United States Patent [19]

Kibby

[11] 3,929,456

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[54] **CARBOTHERMIC PRODUCTION OF ALUMINUM**

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[73] Assignee: **Reynolds Metals Company,** Richmond, Va.

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[21] Appl. No.: **407,271**

Related U.S. Application Data

[63] Continuation-in-part of Ser. No. 250,748, May 5, 1972, abandoned.

[52] **U.S. Cl.** ..... **75/10 R;** 75/68 A

[51] **Int. Cl.²** **C22B 21/02**

[58] **Field of Search** ..... 75/10 R, 10 A, 10 P, 10 V, 75/68 R, 68 A

[56] **References Cited**

UNITED STATES PATENTS

2,829,961	4/1958	Miller et al.	75/68 A
3,607,221	9/1971	Kibby	75/68 A
3,721,546	3/1973	Shiba et al.	75/10 R
3,723,093	3/1973	Shiba et al.	75/10 R

Primary Examiner—M. J. Andrews

Attorney, Agent, or Firm—Glenn, Palmer, Lyne, Gibbs & Clark

[57] **ABSTRACT**

A carbothermic process for the production of aluminum metal contaminated with no more than about 5 weight percent of aluminum carbide in a single furnace operation which comprises contacting a feed comprising an oxide of aluminum and a carbon-containing compound such as aluminum carbide in an open arc furnace wherein heat is applied via an open arc in a manner such that only the surface of the charge is raised to reaction temperature and the aluminum which is formed in the condensed state is then allowed to flow at a temperature sufficiently low to prevent substantial contamination with aluminum carbide.

12 Claims, 4 Drawing Figures

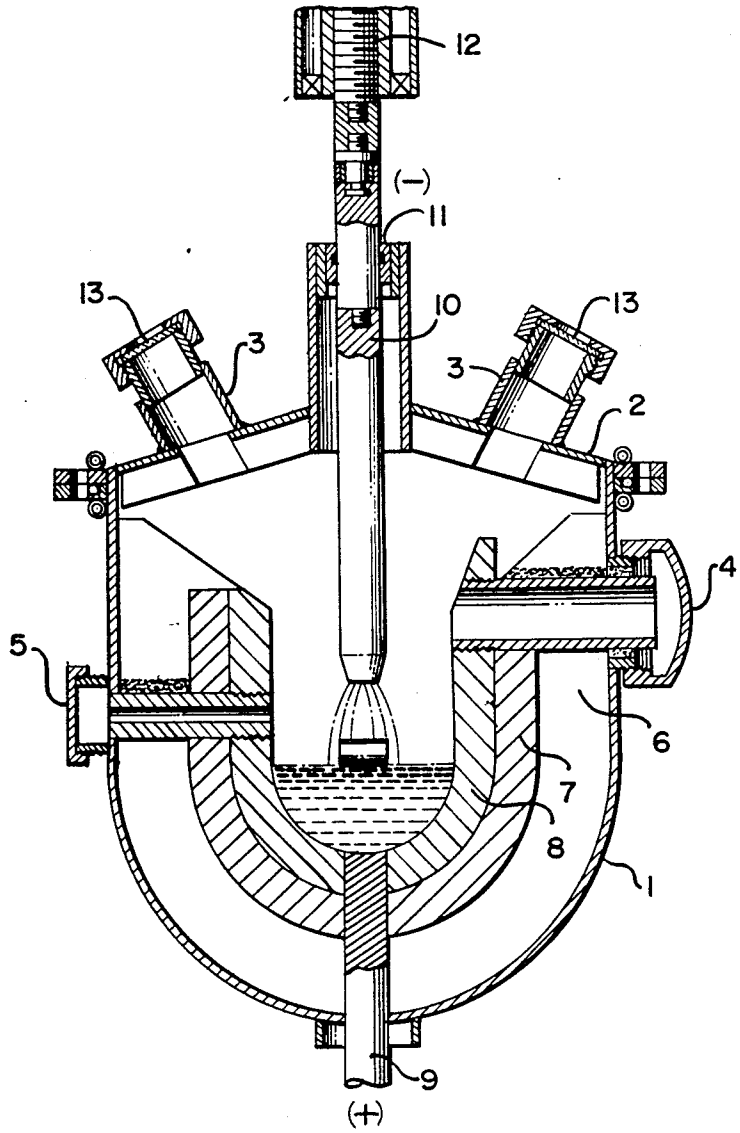


FIG. 4

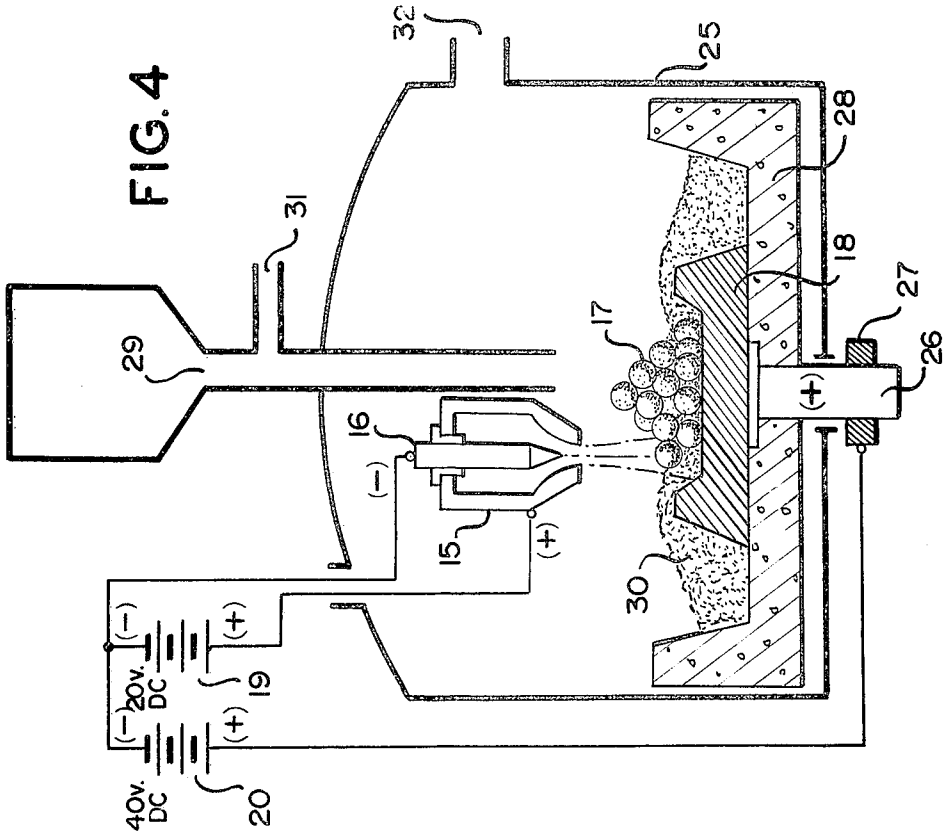

32
25
28
31
18
27
17
29
26
(+)
16
(−)
15
(+)
30
(−)
(+)
20v. DC
19
(−)
(+)
40v. DC
20

FIG. 1

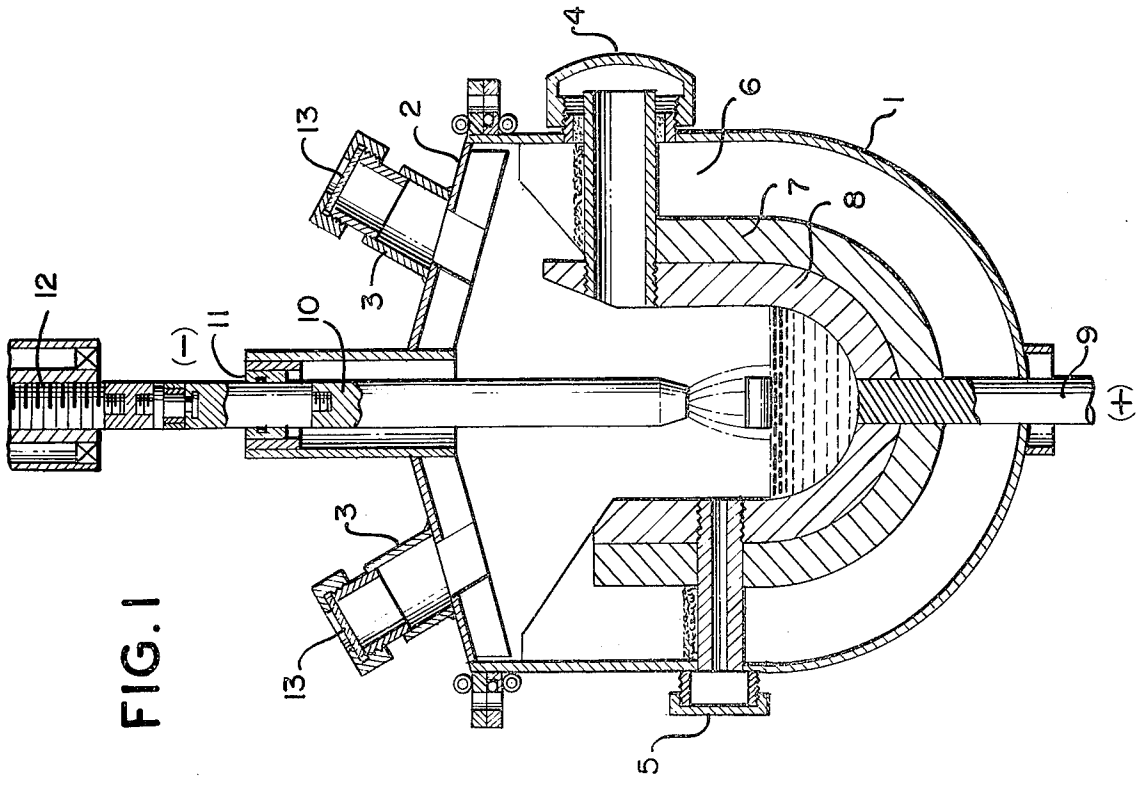

4
6
1
13
2
7
8
12
(−)
11
10
3
9
(+)
3
13
5

FIG. 2

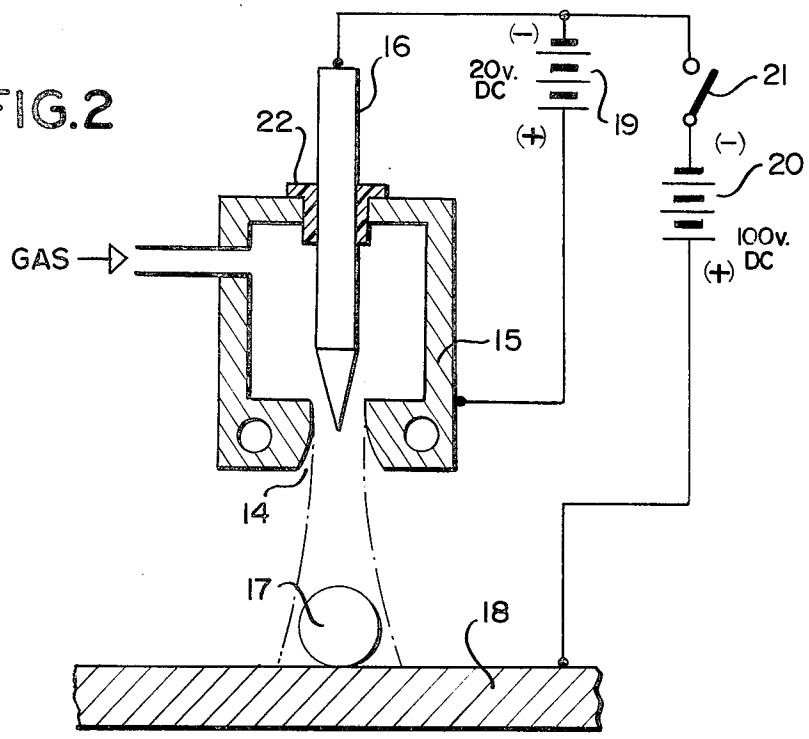
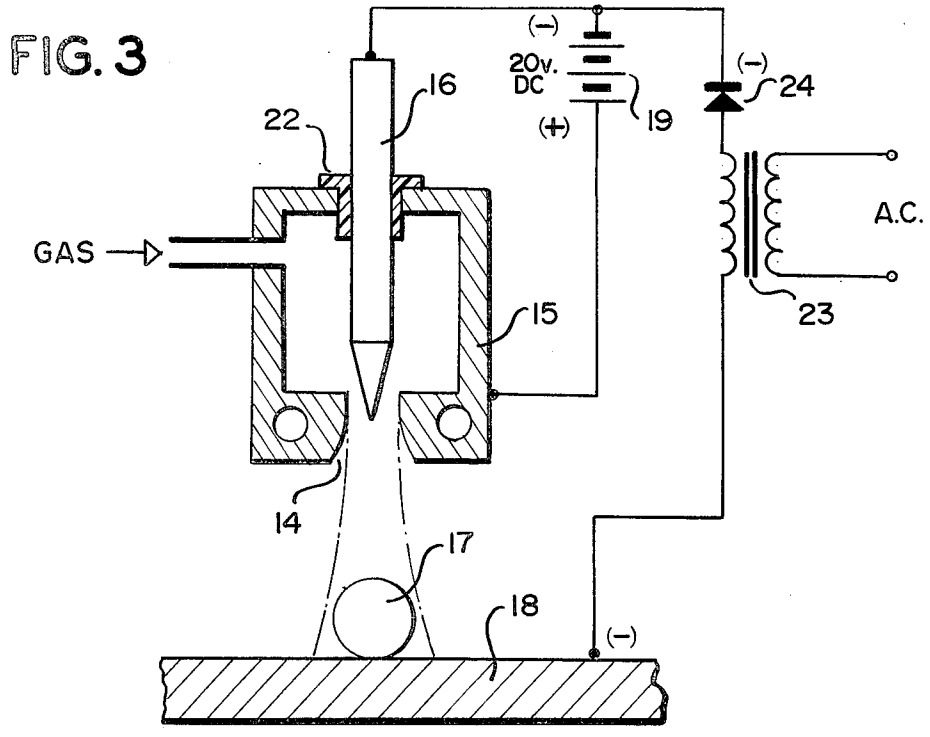

16
(−)
20v.
DC
(+)
19
21
(−)
20
100v.
DC
(+)
22
GAS
15
14
17
18

FIG. 3


(−)
20v.
DC
(+)
19
16
22
(−)
24
A.C.
GAS
15
23
14
17
18
(−)

CARBOTHERMIC PRODUCTION OF ALUMINUM

RELATION TO OTHER APPLICATIONS

This application is a continuation-in-part of Ser. No. 250,748, filed May 5, 1972 now abandoned.

FIELD OF THE INVENTION

This invention relates to a new and improved process for producing aluminum metal having no more than about 5 weight percent of aluminum carbide under carbothermic conditions from feed comprising an oxide of aluminum and carbon containing compounds in a single furnace operation.

DESCRIPTION OF THE PRIOR ART

Even the most cursory inspection of the prior art relating to the thermal production of aluminum will immediately indicate that there has been much activity by many people in an attempt to adequately define a process which would replace the conventional electrolytic method of preparing aluminum. The art has long been aware of the many theoretical advantages which flow from the use of a thermal reduction process for the production of aluminum as opposed to an electrolytic method. Unfortunately, there has been one drawback to the heretofore suggested thermal processes for producing aluminum. That drawback has been that after all was said and done, the simple fact remained that there was no way to produce significant amounts of aluminum in a substantially pure state from a thermal process.

It is to be immediately understood that the difficulty in producing aluminum with respect to thermal processes does not reside in the formation of the aluminum via reduction of the alumina-bearing ore, but rather, in the recovery of the aluminum in a substantially pure state. The patent art, as well as the literature, is full of theories and explanations with respect to various back reactions which can take place between aluminum and the various carbon-containing compounds in the feed. The sum total of all prior art efforts is simply that there is no commerical process today for preparing aluminum other than by an electrolytic process.

In general, it can be stated that the prior art processes for the thermal production of aluminum can be divided into two general categories, i.e., one wherein aluminum is formed in the vapor state and the other being where aluminum never reaches the vapor state, i.e., it is formed as liquid aluminum.

With respect to the processes wherein aluminum is produced in a vapor state, the major difficulty which was encountered was due to the fact that aluminum vapor is extremely reactive with carbon monoxide which is inherently formed in the reaction, thereby producing aluminum-carbon compounds. The patent and literature art contains many teachings directed towards ways of minimizing the reaction of aluminum and carbon monoxide, but, in general, the heretofore proposed solutions have been impractical. One solution to this general problem wherein aluminum is formed in the vapor state is disclosed and claimed in U.S. Pat. No. 3,607,221. Although the process of this patent does result in the production of aluminum in a substantially pure state, nevertheless, extremely high operating temperatures are involved which leads to problems with respect to materials of construction.

The art has long been aware that the aforementioned difficulties with respect to back reaction of aluminum with carbon monoxide could be obviated if, indeed, the aluminum is never in a vapor state. Thus, it is known that if a process is carried out under conditions such that aluminum is formed in the liquid state, then this liquid aluminum will be relatively inert with respect to carbon monoxide, thereby resulting in a process which should be free of back reaction products.

Although there are many processes disclosed and claimed in the prior art wherein aluminum is produced in the liquid state, the simple fact remains that none of these processes have met with success with respect to initially producing aluminum containing low percentages of aluminum carbide. The reason for the failure of the prior art processes can be easily understood when one considers the fact that aluminum carbide is soluble in molten aluminum and the solubility of aluminum carbide in aluminum increases with increasing temperatures. Aluminum carbide is present in a carbothermic reduction process, either due to the fact that it is introduced as a reactant or is inherently formed during the reduction reaction. This is because aluminum is highly reactive with carbon and certain aluminum-carbon compounds to form aluminum carbide. Thus, since the prior art processes by necessity had to be carried out at elevated temperatures in order to form aluminum, the liquid aluminum formed, although relatively unreactive with respect to the carbon monoxide, nevertheless, does dissolve the aluminum carbide which was also inherently present in the system, thereby resulting in carbide-contaminated aluminum. It should be pointed out that aluminum containing greater than about 5 weight percent of carbide contamination is extremely undesirable for many reasons, including the fact that it sets up to a hard non-flowable mass as the temperature is decreased slightly from reaction temperature thereby resulting in severe practical difficulties with respect to transferring it from one place to another except at elevated temperatures. Additionally, it must be realized that electrical energy has been spent to produce the aluminum and if it is contaminated with more than about 5 weight percent of aluminum carbide, the additional energy which must be used in subsequent recycle operations renders the process noncompetitive commercially with respect to power consumption.

A whole body of prior art has arisen on various ways of removing aluminum carbide in admixture with aluminum and there have been many patents and literature articles on this subject.

This invention is not concerned with the removal of aluminum carbide from aluminum but is concerned with a carbothermic process for the preparation of aluminum which never has a substantial amount of aluminum carbide present to begin with. This invention accomplishes that which workers in the art have long hoped could be done and what other learned people have thought was theoretically impossible to do.

In fact, as will be seen from the actual working examples in this application, aluminum has, indeed, been produced in a significant amount via carbothermic reduction, said aluminum having an insignificant amount of aluminum carbide admixed therewith.

SUMMARY OF THE INVENTION

This invention is directed towards the carbothermic reduction of alumina-bearing ores and it has for its significant feature and point of departure from the

heretofore practiced processes certain critical characteristics. At the outset, it is noted that this invention is basically a dual temperature process, i.e., aluminum is formed at a first high temperature reaction zone and thereafter collected at a significantly lower temperature. The lower temperature where aluminum is collected is chosen such that it is physically impossible for aluminum carbide to be dissolved by the aluminum since, as has heretofore been stated, the ability for aluminum to dissolve unreacted aluminum carbide is strictly a function of temperature. Thus, the aluminum is produced at one temperature and is allowed to flow over cool unreacted charge to a second zone maintained at substantially lower temperature wherein the aluminum is physically incapable of dissolving aluminum carbide in appreciable amounts.

An extremely important characteristic of the novel process of the invention resides in the type of heat which is applied on the first zone. The novel process of this invention absolutely and positively requires the fact that only a small portion of the charge be heated to the reaction temperature at any given time whereas the vast majority of the charge must remain at a temperature significantly less than the reaction temperature. This requirement is absolutely contrary to any of the prior art processes which have heretofore been practiced. It should become immediately apparent that if one is conducting a thermal operation, it appears almost logical that one of the main objectives should be to heat the charge in the reaction zone to the reaction temperature as quickly as possible and as uniformly as possible in order to insure a complete reaction. It is not too surprising that the prior art workers strove to accomplish just that. It has now been discovered that if, in fact, this uniform type of heating, above described, is carried out, aluminum in the pure state simply will not be formed. The novel process of this invention does not utilize uniform heating of a charge and, in fact, at the hottest part of the reaction zone, the majority of the charge is not at reaction temperature at any given time and is deliberately kept this way.

In fact, the uniform heating of large portions of the charge material to reaction temperature is perhaps the main reason why the prior art workers failed to produce substantially pure aluminum in a carbothermic process which produced condensed aluminum instead of an aluminum vapor. The reason upon hindsight appears to be rather obvious. If the entire charge is heated to reaction temperature, then the aluminum which is formed and which flows over the charge must contact carbon and aluminum carbide inherently present in the furnace at elevated temperature and a low aluminum carbide containing product is not obtained in a single furnace operation.

It has now been discovered that low aluminum carbide contaminated aluminum can be produced in a single furnace operation providing the charge material is heated non-uniformly, i.e., substantially only the surface of the charge is heated such that this surface-heated material produces aluminum and then the aluminum is allowed to flow over the non-reacted portion of the charge which is not at elevated temperatures and, thus, renders it substantially impossible for the formed condensed aluminum to dissolve aluminum carbide. Quite obviously, another portion of the charge is then exposed to this high heat and the cycle is continued.

DESCRIPTION OF THE DRAWINGS

FIG. **1** represents a furnace suitable for carrying out the instant process. Furnace shell **1** is fitted with lid **2**, and sight tubes **3**. Access tubes **4** and **5** can also be provided. Insulation is shown at **6** and **7**. The crucible **8** is connected to the positive terminal of a D. C. supply through graphite rod **9**. Negative electrode **10** is vertically adjustable through screw mechanism **12** and is insulated from furnace lid **2** through electrically non-conducting vacuum gland **11**.

FIG. **2** represents the configuration of a plasma arc equipped for DC transfer to the charge which can be used for carrying out the instant process.

FIG. **3** represents another configuration of a plasma arc which can be used for carrying out the instant process wherein the arc is equipped for half-wave DC transfer.

FIG. **4** represents a furnace suitable for carrying out the process of the instant invention utilizing the plasma arc configuration of FIG. **2**.

DESCRIPTION OF THE PREFERRED EMBODIMENTS

As has heretofore been stated, the novel process of this invention requires a two temperature operation, a high temperature zone wherein the reaction for the production of aluminum is driven forward and a low temperature zone for transporting and collecting aluminum while preventing it from substantially dissolving any unreacted carbide.

To the furnace is charged an aluminum oxide-bearing material in admixture with a carbon-containing compound which is preferably aluminum carbide and/or carbon. Since it is desired to produce substantially pure aluminum, the aluminum oxide-bearing material is preferably alumina of a high degree of purity, i.e., Bayer alumina, but it is to be understood that the process of this invention is equally operable with impure forms of alumina and aluminum oxycarbides and, although the resulting product will still be free from carbide contamination, it will contain the impurities normally present in the alumina ores.

The ratio of aluminum oxide-bearing compounds to the carbon-containing compounds is preferably adjusted such that the composite charge contains a 1:1 ± 0.05 atomic ratio of carbon to oxygen.

The process of this invention can be carried out at any pressure higher than about 0.1 atmosphere. It has been found that at pressures below about 0.1 atmosphere liquid aluminum is not made under any practical set of conditions. On the other hand, it is recognized from thermodynamic considerations, that vaporization losses decrease as the pressure increases above about 0.1 atmosphere. However, the use of higher pressures requires equipment capable of handling such pressures so it is evident that the choice of pressure above about 1.0 atmospheric involves an economic balance between the energy lost because of vaporization and the cost of equipment. In general, practical pressures for the instant process range from a system pressure of about 0.5 to 10 atmosphere with from 1-5 atmospheres being preferred.

It is also noted that an embodiment of this invention which will be later described involves the use of a plasma torch. In such a situation, the torch itself exerts a pressure depending on power density. Thus, it is recognized that the pressure directly under the torch in the

reaction site can be higher than the pressure away from the torch.

As is well known, the particular pressures and temperatures which are utilized are mutually dependent on each other as is obvious from a rudimentary consideration of the laws of thermodynamics. There exists in the literature works of many authors setting forth temperatures which are necessary for various pressures. However, the exact temperatures which are stated to be necessary for any given pressure vary depending upon the author's interpretation of the thermodynamic data. In general, however, a temperature of about 2500°K is necessary for operation at 1 atmosphere. Therefore, from a practical point of view, it is difficult to specify the exact temperature which is necessary to drive the reaction forward for any given pressure. Additionally and perhaps more significantly, such a recitation of specific temperatures is of no practical significance since in an actual furnace operation the instruments used to measure the temperature utilize optical principles and the charge is hidden from view by the presence of electrodes. The temperatures which are utilized in the novel process of this invention can be described by stating that for any given pressure a sufficient heat must be used to drive the reaction forward but too high a temperature will cause the aluminum produced to volatilize out of the furnace. However, from a practical point of view the reduction of alumina-bearing ores to produce aluminum absorbs heat and the reaction itself controls the temperature.

It has been found that irrespective of all the above-stated principles with respect to temperature and pressure there exists a very convenient manner for carrying out the process of this invention. It has been discovered that an accurate control of the reaction can be accomplished by striking an open arc, as hereinafter defined, to the surface of the charge to be reduced and by regulating the electrical density of the arc striking the charge. It has been found that if the electrical density is maintained between 10–50 kilowatts per square inch of charge struck by the arc, the reaction will proceed in a desirable manner. When the arc density has passed its minimum threshold value the very occurrance of the reaction controls the temperature at higher arc densities because the reaction absorbs heat.

By the expression "electrical density per square inch of charge" is intended to mean measurement of the total electrical power supplied to the arc (i.e., amperes × volts) divided by the area of charge struck by the arc. With respect to the use of a plasma torch, calculations of electrical density ignore the internal power supply and only the transfer current is taken into consideration.

A convenient method of measuring the total area struck by the arc is to use an optical instrument. The portion struck by the arc will glow and its area can be measured. In the case where the surface of the charge is irregular, such area can be measured by striking the arc to the bed of the furnace without putting in any charge.

As has heretofore been stated, one of the criticalities of the novel process of this invention resides in the fact that the charge which is fed to the first high temperature zone does not reach a state of uniform heat as has happened in prior art processes. For reasons which have been previously set forth, it would appear rather obvious that the aluminum which is formed in the system will flow down the charge and if the charge is at an elevated temperature, the aluminum will dissolve unreacted aluminum carbide, thereby resulting in a carbon-contaminated product.

It has now been discovered that one convenient method for producing the type of heating with which this invention is concerned resides in the use of an open arc wherein the adjustable electrode is negative with respect to the charge to be reacted. The term "open arc" is used herein to mean an arc from an electrode which is not in physical contact with the charge to be reacted. In this embodiment, a charge stock is introduced into the furnace and an open arc is struck from a suitable electrode such as a conventional graphite electrode. If the electrical power of the arc is such that it produces an electrical density of 10–50 kilowatts per square inch of charge struck by the arc and, more desirably, 25–35 kilowatts per square inch, then the surface of the charge will be heated to the desired temperatures. However, this condition alone does not ensure that the charge will not be heated in a uniform manner. In order to accomplish this, it has been found that the arc should be an intermittent arc, i.e., it should be on for a period of time and off for a period of time with respect to a given area of charge. This type of operation will hereinafter be referred to as an intermittent operation and what is meant by this expression is the fact that a particular portion of the charge stock is subjected to direct electrical heating via an open arc only from 10–50% of the total time. Thus, by way of a convenient example, the arc can be struck to a charge stock for a period of time of one minute then be turned off for 2 minutes, then be restruck for another minute, etc. In the preferred embodiment of this invention, it is desired that the arc be applied for a period of time ranging from 1/120 to 90 seconds, and thereafter be turned off for the appropriate periods of time such that the heating only occurs from 10 to 50% of the total time.

It is to be immediately understood that there are other ways of achieving this intermittent heating rather than merely turning the arc on and off. Thus, for example a plurality of electrodes can be used over a rather wide surface area and each electrode be turned on and off at appropriate periods of time within the guidelines above set forth. Additionally, the electrode can be left on continuously but moved over the surface of a charge stock by mechanical means such that the total amount of time that the arc strikes a particular surface area is between 10 to 50% of the total time. In like manner, the electrode can be left on continuously and the charge moved in and out of the arc by mechanical means such that the arc strikes a given area 10 to 50% of the total time.

It has been found to be advantageous to use an open D.C. arc with the adjustable electrode negative with respect to the charge. The reason for this is that the negative electrode receives less of the heat while emitting electrons than the anodic charge receives. Under the D.C. arc operation with the movable electrode negative, the charge receives most of the heat and the electrode remains cool enough to avoid excessive volatilization of carbon. This minimizes the opportunity for hot carbon vapor to come into contact with the condensed aluminum product where it could form aluminum carbide, which, in turn, would readily go into solution in the aluminum product.

It has been found that a graphite electrode which is negative with respect to anodic aluminum can melt aluminum without adding more than 0.3% Al_4C_3 to the

aluminum.

The open arc is viewed as desirable because the surface temperature of the charge has an opportunity to decrease rapidly upon arc interruption, thus permitting the majority of the charge to remain at the required low temperature as a result of heat transfer to colder portions of the furnace during periods of arc interruption. During the high temperature operation, above-described, the carbon monoxide which is formed is removed from the system while the aluminum is in a condensed state so that for practical purposes substantially no aluminum compounds are formed via back reaction.

The second stage of the novel process of this invention resides in removing the condensed aluminum at a temperature such that substantial amounts of aluminum carbide simply cannot be dissolved therein. The temperature of the second stage should not exceed 1250°C and preferably should be at a temperature ranging from 670°–1000°C.

In copending application Ser. No. 250,758, filed May 5, 1972, there is disclosed one technique for accomplishing the same which resides in maintaining a liquid pool of aluminum inside the furnace and floating the charge thereon, heating the charge in the manner above described so as to form aluminum in the condensed state and then allowing the formed aluminum to flow into the liquid pool of aluminum which is maintained at the temperatures above stated. As is well known in the art, a liquid metal is an excellent conductor of heat and it will remove heat from the arc and charge to areas where the heat can be lost through furnace walls, roof, and floor in a rapid manner thereby insuring the necessary temperature controls.

It is to be understood, however, that although the maintaining of a liquid pool of aluminum is an effective manner of insuring the temperature control above described, there are other ways of accomplishing the same, such that the maintaining of a liquid pool of aluminum is not absolutely critical in the novel process of this invention.

It has been found, for example, that the action of an open arc has a tendency to blow the aluminum which is formed away from the unreacted charge such that the condensed aluminum rapidly cools and when it passes over the unreacted charge, it is a sufficiently low temperature such that it cannot dissolve appreciable amounts of aluminum carbide.

The formed aluminum can also be removed from the unreacted charge simply by mechanical means. Thus, for example, a sloping hearth can be used such that the formed condensed aluminum immediately flows out of the reaction zone and becomes cooled so that when it passes over the unreacted charge it is at a temperature low enough to prevent dissolving of appreciable amounts of aluminum carbide. Other techniques accomplishing the same result include having a dual level furnace hearth such that the upper and lower level of the furnace hearth is connected via passages which are large enough to allow aluminum to flow from the top level to the bottom level, but small enough to prevent the charge from passing from one level to the other. Thus, when the charge stock which is contained on the uppermost level is struck by the arc and liquid aluminum is formed, the condensed aluminum flows through the charge into the second level of the furnace hearth where there is no aluminum carbide for it to contact.

Thus, in order for the novel process of this invention to be effective, it is required that when the liquid aluminum which is produced flows over an unreacted charge, it must be at a temperature below about 1250°C and preferably at a temperature ranging from 670°–1,000°C. On the other hand, after the condensed aluminum is removed from the unreacted charge or other source of carbon then, quite obviously, it can be at any temperature since there will be no unreacted charge or source of carbon and therefore no aluminum carbide which can be dissolved by the aluminum.

Another significant aspect of the novel process of this invention is the fact that because an open arc is used, it is possible to use a closed furnace rather than having one which is exposed to the atmosphere. The use of a closed furnace has an additional benefit from an environmental point of view since it dramatically minimizes the gases which must be treated to remove the pollutants therefrom in order to comply with environmental standards. The closed furnace also permits utilization of the fuel values of the carbon monoxide released by the process. Thus, although it is not necessary to use a closed furnace in the novel process of this invention in order to produce aluminum, nevertheless, the use of a closed furnace does provide added economic advantages from an environmental and energy conservation point of view thereby improving the overall attractiveness of the process.

As has heretofore been stated, it is necessary to use an open arc in order to carry out the process of this invention and although such open arc can be produced by using conventional graphite electrodes in the manner previously described, a preferred embodiment of this invention resides in using plasma torches in order to provide the open arc.

Although the use of graphite electrodes delivers heat at an appropriate power density and provides a pressurized gas effect which tends to move the aluminum produced at the surface of the charge away from the charge, it does suffer from the disadvantage in that it introduces a small amount of carbon to the product. As has heretofore been stated, it has been found that a graphite electrode which is negative with respect to anodic aluminum can melt aluminum without adding more than about 0.3 weight percent of aluminum carbide to the aluminum. However, the use of graphite electrodes has a further practical operating disadvantage in that if the arc is extinguished, the only practical way to re-establish an arc of this power density is to lower the electrode until it touches and makes electrical contact with the charge. This type of action can lead to difficulties with the charge sticking to the electrode. If too much charge is stuck to the electrode, the electrical discharge properties of the electrode are altered to the detriment of the overall operation. In order to avoid such problems, careful control must be exercised over the arc struck between a carbon or graphite electrode and the charge.

The use of a plasma torch eliminates the abovementioned difficulties which can be experienced when using conventional graphite electrodes in that, quite obviously, no carbon is added to the product and the plasma jet has the advantage that the arc can be established even though the jet nozzle is completely removed from the vicinity of the charge. Additionally, if the jet is extinguished for some reason, it can be reestablished without any physical part of the jet-forming equipment being brought into contact with the charge.

Still another advantage of the plasma jet is that in addition to the normal tendency of the arc column to force the produced aluminum away from the charge, the jet comprises additional gas flow (which is an essential feature of the operation of plasma jets) and this additional gas flow adds to the tendency of the arc jet to remove the product aluminum away from the site of the reaction such that it can cool rapidly and not dissolve appreciable amounts of unreacted charge.

A still greater advantage can be obtained from the use of plasma jets when additional circuits are provided wherein a second power supply is connected between the cathode element of the plasma jet and the hearth so that the arc column is drawn not from a negative electrode to the jet nozzle, but instead from the negative electrode to the hearth. In this mode of operation, very little current flows to the nozzle. Most of the current flows to the hearth. A very high heating rate is established at the site of the reaction even though the nozzle of the jet can be a substantial distance (for example, 3–6 inches) away from the charge. This provides ample opportunity for the charge to pass under the jet without being struck by the casing of the jet apparatus.

If for some reason the transfer current, that is, the current from the negative electrode of the jet to the hearth, is interrupted, then the power supply of the internal jet maintains the jet in normal plasma jet operation between the negative electrode and the positive jet nozzle. This then serves as a pilot light to re-establish the jet through the second power supply to the hearth at any time, without having to move the jet physically, relative to the hearth.

This starting and stopping of the transfer power between the negative electrode and the hearth can be so rapid as to occur as often as **60** cycles per second. In fact, one of the preferred embodiments of the plasma jet application to this invention is to use half-wave DC power (for example, 60 cycles half-wave DC) for the transfer power. In this way, for one-half cycle, the transfer occurs with the interior electrode of the plasma torch negative and the hearth positive. When the voltage of the alternating current supply reverses, rectification blocks the transfer current from the hearth back to the internal electrode of the jet.

It can be seen that with this type of half-wave transfer between the internal electrode of the jet and the hearth, the peak power delivered at the target area, mainly the site of reaction, is about four times the average power delivered to the target area. The rate of heating by the plasma jet to the charge is insignificant when the arc is not transferred to the charge compared to when the arc is transferred. Therefore, on the halfcycle where the arc is not transferred to the charge, the charge can be radiating heat to the relatively cool (for example, 1200°C) walls of the furnace. It can be readily understood, therefore, that the very high temperature required for the reaction (2300°C) only occurs in the very thin layer where the jet is striking the charge and down into the charge body and in the surrounding portions of the charge the temperature is much lower. The high temperature zone is only a small fraction of an inch thick when using half-wave DC jet transfer.

No practical way has been devised as yet to make a simple carbon electrode perform on half-wave DC transfer. Once the arc is extinguished due to the return of the voltage to zero it must be relighted by some method which is not convenient with a carbon or graphite electrode.

FIGS. **2** and **3** illustrate the configuration of a plasma torch which can be used in the novel process of this invention.

In both FIGS. **2** and **3**, Number **14** designates the orifice of the plasma jet casing, Number **15**, the casing; Number **16**, the cathode, or emitting electrode of the plasma jet which is insulated from the casing **15** by insulation **22**.

In a normal, conventional plasma jet application, power supply **19** supplies a negative voltage to electrode **16** with respect to the nozzle **14** and casing **15**. Electrons are emitted from the tip of electrode **16** and the force of the gas between the nozzle and the electrode tip prevents direct discharge between the electrode **16** and the nozzle. Instead, the electrons flow out and then come back and attach to the nozzle **14**, leaving a pencil point-shaped jet which is independent of any other anode surface. In other words, this jet will exist and be maintained without having any other anode surface around.

Now, with respect to FIG. **2**, if a second power supply is connected between electrode **16** and another electrically conducting surface **18**, and extra DC voltage (for example, 100 volts) from supply **20** is connected through switch **21**, then the arc transfers and instead of flowing between electrode **16** and nozzle **14**, it now flows between electrode **16** and the target area **18**. If the charge **17** is within the target area of the transfer arc, it is heated rapidly and efficiently, receiving most of the energy delivered in the arc. Under the transfer mode of operation employing second power supply **20**, the charge is heated much more efficiently and more rapidly then if the simple conventional plasma torch were to be brought into the vicinity of the charge.

FIG. **3** illustrates how a half-wave transfer can be applied. Again, power supply **19** maintains the arc whenever the second power supply **23** and **24** is not applying power between the electrode **16** and the charge **17**. When the AC voltage through transformer **23** is in such a direction as to pass through a rectifier **24** to make the electrode **16** negative with respect to the hearth **18** and charge **17**, then on that half-cycle, current will be transferred from the negative electrode **16** to the charge, delivering heat to the charge. When the AC voltage through transformer **23** reverses such that it would make the electrode **16** positive with respect to the hearth **18** then rectifier **24** blocks passage of current and the arc transfer is extinguished. The arc then reverts to the conventional plasma jet mode being drawn between electrode **16** and nozzle **14** and being maintained by power supply **19**.

As has been stated, the advantage of this type of arc transfer resides in the fact that the charge surface is heated to a temperature (for example, 2300°C) sufficiently high to make the reaction between alumina and carbon proceed to make aluminum in the condensed state and carbon monoxide but on the reverse half of the AC cycle where rectifier **24** blocks the passage of current, the charge is not heated and in fact radiates heat to surroundings which are in the neighborhood of 1200°C. Therefore, the interior portion of the charge remains relatively cool, a condition which is essential to avoid pickup of carbide in the aluminum which has been produced. Likewise, the surrounding charge particles which are not struck by the arc in its transfer mode are not heated to a temperature sufficiently high to impart aluminum carbide to the aluminum which rolls across them on its way to the area of the furnace

where the produced aluminum will be held until tapping.

The following examples represent the best mode now contemplated for carrying out this invention:

EXAMPLE 1

A furnace was constructed to permit electric arc heating under a vacuum or controlled atmosphere. Furnace Shell **1** of steel was fitted with lid **2**, sight tubes **3**, and access tubes **4** and **5** (not used in this experiment). Castable refractory of bubble alumina **6** and carbon flour **7** provided heat insulation. Graphite crucible **8** was connected to the positive terminal of a DC supply through graphite rod **9**. Negative electrode **10** was of graphite and was electrically insulated from the furnace lid through electrically non-conducting vacuum gland **11**. Electrode **10** was vertically adjustable through screw mechanism **12**. A vacuum line (not shown) was connected from the furnace lid **2** through a bag filter to a vacuum pump in order to remove carbon monoxide.

Initially the furnace was heated by the application of an arc of **4** kw. power under a vacuum of 15 inches Hg. below atmospheric i.e., about one-half atmosphere system pressure. The arc struck an area estimated to be about three-fourths inch diameter on the crucible at the location of rod **9**.

When the crucible had been warmed enough to show dull red heat after arc terminations, 443 grams of molten aluminum were added. The arc was struck to this aluminum for several minutes to bring its temperature up to about 1000°C.

One pellet, weighing approximately 8 grams, of a mixture in the ratio 58.5 grams Al_4C_3 to 41.5 grams metallurgical grade Al_2O_3, cold pressed with a binder of 5% starch, was floated on top of the molten pool of aluminum, after the metal had been skimmed. The pellet was positioned directly under the negative electrode.

The systems pressure was reduced to the range of 8–10 inches Hg. below atmospheric. An arc of 30V and 500 amps was applied for 30 seconds, during which time the pellet was seen to react and form aluminum, which coalesced with the starter pool.

With the arc off, the furnace was returned to atmospheric pressure with Argon flowing through the sight tubes, the sight glasses **13** were removed and two additional pellets were floated on the metal pool directly under the negative electrode after the pool was skimmed to expose unoxidized melt. The temperature of the pool was about 1100°C.

The sight glasses were replaced, the system pressure was reduced to 8–10 inches Hg. below atmospheric and an arc of 15 kw. was again struck to cover the intersections of the pellet with the metal pool. Arc target area was estimated to be about three-fourths inch diameter. The arc was applied for 60 seconds, during which time aluminum was formed on the exposed surface of the pellet and at the intersection of the pellet with the starter pool until most of the pellet was consumed, the aluminum produced coalescing with the molten pool.

This cyclical process was repeated until 83 grams of charge had been reacted. At no time was the arc applied for more than 90 seconds. The delay time between arcs, because of the furnace charging operation was from 2 to 5 minutes between each arc application. The molten pool was maintained at a temperature between 1000°C and 1250°C during the run. Systems pressures varied between 4 inches Hg. and 10 inches Hg. below atmospheric.

After solidifying the metal was removed; it weighed 478 grams, indicating a yield of 35 grams of aluminum from a charge of 83 grams of Al_2O_3/Al_4C_3 mix. The surface of the molten pool directly under the arc was undisturbed at the end of the experiment. Three chunks of this undisturbed metal were analyzed for Al_4C_3 content and were found to have analysis of 0.48 wt. %, 0.48 wt. % and 0.28 wt. % of Al_4C_3. The aluminum metal produced was of extraordinary purity compared to metal prepared by prior art single furnace operations.

EXAMPLE 2

The furnace was the same as in Example 1, except that the vacuum was pulled from the access tube **4** instead of the lid. This was done to keep the sight glasses clear during the run.

244 grams of charge of compositions 61.2 wt. % Al_4C_3 and 38.8 wt. % Al_2O_3 (compacted to pellets without starch addition) were reacted in 33 cycles. No arc application exceeded 60 seconds. The minimum delay time was 2 minutes between arc application. The arc intensity was about 12.5 kw. In the first 27 cycles. The cumulative time of arc application was 0.459 hours. The elapsed time of the run was 1.8 hours. The rule was adopted that the maximum time of any arc application would be 60 seconds but the operator would terminate the arc before this time if the charge had completely reacted.

The surface of the molten pool was at temperatures between 823°C and 1180°C for all but the last six arc applications. In some of the last six operations, the power intensity was raised to the range **21–22** kw and, apparently unreacted charges from previous cycles were more completely reacted. The highest temperature of the product pool observed after terminating these higher power applications was 1320°C.

The system pressure during the run ranged from **6** to **11** inches Hg below atmospheric.

The starting aluminum pool weighed 515 grams. The total metal recovered was 617 grams to give a net recovery of metal produced of 102 grams.

Analysis of the aluminum produced showed that it contained only 2 weight percent of aluminum carbide.

EXAMPLE 3

This example illustrates the practice of the invention without the requirement that the charge be engaged with a liquid pool of aluminum. This example is given in reference to FIG. **4** using the plasma arc described in FIG. **2**.

The furnace comprises a gas-tight shell **25**, a rotating electrically conductive hearth of graphite **18**, a connecting post **26** which engages the hearth **18** and conducts current to brushes **27** which go to the positive terminal of the DC power supply **20**. The negative terminal of power supply **20** is connected to the internal electrode **16** of a plasma arc torch. Also connected to electrode **16** is the negative terminal of power supply **19**, the positive terminal of which connects to the casing **15** of the plasma torch. Also rotating with hearth **18** is sub-hearth **28**, composed of an alumina refractory which is of a shape to receive the metal produced by the reaction of the charge under the plasma jet. The hearth rotates at approximately 0.2 revolutions per minute. The casing **15** is offset from the center of the

hearth so that as the hearth rotates, the charge passes under the arc, permitting intermittent application of the arc to the charge.

Briquettes of charge are made from compositions comprising alumina, aluminum carbide, carbon, furnace condensate, and other carbon-aluminum compounds from the process with the sole provision that the composite analysis of the charge has carbon-to-oxygen in the atomic ratio 1:1.

In this example, the charge consists of a furnace condensate having the following analysis:

aluminum	108 pounds
oxygen	32 pounds
carbon	12 pounds

to which is added 204 pounds of alumina and 84 pounds of carbon. The composite final charge has an atomic ratio of carbon to oxygen of 1:1 and analyzed as follows:

aluminum	216 pounds
oxygen	128 pounds
carbon	96 pounds

The final charge is formed into briquettes, or pellets.

These charge pellets **17** are introduced through feed chute **29** to the hearth **18.** Power supplies **19** and **20** are activated and the torch casing is brought to within about 6 inches of the hearth so that the arc is seen to transfer from the torch to the charge pellets. In other words, the arc strikes the charge pellets in a broadening arc pattern as distinguished from the narrowing arc pattern which would be seen if the jet were operating in a simple plasma mode with no arc transfer.

As the hearth rotates and the pellets pass under the charge, they are seen to react to form a bright liquid surface on each pellet struck by the arc. This liquid flows over the curb of the hearth **18** down into the receiving reservoir formed between hearth **18** and hearth **28,** to form a viscous product mass **30.** The carbon monoxide evolved from the reaction is removed from the furnace through tube **31.** The furnace operates at substantially **1** atmosphere pressure. Hearth **18** is controlled to approximately 1,000°C. Approximately 85% of the aluminum content added to the furnace in charge pellets **17** is recovered in the viscous mass **30,** the remainder being lost through vaporization and entrained with the carbon monoxide escaping at vent **31.** This entrained aluminum is captured as furnace condensate by simple cooling and filtration, and this furnace condensate is returned to the charge preparation operation to be recycled with new charge.

An analysis of the viscous mass **30** indicates a composition of aluminum containing 3% aluminum carbide. After filling sub-hearth **28** with products of the reaction of the plasma upon the charge **17,** the plasma jet is moved to strike the melt **30** and the furnace casing is opened at **32** to admit air. Furnace rotation is continued, the mass **30** is returned to a fluid condition by the action of the arc transferred from the plasma torch, and some air is entrained with this arc jet, providing an effective decarbonization action such that after two to three revolutions of the hearth the carbide content has been reduced to the level where the melt in **30** will flow at approximately 900°C. A residue comprising alumina, aluminum carbide, and aluminum is skimmed from the pool **30** and removed from the furnace to be recycled as part of the composition of the charge pellets along with the furnace condensate. Approximately 60% of the weight in the product **30** is recovered as pourable aluminum containing less than 0.2% aluminum carbide. This melt is poured out of the furnace by tilting the furnace in a conventional manner. The torch is then returned to its position for action upon charge pellets **17** which are introduced to continue the operation.

EXAMPLE 4

It is to be understood that it is not necessary to perform the decarbonization reaction within the furnace as set forth in Example 3, supra. As an alternative, the following practice can be used.

While the furnace is in operation as described in Example 3 producing melt **30** from charge pellets **17,** a secondary torch (not shown) is provided to keep the melt **30** in a fluid condition without the admission of air to the furnace. When a product **30** fills the chamber provided for it, it is then tapped at an elevated temperature, i.e., about 1800°C to a container outside the furnace. This product tapped directly from the furnace contains less than about 5 weight percent of aluminum carbide.

If it is desired to reduce the aluminum carbide content to a still lower level, such can be done by passing at least one reactive gas through the tapped furnace product while it is in the molten state so as to react with and separate the aluminum carbide and thereafter recovering aluminum diminished in aluminum carbide content.

From the above examples, it can be seen that the process of this invention is applicable to compounds of aluminum and oxygen other than Al_2O_3. Thus, the expression "an aluminum oxide" is intended to include any compound of oxygen and aluminum, e.g. aluminum tetraoxycarbide. "Aluminum-carbon containing compounds" are intended to include aluminum carbide. The only requirement of the feed is that the atomic ratio of carbon to oxygen is $1:1 \pm 0.05$.

In this connection, it is also noted that although the furnace charge can consist of alumina and carbon, such is not preferred. It is known that when alumina and carbon react, at least one of the intermediate products is an aluminum carbon compound such as aluminum carbide. The optimum conditions for producing aluminum carbon compounds are not necessarily the same as those for producing aluminum. Therefore, if carbon alone is to be used as the reductant, it is preferred that the process be conducted in two separate steps. The first step would involve the reaction of alumina and carbon to form aluminum-carbon containing compounds as is known in the art, and the second step would involve charging the product of said first step together with additional aluminum oxide and carbon such that the feed has an atomic ratio of carbon to oxide of $1:1 \pm 0.5$.

I claim:

1. A carbothermic process for the production of aluminum from an aluminum oxide which comprises:

a. striking an open electrical arc to a portion of the surface of a charge comprising an aluminum oxide and at least one material selected from the group consisting of carbon, aluminum compounds containing carbon, and mixtures thereof;

b. limiting the heating effect of the arc by controlling the time the arc is struck to any given portion of the surface of said charge so that the aluminum formed

by the reaction of the charge is maintained substantially in the liquid state with the proviso that only a small portion of the charge in the reaction zone is heated to reaction temperature while the majority of the charge in the reaction zone is not at reaction temperature at any given time; and

c. causing the liquid aluminum formed under the arc to flow away from the arc and over the non-reacted portion of the charge and be collected, said liquid aluminum containing no more than about five weight percent of aluminum carbide.

2. The process of claim **1** wherein the charge comprises alumina, aluminum carbide, carbon, and other carbon-aluminum compounds and the composite analysis of the charge has a carbon to oxygen atomic ratio of 1:1 ± 0.05.

3. The process of claim **1** in which the open electrical arc is a plasma torch.

4. The process of claim **1** in which the arc is intermittently applied to the charge in a cycle time of 1/120 to 90 seconds.

5. The process of claim **4** wherein the arc is intermittently applied by ending and restarting the arc.

6. The process of claim **1** wherein the energy supplied to the arc provides an average electrical density of from 10 to 50 kw per square inch of charge area struck by the arc.

7. The process of claim **1** wherein temperature of major portion of unreacted charge is below 1,800°C.

8. The process of claim **1** wherein the temperature of furnace parts which contain carbon and which come in contact with the product aluminum are maintained at temperatures below 1,800°C.

9. The process of claim **1** wherein the charge is supported on a rotating hearth in the furnace chamber.

10. The process of claim **1** wherein the system pressure within the furnace chamber is from 0.1 to 10 atmosphere.

11. The process of claim **1** wherein the liquid aluminum removed from the charge is collected in a pool maintained at the temperature of about 670°–1800°C.

12. The process of claim **11** wherein the said collected pool of molten aluminum and the charge and arc extending to the charge are enclosed in a furnace chamber which is substantially closed to the outside atmosphere.

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