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[54] **PROCESS FOR THE PREPARATION OF METALS OR ALLOYS**

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[58] **Field of Search** 75/10, 27, 84.1, 84.4; 13/27, 13/32

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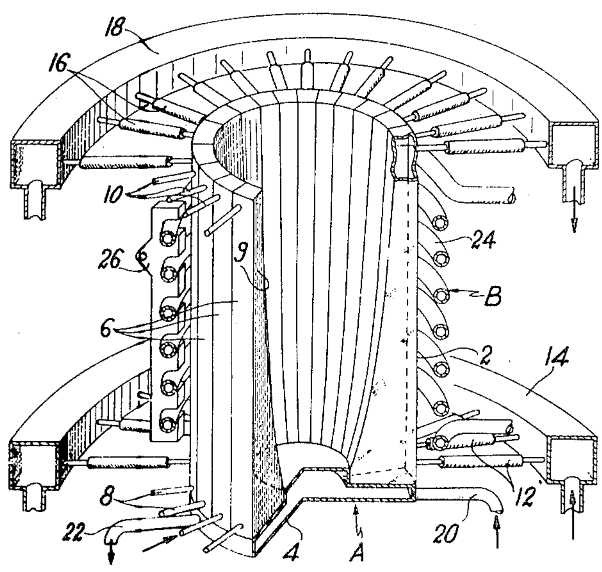
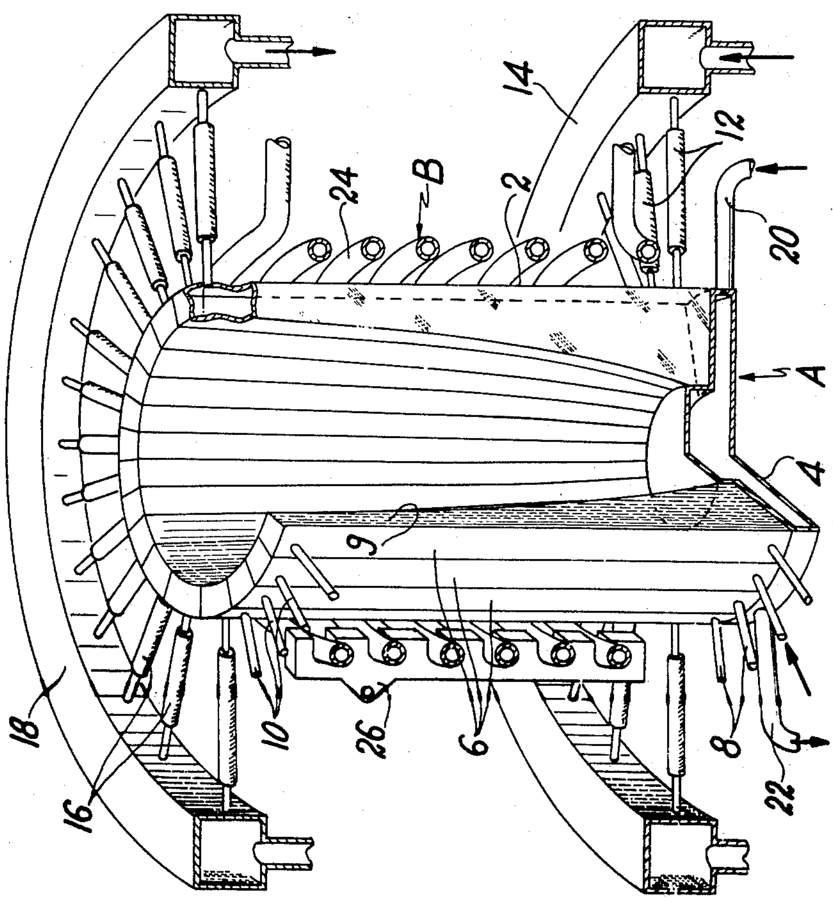
[57] **ABSTRACT**

A reaction mixture is placed in a furnace comprising a vertical vessel which is transparent to electromagnetic fields and in direct contact with the mixture. High-frequency electric currents are induced in said mixture through the vessel and are of sufficient intensity to start the reaction while the vessel is cooled so as to maintain a solid layer of slag in contact therewith. The reaction mass is maintained molten for a sufficient period of time to cause settling of the metal prior to solidification in situ. The vessel is throttled towards its base.

3 Claims, 1 Drawing Figure





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PROCESS FOR THE PREPARATION OF METALS OR ALLOYS

This invention relates to a process for the preparation of a metal or alloy by means of a reaction of the pyrometallurgical or metallothermic type in the presence of a slag in the molten state as well as to a device for carrying out said process. It must be understood that the term "slag" is intended to designate all substances which result from the reaction and which are capable of separating from the metal or alloy by settling.

Except in special cases, the reactions involved in preparation by metallothermic or pyrometallurgical processes have been carried out up to the present time in a crucible with which the reaction mass was in contact. The crucible must meet a certain number of conditions which depend on the properties of the metals to be obtained, on the cost of these latter and on the characteristics of the reaction. It can be mentioned in particular that:

- in the case of preparation of very pure metals, the crucible must be chemically inert with respect to the constituents which take part in the reaction;
- in the case of highly exothermic reactions, the crucible must be constructed of a material which has high resistance to thermal shock and a high melting point;
- in the case of preparation of costly metals, it must be possible to recover the metal or alloy which has penetrated into the crucible or to employ a crucible in which this penetration does not occur.

Moreover, it is evidently preferable to ensure either that the cost price of the crucible is low or that it can be used again. Finally, the crucible must be leak-tight and this is a function which can often be achieved only with difficulty in the case of ceramic crucibles which are formed of a number of assembled parts.

A problem which frequently arises is that no crucible can be found to satisfy all the conditions which are imposed by the reaction. One notable example of this is the preparation of plutonium or of enriched uranium metal by reduction of tetrafluoride. This process is carried out in a calcium fluoride crucible but this latter must subsequently be dissolved in order to recover the 2 to 4 percent of metal which have penetrated into the crucible walls. This requirement makes it necessary to dissolve 6 to 8 kg of calcium fluoride per kilogram of metal prepared. This complementary operation obviously has a heavy incidence on the cost price of the metal.

The aim of this invention is to provide an improvement over the processes of the prior art, especially insofar as the practical requirements referred-to above are complied with in a more effective manner. To this end, the invention proposes a process for the preparation of a metal or alloy by pyrometallurgical or metallothermic reaction in the presence of a slag, this process being characterized in that the reactants are placed in a furnace constituted by a vertical metallic vessel which is transparent to electromagnetic fields and by a winding placed around the vessel in coaxial relation thereto and intended to be supplied with high-frequency electric current, that high-frequency electric currents are induced in the reaction mass through the vessel and are of sufficient intensity to start the reaction while the vessel is cooled so as to maintain a solid layer of slag in contact therewith, that said current and said cooling are then regulated so as to maintain the reaction mass in the molten state for a sufficient period of time to cause settling of the metal or alloy and that the metal or alloy together with the slag are permitted to solidify in situ.

The advantages of this process are immediately apparent: the molten reaction mass is in contact not with a different substance but with its own slag which is maintained in the solid state by the cold wall of the vessel whilst heating is carried out by Joule effect within the midst of the reactants and slag and not by heat transmission through the vessel wall. In consequence, the main causes of contamination are removed and the yield is improved. The vessel can be used again a large number of times without intermediate maintenance or cleaning, which is a very appreciable advantage when the preparation process is carried out in a glove box.

In addition, the invention proposes a device for carrying out the process which has been outlined in the foregoing. Said device comprises a vessel having a vertical axis whose side wall is constituted by tubular metal segments adapted to carry a circulation of cooling fluid and a multiturn inductor coil which is coaxial with the vessel and placed around the wall, said device being especially characterized in that said side wall is delimited by an external cylindrical surface and by an internal surface which is throttled towards the base.

A clearer understanding of the invention will be gained from the following description of a device constituting one embodiment of the invention which is given without limitation and of a number of examples of preparation according to the invention which may be carried out in said device. Reference is made in the description to the single accompanying FIGURE which is a very diagrammatic perspective view of the device and which is partially broken away.

The device which is illustrated in the FIGURE can be considered as being constituted by a vessel A provided with ancillary components and by an indicator B which is associated with a high-frequency current generator (not shown).

The vessel A comprises a side wall as delimited by an external cylindrical surface and by an internal surface forming a space which is throttled towards the bottom in accordance with an arrangement which is comparable with that of a conventional crucible. The openings of the wall are intended to be closed by means of a base **4** and if necessary by a cover (not shown).

The side wall **2** is constituted by a large number of identical segments **6**, each segment being delimited by a portion of the external cylindrical surface, by a portion of the internal surface, by flat top and bottom faces and by lateral faces corresponding to planes which pass through the axis of the vessel. In order that two adjacent segments **6** should be insulated electrically, at least the lateral faces **9** are coated with a layer of insulating ceramic material which is applied by the spray-gun process, for example. After assembly, the segments thus constitute a side wall which is highly permeable to the electromagnetic field. Moreover, the total potential difference produced by induction through the side wall is subdivided in a large number of elements (this would not be the case if provision were made for only a single slot) and there is practically no danger of breakdown resulting from discharge into the reaction mass between two adjacent segments.

The base **4** is constituted by a cylindrical box which is also provided with an insulating lining and shaped in such a manner as to engage in a tight fit within the bottom opening of the side wall **2** in order to prevent leakage. It is apparent that the base **4** could correspond to the side wall and thus be made up of a series of sectors which are insulated from each other.

The side wall **2** and the base **4** are both equipped with a cooling system. Each segment **6** of the cold wall is provided with an inlet **8** for the admission of a liquid coolant at one end of the segment and with an outlet **10** for the discharge of coolant at the other end. All the inlets **8** are connected by means of flexible insulating joints **12** to an admission mainfold **14**. Similarly, all the outlets are connected to a manifold **18** by means of flexible insulating joints **16**.

In a like manner, the cooling system of the base **4** is provided with an inlet **20** for the admission of liquid and with an outlet **22** for the discharge of said liquid.

The inductor B consists of a multi-turn winding **24** which is placed in proximity to the side wall **2** and coaxially with this latter. The turns of said winding are fixed in position relative to each other by means of spacer members of insulating material, only one spacer member **26** being shown in the FIGURE.

Means (not shown) are provided for maintaining the segments applied against each other throughout the duration of the reaction. Said means can consist especially of a strip of insulating material which is wound tightly around the side wall. It would also be possible to clamp the top and bottom of the side wall by means of an insulating collar.

In the device which has just been described, the process in accordance with the invention is carried out as follows:

The base **4** having been placed in position, the reactants are intimately mixed and introduced into the vessel **2**. When the reaction mass has been placed in the vessel, the cooling system of the side wall **2**, of the base **4** and of the inductor coil is put into operation and the inductor coil B is energized. Heating is continued until start-up of the reaction. By reason of the fact that the peripheral portion of the mass is powerfully cooled by the wall **2**, the reaction will start first only in the central portion of the charge. When the reaction is exothermic as is usually the case, the electric power supplied to the inductor B can then be reduced to a very substantial extent and the reaction develops at a rate which can be controlled to a certain extent by regulating the power of the HF heating generator. Moreover, conversion of some constituents to the liquid state frequently results in an increase in electrical conductivity of the reaction mass and this also makes it possible to reduce the intensity of the current supplied to the inductor coil. The reaction takes place without any contact between the reaction mass and either the side wall or the base of the vessel since said mass remains separated from these latter by a thin layer of slag. Any contamination by the metal constituting the segments **6** (usually copper) or by the insulating product with which said segments are coated is thus prevented.

On completion of the reaction, the supply of electric current to the inductor coil is maintained for a period which is a function of the time considered necessary to ensure settling to a sufficient extent. Provision can evidently be made for conventional regulating devices in which a thermocouple is employed as a sensitive element and is immersed in the reaction mass.

When the entire operation has been completed (namely the reaction followed by settling), the supply of current to the inductor coil is cut off and cooling is maintained for the period of time which is necessary to solidify the mass as constituted by an ingot surrounded by a skin of solidified slag. Demolding will be carried out in some cases by removing the base and then displacing the ingot upwards, in some cases by separating the segments or in other cases by opening the wall which is designed in two disconnectable parts. Removal of the mold is facilitated by the presence of the solidified layer of slag which adheres only weakly to the metal walls of the vessel.

There will now be described by way of example a number of different modes of preparation of metals by means of the process according to the invention. Tests have been carried out in a device comprising a high-frequency generator having a rating of 40 kVA and an output frequency of 400 kc/s. The side wall of the vessel was constituted by segments having copper walls coated with alumina by the Schooping or spraygun process.

EXAMPLE 1

Preparation of uranium metal from tetrafluoride UF_4 in powered form and of calcium in granular form.

Quantities varying between 100 and 600 g of uranium were employed in the tests. The uranium recovered in the ingot represented between 98.6 and 99.7 percent of the uranium which was present in the tetrafluoride. The weight of waste material to be recovered was limited to that of the calcium fluoride which had formed. Quantitative analysis resulted in determination of the following impurities:

Cu : 10 ppm
Al : 30 to 40 ppm
Fe : 15 ppm

EXAMPLE 2

Preparation of cerium from powdered $CeCl_3$ and granular calcium.

Yields were higher than 90 percent.

EXAMPLE 3

Preparation of lanthanum from $LaCl_3$ in powdered form and calcium in granular form.

The charges consisted of 150 g of lanthanum and the yield was 90 percent.

In general, the process and the device according to the invention are wholly suitable for the purpose of obtaining metals which are very costly, have a high degree of purity and/or by means of reactions which need not be exothermic. This is particularly the case of metals which must have a sufficient degree of purity to permit their use in nuclear reactions and of very costly metals such as plutonium and highly enriched uranium. It is also worthy of note that the invention can very readily be adapted to application in a glove box and in a controlled atmosphere and therefrom to the preparation of either toxic or radioactive metals or or alloys.

What we claim is:

1. A process for the preparation of a metal or alloy by pyrometallurgical or metallothermic reduction of a chemical compound of said metal or alloy in the presence of a slag, comprising the steps of preparing a reaction mixture from said compound and a chemical reducing agent, placing the reaction mixture in a vertical metallic vessel transparent to electromagnetic fields and in direct contact with the mixture, inducing high-frequency electric currents in said mixture through the vessel for directly heating the mixture until the metallothermic reaction starts, and simultaneously positively cooling the vessel to maintain a solid layer of slag in contact therewith, regulating said current and said cooling to maintain the reaction mass in the molten state, maintaining said current and cooling until substantially all the metal or alloy has settled and then allowing the metal or alloy together with the slag to cool and to solidify in situ.

2. A process according to claim 1 for the preparation of plutonium or uranium by calciothermic reduction from PuF_4 or UF_4, including the step of preparing the reaction mixture by intimately mixing a powdered metal fluoride and granular calcium.

3. A process according to claim 1, said vessel having an internal surface throttled toward its lower end.

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