Oct. 21, 1952 J. C. MARINACE ET AL 2,615,060

CRUCIBLE FOR THE PURIFICATION OF MOLTEN SUBSTANCES

Filed Aug. 14, 1951

Fig. 1.

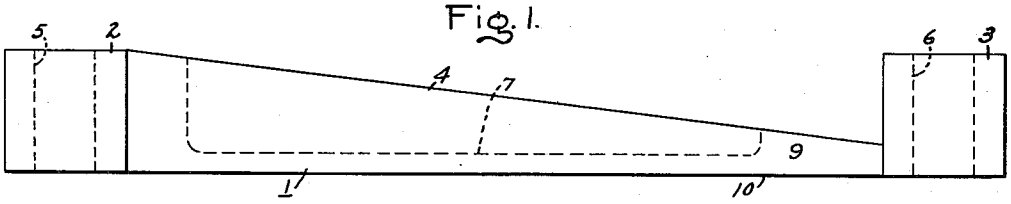


Fig. 2.

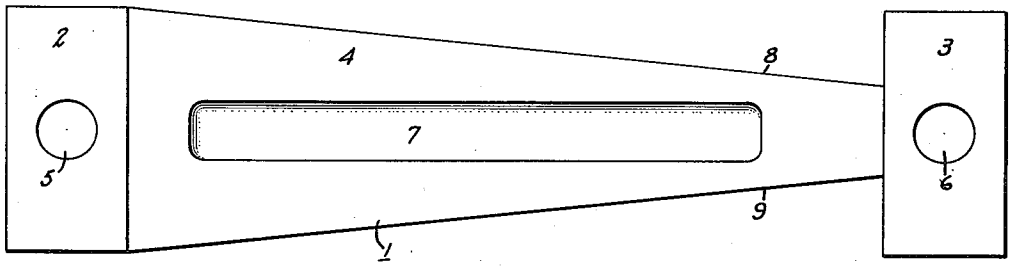


Fig. 3.

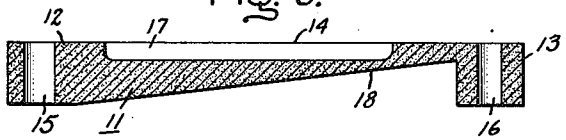
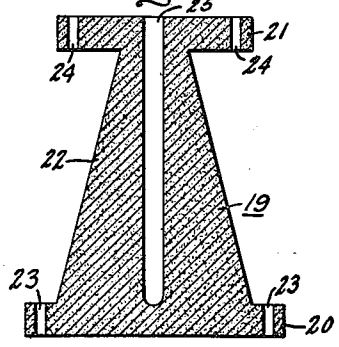


Fig. 4.



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CRUCIBLE FOR THE PURIFICATION OF MOLTEN SUBSTANCES

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2 Claims. (Cl. 13—25)

1

Our invention relates to an improved crucible for the purification of molten substances. More particularly, our invention is in a self-heating crucible of electrically-conducting-resistance material which has specific utility in the purification of metals and similar substances by what may be called the process of differential-directional cooling.

According to a known process for the purification of metals (for example, germanium), a sample of the substance to be purified is heated until it forms a molten ingot extending along a given axis in a mold. Heat to the mold carrying the ingot is then gradually reduced at one end so that the ingot progressively cools, starting in a given direction, along its axis. Obviously, the ingot freezes or solidifies at the cooler end of the mold first. As the ingot is further cooled gradually in this fashion, a liquid-solid interface travels along the ingot in a predetermined direction from one end of the axis of the ingot to the other, because of the differences in temperature between the two ends of the mold. In this process, impurities which prefer or favor the liquid phase of the material being purified are crowded toward the end of the ingot which is last cooled to its solid state. The growth of single crystals or large crystals in the structure of the ingot is promoted by this process, and impurities are readily separated from the completed ingot. One way that this process is carried out is by having a crucible heated in a constant temperature furnace and then moving the crucible gradually out of an opening in one wall of the furnace.

Known apparatus for carrying out this process, which we have called differential-directional cooling, is usually complicated, high in cost, difficult to control, subject to vibration during movement of the crucible, and slow and uncertain to operate. The objects of our invention are to provide a simple apparatus for this process, which is low in initial cost and up-keep, which is easy to control, rapid and convenient in operation, subject to no vibration because there are no moving parts, and in which single crystal or large crystal growth in the ingot is therefore greatly facilitated.

The objects of our invention are accomplished generally by making a crucible generally wedge-shaped or tapered, which crucible is in itself the electrical resistance element for heating the ingot mold and also forms the walls of the mold, the ingot axis extending generally along the axis of taper of the crucible-resistance-mold body. Other objects and further details of that which we believe to be novel and our invention will be clear from the following description and claims taken with the accompanying drawing, in which are illustrated three examples of crucibles embodying the present invention and incorporating the wedge or tapered shape.

2

In the drawing, Fig. 1 is a side view of a crucible according to our invention; Fig. 2 is a plan view of the crucible of Fig. 1; Fig. 3 is a longitudinal sectional view on a reduced scale of a modified form of a crucible according to our invention, and Fig. 4 is a vertical sectional view of another modification.

In carrying out the process for which our crucible is designed, it is desirable that an ingot mold be provided which is heated to a temperature sufficient to melt the sample substance which is to be purified. For this purpose, we provide a crucible body of graphite or other suitable electrical conducting-resistance material, designated generally by the numeral 1 in Figs. 1 and 2. Integral with the ends of the crucible body, we provide conducting terminal blocks 2 and 3. The top face 4 of the body between the terminal blocks is tapered or slanted downwardly from a highest portion adjacent the block 2 to a lowest portion adjacent the block 3. It is understood that the crucible will be placed or used in a substantially horizontal position. Therefore, the end-to-end wedge shape or taper in this form may be called a vertical taper.

In each of the end blocks, we provide suitable conductor terminal structures shown here in the form of holes 5 and 6 in the blocks 2 and 3 respectively. These holes may be used for the reception of cable terminal posts or connector plugs, not shown.

In the body of the crucible, we provide a mold or cavity 7 for the ingot, in the form of a channel extending endwise in the body. This mold opens into the top face 4 of the body, has a generally horizontal bottom, and is shown of uniform width throughout its length. Obviously, the side walls of the cavity may be rounded or tapered to aid in removing the completed ingot.

In addition to the vertical taper spoken of above, we prefer also to taper the sides of the body. For this purpose, the side walls 8 and 9 are tapered or slanted toward each other from a large or widest spacing at the block end 2 to the narrowest spacing at block 3. This may be called a horizontal taper.

In the form of crucible shown in Figs. 1 and 2, the bottom wall 10 of the crucible body is flat and horizontal, and serves as a stable support for the entire crucible.

The combined vertical and horizontal taper results in a preferred reduction in the transverse sectional area of the crucible body between the end blocks of from about 10:1. That is, the sectional area next to the block 2 is about 10 times as great as the area of the body where it joins block 3. This wedge or tapered form is the essence of our invention because it produces uniformly varying differences between cross-sections along the length of the crucible and consequent differences in electrical resistance. Assuming

that suitable power is applied to the terminals, the joule heat generated by current passing through the crucible walls differs from end to end of the crucible, so that the end nearest the block **3** is hotter than the end nearest the block **2**. This is so because the resistance is greatest in the narrow section and least at the wide section. A uniformly varying temperature gradient along the length of the crucible is therefore set up whenever current is applied to the terminals.

In operation, current is passed through the crucible until the sample is melted. The sample may be kept liquid as long as desired. To start cooling, the current is gradually reduced and the larger end of the crucible cools first and freezing or solidifying starts there. The crucible need not be moved. As the current is progressively reduced, the liquid-solid interface travels towards the smaller end adjacent the block **3** and those impurities which prefer the liquid phase are crowded in the ingot mold toward this smaller end of the crucible. The shape of the crucible itself is such that a proper and easily controlled temperature differential will always exist between the two ends of the ingot mold in the crucible. Merely changing the applied current will change the temperature. This increases speed and ease of control in operation of the apparatus when it is used to carry out the differential-directional cooling purification process, and eliminates vibration which might result from moving the crucible.

Obviously, the taper or wedge shape may be produced in other fashions than that shown in Figs. 1 and 2. For example, in Fig. 3, there is shown a crucible body **11** with terminal ends **12** and **13**. The bottoms of these ends serve as supports for the crucible body in this form of the device. The top face **14** in this form of crucible is flat and horizontal. Conductor terminal structures **15** and **16** in the ends **12** and **13** respectively are also provided. The mold cavity is the horizontal channel **17** extending endwise in the body and opening into the top face. The bottom wall in this second form of crucible is designated by the number **18** in Fig. 3 and is slanted upwardly so that the body is tapered from a thickest section at the end **12** to a thinner section at the end **13**. This slanted bottom wall has the same effect as the slanted top wall of the form of Figs. 1 and 2. Tapered side walls, not shown in Fig. 3, but like the side walls **8** and **9** of the first form, may obviously be used to provide further tapering.

In the form of crucible shown in Fig. 4, the electrical conducting resistance body is numbered **19**, and has ends **20** and **21** separated by a central tapered section **22**. The taper may be either conical or pyramidal in outer shape, as long as the required reduction in cross-section from end to end is maintained. Conductor terminal connection details in the form of holes **23** and **24**, in the ends **20** and **21** respectively, are provided as in the other forms.

The ingot cavity **25** in this form of the crucible extends longitudinally of the tapered section, this time in a vertical direction because all of the taper is about a vertical axis. Preferably the ingot mold axis is coincident with the axis of taper of the body so that equal heating is obtained on all sides of the cavity at any given level. For ease in removing a completed ingot from the crucible, the walls of the cavity may be slightly tapered from the upper open end to a narrower section at the closed lower end.

As current applied to the ends is reduced, the bottom end will cool first, in this form of the invention, and the open top of the cavity will allow for possible expansion of the material as it solidifies, the top being the last to become frozen. Obviously, if used with a material which shrinks upon cooling, the taper of the crucible body of this form might be reversed so that the top would cool first and the bottom last.

If desired, the mold cavity may be lined with a material different from that of the body of the crucible itself. For example, if the substance to be melted reacts unfavorably with a graphite crucible, a ceramic "boat" or "pocket" may be snugly fitted into the mold cavity to prevent such reaction. Moreover, if the substance to be melted conducts electricity to such an extent that it upsets the resistance-produced temperature gradient, a non-conducting liner in the form of a poreclain or similar coating for the cavity may be added to any of the forms shown.

As will be evident from the foregoing description, certain aspects of our invention are not limited to the particular details of construction of the examples illustrated, and we contemplate that various and other modifications and applications of the invention will occur to those skilled in the art. It is therefore our intention that the appended claims shall cover such modifications and applications as do not depart from the true spirit and scope of our invention.

What we claim as new and desire to secure by Letters Patent of the United States is:

1. A crucible for the purification of metals comprising a horizontally extending body of electrical resistance material, top, bottom and side faces on said body, electrical terminals at opposite ends of the body, said body having an ingot mold cavity in its top face extending substantially along the length of said face, the faces of said body being uniformly tapered in cross section from end to end in both horizontal and vertical planes from a greatest cross section adjacent one end to a smallest cross section adjacent the opposite end, whereby, upon application of current to said terminals, uniform end-to-end temperature gradient will be impressed on said mold cavity.

2. The crucible of claim 1 in which the reduction in cross-sectional area from end to end is of the order of 10 to 1.

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THEODORE E. LE LOUP.

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