Aug. 29, 1933. L. BURGESS 1,924,151

METHOD FOR THE MANUFACTURE OF BERYLLIUM AND BERYLLIUM ALLOYS

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Fig. 3

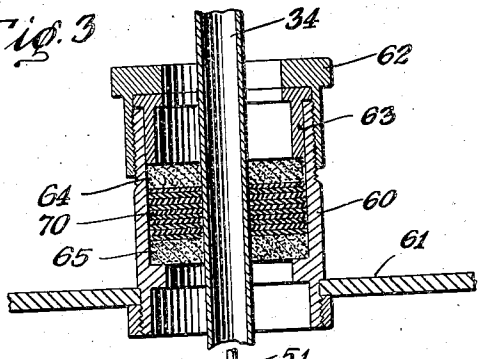


Fig. 2

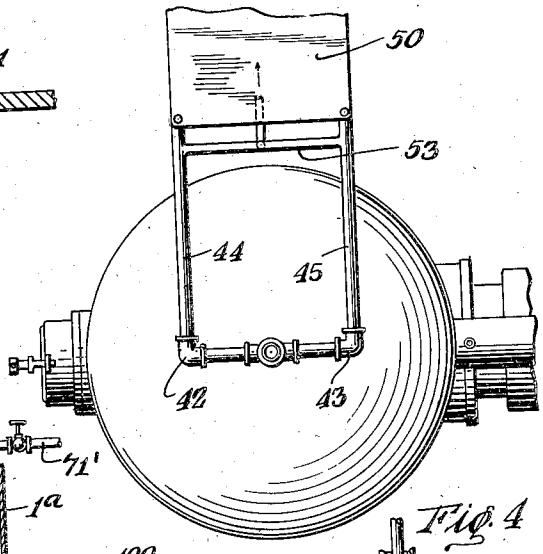


Fig. 1

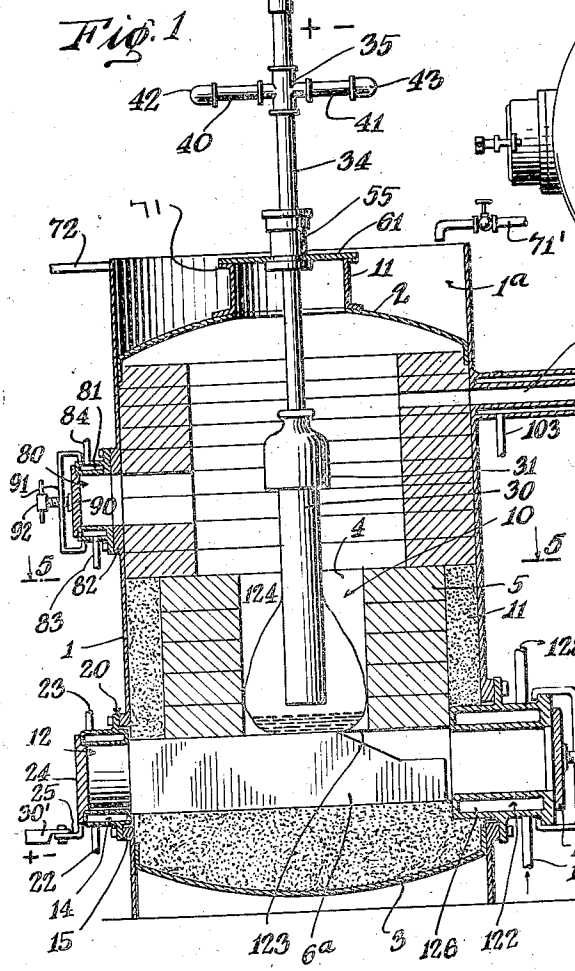
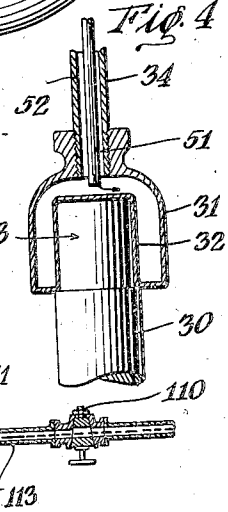


Fig. 4



Inventor

Louis Burgess

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METHOD FOR THE MANUFACTURE OF BERYLLIUM AND BERYLLIUM ALLOYS

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Fig. 5

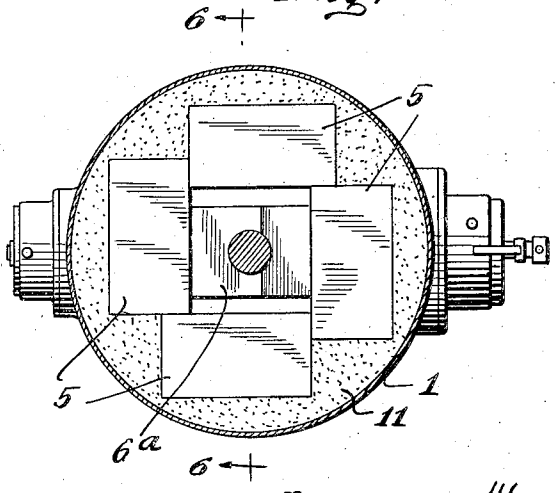


Fig. 6

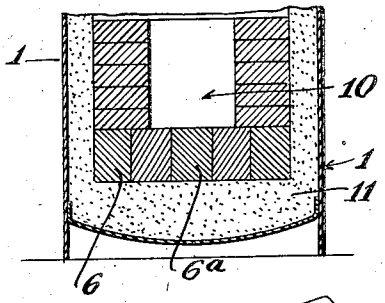


Fig. 7

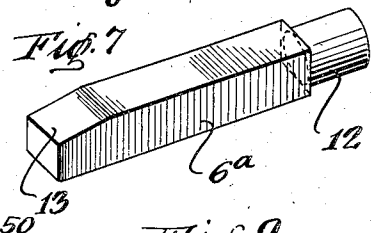


Fig. 8

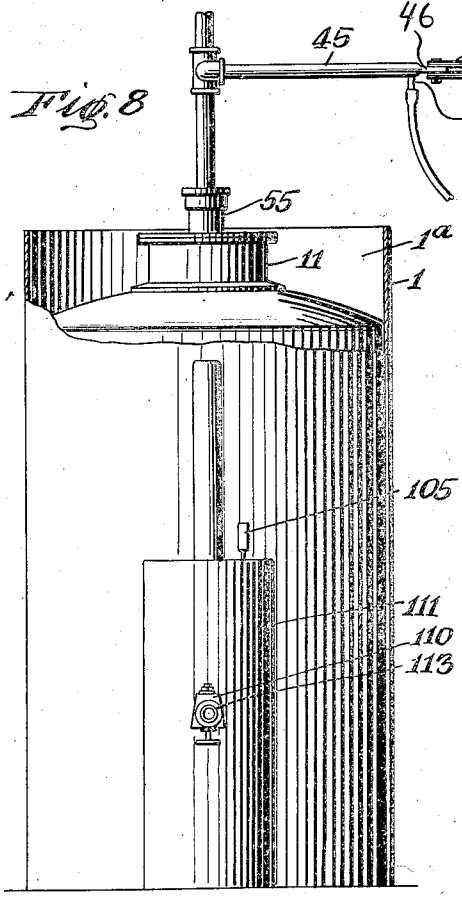
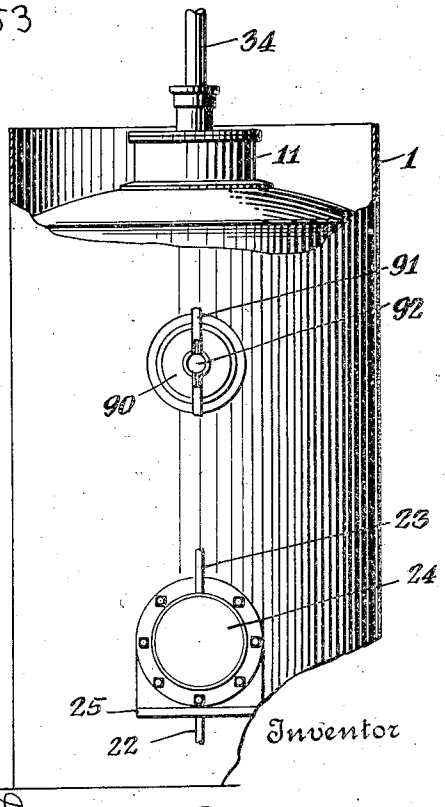


Fig. 9



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METHOD FOR THE MANUFACTURE OF BERYLLIUM AND BERYLLIUM ALLOYS

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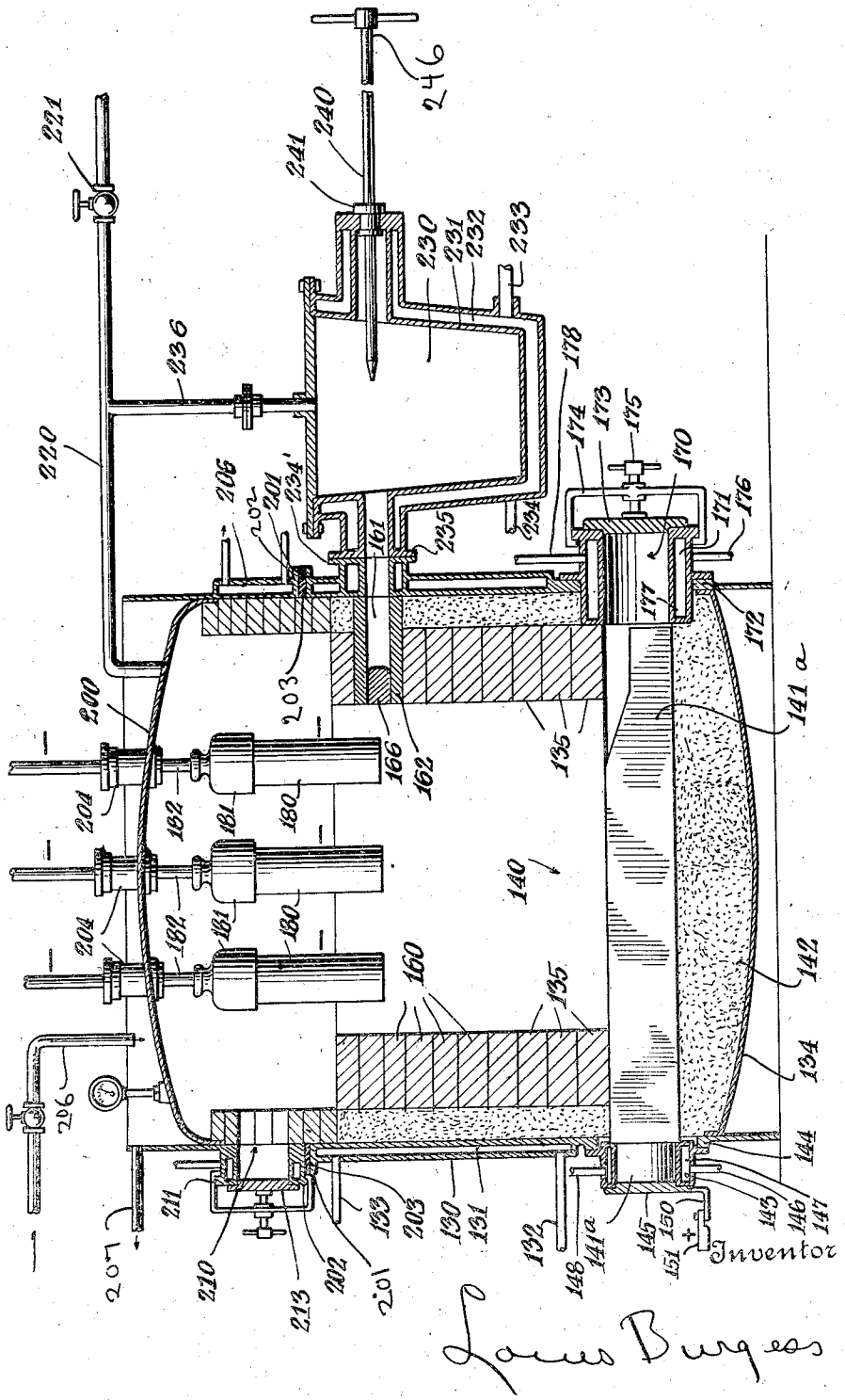
Fig. 10



Patented Aug. 29, 1933

1,924,151

UNITED STATES PATENT OFFICE

1,924,151

METHOD FOR THE MANUFACTURE OF BERYLLIUM AND BERYLLIUM ALLOYS

Louis Burgess, New York, N. Y.

Application December 24, 1930
Serial No. 504,451

31 Claims. (Cl. 204—19)

My invention relates to a process and apparatus for the manufacture of beryllium and beryllium alloys, and will be fully understood from the following description read in conjunction with the drawings, in which,

Fig. 1 is a vertical section through a form of apparatus in which certain features of my invention may be carried into effect.

Fig. 2 is a top view of the construction shown in Fig. 1.

Fig. 3 is a detailed section through part of the construction shown in Fig. 1.

Fig. 4 is a detailed vertical section through another part of the construction shown in Fig. 1.

Fig. 5 is a horizontal section through the construction shown in Fig. 1 on the plane designated by 5—5.

Fig. 6 is a vertical section through the construction shown in Fig. 5 on the plane indicated by 6—6.

Fig. 7 is a perspective view of part of the construction shown in Fig. 1.

Fig. 8 is a side elevation of the construction shown in Fig. 1.

Fig. 9 is a side elevation of the construction shown in Fig. 1 taken at an angle of 180° to the showing in Fig. 8.

Fig. 10 is a vertical section through another form of apparatus in which certain features of my invention may be carried into effect.

One important feature of my invention comprises the manufacture of alloys of beryllium with other metals, under conditions hereinafter more fully set forth. The metals in general to be united with beryllium are those boiling above 2200° C., and particularly the alloys of beryllium with copper. I am aware that alloys of this sort have been heretofore manufactured by reduction effected through the direct application of the heat of an electric arc to a charge containing the oxide together with the necessary amount of carbon to effect reduction. Regardless of the particular type of arc furnace employed for this purpose, I have found that an increase in efficiency and that higher concentrations of beryllium in the resulting alloy may be obtained by the use of a charge in which the material yielding the metal boiling above 2200° C. is disseminated throughout the charge in divided condition and the individual particles or subdivisions of which, or at least a major part of the same, are held in a supporting matrix of carbon. A charge of this sort may be prepared by mixing the oxidized beryllium and the components yielding the metal boiling above 2200° C., or the free metal itself where free metal is added to the charge, in a divided and preferably pulverized condition with a hydrocarbon bonding material and thereafter coking and dividing the same. The materials may, for example, be mixed with high melting point pitch, whereupon the mixture is carefully coked to form a solid mass of carbon throughout which the granules of metal yielding material are disseminated and distributed. The coked charge is then preferably ground to appropriate size, say to pass one inch mesh.

It will be perceived that by this method the charge is formed into a continuous phase of carbonaceous material and a discontinuous phase of metal yielding material which may be the oxide and may include free metal where this is a component of the charge. The percentage of pitch employed may be sufficient to furnish all the necessary carbon, although in the preferred practice the charge may be made by incorporating the necessary amount of coal, coke or other carbonaceous material together with sufficient pitch to furnish a satisfactory bond. Alternatively, certain quickly coking materials may be employed to furnish the bond, such as for example sugars, starches and similar materials adapted to develop a cokey structure, and this cokey structure may be consummated, either by a preliminary heating or in the furnace itself, as the charge is gradually warmed before it enters the zone in which it is subjected to the direct arc heating.

Where the reduction is conducted in the ordinary manner without such a charge, the metal to be alloyed with the beryllium is ordinarily reducible at a temperature below the reduction temperature of beryllium and freely fluid so that it runs quickly down into the pool beneath the electrode. In this case alloying is effected principally through the boiling up of this pool, and the vapor of the metal of higher boiling point is presumably not uniformly distributed through the zone of reduction. With a charge of the type just described, however, the metal is retained and supported by the carbon of the charge so that the metal is released simultaneously with the consumption of the carbon, and for this reason to a large extent simultaneously with the reduction of the oxidized beryllium. The amount of carbon requisite under any circumstances can, of course, be calculated by a competent chemist and should be stoichiometrically sufficient to combine with the oxygen present with the resultant formation of carbon monoxide. If for any reason it is necessary to leave part of the oxide unreduced in order to form a slag, a corresponding deficiency

of carbon will be employed; and conversely, if with the particular charge there develops an excessive tendency to corrode or consume the electrodes and/or other carbon parts of the furnace, a slight excess of carbon may be added. In this case of course the charge will contain some carbon, and the amount of free carbon in the resulting alloy should be kept to a low figure as larger proportions tend to the formation of carbides and the production of an alloy of unduly high melting point.

Metallic beryllium has a relatively low boiling point, and a high vapor pressure at the temperature of reduction. This temperature of reduction is presumably in the region of from 1700 to 2050° C. and is subject to some variation, depending on the amount and character of the alloying metal or metals. Where an effort is made to generate alloys of relatively high beryllium content, say for example in excess of 20, the action is apt to be irregular due to excessive vapor pressures in the furnace shown by a tendency of the metallic vapors to blow out and burn, accompanied by a relatively low recovery and a low electrical efficiency. These difficulties may be to a large extent inhibited by the employment of the method shown and described in my prior Patent No. 1,512,271, issued October 24, 1924. The apparatus shown in Figs. 1 to 8, inc. is a modification of the same.

Referring specifically to Fig. 1, the modified apparatus comprises the shell 1 furnished with end closures 2 and 3, the whole constituting a receptacle or vessel adapted to carry high pressures which may range for example from a few atmospheres up to in certain cases 20 or more atmospheres. The lower part of this vessel contains the pit 4 which retains the charge to be reduced and which also functions to retain the reduced material until the same is tapped off. The side walls of this pit are formed by the carbon blocks 5 which may be arranged in the manner shown in Fig. 5. The floor of the pit may also be formed of carbon blocks 6 laid in parallel in the arrangement indicated in Fig. 6 which is a view in vertical section through the construction shown in Fig. 5 on the plane indicated by 6—6. This pit may be collectively designated by the numeral 10. It is preferably thermally insulated from the shell 1, and this may be accomplished by means of a lining, such as 11, composed of finely divided refractory material of high melting point, such as alumina, bauxite, magnesia, or finely divided carbon. The pit 10 in a single phase furnace constitutes preferably one electrode and for this reason must be suitably placed in electrical communication with one leg of the electrical circuit. This is preferably accomplished in the following manner:

One of the carbon blocks constituting the floor of the pit, in this case the central block 6^a, has one end 12 turned in a lathe to a true cylindrical surface, while the other end 13 is preferably beveled or recessed to form a tapping duct. (See Fig. 7). The block 6^a is then set in a water cooled annulus 14, (Fig. 1) which is bolted or otherwise secured to the flange 15 constituting a part of the shell 1. The connection 20 between the annulus 14 and the flange 15 is of course gas-tight and a bolted connection is preferable, inasmuch as it facilitates the removal of the carbon block 6^a when necessary and its substitution by a new one. The annulus 14 and block 6^a may be joined in electrical contact by the following method:

The end 12 of the block 6^a is first turned to the internal diameter of the annulus 14. The annulus is then expanded by steam or other application of heat and slipped over the end 12 of the block 6^a. On cooling, the tube becomes firmly attached and will maintain effective electrical contact, since the annulus 14 is normally cooler than the block 6^a. The annulus 14 is cooled in normal operation by the introduction of cooling water through the inlet 22 which issues from the educt 23. The annulus 14 is in turn sealed gas-tight by the machined flange 24 which may be extended downwardly to form the lug 25 to which the cable, bus-bar or other electrical connector 30' is suitably attached. The opposed electrode is formed by the carbon rod 30 carried in the electrode holder 31. The construction of this is shown in detail in Fig. 4. It may be a casting of copper or brass internally machined to form the true cylindrical surface 32. The end 33 of the electrode 30 is preferably similarly machined to form a companion surface, and the two are secured together by expanding the electrode holder 31, applying it to the electrode end 33 and then permitting it to cool. The electrode 30 and holder 31 are carried by the tube 34, (Figs. 1 and 4). The tube 34 may be raised and lowered during operation in any suitable manner. This may, for example, be accomplished by the manual control device shown in my prior Patent 1,512,271, or alternatively an electrode regulating device of the mechanical type may be employed. The tube 34 is threadably connected through the cross 35 to the nipples 40 and 41, which are in turn connected through the elbows 42 and 43 to the tubes 44 and 45. The tubes 44 and 45 terminate in flange ends 46 which are bolted to the bus-bars 50, (Fig. 2). The bus-bars are preferably composed of laminated sheet material so that the bus-bars and the tubes 44 and 45 adapt themselves to variations in the height of the pipe 34 and connected electrode. Cooling water is introduced through the internal pipe 51 which discharges into the electrode holder 31, the water returning through the annular space 52 betwen the pipes 51 and 34, and escaping from the system through the pipes 44 and 45 and the connected educt 53, (Fig. 2). It is of course essential that the tube 34 be insulated from the cover 2, and for this purpose the tube reciprocates in a stuffing box 55 (Figs. 1 and 3) of special construction. This comprises the housing 60, threaded through the cover plate 61, which housing carries the cap 62 and the gland 63. The gland 63 serves to compress the washers 64 and 65 of suitable insulating material, such as asbestos wood. Between the washers 64 and 65 are a number of washers of yielding material 70 composed, for example, of sheet rubber or similar composition. The housing 60 and the metallic connected parts are spaced away from the tube 34 to effectively insulate the same. In addition, the cover 61 may also be insulated from the flange 71, (Fig. 1), although this is not ordinarily necessary.

As shown in Fig. 1, the shell 1 carries the upward extension 1^a to which water may be admitted through the pipe 71', flowing off through the educt 72, thereby forming a pool of water on the roof of the furnace which prevents overheating at any point. Charging of the furnace is accomplished through the manhole 80, comprising the water cooled annulus 81, connected to the shell 1 by the flange 82. Cooling water may be admitted through the inlet 83 and withdrawn

through the educt 84. The hole 80 is sealed by the removable cover 90 which may be secured in position by means of the dolly-bar 91. A paper gasket is ordinarily employed to make the gas-tight joint between the plate 90 and the machined surface at the end of the annulus 81. The cover plate 90 is removed by loosening the turn-screw 92 carried by the dolly-bar and swinging or lifting the plate 90 from position. The current is ordinarily on during the charge period so that cold charge will not fall in under the electrode 30 and prevent the institution of an arc. As soon as the pit 10 has been completely filled with charge the cover plate 90 is replaced and secured firmly in position. The requisite pressure builds up quickly in the furnace and the excess gas over that necessary to maintain the pressure is removed through the side outlet 100. This is water cooled by means of the jacket 102 to which cooling water is introduced by means of the inlet 103. This cooling water passes off through the jet 104 to scrub and quench the gases withdrawn from the furnace. These gases ordinarily contain small abrasive particles which injure a valve seat, but by quenching them with water in the manner indicated the difficulty is entirely overcome. The actual pressure in the furnace can be determined by means of the pressure gauge 105 and the excess gas released by manipulation of the valve 110 which may be of the globe seat type. The water and gas pass into the tank 111 in which the water accumulates up to about the level 112.

In the normal operation of the furnace in combination with this apparatus, the valve 110 is opened to a point sufficient to hold the pressure as shown by the gauge 105 at a predetermined point. It will immediately be perceived that before the gas can pass out through the pipe 113 and valve 110 the water must be vented from tank 111 up to the level 112. In this way the disposition of the water takes place automatically so long as the gas is vented. After operating the furnace for a period, the current is discontinued and the gas pressure is gradually dropped to atmospheric. As soon as this has occurred, the cover plate 120, Fig. 1, is removed by loosening the dolly-bar 121, and tapping is effected through the water cooled annulus 122 into a tapping pot or buggy. Cooling water is introduced to the water jacket 126 of the annulus 122 by means of the inlet 127. The water leaves the jacket through the educt 128. When all the metal has been withdrawn in this manner, the tapping outlet 123 is sealed. This may easily be accomplished by throwing in some cold charge and tamping the same in position. The current is again applied and fresh charge is introduced through the charging port 80. In practical operation, the reduction of a particular batch of charge is preferably carried only so far as will leave a basket 124 of unreduced material standing in the furnace. Before introducing the fresh charge, a workman ordinarily reaches in with a bar through the charging port 80 and breaks down the basket formation in the furnace so as to permit the fresh charge to entirely fill the pit 10.

Where the furnace hereinbefore described is to be operated in combination with the steps hereinafter described for the isolation of beryllium from the alloy generated, the charge is so proportioned as to furnish an alloy of copper and beryllium in the desired proportions. Such alloys should contain in excess of 15% beryllium based on the total copper and beryllium present, and may contain in addition several percent of silicon. The latter ingredient is recommended where subsequent electrolytic refining is to be carried out at relatively low temperatures, say below 1000° C., for the purpose of keeping the alloy fluid as the beryllium content is reduced. The current densities, voltage and amperage may be determined by comparison with electric furnace practice and similar materials reduced in this type of furnace, for example ferro-silicon. In the furnace illustrated, in which the vertical electrode is six inches in diameter and the pit is approximately two feet square (other dimensions in proportion), the reduction may be successfully carried out with a voltage of about 60 and an amperage of about 2500, although higher voltages and amperages may be employed; and for large furnaces voltages in excess of 100 may be found highly desirable. The duration of the run is preferably less than required to completely reduce the charge in the pit and should leave standing a basket of unreduced material. For the manufacture of alloys containing in excess of 20% of beryllium, I have found pressure of the order of 4 to 10 atmospheres to be helpful. In the furnace illustrated, runs of about one hour duration at 60 volts and about 2500 amperes can be carried out.

After drawing off the alloy containing beryllium and copper, it should be carefully separated from slag or non-metallic inclusions, and for this purpose is preferably held at a molten temperature in a refractory vessel until substantially complete separation has taken place. While I have hereinabove described the preferred method of generating an alloy suited for electrolytic refining, it will be understood that in combination with such electrolytic refining I may substitute some other method of preparing the alloy; conversely, I may also within the purview of my invention employ the reduction furnace hereinabove described to manufacture beryllium alloys for any purpose whatsoever.

In operating the reduction furnace, I may employ in whole or in part the ores of beryllium, particularly those carrying relatively high beryllium contents, but I preferably add the oxidized beryllium in the form of the anhydrous oxide and this may be conveniently prepared by the method covered by my prior U. S. Patent 1,418,527, issued June 6, 1922.

Where an alloy containing both beryllium and aluminum is required, aluminum oxide will be added to the furnace charge. The beryllium oxide may be replaced with alumina where a corresponding alloy containing aluminum instead of beryllium is desired.

For the electrolytic refining I preferably employ the apparatus illustrated in Fig. 10. This comprises the shell 130, surrounded by the water jacket 131, through which cooling fluid may be circulated by means of the pipes 132 and 133. The lower part of the shell 130 is sealed by the end closure 134. The lower part contains a pit built up of carbon blocks 135 laid in substantially the same manner as the blocks 5 shown in Fig. 5. The floor of the pit 140 is sealed by the carbon blocks 141 and similar blocks arranged in parallel in the same manner as the blocks 6ª and 6 in Fig. 6. In this case, inasmuch as fluid metal washes the surface of the carbon blocks, they should be carefully sealed in position and may be laid with heavy asphalt and thereafter carefully heated to coke or carbonize the asphalt. Be-

tween the blocks constituting the pit 140 and the shell 130 there is firmly tamped a lining of refractory material 142, such as bauxite or magnesia. The rod 141ª is sealed to the water cooled annulus 143 which is bolted to the flange 144. The annulus is provided with a gas-tight cover 145. Cooling water may be introduced through the pipe 146 to the jacket 147. The water escapes through the educt 148. The cover 145 may terminate in the lug 150 to which cables may be attached to provide the necessary electrical connection. For this purpose cable lugs such as 151 may be bolted to the lug 150. The upper part of the pit 140 is composed of bricks 160 of refractory material, preferably bonded beryllium oxide. An overflow or tapping outlet 161 is provided adjacent the upper part of the shell 130, and for certain uses this may be provided with an insulating liner 162. The bricks 160 are preferably snugly joined and the space between the shell and bricks is also filled with a tightly tamped insulating material, such as bauxite. Any molten metal in the lower part of the pit 140 may be withdrawn through the tapping duct 170 defined by the water cooled annulus 171, secured to the flange 172, and normally sealed by the cover plate 173, locked in position by the dolly-bar 174 and turn-screw 175. Cooling water may be introduced through the pipe 176 to the water jacket 177: the water escaping through the educt 178. Dipping into the pit 140, adjacent the upper portion thereof, are a number of electrodes 180, preferably graphite, carried by electrode holders 181 which are similar to 31 hereinbefore described. The holders 181 are in turn carried by the tubes 182 provided with an internal pipe through which cooling water is introduced and with a suitable educt for the cooling water. The tubes 182 are suitably connected to the negative side of an associated electrical circuit. There is connected to the upper part of the shell 130 the domed cover 200 by means of the companion flanges 201 and 202 carried by the cover and shell, respectively. An insulating gasket 203 is preferably laid between the two and the bolts or other connecting means are suitably insulated to prevent electrical contact between the shell 130 and cover 200. The cover is preferably provided with an annular water jacket 206. The stuffing-boxes 204 are preferably of the same type shown in detail in Fig. 3. The cover carries a side outlet 210 defined by the flange 211 which may be sealed gastight by means of the cover 213. The roof 200 may be cooled by the introduction of water through the pipe 206, the excess flowing off through the pipe 207.

In one mode of operating the device, the cover may be entirely removed and the electrodes 180 suspended in any suitable convenient manner. In this mode of operating the device the pit 140 is first charged with a layer of the beryllium copper alloy which may contain silicon as aforesaid. The upper level of this pool should extend above the level of the carbon blocks 135. There is then poured into the furnace an electrolyte of lesser density than the said alloy. In this electrolyte all the metal compounds present, excepting beryllium, should require a higher E. M. F. for electrolytic decomposition than the beryllium compounds present. If, however, the simultaneous production of both aluminum and beryllium is desired, aluminum salts should be present. A fluoride electrolyte is preferred. This may conveniently be composed of beryllium fluoride, fluorides of the alkali metals sodium and potassium, and fluorides of the alkaline earth metals barium and strontium. In general, the beryllium fluoride may constitute about 20% of the mixture, and each of the other groups may represent about 40% of the mixture. Some latitude may be exercised in the selection of individual compounds within the groups mentioned. For example, the substitution of potassium fluoride for sodium fluoride will result in increasing the gravity of the electrolyte. The same result may be obtained by increasing the proportion of the barium fluoride at the expense of the strontium. A few percent of beryllia may be added. Preferably, the electrolyte is charged to the depth of several inches. The electrolyte may be denser than metallic aluminum at the operating temperature. This may be insured for any particular temperature of operation by a selection of the components of the electrolyte within the groups mentioned. The density of the electrolyte may be increased by increasing the proportion of potassium and barium fluorides present at the expense of the sodium and strontium, respectively. For this purpose, either the gravity of test batches may be determined by experiment, or such test batches may simply be held at the operating temperature, and it may then be practically ascertained whether molten aluminum will remain afloat on a batch of particular composition. The result may, of course, be insured by forming the electrolyte entirely of beryllium, barium and potassium fluorides in the proportions mentioned, but the substitution of strontium and/or sodium fluorides may be imperative when alloys high in beryllium are used as anode which are of low density and which in certain proportions may approach the gravity of the electrolyte. In this case the density of the electrolyte must be reduced while remaining however above the density of the cathode layer floating thereon. There is then poured into the furnace molten aluminum to the depth of a few inches. The electrodes 180 make contact with the molten aluminum, and on passing a current from the anode layer of copper beryllium alloy through the electrolyte into the said aluminum layer the beryllium is caused to migrate selectively from the anode layer into the aluminum cathode layer, thereby building up and forming an aluminum beryllium alloy.

This method of starting up the electrolysis with a cathode of aluminum is relatively simple. Where, however, the anode layer contains a high beryllium content, it may be necessary to choose an electrolyte which floats thereon, but which is in turn incapable of floating metallic aluminum at the operating temperature. In this case, the upper cathode layer is preferably formed of an aluminum beryllium alloy of lesser density than the electrolyte. In either case, it is feasible to add aluminum continuously to the upper cathode layer while simultaneously withdrawing an aluminum beryllium alloy of any predetermined desired composition. This may be accomplished continuously or periodically. It may be accomplished periodically by tapping through the outlet 161 or continuously by means of an overflow trough provided with an adequate refractory insulating liner. The temperature during this stage of operations may vary from 800 to 1200° C., although I prefer a temperature in the neighborhood of 1000° C. and a current density of approximately 1000 amperes per square foot of cross sectional pit area. The voltage may vary between 3 and 8 volts between terminals. The voltage will, of course, be increased by increasing the depth of the electrolyte

layer and should be sufficient to insure sufficient wattage consumed in the furnace to maintain the temperature at the predetermined point. I prefer to work with an anode layer containing at least 50% of copper. Where the operation is to be started with a cathode layer of pure aluminum or where an alloy containing a minor proportion, say not over 15%, of beryllium is to be made, the electrolyte may have the following composition:

	Per cent
Beryllium fluoride	20–25
Barium fluoride	35–40
Sodium and potassium fluorides	35–40

The ratio of potassium to sodium to be so adjusted that the electrolyte has a density in excess of that of aluminum at the working temperature.

If the removal of the cathode alloy is intermittently practiced, it may completely be withdrawn from time to time through the duct 161 which is ordinarily sealed with a plug of refractory material. The copper beryllium alloy now of reduced beryllium content may then be withdrawn through the tapping outlet 170. This outlet is sealed as soon as electrolyte begins to issue. Fresh copper beryllium alloy is then charged in molten condition together with electrolyte if required to replenish the quantity in the furnace, and a cathode layer of aluminum or aluminum beryllium alloy is then added whereupon the electrolysis may be resumed.

Other metals lighter than aluminum may be employed in forming the cathode layer, as for example lithium and/or magnesium. In this case, however, the added lithium and/or magnesium should be protected by surrounding it and the cathode layer with an atmosphere of an indifferent gas, such as hydrogen, or an inert gas.

Where making alloys of high beryllium content, the operating temperature should be increased for best results. In this case, owing to the lesser density of the cathode layer, the relative amount of beryllium fluoride in the electrolyte may be increased. In this case, I may for example work with an electrolyte containing 30 to 40% of beryllium fluoride, the balance approximately equally divided between barium and sodium fluoride. As the temperature is increased, however, the fluorides tend to volatilize. I find, however, that this tendency may be inhibited by conducting the electrolytic operation under pressure. The operation may, for example, be carried out under pressure of a gas in excess of atmospheric where the temperature of the electrolyte is in excess of 1000° C. Any pressure in excess of atmospheric is beneficial, although pressures of a few atmospheres, say from 1 to 3 or higher, are recommended. The pressures should be maintained by means of an indifferent gas, although other gases may be employed inasmuch as the cathode layer is ordinarily protected by a skin of solidified electrolyte. Air may be employed, but the best results are obtained by the use of hydrogen or an inert gas. Carbon monoxide may also be employed. For commercial practice hydrogen is preferred. In this case, the operation is conducted with the cover in place and with the plate 213 tightly secured to prevent the escape of the gas. The predetermined pressure is maintained by passing the gas in through the pipe 220 controlled by the valve 221. The gas may conversely be withdrawn through this pipe if for any reason the pressure becomes excessive. By operating at pressures in excess of atmospheric it is possible to operate at temperatures above the melting point of beryllium metal. The melting point of beryllium appears to lie between 1275 and 1300° C. A pressure of one atmosphere gauge appears to be sufficient to permit the attainment of the melting point, but to insure sufficient fluidity and to provide for variations of temperature during operation, I prefer pressures of at least two atmospheres gauge. The voltage and amperage must of course be adjusted to maintain the predetermined temperature. The temperature may also be maintained at any predetermined point by passage of alternating current through the electrolyte by means of a circuit which is separate from the direct current employed for electrolysis. In this case the cathode layer may be gradually built up by electrolysis under pressure so that metal need not be supplied to form a cathode layer at the commencement of the operation. This procedure enables the manufacture of substantially pure beryllium metal: in this case the temperature is maintained above the melting point of beryllium and the pressure within the apparatus in excess of the vapor pressure of beryllium at the melting point.

Under these circumstances, if the furnace were permitted to cool the beryllium metal would solidify and its removal would present considerable difficulty. I, therefore, effect the removal of the beryllium metal in the following manner: There is connected to the furnace a pressure tight tapping pot 230. This comprises the iron vessel 231, surrounded by the water jacket 232, through which water may be circulated by means of the pipes 233 and 234. The flange 234′ carried by the electrolytic cell is connected to the flange 235 carried by the tapping pot 230 to form a detachable gas-tight connection. The pressure in the tapping pot is held at the same pressure as that obtaining in the electrolytic cell by means of the pipe 236. The tapping duct 161 is normally sealed by a plug 166 of refractory material. After the electrolysis has been continued for a period and a layer of beryllium metal has accumulated in the cell, the plug of refractory material in the tapping duct 161 is dislodged by means of the bar 240, operated from without through the stuffing-box 241 by means of the handle 246, whereupon the beryllium flows in the tapping pot 231. Any alternative externally controllable means for opening a passageway through the tapping duct may be employed. When this operation has been completed, the furnace is permitted to cool to about 1000° C., whereupon the pressure in the electrolytic cell is reduced to atmospheric. The impoverished anode layer is then withdrawn. A new plug 166 is inserted in the duct 161. Fresh anode metal may then be supplied through the inlet 210. Any electrolyte lost is then replenished. The tapping pot 230 is then removed and replaced with an empty receptacle. When the ingot of beryllium has cooled sufficiently it may be removed from the pot first employed. The pressure is then restored and the electrolysis resumed.

The term material containing metallic beryllium, as employed in the claims, connotes either substantially pure beryllium, or material consisting in part of free beryllium such as the alloys thereof.

The foregoing description is for purposes of illustration and since the invention may be carried out in a variety of forms other than those specifically described, it is my intention that the

invention be limited by the appended claims or their equivalents in which I have endeavored to claim broadly all inherent novelty.

I claim:

1. Process of forming by electrolysis cathodic material containing metallic beryllium, which comprises maintaining a fluid electrolyte containing a dissolved beryllium compound and metallic fluorides of higher dissociation potential in contact with anodic material, consisting of a fluid alloy containing beryllium and copper, and electrolyzing the same.

2. Process according to claim 1, carried out under pressure of a gas in excess of atmospheric.

3. Process according to claim 1, carried out under pressure of an indifferent gas in excess of atmospheric.

4. Process according to claim 1, carried out under pressure of a gas in excess of atmospheric and with the electrolyte at a temperature in excess of 1000° C.

5. Process of forming by electrolysis beryllium alloys, which comprises maintaining a fluid electrolyte containing a dissolved beryllium compound and metallic fluorides of higher dissociation potential in contact with anodic material, consisting of a fluid alloy containing beryllium and copper, and in contact with a fluid cathode layer containing a metal forming with beryllium an alloy fluid at the temperature of electrolysis, and electrolyzing said electrolyte in series with said anode and cathode.

6. Process according to claim 5, carried out under pressure of a gas in excess of atmospheric.

7. Process according to claim 5, carried out under pressure of an indifferent gas in excess of atmospheric.

8. Process according to claim 5, carried out under pressure of a gas in excess of atmospheric and with the electrolyte at a temperature in excess of 1000° C.

9. Process of forming by electrolysis beryllium alloys, which comprises maintaining a fluid electrolyte containing beryllium fluoride and metallic fluorides of higher dissociation potential in contact with anodic material, consisting of a fluid alloy containing beryllium and copper and in contact with a fluid cathode layer containing a metal forming with beryllium an alloy fluid at the temperature of electrolysis, electrolyzing said electrolyte in series with said anode and cathode, at least intermittently replenishing the metal other than beryllium in said cathode layer, and at least intermittently withdrawing a part of the alloy of beryllium with said metal contained in said cathode layer.

10. Process of forming by electrolysis material containing metallic beryllium, which comprises maintaining a fluid anode layer of an alloy containing copper and beryllium, maintaining a fluid electrolyte layer of lesser density floating thereon, said electrolyte layer containing beryllium fluoride and metallic fluorides of higher dissociation potential, maintaining a cathode in contact with the upper surface of said electrolyte, and electrolyzing said electrolyte in series with said anode and cathode, thereby depositing metallic beryllium at the cathode.

11. Process according to claim 10, carried out under pressure of a gas in excess of atmospheric.

12. Process according to claim 10, carried out under pressure of an indifferent gas in excess of atmospheric.

13. Process according to claim 10, carried out under pressure of a gas in excess of atmospheric and with the electrolyte at a temperature in excess of 1000° C.

14. Process of forming by electrolysis material containing metallic beryllium, which comprises maintaining a first layer of fluid alloy containing beryllium and copper, maintaining floating thereon a fluid electrolyte layer of lesser density, containing beryllium fluoride and metallic fluorides of higher dissociation potential, maintaining floating on said electrolyte layer a second layer of fluid metal of lesser density than said electrolyte layer, and electrolyzing said electrolyte in series with said first layer as anode and said second layer as cathode, thereby forming an alloy containing beryllium.

15. Process according to claim 14, carried out under pressure of a gas in excess of atmospheric.

16. Process according to claim 14, carried out under pressure of an indifferent gas in excess of atmospheric.

17. Process according to claim 14, carried out under pressure of a gas in excess of atmospheric and with the electrolyte at a temperature in excess of 1000° C.

18. Process of forming by electrolysis material containing metallic beryllium, which comprises maintaining a first layer of fluid alloy containing beryllium and copper, maintaining floating thereon a fluid electrolyte layer of lesser density, containing beryllium fluoride and metallic fluorides of higher dissociation potential, maintaining floating on said electrolyte layer a second layer of fluid metal of lesser density than said electrolyte layer, and electrolyzing said electrolyte in series with said first layer as anode and said second layer as cathode, thereby forming an alloy containing beryllium, at least intermittently replenishing the metal other than beryllium contained in said second layer, and at least intermittently withdrawing part of the alloy of beryllium with said metal contained in said second layer.

19. Process of forming by electrolysis material containing metallic beryllium, which comprises maintaining a first layer of fluid alloy containing beryllium and copper, maintaining floating thereon a fluid electrolyte layer of lesser density than said alloy containing beryllium fluoride and metallic fluorides of higher dissociation potential, maintaining floating on said electrolyte layer a second layer of fluid metal of lesser density than said electrolyte layer, said second layer consisting predominantly of aluminum and beryllium, and electrolyzing said electrolyte in series with said first layer as anode and said second layer as cathode.

20. Process according to claim 19, carried out under pressure of a gas in excess of atmospheric.

21. Process according to claim 19, carried out under pressure of an indifferent gas in excess of atmospheric.

22. Process according to claim 19, carried out under pressure of a gas in excess of atmospheric and with the electrolyte at a temperature in excess of 1000° C.

23. Process of forming by electrolysis material containing metallic beryllium, which comprises maintaining a first layer of fluid alloy containing beryllium and copper, maintaining floating thereon a fluid electrolyte layer of lesser density than said alloy containing beryllium fluoride and metallic fluorides of higher dissociation potential, maintaining floating on said electrolyte layer a second layer of fluid metal of lesser density than said electrolyte layer, said second layer consisting

predominantly of beryllium and aluminum, electrolyzing said electrolyte in series with said first layer as anode and said second layer as cathode, at least intermittently adding aluminum to said second layer and at least intermittently withdrawing part of the alloy of beryllium and aluminum contained in said second layer.

24. Process of forming by electrolysis material containing metallic beryllium, which comprises maintaining a layer of fluid alloy containing beryllium and copper, maintaining floating thereon a fluid electrolyte layer of lesser density than said alloy containing beryllium fluoride and metallic fluorides of higher dissociation potential, maintaining said electrolyte layer at a temperature at least equal to the melting point of beryllium metal, maintaining floating on said electrolyte layer a layer of beryllium metal in liquid phase, and electrolyzing said electrolyte in series with said alloy layer as anode and said beryllium layer as cathode while maintaining said anode, electrolyte and cathode under pressure of a gas in excess of the vapor pressure of beryllium metal at the temperature maintained.

25. Process according to claim 24, carried out under pressure of a gas indifferent to beryllium metal.

26. Process according to claim 24, carried out under pressure of a hydrogen gas.

27. Process according to claim 24, carried out under pressure of a gas in excess of one atmosphere gage.

28. Process according to claim 24, carried out under pressure of a gas in excess of two atmospheres gage.

29. Process of forming by electrolysis material containing metallic beryllium, which comprises maintaining a layer of fluid alloy containing beryllium and copper, maintaining floating thereon a fluid electrolyte layer of lesser density than said alloy containing beryllium fluoride and metallic fluorides of higher dissociation potential, maintaining said electrolyte layer at a temperature at least equal to the melting point of metallic beryllium, maintaining floating on said electrolyte layer a layer of metallic beryllium in liquid phase, electrolyzing said electrolyte in series with said alloy layer as anode and said beryllium layer as cathode while maintaining said alloy layer, electrolyte and cathode under pressure of a gas in excess of the vapor pressure of metallic beryllium at the temperature maintained, and at least intermittently withdrawing metallic beryllium in liquid phase from the said cathode layer.

30. Process according to claim 29, carried out under pressure of a gas indifferent to beryllium.

31. Process according to claim 29, carried out under pressure of a hydrogen gas.

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