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METHOD OF TREATING MELTABLE MATERIAL BY FLOATING ZONE-MELTING

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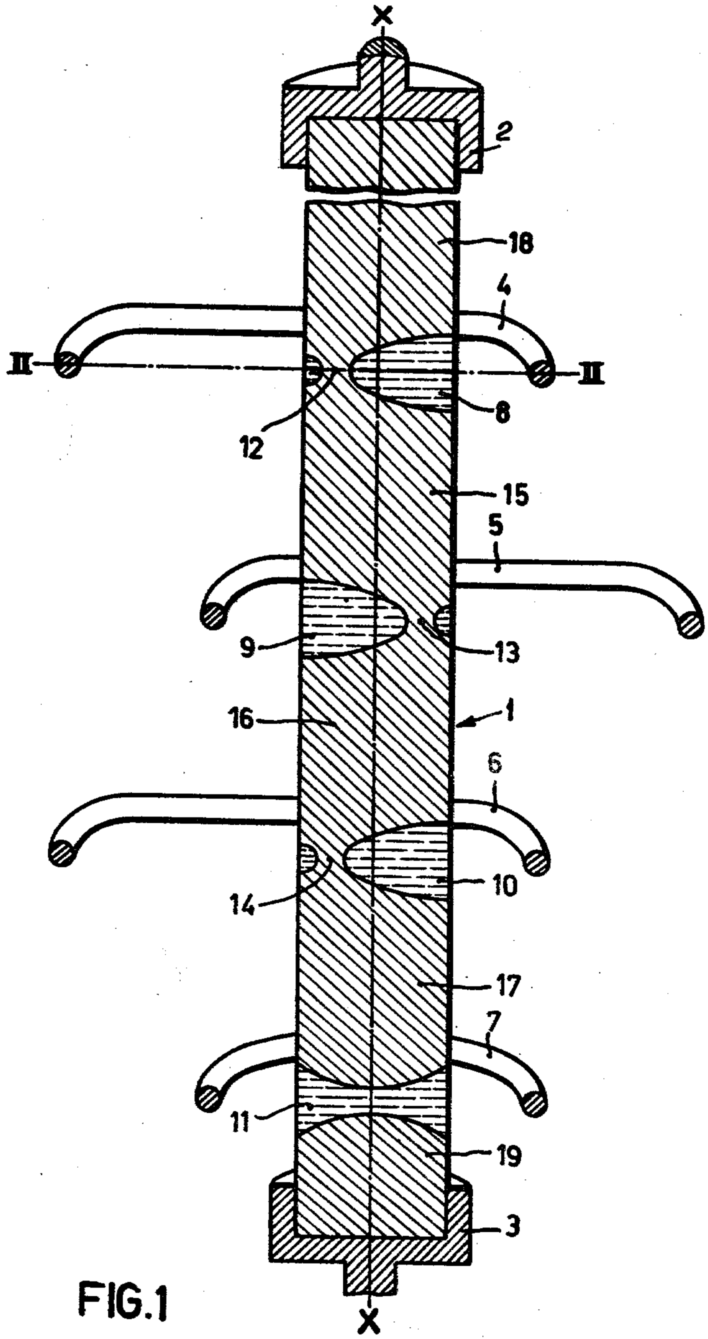

FIG.1

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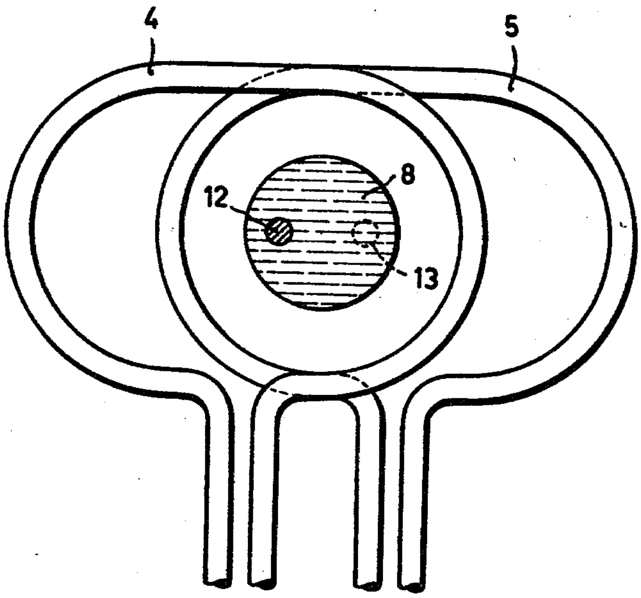

FIG. 2

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METHOD OF TREATING MELTABLE MATERIAL BY FLOATING ZONE-MELTING

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4 Claims. (Cl. 23—293)

In zone-melting material, in particular for its purification, it is known to pass successively a number of molten zones through a rod consisting of this material. If more than one zone is simultaneously passed through a rod, the rod is divided into a number of separate solid parts, so that this method of zone-melting can be used only if the rod is supported throughout its length, for example in a crucible. In floating zone-melting, it is evidently not possible to melt simultaneously more than one zone in a rod because the part lying between these zones would not be supported at all. When using floating zone-melting it is therefore common practice to pass a single molten zone a number of times through the rod, which is very time-consuming and expensive.

Floating zone-melting methods are known also, in which a partial molten zone is passed through a rod, by which is understood here a zone which does not occupy the whole cross-section of the rod but in which a preferably eccentric part thereof, hereinafter termed "unmelted core," remains in the solid state so that the parts of the rod on either side of such a partial zone are permanently linked together. In order to purify these parts of the rod too, it has been proposed to rotate the rod each time after passing a zone about its longitudinal axis through angles of for example 180° or 120° and again passing a partial zone through the rod. In order to purify the material throughout its cross-section by passing one partial zone, it has been proposed in addition to pass the zone through the rod along a helical line having a short pitch. However, both methods are time-consuming and not very efficient.

The condition that the parts of the rod on either side of a partial zone are permanently linked together was used in these known methods by simultaneously passing two or more zones through the rod, in which the non-melted cores, in the longitudinal direction of the rod, were the same. This method, too, is time-consuming.

The invention, which consequently relates to a method of zone-melting a rod without the use of a crucible, in which two or more partial zones are simultaneously passed through the rod in the longitudinal direction, has inter alia for its object to mitigate the above drawbacks.

It is based on the recognition that by suitable arrangement of the zones with respect to each other a considerable purification throughout the cross-section of the rod can be obtained already after once passing these zones.

According to the invention, these zones, in the longitudinal direction of the rod which is in a vertical position, overlap each other in a manner such that together they cover at least once the whole cross-section of the rod.

Preferably, more than two partial molten zones are simultaneously passed through the rod, in which each time two successive zones together cover at least the whole cross-section of the rod. The non-melted cores associated with two successive zones are preferably situated on either side of the axis of the rod but may also be arranged in a different manner.

Preferably, at the same time a complete molten zone situated behind the other zones is also passed through the rod so as to remove inhomogeneities, if any.

The effective action of each partial zone will be the larger according as an area of the cross-section of the unmelted core associated with each zone is smaller. However, this area should be large enough to form a rigid connection between the parts of the rod on either side of the zone. Preferably, this area is at most one fourth and at least one hundredth of the area of the cross-section of the rod. It has appeared that for practical purposes the choice from one tenth to one twentieth is readily usable.

In order that the invention may be readily carried into effect, one embodiment thereof will now be described in greater detail, by way of example, with reference to the accompanying drawings, in which:

FIGURE 1 diagrammatically shows a vertical sectional view of a device for floating zone melting, and

FIGURE 2 is a horizontal sectional view of this device taken along the line II—II of FIGURE 1.

In FIGURE 1, 1 is a vertically provided rod-shaped body of meltable material, such as a meltable metal or semi-conductive material, for example silicon, which is rigidly secured at its upper end to a holder 2 and at its lower end to a holder 3. The device further comprises four coils which are connected to a high-frequency generator not shown. The coils 4, 5 and 6 enclose a plane of a somewhat elongated shape (see FIGURE 2) and are provided eccentrically with respect to the axis of the rod, shown in FIGURE 1 by the dot and dash line X—X, while the smaller lower coil 7 is circular and provided coaxially with respect to this axis of the rod. The coils 4 and 6 are provided straight below one another, while the coil 5 assumes a position with respect to these coils shifted through an angle of 180° about the axis X—X of the rod.

As a typical example of suitable coil dimensions, for a rod of 2 cm. in diameter, coil 7 may have a diameter of 35 mm. and the longer and shorter dimensions of the coils 4 to 6 may be 55 and 35 mm. respectively. As shown in FIGURE 2, the coils 4 to 6 may be arranged so that the rod 1 is approximately concentric with one end of the coil.

Preferably, the coils are connected in series but they may alternatively be connected in a different manner, if desired. They are preferably connected so that the high-frequency current from the generator will flow through each pair of successive coils in a mutually opposite direction of rotation.

By energization of the high-frequency coils 4, 5, 6 and 7, four molten zones are formed, 8, 9, 10 and 11 respectively, namely three partial zones 8, 9, and 10 which enclose the unmelted cores 12, 13 and 14 respectively, and one zone 11 which, owing to the strong energy absorption of the field of the small coil 7, extends over the whole diameter of the rod. The unmelted cores 12, 13 and 14 lie outside the axis of the rod X—X, notably the core 13 lies at a side of the axis X—X opposite to the side where the cores 12 and 14 are situated.

For the purification of the material of the rod 1, the set of coils is gradually moved upwards with respect to that rod. A typical rate for moving all of the coils for silicon may be about 0.2 cm. a minute. The molten zones transverse successive the rod-shaped body from the bottom to the top. The parts of the rod 15, 16 and 17 lying between the zones are firmly connected to the uppermost part of the rod 18 which is attached to the holder 2 by the unmelted cores 12, 13 and 14 while the part of the rod 19 connected to the lower holder 3 is separated from the uppermost parts by the molten zone 11. The holder 3 with the part of the rod 19 may be rotated, if desired. However, the holder 2 with the parts 15–18 connected to it is not rotated. During the upward movement of the zones 8–11 the material of the core 12 is melted again by the zone 9, the material of the core 13 by the zone 10, and the material of the core 14 by the zone 11.

The material of the part of the rod **19** has become an approximately equally large purity as when a complete molten zone would have been passed through the rod **1** three times, but in less than half of the period which is required for this latter treatment as will be explained below with reference to a calculation.

In zone melting a rod consisting of meltable material, the concentration change of an impurity in this material after passing each zone is to be indicated by a constant factor per zone, the so-called reduction factor α. This concentration change may be the result of segregation and/or evaporation of the impurity. In general, the reduction factor α has a different value for each impurity in the meltable material. In general, it will be smaller than unity.

If a number, n, of complete molten zones is successively passed through the rod, the concentration of an impurity in the treated material will have become a value of $\alpha^n C_0$, in which formula C_0 is the initial concentration of the impurity.

In the method according to the invention, partial molten zones are used which only partly occupy the cross-section of the rod. The relation between the area of the cross-section of the unmelted core of each of these partial zones and the area of the total cross-section of the rod is indicated by the factor x. In the present case, the factor x is the same for all the partial zones. In addition, the partial zones are chosen so that each pair of successive zones occupies at least the whole cross-section of the rod and three of such partial zones are used. In Table I, the concentrations of an impurity after passing each of these zones is summarized in formulae.

Table I

After passing	Concentration in the core	Concentration outside the core
1 zone	C_0	αC_0
2 zones	αC_0	$\left(\frac{x}{1-x}\alpha+\frac{1-2x}{1-x}\alpha^2\right)C_0$
3 zones	$\left(\frac{x}{1-x}\alpha+\frac{1-2x}{1-x}\alpha^2\right)C_0$	$\left\{\frac{(2x-3x^2)}{(1-x)^2}\alpha^2+\frac{(1-2x)^2}{(1-x)^2}\alpha^3\right\}C_0$

If these three zones are succeeded by a molten zone which occupies the whole cross-section of the rod, the concentration of an impurity, after passing this zone, is expressed by the following formula:

$$\alpha^2 c_0\left\{\frac{x^2}{1-x}+\frac{3x-5x^2}{1-x}\alpha+\frac{(1-2x)^2}{1-x}\alpha^2\right\}$$

In Table II, the relation C/C_0, calculated by means of the latter formula and with different α, between the concentration C after, and the concentration C_0 before a zone melting treatment with the above four zones, is compared with the concentration relation C/C_0 in the treatment with a number of complete molten zones.

Table II

α	C/C_0 after three partial zones +one complete zone		C/C_0 after n complete zones		
	for $x=0.25$	for $x=0.1$	for $n=2$	for $n=3$	for $n=4$
0.03	9×10^{-5}	1.8×10^{-5}	90×10^{-5}	2.7×10^{-5}	0.08×10^{-5}
.1	1.4×10^{-3}	0.46×10^{-3}	10×10^{-3}	1×10^{-3}	0.1×10^{-3}
0.3	2.6×10^{-2}	1.4×10^{-2}	9×10^{-2}	2.7×10^{-2}	0.8×10^{-2}
0.5	1.15×10^{-1}	0.82×10^{-1}	2.5×10^{-1}	1.25×10^{-1}	0.62×10^{-1}

It goes explicitly forward from Table II that a purification efficiency may be achieved with the four simultaneously used zones which is comparable with or better than the result of the use of three complete molten zones.

When using more partial molten zones simultaneously, a considerably better result may even be obtained.

It is noted in addition that the distance between two successive coils with respect to the total length of the rod may be rather small. For example, a length of rod of from 40 to 50 cm. and a distance between successive coils of 4 cm. may be used. The combination discussed in the example of four successive coils as a result occupies only 12 cm. of the total rod length, so that one passage of the four zones produced by these coils may consume less than half the period required for passing three complete molten zones through the rod one at a time.

What is claimed is:

1. A method of floating-zone melting and treating of a rod-like body of meltable material selected from the group consisting of metals and semiconductors, comprising the steps of supporting the body in a vertical position, establishing in a first transverse region of the body a first molten zone that occupies more than half but less than the whole cross sectional area of the body leaving a first solid unmelted region in said first transverse region, establishing in a second transverse region of the body spaced above the first transverse region a second molten zone that lies directly above the first unmelted region adjacent the first molten zone and that also occupies more than half but less than the whole cross sectional area of the body at the second transverse region leaving a second solid unmelted region in the said second transverse region that lies directly above the first molten zone, and simultaneously passing the two spaced molten zones longitudinally through the body maintaining the overlying relationship of the molten zones and unmelted regions in the different transverse regions such that the molten zone in each transverse region sweeps through the same section of the rod-like body that remains unmelted in the other transverse region in order to treat the whole of the body cross section in a single pass by a melting treatment.

2. A method as set forth in claim 1 wherein there is established, spaced from the other molten zones, a third molten zone that occupies the whole cross sectional area of the body, and all three molten zones are simultaneously passed through the body in a direction such that the third molten zone follows after the first and second zones.

3. A method of floating-zone melting and treating of a rod-like body of meltable material selected from the group consisting of metals and semiconductors and having a longitudinal axis, comprising the steps of supporting the body in a vertical position, establishing in a first transverse region of the body a first molten zone that occupies more than half but less than the whole cross sectional area of the body leaving a first solid unmelted region in the first transverse region located off the longitudinal axis, establishing in a second transverse region of the body spaced above the first transverse region a second molten zone that lies directly above the first unmelted region adjacent the first molten zone and that also occupies more than half but less than the whole cross sectional area of the body at the second transverse region leaving a second solid unmelted region in the second transverse region that lies directly above the first molten zone and is also located off the longitudinal axis, and simultaneously passing the two spaced molten zones longitudinally through the body maintaining the overlying relationship of the molten zones and unmelted regions in the different transverse regions such that the molten zone in each transverse region sweeps through the same section of the rod-like body that remains unmelted in the other transverse region in order to treat the whole of the body cross section in a single pass by a melting treatment.

4. A method as set forth in claim 3 wherein, in each transverse region, the cross-sectional area of the unmelted region is between one-fourth and one-hundredth of the total cross-sectional area of the rod-like body.

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