

Jan. 27, 1970

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VAPORIZING CRUCIBLE

3,491,992

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3 Sheets-Sheet 1

Fig_2

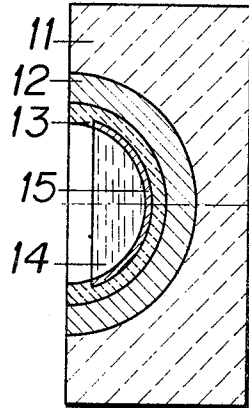
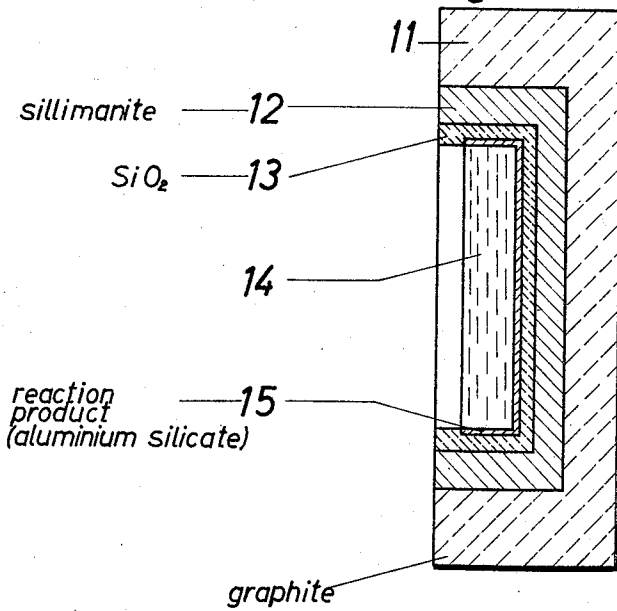


Fig_1



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Fig_6

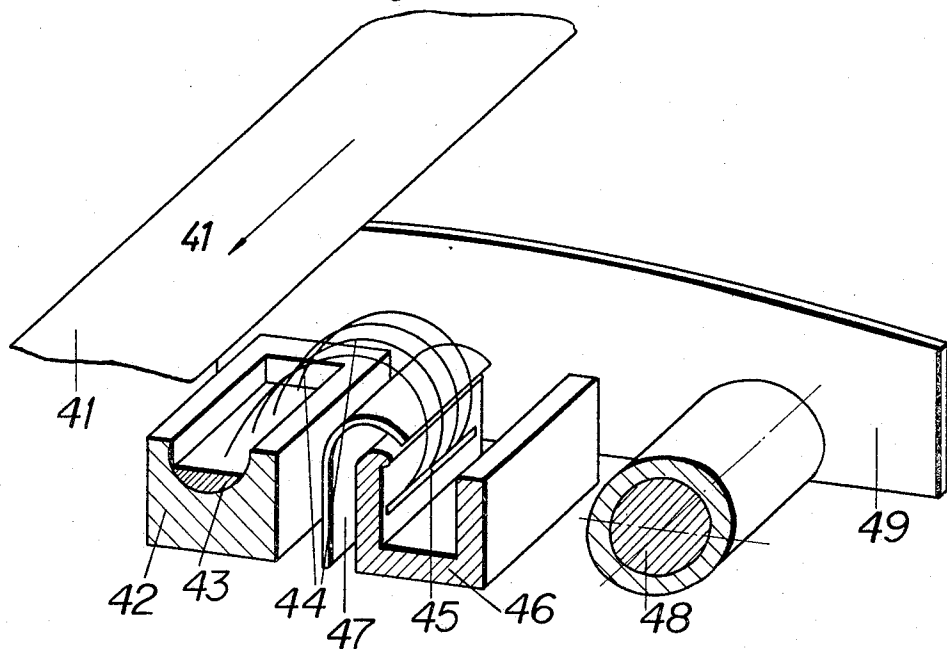
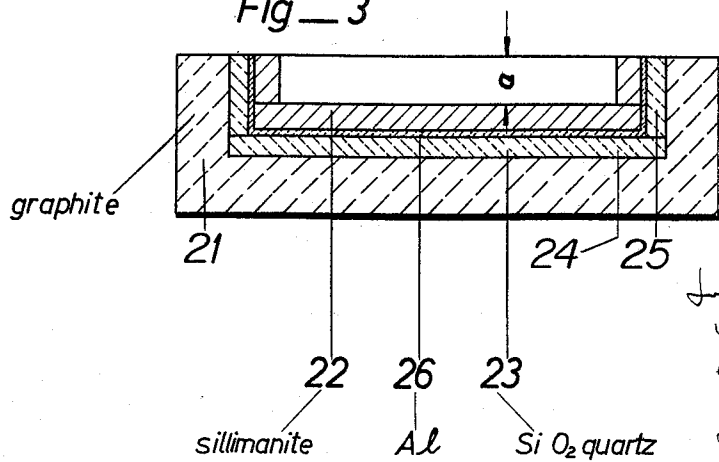


Fig_3



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Fig_5

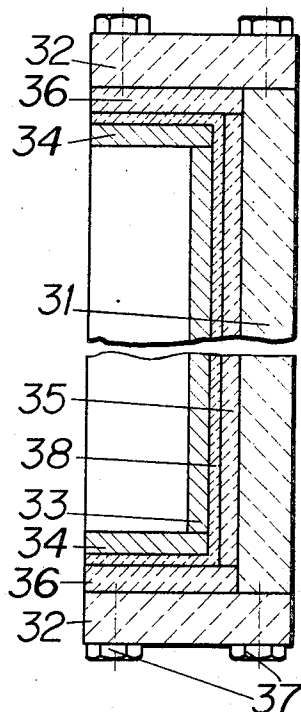
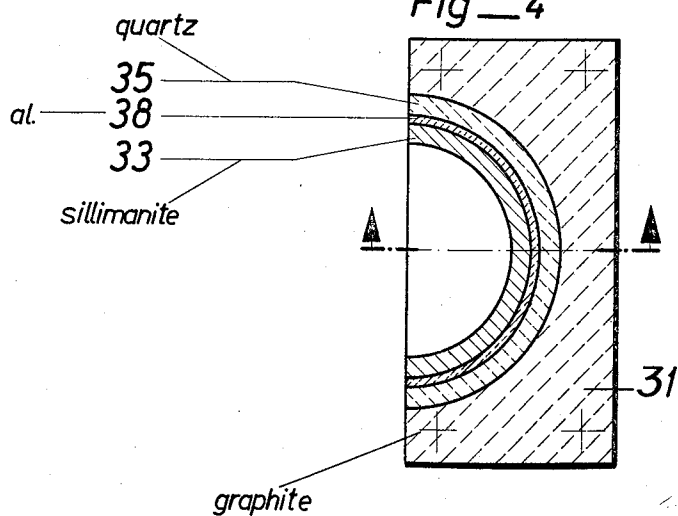


Fig_4



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3,491,992

VAPORIZING CRUCIBLE

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7 Claims

ABSTRACT OF THE DISCLOSURE

A crucible for vaporizing aluminum comprises a solid casing of refractory material with a lining of aluminum silicate in contact with the metal to be vaporized. In addition the crucible can have intermediate layers of silicon dioxide, aluminum foil, and an additional layer of aluminum silicate.

The present invention relates to a crucible for vaporizing large quantities of aluminum or similar light metals under a vacuum.

The technical development of vapor-coating under a vacuum may be regarded as being distinguished by three important stages:

The oldest and simplest stage consists of vapor-coating individual articles, for example, glass plates with silver or aluminum for producing highly reflective mirrors or with oxides for producing mirrors which are transparent to light of a certain wave length. This method consists of filling small quantities of the respective material which usually are accurately weighed, for example, a few tenths of a gram, into the crucible and of then vaporizing the material almost completely by a heat output of a few hundred watts. This vaporizing process requires only a few minutes and thereafter the apparatus is opened for the removal of the coated articles and the insertion of new articles to be coated. Since the apparatus must be opened frequently and thus permits the crucibles to be quickly exchanged, these vaporizing crucibles may be made of an inexpensive structure which only needs to last a relatively short length of time. This applies in principle also to a process in which by the use of pivotable holding means it is possible after each evacuation of the apparatus to coat several series of articles successively.

The second stage in the development of vapor-coating under a vacuum consists in the continuous coating of sheet materials of greater lengths. In order to hold sufficient quantities of the material to be vaporized, the crucibles employed in these methods must usually be of a larger size than that of the small crucibles as described above. Furthermore, in order to permit them to be refilled, these crucibles must be made so as to last for a greater length of time. There have been numerous proposals according to which this may be accomplished. Thus, for example, it has been proposed to provide a crucible which consists of a ceramic material or of graphite or metal and to provide it with a coating of oxides or halogenides. Such crucibles have proved successful under normal requirements and they are large enough and possess sufficient stability to permit quantities up to approximately 20 to 30 g. of aluminum, for example, to be vaporized, at temperatures of 1200 to 1300° C. at which the metal vapor pressure amounts to approximately 10^{-2} mm Hg. The heating is usually carried out either directly or inductively at an energy of a few kilowatts.

The third and newest stage in the development of vapor-coating consists in heating the surface of the material to be vaporized directly by means of electron beams which intensifies the vaporizing process tremendously as compared with the process according to the previous

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methods. According to this method, ribbonlike electron beams are directed with considerable energy, for example, of several hundred kilowatts, upon the surface of the material which is thereby heated and vaporized. Aluminum is thus heated to approximately 1600° C. and reaches a vapor pressure of approximately 1 mm. Hg. Such vaporizing means are very suitable for continuously vapor-coating wide bands or sheets of metal, for example, of steel. This may be done by conducting a band or sheet of a width of approximately 1 m. at a speed of more than 5 m./sec. past the vapor source and thus providing it with a coating of a thickness of up to approximately 3 to 5 μ m. The quantity of metal to be vaporized may amount in such a case to several kilograms per hour, that is, to many times the quantities as may be vaporized in the other methods as previously discussed.

These temperatures and other stresses to which they are subjected are, however, too high for the usual crucibles, even if they are provided with a protective oxygen or halogenide coating, since despite these coatings they react with the material to be vaporized and do not permit the vapor-coating process to be safely continued for a great length of time. It was then found that such a reaction between the material to be vaporized and the material of the crucibles may be prevented by employing crucibles which are made of copper and are very strongly cooled. Such copper crucibles permit sufficient quantities of many different metals and even copper to be vaporized.

Unfortunately, however, it has been found that such cooled copper crucibles cannot be employed for vaporizing aluminum since this metal because of its specific properties, such as its low surface tension and its good heat conductivity, wets and heats the copper crucible so that, even if the amount of energy supplied is considerably increased, the rate of vaporization decreases and the crucible will be destroyed.

It is an object of the present invention to provide a vaporizing crucible which has a high durability and may be bombarded with electron beams of a high intensity for a considerable length of time without causing any reaction with the metal to be vaporized and which permits large quantities of light metals and particularly of aluminum to be vaporized. The particular manner in which this object is attained according to the invention will hereafter be described with reference to aluminum, although the invention is likewise applicable to metals with properties similar to those of aluminum.

The important feature of the present invention for attaining its objects, namely, to provide a vaporizing crucible for intensively vaporizing light metals, especially aluminum, preferably by means of electron beams or by inductive heating, consists in producing such a crucible of the reaction product which is formed when a liquid metal, preferably aluminum, remains for a lengthy period of time in contact with silicon dioxide, especially in the form of quartz, at temperatures which are preferably higher than 1200° C. and lie, for example, between 1200° and 1400° C. assuming by way of example that the materials employed consist of aluminum and quartz, an increasing layer of an aluminum silicate will then be formed at the contact surface between these two materials. This layer will automatically grow so as to form a crucible which is resistant to the action of aluminum when being vaporized at the required vaporizing temperature. The aluminum which is to be vaporized is preferably melted in a quartz crucible which is embedded in a crucible which consists of an aluminum silicate and, in turn, rests within a graphite crucible.

Instead of producing the aluminum silicate lining in the manner as just described within the quartz crucible itself, it is, of course, also possible to line a quartz crucible with an aluminum silicate which is formed by a

separate reaction or with a natural aluminum silicate. An aluminum silicate which has proved especially successful consists of sillimanite.

If according to the last-mentioned modification of the invention a separate lining of an aluminum silicate is employed, it is advisable to provide between this lining and the crucible itself a thin layer of the metal which is to be vaporized, for example, in the form of a foil. The crucible itself rests also in this case within a graphite crucible.

The features and advantages of the present invention will become further apparent from the following detailed description thereof which is to be read with reference to the accompanying drawings, in which—

FIGURE 1 shows a cross section of a crucible according to one embodiment of the invention;

FIGURE 2 shows a cross section of a similar crucible of a different shape;

FIGURE 3 shows a cross section of a crucible according to a modification of the invention;

FIGURES 4 and 5 show respectively a cross section and a longitudinal section of a modification of the crucible according to FIGURE 3; while

FIGURE 6 shows a cross section of the principal elements of a vaporizing apparatus and illustrates the operation thereof.

Referring to the drawings, the vaporizing crucible according to the invention as illustrated in FIGURE 1 consists of a graphite crucible 11 which encloses a crucible 12 which consists of an aluminum silicate, preferably sillimanite, and within which a quartz crucible 13 is embedded which contains molten aluminium 14. This molten aluminium then reacts with the quartz of the crucible 13 so that, when the quartz is completely converted into an aluminum silicate at least at the side which is in contact with the molten metal, a solid intermediate wall 15 of aluminum silicate is formed adjacent to the inner surface of the crucible 12 which consists of the kindred sillimanite. The entire unit then forms a vaporizing crucible of a very high durability. It also has the special advantage of possessing self-curing properties insofar as, if a crack occurs in the aluminum silicate wall 15, any liquid aluminium which might run through the crack will react with the sillimanite cover 12 and thereby form a reaction product which tightly seals the crack so that no further liquid metal can pass therethrough.

FIGURE 2 illustrates a vaporizing crucible of the same structure as that according to FIGURE 1, but of a semicylindrical inner shape.

FIGURE 3 shows a modification of the invention, in which the vaporizing crucible consists of a crucible body 23 of silicon dioxide in the form of an opaque quartz ware which may be composed of a bottom plate 24 of a thickness of approximately 5 mm. and side walls 25 of the same thickness. The crucible body 23 may, however, also consist of a single piece of material of a corresponding shape. This body 23 is at first lined with an aluminum foil 26 of a thickness of, for example, 0.2 to 0.4 mm. The crucible lining 22 is made of sillimanite and, like the quartz crucible 23, it may be molded as an integral element or it may be composed of several parts which is generally preferred as the more simple type of construction. The depth *a* of this lining 22 should usually amount to approximately 20 to 40 mm. In order to facilitate its installation, the crucible body 23 is fitted into a graphite casing 21 which also serves for holding the parts of the crucible body together. Although the sillimanite lining 22 will be wetted by the liquid aluminium during the vaporizing process, practically no reaction will occur between it and the highly heated metal. Furthermore, silicates and especially sillimanite possess an excellent resistance to sudden changes in temperature and are in this respect quite the opposite to other oxides, such as aluminum oxides, which are also quickly eroded by liquid aluminum. These properties of the sillimanite lining 22

have the result that, if this lining cracked because of extensive use or frequent temperature changes, it will only break into pieces of a large size. These pieces are held in place and remain stuck together by the aluminum foil 26, the lower side of which facing the quartz crucible 23 forms a silicate layer while its upper side wets the sillimanite lining 22. Therefore, the broken pieces of this lining cannot pass into the metal which is to be vaporized. If, on the other hand, as was found by experiments, the lining is made of alumina (Al_2O_3), it would fall apart into many small pieces which would at least partly pass into the molten metal. The aluminum foil 26 also levels uneven surface portions of the two crucible parts 22 and 23. Although there is a possibility that, if the sillimanite lining 22 cracked, liquid aluminum might penetrate through these cracks, this cannot endanger the safety of operation of the vaporizing process since the aluminum which has penetrated will then react with the silicon dioxide of the crucible body 23 and form silicates which will again seal the cracks. The feature of embedding the crucible body 23 within the graphite casing 21 has not only the mechanical advantages as stated above, but also the very important further advantage that, if the rare event occurs that the sillimanite lining 22 as well as the crucible body 23 crack at the same place, the liquid metal which then penetrates into the graphite casing 21 will react with the graphite and thereby form a metal carbide which seals the crack. The graphite casing 21 therefore also increases the safety of operation of the vaporizing process. This casing 21 may also consist either of an integrally molded body or it may be composed of several parts which are secured to each other.

FIGURES 4 and 5 show a cross section and a longitudinal section of a vaporizing crucible of the same type as shown in FIGURE 3, but of a different shape. It consists essentially of semicylindrical parts which are provided with end plates. The quartz-ware crucible is composed of the semicylindrical part 35 and the end plates 36, and the sillimanite lining is composed of the semicylindrical part 33 and the end plates 34. Even the graphite casing consists of several parts, namely, the cylindrically based part 31 and the end plates 32 which are secured to the part 31 by screws 37. The sillimanite lining 33, 34 is also in this case separated from the quartz body 35, 36 by an aluminum foil 38. Each end of such an elongated crucible may be placed between the poles of a transverse electron gun and the metal contained in the crucible may thus be vaporized very intensively by being bombarded simultaneously by two guns. A larger round crucible of the type as shown in FIGURE 3 may thus be heated from different sides by an even larger number of transverse electron guns.

The vaporizing crucibles according to the invention have already been tested extensively in actual practice and have proved extremely successful. They may be employed in all cases in which light metals should be vaporized intensively. This also includes not only the process of vapor-coating long strips or sheets of steel, but also, for example, the process of cleaning metals by distillation in a vacuum.

FIGURE 6 finally illustrates the principles of construction of a vaporizing apparatus which comprises a ribbonlike electron source which is produced by a transverse electron gun, and a vaporizing crucible according to the invention. This vaporizing apparatus is mounted in a boiler, not shown, which is evacuated by high-vacuum pumps. The strip 41, for example, of steel, which is to be vapor-coated travels above and past the vapor source which consists of the crucible 42 which contains the metal 43 to be vaporized. This metal is heated by ribbonlike electron beams 44 which are produced by a transverse electron gun which consists of a heated cathode 45, a focussing electrode 46, and an anode 47. The cathode and the focussing electrode are acted upon by a negative potential of approximately 10 to 20 kv., while

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the anode 47 and the other parts as illustrated have a ground potential. The transverse gun is provided with a permanent magnet or an electromagnet 48, the pole piece 49 of which together with a second pole piece, not shown, produce a magnetic field of a particular shape. When the electron rays 44 emerge from the cathode 45, they are deflected by this magnetic field in a manner so as to impinge in the form of a substantially circular beam upon the metal 43 which is thereby strongly heated and vaporized. The employment of a transverse electron gun has not only the advantage that the heating energy which is supplied may be increased practically to any desired value, but that the operation of the apparatus is also very secure insofar as all parts of the electron gun are located underneath the upper edge of the crucible and are therefore protected from the formation of undesirable condensate layers.

Having thus fully disclosed my invention, what I claim is:

1. A crucible for holding an aluminum-containing metal to be vaporized, comprising a solid refractory casing, a solid lining in contact with said metal, the lining consisting of a reaction product of silicon dioxide and aluminum formed at a temperature higher than 1200° C., a silicon dioxide layer underlying the lining, and an aluminum silicate layer intermediate the refractory casing and the silicon dioxide layer.

2. The crucible of claim 1, wherein the casing is of graphite.

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3. The crucible of claim 1, wherein the silicon dioxide is quartz.

4. A crucible for holding an aluminum-containing metal to be vaporized, comprising a solid refractory casing, a solid aluminum silicate lining in contact with said metal, a thin aluminum layer underlying the lining, and a silicon dioxide layer intermediate the refractory casing and the thin aluminum layer.

5. The crucible of claim 4, wherein the casing is of graphite.

6. The crucible of claim 4, wherein the silicon dioxide layer is of quartz.

7. The crucible of claim 4, wherein the aluminum silicate is sillimanite.

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