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## METHOD OF IMPROVING ALUMINUM AND ALLOYS THEREOF

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This invention relates to aluminum and alloys thereof, and more particularly to an improved process for imparting desirable characteristics to such metals.

An object of my invention is to provide an improved process for treating aluminum and alloys thereof to improve their characteristics.

A further object of the invention is to provide a simple, effective, inexpensive, more exact and more rapid process for introducing boron into molten aluminum and alloys thereof.

Another object of this invention is to provide an improved process for treating aluminum and alloys thereof with boron, whereby the treated metal will exhibit superior properties for casting, forging, rolling, drawing, extruding, welding and other purposes.

An additional object of the invention is to provide an improved method of treating aluminum and alloys thereof to impart improved characteristics thereto, which treatment may accompany the fluxing of the molten metal and require no additional steps or equipment.

Another object of the invention is to provide an improved method of treating aluminum and alloys thereof with boron to impart improved casting properties thereto such as better mold filling characteristics in sand and metal molds, uniform and fine grain structure and homogeneous characteristics, with substantial reduction of porosity and shrinkage cracks.

A further object of my invention is to provide an improved and simple process for treating aluminum and alloys thereof with boron to impart to such metals improved characteristics which are relatively permanent and do not disappear upon remelting the treated metal or upon over-heating molten treated metal.

Another object of this invention is to provide a simple and convenient process of treating aluminum and light alloys thereof with boron to obtain a substantially permanent refinement of grain size and improvement of physical and mechanical properties of the treated metal.

A further object of the invention is to provide an advantageous method of treating secondary aluminum and alloys thereof to impart to such metal advantageous characteristics for casting and other purposes.

Another object of my invention is to provide an improved method of imparting substantially permanent improvement with respect to grain refinement and physical and mechanical properties to aluminum and alloys thereof by the

treatment thereof with boron in a normally solid, non-metallic form.

A further object of this invention is to provide an improved method of treating aluminum and alloys thereof with boron to impart relatively permanent improvement in physical and mechanical characteristics, which method may be carried out by the use of boron in an inexpensive and readily available form and which may be carried out at relatively low temperatures and in a relatively short time.

Another object of my invention is to provide an improved product resulting from the treatment of aluminum or alloys thereof with a non-metallic boron compound.

An additional object of this invention is to provide an advantageous composition wherein a boron compound may be dissolved and maintained in cooperative relation with the molten aluminum or alloy bath to cause a rapid reaction between said boron compound and aluminum.

Other objects of the invention will be apparent to those skilled in the art from the disclosures herein made.

Fine grain size and uniform dispersion of alloy constituents are extremely desirable in aluminum and light alloys thereof. The desirability of fine uniform grain size throughout the cross section of all types of castings made from such metals is apparent. Furthermore, the presence of fine uniform grain size is very desirable in various aluminum alloys which are to be forged, rolled, welded, or used for other purposes. Such fine grain structure of aluminum and aluminum alloys in general has not been obtainable in the past until the discovery that treatment of aluminum or aluminum alloys in molten condition with boron under proper conditions would impart properties to the molten metal such that, upon solidification, a marked grain refinement of the aluminum and alloyed constituents occurred. It was found, moreover, that such improvement of the aluminum or aluminum alloy was relatively permanent in that the treated metal exhibited marked grain refinement and improved physical and mechanical properties even after repeated melting and solidification. However, the previous methods of treating aluminum and aluminum alloys with boron to accomplish the desired improvement of the metal have been relatively complicated, difficult and expensive. My invention, on the other hand, contemplates an inexpensive, simple and effective method of treating aluminum and

alloys thereof with boron which may be carried out in relatively short times and at relatively low temperatures to effect a substantially permanent improvement of physical and mechanical properties.

The more common boron compounds, such as borax and boric oxide, will react with metallic aluminum to form metallic boron and aluminum oxide. For example, I have found that if boric oxide is disposed on the surface of a bath of molten aluminum and the aluminum maintained in molten condition at approximately 900 degrees C. for about an hour and then allowed to solidify without pouring, a deposit of crystalline metallic boron is found on the surface of the aluminum. This reaction between the boron compound and the aluminum, which results in the production of metallic boron and aluminum oxide, proceeds relatively slowly due to the high viscosity of the melted boron compound and also due to the deficient ability of said boron compound to dissolve the aluminum oxide resulting from the reaction.

My invention contemplates the promotion and acceleration of the reaction between a boron compound and molten aluminum and the dispersion of the resulting metallic boron through the molten metal. Thus, I have discovered that the reaction between a boron compound and molten aluminum will take place much more rapidly and at considerably lower temperatures if the boron compound be dissolved in a molten flux which is also capable of dissolving the aluminum compound or compounds resulting from the reaction. Under these conditions the boron compound is in a much more reactive state, and can react more readily with the molten metal with which the flux is in contact, and the metallic boron produced by the reaction can more readily diffuse into the molten metal. Furthermore, the solution of the aluminum compound or compounds resulting from the reaction and the diffusion thereof into the flux, and the consequent removal of said aluminum compound or compounds from the contacting surface of the flux and molten metal where, according to my present opinion, the reaction takes place, greatly assists in causing the reaction to go forward more rapidly to substantial completion. To further increase the rate of reaction I prefer to agitate the molten metal and flux whereby the contacting surfaces of flux and molten metal are greatly increased.

It is quite possible that the boron compound takes part in reactions other than that hereinabove mentioned. Such other reactions, however, I believe to have no deleterious effect on the reaction above outlined.

The boron material, wherewith my invention is carried out and which is incorporated in a suitable flux coating, may comprise one or more of a large variety of boron compounds. For example, I have found boric acid, borax, boron oxide and calcium boride to be very satisfactory. Alkali, alkaline earth, and other borates, boron sulphide, boron phosphate, boron nitride and boron carbide likewise may be employed. Furthermore, double salts may be successfully used, such as ammonium boro-fluoride and potassium boro-fluoride. The above boron compounds are merely illustrative. In general I have found most solid and many liquid inorganic boron compounds to be satisfactory for the purpose of this invention. The boron material is preferably present in appreciable amount up to  $1\frac{1}{2}$  per-

cent by weight of the metal bath. I have employed boron compounds in amounts as low as a small fraction of one percent with satisfactory results. If desirable, boron compounds in excess of  $1\frac{1}{2}$  percent, say up to 3 percent, may be used. It will be understood that the boron compound may be introduced into the flux covering after the flux is melted, or may be incorporated with the flux materials in making up the flux.

The flux with which the molten metal bath is treated during the carrying out of my invention and into which the boron material is introduced may be of widely varying composition, the requisites being that it be molten and not too volatile at the temperature of the metal bath, and that said flux be capable of dissolving the particular boron compound being used, and of dissolving aluminum oxide and other compounds formed by the reaction between the boron compound and aluminum.

The flux preferably has a melting point relatively close to that of the metal, in order that the molten bath need not be heated too highly. Thus, I have found a flux consisting of potassium chloride, sodium chloride, cryolite and calcium fluoride to be very satisfactory. I have also employed a flux consisting of calcium chloride with good results. Moreover, many other fluxes having characteristics outlined above may be employed. For the purpose of this invention the fluxes preferably should be present in appreciable amounts up to 3 percent by weight of the metal bath. I have employed fluxes in amounts as low as a small fraction of one percent with satisfactory results. If desired, flux in amounts in excess of 3 percent may be used.

In carrying out my invention, the boron material consisting of one or more boron compounds is incorporated in the flux, either before or after the flux has been associated with the molten bath. The metal bath preferably is maintained at relatively low temperatures and need not be heated to temperatures much more than 100 degrees C. higher than the melting point thereof. In many cases I have carried out the invention at temperatures relatively close to the melting point of the metal bath and less than 100 degrees C. higher than the same. The metal bath and flux may be agitated for a short time, and may then be permitted to stand longer before pouring, if desired.

The result of the reaction between the boron compound or compounds and the metal of the bath, as previously stated, I believe to be the formation of finely divided metallic boron, which boron becomes disseminated throughout the metal of the bath.

While the mechanism whereby the aluminum or aluminum alloy is improved by the boron is not definitely known, it is believed that during solidification boron forms a very large number of crystal nuclei throughout the molten bath, resulting in the growth of a very large number of relatively small crystals of aluminum and alloyed constituents rather than a small number of relatively large crystals.

The experience which I have had indicates that the time required to carry out my improved method will depend upon the composition of the molten bath or alloy, the temperature of the bath, and the boron material used. For example, I have found that aluminum-calcium alloys require a relatively short period of time for their modification as compared with aluminum-silicon

alloys. Between these extremes I have found aluminum-magnesium alloys, aluminum-zinc alloys, aluminum-copper alloys, and unalloyed aluminum to require increasingly longer times.

5 By way of specific example, I have carried out my invention as follows: An alloy consisting of 92 parts by weight of aluminum and 8 parts copper, having a melting temperature of about 620 degrees C., was melted in a suitable container. A flux consisting of 8.2 parts by weight of potassium chloride, 33.3 parts sodium chloride, 8.2 parts cryolite, and 38.8 parts calcium fluoride was prepared. To this flux was added 11.2 parts boric acid. The flux to which had been added the boric acid was disposed on the surface of the metal bath, which was maintained at a temperature of about 700 degrees C. The total of the flux plus boron material was 1 percent of the weight of the molten metal bath. The molten metal and flux were agitated vigorously for one-half minute, and after 3 minutes the metal was poured. The resulting solidified metal was found to exhibit an extremely fine grain size and to have higher tensile strength, greater elongation, and somewhat greater hardness than corresponding alloys not subjected to suitable boron treatment. Moreover, the improvement in properties persisted throughout several subsequent operations of remelting and resolidification. In the foregoing example, the boric acid reacts with one or both of the fluorine compounds and produces boron fluoride which reacts with the aluminums.

Further by way of illustrating my invention, an alloy consisting of 90 parts by weight of aluminum and 10 parts silicon, having a melting temperature of about 590 degrees C. was melted in a container. A flux of calcium chloride to which had been added boric acid was disposed on the surface of the metal bath, the calcium chloride and boric acid being in proportions of 70 to 30. The boric acid was present in amount of 1 percent of the weight of the molten metal bath, the latter being maintained at a temperature of about 700 degrees C. The bath and flux were agitated vigorously for 5 minutes, and after 10 minutes the metal was poured. The resulting solidified metal showed the improved characteristics referred to above in connection with the previous specific example.

It will be noted that I have provided an improved method, not requiring the use of expensive alloying materials, whereby there may be imparted to aluminum and alloys thereof improved mechanical and physical properties such as greater tensile strength and elongation, increased elasticity and resistance to fatigue, low gas content, decreased crystalline shrinkage upon solidification, and decreased grain size of the aluminum constituent. Moreover, such improvement takes place without impairment of the thermal or electrical conductivity of the metal, while yet facilitating subsequent heat treatment thereof. Furthermore, the influence of undesirable impurities such as iron is decreased, while the treated metal in molten form exhibits greater fluidity whereby its casting properties are improved.

It will be observed that I have provided an improved method of treating aluminum and its alloys with boron which needs no additional equipment beyond that used in present day practice, and which has the combined advantages of economy, with speed and ease of operation. Furthermore, the fact will be duly

appreciated that my improved process may be practiced in connection with the usual step of fluxing with the obvious attendant advantages.

It will be seen that my invention has great commercial advantages in that aluminum and aluminum alloys of either virgin or secondary sources, or combinations thereof, may readily be provided with those properties of fine grain size, high tensile strength and elongation, which are of so much importance in respect to metals which are to be subjected to forging, rolling, drawing, extruding, welding and other purposes, and the production of more perfect billets for such purposes is made possible.

My invention furthermore makes readily available aluminum and aluminum alloy sand and metal mold castings of uniform grain size throughout their cross-section, making unnecessary such careful control over pouring and metal mold temperatures as has hitherto been necessary, and making the manufacture of more difficult castings commercially practicable. Furthermore, by my invention, the use of secondary aluminum and alloys, by reason of the relative ease with which the improvement of their characteristics may be obtained, becomes more feasible for casting purposes with attendant increasing value of such secondary aluminum, on the one hand, and on the other hand, decreasing the cost of both sand, and chill cast or metal mold aluminum castings by reason of the fact that more secondary metal may be employed therein with equal, if not better, results than has hitherto been readily possible by the use of more expensive metal.

It will further be noted that metal treated according to my invention may be poured at temperatures considerably higher than heretofore possible to produce solid sound castings relatively free of shrinkage cracks, gases and porosity, and having in many instances a superior surface finish.

Furthermore, it is to be understood that the particular materials and compounds disclosed, and the procedure set forth, are presented for purposes of explanation and illustration and that various equivalents can be used and modifications of said procedure can be made without departing from my invention as defined in the appended claims.

What I claim is:

1. The method of improving the characteristics of aluminum and alloys thereof, which comprises maintaining the aluminum or aluminum alloy to be treated in molten condition and at a temperature relatively close to its melting point, holding a fused solution of a boron compound dissolved in a flux containing stable ingredients, in contact with the molten metal for a sufficient length of time to introduce a sufficiently small amount of boron into the metal so that, upon solidification, the aluminum constituent will exhibit relatively fine grain size, said flux being molten at a temperature not substantially more than 100° C. higher than the melting point of aluminum and being capable of dissolving aluminum compounds resulting from the reaction of aluminum and the boron compound.

2. The method of improving aluminum and alloys thereof, which comprises melting the metal to be treated, disposing on the surface of said molten metal a flux comprising sodium chloride, potassium chloride, cryolite and calcium fluoride, dissolving in said molten flux an

appreciable amount, up to about 3 percent of the weight of the molten metal, of boron compound of an arbitrary class consisting of alkali borates, alkaline earth borates and boric acid, and agitating said molten metal and flux to produce a more vigorous reaction between the boron compound and the molten metal.

3. The method of improving aluminum and alloys thereof, which comprises disposing a flux of calcium chloride on the surface of a molten bath of the metal to be treated, dissolving boric acid in said flux in an appreciable amount up to about 3 percent of the weight of the molten metal, agitating said bath for several minutes to promote the reaction between the boric acid and aluminum, and permitting said metal to remain molten for several minutes longer to permit the dissemination of boron formed from said reaction through said molten metal.

4. The method of improving aluminum and alloys thereof, which comprises preparing a molten bath of the metal to be treated, associating with said molten metal a flux of the composition, sodium chloride about 33.3 parts, potassium chloride about 8.2 parts, cryolite about 8.2 parts, and calcium fluoride about 38.8 parts, dissolving in said flux about 11.2 parts boric acid, said flux being in appreciable amount up to about 3 percent of the weight of the molten metal, and maintaining said bath molten for several minutes to permit substantial reaction between said boric acid and molten metal.

5. The method of improving aluminum and alloys thereof, which comprises treating the metal in a molten state with a boron compound dissolved in a molten flux comprising sodium chloride, potassium chloride and cryolite, the solution of the boron compound in the flux being maintained in contact with the molten metal for a sufficient length of time to cause an improvement in the physical properties of the metal.

6. The method of improving aluminum and aluminum alloys, which comprises treating the molten metal with a boron compound dissolved in a molten flux which is capable of dissolving reaction products of aluminum and the boron compound, and which does not have a melting point substantially more than 100° C. higher than the melting point of aluminum, the solution of the boron compound in the flux being maintained in contact with the molten metal for a sufficient length of time to cause an improvement in the physical properties of the metal and to introduce a sufficiently small quantity of boron into the metal in a dispersed condition so that upon solidification the aluminum constituent of the treated metal will exhibit relatively fine grain size.

7. The method of improving aluminum and aluminum alloys, which comprises treating the metal in a molten state with boric acid dissolved in a molten flux which does not have a melting point more than substantially 100° C. higher than the metal being treated, the solution of the boric acid in the flux being maintained in contact with the molten aluminum for a sufficient length of time to cause an improvement in the physical properties of the metal and to introduce a sufficiently small quantity of boron in a dispersed condition into the metal so that upon solidification the aluminum constituent of the metal will exhibit relatively fine grain size.

8. The method of improving aluminum and alloys thereof, which comprises treating the metal in a molten state with boric acid dissolved in a molten flux comprising sodium chloride, potassium chloride and cryolite, and causing relatively large quantities of the flux and molten aluminum to come in contact with each other, the solution of the boric acid in the flux being maintained in contact with the molten metal for a sufficient length of time to cause an improvement in the physical properties of the metal.

9. The method of improving the characteristics of aluminum and alloys thereof which comprises holding a flux having a boron compound dissolved therein in contact with molten aluminum or an aluminum alloy, said flux being present in sufficient amount to dissolve the boron compound but not exceeding substantially more than three percent (3%) of the weight of the molten bath, and maintaining said flux in contact with the molten bath for a sufficient length of time to cause a modification of the aluminum constituent of the metal and to introduce a sufficiently small amount of boron in a dispersed condition into the molten metal so that upon solidification the aluminum constituent of the metal will exhibit relatively fine grain size.

10. The method of improving the characteristics of aluminum and alloys thereof which comprises holding a flux having a boron compound dissolved therein in contact with molten aluminum or an aluminum alloy, said flux having a melting point not substantially in excess of 100° C. higher than the melting point of aluminum and being present in sufficient amount to dissolve the boron compound but not exceeding substantially more than three percent (3%) of the weight of the molten bath and maintaining said flux in contact with the molten bath for a sufficient length of time to cause modification of the aluminum constituent of the metal and to introduce a sufficiently small amount of boron in a dispersed condition into the molten metal, so that upon solidification the aluminum constituent will exhibit relatively fine grain size.

11. The method of improving the characteristics of aluminum and alloys thereof which comprises bringing a flux having a boron compound dissolved therein in contact with molten aluminum or an aluminum alloy, the amount of boron dissolved in said flux ranging from appreciable amounts up to three percent of the weight of the molten bath and the amount of flux being sufficient to dissolve the boron compound but not substantially in excess of that amount, and maintaining said flux in contact with the molten bath for a sufficient length of time to cause a modification of the aluminum constituent of the metal and to introduce a sufficiently small amount of boron in a dispersed condition into the molten metal, so that upon solidification the aluminum constituent will exhibit relatively fine grain size.

12. The method of improving aluminum and alloys thereof, which comprises treating the metal in a molten state with a boron compound dissolved in a molten flux containing stable ingredients which does not have a melting point substantially more than 100° C. higher than the melting point of aluminum, the amount of boron compound dissolved in the flux being present in appreciable amounts but not exceeding three percent of the weight of the molten bath, and maintaining the solution of boron compound

dissolved in said flux in contact with the molten metal for a sufficient length of time to cause an improvement in the physical properties of the metal and to introduce a sufficiently small amount of boron in a dispersed condition into the molten metal so that, upon solidification, the aluminum constituent will exhibit relatively fine grain size.

13. The method of improving the characteristics of aluminum or aluminum alloys which comprises holding a boron compound and a fluorine compound dissolved in a flux in contact with molten aluminum or an alloy thereof and causing the boron fluoride formed by the reaction to pass in contact with the molten bath for a sufficient length of time to introduce a sufficiently small amount of boron in a dispersed condition into the metal, so that upon solidification the aluminum constituent of the metal will exhibit relatively fine grain size.

14. As a new and useful product, improved

cast metal of aluminum or alloys thereof resulting from the treatment of said metal in molten form with a normally solid boron compound to introduce a sufficiently small quantity of boron in a dispersed condition into the metal so that, upon solidification, the aluminum constituent of the metal will be characterized by having a fine, modified grain structure which persists throughout repeated melting and re-solidification of said metal.

15. As a new and useful product, aluminum or alloys thereof treated while in molten condition with boric acid in molten dissolved condition to introduce a sufficiently small quantity of boron into the metal so that, upon solidification, the aluminum constituent will exhibit a refined modified grain structure and increased physical and mechanical properties capable of persisting throughout repeated melting and solidification of the treated metal.

WALTER BONSACK.

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