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PROCESS OF COATING A REFRACTORY BODY WITH BORON NITRIDE AND THEN REACTING WITH ALUMINUM

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This invention relates to coating and more particularly to the coating of various substrates with aluminum. This application is in part a continuation of the copending application of Baer et al., Serial No. 786,675, filed January 14, 1959, now abandoned.

In the vacuum evaporation of aluminum where aluminum is heated to an elevated temperature on the order of 1200 to 1300° C., or above, one of the principal technical problems to be solved has been to find a crucible which is resistant to attack by the high-temperature aluminum. The same problem is encountered when a portion of the aluminum-evaporating source which is at elevated temperatures is exposed to a high density of aluminum vapors. This situation arises in those cases where it is desired to deflect or concentrate the flowing aluminum vapors to provide, for example, lateral or downward evaporation.

Some progress has been made in providing aluminum-resistant structures, but these structures have not always been adequately simple to construct or cheap to maintain. While some progress has been made, great difficulty has been experienced in providing a completely satisfactory method for producing such a source, particularly one which is of a complex geometric shape. Equally, the art has only with difficulty been able to produce a stream of aluminum vapors which is directed laterally or downwardly from the source. Such a stream is particularly useful when discrete objects such as nuts and bolts and powders are to be coated, or when two sides of a continuous substrate are to be coated at the same time.

Accordingly, it is a principal object of the present invention to provide a method for producing a source of aluminum vapors which can have wide latitude of design and still be reasonably simple and inexpensive to construct.

Still another object of the invention is to provide a source of aluminum vapors of the type described above which is simple to manufacture and use.

Another object of the invention is to provide a source of the above type which can be produced from relatively inexpensive materials.

Still another object of the invention is to provide a means for confining high-temperature aluminum vapors to produce a stream of aluminum vapors much more concentrated than can be produced by previously-known techniques.

Still another object of the invention is to provide a mechanism for providing a high intensity stream of aluminum vapors capable of providing rapid coating of a discrete object with minimum transfer of heat to the object.

Other objects of the invention will in part be obvious and will in part appear hereinafter.

The invention accordingly comprises the process involving the several steps and the relation and the order of one or more of such steps with respect to each of the others and the apparatus possessing the features, properties and the relation of components which are exemplified in the following detailed disclosure, and the scope of the application of which will be indicated in the claims.

For a fuller understanding of the nature and objects of the invention, reference should be had to the following detailed description.

In the present invention, a container for confining molten aluminum at elevated temperatures is provided by furnishing a refractory base which is in the form of a crucible or other structure suitable for confining a substantial pool of molten aluminum. This base is preferably formed of carbon or a refractory oxide such as magnesia, alumina or zirconia, or a refractory silicate such as zircon and the like. The base preferably does not contain any substantial quantities of materials which will outgas as the base is heated in a vacuum chamber to the elevated temperature of 1200° C. and above, which is necessary for aluminum evaporation. For convenience, the invention will be initially described in connection with the utilization of a carbon crucible without attempting thereby to limit the invention.

A carbon crucible, which may be in the form of a cylindrical bucket or long boat, is formed as a solid piece or built up out of smaller pieces of carbon suitably attached together by carbon pins or the like. The whole interior surface of the crucible, which is to be exposed to molten aluminum or aluminum vapors while the surface is at an elevated temperature, is then coated with a slurry of boron nitride to a thickness on the order of $\frac{1}{64}$ inch. This slurry is then dried, such as by baking in an oven, at a temperature on the order of 200° F. to drive off the water or other medium for forming the liquid phase in the slurry. The thus prepared crucible is positioned in a vacuum chamber and suitably supported so as to be heated by an induction coil, for example. Such a crucible has an extremely long life and is capable of operation at elevated temperatures in contact with molten aluminum for many hours.

While the exact reason is not fully understood, it has also been found that the addition of titanium, zirconium, hafnium, vanadium, niobium or tantalum to the molten aluminum tends to maintain an appreciably higher evaporation rate. This is believed to be the result of minimizing any appreciable concentration of aluminum carbide in the melt due to pin hole porosity of the boron nitride coating which would otherwise permit penetration of the coating and attack of the crucible by the aluminum.

After a period of operation of the crucible, it was found that the boron nitride layer has been converted to an extremely hard, dense compound whose identity has not been clearly established. This is believed to be a reaction product between boron nitride and aluminum, and it has been found to form in the vapor state as well as in the liquid state, that is, by reaction of boron nitride with aluminum vapors as well as the reaction of boron nitride with molten aluminum.

This surface can also be formed, for example, when a cover is desired over the crucible to provide lateral or downward evaporation from the crucible. In this case, a deflecting top is provided over the crucible, a suitable opening being left between one portion of the cover and the main body of the crucible. This cover is also preferably formed of a refractory material, such as carbon, similar to the material of the crucible. It is equally coated with a layer of boron nitride and dried. The abutting surfaces of the crucible and cover are coated with a boron nitride layer. The crucible and cover are placed in the vacuum system, the crucible being charged with aluminum. The crucible and cover are then heated to about 1200° to 1300° C. and the boron nitride coating on the cover is reacted with aluminum vapors coming from the pool of aluminum confined by the source. These aluminum vapors react with the boron nitride coating at the elevated temperature of the cover. After a few minutes of operation, the aluminum vapors have reacted with the boron nitride coating to form a dense, hard surface which appears to be substantially impervious to and unreactive with aluminum vapors for long periods of time thereafter. The joint between the cover and the crucible has been converted to a hard, dense mass which is tight to liquid aluminum as well as to the aluminum vapors.

As a result of the techniques described above, it is possible to produce sources for molten aluminum having a wide range of geometric configurations and a control of aluminum vapors which permits upward, sideways or downward direction of the vapors. The above described method of forming a vapor-tight joint can also be used to form joints which also withstand molten aluminum, such as when a large source is to be built from a number of pieces of carbon.

In one preferred embodiment of the invention, a slurry is prepared by mixing 55 grams of powdered boron nitride in 121 cc. of water. This makes a paste having the consistency of whipped cream. This paste is then applied, such as by a brush, to all those surfaces of a carbon crucible which are to be exposed to molten aluminum or aluminum vapors. This coating is preferably 1/64 inch thick. The thus coated carbon body is air dried at about 200° F. Several coatings can be applied in sequence. In one preferred embodiment of the invention, a carbon crucible, having an internal diameter of 4 and a depth of 2 inches, is thus treated with boron nitride paste and air dried. The crucible is placed in a vacuum coating tank and is charged with 550 grams of aluminum. The crucible is then brought up to elevated temperatures on the order of 1200 to 1300° C. At the end of 35 minutes the major percentage of the aluminum has been evaporated. The tank is opened after the crucible is cooled to about 800° C. Another 250 grams of the solid aluminum is added to the remaining molten aluminum in the crucible. 10 grams of titanium are also added to the melt at this time. The chamber is evacuated again and the crucible is brought up to operating temperature. The aluminum evaporation rate in the second run is found to be equal to or greater than the aluminum evaporation rate in the first run, the effect of the titanium addition being to remove any small amounts of aluminum carbide formed by penetration of aluminum through pinholes or cracks in the boron nitride coating.

In another embodiment of the invention a carbon crucible having an internal diameter of 2 inches and a depth of 2½ inches is provided with a ½ inch hole near the top thereof. This crucible is also provided with a close fitting carbon cover. The inner surfaces of the crucible and cover (as well as the mating surfaces of the crucible and cover) are then coated with boron nitride and heated as above. This provides a source which produces a concentrated stream of aluminum vapors travelling laterally from the hole near the top. If the crucible is tilted somewhat the stream of vapors can be directed downwardly as well as laterally. This arrangement is particularly suited for coating discrete objects such as nuts and bolts or powders which are most conveniently coated from above while being supported on a vibrating tray or the like.

While several preferred embodiments of the invention have been described above, numerous modifications thereof can be employed without departing from the spirit of the invention. The slurry of boron nitride can be prepared using numerous vehicles other than water. However, from the standpoint of cost and freedom from residual material which might outgas in the vacuum system, water is preferred. Equally wetting agents or binding agents can be added to the boron nitride, but these have been found to be unnecessary. For complex structures they can be helpful.

Similarly, carbon is a preferred material from the standpoint of structural strength at elevated temperatures and freedom from decomposition at elevated temperatures. Other refractory substances can be employed; for example, refractory oxides such as magnesia, alumina and zirconia or refractory silicates such as zircon can be protected by the application of a boron nitride coating. While metals can be given a temporary coating with boron nitride, the high solubility of all metals in molten aluminum precludes their use since any pinhole or crack in the boron nitride coating causes rapid failure of any of the metals.

While specific forms of apparatus have not been illustrated, the improved aluminum vapor source can be employed in many types of coating devices such as those shown in the following U.S. Patents: 2,622,041, 2,643,201, and 2,879,739 and the copending application of Cerych, Clough, and Steeves, Serial No. 795,424, filed February 25, 1959, to mention only a few of its uses.

Since certain changes can be made in the above process and apparatus without departing from the scope of the invention herein involved, it is intended that all matter contained in the above description shall be interpreted as illustrative and not in a limiting sense.

What is claimed is:

1. The process of forming a container for confining molten aluminum at elevated temperatures on the order of 1200° C. and above, which container is also resistant to attack of aluminum vapors at said elevated temperatures, the process comprising the steps of forming a slurry containing boron nitride powder, applying a layer of the boron nitride slurry to those surfaces of a container formed of a refractory material selected from the group consisting of carbon and the refractory oxides and silicates which are to be exposed to aluminum at said elevated temperatures, drying said boron nitride layer and thereafter reacting said boron nitride layer with aluminum at a temperature on the order of 1000° C. to 1300° C. to form a hard solid reaction product of said boron nitride and aluminum, said reaction product being unreactive with aluminum and being wettable by aluminum.

2. The process of preventing erosion of a carbon body by molten aluminum which comprises the steps of forming a slurry containing boron nitride powder, applying a layer of the boron nitride slurry to the surface of said carbon body, drying said layer and reacting said boron nitride layer on the carbon body surface with aluminum vapors at a temperature on the order of 1000° C. to 1300° C. to form a reaction product which is unreactive with aluminum and wettable by aluminum.

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