

March 17, 1959

HIROSHI ISHIZUKA
APPARATUS FOR CONTINUOUS VACUUM REFINING
OF SPONGE METALLIC TITANIUM

2,878,008

Filed July 22, 1955

4 Sheets-Sheet 1

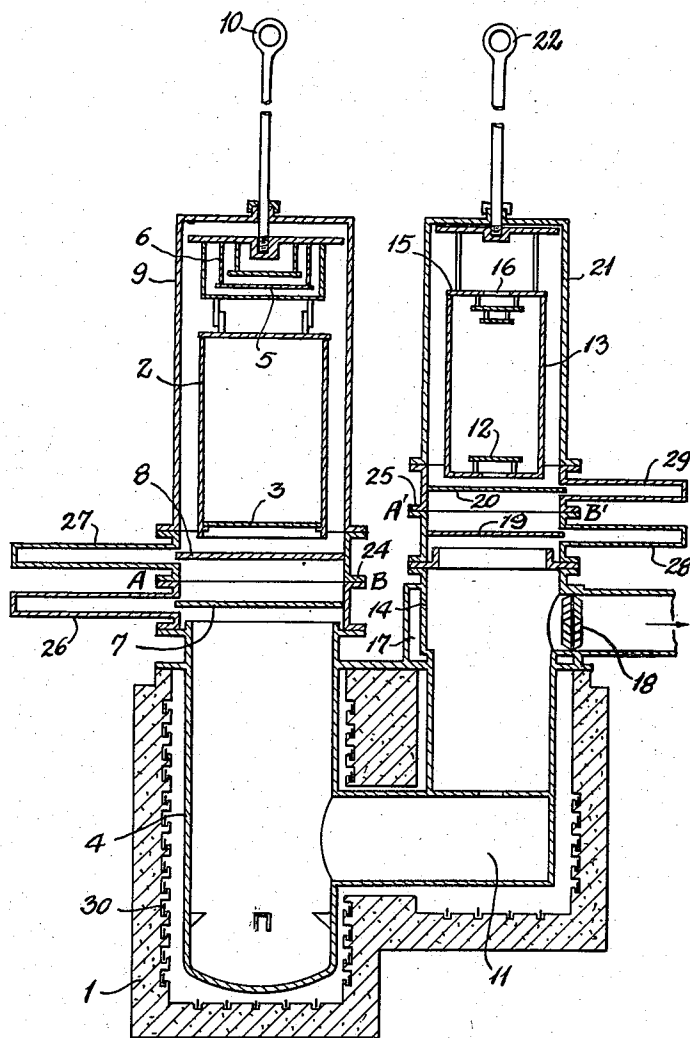


FIG. 1.

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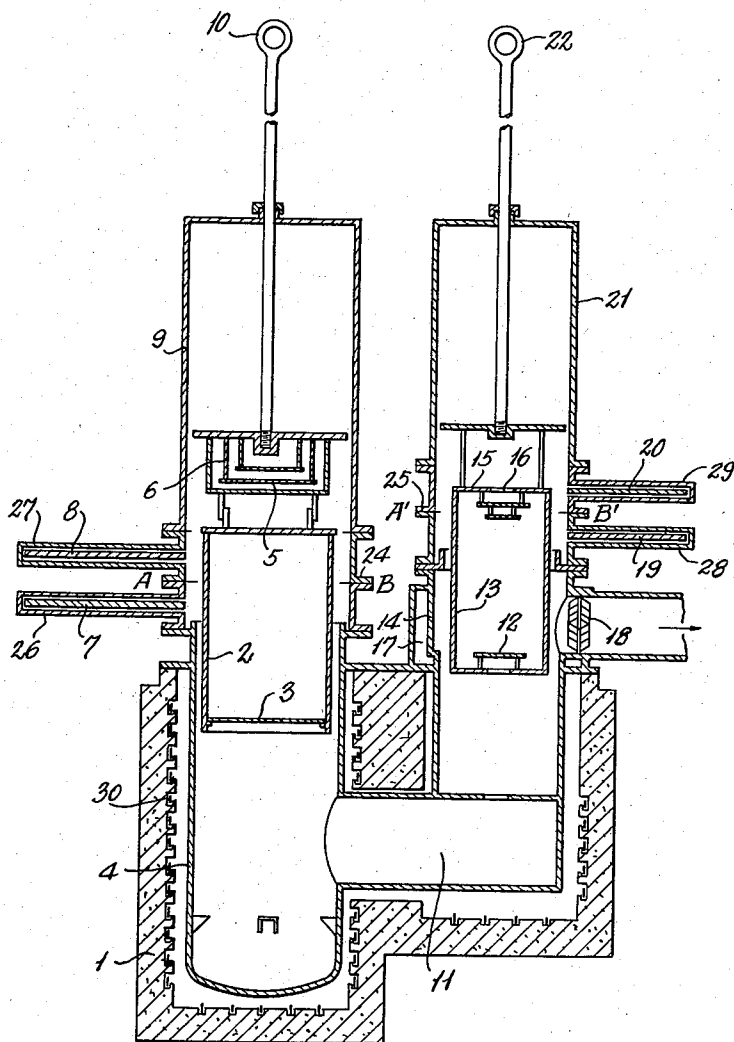


FIG. 2.

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4 Sheets-Sheet 3

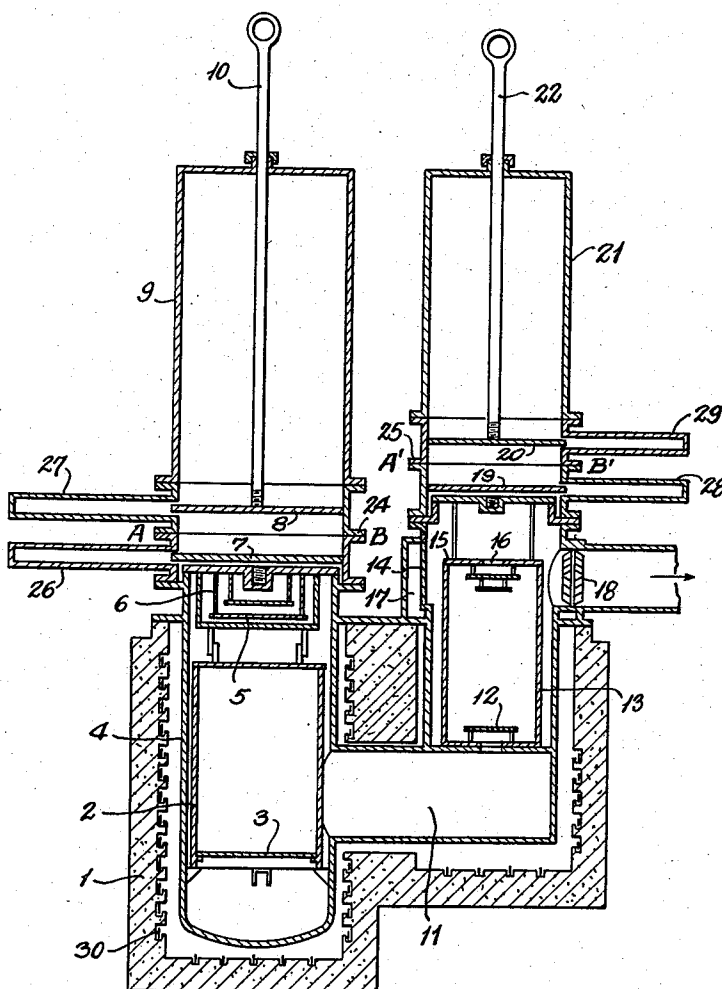


FIG. 3.

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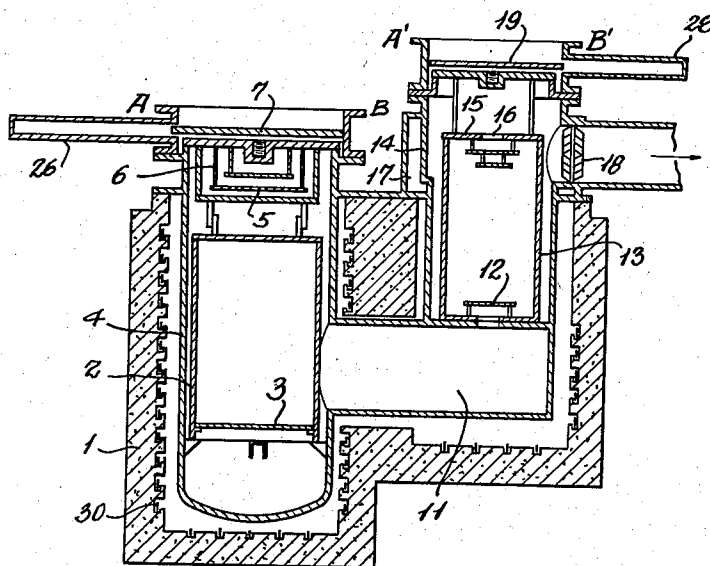


FIG. 4.

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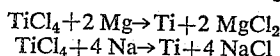
APPARATUS FOR CONTINUOUS VACUUM REFINING OF SPONGE METALLIC TITANIUM

Hiroshi Ishizuka, Ashiya-shi, Japan

Application July 22, 1955, Serial No. 523,859

2 Claims. (Cl. 266—16)

The present invention relates to an apparatus for refining spongy titanium continuously under vacuum, more particularly to an apparatus for the continuous production of high purity spongy titanium by a process wherein titanium tetrachloride (TiCl_4) is reacted with metallic magnesium (Mg) or sodium (Na) in a reaction cylinder in accordance with the following equations:



The spongy titanium formed by this reaction deposits in the reaction cylinder and is further treated at elevated temperatures under vacuum to separate completely the unreacted magnesium (Mg) or sodium (Na) as well as magnesium chloride (MgCl_2) or sodium chloride (NaCl) formed as by product of the reaction and which are present in the deposited spongy titanium.

The currently used process for producing metallic titanium industrially is performed by reacting TiCl_4 with Mg or Na at high temperatures in an inert gas atmosphere.

In this process, the above given reaction is carried out in a reaction cylinder, usually of iron, to collect therein spongy titanium which adheres to the inner wall of the reaction cylinder. In the case in which Mg is used as the reducing agent in the above given reaction, an excess of Mg is used. As a result, the spongy titanium obtained contains a small amount of unreacted magnesium admixed therewith. The magnesium chloride (MgCl_2) obtained as by-product of the reaction cylinder is preferably removed from the bottom of the reaction, but there still remains a small amount of it in the spongy titanium. This magnesium chloride has to be removed.

The usual practice for this removal is the vacuum separation process. The present inventor also carries out this practice. That is, in the usual process the reaction cylinder in which TiCl_4 and Mg are reacted together consists of an inner cylinder and an outer cylinder. Spongy titanium precipitates and adheres to the inner wall of the inner cylinder which is then cooled to a safe temperature. The cooled inner cylinder is then inserted into the vacuum separation cylinder. The vacuum separation cylinder is a top-sealed cylinder provided with a condensing portion underneath, i.e. an outer-separating cylinder. The said inner cylinder containing therein spongy titanium adherent to the inner wall, is first placed into said outer separating cylinder which is then evacuated to high vacuum, e.g. 10^{-5} to 10^{-6} mm. of Hg. Then, the outer-cylinder portion corresponding to the inserted inner cylinder is heated exteriorly in a heating furnace at elevated temperatures, for example, 900° to 1000° C. for more than twenty hours. The Mg and MgCl_2 contained in the spongy titanium are then volatilized and introduced into the lower condensing portion wherein they are condensed and solidified.

The resulting spongy titanium then is of a high degree of purity.

After completion of the treatment, first the heating furnace is removed and cooled sufficiently. Thereafter,

2

the outer-separating cylinder is removed, and the inner cylinder containing the adherent spongy titanium is shifted to peel off and collect the spongy titanium.

However, the spongy titanium is apt to come in contact with the ambient air during the above given operation and the large surface area of the spongy titanium is likely to be readily degraded by reacting with oxygen, nitrogen, hydrogen, etc. Thus, unless the spongy titanium is rapidly cooled, it is likely to burn rapidly.

This operation requires a long period of time and the ratio of installation to the production is high. In large scale production especially much reaction equipment, vacuum separation apparatus etc. is required.

This invention relates to an apparatus which permits the continuous vacuum refining of the titanium obtained from the reduction reaction. Its industrial value is apparent from its practical operation.

According to this invention, the reduction of titanium is performed in an apparatus whereby the reaction receptacle in which spongy titanium is adherent is brought to the vacuum separation apparatus, even at high temperatures before it is cooled to the temperature required by the prior art process. The said reaction receptacle is hung down within a casing kept in vacuum or in inert gas atmosphere.

Or, the reaction inner receptacle encased in the casing held at high temperature and in vacuum or in inert gas, e.g. argon atmosphere is lowered into the interior of the vacuum refining apparatus according to this invention.

In the accompanying drawings given as illustration of the invention:

Fig. 1 is a sectional elevation of the apparatus illustrating the first position of said apparatus, where the upper portions of two outer casings have just been placed respectively on the corresponding lower portions of the casings after two pairs of lids have been closed;

Fig. 2 is a similar view of Fig. 1 illustrating the second position of the apparatus, where the two inner receptacles are descending to the respective bottoms of the vacuum distillation chamber and the condensing chamber, after the two pairs of lids have been removed laterally to their casings;

Fig. 3 is a view similar to Fig. 1 illustrating the third position of the apparatus, where the two inner receptacles are set respectively in their operative positions, the two hanging rods having been detached from the respective top covers of the two inner receptacles and the two pairs of lids having been removed, ready for removing the two upper portions of the outer casings;

Fig. 4 is a view similar to Fig. 1 illustrating the fourth position (i.e. the working position), where the two upper portions of the outer casings are removed and the distillation and the condensing apparatuses are in operation. (After completion, the above order is reversed.)

Referring to the drawings, for example, the inner receptacle casing 9 holding the inner reaction receptacle at a high temperature in an inert gas atmosphere, is placed on flange 24; the inner receptacle 2 is lowered by actuating a lateral hanging metal 5 as well as a longitudinal hanging metal 6 and a hanging rod 10 and put into an outer separation receptacle 4. The outer receptacle 4 is held in a heating furnace, and is adapted to be heated to the required temperature.

A bottom plate 3 for the inner receptacle is provided in the lower portion of the inner receptacle 2. Spongy titanium adheres to the inner wall of the inner receptacle above the bottom plate and forms in lump form.

The inner receptacle 2 having been prepared for the vacuum distillation treatment, that is, immediately after completion of the reduction reaction, is suspended by means of hanging rod 10 (for inner receptacle) in the

casing 9, is made air tight with bottom lid 8 at the bottom, is transferred from the reduction procedure and attached in an air-tight manner on the outer receptacle 4 above line A—B. Then the bottom lid (for inner receptacle) is withdrawn and placed in bottom lid chamber 27 provided laterally, and an outer receptacle lid 7 is shifted to the corresponding position in the air-tightened outer receptacle lid chamber 26 arranged laterally above outer receptacle 4. The inner receptacle 2 is then lowered as illustrated in the drawing by actuating hanging rod 10 and placed into outer receptacle 4. The outer receptacle lid 7 is then moved to make a sufficiently air-tight seal. Then the vacuum separation is carried out.

After the vacuum separation treatment has been completed, the outer receptacle lid 7 is opened and the inner receptacle is raised into the inner receptacle casing 9 mounted above. The inner receptacle is sealed with the bottom lid 8 for said casing and then it is separated along line A—B and is shifted for the following step.

A suction gas flue is provided on the side wall of outer receptacle 4, which leads to an inner condensing receptacle 13. In said inner condensing receptacle 13, a condensation-plate 12 is provided so that gases introduced by suction may be set sufficiently in the condensing receptacle 13 and condensed and that the condensate may be prevented from falling in the suction flue 11.

An outlet 16 for suction gas is provided on the inner receptacle lid 15 for condensing on top of condensing inner receptacle 13, and the inner receptacle 13 is appropriately suspended by means of hanging rod 22, thus being adapted to be taken out of or inserted into the condensing receptacle 14.

The condensing receptacle 14 can be cooled sufficiently by a condensation cylinder 17.

In the gases separated by sufficient condensation in the condensing inner receptacle 13 in a condensation receptacle, there is practically no solid substance, but in some cases presumably there may be a small quantity of solid materials. In order to avoid the above-said contamination, a lamellar preventing sheet 18 is provided, through which gases are passed and led to the vacuum pump.

When the sufficiently condensed and separated mixture of Mg and $MgCl_2$ has been sufficiently deposited in the condensation inner receptacle, the inner receptacle is required to be exchanged. For this purpose, a condensation inner receptacle casing 21 is provided, wherein the condensation inner receptacle is suspended in air-tightness. The condensation inner receptacle casing 21 is placed on the flange 25 on line A'—B'. Then the condensation inner receptacle is suspended in said casing to be removed. A new inner receptacle is then suspended and inserted.

For example, in order to withdraw the inner receptacle 13 containing Mg and $MgCl_2$ adherent therein, out of the condensation receptacle 14, the condensation inner receptacle casing 21 is mounted along line A'—B' and a vacuum is created in said casing. The lid 19 for condensing inner receptacle casing 21 is shifted into the lid chamber 28 provided laterally with a condensation inner receptacle lid which is arranged in air-tightness on the inner condensation receptacle and accommodated in the position of inner condensation receptacle lid 19. The inner condensation cylinder 13 is attached after the hanging rod 22 is inserted and then is received by the inner condensation receptacle casing. The lid is air-tightly sealed by shifting the casing bottom lid 20 in the bottom lid chamber 29 for the inner condensing receptacle casing, the casing whereupon is separated on the line A'—B' and is removed.

Then, the inner condensing receptacle casing containing a new inner condensing receptacle is set on line A'—B' above the condensation receptacle, the bottom lid for inner condensation receptacle is opened and the conden-

sation receptacle lid is opened, the inner condensing receptacle is lowered and fixed and then the lid is shut and the condensation is further continued.

Thus, the inner receptacle 2 having spongy titanium admixed with Mg and $MgCl_2$ is put into the outer receptacle 4. When heating is carried out at high temperatures, e. g. 900° to 1000° C. in high vacuum, for instance, of 10^{-5} to 10^{-6} mm. of Hg, Mg and $MgCl_2$ are volatilized and enter into the inner condensation receptacle 13 through suction flue 11 and are condensed and adhere in the inner condensation receptacle 13.

Heating elements 30 have been added in several locations in the drawings for the sake of illustration.

According to this invention, as above-said, the reaction receptacle in which spongy titanium adheres can be transferred even while being kept at a high temperature in an inert gas atmosphere, directly to the apparatus for the vacuum refining and fixed. After high vacuum and high temperature treatment has been completed and the admixed $MgCl_2$ has been gasified and separated, the spongy titanium is withdrawn and shifted adherent in the reaction receptacle at a high temperature before it has been cooled to a low temperature and the operation can thus be carried out continuously. Moreover, the thus gasified Mg and $MgCl_2$ are condensed in a connected condensation receptacle and are condensed and deposited into a movable receptacle which is withdrawn and charged. The vacuum separation is, thus, performed continuously.

According to the present invention, the charging and withdrawal of the inner receptacle for the vacuum separation can be carried out rapidly in vacuum or inert gas atmosphere and can also be performed at elevated temperatures by putting the inner receptacle containing spongy titanium therein adherent into the vacuum separation receptacle and by performing the charging and withdrawal of said inner receptacle under exclusion of contact with atmospheric air. And as the inner receptacle is withdrawn and a fresh receptacle is replaced, after the vacuum treatment has been completed, the operation can be made continuously with out contact with the outside air.

Moreover, the condensation of vacuum separated Mg and $MgCl_2$ is effected in a connected condensation receptacle. The shifting of the receptacle, i. e., the condensation receptacle containing Mg and $MgCl_2$ condensed therein and adherent thereto, can be carried out continuously without allowing contact with the outside air as in the above mentioned case relative to the inner receptacle.

The present invention relates to an apparatus wherein the usual batch procedure is carried out continuously and rapidly while avoiding the deterioration of the quality of the products. With this apparatus, a high grade product can be easily obtained industrially and economically. According to this apparatus, the absorption of O, N and H_2O does not occur, the purity of product is high, the damage of pumps due to the presence of HCl is low, economy in heat, labor and time can be achieved and the mass production of uniform products is assured at high yield. The apparatus according to this invention can be used for the vacuum separation not only in connection with the reduction of $TiCl_4$ by Mg but also with the reduction by means of Na.

The present invention which is applicable also to other similar metals enables the continuous operation which has heretofore been considered impossible and aims particularly to provide continuous production of titanium especially in vacuum refining. Thus the great industrial value of the present invention is readily apparent.

What I claim:

1. Apparatus for continuous vacuum distillation which comprises, in combination, a distillation apparatus provided with an outer casing to accommodate therein an

5

inner receptacle in which the mixture to be subjected to vacuum distillation is contained therein adherent to the wall, a pair of lids provided above said inner receptacle when said inner receptacle is in its operational position within the distillation apparatus, said pair of lids being adapted to keep said outer casing air-tight, and a separate condensation apparatus connected to said distillation apparatus, said condensation apparatus comprising an outer casing to accommodate therein an inner receptacle, adapted to receive substances which have been gasified by the vacuum distillation performed in said vacuum distillation apparatus, a pair of lids provided above the last mentioned inner receptacle when said last mentioned inner receptacle is in its operational position within the condensation apparatus, said last mentioned pair of lids being adapted to keep the last mentioned outer casing air-tight, and there is provided a vacuum pump for evacuation, whereby said distillation apparatus and said condensation apparatus are connected together, means adapted for mounting thereon an inner receptacle encased in an inner receptacle casing and means for introducing said inner receptacle into said distillation apparatus while maintaining exclusion of air from said distillation apparatus.

2. Apparatus for continuous vacuum distillation which comprises, in combination, a distillation apparatus provided with an outer casing to accommodate therein as inner receptacle in which the mixture to be subjected to vacuum distillation is contained therein adherent to the wall, a pair of lids provided above said inner receptacle

6

when said inner receptacle is in its operational position within the distillation apparatus, said pair of lids being adapted to keep said outer casing air-tight, and a separate condensation apparatus connected to said distillation apparatus, said condensation apparatus comprising an outer casing to accommodate therein an inner receptacle, adapted to receive substances which have been gasified by the vacuum distillation performed in said vacuum distillation apparatus, a pair of lids provided above the last mentioned inner receptacle when said last mentioned inner receptacle is in its operational position within the condensation apparatus, said last mentioned pair of lids being adapted to keep the last mentioned outer casing air-tight, and there is provided a vacuum pump for evacuation, whereby said distillation apparatus and said condensation apparatus are connected together, means for air-tightly encasing an inner receptacle in a casing so as to permit the removal of said inner receptacle encased in its casing from the distillation apparatus while maintaining exclusion of air from said distillation apparatus.

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