

Aug. 3, 1926.

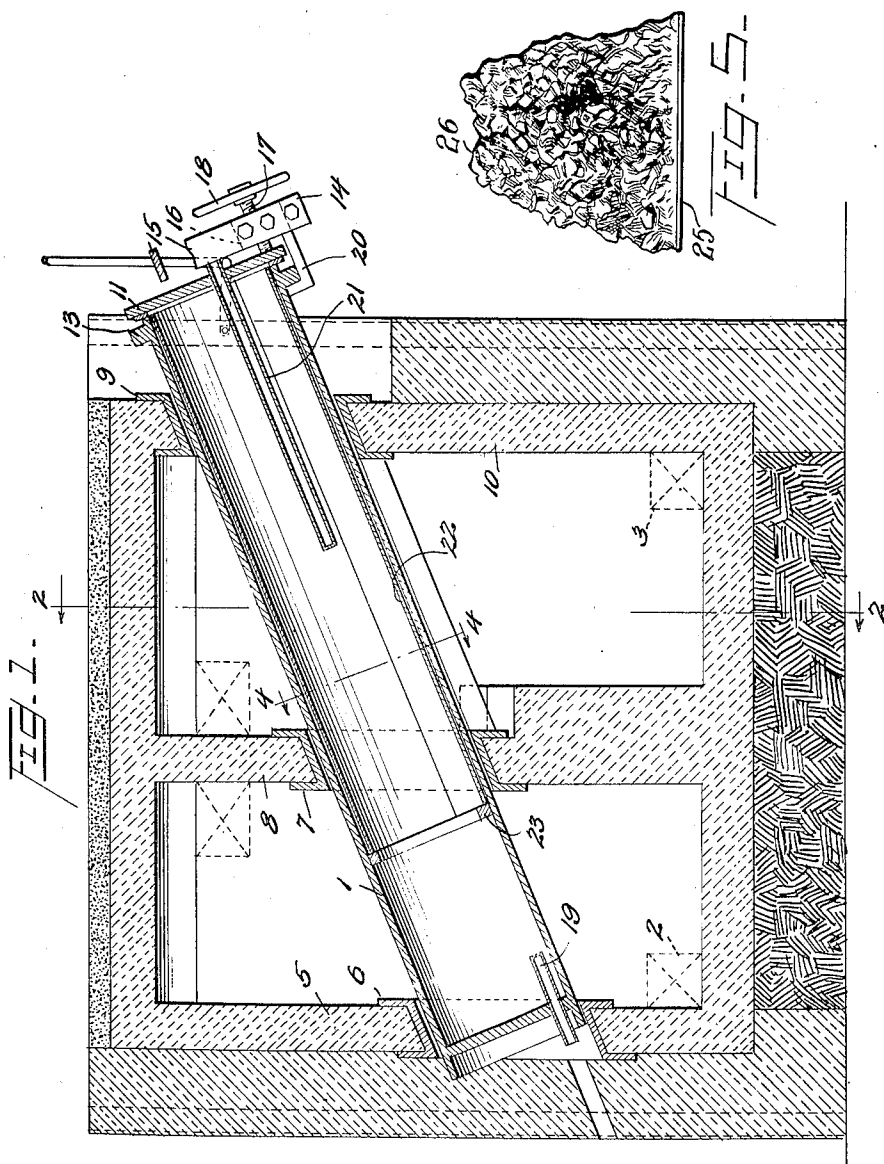
1,594,346

H. E. BAKKEN

MAGNESIUM PRODUCT

Original Filed Nov. 1, 1922

2 Sheets-Sheet 1



H. E. Bakken ^{Inventor}
By John Boyley _{Attorney}

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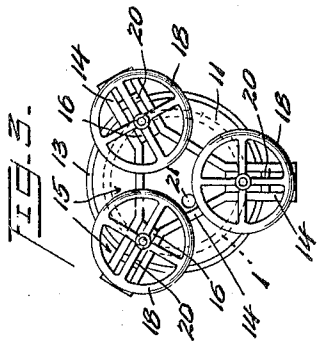
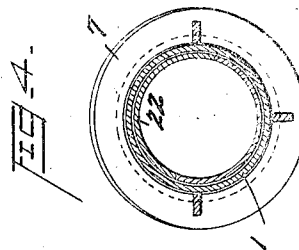
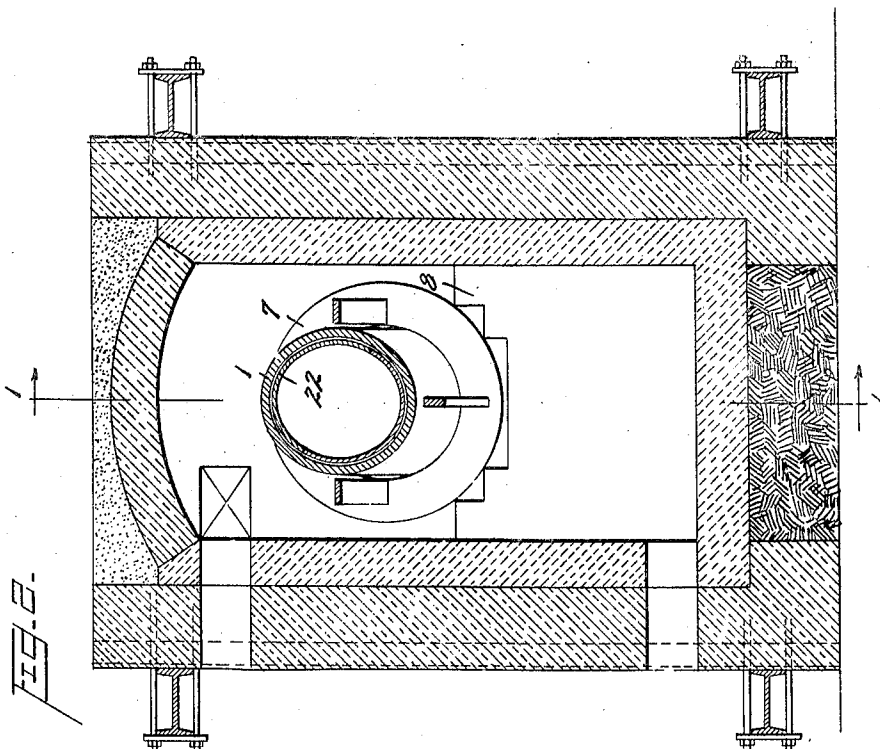
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2 Sheets-Sheet 2



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UNITED STATES PATENT OFFICE.

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MAGNESIUM PRODUCT.

Original application filed November 1, 1922. Serial No. 598,292. Divided and this application filed December 6, 1924. Serial No. 754,415.

My invention relates to the production of metallic magnesium in a substantially chemically pure state and in such a unique form that its use in subsequent processes of manufacture is greatly facilitated. In the processes that are at present practiced, magnesium, when produced from ores containing other metals and metalloids, is likely to be contaminated with them. For use in the mechanical arts, these impurities should be removed, as it is found that they exert a marked influence on the physical and chemical properties of the metal even when present in only a limited degree. Some prior processes for producing pure magnesium involve the production and condensation of magnesium vapor. The vapor is condensed either in liquid or powder form depending on the conditions of condensation. If condensed in liquid form, impurities such as sodium, which may travel into the condenser from the metal being vaporized, are likely to be present in the molten liquid. If condensed in the form of powder, further manipulation of the product involves difficulties. Powder is very difficult to melt without excessive losses and is very easily oxidized, due to the large amount of surface exposed.

My process is particularly adapted for use in refining crude metal. It is applicable also to obtaining pure metal from magnesium scrap, such as castings, alloys, etc.

This application is a division of my application Serial No. 598,292, filed November 1, 1922, wherein I have described and claimed my said process.

My invention comprises the purification of magnesium by converting the solid metal directly into the state of vapor and then condensing it directly into the crystallized state without intermediate liquefaction, and in particular it consists in the discovery of suitable means and conditions whereby such purification can be made practically operable and capable of being commercially carried out so as to produce pure magnesium at a reasonable cost, and in the discovery and production as a new commercial product, of a new form of magnesium of high purity, easily capable of further manipulation.

It is known that magnesium boils at atmospheric pressure at about 1120° C. Purification by distillation at this temperature,

however, is in practice very difficult because of the fact that the necessary apparatus deteriorates rapidly at such a high temperature. By reducing the pressure in the apparatus, i. e. by carrying on the distillation under diminished pressure, the boiling point of the metal can be reduced, until it coincides with the melting point at a pressure of approximately 2 millimeters of mercury, thus reducing the difficulty found in producing suitable apparatus of a sufficient size to handle commercial quantities of metal, and strong enough to satisfactorily withstand the external pressure involved.

I have discovered, however, that if the pressure inside the apparatus is still further reduced, say to less than about 1 mm., rapid sublimation of the magnesium may be accomplished at temperatures which are within the range where ordinary steel apparatus will function satisfactorily. In order to obtain rapid sublimation, it is necessary to so adjust or regulate the operating pressure that the vapor pressure of the magnesium at the temperature of the solid metal being sublimed is slightly greater than the total pressure within the condenser. Under these conditions, the evolution of magnesium vapor will be rapid and if the proper condensation facilities are provided the rate of sublimation is only limited by the rate at which heat can be supplied to the metal being sublimed. This point at which the vapor pressure of the solid magnesium becomes greater than the absolute pressure of the system is hereinafter called the "sublimation point."

By virtue of operating at or above the sublimation point, I not only produce rapid sublimation, but by the rush of magnesium vapor toward the condenser I sweep out and away the residual amount of air or other gas in the apparatus, and thus reduce very much the tendency of this gas to combine with the magnesium and render the product impure.

One of the great advantages of my method of sublimation as compared with vacuum distillation of magnesium is that not only can it be made to be as rapid as the distillation but it also gives a considerably purer product. In vacuum distillation I have

found that no matter how carefully the distillation is carried on, there is always a certain proportion of non-volatile impurities carried over into the condensed metal. I ascribe this phenomenon to the well-known tendency of gases or vapors to carry with them minute droplets or spray of any liquid through which they pass or from which they are evolved, particularly if they are evolved rapidly. This spray of impure magnesium carried over from the boiling liquid contaminates the distillate, but its formation appears to be prevented by subliming rather than distilling the magnesium so as to avoid the phenomenon of the evolution of bubbles from a liquid.

It will be apprehended that the process is capable of being carried out in a number of different types of apparatus and that the one shown and described is merely illustrative of the principles of the invention. This embodiment, however, has been found to be practical in construction and operation and efficient in use.

Referring to the drawings for a more complete disclosure of the invention:

Figure 1 is a vertical longitudinal section of the furnace;

Figure 2 is a section on the line 2—2 of Figure 1;

Figure 3 is a detail of the cover and closing mechanism for the retort;

Figure 4 is a section on the line 4—4 of Figure 1; and

Figure 5 is a view showing a full size fragment of my new product.

The iron retort 1 is suitably supported and inclosed in a brickwork and may be heated by burners projecting through the openings 2 and 3 in the lower corners of the combustion chamber, or in any other suitable manner. The retort at its lower end is supported by the vertical brick wall 5 and by the wall collar 6. Intermediate the ends, it is supported by a wall collar 7 carried by the center wall 8. At its front end it is supported by the wall collar 9 carried by the front wall 10. In such a construction, the greater part of the retort is in the zone of heat, the remainder extending out into the air to provide a cold end therefor, which may be provided with cooling coils.

The charging end of the retort is closed by a suitable door 11 which is so constructed as to make the same as gas tight as possible. A suitable construction comprises lugs 13 integral with the retort and a series of hooks 20 welded to the lugs and clamped between the bars 14 of the framework 15. Carried between the said bars are screw blocks 16 which engage with the screws 17 of the hand wheels 18. Rotation of the said wheels in a suitable direction will loosen or tighten the cover of the retort.

For determining the temperature at the

bottom of the retort a pyrometer well 19 is provided and a similar pyrometer well 21 is provided at the top of the retort.

For facilitating removal of the deposited crystals, a liner 22 is located in the cooler end of the retort.

This liner consists of a pipe somewhat smaller than the inside diameter of the retort, so that it can be easily slipped in and taken out. The liner is open at both ends, and at its lower end rests on the annular ring 23, integral with the pot, the other end extending up to the closing flange. The purpose of the ring 23 is to keep the liner from slipping down into the retort and also to form a seal so that gaseous magnesium does not travel up between the liner and the retort wall. The liner may be made in one piece or split longitudinally into two halves, to facilitate removal of the deposited crystals.

Operation.

The pot is charged with a suitable quantity of crude, alloyed or impure magnesium, sealed and vacuum applied. When the manometer shows that the desired degree of vacuum has been obtained, heat is applied, care being taken to maintain the vacuum. After a period of time, the temperature will rise to about 600° C. Under the residual pressure employed the temperature will remain practically constant at about 600° C. Further application of heat merely increases the rate of sublimation and does not appreciably change the temperature of the metal being sublimed. Thus it is impossible to melt the metal with any reasonable application of heat. When sublimation is complete as indicated by an abrupt rise in temperature, the source of heat is cut off and the retort allowed to cool while still under the same vacuum.

Under the conditions outlined, the magnesium does not melt but passes directly from the solid into the vapor state. The vapor passes into the cooler portion of the retort where it condenses on the liner. When opened, the inner surface of the liner will be found to be covered with crystallized magnesium in the form of a loosely cohering mass of aggregated crystals which can be removed in a suitable manner. Analysis of the residue in the bottom of the retort will show that nearly all the magnesium has been vaporized. Analysis of the crystals of magnesium will show that the magnesium content may be as high as 99.989%.

The best available data indicate that the vapor pressure of magnesium decreases from one atmosphere or 760 mm. of mercury at 1120° C., which is ordinarily spoken of as its boiling point, to approximately 2 mm. at its melting point, 651° C. Below the melting point the vapor pressure de-

creases so that after a further drop of about 270° C. it is about 0.001 mm. If crude, impure, or alloyed magnesium is heated at a temperature below its melting point, the magnesium will vaporize so as to produce a partial pressure of magnesium vapor, determined by the temperature. If this heating is done in the presence of an inert gas such as argon, at atmosphere pressure or under diminished pressure, but still at a total pressure greater than the vapor pressure of magnesium at the temperature employed, this magnesium vapor will diffuse through the inert gas; and if another part of the apparatus is cooler than the metal being heated, magnesium will gradually condense in this portion of the apparatus. This rate of sublimation is very slow, but I have found that if the total pressure within the condenser is reduced to a point below the vapor pressure of the magnesium at the temperature employed (as measured at 19, Fig. 1), rapid sublimation takes place. Instead of having to slowly diffuse through the inert gas present, the magnesium vapor will then be rapidly evolved at a sufficient pressure to sweep this gas away and into the condenser; where, by a suitable regulation of the condenser temperature, I continuously condense this vapor in the form of a very pure crystallized mass. In carrying out my process, solidification of the magnesium vapor without crystallization may occur if the condenser is too cold. The temperature of the condenser, therefore, must be high enough to permit continued condensation and re-evaporation of the molecules such as is necessary in the production of crystals from any vapor.

It is, of course, necessary that the temperature in the condensing area should be below that of the metal being sublimed, since the difference in vapor pressure due to this difference in temperature is the driving force causing the rapid transfer of the magnesium vapor from the subliming to the condensing end of the system. This difference in pressure must be equal to the sum of the partial pressure of the inert gas present in the condenser and the pressure required to overcome the frictional resistance and force the vapor from the subliming to the condensing end. This importance of these factors is seen by the fact that calculation shows that if magnesium be sublimed in an apparatus such as the one shown in Fig. 1, having a cross-sectional area of one square foot at the section 4—4, at a temperature of 600° C. and a pressure of 0.75 mm., the linear velocity of the magnesium vapor which would be necessary to sublime 100 pounds of magnesium in 24 hours would be in excess of one-half mile a minute. Diffusion, such as would take place if the total pressure within the apparatus were greater than the vapor

pressure of magnesium at the temperature employed, is known to take place relatively slowly, and it is evident that my method of sublimation of magnesium will be much more rapid than one which depends upon diffusion.

The temperature of sublimation and conversely of solidification may vary from approximately 300° C. at .001 mm. pressure to 651° C. at approximately 2 mm. pressure. The most favorable operating conditions at the present time have been found to be a temperature of approximately 600° C. and a pressure of 0.5 to 0.2 mm. Under such conditions I am able to sublime 100 pounds of magnesium in 4 hours in an apparatus having a cross-section of approximately one square foot.

When the liner is removed it is found that impurities, such as sodium, which are more volatile than magnesium and which therefore vaporize and go into the condenser, are segregated at the upper or cooler end of the liner and may be readily removed.

The specimen shown in Figure 5 is a fragment of the crystallized mass which adheres to the surface of the liner along the line 25. The surface 26 may show fully developed crystal faces of magnesium. The crystals vary in size and some may have crystal faces as large as one-fourth of a square inch in area or more. Being coherent, the masses can be readily handled but since they are only loosely coherent, they can be readily broken into pieces suitable for subsequent manufacturing operations.

In using the term "sublimed crystals of magnesium" in the claims, I have reference to a product produced by a process wherein the crystals are quite free to grow in part, and where during this growth the faces of many of the crystals assume their typical external shape.

Magnesium which has solidified from the liquid state is crystalline but not crystallized. It is composed of closely coherent grains having a crystalline structure and which while forming were not free to grow and assume their typical external shape.

Crystallized metal as produced by my process is exceedingly pure. By virtue of the absence of reactive agents such as nitrogen and oxygen, the crystallized mass is substantially free from admixed oxides or nitrides.

The crystallized metal can be subjected to various further operations. It can be satisfactorily melted or it can be placed without melting in an extrusion press and extruded directly into any of the customary structural shapes such as wire, bars, rods, etc., having physical properties equivalent to those of similar shapes extruded from cast billets. The crystallized metal can also be com-

pressed directly into briquets at a temperature of about 200° C. or even lower if sufficient pressure is available. Both the direct extrusion and direct compression give a substantially pure metal in marketable form. Such metal is substantially free from admixed magnesium oxide and magnesium nitride which might not be the case if the metal were melted before being extruded or compressed.

On account of its extreme purity, crystallized magnesium as made by my process is more highly resistant to corrosion by moisture or the atmosphere than any other commercial magnesium of which I have any knowledge. This resistance to corrosion is maintained through any subsequent manipulation or process which does not allow the formation or introduction of impurities.

I claim:

1. As a new commercial product, a mass of sublimed crystals of magnesium.

2. As a new commercial product, coherent aggregates of sublimed crystals of magnesium.

3. As a new commercial product, loosely coherent aggregates of sublimed crystals of magnesium.

4. As a new commercial product, a mass of sublimed crystals of magnesium, substantially free from non-metallic impurities.

5. As a new commercial product, a mass of sublimed crystals of magnesium containing less than 0.1% of other metals.

6. As a new commercial product, a mass of sublimed crystals of magnesium containing less than 0.02% of other metals.

In testimony whereof I affix my signature.

HERMAN E. BAKKEN.