Feb. 19, 1946. J. M. MERLE 2,395,286

PROCESSES FOR CHEMICALLY PURIFYING AND REFINING METALS

Filed Jan. 31, 1944

Fig. 3

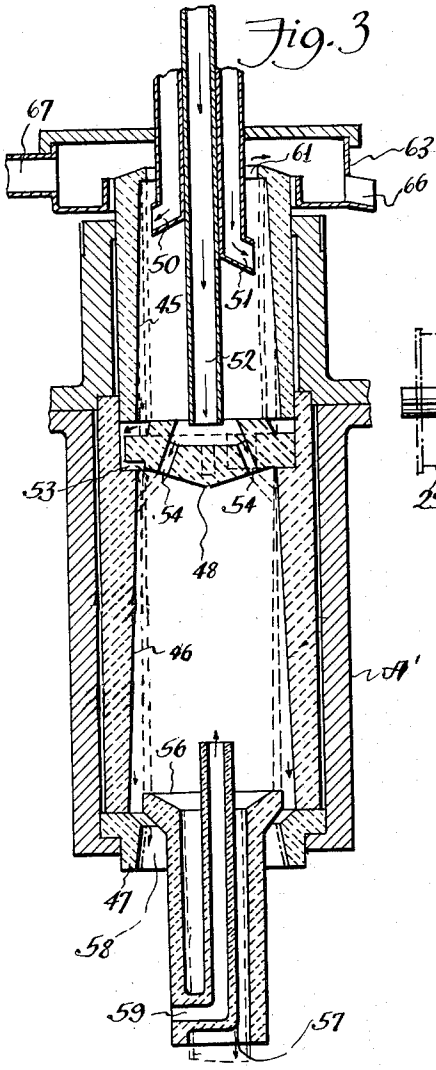


Fig. 1

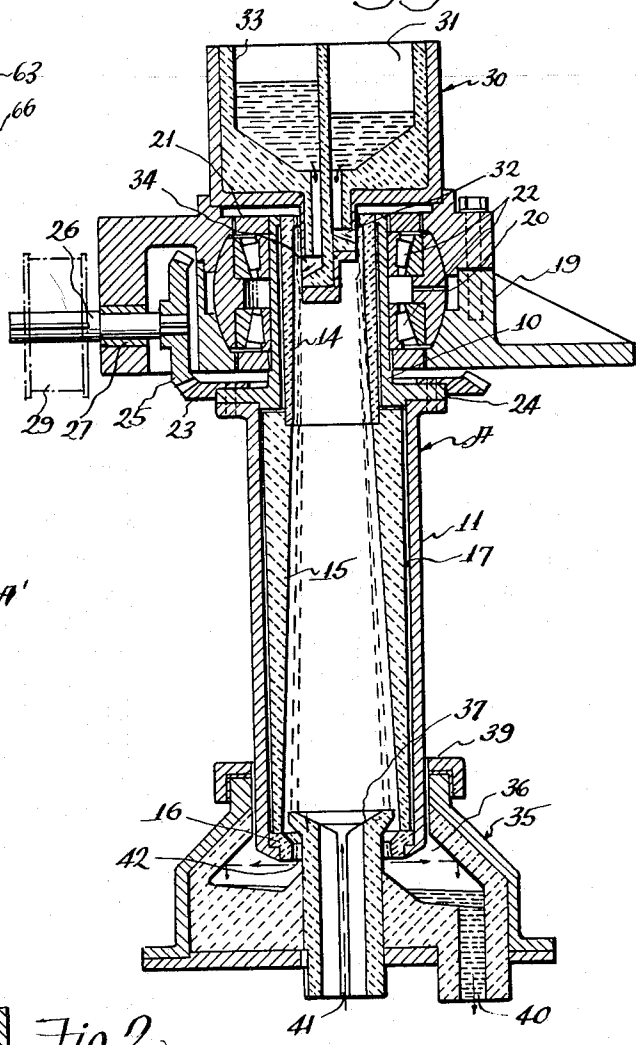
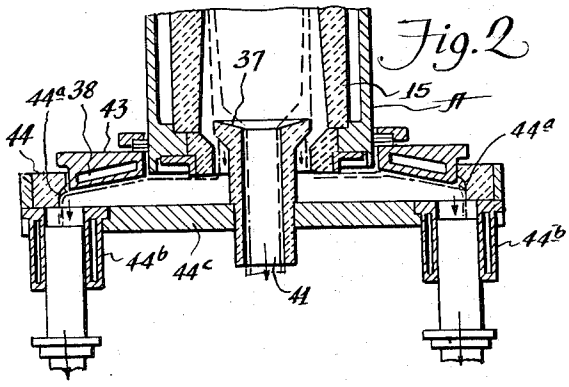


Fig. 2



INVENTOR
Joseph M. Merle
BY
Charles M. Lindroth
ATTORNEY

Patented Feb. 19, 1946 2,395,286

UNITED STATES PATENT OFFICE

2,395,286

PROCESSES FOR CHEMICALLY PURIFYING AND REFINING METALS

Joseph M. Merle, Chicago, Ill.

Original application July 19, 1941, Serial No. 403,124. Divided and this application January 31, 1944, Serial No. 520,444

9 Claims. (Cl. 75—27)

This invention relates to processes for chemically purifying and refining metals and other substances in fusion and is a division of application Serial No. 403,124, filed July 19, 1941.

The principal objects of the present invention are to provide a new and improved method of refining metal and other substances in a liquid state in a more facile, economical, and efficient manner than formerly, which consists in depositing a mixture of purified metal oxide and aluminum powder on a rapidly moving film forming member, igniting the powder mixture and forming the molten metal liberated at a temperature much higher than its melting point into a thin film by centrifugal action, and in continuously separating and collecting the pure metal and the products of reaction.

A more specific object of my invention is to provide a new and improved process for refining metal which consists in depositing a batch or continuous flow of a mixture of purified metal oxide powder and aluminum powder on a rapidly moving film forming member, igniting the powder mixture and forming the molten metal liberated into a film by and under centrifugal action of sufficient magnitude to dissociate the isotopic components into superimposed layers by centrifugal action, and in subjecting the separated metal isotope to the action of a molten or powdered material capable of forming a stable component with it.

Other objects of my invention will appear from time to time as the following specification proceeds and with reference to the accompanying drawing wherein:

Figure 1 is a vertical sectional view taken through a film forming refining machine constructed to effect the refining process of my invention;

Figure 2 is a detail fragmentary vertical sectional view showing a rotary disk atomizer at the discharge end of the refining machine, arranged to propel the metal coming from the refining machine into molds; and

Figure 3 is a partial fragmentary vertical sectional view taken through a modified form of apparatus for effecting the process of my invention.

Referring now in particular to Figure 1 of the drawing, one form of the device for carrying out the process of my invention includes a rotary working cylinder A rotatably driven at a high rate of speed and adapted to form the substance to be refined into a thin film on its inner cylindrical surface.

The rotary working cylinder A is shown as being constructed in two parts 10 and 11, and is preferably made of steel or alloy steel, machined all over and perfectly balanced for high speed rotation. In this metal working cylinder various refractory linings, tubular or of other shapes, can be assembled, such linings being indicated by reference characters 14, 15 and 16. These linings can be insulated from the metal cylinder by an insulating lining or by an air space indicated at 17. These linings, either made in the machine by ramming and compressed by centrifugal action, or made of assembled bricks, may have different compositions which may best be suited to withstand the action of the molten metals or molten slags coming in contact with their working surfaces, or to participate in the refining action by combining with some of the metalloids or impurities existing in the metal being treated. For example, lining 14 may be made of silica brick or rammed silica sand, while lining 15 may be made of magnesite brick or rammed sintered magnesite and dolomite, and lining 16 may be a brick of zirconium oxide, to prevent erosion.

The working cylinder A is supported in a machine frame 19 by a self-aligning cradle 20 having a convex bearing surface engaging said frame, to permit limited pivotal movement thereof about axes perpendicular to the axis of rotation of said cylinder. A flanged retaining member 21 and bearings 22, 22 are provided to mount said cylinder in said cradle. A bevel ring gear 23 is bolted to the upper side of a flanged lower portion 24 of the upper cylindrical portion 10, and is meshed with a bevel gear 25 on a shaft 26. Said shaft is mounted in a bearing 27 in the frame 19. A pulley 29 is shown by broken lines as being keyed on said shaft, for driving said shaft. Said pulley may be driven from a suitable motor (not shown) by means of a belt either directly from said motor or through a geared reduction drive or a usual form of variable speed drive. While a pulley is herein shown as being provided to drive said shaft, it should be readily apparent to those skilled in the art that various other well known forms of drive means may be provided for driving said shaft, if desired. Since the speed of rotation of said cylinder must be sufficient to hold the molten metals and substances treated in films or layers against the inside of the cylinder, it should be understood that the axis of rotation of said cylinder may be horizontal or at various angles between the horizontal and vertical, as well as the vertical, if desired.

The means for pouring the metal and reagents

into the inside of said cylinder includes a chamber 30 divided into a pouring box 31 having a nozzle 32 depending within said cylinder, and a pouring box 33 having a depending nozzle 34 extending within said cylinder a greater distance than the nozzle 32. The discharge ends of the nozzles 32 and 34 are inclined downwardly and outwardly towards the inner periphery of the lining 14, to direct a regulated amount of molten metal or slag, or other powdered material employed in the process, against the inner wall of said working cylinder at a constant rate of speed and at selected levels, so each material will be immediately picked up and formed into a film by said cylinder as it is delivered from its respective nozzle.

The delivery end of said cylinder freely rotates within a collector 35 which includes two receptacles, one of which receptacles 36 serves to receive the refined metal and the other of which receptacles serves as a skimmer 37, for the slag.

The receptacle 36 is preferably of a circular form and as herein shown is sealed from contact with the air by means of a seal indicated generally by reference character 39 and disposed between the top of the collector 35 and the outer wall of the cylinder A. Said seal may be of any well known construction, such as asbestos composition, or mercury, or may be constructed from any other suitable form of sealing means.

The receptacle 36 may have an inclined bottom for quick aggregation and drainage of the continuously expelled molten metal, through an opening 40 which conducts the metal to a ladle or mold. Said receptacle may also have a number of radial openings (not shown), leading to as many molds. The slag is drained through the stationary circular skimmer 37, which collects the separated slag so it will run down through an opening 41 by gravity. The refined molten metal enters the receptacle 36 through an annular opening 42 formed between the inner periphery of the refractory lining 16 and the outer periphery of the skimmer 37.

The circular skimmer 37 may be made of any suitable refractory material capable of withstanding the action of the slag formed.

Figure 2 shows a water-cooled rotary disk atomizer 43 attached to the lower end of the working cylinder A. The lower surface of the disk of this atomizer is adapted to receive and spread the metal coming from the outlet 42, by centrifugal action, and is herein shown as being of a substantially frusto-conical formation, provided with passageways 38 therein for the circulation of a cooling medium, such as water. The purpose of said atomizer is to supercool the metal and free it from all contained gases before it is distributed into molds. In this figure the outer periphery of the disk 43 is encased by an annular refractory ring 44 having slots 44*a*, 44*a* formed in the inner periphery thereof. The inner periphery of said refractory ring forms a baffle, and the metal propelled to the inner periphery of said baffle by centrifugal action is forced down the slots 44*a*, 44*a* leading to the center of water-cooled continuous molds 44*b*, 44*b* for billets or slabs. As previously described, the refined metal forms a liquid air-seal around the center of the skimmer 37, leading the slag out. The molds 44*b*, 44*b* may be mounted in a plate 44*c* abutting the bottom of the baffle 44, to effect cooling and molding of the metal in a condition sealed from the open air.

In Figure 3 a rotary working cylinder A' is provided, which is like that shown in Figure 1. This cylinder is provided with refractory linings 45, 46 and 47. Said linings may be insulated from the metal working cylinder A' by an insulating lining or by an air space, as in the device illustrated in Figure 1. These linings may be made from the same materials as the linings 14, 15 and 16 shown in Figure 1. A refractory brick partition 48 is mounted adjacent the upper end of the lining 46 in a recessed shouldered portion thereof, just beneath the lower end of the lining 45.

The supporting means for the working cylinder A and the drive mechanism therefor may be the same as that shown for driving the working cylinder A shown in Figure 1, so will not herein be shown or described in connection with Figure 2.

A pouring chamber (not shown) divided into a plurality of pouring boxes similar to that shown in Figure 1 may be provided. A nozzle 50 may depend from one pouring box and extend within the refractory lining 45 of the cylinder A'. Another nozzle 51 may depend from another pouring box, as shown in Figure 1, and extend within said cylinder a greater distance than the nozzle 50. A third nozzle 52, leading from a third pouring box (not shown), may extend within the cylinder A' between the nozzles 50 and 51. Said nozzle 52 opens directly into the center of the refractory brick lining 48 and extends a substantial distance beneath the nozzles 50 and 51. The discharge ends of the nozzles 50 and 51 are inclined downwardly and outwardly towards the inner periphery of the lining 45, like the nozzles 32 and 34 in Figure 1.

The refractory brick partition 48 has peripheral slots 53, 53, which with the inside of the refractory lining 46, form passageways for the downward flow of the metal. These slots are so placed in relation to the internal diameter of the lining 46 that the levels of metal and slag in the lining 45 will be kept constant while the refined metal produced will flow through said passageways at a constant speed. Another series of slots 54, 54 is provided in said refractory brick partition to permit molten material supplied through the tube 52 to be impinged on the inside of the refractory lining 46, or on the film of molten metal flowing down said lining.

The delivery end of the cylinder A' rotates freely within a collector (not shown). Said collector may be like that shown in Figure 1, and may include a receptacle for the refined metal and a central receptacle 56, which serves as a skimmer for the slag. Said collector may also include a rotary disk atomizer like that shown in Figure 2. Said central receptacle or skimmer is stationary and serves to collect the separated slag so it will run down through an opening 57 of said slag receptacle by gravity. The refined molten metal is delivered through an annular opening 58 between the inner periphery of the lining 47 and the outer periphery of the skimmer 56. Said slag receptacle or skimmer is shown as having an integrally formed pipe 59 extending through its center, to permit a gas or oil flame to preheat the refractory lining before operation of the device, to keep the treated metal at the proper temperature during the refining operation, if necessary. Said slag receptacle may be made of any suitable refractory material capable of withstanding the action of the slag formed.

The molten slag may also be delivered from

the feeding end of the working cylinder A through an opening 61, which throws the slag into a special slag box 63, provided with a drain 66 and a stack pipe 67, for free elimination of the gases.

Referring now to several refining operations which form the subject matter of my invention, the general principle of refining has been clearly described in my parent application Serial No. 403,124, now Patent No. 2,355,885, so need not herein be described in detail. A mixture of metal oxide and aluminum powder may be supplied to the inside of the refractory brick lining 14 of the working cylinder A, through the nozzle or tube 34 and ignited while said working cylinder is rotating as by initially supplying magnesium powder, to act as a priming agent. The magnesium powder may be supplied separately through the nozzle 32 if desired. The chemical reaction taking place between the metal oxide and the aluminum powder creates the heat which produces the molten metal and the molten slag and is continuous and self-propagating to the incoming stream of powdered mixture. Ignition may also be accomplished by means of a blow torch or flame applied through the opening 32, or may be accomplished by a flame introduced through the pipe 59 when using the apparatus shown in Figure 3. A continuous supply of this mixture may be kept supplied through this nozzle, or if the mixture is too unstable, part of it such as the metal oxide can be supplied in the proper proportions through the nozzle 34 and the remainder such as the aluminum or magnesium or other powder through the nozzle 32. Under these conditions the reaction can be continuous, and as long as it continues, the molten metal can be reduced from the oxide if a single metallic oxide or ore has been used. Where several metallic oxides or ores have been introduced through the nozzle 34 in predetermined proportions, a film or layer will be formed against the lining 14 and will flow down said lining. The film formed can then be subjected to the action of a selected molten slag supplied through the nozzle 32. This slag is formed into a thin film moving downwardly inside of the slightly conical refractory lining 14. The molten metal being slightly under the discharge level of the slag will be formed into a thin film moving downwardly over the film of slag.

On account of the lighter specific gravity of the molten slag, and the fact that the slag is introduced into the working cylinder above the molten metal, this slag will first contact the film of molten metal on the surface of the metal closest to the inner periphery of the refractory 14 and then will gradually be forced mechanically through the metal under the centrifugal action exerted, thus submitting every particle of the metal film treated to the action of the refining slag. On its further downwards motion, this slag containing all the sulphur and phosphorus in the form of sulphides and phosphates, as well as all impurities and inclusions contained, will be completely separated from the treated metal, and the refined and centrifugally cleaned metal will continuously flow away through the openings 42 and 40 to a ladle or mold, while the used slag will be continuously drained through the skimmer sleeve 41 and received in a container, for further use or sale.

By proper disposition of the conicity of lining 15 with the outside diameter of skimmer 41 and the diameter of circular outlet 42, clean metal only is continuously expelled into collector 35 and a fixed quantity of molten metal is constantly held inside the refining machine in the form of a slightly conical film or layer whose weight is insignificant in relation to the total weight of the working rotary cylinder. This material being a liquid is self-leveling and has a balancing effect on the rotation of the working cylinder.

In the machine shown in Figure 3 a mixture of metal oxide and aluminum powder may be supplied through the tube 52 inside of the refractory brick lining or partition 48 and ignited while the machine is rotating. A continuous supply of this mixture may be kept supplied through the nozzle 50, or if the mixture is too unstable, part of it, such as the metal oxide, can be supplied in the proper proportions through the nozzle 50, and the remainder, such as aluminum or magnesium or other powder, through the nozzle 51. Several metallic oxides or ores may be introduced in predetermined proportions, causing a film or layer to be formed against the lining 45. This film will flow through the slots 53 of the refractory 48, after the controlled level has been established, downwards into the lining 46. The film formed in this last mentioned lining can then be subjected to the action of a selected molten slag supplied through passageways 54, 54 of refractory 48. The balance of the operation would then be exactly as previously described in connection with the operation of the machine shown in Figure 1.

While these procedures may be applied to the refining of iron or steel, they are generally applied to the reduction of high melting point metals, such as, tungsten, chrome, tantalum, titanium, molybdenum, vanadium, platinum, uranium, etc. It enables these metals to be obtained individually in the molten state and enables them to be refined and cast directly into appropriate molds. It also enables them to be supercooled by the rotary disk atomizer shown in Figure 2, and to be impelled into the molds in a supercooled condition, as has been previously described. By a mixing of the metallic ores or oxides used, it enables exact alloys or mixtures between these high melting point metals to be obtained in a refined condition and to be directly cast in appropriate molds.

On account of the fact that the chemical reaction used in refining high melting point metals, is highly exothermic, these metals are obtained or liberated from their oxides in their liquid condition at temperatures much higher than their melting points. Since these metals are subjected to centrifugal force upon their liberation, when their atomic cohesive forces are reduced under the high temperature generated, it is possible by properly graduating the centrifugal action, to separate pure metals into their isotopic components, if any, by multiplying the effect of their difference in atomic weight. For instance, by this procedure it becomes possible to reduce uranium ore or oxide and to obtain pure uranium, and also to increase the speed of rotation of the chamber A up to the point where the lighter isotope U–235 will be separated from the heavier component U–238 at the high temperature of the metal liberated, and under the centrifugal force exerted, and this lighter isotope will be formed into a film inside of the film of heavier uranium, where it can be combined with an adequate metalloid, such as, silicum, calcium, sodium, etc., capable of

forming a stable component with it, which can be collected and handled. In this case this component would be made to flow through skimmer 56 to be collected with and under the refining slag, while the heavier uranium isotope would be expelled into the collector through opening 81.

While I have herein shown and described one form in which my invention may be embodied, it will be understood that the construction thereof and the arrangement of the various parts may be altered without departing from the spirit and scope thereof. Furthermore, I do not wish to be construed as limiting my invention to the specific embodiment illustrated, excepting as it may be limited in the appended claims.

I claim as my invention:

1. A process for refining metals which consists in depositing a batch of a mixture of powdered metal oxide of the type that can be reduced by aluminum, and aluminum powder on a rapidly moving film forming surface, in igniting the powdered mixture and forming the liberated molten metal into a thin film by centrifugal action, and in separating and collecting the pure metal from the products of reaction.

2. A process for refining metals which consists in continuously depositing a mixture of powdered metal oxide of the type that can be reduced by aluminum, and aluminum powder on a rapidly moving film forming surface, in continuously igniting the powdered mixture, in continuously forming the liberated molten metal into a thin film by centrifugal action, and in continuously separating and collecting the pure metal and the products of reaction by centrifugal action.

3. A process for refining metals which consists in continuously depositing a purified metal oxide powder of the type that can be reduced by aluminum, on a rapidly moving film forming surface, in separately depositing a metallic aluminum powder on said film forming surface, in continuously igniting the mixture, in continuously forming the liberated molten metal into a thin film by centrifugal action, and in separating the products of reaction by centrifugal action and then casting the pure metal into a mold.

4. A method of making metallic alloys or mixtures of predetermined composition which consists in depositing a mixture of purified metal oxide of the type that can be reduced by aluminum, and aluminum powder on a rapidly moving film forming surface, in igniting the powder mixture and forming the liberated molten metal alloy into a thin film by centrifugal action, and in separating the molten metal alloy from the products of reaction by centrifugal action and then casting the molten metal alloy into a mold.

5. A process for refining metals which consists in continuously depositing a mixture of purified metal oxide of the type that can be reduced by aluminum, and aluminum powder on a rapidly moving film forming surface, in continuously igniting the powdered mixture, in continuously forming the liberated molten metal into a thin film by centrifugal action, in continuously separating the pure metal from the products of reaction, and in subsequently forcing a film of molten refining slag moving at a high velocity, in contact with the film of molten metal by centrifugal action, and in separating and collecting the products of reaction from the pure metal by centrifugal action.

6. A process for refining metals which consists in depositing a batch of a mixture of purified metal oxide powder of the type that can be reduced by aluminum, and aluminum powder on a rapidly moving film forming surface, igniting the powder mixture and forming the molten metal liberated at a temperature much higher than its melting point into a thin film by centrifugal action of sufficient magnitude to dissociate the isotopic components of the molten metal, in separating the isotopic components of the molten metal into superimposed layers by centrifugal action, and in separately collecting the isotopic components.

7. A process in accordance with claim 6 wherein the separated metal isotope layer is subjected to the action of a molten or powdered material capable of forming a stable component with it.

8. A process in accordance with claim 6 wherein the liberated molten metal is uranium and the lighter isotope separated is uranium 235.

9. A process in accordance with claim 6 wherein the separated metal isotope layer is subjected to the action of a molten or powdered material capable of forming a stable component with it, and wherein the liberated molten metal is uranium and the lighter isotope separated is uranium 235.

JOSEPH M. MERLE.