E. WEINTRAUB.

REDUCTION OF CHEMICAL COMPOUNDS.

APPLICATION FILED JULY 20, 1910.

1,019,394. Patented Mar. 5, 1912.

Fig. 1.

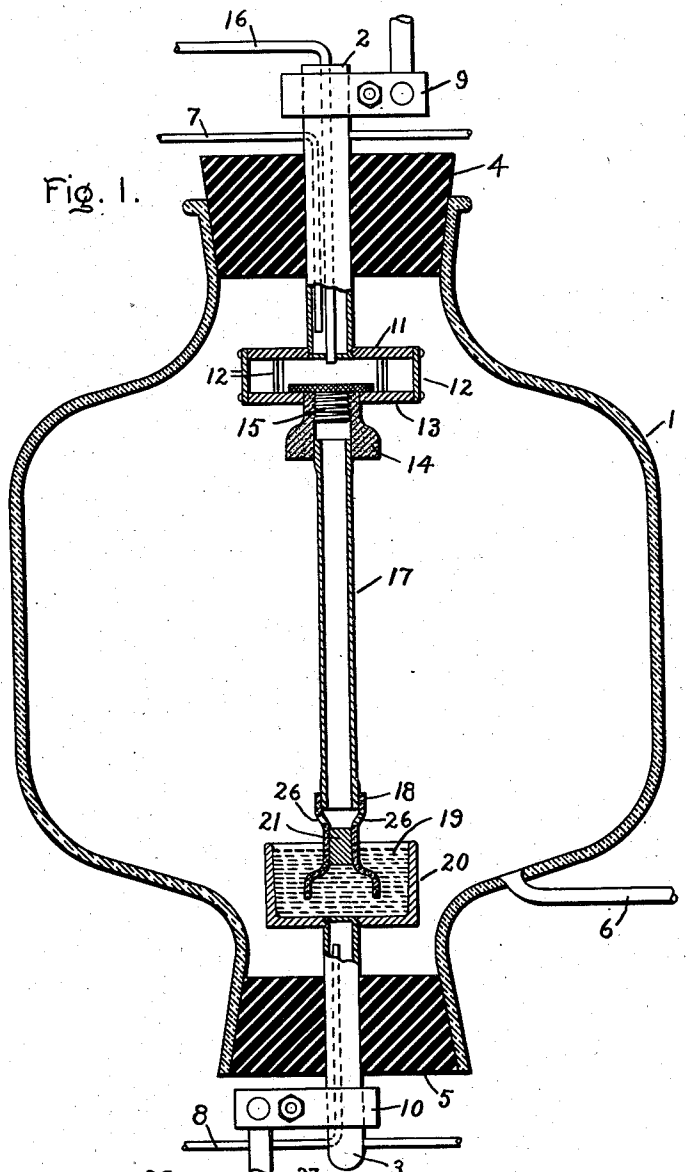
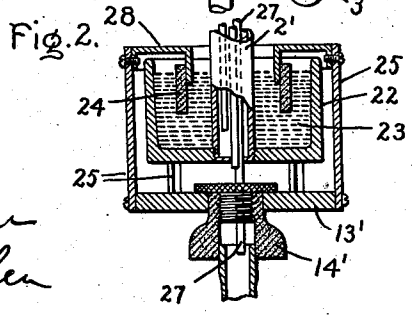


Fig. 2.



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UNITED STATES PATENT OFFICE.

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REDUCTION OF CHEMICAL COMPOUNDS.

1,019,394. Specification of Letters Patent. Patented Mar. 5, 1912.

Application filed July 20, 1910. Serial No. 572,830.

To all whom it may concern:

Be it known that I, EZECHIEL WEINTRAUB, a citizen of the United States, residing at Lynn, county of Essex, State of Massachusetts, have invented certain new and useful Improvements in Reduction of Chemical Compounds, of which the following is a specification.

My present invention relates to the reduction of metals and metalloids and comprises an improved method and apparatus suitable especially for the production in a pure state of elements such as boron, titanium, thorium, uranium, tungsten, molybdenum, and others.

According to my present invention, a mixture of a volatile reducible compound of the respective element to be reduced, as for example, a halid, is mixed with a suitable reducing gas such as hydrogen. The mixture is brought into contact with a thin refractory tube heated to the temperature of reduction. The reducing action thereby initiated results in the deposition of the element on the surface of the refractory tube. The reducible mixture is preferably brought into contact with the tube on its outer surface, the reduction being continued until a layer of substantial thickness has been deposited. The original tube of refractory material is then removed by mechanical means leaving a tube of reduced element. The reduction may, however, take place on the inner surface of the refractory tube and may be continued until the tube becomes practically filled with deposited material so that upon removal of the shell there is left behind a solid, or nearly solid, stick of the reduced element.

My invention is also applicable to the production of alloys which result from the addition of a volatile compound of the desired alloying element or elements to the reduction mixture, for example, the process may be employed for the production of boron carbon alloys as described in my application, Serial No. 524,950, filed Oct. 27, 1909. Alloys of different refractory metals may be produced in like manner by employing a mixture of volatile reducible compounds of the same, thereby depositing the metals simultaneously. The process is particularly applicable to the production of pure boron. The boron thus obtained is exceedingly hard, and very strong. With it glass may be cut with the same ease as with diamond, and even in thin sheets it can be broken only with difficulty. The process is also useful for the reduction of difficultly reducible metals, such as titanium in a state of high purity.

An apparatus suitable for producing the effects above outlined is shown in the accompanying drawings, of which—

Figure 1 is a sectional elevation of the reducing chamber and Fig. 2 shows in detail a modified clamp for the heater.

The reduction chamber comprises a gas-tight envelop, 1, which may conveniently consist of glass, provided at top and bottom with insulated terminals 2, 3, passing through rubber stoppers 4, 5, making gas-tight joints. The removal of air and other gases from the chamber takes place through the tube 6. The terminals are cooled by the passage of a suitable fluid such as water introduced by tubes 7, 8. Clamps 9, 10 serve to make the necessary electrical connections. The tubular terminal 2 is screwed into a copper plate 11 which is attached by connecting strips 12 to another metal plate 13. To this plate 13 a graphite clamp 14 is firmly attached by the screw 15 as clearly shown. The tube 16 ending between the two copper plates 11, 13 serves to introduce the reaction mixture which is then distributed by diffusing out between the plates 11, 13. The thin-walled heater tube 17 consists of carbon or graphite, prepared either by carbonizing tubes of paper, or by turning down carbon or graphite tubes to the desired thickness in the lathe. The tube 17 is conveniently attached to the clamps by friction and a small amount of carbon paste. It is attached at its lower end to a graphite clamp 18 dipping into a body of mercury 19 contained in a small vessel 20 supported by the terminal 3. A small platinum plug 21 attached to the clamp 18 is used to weight the lower end of the tube, thereby preventing its being broken by the upward pressure of the mercury. The mercury connection is used to enable the tube to readily expand when heated.

In some cases it is desirable to support the clamp carrying the upper end of the tube as well in mercury for the purpose of allowing free expansion and contraction. A modified form of connection designed to carry the upper end of the tube in mercury is shown in Fig. 2. The terminal 2′ in this case is screwed into a small container 22

containing a body of mercury 23 in which floats a graphite ring 24, attached to a plate 28, which carries copper strips 25. These copper strips in turn are attached to the plate 13′, which supports the graphite clamp 14′ holding the tube.

Means should be provided for equalizing the gas pressure in the inside of the tube at the beginning of the operation. This can be conveniently done by providing small holes as indicated at 26 in clamp 18 in Fig. 1 or by providing a tube 27, communicating with the inside of the tube as shown in Fig. 2. This tube can be attached to a supply of hydrogen, in order to allow the pressure to adjust itself and the connection may be later shut off when the equilibrium in gas pressures has been established.

In an apparatus as described above the process is carried out as follows: For the production of boron, for example, a mixture of reducible boron compound, such as boric chlorid, and hydrogen is introduced into the chamber 1 by means of tube 16, displacing the gas in the vessel, which may conveniently consist of hydrogen. Current is then supplied to the heater tube which is thereby heated to the temperature necessary to cause chemical action between the boric chlorid and hydrogen, resulting in the deposition of boron upon the surface of the tube and the formation of hydrochloric acid gas which is removed through the tube 6, together with unused hydrogen, or any unreduced boric chlorid which may remain. The reduction is continued until the desired thickness of boron has been deposited, the coated tube is removed and the material of the original tube, which has formed a brittle substance consisting of carbon and boron is removed mechanically.

When it is desired to deposit an alloy of boron and carbon on the tube, a mixture of boric chlorid, hydrogen and a volatile carbon compound such as gasolene or carbon tetra-chlorid is employed, the proportion of carbonaceous gas depending, of course, upon the amount of carbon desired in the alloy. This simultaneous deposition of the two elements is a convenient method of producing solid solutions, or alloys of carbon and boron of any desired predetermined proportion. As already indicated alloys of refractory metals can be produced in a similar manner by using mixtures of volatile compounds of the same with hydrogen, or other reducing gas.

In case the reduced element is to be deposited upon the inside of the tube, the gaseous mixture is admitted to the inside of the tube through pipe 27, Fig. 2, and the hydrogen used for washing the tube is admitted or withdrawn through tube 16. The deposit on the inside of the tube can be built up until the passageway becomes entirely filled. Thereafter the tubular envelop of carbon and carbid may be ground off, leaving a substantially solid stick consisting of boron or of boron associated with another element, in accordance with the chemical composition of the gaseous mixture originally supplied to the tube.

What I claim as new and desire to secure by Letters Patent of the United States, is,—

1. The process which consists in progressively depositing on the wall of a heated tube, a dense uniform coating which chemically unites with the material of said tube to form a brittle substance, and subsequently removing said brittle substance to leave a uniform body of deposited material.

2. The process which consists in depositing on a carbon tube heated to incandescence, a thick uniform layer of material capable of uniting with the carbon of said tube at the depositing temperature to form a brittle material, said deposition being effected from a mixture of reducible halid and hydrogen and subsequently removing said brittle material.

3. The process which consists in progressively supplying a mixture of boric chlorid and hydrogen to a carbon tube heated with current to the temperature at which reduction of the boric chlorid takes place, with deposition of boron on the wall of the carbon tube and chemical union therewith, and subsequently removing the combined carbon and boron to leave a dense uniform body of boron.

4. The process which consists in progressively applying a mixture of reducible gas and reducing gas to the inside of a thin-walled refractory tube heated to the temperature at which reduction occurs, continuing the operation until the tube is substantially filled with deposited material and then removing said tube to leave a body of pure deposited material.

5. An apparatus for the reduction of compounds comprising a thin-walled carbon tube, means for supplying current to said tube to heat the same to incandescence, said means including a connection permitting free expansion of said tube with thermal changes, and means for bringing in contact with said tube a mixture of reducible compound and reducing gas.

6. The combination of an envelop, fluid cooled electrical terminals therefor, a stationary carbonaceous tube arranged in circuit between said terminals, means of washing said tube with hydrogen and means for bringing into contact with said tube a mixture of gases chemically active on one another at an elevated temperature.

7. In a reducing apparatus, the combination of an envelop, a fragile heater tube mounted therein, means for conducting current to said tube to heat the same, said means

comprising a contact arranged to permit free thermal expansion of said fragile tube, means for bringing into contact with said tube a reduction mixture, and means for washing said tube with hydrogen.

8. In a reducing apparatus, the combination of an envelop, a liquid metal terminal therein, a fragile carbon heater tube having its lower end floating in said liquid metal, and means for supplying a reduction mixture to said envelop.

9. In a reducing apparatus, the combination of an envelop, a refractory heater tube mounted therein, means for supporting each end of the tube and conducting current thereto, one at least of said supporting means comprising a body of mercury arranged to allow free changes in length of said tube.

In witness whereof, I have hereunto set my hand this first day of July, 1910.

EZECHIEL WEINTRAUB.

Witnesses:
H. C. Coxe,
M. S. Hinland.