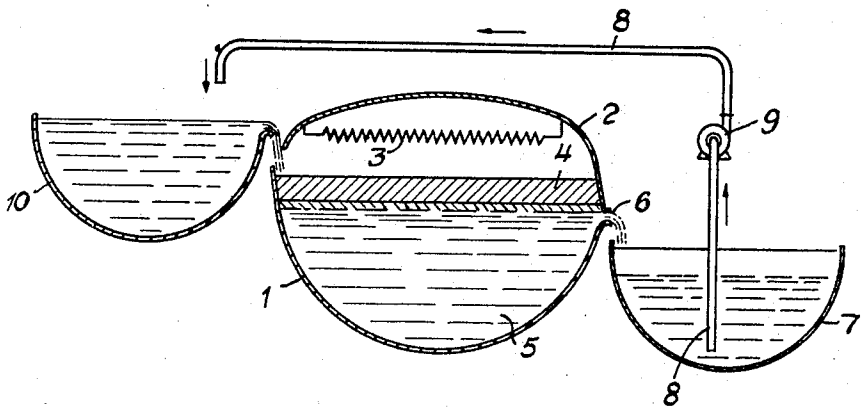
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METHOD OF PRODUCING OR REFINING ALUMINUM

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METHOD OF PRODUCING OR REFINING ALUMINUM

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The present invention refers to a method of producing or refining aluminum.

In the reduction of aluminum silicate or other raw materials containing aluminum oxide, such as clay, kaolin, bauxite and so forth, with the aid of carbon in the electrothermal way, alloys will be obtained, which in addition to a considerable quantity of aluminum also contain large quantities of silicon, iron, titanium and other elements. Hitherto, these products would principally be used as auxiliary agents in metallurgy, but it has also been proposed to employ them as primary materials for the production of aluminum, for instance by separating the iron and the silicon through liquation. No satisfactory results would be obtained in this way, however.

Products of a similar kind as the above-mentioned Al-Si-alloys, which are obtained in the reduction of aluminum silicate and so forth, are now also to be found, however, in the form of waste or scrap alloys of aluminum from the generally employed aluminum alloys of the type AlSi, which may also show a series of other alloy constituents.

Hitherto, it would be difficult or impossible to refine scrap alloys of aluminum, inasmuch as this metal is more electro-positive than the remaining alloy constituents employed. Here, magnesium forms an exception, but this metal can be removed as an alloy constituent from aluminum with the aid of certain salts, which are added in the molten state. Hitherto, no satisfactory thermal method was known for the separation of the other ordinary alloy constituents, such as copper, nickel, cobalt, chromium, titanium, iron and silicon, zinc, although a limited refining, as stated above, may be obtained by separation through liquation, such refining being not carried out to a satisfactory extent, however.

As will appear from the above, it would be of great importance to be able to provide a method both to work the Al-Si and Al-Si-Fe-alloys produced in a purely thermal way into pure aluminum, and to obtain the aluminum from various scrap alloys to be found in the market.

The present invention has for its object to provide a method, by means of which this problem is to be solved. With the aid of this method it will also be found possible in a simple and inexpensive manner to produce aluminum of a degree of purity hitherto unobtainable in technology.

This method is distinguished by heating of aluminum alloys containing considerable quantities of, for instance, silicon, iron and other elements together with a metal, preferably lead, which dissolves the aluminum but not the other alloy constituents, to high temperatures, an appreciable portion of the aluminum in the alloy to be refined being then dissolved in the lead, whereas the remaining alloy constituents remain undissolved, the lead-aluminum- alloy forming being then separated from the undissolved alloy constituents and cooled down rather slowly, and the dissolved aluminum being then precipitated and subsequently separated from the lead. The aluminum obtained, which may contain certain quantities of silicon and other substances, if desired, may then, if required, be dissolved anew in lead at high temperatures; upon precipitation and separation from the lead, an aluminum will then be obtained which is considerably purer than that which was first obtained. By continuing in this manner, it is possible little by little to reach an aluminum which may be designated super-aluminum.

It has been found, however, that as a rule an aluminum of an exceeding high degree of purity will be obtained upon a single extraction, particularly if certain working directions be followed, which will be treated more closely hereinafter.

The method is very simple, and it is thus based on the circumstance that aluminum is dissolved more readily by lead at higher temperatures than silicon and the remaining alloy constituents, and that the aluminum dissolved in the lead separates from the lead, when the latter is cooled down slowly to lower temperatures.

The invention will be explained more fully in the following.

It has been assumed previously that lead and aluminum do not form any alloy. When these two metals are melted together, two layers will be obtained, lead lowermost and aluminum topmost, without any alloy being formed. Investigations during the last years have shown, however, that there is still a mutual solubility which increases with an increasing temperature. Thus it is known now that there is a uniformly augmenting solubility of lead in aluminum, up to 30% at 1200 degrees centigrade. On the other hand, the solubility of aluminum in lead is comparatively limited. At the melting point of aluminum, the solubility is practically naught, whereas it is 1.5% at 1000 degrees centigrade and about 2% at 1200 degrees centigrade. It has been found, however, that the solubility of silicon in lead is still much less, and in the tests carried out by the inventor it has been proved that at higher temperatures aluminum is preferably alloyed with the lead, and that this aluminum when slowly cooled down to a point below the melting point of

aluminum flows up on top of the surface of the lead bath, whence it can be removed by simple means, such as scraping and the like.

As an example of carrying the method into effect the following may be cited:

500 grams of lead were melted in a crucible, and to this were added 10 grams of eutectic silumin containing 12.8% Si, the whole being then heated to 1000 degrees centigrade. Upon stirring for about 15 minutes the mixture was cooled down, and a lump of remaining undissolved alloy could then be separated from the lead. The lead with dissolved aluminum was now slowly cooled down further to below 650 degrees centigrade. Meanwhile the aluminum was precipitated and rose to the top, whereupon it was removed and analyzed. Disregarding immaterial quantities of lead and iron this test showed a percentage of silicon of only 0.21.

Thermal refining of aluminum alloys

[Non-recurrent treatment]

No.	Alloy comprised		Time in minutes	Temperature °C.	Refined aluminum						
	Species	Analysis			Fe %	Si %	Cu %	Ni %	Mn %	Mg %	Zn %
1	Silumin	12.8% Si 0.36% Fe	10	900	Trace	2.72					
2	Al-Cu	2.55% Cu 0.15% Si 0.20% Fe	15	900	...do...	<0.01	0.03				
3	Al-Cu+Silumin	1:1	15	900	...do...	<0.01	Trace				
4	Al-Ni	2.05% Ni 0.15% Si 0.25% Fe	15	900	...do...	<0.01		Trace			
5	Al-Fe	2.20% Fe 0.18% Si	15	900	...do...	<0.01					
6	Al-Mg	4.2% Mg 0.20% Si 0.18% Fe	15	900	...do...	<0.01				0.12	
7	Mg-Al+Silumin	1:1	15	900	...do...	<0.01				Trace	
8	Al-Zn	6.30% Zn 0.18% Si 0.18% Fe	15	900	...do...	<0.01					0.18
9	Al-Mn	0.93% Mn 0.26% Si 0.39% Fe	15	900	...do...	<0.01			0.05		
10	Al-Mn+Silumin	1:1	15	900	...do...	<0.01			Trace		
11	Duraluminum		15	900	...do...	<0.01	0.02		Trace	Trace	
12	Y-alloy (Cu-Ni)		15	900	...do...	<0.01	0.34	Trace			
13	Y-alloy+Silumin	1:1	15	900	...do...	<0.01	Trace	Trace			

At the following treatment of the aluminum obtained it could be refined further, so that the percentage of silicon thereof could be lowered to a few hundredths of a percent only.

The term Silumin as used in the above example and throughout this specification constitutes a trade name for a commercial eutectic mixture of approximately 87% aluminum and 13% silicon (see British Chemical Abstracts, B 1926, p. 831 and B 1931, p. 722).

During the continued work of the inventor it has been found possible, while observing certain directions for the working process, in one cycle of operation to produce aluminum containing exceedingly small quantities of silicon, iron and other impurities, so that the product can be regarded as chemically pure aluminum.

It has been found that if it be seen to that the alloy forming the starting material contains elements forming compounds with silicon, perhaps also with other alloy constituents, the latter will be fixed as silicides and remain in the undissolved portion, so that the refining product will be entirely free from silicon and from the elements tied up thereby. As such elements may be particularly mentioned Mg, Fe, nickel, copper and manganese. In carrying the method into effect in this manner it is thus seen to in the selection of a suitable primary material that the latter will always contain one or more of such elements. If the primary material does not contain any of the elements furthering the refining, small quantities thereof are added to the charge.

On the other hand, if the primary material contains only small quantities of silicon or no silicon at all, as in copper-aluminum-alloys, aluminum alloys containing silicon, such as Silumin, are added to the charge, whereby a complete removal of all alloy constituents, such as copper, manganese, nickel, iron, chromium, titanium, magnesium and so forth, is obtained.

To obtain best results, the percentages of the various constituents in the primary charge are thus controlled in accordance with the working directions above described.

The results of a few analyses have been compiled in the following table, said analyses referring to the products obtained, it being possible to derive from said results what effect will be obtained when said refined elements are present.

It will be seen from test No. 2 that the finished product contains only 0.03% Cu. Here, the primary material was a copper-aluminum alloy. In test No. 3, the same alloy was used as primary material, but with the addition of silicon, and by reason of the increased quantity of silicon even the small copper percentage from test No. 2 (0.3%) had disappeared altogether, so that only a trace thereof could be shown to be present.

Furthermore, it appears from test No. 6 that with the pure Al-Mg-alloy with a low content of silicon a final product with a content of 0.12% Mg was obtained. This percentage was reduced to a mere trace only when the same alloy was treated together with silicon (test No. 7).

In test No. 9, the final product contained 0.05% Mn, but even this low percentage was reduced to a trace only when the same primary material was employed together with silicon (test No. 10), the formation of manganese-silicide being thus promoted.

In test No. 12, where the primary material was a copper-nickel-aluminum-alloy, the final product contained 0.34% Cu, which percentage was reduced to a trace, however, when the same alloy was treated together with silicon (test No. 13). In test No. 11, where the primary material contained, interalia, Mg and Si, the magnesium remained in the original alloy as magnesium silicide.

If magnesium is present in the alloy while the quantities of silicon therein are inconsiderable,

the magnesium enters into the lead. This is also the case with respect to zinc. Zinc does not form silicide, but the refining may be carried into effect in a satisfactory manner, as it is possible to extract for a long time, before, with an ordinary aluminum scrap alloy, sufficient zinc is dissolved in the lead that it is necessary to refine it out. This may then be effected by cooling to 400 degrees centigrade, the major portion of the zinc then crystallizing out on the surface.

It will be seen from the statements in the preamble that the solubility of aluminum in lead amounts to about 1.5% at 1000 degrees centigrade. By making use of a special working method it is possible, however, to obtain a much higher solubility, it having been found that there is formed, in the border zone between the lead bath and the charge of the aluminum alloy under treatment flowing on top of it, a lead layer which is much richer in aluminum than corresponding to the ordinary solubility of the lead at the prevalent temperature. Therefore, if this lead layer specially rich in aluminum is continuously removed, it is possible to take advantage of this layer formation, so that an increased yield of extracted aluminum is obtained in a definite quantity of lead. That this is a matter of fact will appear therefrom that the inventor has succeeded on various occasions to dissolve 3% of aluminum at about 900 degrees centigrade, whereas the solubility at this temperature really is at about 1%.

In consequence of this it may be advantageous to avoid working with powerful stirring of the lead-aluminum-alloy in such manner that a uniform supply of the aluminum alloy is had at the surface of the bath and the layer specially rich in aluminum is continuously removed, while the rest of the lead then always holds about 1–2% of aluminum, corresponding to the solubility at the prevalent temperature.

The annexed drawing illustrates by way of purely diagrammatic representation a contrivance for carrying the method according to the invention into effect.

1 designates a container in which the extraction of aluminum takes place. This container or furnace is provided with a lid 2, wherein or beneath which is provided an electric heating device 3, such as a resistance heater adapted constantly to keep the charge 4 and the lead 5 at the desired temperature. Provided in the lower portion of the container is a quantity of lead holding about 1–1.5% of aluminum, if an operating temperature of 1000 degrees centigrade is maintained. The aluminum scrap alloy to be refined is supplied continuously onto the surface of the lead bath through means not illustrated, and underneath this alloy there is formed a relatively thin lead layer which is particularly rich in aluminum. The formation of this Al-rich layer is promoted by the heating from above, since the upper layers will obtain a specially high temperature thereby.

This layer is indicated by the hatched portion. This layer which is rich in aluminum is drawn off continuously through the tap pipe 6, which is provided with a cut-off device preventing the aluminum alloy also from flowing out. The lead which is rich in aluminum flows down into a cooling receptacle 7, wherein it is cooled comparatively slowly to about 450 degrees centigrade, whereat the aluminum dissolved in the lead is precipitated and flows up on top of the surface of the lead bath, whence it is removed with the aid of some or other suitable contrivance, such as scraping devices or the like. It may be advantageous to make use of a plurality of cooling receptacles and to utilize the quantity of heat liberated in the cooling for various purposes.

Provided in the bottom of the receptacle 7 is a pipe 8, and this pipe communicates with a lead pump 9 adapted continuously to convey the lead from which the aluminum has been separated over to a store receptacle 10 whence the lead is supplied continuously to the extraction furnace 1. During the conveyance of the lead from the receptacle 7 to the receptacle 10 the temperature falls somewhat, so that the lead in the receptacle 7 attains a temperature of approximately 400 degrees centigrade.

It should be stated that it is possible to make use of certain alloys of lead and other elements as a lead extracting agent. In the above-mentioned method, the quantities of aluminum dissolved in the lead are rather limited on account of the low solubility of aluminum in the lead at the temperatures brought into consideration. Certain advantages may be obtained, therefore, by the use of lead alloys adapted to increase the solubility of the aluminum.

It is understood that the primary alloy to be refined or from which aluminum is to be produced may be constituted either by an alloy produced by chemical reduction of aluminum silicate and the like with carbon or by a scrap alloy containing the ordinary, above-stated alloy constituents. It is also possible to start with advantage from ordinary electrolytic aluminum and to work the latter according to the invention for the production of super-aluminum.

After non-recurrent treatment, the crystallized aluminum will contain some lead. The solubility of lead in solid aluminum is only 0.2%, however, and it has now been found that it is possible to remove the excess lead by plastic deformation at about 500 degrees centigrade by the excess lead being pressed in the molten state up to the surface so as permit of being removed. At subsequent remelting the percentage of lead may be reduced to 0.2% Pb. Such contamination, however, has no influence on the electric or mechanical properties of the aluminum. On the contrary, it has been found that a small addition of lead increases the corroding resistance and, to a certain extent, the capability of the metal of being rolled in the cold state.

The importance of being able inexpensively to produce entirely pure aluminum on a comercial scale will apear from the following: 99.5%-aluminum is known to be used in hardened condition as electric wiring material. It then possesses a conductivity which is 62.5% of that of copper, whereas an aluminum containing 99.97% Al has a conductivity which is 64.6% of that of copper. According to the above, aluminum holding 99.8% Al and 0.2% lead would have the same conductivity as aluminum of 99.97%.

In this connection it should be stated that in the so-called three layer electrolysis ordinary electrolytic aluminum may be refined up to pure aluminum (from 99.5 to 99.99% Al). According to this method, copper is added to the aluminum, and this copper alloy forms the anode. The electrolyte frequently consists of cryolite in combination with barium fluoride to make the bath heavier than aluminum. The pure aluminum now collects in the course of the electrolysis on the surface of the electrolyte whence it can be removed. However, a drawback of this method is that it con-

stantly supplies Si and Fe to the copper-aluminum at the supply of crude aluminum, the percentage of these elements being then gradually increased so much that the copper alloy will finally have to be exchanged and fresh alloy added. After that, the copper must be refined out of the used alloy to permit of being used anew. This makes that the tension conditions are displaced in the electrolysis, which is also rendered expensive by the constant exchange of anode material.

On the other hand, if refining with the aid of lead is made use of, a metal will be produced which holds 99.8% Al and 0.2% Pb without any other impurities. If it is desired to refine this metal up to 99.99%, the aluminum may be alloyed with the copper as usually. By the addition of more and more aluminum, the percentage of lead in the anode is increased, but since this metal is practically insoluble both in aluminum and in copper, the excess of lead falls below the copper alloy, whence it may be removed, without the electrolysis being interrupted. Hereby the electrolysis may be carried out practically without any interruptions in operation, at constant tension conditions and without exchange of the anode material. The three layer electrolysis is thus simplified to a great extent and may be operated under altogether other economical conditions than hitherto.

There are metals other than lead which may be used in a similar manner as extraction agents, such as bismuth, cadmium and thallium. Bismuth dissolves comparatively larger quantities of aluminum at higher temperatures than does lead, and this is also the case with cadmium. For economical reasons the inventor has found, until further notice, that lead is nevertheless to be preferred. It may appear surprising that an extraction process based on comparatively so small a solubility of the metal to be extracted in the extraction agent may be carried out in an economically satisfactory manner. Calculations have shown, however, that the expenses, particularly when the process is carried out continuously, become comparatively small, which partly depends on the fact that the specific heat of the lead is very low. The costs of energy are limited, on the whole, to the covering of the loss of heat involved by the necessary cooling of the heated body of lead to precipitate the aluminum, that is to say from the operating temperature to the precipitation temperature of about 450 degrees centigrade.

It has been found, furthermore, that this process can be materially improved by the extraction procedure being carried out under pressure, inasmuch as the solubility of the aluminum in the extraction metal, such as lead, is increased thereby. A pressure of a few atmospheres entails good results. In the method according to the invention, the extraction container is formed altogether closed, and is provided with suitable sluicing means to introduce the raw material and to draw off the lead-aluminum-alloy.

What I claim is:

1. The method of refining aluminum which comprises heating a mass comprising an aluminum containing alloy and an aluminum dissolving metal from the group consisting of lead, bismuth, cadmium, thallium and mixtures thereof, with the solution of at least a portion of the aluminum in the metal, separating the portion of the aluminum alloy not dissolved in said metal from said aluminum-metal solution, thereafter concentrating the aluminum of said solution by cooling said solution and, subsequently, separating the concentrated aluminum from said metal.

2. The method according to claim 1 wherein the concentrated and separated aluminum is dissolved in an aluminum-dissolving metal as defined in claim 1, and is again concentrated and separated from said metal, whereby a purer product is obtained.

3. The method according to claim 1 wherein said aluminum-dissolving metal is an alloy of metals from the group consisting of lead, bismuth, cadmium and thallium, said alloy having a greater capability of dissolving aluminum at high temperatures than any of its pure elemental components separately.

4. The method according to claim 1 wherein the aluminum-dissolving metal and the aluminum alloy are heated to a temperature in excess of 1000 degrees centigrade.

5. The method according to claim 1 wherein the refining process is operated in a continuous manner by continuously removing the aluminum-metal solution formed in the process, from the heated mass, and, then, returning said metal, after separation of the same from said solution by cooling, to said heated mass.

6. The method of refining aluminum which comprises heating an aluminum alloy to a high temperature together with lead, whereby at least a portion of the aluminum of said alloy is dissolved in the lead, continuously withdrawing the portion of the resulting aluminum lead solution at the border zone between the lead and the aluminum alloy from the heated mass, cooling this aluminum-lead solution to concentrate the aluminum therefrom, removing the concentrated aluminum from the remainder of the solution, and returning said remainder to said heated mass.

7. The method of claim 1 wherein the heated mass is subjected to vigorous stirring.

8. The method of claim 1 wherein said aluminum alloy is an aluminum-silicon alloy.

9. The method of claim 1 wherein said recovered aluminum is relieved of dissolved quantities of said aluminum-dissolving metal by subjecting said recovered aluminum to plastic deformation of about 500 degrees centigrade, and pressing out said metal in the molten state from said aluminum.

10. The method according to claim 1 wherein said aluminum alloy in a finely divided and heated state is added to said metal in a molten state to form said heated mass.

11. The method according to claim 1 wherein said aluminum-dissolving metal is lead.

12. The method according to claim 1 wherein said aluminum-dissolving metal is bismuth.

13. The method according to claim 1 wherein said aluminum-dissolving metal is cadmium.

14. The method according to claim 1 wherein said refining process is carried out at super-atmospheric pressure.

15. The method according to claim 1 wherein said refining process is carried out at between 2 and 3 atmospheric pressure.

16. The method according to claim 1 wherein said refining process is conducted in a continuous manner and under pressure.

17. The method of claim 1, wherein the composition of said aluminum-containing alloy comprises silicon and a metal which forms with

silicon a material which is more insoluble in molten aluminum than is silicon.

18. The method of refining aluminum containing as an impurity silicon and a metal which forms with silicon a material which is more insoluble in aluminum than is silicon, which comprises heating said impure aluminum with lead to at least 1000° C. with the solution of at least a portion of the aluminum in the lead, separating the portion of the impure aluminum not dissolved in the lead from the resulting aluminum lead solution and, thereafter, separating the aluminum from said aluminum-metal solution by cooling the solution.

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