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METHOD OF AND APPARATUS FOR CIRCULATING LIQUID METALS IN FUSED SALT ELECTROLYSIS
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FIG. 1

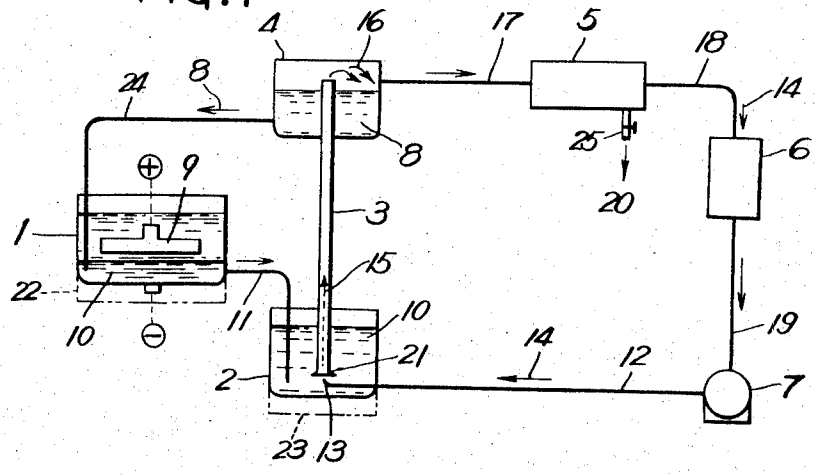
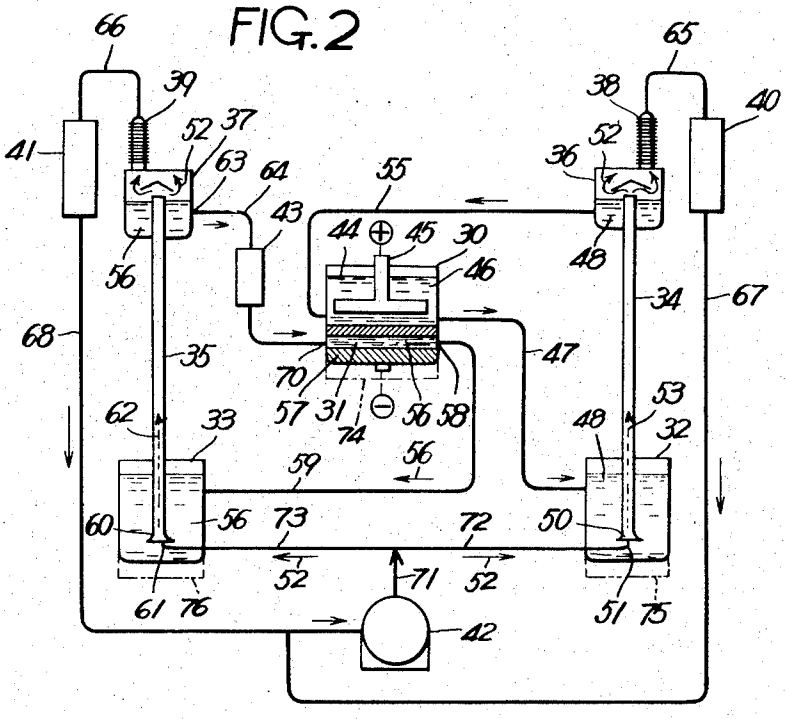


FIG. 2



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3,728,234

METHOD OF AND APPARATUS FOR CIRCULATING LIQUID METALS IN FUSED SALT ELECTROLYSIS

Hidetami Sakai, Funabashi, and Muneyoshi Arita, Sakura, Japan, assignors to Daiki Engineering Co., Ltd., Tokyo, Japan

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U.S. Cl. 204—68 3 Claims

ABSTRACT OF THE DISCLOSURE

A method of and an apparatus for circulating a liquid metal in fused salt electrolysis comprising passing the fused alloy of the cathode liquid metal and the electrolytically liberated metal both coming out of the cell and the liquid metal for controlling the temperature of the cell through a lift pipe by making a mixture of an inert gas and the liquid metals by the jetting force of the gas, said mixture being small in specific gravity, lifting said liquid metals into a head tank positioned at a level high enough for recirculating the liquid metals into the electrolytic cell by gravity, separating the liquid metals from the inert gas in said head tank, recirculating the liquid metal necessary for electrolysis into the cell, condensing and separating the electrolytically liberated metal contained in the inert gas from the gas, and returning the inert gas into the lift pipe for further use by circulation.

This is a divisional of application Ser. No. 18,103 filed Mar. 10, 1970, now U.S. Pat. No. 3,642,603.

BACKGROUND OF THE INVENTION

In the method of producing an alkali metal and an alkaline earth metal with a conventional fused salt electrolytic equipment the fused alloy of the cathode liquid metal and the electrolytically liberated metal is distilled and separated and the liquid metal is recirculated into the electrolytic cell to be used as the cathode. In this circulating line a mechanically circulating pump is employed. In some case, the electrolytic cell is provided with a flow passage at a side or at the bottom thereof through which a liquid metal is passed as the medium for heat to control the temperature of the cell so that the conditions of electrolysis can be stabilized. In the circulating line, too, a mechanically circulating pump is employed.

The electrolysis of fused salt is operated at a considerably high temperature. The alloy of the cathode liquid metal and the electrolytically liberated metal both coming out of the cell and the metal for controlling the temperature of the cell must be circulated to each process of the equipment at high temperatures. However, there are various troubles about the circulation of the liquid metals at high temperatures. For instance, there are the problems of corrosion of the materials of the equipment with the liquid metal, the alloy of the liquid metal and the liberated metal, and the temperature controlling circulating liquid metal, the heat resistance of the equipment itself, and the heat insulation of the high temperature equipment. Also the mechanically circulating pump such as an electromagnetic pump or a centrifugal pump is liable, to get troubles at its shaft seal and other parts. Especially, the complexity of the construction of the equipment increases the possibility of troubles and it is often difficult to secure the safety of operation. All these undesirable factors cause the service life of the equipment to be shortened and the cost of fused salt electrolysis to be raised.

SUMMARY OF THE INVENTION

The present invention mainly comprises circulating the liquid metal into fused salt electrolytic cell in which a fused metal is used as the cathode by the jetting force of an inert gas instead of using a mechanically circulating pump in the liquid metal circulating line. This method of circulating the liquid metal simplifies the construction of the equipment, facilitates the selection of heat resistant materials and heat insulating materials, and assures the safety of operation, thus resulting in many advantageous effects.

More specifically, this invention provides a method of and an apparatus for circulating the liquid metal into said fused salt electrolytic cell comprising passing the fused alloy of the cathode liquid metal and the electrolytically liberated metal produced in the gap between the cathode and anode through the lift pipe together with the inert gas by the jetting force of the gas, making a mixture of the alloy and the inert gas in the lift pipe by the jetting force of the gas, said mixture being small in specific gravity, transporting the electrolytically liberated metal vapor alone into the inert gas in said lift pipe lifting said mixture into the head tank, separating the cathode liquid metal from the inert gas containing the electrolytically liberated metal in the head tank, condensing and separating the electrolytically liberated metal from the inert gas, cooling, compressing and circulating the inert gas for repeated use, and returning the liquid metal remaining on the bottom of said head tank to the electrolytic cell for re-use.

BRIEF DESCRIPTION OF THE DRAWING

FIG. 1 shows diagramatically the circulating line of a fused salt electrolytic equipment suitable for carrying out the method of the present invention.

FIG. 2 shows diagramatically the circulation device for the alloy of fused metals and the temperature controlling liquid metal in the electrolytic cell for the fused salt electrolytic equipment.

DESCRIPTION OF PREFERRED EMBODIMENTS

Referring to FIG. 1, the electrolytic cell **1** is heated by the heater **22**. The receiver **2** for receiving the alloy produced by electrolysis is also heated by the heater **23**. **3** is the lift pipe standing upright in said receiver **2**, **4** the head tank, **5** the condenser, **6** the heat exchanger also serving as gas cooler, and **7** the gas compressor, respectively. The alloy **10** of the cathode liquid metal **8** and the electrolytically liberated metal **20** produced by electrolysis in the gap between said cathode liquid metal **8** and the anode **9** made of graphite for instance in the electrolytic cell **1** enters the receiver **2** placed at a level lower than that of the cathode liquid metal **8** through the pipe **11**. In the receiver **2** is standing the lift pipe **3**. The lift pipe **3** has a length equal to the sum of the height of that part of the receiver **2** which is immersed in the alloy **10** contained therein and the length of the part lifting the alloy **10**. Said length of the lift pipe **10** should be such that when the lift pipe **3** is planted upright in the receiver **2** the upper end of said lift pipe **3** will come to a level at least higher than that of the cathode liquid metal **8** in the electrolytic cell **1**. In other words, since the lift pipe needs to have a head large enough to return the alloy **10** to the electrolytic cell **1**, it should have a length equal to the sum of a length a little greater than the height of the level of said cathode liquid metal **8** and an optional length added. Immediately below the lift pipe **3** is provided the nozzle **13** of the inner gas conduit **12**, and when a suitable volume of the inert gas **14** is jetted out at said nozzle **13** into the liquid alloy **10** the mixture **15** of the alloy **10** and the inert gas **14** is pro-

duced in the lift pipe **3**. This mixture **15** is smaller than the alloy **10** in specific gravity. The buoyancy created by the difference in specific gravity and the jetting force of said inert gas **14** combine to serve as a driving force to lift the mixture **15** through the lift pipe **3** into the head tank **4** provided at the upper end of said lift pipe **3**. At the same time the electrolytically liberated metal **20** alone has its vapor transported into the inert gas **14** by the effect of the pressure at its temperature. The liquid metal **8** is separated from the inert gas **14** containing the liberated metal **20** in the head tank **4**. The inert gas **14** containing the liberated metal **20** is introduced into the condenser **5** through the pipe **17** connected to the upper part of the head tank **4**, and the liberated metal **20** is condensed and separated from the inert gas **14** in said condenser **5** and then taken out at the discharge pipe **25**. On the other hand, the separated inert gas **14** is transported into the heat exchanger/gas cooler **6** through the pipe **18** and cooled there to a suitable temperature. Then it is conveyed further to the compressor **7** through the pipe **19** to be made into compressed inert gas **14** and finally blown into said lift pipe **3** through the pipe **12** for recirculation. The liquid metal **8** left behind in the head tank **4** flows down by gravity into the electrolytic cell **1** as the cathode liquid metal **8** for recirculation through the pipe **24** connected to a side of the head tank **4** at a level lower than the upper end of the lift pipe **3**.

In the case where the vapor pressure of the alloy **10** of the cathode liquid metal **8** and the electrolytically liberated metal **20** is low, the separation of the liberated metal **20** from the inert gas **14** containing the liquid metal **8** is facilitated by heating said alloy **10** by the heater **23** provided under the receiver **2**. If the inert gas **14** coming out of the compressor **7** is passed into the gas cooler **6** for heat exchange and blown into the nozzle **13** in a warmed state, the heat may be economized. In the condenser **5** the inert gas **14** is cooled to the degree that the liberated metal **20** can be taken out at a temperature a little higher than the melting point of said liberated metal **20**. As the liberated metal **20** thus condensed and separated generally contains slight quantities of the cathode liquid metal **8**, it is advisable to rectify the liberated metal **20** by a rectifier (not shown) to obtain high purity liberated metal **20**. It is possible to produce compounds of the liberated metal **20** by adding other metals directly to the inert gas **14** containing the liberted metal vapor **20**.

This equipment can not only lift the alloy **10** in the receiver **2** to required height through the lift pipe **3** by the jetting force of the inert gas **14** but also make a mixture **15** of the alloy **10** and the inert gas **14** by said jetting force and transport only the liberated metal vapor **20** into the inert gas **14** to separate the liberated metal **20** from the cathode liquid metal **8** without resorting to distillation.

The cathode liquid metal used in the present invention may be lead, tin, bismuth, or alloy of two or more of these, the fused salt may be a halide of any of alkali metals or alkaline earth metals or a combination of these, and the inert gas may be argon or nitrogen.

Now an embodiment of this invention will be described in detail by referring to FIG. 1. In the fused sodium chloride electrolytic equipment employing fused lead as cathode, the horizontal electrolytic cell **1** has a metal bottom plate and corrosion resisting brick-lined side plates. Fused lead and fused sodium chloride are introduced into this electrolytic cell **1**, and said sodium chloride is electrolyzed at a temperature of 850° C. to produce a lead alloy **10** containing about 10 percent by weight of sodium. Said alloy is introduced into the receiver **2** 150 mm. inside diameter and 1,500 mm. high, in which stands said lift pipe 25 mm. in inside diameter and 3,000 mm. long. Said head tank 800 mm.-by-800 mm. square and 1,500 mm. high is installed with its bottom at a level 300 mm. below the top of said lift pipe **3**. The lower end **21** of

said lift pipe **3** is open in the shape of a funnel, 100 mm. below which is arranged the pipe **12** having said nozzle 12 mm. in inside diameter for blowing out nitrogen gas. When the nitrogen gas is jetted out at this nozzle at a pressure of about 1.4 kg./cm.^{2} g. and at a rate of 1.0 liter/second, the fused lead is lifted into the head tank **4** at a rate of 0.11 liter/second. By cooling the nitrogen gas containing the sodium vapor to 350° C. in the condenser **5**, we obtained 1.17 kg./hour of sodium. The nitrogen gas was cooled to 50° C. in the gas cooler **6**, compressed at 2 kg./cm.^{2} g. in the oil-free compressor **7**, and recirculated. The fused lead in the head tank **4** was returned to the electrolytic cell **1** for recirculation. In case the capacity of the lift pipe to lift the fused lead is large for the electrolyzing capacity of the cell, part of the fused lead can be conveniently circulated between the receiver **2** and the tank **4**.

FIG. 2 shows a flow sheet of the said equipment installed together with a device for circulating a liquid metal for controlling the temperature of the electrolytic cell. The cathode liquid metal, fused salt and inert gas used in this embodiment are the same as those used for the previous one. **30** is the electrolytic cell provided at its bottom with the flow passage **31** for passing the temperature controlling liquid metal and with the heating section **74**. **32** is the receiver for receiving the liquid metal remaining in the distiller, said receiver **32** having the heating section **75**. **33** is the receiver for receiving the liquid metal for controlling the temperature of said electrolytic cell **30**, said receiver **33** having the heating section **76**. **34** and **35** are the lift pipes each planted in the receivers **32** and **33**, **36** and **37** the head tanks, **38**, **39**, **40** and **41** the inert gas coolers, **42** the gas supplying compressor, and **43** the heat exchanger for the liquid metal, respectively. The circulating line for the alloy **46** of the cathode liquid metal **44** and the electrolytically liberated metal produced by electrolysis in the gap between cathode liquid metal **44** and the anode **45** in the electrolytic cell **30** is the same as that for the previous embodiment. Namely, the alloy **46** is introduced into the receiver **32** through the conduit **47**. The receiver **32** is so installed that the level of the liquid metal **48** contained in said receiver **32** will come to a level lower than that of the liquid metal remaining on the bottom of the cell **30**. In the receiver **32** is planted the lift pipe **34**, the upper end of which should come to an optional level higher than that of the cathode liquid metal **44**. The orifice **50** at the lower end of the immersed lift pipe **34** is fronted from below by the nozzle **51** of the conduit **72** coming from the compressor **42**. The inert gas **52** jetted out flows into said lift pipe **34** through the orifice **51** and makes a mixture **53** of the liquid metal **48** and the inert gas **52** having small specific gravity. Said mixture **53** rises in the lift pipe **34** by the jetting force of the inert gas **52** and is lifted into the head tank **36** provided above, where the liquid metal **48** and the inert gas **52** are separated from each other by the difference of specific gravity. The liquid metal **48** thus separated in the head tank **36** flows down into the electrolytic cell **30** by gravity through the connecting pipe **55** and is circulated again as the cathode liquid metal **44** to make the alloy with the electrolytically liberated metal.

As for the circulation of temperature controlling liquid metal **56**, the temperature controlling liquid metal **56** coming from the bottom plate **57** of the cell **30** or through the outlet **58** of the tunnel-shaped flow passage **31** provided in said bottom plate **57** is introduced into the receiver **33** installed at a level lower than that of said outlet **58**. In the receiver **33** is planted the lift pipe **35** as in the case of the above-mentioned receiver **32**. The inert gas **52** supplied through the conduit **73** of said compressor **42** is blown at the nozzle **61** into the lift pipe **35** through the orifice **60** at the lower end thereof. The mixture **62** of the liquid metal **56** and

the inert gas **52** is lifted into the head tank **36** provided above the lift pipe **35** through said lift pipe **35**. The liquid metal **56** separated from the inert gas **52** in said head tank **36** flows down through the pipe **64** connected to the outlet **63** of the head tank **36** and enters the heat exchanger **43**. In order that the electrolytic cell **30** containing said liquid metal **44** can be kept at a suitable temperature, said liquid metal **56** heated or cooled in said heat exchanger **43** is discharged from the heat exchanger **43** and flows by gravity into the inlet **70** of the flow passage **31** of the cell **30** and then, after controlling the temperature of the electrolytic cell **30** to a level suitable for electrolysis of the fused salt, returns to the receiver **33** through the pipe **59** for recirculation.

The inert gas **52** used as driving force in the circulating lines of the cathode liquid metal **44** and of the temperature controlling liquid metal **56** is separated in the head tanks **36** and **37** respectively. The metal vapor contained in small quantities in the inert gas **52** is collected and separated in the coolers **38** and **39** installed above the head tanks **36** and **37** respectively to prevent the metal vapor from solidifying and choking the circulating lines. After being made free from the metal vapor the inert gas **52** is further introduced in the coolers **40** and **41** through the pipes **65** and **66** to be cooled to a temperature substantially suitable for giving no troubles to the compressor **42**. Then the inert gas **52** is returned to the compressor **42** through the pipes **67** and **68** and compressed to a specified pressure high enough to work as the driving force for circulation. During all this time the operation is carried out with the pressure kept slightly higher than the atmospheric pressure to prevent the entry of air into the inert gas piping. The inert gas **52** at said specified pressure is blown into the lift pipes **34** and **35** through the nozzles **51** and **61** provided below them to circulate the cathode liquid metal and the temperature controlling liquid metal.

In the embodiment shown in FIG. 2 the numerical values are the same as those for the previous one and liquid lead is circulated in the temperature control circulating line. It was possible to stabilize the operational conditions of the electrolytic cell by keeping the temperature of liquid lead at 900° C. at its inlet and at 850° C. at its outlet by means of the heat exchanger **43**. The fused salt, therefore, was electrolyzed in a highly stabilized state.

Since both equipment shown in FIG. 1 and FIG. 2 do not need the use of mechanically circulating pumps in the liquid metal circulating lines, no severe conditions of materials or complicated constructions are required of the whole equipment, the design of the equipment is

simplified with regard to influences of thermal stress, and no special kind of insulating means are necessary for the piping. For these reasons, relatively low-priced materials may be used, the repair and replacement are easy, the safety of operation is high, and the service life is long.

What we claim is:

1. A method of circulating a liquid metal is fused salt electrolytic equipment comprising making a mixture of the alloy of cathode liquid metal and electrolytically liberated metal coming out of the electrolytic cell and an inert gas by the jetting force of said inert gas, lifting the mixture into a head tank through a lift pipe and separating the liquid metal from the inert gas in said head tank, the liquid metal being returned to the electrolytic cell and the gas being jetted again into the lift pipe.

2. A method of circulating a liquid metal defined in claim **1**, wherein a liquid metal controlling the temperature of the electrolytic cell is circulated by the use of another receiver, lift pipe and head tank, an inert gas being jetted into the lift pipe in the receiver to lift the liquid metal into the head tank together with the inert gas, the liquid metal being flowed into the receiver through the electrolytic cell and the inert gas being jetted again into the lift pipe.

3. A method of circulating a liquid metal defined in claim **1**, comprising a separation system wherein the alloy of cathode liquid metal and electrolytically liberated metal produced in the gap between the cathode liquid metal and the anode is passed through the lift pipe together with the inert gas to make a mixture of alloy and gas, only the liberated metal for its vapor pressure being transported from the mixture into the inert gas, the remnant alloy being retuned to the electrolyitc cell for circulation and the inert gas being separately circulated.

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JOHN H. MACK, Primary Examiner

D. R. VALENTINE, Assistant Examiner

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