Sept. 20, 1960 H. SCHMITT 2,953,506

METHOD AND APPARATUS FOR REFINING ALUMINUM

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FIG. 2

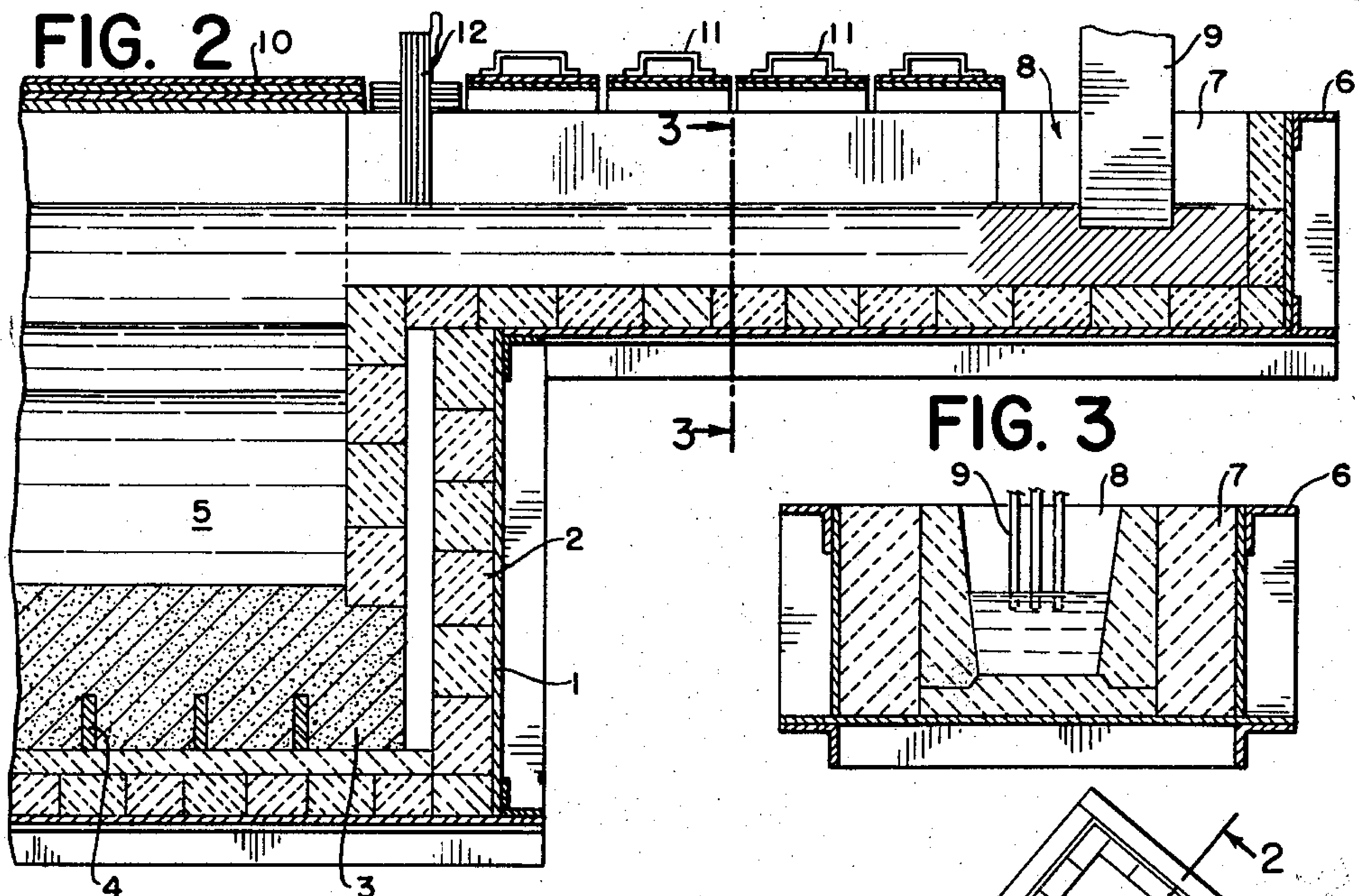

FIG. 3

FIG. 1

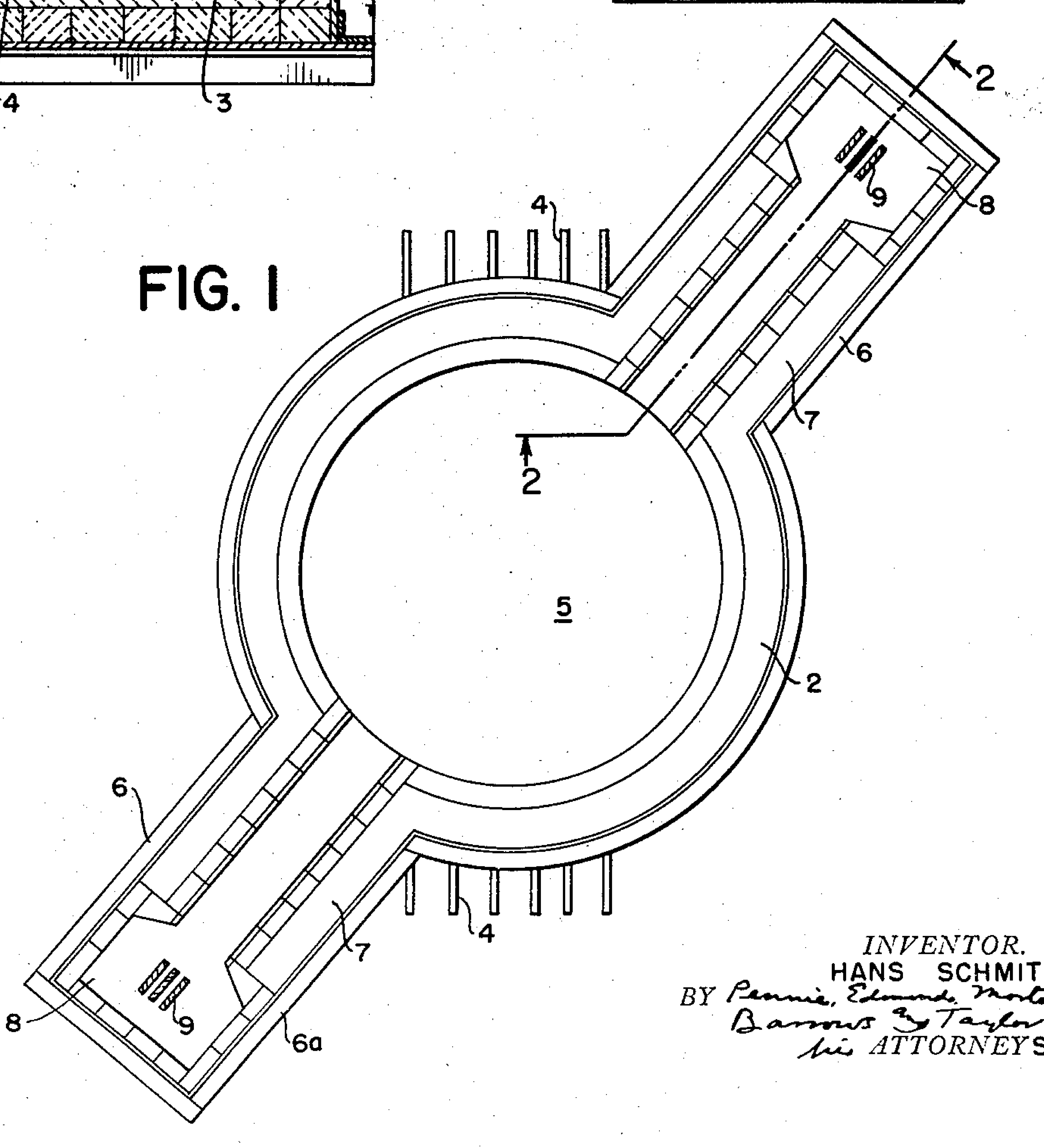

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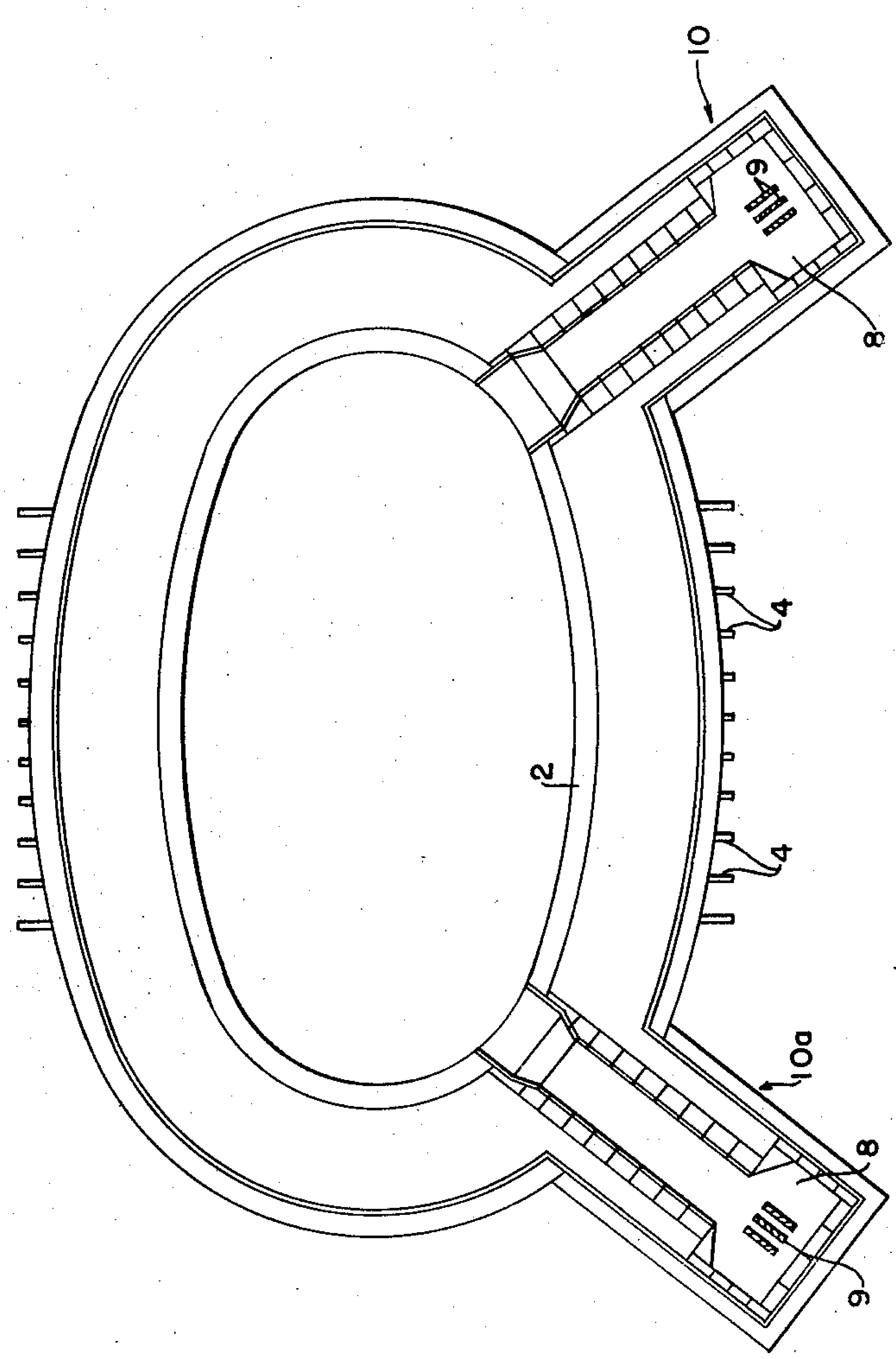
FIG. 4



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METHOD AND APPARATUS FOR REFINING ALUMINUM

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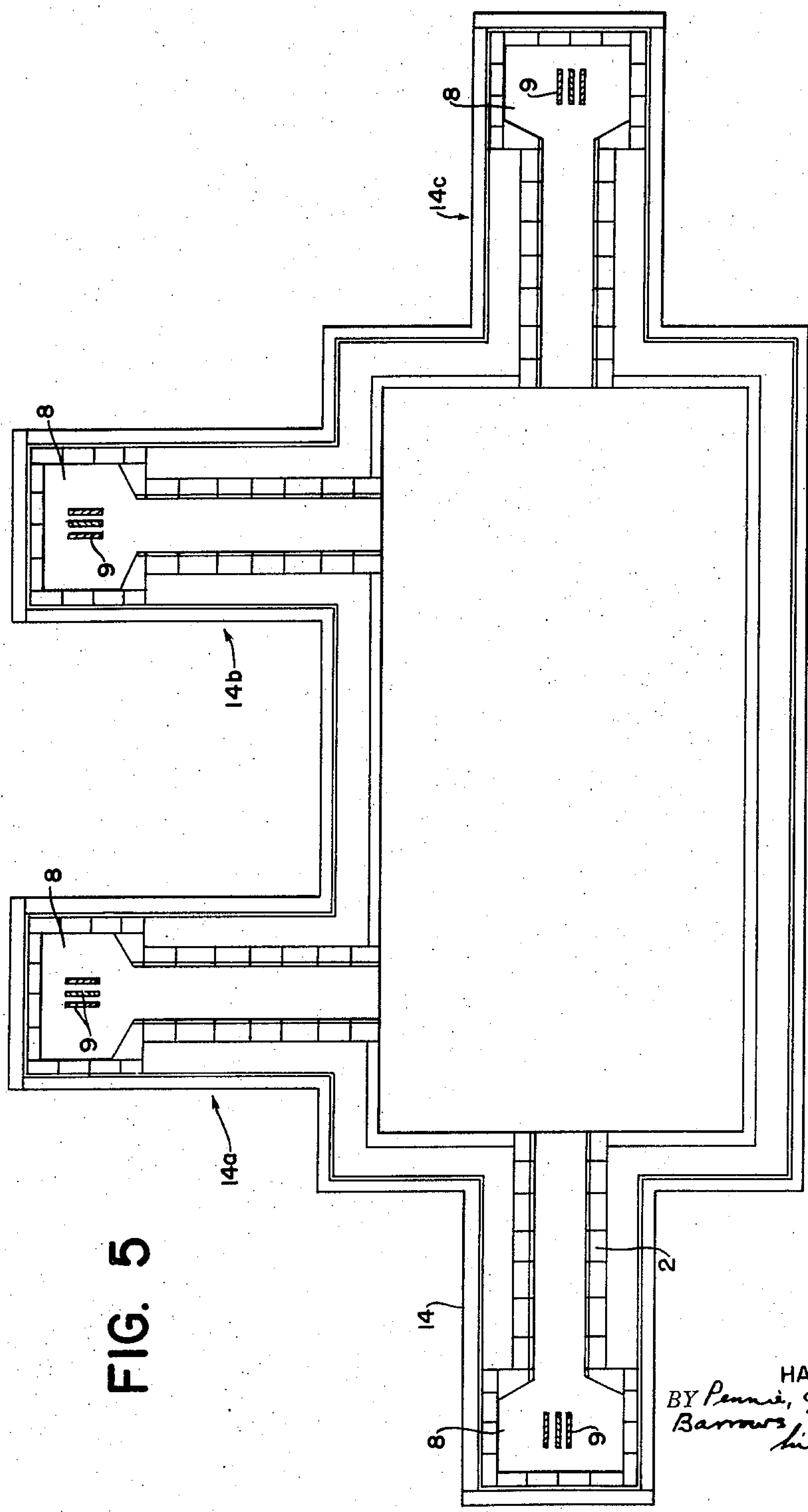

FIG. 5

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METHOD AND APPARATUS FOR REFINING ALUMINUM

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It is known, that it is possible to refine virgin or scrap aluminum in the three-layer electrolysis to a purity of at least 99.99%. The contaminations present in the purest aluminum are for example the following:

	Percent
Fe	0.0005 to 0.002
Si	0.002 to 0.005
Cu	0.0005 to 0.002
Zn	0.0005 to 0.002
Ti	0.0001 to 0.003
Mg and other elements	Traces

The refining cells used at present in the aluminum industry are working with current intensities between 8,000 and 20,000 amperes, in a few cases with higher current intensities of, for example, 40,000 amperes. A refining cell consists usually of a pot built up from magnesite bricks, the bottom being lined with carbon serving for the anode current supply. The pot contains as anode a molten aluminum alloy with a copper content of about 25 to 35%. On this molten alloy swims the electrolyte which is composed either of pure fluoride salts or of a mixture of fluoride or chloride salts of the alkaline and the alkaline earth metals. The refined aluminum, separated in the molten state through the effect of the current, swims on the electrolyte and serves as cathode, the cathodic current supply being provided through graphite electrodes plunged into the cathode metal.

Metal of equal or even better purity may be obtained too, in refining cells with a cathodic current supply in the form of a molten metal connection, of which a favourable design is described in the pending application of Hans Schmitt Serial No. 630,830 now Patent No. 2,866,743. The refining cell described in that application is provided with a radially disposed channel at the height of the upper cathodic layer, filled with molten cathode metal. The solid current conductors are engaged in the end of the channel opposite to the cell, before filling the channel with cathode metal. After the filling, the metal solidifies around the solid current conductors, at the end of the channel opposite to the cell, and remains liquid in the rest of the channel.

The refining cells with a molten metal connection for the cathode current have the advantage over the cells working with graphite electrodes, that although the purest graphite is used for the electrodes there are always contaminations getting from the graphite electrodes into the cathode metal.

It is known that the electrolyte must be purified if aluminum of the highest possible aluminum content is to be obtained. The purification is usually performed by mixing and melting the electrolyte components and subjecting the melt to electrolysis in a pre-refining cell from which the purified electrolyte is transferred to a refining cell in which the metal of highest purity is to be produced. It is also possible to purify the electrolyte metal in two or even in three steps by transferring it from one to another pre-refining cell before it is used in the

last refining cell for the production of the purest metal.

Through this purification generally the amount of the contaminations, which is present in the fluoride and chloride salts for example as iron oxide and silica, is reduced substantially. The analysis of a purified fluoride electrolyte showed for example a SiO_2-content of only 0.02% and a Fe_2O_3-content of only 0.03%. In these pre-refining cells one obtains an aluminum of a lower purity which is satisfactory for many purposes or can be further refined.

Furthermore, it is known to operate the refining cell for the production of aluminum of the highest purity that an electrolyte layer having a height of 12 cm. or more is desirable. Due to the thick electrolyte layer a mixture of the anode metal with the cathode metal which may happen from movements and drifts in the anode or in the electrolyte layer is avoided as far as possible.

A thick layer of cathode metal together with the thick layer of electrolyte are also advantageous for producing a purest aluminum.

The layer of cathode metal should be in the order of 15 to 20 cm. but not exceeding 30 cm. Necessary operation of the refining cell, during which the heat insulating covers must be removed, for example for changing the electrodes or for drawing out the yielded aluminum, cause great losses of heat through convection and radiation which may lead to a considerable temperature drop within the cell. Such a reduction of the temperature favours the formation of crusts of the electrolyte, especially if the liquidus point of the electrolyte is reached, which crusts may get into the cathode metal causing a contamination. It may be added that the greater the formation of crusts the more the cell must be skimmed. Each skimming however means a loss of electrolyte which must be replaced by introducing fresh electrolyte with which new contaminations get into the bath.

It is also necessary in order to get the purest possible aluminum in the three-layer electrolysis that the crude metal which is to be refined, added to the anode metal should already have a purity of possibly over 99.0%, and that all instruments used in connection with the refining cell should be made of the purest graphite.

If all these conditions are fulfilled and by operating very carefully it is possible with the refining cell known up to now to get an aluminum with a purity of maximum about 99.9975%. Such a very pure aluminum contains for example still the following contaminations:

	Percent
Fe	0.0005
Si	0.001
Cu	0.0005
Other contaminations as Zn and Fi, together	0.0005

For certain applications, there is now interest in an aluminum the purity of which is still higher than that of aluminum obtained up to now with the three-layer electrolysis.

It was the object of the research work which leads to this invention to develop a refining cell for the three-layer electrolysis and a method of operating this cell, with which it would be possible to get an aluminum with a purity higher than ever produced up to now by three-layer electrolysis, the wanted purest aluminum being of a purity of 99.998% and more, in the average of 99.999%, whereby it is assumed that besides the above mentioned foreign metals also neglectable traces of other elements may be present.

The invention of this application is based on the results of this research work during which the surprising discovery was made that the problem of the reduction of the last contaminations in the refined aluminum to a minimum is related with the distribution of the current flow in the cell.

The refining cells with molten metal connection as heretofore constructed are provided with one channel or if two channels were used they were arranged with their longitudinal axes at an acute angle. This design was used so as to lead the current conductors in a simple form and as closely together as possible to the cell and to keep free the side of the cell outside the acute angle between the channels for other purposes, for example for the arrangement of a forehearth, for carrying out measurements or for drawing out the metal.

My investigations have shown that in cells with the known arrangement of the channels the current does not flow evenly through the bath. In a cell with channels arranged in an acute angle the flow lines of the current are crowded at the side of the channels.

According to the present invention, an even distribution of the current is attained when the refining cell is provided with at least two side channels filled with cathode metal for the cathodic current supply, whereby the horizontal lines joining the center of the cell with the center of the mouth of two of these channels into the vessel of the cell make an angle of at least 90° without consideration of any other channel which could be disposed between these two channels. Preferably the number of side channels will not be greater than four.

At their abutting point with the wall of the cell, the channel can make any angle with this wall, but in practice the most favourable arrangement is that the channels are perpendicular or nearly perpendicular to the wall of the cell.

By round cells provided with radially disposed channels, the longitudinal axis of the channels is always going over the center of the cell. According to the invention, such radially disposed channels must be arranged in such a way that the longitudinal axes make an angle of at least 90°. Further channels may then be arranged at any other point of the perimeter of the cell. Also elliptic or polygonal refining cells may be provided with side channels for cathodic current supply according to the present invention. In the case of rectangular cells provided with two channels, these must be connected to two neighbouring or opposite sides of the cell. If there are three or four channels, these are distributed preferably on three sides of the cell. Such a disposition of the connection channels insures an even flow of the current through the bath.

In the accompanying drawings I have illustrated cells of different shapes with two or more channels for conducting the cathode to the cell with the channels arranged in accordance with the invention.

Referring to the drawings:

Fig. 1 is a plan view of a cell of circular cross section;

Fig 2 is a vertical section on line 2—2 of Fig. 1;

Fig. 3 is a vertical section on line 3—3 of Fig. 2;

Fig. 4 is a plan view of a cell of elliptical cross section with channels arranged in accordance with my invention; and

Fig. 5 is a plan view of a cell of rectangular cross section having four cathode channels arranged in accordance with my invention.

Referring to the drawings, particularly to Figs. 1–3, **1** indicates the outer metal wall of the cell which is lined in the customary manner with bricks **2** of magnesite or other refractory meterial. The bottom of the cell consists of a slab **3** of carbon supported on bus bars **4** through which the current is supplied to the anode of the cell, which in the customary process consists of a layer of molten alloy of aluminum and copper indicated at **5**.

The cell is provided as shown with two oppositely arranged radially extending channels **6**, **6*a*** each of which is shown in Fig. 3 as provided with a refractory lining **7** shaped to provide an enlarged cross sectional area **8** at the outer ends of the troughs into which enlarged area the bus bars **9** for the cathode extend in the manner shown.

In carrying out the electrolytic operation the quantity of electrolyte which consists of fluoride salts of the alkali or alkali earth metals, or mixtures of such salts with chloride salts of said metals and the quantity of copper-aluminum alloy charged to the cells is such that the top surface of the electrolyte will be below the bottom of the channels **6** so that the channels in operation will contain only the molten refined aluminum.

The cell is provided with a heat insulating cover **10** and the channels **6** and **6*a*** have sectional covers **11**. The channels are also provided with gates **12** for reducing the loss of heat from the surface of the metal in the cell.

The structure so far described, except for the provision of the second channel **6*a*** arranged at a point diametrically opposite the channel **6**, is the same as that disclosed in the above mentioned pending application. In that application one channel only is provided for supplying cathode current to the cell through an electrode consisting of the molten refined metal, but in practice, as stated above, such cells have sometimes been provided with a second channel at the same side of the vessel and with its radial axis at an acute angle to the radial axis of the other channel.

At the time the above mentioned pending application was filed it was not known that the number and arrangement of the channels bore any relation to the degree of purity of the refined aluminum which could be obtained by the operation of the cell, and subsequent use of the cells with one channel and the cells with two channels the mouths of which into the cell being disposed one near the other and this channel making an acute angle has demonstrated that there is no difference in the purity of the aluminum obtained therein under like conditions of operation, whereas with the channel arrangement shown in Figs. 1–3 aluminum of an average purity of 99.999% has been obtained.

It is not essential, however, that the two channels be diametrically opposite and in Fig. 4 I have shown a cell which is of elliptical cross section with two conducting channels **13** and **13*a*** at the same side of the cell. In this cell, the horizontal lines joining the middle point of the vessel with the center of the mouth of the two channels make an angle greater than 90°. With this cell of Fig. 4 the same degree of purity can be obtained as with the cell of Figs. 1–3 under the same operating conditions.

In Fig. 5 I have shown a modified cell structure wherein the cell itself is of rectangular cross section with four outwardly extending channels **14**, **14*a***, **14*b*** and **14*c***, respectively, arranged at three sides of the rectangle. The channels **14** and **14*c*** project from opposite ends of the cell with their vertical axes in the same vertical plane. The other two channels **14*a*** and **14*b*** extend outwardly from the same side wall of the channel and have their longitudinal axes parallel and at right angles to the axes of the other two channels. I found with this arrangement also a substantially uniform distribution of the current is obtained with a resulting purity of the refined aluminum equal to that obtained with the other two cells.

My experience has demonstrated that it is essential in order to obtain aluminum of a purity such as herein specified, that the channels for the conduction of cathode current be disposed in such a way that the horizontal lines joining the center of the cell with the center of the mouth of two of these channels into the vessel of the cell make an angle of at least 90°, whereby the further channels can be disposed at any other point of the perimeter of the cell. It is of course advantageous to dispose the further channels in the same angle relation to each of the first ones in order to obtain as far as possible a symmetrical arrangement of the channels around the cell.

During the operation of the cell the current flow must in no case be interrupted or reduced, as otherwise the distribution of the current in the cell will be disturbed. The operations for cleaning and removing the slags in a channel must therefore be carried out without switch-off the current in the channel to be cleaned.

The refining cells with molten metal current connections have the advantage over the cells working with graphite electrodes in that the operations at the cell are reduced to a minimum and that the cells must be opened only for drawing out the refined metal.

It is known by experience that cells of such a kind seldom must be skimmed and that they show a minimal use of electrolyte. Hence if these cells are constructed in accordance with this invention, an even distribution of the current flow can be maintained throughout the entire refining and aluminum with the highest possible aluminum content can be obtained.

For example, in a round refining cell working with a current intensity of 18,000 amperes and with two radially disposed cathodic molten metal connections, the axis of which make an angle of 90° it is possible to obtain in continuous working a purest aluminum with an average purity of 99.999%. It is of course the same with refining cells of a higher current intensity, for example of 60,000 or even up to 100,000 amperes.

Such refined aluminum has for example the following composition.

	Percent
Fe	0.0004
Si	0.0002
Cu, Zn and other impurities	0.0004
Al	99.999

Of course, by operating the new cells according to this invention, it is always indispensable in order to obtain a refined aluminum with maximum purity, to observe the precaution known up to now and set in the introduction of this description, namely to use a purified electrolyte, a high cathode metal layer, a high electrolyte layer and also tools of purest graphite and to add to the anode metal aluminum of at least 99.0%. Furthermore, it is advantageous to use refining cells working at high current intensity of at least 18,000 amperes.

When these conditions are observed, the refining cells according to the invention allow to increase the purity of the refined aluminum up to over 99.9975% to at least 99.998% and even to 99.999% in the average.

I claim:

1. An electrolytic cell for refining aluminum according to the three-layer process, the cell being provided with side channels filled with cathode metal for the cathodic current supply, which refining cell is provided with at least two such channels, whereby the horizontal lines joining the center (or: the vertical axis) of the cell with the center of the mouth (or: entrance) of two of these channels into the vessel of the cell make an angle of at least 90°, without consideration of any other channel which could be disposed between these two channels.

2. An electrolyte cell according to claim 1 in which the number of the channel is at most four.

3. An electrolytic cell according to claim 1, wherein said channels are two in number and arranged so that they debouch into the vessel of the cell at points diametrically opposed with respect to the geometric center of the cell.

4. An electrolyte cell according to claim 1, which cell is round in horizontal section and is provided with two to four radially disposed side channels, two of them making an angle of at least 90°, without consideration of the disposition of the further channels.

5. An electrolytic cell according to claim 1, which has the form of a rectangle and which is provided with at least three channels disposed at these sides of the rectangle.

6. In a method of operating an electrolytic cell for refining aluminum by three-way electrolysis the step which consists in transmitting cathode current to the cathode layer of refined aluminum from a plurality of molten metal connectors disposed horizontally at the perimeter of the vessel, so that the horizontal lines joining the center (or: the vertical axis) of the vessel of the cell with the center of the mouth (or: entrance) of two of these connectors into this vessel make an angle of at least 90°, without consideration of any other connectors which could be disposed between these two connectors.

7. The method of claim 6 wherein the connectors are two in number and debouch in the vessel at points diametrically opposed with respect to the geometric center of the cell.

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