Nov. 10, 1959 J. B. BRENNAN 2,912,321

CONTINUOUS CASTING AND REFINING OF MATERIAL

Filed Sept. 4, 1956 3 Sheets-Sheet 1

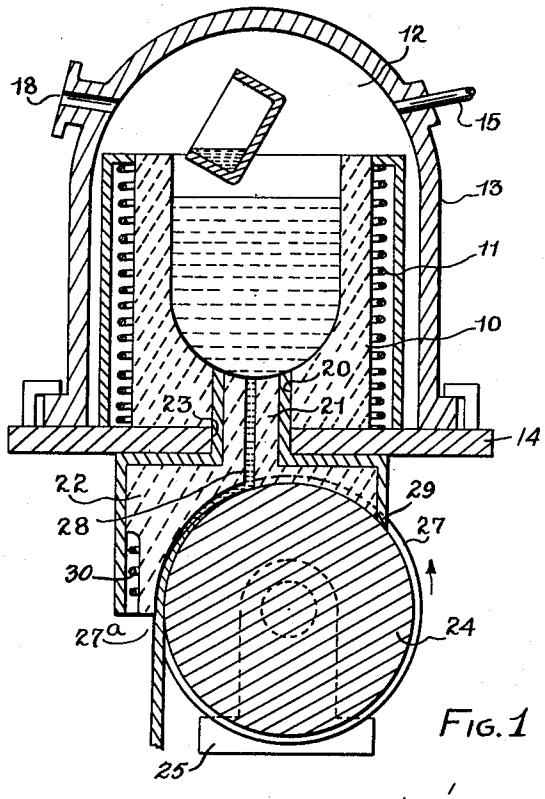


FIG. 1

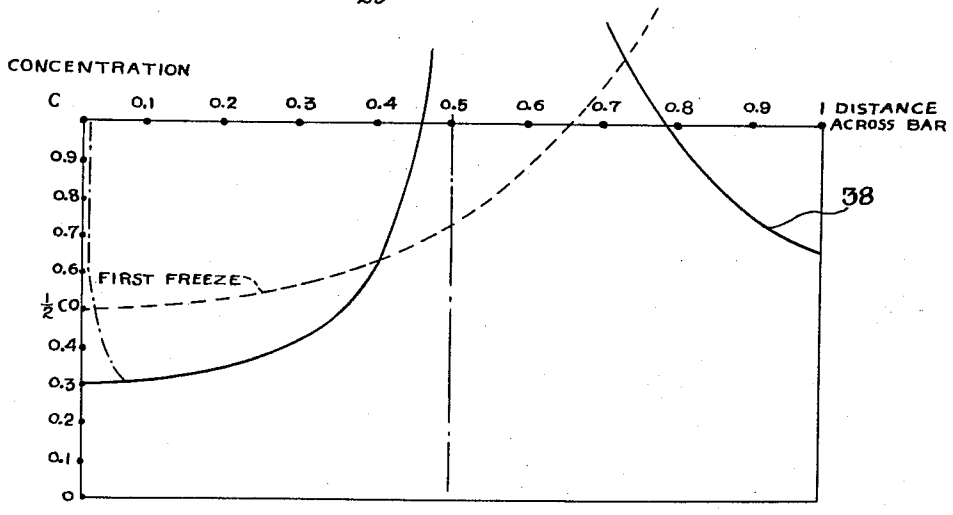


FIG. 6

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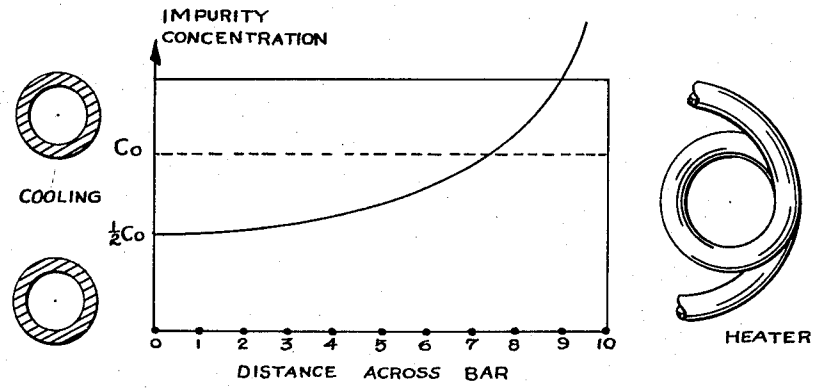


FIG. 2

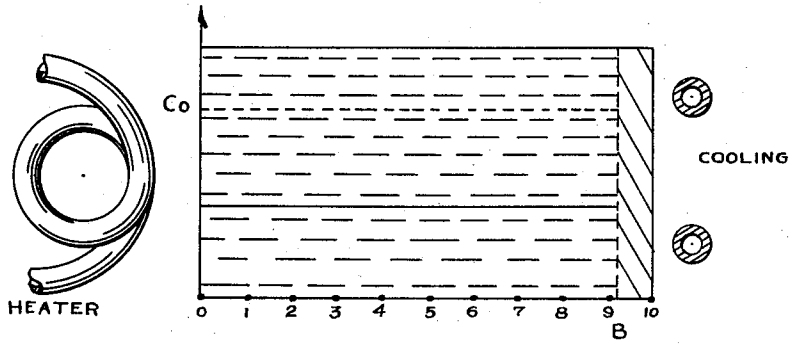


FIG. 3

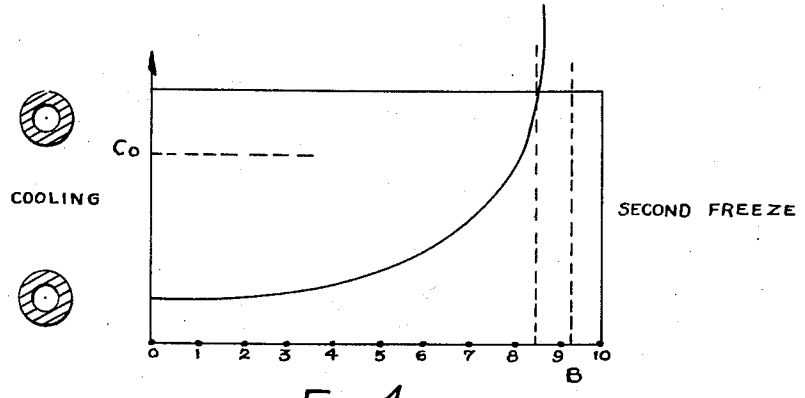


FIG. 4

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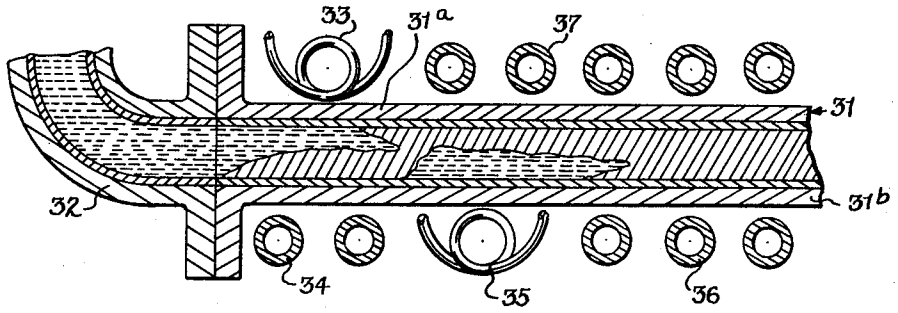


FIG. 5

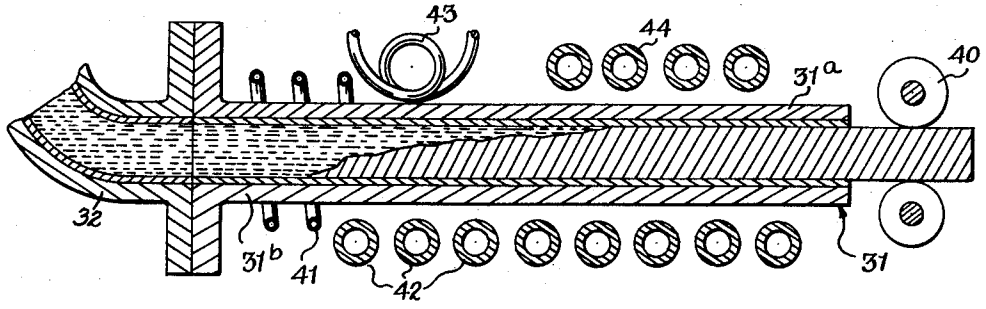


FIG. 7

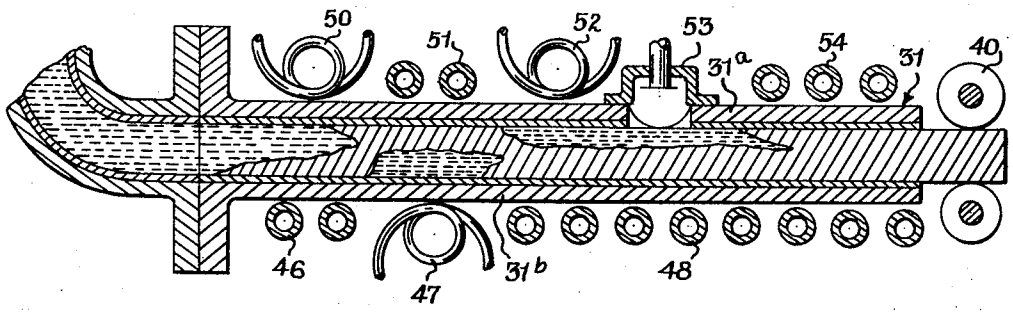


FIG. 8

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2,912,321

CONTINUOUS CASTING AND REFINING OF MATERIAL

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Application September 4, 1956, Serial No. 607,640

6 Claims. (Cl. 75—63)

The present invention relates to the separation of metals and, more particularly, to a method of an apparatus for purifying a material, particularly a metal, having a different melting point impurity therein. The present invention contains subject matter common with that of my application Serial No. 343,563, filed March 20, 1953, and issued on February 26, 1957 as Patent No. 2,782,473.

The principal object of the present invention is to provide a new and improved method of reducing the impurity content of a material, in which method a solid body is formed from a melt of the material by controlled freezing with the body having a higher concentration of the impurity in one zone of the body.

Another object of the present invention is to provide a method of separating the components of different melting points of a body of material, which components are combined as a solid or molten mixture, in which method the body, if solid, is rendered molten, and the molten body progressively solidified or frozen from one side or portion of the body so that the lower melting point component tends to remain in the molten or liquid phase during the solidification, while the higher melting point component tends to solidify first and to concentrate in that portion of the body which is first solidified.

A further object of the present invention is to provide a new and improved apparatus for concentrating metals of different melting points present in a melt, in which method the melt is progressively solidified or frogen from one side thereof and remelted and again solidified progressively from one side thereof to produce a solid body in which the particular metals are concentrated in different zones or portions of the solid body.

A still further object of the present invention is to provide a new and improved method of purifying a metal containing as an impurity another metal of a different melting point, in which method the metal to be purified is melted and is then progressively solidified or frozen from one portion or side of the body to cause the impurity to concentrate in one zone or portion, and in which a higher purification of the metal may be obtained by additionally melting and refreezing the portion of the frozen body which contains the highest concentration of the purified metal.

Further objects and advantages of the present invention will be apparent from the following detailed description of the preferred embodiment thereof made with reference to the accompanying drawing forming a part of this specification and in which:

Fig. 1 is a vertical sectional view of an apparatus which may be used to continuously cast metal being purified in accordance with the present invention;

Fig. 2 is a graph showing the distribution of an impurity in a bar cast by the apparatus of Fig. 1;

Fig. 3 is a graph showing the depth of heating for additionally purifying the bar of Fig. 2;

Fig. 4 is a graph showing the impurity distribution after the heating schematically illustrated in Fig. 3;

Fig. 5 is a sectional view of a portion of an apparatus for first solidifying a melt from one portion thereof toward another portion thereof and subsequently remelting and resolidifying the first-mentioned portion;

Fig. 6 is a graph showing the impurity distribution in a metal having a higher impurity melting point and which has been treated in the apparatus of Fig. 5;

Fig. 7 is a sectional view through a part of a continuous casting apparatus for progressively solidifying a melt from one side thereof; and

Fig. 8 is a sectional view through a part of a continuous casting apparatus which is similar to Fig. 7, but which additionally includes means for alternately remelting and resolidifying the sides of the initially solidified melt to increase the concentration of the different components of the material.

The present invention contemplates the provision of a new and improved method of concentrating and separating components of a melt having different melting points in which the melt is solidified or frozen under controlled conditions to provide a body having the components to be separated concentrated in different zones or portions of the body.

If a melt which contains components of different melting points is gradually cooled proceeding from one side of the melt, the component having the highest melting point will tend to solidify first and the component having the lower melting point will tend to remain in the liquid phase. As a result, if the melt is cooled progressively from one side, the portion of the frozen body adjacent one side will have a high concentration of the higher melting point component, while the portion of the body adjacent the opposite side of the melt will have a high concentration of the lower melting point component. If further concentration is desired, say of the highest melting point component, it is obtainable by remelting the portion of the frozen body in which the highest melting point component is concentrated without melting the other portion of the body and then once again solidifying the melted body in the manner described to form a second zone of concentration for the low melting point component adjacent the first zone formed by the first freezing. After the melting and freezing process has been completed, the final bar is cut to remove the area of high impurity concentration so as to leave a high purity casting.

The present invention is applicable for separating and purifying various materials but is particularly suitable for purifying metals. As a specific example, copper may contain aluminum as an impurity and the following description is made with reference to increasing the purity of copper by eliminating at least a portion of the aluminum impurity. It is to be understood, however, that the process is equally applicable to other metals and materials having different melting point components and the material to be separated may have a higher or lower melting point than the other metal or metals.

Fig. 1 of the drawing illustrates one type of apparatus which may be used to carry out the method of the present invention. Referring to Fig. 1, a crucible **10** is provided in which the melt of a material to be treated, in this case aluminum having a a copper impurity, is poured. The crucible **10** is surrounded by an induction heating coil **11** and is located within a chamber **12** formed by a removable dome-shaped housing **13** positioned on a support plate **14**. The dome-shaped housing **13** has a conduit **15** leading into the chamber **12** through which the chamber **12** may be evacuated and through which an inert atmosphere, such as helium, may be introduced into the chamber. A sight glass **18** is also provided to enable the operator to view the interior of the chamber **12**.

The crucible **10** has a vertical opening **20** through the

bottom thereof which communicates with the interior of the crucible and in which a cylindrical portion 21 of a block 22 is seated. The cylindrical portion 21 extends through an opening 23 in the plate 14, and the main body portion of the block 22 is located below the plate 14. The lower portion of the block 22 is curved to fit closely adjacent to a wheel 24 which is rotatably supported adjacent the lower side of the block by a pedestal 25. The wheel 24 has a circumferentially extending groove 27 therein which forms with the block 22 a die cavity 27*a* that communicates with a vertical passage 28 extending upwardly through the cylindrical portion 21 and opening into the interior of the crucible 10. The molten material flows from the crucible 10 through the passage 28 into the die cavity formed by the groove 27 as the wheel is rotated. The block 22 includes a dam portion 29 which projects into the groove 27 adjacent the passage 28 to prevent the melt from flowing opposite the direction of rotation of the wheel. The melt in the die cavity first cools adjacent the surface of the wheel and then progressively solidifies proceeding outwardly in a radial direction. In order to assure that the metal solidifies from the side adjacent the wheel, an induction heating coil 30 is positioned adjacent the exit end of the groove 27 to keep the portion of the material being solidified, which is remote from the wheel 24, in a liquid phase until just before it leaves the die cavity.

If the concentration of aluminum in the copper in the crucible is designated by C_0, the frozen bar or material which comes off the wheel 24 will have an impurity distribution as shown in the graph of Fig. 2 with the side of the bar, which was adjacent the wheel, being of the highest purity copper and with the aluminum being concentrated adjacent the opposite side. In the graph, the horizontal coordinant indicates unit distances across the bar. The graph shown is for a copper-aluminum melt having a segregation constant k of 0.5 where the segregation constant is defined as the ratio of the concentration of impurity in the solid to the concentration in liquid at a given point during the freeze. Where k is greater than 1, the impurity concentration will be less in the liquid than in the solid, and where k is less than 1, the impurity concentration is less in the solid than in the liquid.

It can be shown that the concentration of impurity in the material freezing at any given point is given by:

$$\frac{di}{dv'}=\frac{ki}{v'} \tag{1}$$

where i is the total amount of impurity remaining in the material still molten and v' is the volume still molten.

Solving this differential equation and evaluating the constant, we obtain:

$$C=kC_0v^{k-1} \tag{2}$$

where C_0 is the initial concentration of the fluid metal as it leaves the crucible;

k is the segregation constant of the material;

C is the concentration of impurity at any point in the metal; and

v is the volume of metal which already solidified before the metal at the chosen point.

It can therefore be seen, from the above, that the average impurity concentration in the lower half after processing is:

$$C_L=2(1-\tfrac{1}{2}k) \tag{3}$$

and in the upper half:

$$C_u=2^1-k \tag{4}$$

The impurity distribution for the lower half can be found by substituting C_L for C_0 in Equation 2 (for a volume of 0 to 0.5). Assuming a segregation constant of 0.5, the average copper concentration in the part of the bar first frozen will be 141% greater than the average concentration of the initial melt.

As is shown in Fig. 4, if it is desired to increase the purity of the bar formed by the first freezing, the highest purity portion of the bar can be again melted and in the case of the material represented by the graphs in Figs. 3 and 4, the depth of melting should be approximately to the point B on the graph of Fig. 3. The zone to the right of the point B is the high impurity zone and, therefore, is left in a solid state. If the bar is then again progressively cooled from the left-hand or high purity side, as indicated in Fig. 4, the final purity distribution will be similar to that shown by the curve of Fig. 4, and a second high impurity zone will be formed immediately adjacent to the first high purity zone. Therefore, by remelting and resolidifying the high purity zone, the purity thereof can be increased to the desired point.

After a zone of desired purity is produced, the remainder of the bar can be cut away leaving the high purity material.

In the case of low melting point impurities which tend to remain in the liquid phase, the heating coil 30 may keep the impurity from solidifying so that it may be drained off by a suitable drain, not shown, adjacent the lower end of the block 22. This method eliminates the need for later separating out the impurity.

Fig. 5 illustrates an alternative apparatus which may be used to concentrate the impurity to one side of a bar. Molten material, in this case aluminum with a copper impurity from a crucible, not shown, is introduced into one end of a die 31 through a conduit 32. An induction coil 33 is positioned adjacent one side 31*a* of the die 31 at the entrance end thereof and cooling coils 34 are positioned adjacent the opposite side 31*b* of the die 31 in opposition to the coil 33. The cooling coils 34 cause the material adjacent the cooling coils to solidify first, while the heating coil 33 maintains the material adjacent the side 31*a* of the die in a liquid phase. The impurity concentration of the metal between the cooling coils 34 and the heating coil 33 will be as illustrated in the graph of Fig. 6 by the dotted line.

Immediately adjacent the cooling coil 34, and toward the exit side of the die, is a heating coil 35, and adjacent to the heating coil 35 and at the exit end of the die are additional cooling coils 36. On the side 31*a* of the die, cooling coils 37 are provided between the heating coil 33 and the exit end of the die. When the bar moves from a position between the heating coil 33 and the cooling coils 34 to a position between the heating coil 35 and the opposed cooling coils 37, the portion of the bar adjacent the side 31*b* will be melted and the portion adjacent the cooling coil 37 will be solidified. The solidifying of the metal adjacent to the side 31*a* will be progressively inwardly from the side 31*a* and will cause an additional concentration of the impurities to occur near the center of the bar, as is shown by the curve 38 in Fig. 6. As the liquid material adjacent side 31*b* moves opposite the cooling coils 37, it will solidify beginning at the side 31*b* and moving inwardly to cause a further concentration of impurities at the center of the bar. The final impurity distribution is shown by the solid line curves in Fig. 6.

A die setup similar to that just described may be utilized for providing a single freeze for purifying the bar in a continuous casting method. Such a setup is shown in Fig. 7 and, referring to this figure, exit rolls 40 are provided to draw the material from the die 31. The entrance of the die 31 is connected to a crucible, such as the crucible 10 shown in Fig. 1, by the conduit 32, and a heating coil 41 is positioned about the conduit at its entrance end to assure that the melt moves into the die in a liquid phase. Next to the heating coil 41, and adjacent to the side 31*b* of the die, are positioned cooling coils 42, while a heating coil 43 and cooling coils 44 are positioned adjacent the side 31*a* of the die proceeding from the heating coil 41 to the exit end of the die in the order given. As is shown in the drawing, the melt begins to freeze adjacent the side 31*b* and progressively solidifies

from the side 31*b* toward the side 31*a* as the bar moves past the heating coil 43. The impurity distribution in the bar will be substantially the same as that shown in Fig. 2.

Fig. 8 illustrates a device where one side of the melt is alternately solidified, melted and solidified, while the other side is respectively maintained in a melted condition, solidified, melted, and again solidified so that the impurity concentration, assuming that the impurity has a higher melting point than the metal of the final material, as in the case with copper having an aluminum impurity, is highest in the central portion of the bar. The apparatus shown comprises the die 31 which is connected to a crucible by the conduit 32 and which has adjacent its side 31*b*, proceeding from the entrance end to the exit end, cooling coils 46, a heating coil 47, and cooling coils 48. Adjacent the side 31*a*, a heating coil 50 is positioned opposite the cooling coils 46, cooling coils 51 opposite the heating coil 47, and a heating coil 52 and an evacuation chamber 53 and cooling coils 54 are positioned opposite the cooling coils 48 in the order named proceeding from the cooling coils 51 to the exit end of the die.

When the material is between the cooling coil 46 and the heating coil 50, the material adjacent side 31*b* is solidified, while that adjacent side 31*a* is maintained in a liquid phase, and when the material moves to a position between the heating coil 47 and the cooling coil 51, the material which had remained molten is solidified, while the previously-solidified material is again melted. When the material moves from between the cooling coil 51 and the heating coil 47, the melted portion of the material which was adjacent the coil 47 is solidified, and the solid portion which was adjacent the coils 51 is melted as it passes the heating coil 52 and then solidified as it passes by the cooling coils 54. In all cases, the solidification of the material takes place inwardly from the adjacent side. The melted material between the heating coil 52 and the cooling coil 54 is preferably subjected to a vacuum which is drawn in the vacuum chamber 53 communicating with the interior of the die. The melted material opposite the vacuum chamber 53 is not drawn into the vacuum chamber since the melted portion adjacent the portion is surrounded by solid material. It will also be noted that a solid material in the die 31 extends from the entrance end continuously to the exit end enabling the bar, including the melted portions thereof, to be drawn through the die by the rolls 40.

It can now be seen that the present invention provides a new and improved method of, and apparatus for, concentrating different melting components of a material in various zones of the material or body formed by melting and solidifying the material and that the process is particularly useful for eliminating an impurity having a higher melting point and also concentrating an impurity of a material in a particular portion of a body of the material so that this portion may be eliminated and a high purity material produced.

While the present invention has been described as concentrating an impurity having a higher melting point than the material in which it is present as an impurity, it will be well understood by those skilled in the art that the method is equally applicable to separating impurities having a lower melting point. In this latter case, the impurity will solidify first and will be concentrated in that part or zone of the solidified body which is first frozen. It will be also understood that if a metal has present therein as impurities a first impurity metal of lower melting point and a second impurity metal of high melting point, the material is melted and progressively solidified, beginning with one side thereof, the resultant frozen body will have a higher concentration of the high melting point metal adjacent the side first frozen and a zone adjacent the side last frozen which has a high concentration of the low melting point metal. A high purity form of the desired metal is then obtainable by eliminating the side portions containing the high concentrations of the first and second impurity metals.

It can now be seen that the objects heretofore illustrated and others have been accomplished and that the present invention provides a new and improved method of an apparatus for concentrating the various components of a mixture in certain zones of a solidified body to enable these components to be eliminated.

While the preferred embodiment of the present invention and other alternatives have been described in considerable detail, it is to be understood that it is hereby my intention to cover all modifications, constructions and arrangements which fall within the ability of those skilled in the art and the spirit and scope of the appended claims.

Having thus described my invention, I claim:

1. The method of continuously casting material comprised of components having different melting points, which method comprises providing a mold cavity having a charging point and a delivery point and walls extending lengthwise from the charging point to the delivery point, continuously supplying a melt of said material to said cavity at said charging point and moving said material from said charging point to said delivery point, freezing said melt from one side thereof disposed adjacent one of said walls progressively inwardly toward the opposite side of the melt as the material is moved toward the delivery point and discharging the material at the delivery point in the form of a continuously frozen strip having zones of concentration of different melting point components extending lengthwise of the strip.

2. The method of continuously casting material comprised of components having different melting points, which method comprises providing a mold cavity having a charging point and a delivery point and walls extending lengthwise from the charging point to the delivery point, continuously supplying a melt of said material to said cavity at said charging point and continuously moving said material from said charging point to said delivery point, applying heat to one side of the melt in the cavity for a portion of the length of the mold cavity adjacent the charging point thereof and cooling the material adjacent the wall on the opposite side of said one side to progressively freeze the melt from said wall inwardly toward said one side to form a frozen strip having zones of concentration of different melting point components extending lengthwise of the strip.

3. The method as defined in claim 2 and which further comprises the step of separating a certain zone from the frozen strip.

4. The method of continuously casting material comprised of components having different melting points, which method comprises providing a mold cavity having a charging point and a delivery point and walls extending lengthwise from the charging point to the delivery point, continuously supplying a melt of said material to said cavity at said charging point and moving said material from said charging point to said delivery point, applying heat to one lengthwise side of the melt in the cavity for a portion of the length of the mold cavity adjacent the charging point thereof and simultaneously cooling the opposite side of the material in the cavity to progressively freeze the melt from said opposite side inwardly toward said one side to form a frozen strip having zones of concentration of different melt point components extending lengthwise of the strip and again heating the frozen strip from one side thereof to melt the strip to a depth of one of said zones and subsequently cooling the remelted material progressively inwardly from its outer side.

5. The method of purifying a first metal having a second metal of a different melting point present therein as an impurity comprising the steps of forming a melt of the metals, providing a mold cavity for continuous

casting, continuously charging the mold cavity with the melt and as the melt moves from the charging point progressively solidifying as the melt passes a first station a first portion of the melt from one longitudinal side thereof while maintaining the second portion of the melt adjacent to the opposite longitudinal side in a liquid phase, progressively solidifying at a following station the second portion of the melt from said opposite side thereof and remelting the first portion of the melt as the latter moves by the station, and as the melt moves by a third station progressively solidifying the first portion from said one side.

6. The method of purifying a first metal having a second metal of a different melting point present therein as an impurity comprising the steps of forming a melt of the metals, providing a mold cavity for continuous casting, continuously charging the mold cavity with the melt and as the melt moves from the charging point past a first station progressively solidifying a first portion of the melt from one longitudinal side thereof while maintaining the second portion of the melt adjacent the side opposite said one side in a liquid phase, progressively solidifying as the melt moves by a second station displaced from the first station in the direction of movement of the melt the second portion of the melt from said opposite side thereof and remelting the first portion of the melt, again progressively solidifying the melt as the latter moves by a third station displaced from the second station in the direction of movement of the melt the first portion from said one side of said first portion, and separating out the portion having the highest concentration of said first metal.

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