

Aug. 30, 1949.

A. F. JOHNSON

2,480,474

METHOD OF PRODUCING ALUMINUM

Filed Dec. 14, 1945

8 Sheets-Sheet 1

Fig. 1.

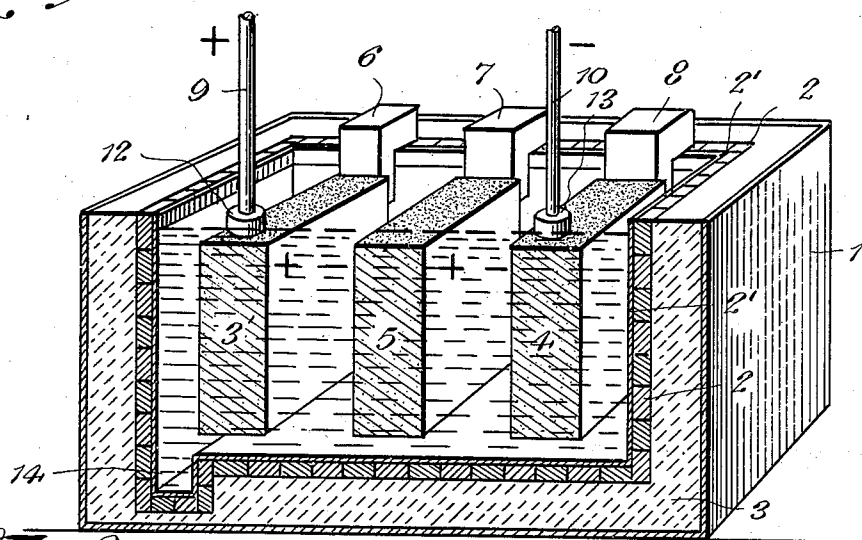
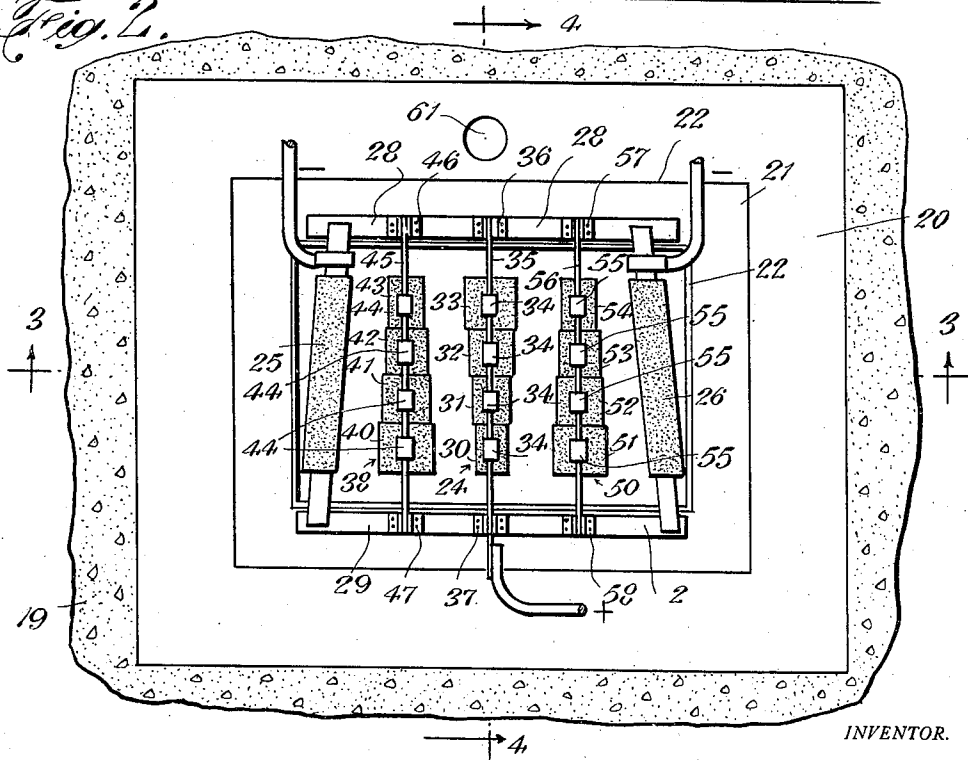


Fig. 2.



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Fig. 3.

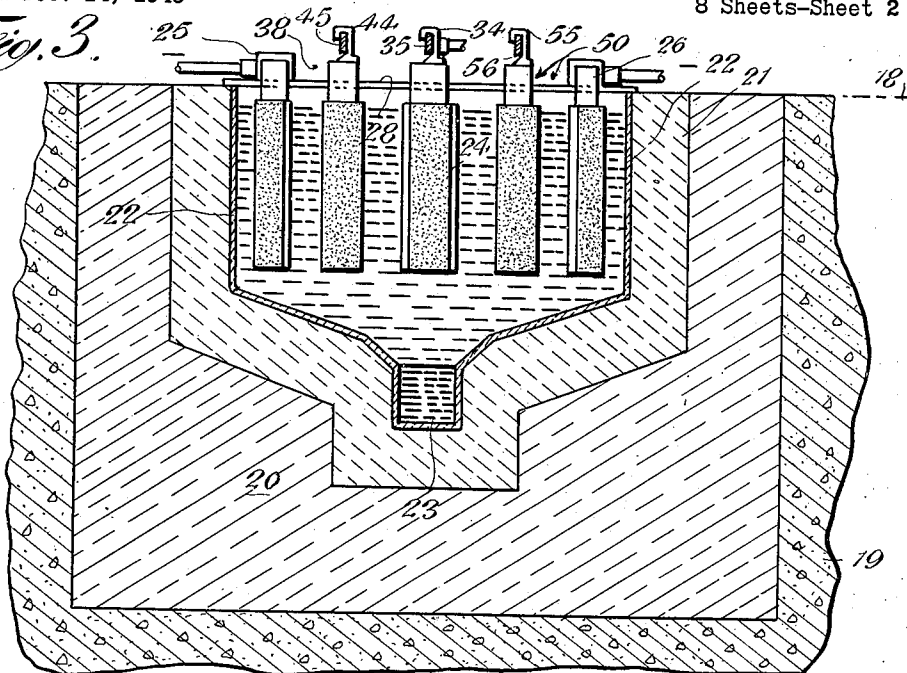
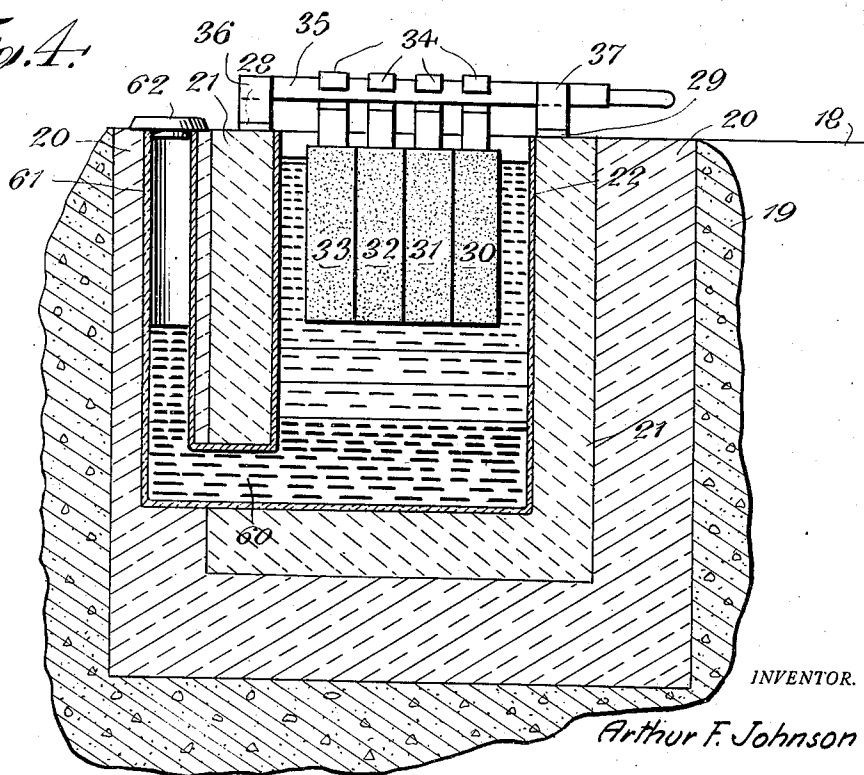


Fig. 4.



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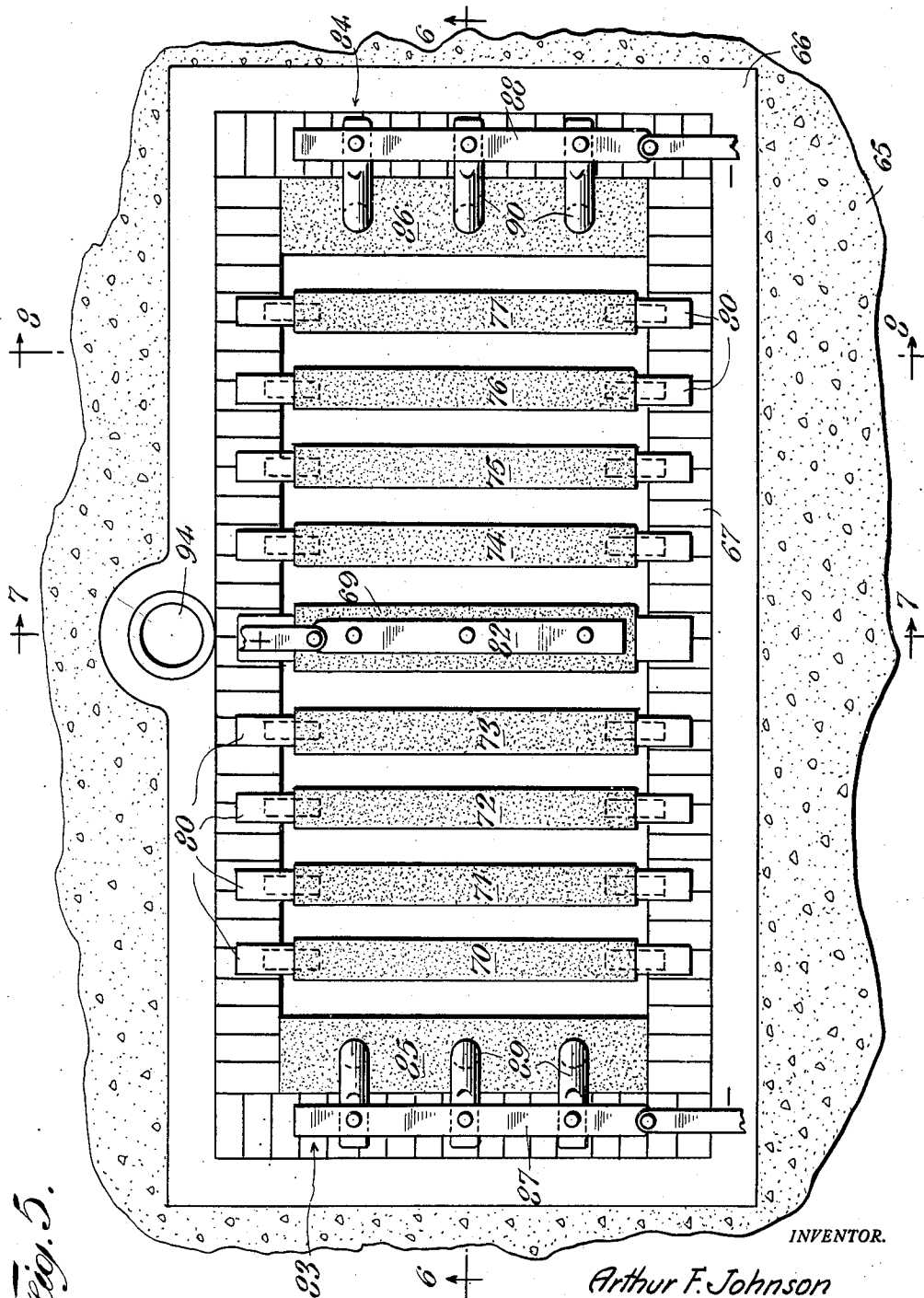


Fig. 5.

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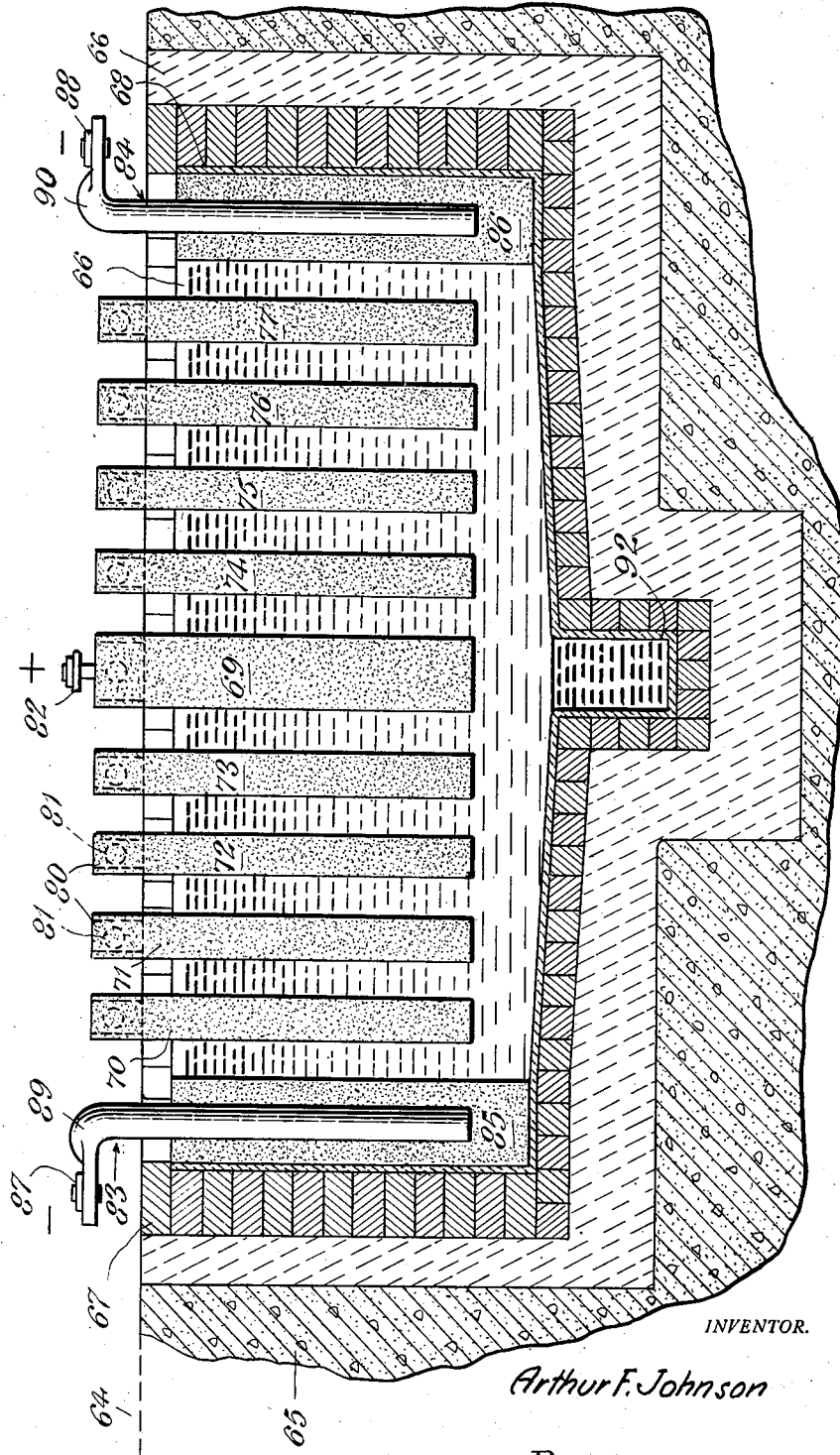
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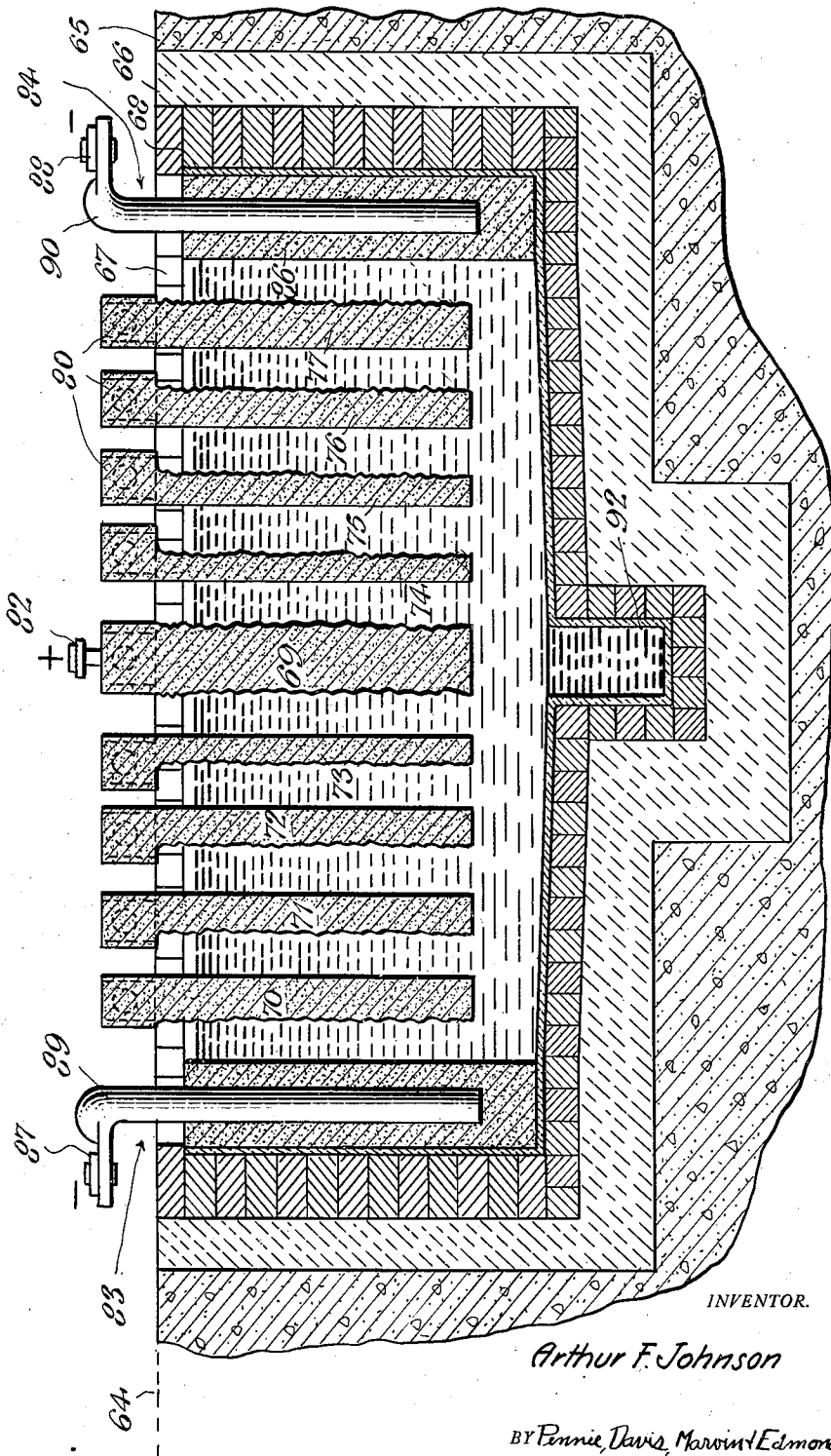
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Fig. 6.a



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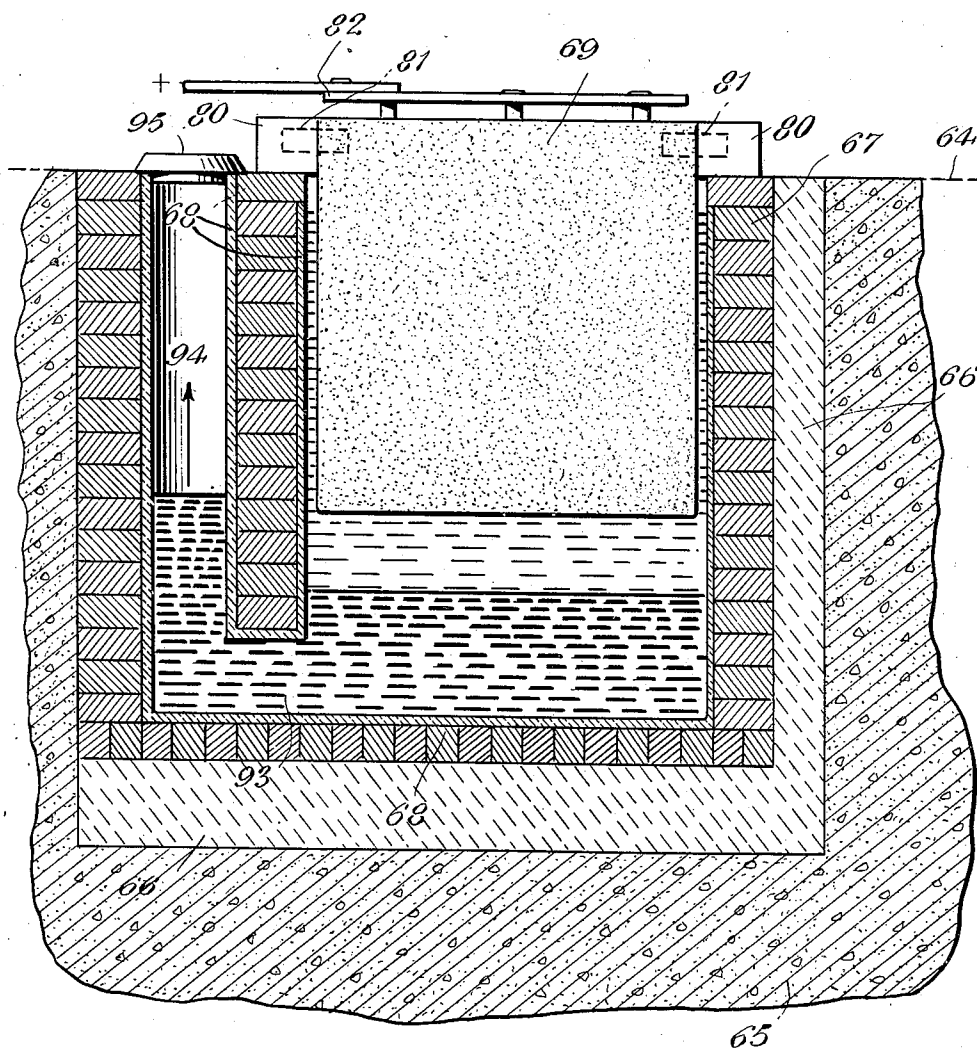
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METHOD OF PRODUCING ALUMINUM

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Fig. 7.



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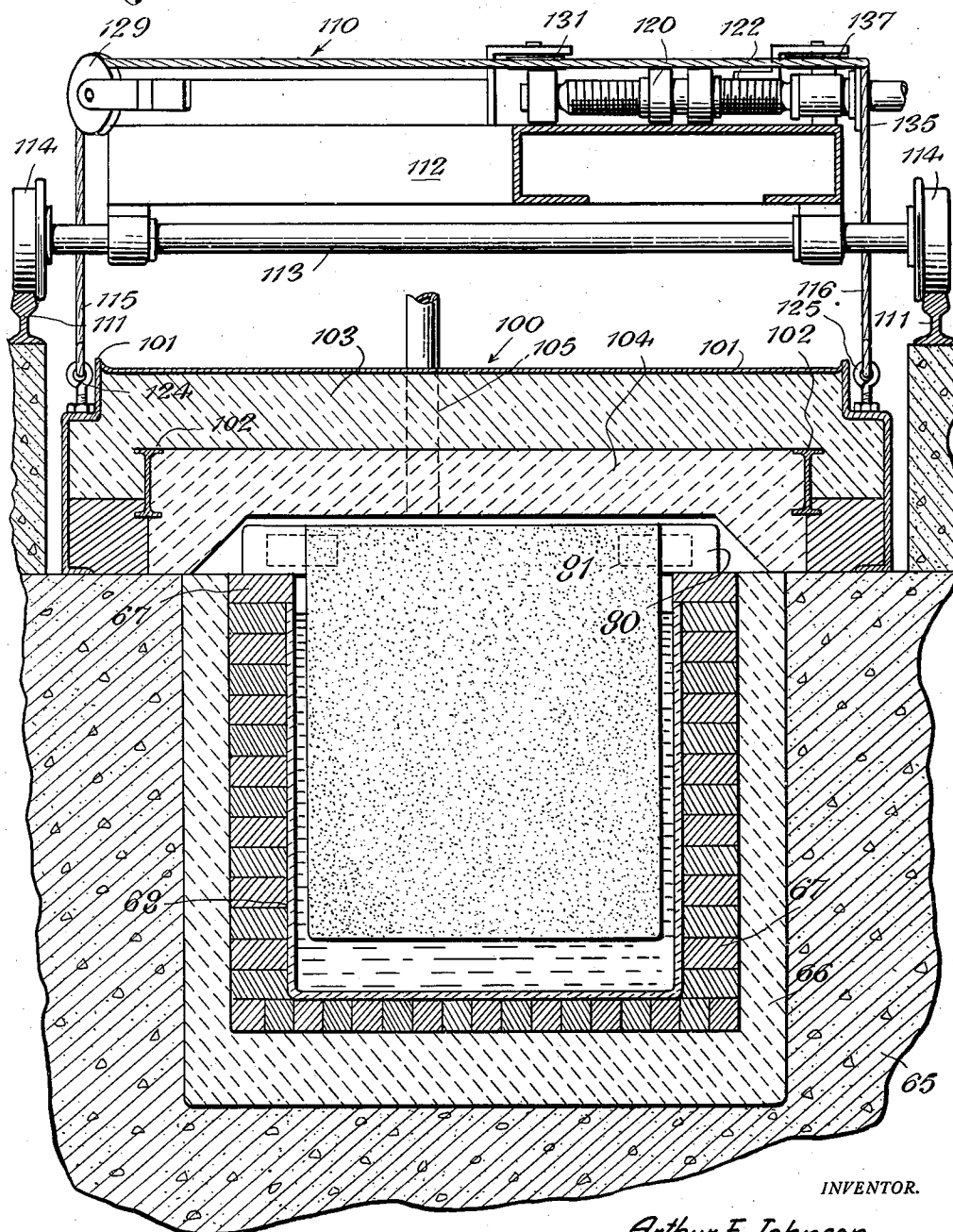
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Fig. 8.



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Fig. 9.

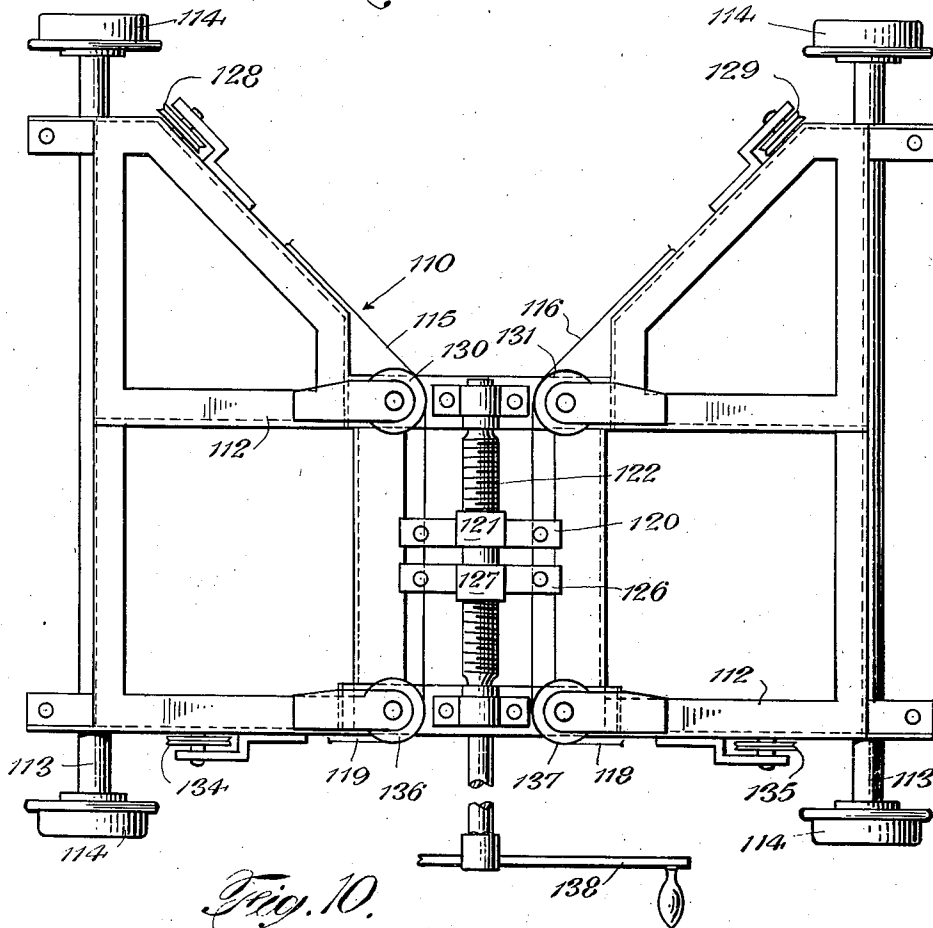
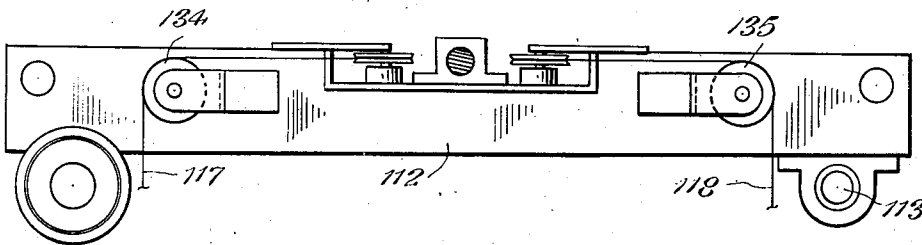


Fig. 10.



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UNITED STATES PATENT OFFICE

2,480,474

METHOD OF PRODUCING ALUMINUM

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Application December 14, 1945, Serial No. 634,903

4 Claims. (Cl. 204—67)

1

This invention relates to the electrolytic production of aluminum from aluminiferous fusions. The invention is concerned with the electrolytic reduction of alumina-containing ore in solution, such as alumina or bauxite dissolved in cryolite or other suitable salts, and provides an improved method for producing aluminum from such aluminiferous fusions. The invention eliminates many of the drawbacks inherent in the universally used method and provides a method possessing many advantages among which are the utilizing of lower voltages with overall higher electrical efficiencies, saving in labor, heat and electrodes, better feeding and metal removal.

The cells now used comprise a refractory lined steel shell the bottom carbon lining of which is the cathode, and anodes of carbon which are suspended in a bauxite fusion. The cells are rectangular shallow vessels, of small capacity, supported above the floor with the electrodes connected in parallel, each cell using about 5 volts. The aluminum collects on the bottom and is periodically tapped by flowing it into a ladle. The heat from the cell walls and exposed molten and crust-covered bath not only is an economic loss but it creates very undesirable working conditions. The cell lining is subjected to changes in temperature due to variations in the kilowatts used per cell with resulting contraction and cracking. These cracks or fissures become filled with molten bath which solidifies in time causing a growth of the lining and a bulging of the shell. Under this great force, the shells bloat out of shape and must be replaced before they split and empty the molten bath on the floor. One of the main difficulties with such cells is the irregularity in space between the conducting bottom and anodes due to corrosion and erosion with a resulting unpredictable current density through the bath.

Heretofore, it has not been possible to increase the productive capacity of an aluminum cell without increasing the current load. In carrying out the method of my invention, I utilize a cell of increased productive capacity constructed to carry a higher voltage rather than a higher current. Since direct current electrical installations in aluminum plants are feasible only at voltages of 500 volts or more, the present aluminum plants must have about 100 cells in series, each requiring about 5 volts to operate. According to my invention, I may provide a few large cells, each using, say, 15 or more volts.

In a simple form, the apparatus used in practicing my invention comprises an electrolytic aluminum reduction cell with an anode, a cathode and

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one or more intermediate electrodes in the path of current flow from the anode to the cathode. I may use any suitable number of such intermediate electrodes between an anode and a cathode, say, 2, 3, 4, 10 or more. The cell is provided with a non-conducting lining in which the flow of current is efficiently confined to a path through the bath and electrodes and in which the electrodes may be submerged to minimize heat loss and corrosion. In practicing the improved method several reduction stages are obtained in a single cell.

One form of apparatus which may be used in carrying out the improved method of producing aluminum comprises a single cell with one common electrode, either an anode or a cathode, which supplies current to, or receives current from, two groups of intermediate electrodes, each having an independent anode or cathode as required by the common electrode used. I may take advantageous use of the higher temperature of the anode (caused by carbon oxidation) to effect a control over the bath temperature, and to this end, I place the anodes at the ends of the cell with a common cathode in the center. I may also use an anode as the common electrode and place two independent cathodes at the ends of the cell. In these forms of apparatus, the cell comprises two groups of electrodes in parallel, each having one or more intermediate electrodes in series.

As the cell does not require a conducting bottom, it may, with several advantages, be constructed in the earth of formed materials, for example a monolithic concrete structure with a refractory, and preferably also insulating lining, with the top at about floor level and in a permanent setting. While I may line the cell with any suitable material which is chemically inert to the bath irrespective of its conductivity, I have found it is more efficient to use a lining which is substantially non-conducting at the voltages I use for electrolysis. One of the important features of the cell which is used in the new method is that it may be arranged with the electrodes in upright or inclined positions with variable spaces between the active faces with the result that the liberated aluminum can trickle to the bottom where it is out of electrical contact with the electrodes and the gas can easily escape upward. While I prefer to lead the metal into a well from which it may be pumped out of the cell, I may also adjust the density of the bath to effect a floating of the aluminum.

The electrodes, and especially the cathodes, are formed of graphite, but the anodes, for pur-

poses of economy, are formed of carbon, such as petroleum coke, and are suspended from above or otherwise supported in their upright positions, preferably entirely within the bath where they are out of contact with oxidizing gases.

The intermediate electrodes which are used in practicing my invention are, in effect, each a combination anode and cathode by reason of the fact that the current flows in one side from the anode and out the opposite side to the cathode giving it a cathode face and an anode face. In a number of large scale experiments, I have confirmed my discovery that a single electrode suspended between an anode and a cathode, can function as an anode and a cathode and that the anode face decomposes in combination with the oxygen of the alumina. The intermediate electrode may be formed of carbon with a graphite cathode face. However, from a technical point of view, it may be formed entirely of graphite.

One of the important improvements of my invention is the use of relatively low voltages between adjacent anode and cathode faces. I may use only a sufficient voltage to provide for decomposition of the alumina and the small heat required to maintain fusion. Suitable voltages may be in the order of from 2 to 4 volts. By reason of the deep bath, large electrode surfaces and close spacing between the active electrode faces, I may use a low IR drop and thus prevent the generation of useless heat. In view of the several series voltage drops which I may use in a single cell, I may use a very large cell operating with an overall voltage of from 10 to 15 volts or more and with parts in parallel.

In one of its more complete and advantageous embodiments, the new method utilizes an electrolytic reduction cell for aluminiferous fusions comprising a vessel lined with material non-conducting at the voltages used, with electrodes in series, at least one of which is an intermediate electrode in circuit through the bath and with means for removing the metal from contact with the electrodes. Such cell also includes means for adjusting the spacing of the electrodes to compensate for the thinning of the electrodes due to the decomposition of the carbon to maintain the desired IR drop in the bath from anode face to cathode face and thus save a useless loss of heat. In contradistinction to the use of present cells with their undue loss of heat through the exposed shell, the anode, the open top and even in the metal removal, I control the generation of heat, and accordingly, provide only enough heat to keep the bath at an efficient operating temperature. I can effectively eliminate the loss of heat through the sides and bottom by eliminating the flow of electricity therethrough and by the use of thermal insulation, through the electrodes by conductivity because I may submerge them in the bath, and by radiation from the top because I may cover the top. Since I may cover the top, I can prevent the loss of heat by radiation from the bath top, prevent the formation of a frozen crust on the bath, giving freedom for electrode shifting and control the escape of gas. The breaking of the crust on present cells to add fresh alumina or to shift electrodes requires a large part of the labor in the operation. Not only does this thermal control result in a saving of electricity but it provides a clean plant free of the unbearable heat now suffered with the objectionable, even harmful, gases under control. In operating according to my invention, I may

use exceptionally large reduction cells of much greater horizontal area and depth than are now possible. This large construction increases the thermal efficiency and permits the efficient use of mechanical feeders and improved metal removal.

The above and other novel features of my improved method of producing aluminum will be better understood by reference to the accompanying drawings which disclose different forms of electrolytic cells which may be used in carrying out the invention.

In those drawings:

Fig. 1 is a sectional elevational view of one form of an electrolytic cell which may be used;

Fig. 2 is a plan view of another form of electrolytic cell which may be used;

Figs. 3 and 4 are views along lines 3—3 and 4—4 of Fig. 2, respectively;

Fig. 5 is a plan view with parts removed of another form of electrolytic cell which may be used in practicing the invention;

Figs. 6, 6a and 7 are views along lines 6—6 and 7—7 of Fig. 5, respectively;

Fig. 8 is a view along line 8—8 of Fig. 5 showing the cell cover in position and partly in section;

Fig. 9 is a plan view of the cell cover, and

Fig. 10 is a sectional view taken at right angles to Fig. 8 showing details of the cell cover.

The electrolytic reduction cells illustrated in the drawings are for the reduction of aluminum oxide to aluminum. Alumina or other aluminiferous material is dissolved in fluoride fusion consisting of cryolite to which has been added various amounts of calcium fluoride and soda ash or aluminum fluoride to lower the fusion point. In the preferred embodiment of the invention, the salts are added to provide a bath having a specific gravity lower than the specific gravity of aluminum, so that aluminum produced in the cell will flow to the bottom thereof. Hereinafter, the charge in the cells subjected to electrolytic reduction will, for convenience, be referred to simply as the "fusion" or the "bath."

The simple form of cell illustrated somewhat diagrammatically in Fig. 1 comprises a steel shell 1 with a refractory lining formed of an electrically non-conducting material, such as fused aluminum oxide or fused magnesium oxide bricks 2, the interior surface of which is preferably covered with a layer of aluminum nitride containing material 2' which prevents solution of the lining in the bath. Between the steel shell and the refractory bricks, I prefer to use a thermal insulation such as powdered alumina 3. The anode 3' and cathode 4 are suspended in the bath and are preferably completely submerged. The intermediate bipolar electrode 5 is also suspended and submerged in the bath between the anode and the cathode and in the path of current flow therebetween. As a matter of convenience, both for supporting the electrodes and submerging them, the iron weighting and supporting lugs 6, 7 and 8 are attached to the electrodes by any suitable means as bolts or pins (not shown). These lugs simply rest upon the electrically non-conducting edge of the refractory and support the electrodes in relatively adjustable positions with respect to each other.

The three electrodes may be formed entirely of graphite. They may also be formed of any suitable form of carbon and it is advantageous that the anode and the intermediate electrode be formed at least in part of carbon for economic

reasons. The intermediate electrode has an anodic face and a cathodic face and may be so formed that the cathodic face is of graphite and the anodic face of carbon. In this connection most of the mass of the intermediate electrode may be formed of carbon with a very thin veneer of graphite for the cathodic face. The anode and cathode are connected to suitable electrical conductors 9 and 10 in the form of copper or other metal bars with refractory or cast iron sleeves 12 and 13 for protecting the conductors where they contact the bath.

It will be noted that the electrodes are arranged in vertical positions. While vertical positions may be used, the electrodes may be in any effective inclined position providing for the unobstructed upward movement of liberated gas and the downward flow of reduced aluminum. It is important so to arrange the position of the electrodes that the released gas follows the anode face and does not flow over to the cathode area where it can oxidize the reduced aluminum. It is important that the aluminum be removed from contact with the electrodes, and to this end, the aluminum is permitted to flow to the bottom of the receptacle and to accumulate at a place out of electrical contact with the electrodes. In the simple form of shell shown in Fig. 1, the bottom slopes towards the left and the metal is free to flow in that direction and enter the trough or well 14 from which it may be removed from time to time.

A characteristic feature of the operation of a cell in accordance with my invention is the use of such relatively low voltages as from 2 to 4 volts between the immediately adjacent electrodes. In the case of Fig. 1, the voltage between the anode 3' and the intermediate electrode 5 would be from about 2 to 4 volts and between the intermediate electrode and the cathode 4 also from about 2 to 4 volts. By placing the electrodes quite close together, for example, around 2 inches, I may use such low voltages, and I prefer to use such current densities that the IR drop through the electrolyte boundaries around the electrodes is large with respect to, or larger than, the decomposition voltage. I may use current densities through the bath between the electrodes of from 1 to 10 amperes per square inch depending on the relative resistivity of the bath and electrodes employed. With petroleum coke electrodes, the allowable current density varies from 1 to 10 amperes per sq. in. The cell is so constructed and controlled as to direct the current through the bath from the anode to the cathode without flowing into the refractory. Factors which influence this flow of current are the thickness of the electrodes with respect to the distance of travel through the bath, the current density, the voltage, and the non-conductivity of the pot lining at the voltages and temperatures used.

During the operation of the cell, the bath is usually at a temperature of around 920° C. to 1000° C. and the carbon of the anode and the anodic face of the intermediate electrode is consumed in combination with the oxygen of the alumina. By reason of the upright position of the electrodes, the gas rises and escapes through the upper surface of the bath. Reduced aluminum is deposited on the cathodic face of the intermediate electrode and on the cathode. This metal coalesces in small drops and is free to roll down the cathode face to the bottom of the receptacle and flow into the well 14. As the opera-

tion continues, the anode and intermediate electrode become progressively thinner. In order to maintain the desired uniform electrical characteristics, it is accordingly necessary to make periodic adjustments in the positions of these electrodes. This may be done simply by pushing the anode and intermediate electrode in the direction of the cathode and ultimately, when they become too thin, removing them for replacement with fresh electrodes.

The simple cell of Fig. 1 is a somewhat diagrammatic for the purpose of illustrating certain fundamental principles and features of apparatus which may be used in the method of the invention. One important characteristic of the invention is that the current is passed from electrode to electrode without flowing through the cell walls and the collected aluminum is not in electrical contact with the electrodes.

The cell of Figs. 2, 3 and 4 is formed in the earth with the upper surfaces of the confining vessel approximately even with the floor level 18. A monolithic body of concrete 19 is poured in between a rectangular form and the surrounding earth to form the structural support for the sides and bottom. Within this concrete mass a lining of such refractory material as powdered alumina 20 for heat insulation is formed. The pot lining 21 for the bath receptacle is formed of non-electrical conducting material, preferably fused aluminum oxide with a protective lining of aluminum nitride 22. As best shown in Fig. 3, the bottom of the receptacle slopes towards the center wherein the metal collecting well 23 is provided.

In this form of the apparatus which may be used, the anode 24 is arranged in the center with cathode 25 at one end and cathode 26 at the other end. The cathodes 25 and 26 are preferably of graphite supported upon the insulating members 28 and 29 resting upon the top of the lining. The anode comprises several anode sections 30, 31, 32 and 33 formed of graphite or carbon, each of which is suspended by means of a hook 34 bolted thereto on the steel bar 35 which is supported in brackets 36 and 37 resting on the insulation 28 and 29. Spaced between the anode and cathode 25 is the intermediate bipolar electrode 38 comprising separate sections 40, 41, 42 and 43 of graphite or carbon, each of which is suspended by means of a hook 44 on the steel bar 45 which rests in the brackets 46 and 47 on the insulation 28 and 29. Between the anode and the cathode 26, the intermediate bipolar electrode 50 is located comprising the separate sections 51, 52, 53 and 54 of graphite or carbon suspended by the hooks 55 from the steel rod 56 attached to the brackets 57 and 58 resting upon the insulation 28 and 29.

As best shown in Fig. 2, it will be seen that the cathodes 25 and 26 are at angles converging on the plane of the anode and that the anode sections 30 to 33 are of progressively decreasing width. The electrode sections 40 to 43 of the intermediate electrode 38 and the electrode sections 51 to 54 of the intermediate electrode 50 are of progressively decreasing width. As the intermediate electrodes are consumed in combination with oxygen and become thinner, they are moved along the bars 45 and 56, as shown in Fig. 2, towards the upper edge of the figure. The sections in the positions of sections 43 and 54 are removed when they become too thin and the other sections are pushed upward, as viewed in Fig. 2, to make room for fresh sections placed

in the positions of sections 40 and 51. Similarly, the sections of the anode are shifted towards the bottom, as viewed in Fig. 2, the section 30 being removed when consumed, and after the other sections are shifted to the front, a fresh section is placed in the position of section 33. In this arrangement for the manipulation of the electrodes, a substantially uniform distance is maintained between the active faces of the electrodes and the uniformity of the operation is facilitated.

The electrodes of Figs. 2 to 4 may be in the neighborhood of 20 inches in depth and in the aggregate approximately 24 inches in horizontal length. The fresh sections of intermediate electrodes and anode may be approximately 6 inches square in cross section. When these sections are consumed, they may be a little more than 1 inch thick. They are removed before they become so thin as to break off and fall into the bath.

As best shown in Figs. 3 and 4, the upright positions of the electrodes permit the gas to escape freely and the metal to fall to the bottom of the receptacle out of electrical contact with the electrodes. The aluminum which accumulates in the well 23 flows into the lateral extension 60 from which it may be removed by means of a pump inserted in the upright duct 61 after removal of the cover 62. By reason of the weight of the bath resting upon the aluminum in the well 23, the aluminum is forced upward a considerable distance in the removal duct 61, as shown in Fig. 4, and is therefore accessible for removal.

The arrangement of electrodes shown in the apparatus of Figs. 2 to 4 comprises a parallel circuit with the current entering a common anode and splitting to flow into the two independent cathodes 25 and 26. While I have shown but one intermediate electrode in each series circuit between the anode and the cathode, I may use two or more intermediate anodes in the series circuit.

The reduction cell illustrated in Figs. 5 to 10 is of a large type formed mostly below the floor level 64. The receptacle for the bath comprises a monolithic supporting structure of concrete 65 cast between a form and the surrounding earth, a thermal insulating refractory 66 consisting of powdered alumina or other suitable heat resistant material and a layer of dense fused alumina bricks 67 of high electrical resistivity. In order to protect the bricks from solution in the bath, I have found it advantageous to line the bricks with a layer of aluminum nitride containing material 68. The upper inner edge portion of the receptacle is covered with a row of the electrically non-conducting bricks 67. The anode 69, the intermediate electrodes 70, 71, 72 and 73 and the intermediate electrodes 74, 75, 76 and 77 are supported upon these bricks 67. In order that the anode and the intermediate electrodes will be held in the bath and prevented from floating, they are each attached at their upper ends to iron weights 80 by means of alloy steel bars 81. The weights 80 not only hold the electrodes down in the bath preventing them from floating but they provide a simple support and a convenient means for shifting the intermediate electrodes along the bricks 67 in the direction of the anode 69. In this form of cell the anode is preferably formed of carbon because of its lower rate of decomposition. The power conductor 82 is bolted to the anode.

The cathodes 83 and 84 are preferably formed of slabs of graphite 85 and 86 fitted snugly into the end walls of the receptacle. Electrical contact with the conducting bars 87 and 88 is effected

by means of the rods 89 and 90 embedded in the graphite.

This form of cell also comprises a parallel electrical circuit in which the current enters the cell through the anode, divides and flows in one series circuit through the intermediate electrodes 70 to 73 and out through the cathode 83, and flows in another series circuit through the intermediate electrodes 74 to 77 and out through the cathode 84. In the operation of this cell the anode and the intermediate electrodes are consumed in combination with oxygen and become progressively thinner. As shown in the drawings, the intermediate electrodes have both anode and cathode faces and in a satisfactorily controlled operation the current density and spacing is so adjusted as to give approximately from 2 to 4 volts between the active faces with the result that each series circuit has an overall voltage of approximately 10 to 20 volts. The intermediate electrodes are preferably formed of carbon with a graphitic cathode face so that the bulk of the carbon consumed is carbon comprising the anode face. Fig. 6 shows a cell with fresh electrodes and spaced as at the beginning of an operation. Fig. 6a shows the condition of the electrodes during operation. As the intermediate electrodes and anode become progressively thinner, the intermediate electrodes and cathode are shifted towards the anode while maintaining the proper space therebetween. As they reach the position of electrodes 73 and 74, they are removed and fresh intermediate electrodes, varying in thickness from 4 to 6 inches, are placed in the positions of electrodes 70 and 71.

The bottom of the receptacle slopes towards the center so that the metallic aluminum may flow into the collecting trough 92 and enter the lateral extension 93 and rises in the metal removal duct 94 which is protected from the atmosphere by the removable cover 95. By means of any suitable pump or syphon, the metal may be removed from time to time.

My invention contemplates the utilization of a cell similar to the cell of Figs. 6 to 10 in which a common cathode is used in the central position of anode 82 with independent anodes in the positions of the cathodes 83 and 84. In this form of cell the series circuit flows in the opposite direction, and since the anode is consumed, it is considerably hotter than the cathode by reason of the oxidation of the carbon and thus provides suitable bath temperatures at the ends of the cell.

As best shown in Figs. 8 to 10, the cell of Fig. 5 may be provided with a removable cover to prevent the radiation of heat from the cell and to prevent a frozen crust from forming on the top of the bath, thus assuring the free movement of the intermediate electrodes, and a control over the escape of liberated gases. The cover 100 comprises a steel exterior supporting frame 101 with interior reinforcing I-beams 102. The upper part of the cover is filled with rammed alumina 103 and the inner part of the cover that is exposed to the open cell is lined with a thick layer of carbon paste 104. Stabilizing bars may be embedded in the refractory at suitable places to help anchor the refractories to the steel frame. The cover is arranged to lay flush over the top of the cell and there is sufficient clearance under the arched refractory lining for the electrodes and their electrical connections. Gas accumulating under the cover may escape through duct 105 and be led away through a suitable pipe.

The cover can be moved from its position over the cell by means of the hoist 110 which travels

in a lengthwise direction of the cell on the rails 111. The hoist comprises a structural steel frame 112 with axles 113 at the ends on which are mounted rail wheels 114 which travel on the rails 111. The cables 115 and 116 are each connected at one of their ends to a bar 120 attached to a threaded nut 121 which travels on the threaded shaft 122 and at their other ends to eye-bolts 124 and 125 which connect to the frame 101 of the cover (Fig. 8.) The cables 117 and 118 are each attached at one end to the bar 126 fixed to the threaded nut 127 which travels on the threaded shaft 122 and at the other ends to eye-bolts (not shown) but similar to 124 and 125 attached to the cover as just described. The cables 115 and 116 pass upwardly over the pulleys 128 and 129 and then horizontally over the pulleys 130 and 131 to the bar 121. The cables 117 and 118 pass upwardly from the cover over the pulleys 134 and 135 and 136 and 137 to the bar 126. The shaft 122 may be rotated by hand by means of the crank 138. As shown in Fig. 9, the threads on shaft 122 are both right and left, so that by turning the crank in one direction, the threaded nuts 121 and 127 move toward each other, while turning it in the opposite direction causes them to move away from each other. In the position shown in Fig. 9, the cables have been pulled in towards the center and thus hold the cover in an upward suspended position. By pushing the hoist along the rails 111, the cover may be removed from its position over the cell. By turning the crank 138 to spread the nuts 121 and 127 apart, the cover may be lowered to the position over the cells shown in Fig. 8. The track 111 may be of sufficient length to completely remove the cover from its position over the cell, or only sufficiently long to expose, say, one-half of the cell at a time.

In the appended claims, "alumina" is used in a generic sense and includes its hydrous forms, such as bauxite, as well as the anhydrous form.

I claim:

1. In a method of producing aluminum by the electrolysis, of a fused bath of alumina dissolved in cryolite having a specific gravity lower than the specific gravity of aluminum and at a temperature higher than the melting point of aluminum, in an electrolytic cell, the improvement which comprises maintaining the anode and the cathode in generally vertical position, maintaining at least one generally vertically-positioned bipolar electrode between the anode and the cathode, passing the electric current through the fusion from the anode to said bipolar electrode

and to the cathode in series, whereby electrolytic decomposition of the alumina is effected and aluminum in a molten state is liberated at the cathode and at the cathodic face of said bipolar electrode and flows downwardly to the bottom of the cell, and relatively moving said electrodes towards one another as the electrolytic decomposition proceeds to compensate for consumption of said anode and the anodic face of said bipolar electrode.

2. The improvement in the method of producing aluminum by electrolysis as set forth in claim 1 in which the flow of electric current between the several electrodes is maintained without flowing through the fusion to the cell receptacle.

3. The improvement in the method of producing aluminum by the electrolysis as set forth in claim 1 in which the cathode and the bipolar electrode are spaced from the bottom of the cell so that liberated molten aluminum will flow from the cathode and from the cathodic face of said bipolar electrode to the bottom of the cell, and removing accumulated liberated aluminum from the bottom of the cell to keep it out of contact with said cathode and said bipolar electrode.

4. The improvement in the method of producing aluminum by the electrolysis as set forth in claim 1 in which the electrolytic cell has a refractory lining, the voltage from electrode to electrode is from about 2 to 4 volts in the series circuit, and in which the flow of current from the electrodes through the fusion to the refractory lining is prevented.

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