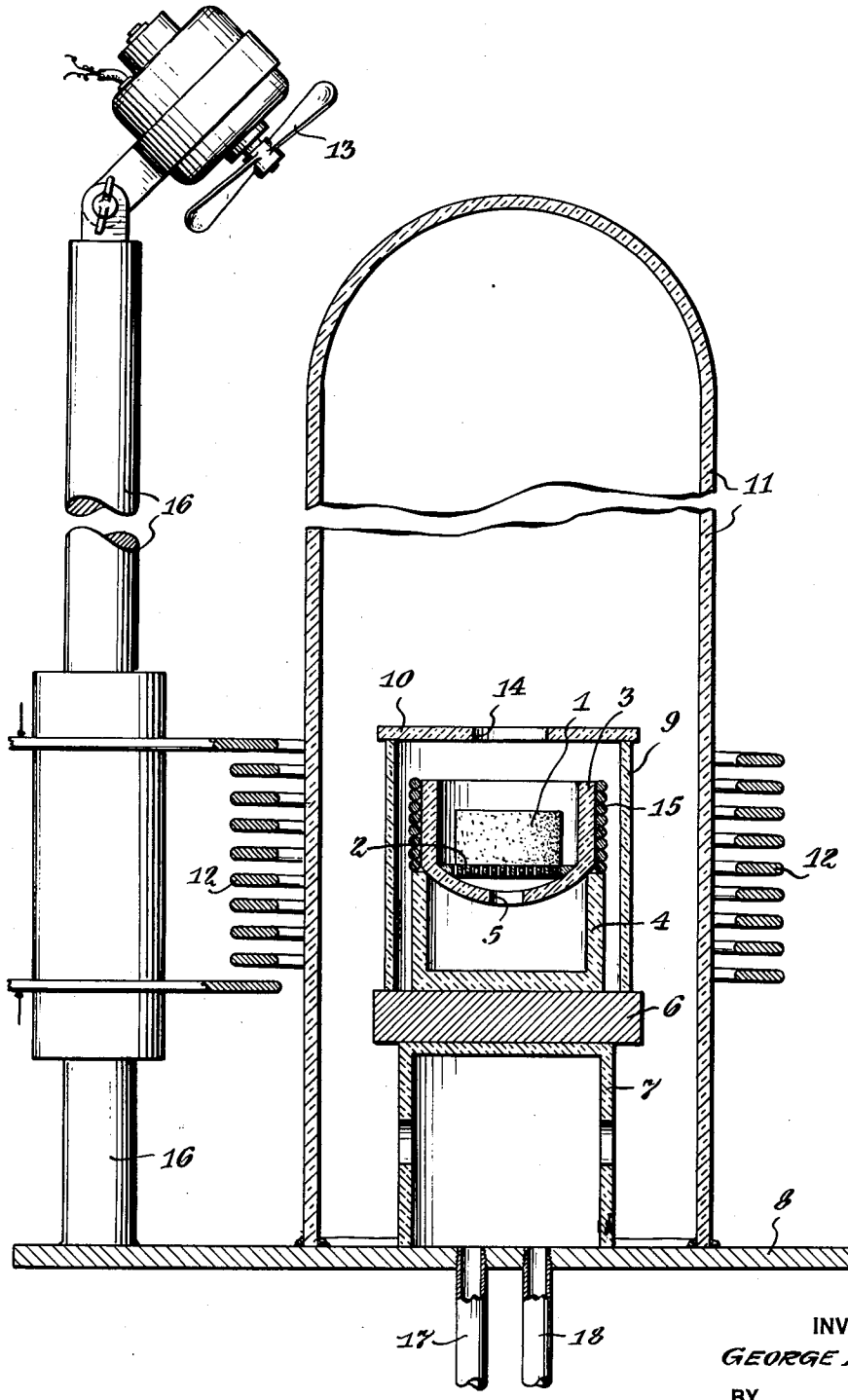
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PROCESS OF VACUUM REFINING URANIUM

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PROCESS OF VACUUM REFINING URANIUM

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This invention relates to methods for refining uranium and more particularly to methods for melting and casting uranium in substantially pure coalescent form.

In a copending application of William C. Lilliendahl et al., Serial No. 478,270, filed March 6, 1943, now Letters Patent No. 2,690,421 issued September 28, 1954, a method is described whereby uranium may be prepared in the form of a compressed coherent powder. Briefly, the method thereof includes the electrolysis of potassium uranous fluoride, KUF_5, uranium tetrachloride, UCl_4, or uranium tetrafluoride, UF_4, in a fused salt bath of sodium and calcium chlorides in which uranium is deposited on the cathode in the form of a powder, the separation of the uranium powder from soluble impurities by washing with water and acid, and finally the compression of the uranium powder into a slug or button while protecting it from oxidation. By heating said uranium button to sintering temperature in a high vacuum as described in United States Letters Patent No. 1,814,719 to J. W. Marden et al., many vaporizable impurities may be eliminated from the button, but nonvaporized impurities remain in the sintered mass. Such impurities cannot be tolerated in certain uses of uranium.

The present invention provides a method of separating the uranium from vaporizable impurities, allowing the nonvaporized impurities to form a slag, and collecting or casting the molten uranium in a suitable mold while separating out the slag. Since uranium is extremely active, especially at high temperatures, the melting and separating are preferably carried out in a high vacuum of the order of 10 microns of mercury or less.

An object of the invention is to provide methods for converting impure uranium into a cast form substantially free from vaporizable and nonvaporizable impurities.

It is another object to provide methods for producing uranium metal in a substantially pure and ductile form that may be conveniently handled and easily worked.

A further object is to provide methods for melting and collecting molten uranium in a high vacuum.

Additional objects will be apparent from the following description and drawings in which the single figure is a vertical section of a preferred form of melting and separating apparatus.

The preferred form of apparatus shown in the drawing is fully assembled and ready for the melting to begin. An impure uranium button **1** is supported on a strainer **2**, which provides a support on which the slag will be retained when the molten metal flows away from the button. The strainer or bridge member **2** shown in the drawing may comprise a plurality of spaced, parallel bars, resting in a crucible **3**, which latter is provided with a funnel-shaped lower portion extending into and supported by a mold **4**. The mold **4** may be made in any shape in which it is desirable to cast the metal. An opening **5** in the bottom portion of the crucible **3** allows the molten metal to flow into the mold **4**. The mold **4** is supported on a massive tungsten block or plate **6** that in turn rests on a support **7**. A bronze plate **8** provides a mounting

for the entire assembly. A radiation shield **9** and cover **10** surround and cover the crucible and mold.

The crucible, mold and supports are enclosed by an envelope **11** constructed of a refractory glass, such as "Vycor," a 96% silica glass having a softening point of about 1500° C. Water-cooled "Pyrex" glass may be used but has a shorter life than "Vycor." This is due to the fact that "Vycor" glass withstands acids used to remove condensed impurities better than "Pyrex" glass. The envelope **11** should be of such a height that it extends well above the cover **10**, thus providing a space above the crucible **3**, in which vaporized impurities may be dispersed and cooled at points where they will not recombine with or contaminate metal in the mold **4**. The inner upper wall of the "Vycor" envelope **11** provides a surface on which vaporized impurities may be condensed at a suitable distance from the hot metal in mold **4**. An induction heating coil **12** connected to a suitable source of high frequency current extends around the envelope **11**, and is slidably mounted for vertical movement on a standard **16**. A pair of pipes **17** and **18**, in open communication with the interior of the envelope **11**, are connected to a vacuum pump and to a source of inert gas, respectively, neither of which is shown.

In order to secure a homogeneous casting of high purity, great care must be used in choosing the materials of which the apparatus is made. All materials must be inert with respect to uranium, and they should be highly refractory since the melting point of uranium appears to be about 1133° C. All parts of the apparatus that are directly contacted by the molten metal, such as the crucible **3**, bridge **2** and mold **4**, are preferably made of beryllia or thoria, as these refractory oxides are inert with respect to molten uranium. Carbon, graphite, silicon carbide, or alumina, or such materials lined with beryllia or thoria may be used for these parts. The support **7**, as well as the shield **9** and cover **10**, is preferably constructed of silica or alumina. Before use, each piece of the apparatus should be prebaked at a temperature above the melting point of uranium in order to drive out any entrapped or occluded gases, or other vaporizable material.

In the preferred procedure, all parts of the apparatus are pre-baked and assembled with a uranium button **1** on bridge **2**, as described hereinbefore. The envelope **11** is hermetically sealed to base **8** by suitable means, such as "Apiezon" sealing compound, and is then evacuated by means of a vacuum pump connected to pipe **17** until the pressure within the envelope is less than 10 microns of mercury. The pressure should be held to 10 microns or less throughout the melting and collecting in order to prevent contamination of the melt. The coil **12** is initially lowered below the position shown, and connected to a high frequency current supply to inductively heat the massive tungsten block **6** which, in turn, heats the mold **4** until these parts have reached a temperature above the melting point of uranium. This preheating of the mold and its support has been found effective to prevent the molten metal from freezing immediately when it flows into and strikes the mold. Quick freezing causes an undesirable uneven bottom surface on the casting and hence should be avoided. The massive tungsten block **6** retains enough heat so that the bottom of the mold remains hot when the heating coil **12** is moved upward in the next step.

After the block **6** and the mold **4** have been preheated, the coil **12** is raised to the position shown in the drawing, in which the coil surrounds the crucible **3** and the mold **4**. As the temperature of button **1** is raised above the melting point of uranium, the film or sac formed of surface impurities is broken, and the uranium metal begins to flow from the mass through the bridge **2** and into the mold

4 and continues to flow until all the uranium metal that will flow is separated from the unvaporized impurities. Vaporizable impurities such as sodium, calcium, and probably magnesium, cadmium and zinc become vaporous and are liberated from the molten metal both in the crucible 3 and in the mold 4. The term "vaporizable impurities," as used above and also in the appended claims, refers to those impurities in a mass of impure uranium that becomes vaporous or gaseous by sublimation of otherwise at temperatures of the order of 1080° C. The nonvaporized impurities such as the oxide and fluoride dross having considerably higher melting points than that of uranium remain supported on the bridge 2 in the form of slag, and impurities that vaporize are drawn off by vacuum or condense on the inner surface of the envelope 11. Vaporizable impurities may condense on the inner upper surfaces of the envelope 11 away from the heating coil 12 without the use of an external air blast from fan 13, but better results are generally obtained by cooling the envelope with the air blast.

When the metal has ceased to flow, the coil 12 is further raised until its lower end is approximately on a level with the mid-point of the mold 4. The heating is continued in this position from five to ten minutes, after which the coil 12 is disconnected. This final heating is necessary in order to insure that the casting will solidify or freeze from the bottom to the top. Castings that freeze downwardly from the top surface are apt to contain undesirable voids or gas pockets.

Uranium metal has a density of about 18.7 grams per cubic centimeter, and, therefore, a relatively large mass of it may be heated in a small crucible and cast in a small mold. However, such a mass takes considerable time to cool, especially in a vacuum, and uranium should be cooled to room temperature before exposure to the air in order to hold oxidation to a minimum. I have discovered that the cast metal can be cooled in a relatively short period of time in the presence of an inert gas such as helium or argon. This may be done by introducing the inert gas into the envelope 11, through the pipe 18, immediately after the coil 12 is disconnected, or removed from the envelope 11. The inert gas pressure inside the envelope is increased to one-half to three-quarters of an atmosphere, and, as the apparatus gradually cools, more gas is added. The gas conducts the heat from the metal to the glass envelope 11, and a blast of air from the fan 13 is directed at the outer surface of the envelope to hasten cooling. Such cooling also aids in controlling vaporized impurities given off from the button and the hot metal by causing at least some of them to condense on the cooled glass envelope. Cooling also lowers the vapor pressure of such impurities, and reduces the danger of having them recombine with the hot metal.

The shield 9 and the cover 10 are used to reduce the head lost by radiation from the crucible 3, thereby reducing the time and energy necessary to complete the melt. An opening 14 is provided in the cover 10 to allow for free escape of vaporized matter from the crucible to the envelope 11.

An electrical conducting material, such as a tungsten heating coil 15, surrounding the crucible 3, may be provided for assisting the melting of a button of uranium powder that contains an unusual amount of impurities. When the uranium button is melted in a beryllia, thoria or an alundum crucible, only the button is heated by induction because such a crucible is practically a non-conductor of electric currents. Moreover, if the metal powder is very impure, it is also a poor conductor and heats slowly. With the tungsten coil 15 around the crucible, the coil 15 is heated immediately by induction from coil 12. The heat from the coil 15 is conducted to the crucible

3, which in turn heats the metal button 1 by radiation, thus starting the melt. As soon as the metal begins to fuse, its conductivity increases, and the melting proceeds at a faster rate. The button or slug 1 may be "seeded" with uranium metal, that is, it may have small pieces of uranium metal dispersed therein, to facilitate the heating of the button by induction at the start of the melt.

Uranium cast by the herein described method will form a solid uranium casting without voids. Because the cast metal thus produced is very pure, it is ductile and easily worked.

Although a preferred method has been described, it will be understood that variations and modifications may be made in the method without departing from the scope of the invention as defined by the appended claims.

I claim:

1. The method of refining uranium comprising the steps of supporting a mass of uranium containing impurities upon a strainer in a crucible having an opening in the bottom thereof communicating with a mold, heating said mass until the uranium metal begins to flow therefrom vaporizing and removing at least a portion of the vaporizable impurities in said uranium, condensing the vaporized impurities on a cooled surface above and adjacent the mass of uranium containing impurities, passing the molten uranium through said strainer while retaining the non-vaporized impurities in said crucible, and collecting the molten metal in said mold, said heating and collecting being conducted in a high vacuum.

2. The method of refining uranium comprising the steps of supporting a mass of uranium containing impurities upon a strainer in a crucible having an opening in the bottom thereof communicating with a mold, heating said mass until the uranium metal begins to flow therefrom vaporizing and removing at least a portion of the vaporizable impurities in said uranium, condensing the vaporized impurities on a cooled surface above and adjacent the mass of uranium containing impurities, passing the molten uranium through said strainer while retaining the non-vaporized impurities in said crucible, collecting said molten uranium in the mold, said heating and collecting being conducted in a vacuum within a refractory envelope, and cooling the metal in said mold by introducing an inert atmosphere into said envelope, and by forcing air against the outer surface of said envelope to conduct heat therefrom.

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