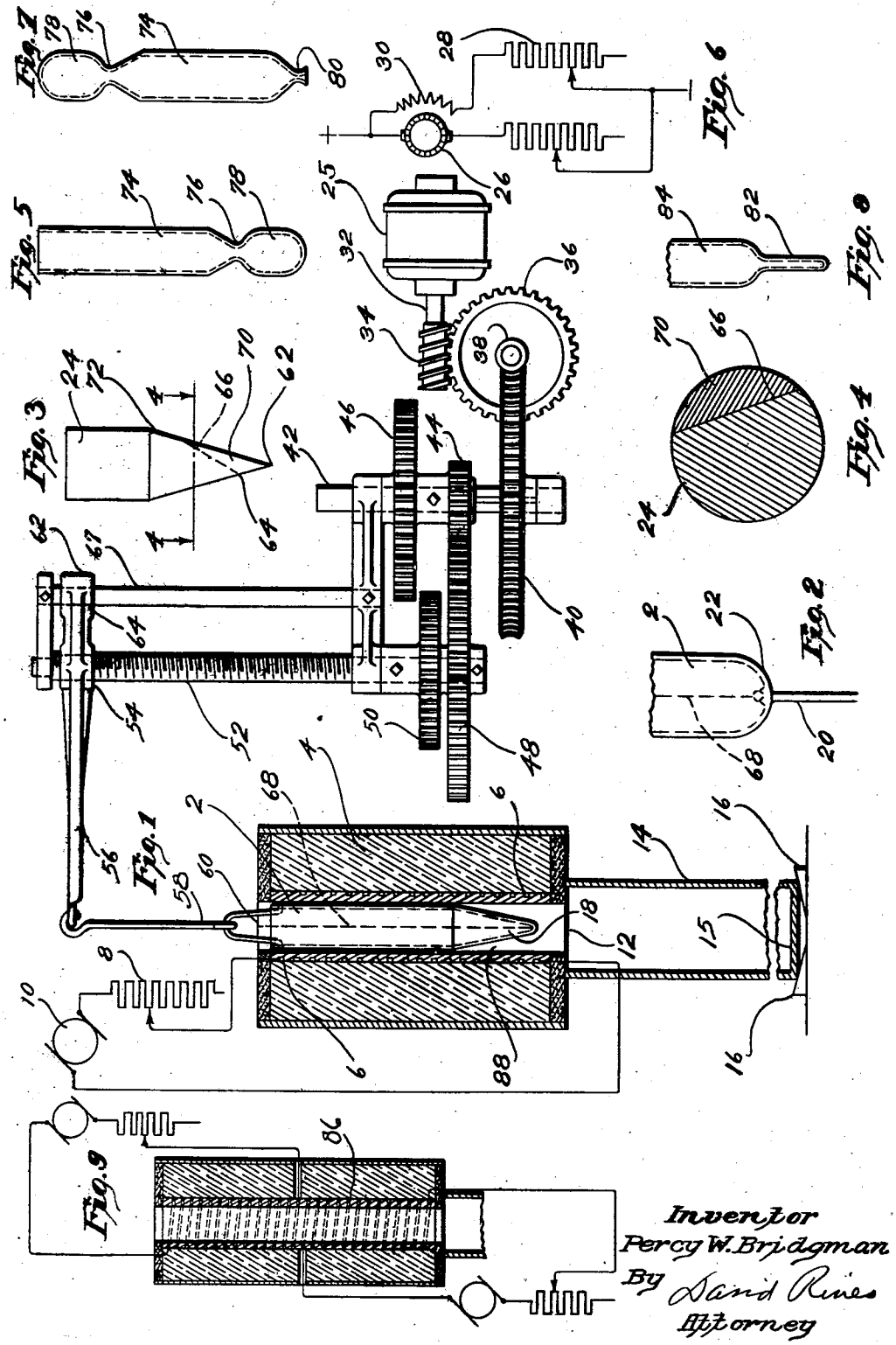
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CRYSTALS AND THEIR MANUFACTURE

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CRYSTALS AND THEIR MANUFACTURE

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The present invention relates to methods of and apparatus for making crystals, particularly large single crystals. The invention relates also to methods of and apparatus for purifying crystalline substances; also, to methods of and apparatus for molding, and to the molded substances, more particularly, molded large single crystals, thereby produced. The present application is a continuation in part of applications, Serial No. 687,147 filed January 18, 1924, and Serial No. 36,640, filed June 12, 1925.

To the layman, the word "crystal" connotes a regular, external form, comprising plane surfaces arranged along the crystal axes according to some law of symmetry. Not all substances having such regular form, however, are crystals; and a crystal is none the less a crystal because it has lost its external form, as by machining, or because it never had such form to start with. The distinguishing characteristic of a crystal is its internal structure. It will conduce to an understanding of the present invention, therefore, to bear in mind that in a crystal of any substance, the molecules are arranged throughout that particular body according to a definite law of regularity that is characteristic of the particular substance in question; though the fact should not be lost sight of that some substances may crystallize according to more than one law.

Many crystals are weak along their planes of cleavage or other characteristic planes,—so much so that the mere act of machining, to change their exterior contour, is liable to cause the parts of the crystal to slide or otherwise move relatively to each other, thereby destroying the regularity of the crystalline arrangement, and the unitary nature of the crystal. It is therefore fortunate that many crystalline substances, like the metals, are constituted of small crystal grains that are intimately bound together, helter-skelter, with their axes differently oriented, and biting into the impurities that are bound together with them; for this arrangement enables the metals to resist successfully the stresses to which they are subjected, in use. In the form of a single, large crystal, made in accordance with the present invention, the same metal will be found to be unable to withstand such stresses, but will yield along its gliding planes. A metal crystal of circularly cylindrical form, for example, if subjected to compression, may become shorter in length, but elliptical in cross section, or of some other cross-sectional shape, depending upon the nature of the crystalline substance. Although weak mechanically and, therefore, not so desirable as an ordinary, multicrystalline metal where mechanical strength is necessary, a single crystal has many properties which multicrystalline metal does not have. Among these are lower electrical resistance, much greater thermo-electric power in one direction than others, and much greater thermal expansion. It is, therefore, desirable, for many purposes, to be able to produce large, single crystals; and according to the purpose for which these crystals are to be used, it is desirable to be able to give them any external form, and, also, sometimes, to orient the crystal axes in any desired way with respect to the external form. Since, in general, a single crystal is exceedingly weak mechanically, it can not be easily or economically machined to give it the desired external form, as machining tends to destroy the regularity of the internal crystalline arrangement. It is therefore convenient to give it the external form desired by some operation of molding. A single crystal of the substance will naturally contain fewer impurities; for there is greater opportunity for impurities to become lodged between many irregularly small crystal grains than for them to become crystallized as part of a single crystal. A single crystal, furthermore, will not have the pipe and blow holes that are ordinarily so readily formed in the metals, as ordinarily manufactured.

It is therefore an object of the present invention to provide an improved method of, and apparatus for, making crystals. It is a further object to provide an improved method of and apparatus for purifying substances, particularly metals. Another object is to improve upon present-day methods of, and apparatus for, manufacturing moldings,

such as moldings of the metals, so as to avoid the formation of pipes and blow holes. It is a further object to control the orientation of the crystal axes with respect to the mold, where this may be desirable. Other and further objects will be explained hereinafter in connection with the accompanying drawings, and will be particularly pointed out in the appended claims.

In the accompanying drawings, Fig. 1 is a diagrammatic view, partly in section, of apparatus constructed according to a preferred embodiment of the present invention; Fig. 2 is a detail view of a modified mold; Fig. 3 is a fragmentary view of a crystal made with the mold shown in Fig. 1; Fig. 4 is a section taken upon the line 4—4 of Fig. 3, looking in the direction of the arrows; Fig. 5 is a view of a further modified mold; Fig. 6 is a diagrammatic view of circuits and apparatus for controlling the motor of Fig. 1; Figs. 7 and 8 are views of still further modified molds; and Fig. 9 is a view of a modified apparatus.

The crystal of the present invention may be made in many ways. According to the preferred method of the above-designated applications, the first step is to obtain, in molten form, the substance a molded crystal of which is desired or which it is desired to purify. The invention may, however, in its broader features, be practiced by first dissolving the substance in a suitable solvent. Most metals must be heated to melt them, and a vertically disposed mold 2, within which the substance is adapted to be contained, is therefore diagrammatically shown within a vertically disposed, cylindrical, electric furnace 4. The mold 2 must, of course, be constituted of a refractory substance such that it shall withstand the heat of the furnace, while permitting the melting of the metal, and the refractory substance must not be such as to become dissolved in the molten substance, or otherwise impart impurities thereto. For metals that liquefy at comparatively low temperatures, about 600° C., more or less, such as lead, tin, zinc, cadmium, tellurium, bismuth, antimony and the like, the mold 2 may be constituted of glass. If the crystal is to be molded to a complicated shape, it may be desirable to make the mold of graphite, which can be more readily formed to complicated shapes than glass. For substances that liquefy above the softening point of glass, the mold 2 must be constituted of some other material suitable to the purpose in hand. Before the furnace 4 is heated, the position of the illustrated mold 2 therein is suitably adjusted so that the furnace, after heating, may melt, and maintain melted, all parts of the substance contained in the mold 2. If a very long mold is used, longer than the furnace, metal may be fed into the top of the mold as the mold is passed through the furnace, melting the metal not all at once, but continuously. In either case, the metal will be thoroughly melted above the surface of deposition so that crystallization may take place from a liquid from which all nuclei of the solid have been removed by thorough melting, as will be understood from the description to follow. Any well known means may be employed for controlling the temperature of the furnace, such as a heating coil 6, in series with a variable resistor 8 and a source of energy 10. The mold is now gradually lowered into the cooler, outside air (Fig. 1) through an opening 12 in the bottom of the furnace 4, without affecting the operation of the furnace. In practice, to avoid convection air currents, the mold may be protected from direct contact with the outside air by a pipe 14 or the like that is maintained tightly against the bottom of the furnace 4 in any desired way, as by means of wedges 16. The bottom 15 of the pipe 14 is closed to prevent air currents from streaming upward through the pipe from below. The layers of the molten substance in the lower portions of the mold 2 will become solidified or crystallized upon leaving the furnace 4. A saturated solution may be used, instead, of a substance such that its solubility decreases as the temperature falls. The solute substance will then crystallize out as the mold leaves the furnace, depositing the crystal of the desired substance on the surface already crystallized. The deposition takes place at that surface whose temperature is the saturation temperature. Thus, sugar may be crystallized out of a saturated solution of sugar in some solvent, and salt out of a saturated salt solution. The solute will crystallize out of the solvent. Of course, the solvent may also be crystallized out, yielding a purified substance. An advantage of crystallizing from solution is that it enables one to obtain crystals of some substances that can not be conveniently obtained from the melt, such as salt and sugar. Sugar decomposes before melting, and the melting point of salt is inconveniently high, and, furthermore, is liable to become shattered by thermal contraction, as will be mentioned later.

If the bottom of the mold is of considerable magnitude, the metal will usually start crystallizing from a number of different nuclei at various places on the bottom, as, for example, at 62 and 64, Fig. 3, and therefore the casting may consist of several grains, with a plane of separation 66 between them. In the case of some metals, however, crystallization may be more rapid from grains which start with a particular orientation, so that some grains grow at the expense of others, and it may result that in the upper part of the mold, one grain has gained the upper hand. When this happens, the casting is, beyond a certain point, all one crystal grain. If it is desired merely

to obtain a cylindrical casting of no particular length, the lower end of the casting may be cut off, and the upper end affords the single, molded crystal desired. But in the case of many metals, the tendency for one grain to grow at the expense of others is not very strong, so that many grains may persist through the entire length of the casting at small angles to the axis 68 of the mold, if they ever get started; or it may be necessary to have the entire contents of the mold of one grain, without discarding any part. Hence, in most cases, it is desirable to rely on something more positive than mere chance to compel the crystallization to start from a single nucleus. This may be done in a variety of ways. It is found that if the lower end of the mold is drawn out to a sharp point, as at 18, Fig. 1, crystallization, in the case of many metals, is much more likely to start from a single nucleus at the sharp end. In order to function properly, the metal must run down cleanly into the sharp point; this may be conveniently accomplished by filling the mold in a vacuum, running the molten metal through a properly disposed filter to remove stray particles of mechanical dirt. In fact, with any method, it is necessary to have the metal mechanically clean, because it is found that particles of dirt are very apt to act as centers of new nuclei. If a mold with a pointed end 18 is used, the taper above the point must be so gradual that, when the mold emerges from the furnace, the temperature increases smoothly at greater and greater distances from the point, else new nuclei will be formed above the original surface of crystallization.

Crystallization from a single nucleus may also be effected by leading through the bottom wall of the mold 2 a pointed metal or other wire 20, Fig. 2 (depending upon the temperature of the molten substance that it is desired to crystallize), of better thermally conducting material than that of which the mold 2 is constituted. This effects cooling of the molten metal around the pointed wire 20 by conduction of the heat from the interior of the mold 2 to the outside, resulting in the formation of a single nucleus at the point where the wire 20 is positioned. If desired, the wire 20 may be artificially cooled from outside, so as to start the formation of a crystallized nucleus before it is commenced to lower the mold 2 out of the furnace 4. When the wire 20 is employed, the bottom of the mold need not necessarily be sharp pointed, as shown at 18, but may be rounded, as illustrated at 22 in Fig. 2.

By suitable manipulation and control, as the mold 2 is gradually lowered out of the furnace 4, the successive horizontal layers of the metal in the mold 2 may be caused to build up, one after the other, upon the crystal nucleus formed at the bottom of the mold, forming a single, unitary large crystal 24 having the shape of the mold. The shape of the mold thus determines the shape of the crystal. All that is necessary to effect this result is to lower the mold 2 out of the furnace 4 at a speed so low as to afford each successively lowered horizontal layer of the molten or dissolved substance time to crystallize before the next higher layer commences to crystallize. All parts of the substance having the crystallizing temperature will then lie on a single, horizontal surface of deposition that advances into the molten or dissolved substance at a speed less than the crystallizing velocity. The lowering is determined by two factors. In the first place, the speed must be less than (or, possibly, equal to) the maximum velocity of crystallization of the substance. This is a characteristic of the substance and the particular orientation of the crystal axes in the nucleus from which the crystallizing growth commenced. In the second place, the speed must be low enough so that the heat set free by the crystallizing substance, due to its latent heat of melting, may be conducted away into the surrounding atmosphere, so that the crystal surface on which the new crystal molecules become deposited will not tend to rise to a higher temperature than the melting point. This speed therefore depends upon the size, the design and the materials of the apparatus and will, in general, be less for large molds and for molds made of poorly conducting substances. Both of these factors may, for brevity, be included in the general statement that the speed must be lower than the crystallizing velocity of the substance. Similar considerations apply to crystals deposited from solution, as will be understood by persons skilled in the art.

There is, of course, no harm done in lowering the mold more slowly than the said crystallizing velocity; but a greater speed of lowering will cause the higher layers of the liquid to commence crystallizing from new nuclei while the lower layers are still liquid, with the result that the finally solidified mass may be constituted of a number of crystals, and not of a single crystal. As the layers of the liquid in the conical end portion 18 of the mold 2,—if a mold of varying diameter is employed—will naturally crystallize more rapidly than the upper layers, because being of smaller area, the mold 2 may be lowered at greater speed, to start with. The speed of lowering may be controlled in any desired way, as by clock-work, or by means of a variable-speed motor 25 having an armature 26, and the speed of which may be regulated automatically, or by hand, by varying the resistor 28 in series with a field-magnet winding 30, diagrammatically indicated in Fig. 6. The motor is adapted to rotate a shaft 32 that is connected by a

worm 34 to a gear 36. The gear 36 is concentric with a worm 38 that meshes with a gear 40. This double worm-gear arrangement provides for a lowering speed of the mold 2 as small as desired; say, one-half inch per hour. The conditions thus established are highly favorable to a similar orientation of the molecular crystal axes throughout the mold, and, therefore, to the formation of a single crystal in the mold. The gear 40 is rigid with a shaft 42 upon which are slidably mounted two gears 44 and 46. The gears 44 and 46 are keyed to the shaft 42 so as to rotate therewith, and are adapted selectively to mesh with gears 48 and 50, respectively, thus providing for a further range in the lowering speed of the mold 2. The gears 48 and 50 are mounted upon a threaded shaft 52, the threads of which engage in a non-rotating nut 54. The rotation of the shaft 52, therefore, effects the raising and lowering of an arm 56 that is integral with the nut 54, and from which the mold 2 is suspended by a link or cord 58 that passes through an opening 60 at the top of the furnace 4. An extension 62 of the nut 54 is provided with an opening 64 within which a rod 67 is slidably mounted to guide the arm 56, and the mold 2 that is suspended therefrom, against swinging movement.

It has been found by experience that, in some cases, and with some materials, a nucleus of one orientation will grow at the expense of nuclei of other orientations, so that that nucleus will eventually prevail, even though there may have been several nuclei initially present.

This mold 74, though of somewhat more complicated construction, is more certain in operation, and more desirable for some metals. This mold consists of two portions, a lower chamber 78, the function of which is to start the crystallization, and which may be cut from the rest of the mold when the casting is completed, and the main part of the mold 74. The chamber 78 is separated from the part 74 by a narrow neck 76. Crystallization starts at the lower end of the chamber 78 from a number of nuclei; but because of the slow rate of deposition, it will be found that the grains which grow from these nuclei are not microscopic in size, as in the case of many ordinary castings, but are larger, perhaps of the order of a millimeter or more in diameter. If, however, the neck at 76 is made smaller than these grains, perhaps ¼ millimeter in diameter, it will result that, except under especially unfavorable conditions, only one of the grains in the chamber 78 is properly situated to grow through the neck 76, so that crystallization in the chamber 74, which is the main crystallizing chamber, and has the shape to which it is desired to mold the crystal, starts from a single grain at the neck 76, and therefore the entire main part of the mold crystallizes as a single grain. The mold may be readily filled by first exhausting the air from the mold, and then allowing the molten metal to flow in, driven by external air pressure. The mold 74, with the substance that it is desired to crystallize, in solid form, is placed in the furnace 4, after which the air is exhausted by a pump from the mold 74. The furnace 4 is then heated to melt the substance contained in the mold 74. The molten substance readily drops into the vacuum of the mold 74 to the bottom of the mold, and through the small, restricted neck 76, into the enlarged chamber 78 at the bottom of the mold. This structure, therefore, renders it possible to prevent air becoming trapped in the body of the mold 74, which is particularly necessary when a small mold is used to manufacture small crystals. The method of crystallizing is the same as before described, the small neck 76 insuring that the substance shall crystallize from a nucleus at that point, into a single crystal.

A modification of the procedure outlined above has been found desirable in the case of bismuth, which differs from the other metals mentioned above in that it expands on solidification, instead of contracting. It has been found that it is much more difficult to produce single crystals of bismuth by the method above than of the other metals. The reason is that the solid bismuth, which is deposited on the bottom of the mold, is lighter than the liquid above it, so that particles of the solid tend to detach themselves from the surface of deposition and, rising through the liquid, act as new nuclei of crystallization of an orientation different from that of the original crystal. This difficulty may be avoided by causing the solid to deposit itself on the top of the mold instead of the bottom. In order to effect this, the mold may be raised through the top of the furnace, instead of through the bottom, using for the purpose a special form of mold. This mold is shown in Fig. 7 and is essentially the mold of Fig. 5 inverted, though the other illustrated molds could equally well be used inverted. At the upper end, is a small chamber 78, connecting by a narrow neck 76 with the main crystallizing compartment 74, which is provided at the lower end with a narrow opening 80. The tube is filled with molten metal by first exhausting the air, and then allowing the molten metal to flow in through the bottom under external air pressure, thus filling the entire tube by barometer action. The neck 80 is so small that air bubbles can not pass into the chamber, and the liquid is maintained in position by the external air pressure. As crystallization proceeds, the volume of metal in the mold increases, and liquid metal drops out through the neck 80 in compensation. Crystallization may start in many nuclei at the extreme upper end of the mold,

but the neck or channel 76 is so fine that only one of the crystals produced from these nuclei grows into the neck, so that crystallization in the main crystallizing compartment 74 starts from a single nucleus, and only one crystal grain is formed in the main compartment.

The procedure outlined for the other metals is now duplicated for bismuth (or other similar metals) with the exception that, wherever formerly the mold was lowered, here it is to be raised. It will be understood that the shield 14 to prevent the access of air currents to the outside of the mold must now be placed about the upper end of the furnace, instead of the lower end, and that there may be other similar changes which may be readily made, but need not be mentioned in detail. It will also be understood that a similar procedure will apply when crystals are deposited from solution, where the crystallized substance is lighter than the liquid solvent.

If crystallization starts at haphazard, as described above, the crystal axes will have a haphazard orientation with respect to the mold. It will be found, in practice, however, that there is usually a preferred direction of orientation, so that the majority of the castings made by this method will have the axes approximating to some special direction. For the low-melting metals mentioned above, it will be found that the principal cleavage or slip plane prefers to stand perpendicular to the plane on which the crystal is deposited. The strength of this preference is much greater in crystals of large than of small diameter; cases of highly oblique incidence of the cleavage planes are not uncommon in the smaller-diameter castings. In order to control the direction of the crystalline axes, the mold illustrated in Fig. 8 may be used. It is, in the first place, necessary to start from a seed crystal of the desired orientation. This seed may be obtained by selecting from a large number of castings of small diameter one which has the desired orientation. These castings may, if desired, be made of one of the methods already described. This seed is placed in a long slender projection 82 attached to the lower end of the main part of the mold 84. The main part of the mold is now placed in the electric furnace, with the projection 82 extending out into the air. The mold is now charged with metal and melted. After the charge is completely melted, the mold is drawn up partially into the furnace, so that the upper part of the seed crystal is melted, but not the lower part. The mold is now lowered in the regular way by the slow lowering mechanism, and crystallization begins from the upper end of the seed crystal, which forces the material freshly deposited to have the same crystal orientation as itself, so that the orientation of the crystal formed in the main part of the mold 84 is thereby controlled.

Some substances, particularly various salts, become torn into a number of pieces as the mold is lowered out of the furnace and into the outside air. This is because the temperature gradient between the furnace and the outside air is so great that the newly solidified substance, which is less strong that the metals, can not withstand the differential thermal expansion resulting from their passing through this region. Single crystals of these substances may, however, be produced, by lowering the mold from the furnace 4, not into the outside air, but into a second furnace 86, as shown in Fig. 9. The furnace 86 preferably comprises two windings, each with its own control, as illustrated, with no separating material between the two parts of the double furnace. The furnace 4 is maintained heated to a temperature slightly above the melting temperature, and the furnace 86 at a temperature slightly below. After the mold has been completely lowered into the furnace 86, the mold is held stationary and the temperature of the furnace 86 is slowly reduced to that of the outside air. Large thermal distortions resulting from steep temperature gradients are thus avoided and the crystals are thus prevented from becoming shattered. It will be readily understood that multicrystalline, unshattered solids may similarly be produced at a more rapid rate by increasing the speed of lowering of the mold.

It has been stated that crystals may be produced either from the molten state or from solutions. There is still another method of production. Some metals, like zinc and tin, crystallize from the melt in one crystalline form at the freezing temperature (232° C. in the case of tin) but, on further cooling, as the mold is lowered further from the furnace (to 161° in the case of tin) the crystal formed from the melt is spontaneously transformed to another sort of crystal. If the crystal resulting from direct solidification out of the melt were to be maintained in the form thus produced, as by means of the apparatus illustrated in Fig. 9, it would be of quite a different character from that obtainable by lowering the mold directly from the furnace 4 into the outside air. The space lattices of the two crystals are in general quite different. The final crystal of such a substance is therefore the product of two transitions, first from the melt to the one form of solid, and then, at a temperature between the melting point and ordinary temperatures, from this form of solid to the other form of solid. It is just as necessary to control the second transition, from the high-temperature form of solid to the low-temperature modification, as it is to control the first transition, if a single crystal is to be produced; and exactly the same methods of control above described

are applicable. In the apparatus of Fig. 1, the two transitions take place successively and automatically at the appropriate temperatures as the mold is lowered out of the furnace and into the outside air. The first transition may, for example, take place at 88, Fig. 1; and the second transition when the mold leaves the furnace 4.

In the case of zinc and tin, in which there is little internal resistance to the change from the high-temperature form to the low-temperature form, the final crystal is always of one form at room temperature. Some substances, however, in which the internal resistance to the transition is greater, may exist at room or other temperatures in two or more distinct forms. The production of one or another of the desired forms may be controlled in the apparatus (Fig. 9) by suitably controlling the lowering of the furnace 86 to room temperature after a single crystal has been produced from the melt.

There is also another way to handle such substances. When a bar of ordinary tin, which has a transition at 161° C., is heated to between 161° C. and its melting temperature, 232° C., it will be transformed into the high-temperature modification. If this bar is now placed in an electric furnace maintained at 200° C., and slowly lowered, as above described in connection with the lowering of molds, the lower end of the bar will begin to become transformed into the low-temperature modification at 161° C. The lower end of the bar may be made pointed, like the lower end of the mold shown in Fig. 1, and if the lowering is slow enough, a single crystal will be produced in the same manner as before described. It is thus possible to produce crystals of some substances without melting and without using molds.

In all cases, the substance is moved from a higher-temperature region to a lower-temperature region in which the molecules have a different arrangement. On passing from the region in which the higher temperature prevails into the lower-temperature region, the molecules rearrange themselves. If the operation is slow enough, they will so rearrange themselves that they shall all form parts of the same crystal lattice, or one large crystal grain. This is true whether the high-temperature condition is that of molten metal, or a solution or another crystal modification. In the two first-named cases, a mold is necessary; in the third, it is not.

In order to avoid circumlocution of language the term "freezing temperature" or its equivalent will be employed in the claims to denote "freezing or transition temperature", including the change from one crystal solid to another, including also, separation from solution.

Crystals made in accordance with the present invention are much purer than the mother substance from which the crystals are produced. This is because, though some impurities readily crystallize into the crystal, many do not. Such impurities will therefore be found concentrated at the top of the crystal, and can be removed with facility. The invention therefore provides a very efficient means of purifying substances that lend themselves to crystallizing. If it is desired to have the purified substance in the form in which it is ordinarily used, it may be remelted and allowed to solidify in the ordinary manner. It should be noted, however, that when the described method is used for purposes of purification, the speed of lowering depends not only upon the two factors already explained, but also upon the rapidity with which the impurity will diffuse away from the surface of deposition, towards the top of the mold, through the molten substance. For this reason, the speed may be less than when the only object in view is to form a single crystal, whether or not relatively pure. On the other hand, if the sole object aimed at is purification, and if the impurity is such that it diffuses rapidly, the lowering may be more rapid than is required for the production of a single crystal. Under some circumstances, it may be desirable to stir the liquid to hasten the segregation of the impurities.

Not only is it possible to obtain very pure, single crystals, of any desired shape and size, by the method of the present invention, but the molded crystals so produced will be free from pipes, blow holes, or other flaws. The unitary character of the crystals is readily demonstrated in various ways, for the crystals possess properties that are not possessed by similar masses of the same substances that are solidified in the ordinary way.

It will be understood that the invention is susceptible of considerable modification and change by persons skilled in the art, and all such are considered to fall within the spirit and scope of the invention. It is therefore intended to express in the appended claims all the novelty that the invention may possess.

The molded crystal made in accordance with the present invention is not claimed herein, as it forms the subject matter of the aforesaid application, Serial No. 36,640.

What is claimed is:

1. A method of solidifying a molten substance that comprises placing a seed crystal adjacent to the substance, melting a portion of the seed crystal adjacent to the substance, and advancing the surface of solidification from the seed crystal into the substance.

2. A method of crystallizing a molten substance that comprises placing a seed crystal adjacent to the substance with the axis of the seed crystal oriented in a predetermined di-

rection, advancing the surface of crystallization of the substance from the seed crystal outward into the molten substance, whereby the axis of the resulting crystal will have the same orientation as the seed crystal, and limiting the speed of advance of the said surface into the molten substance to a value smaller than the crystallizing velocity of the substance.

3. A method of solidifying a molten substance that comprises placing a seed crystal adjacent to the substance with the axis of the seed crystal oriented in a predetermined direction, melting a portion of the seed crystal adjacent to the substance, advancing the surface of deposition from the seed crystal into the substance, whereby the axis of the resulting crystal will have the same orientation as the seed crystal, and controlling the speed of advance of the surface.

4. A machine for crystallizing a substance having, in combination, a mold having a body portion and a slender projecting portion, the projecting portion being adapted to contain a seed crystal with the axis of the seed crystal oriented in a predetermined direction, and the body portion being adapted to contain a substance, means for melting a portion of the seed crystal adjacent to the substance, and means for crystallizing the substance from the seed crystal outward with the substance, whereby the axis of the resulting crystal will have the same orientation as a seed crystal.

5. A machine of the character described having, in combination, a mold having a body portion adapted to contain a substance and a slender projecting portion adapted to contain a seed crystal, means for relatively moving a portion of the projecting portion from a region that is maintained below the freezing temperature of the substance into a region that is maintained at or above the freezing temperature of the substance, whereby a portion of the seed crystal is melted, and means for moving the mold from the second-named region into the first-named region to crystallize the substance.

6. A method of crystallizing a molten substance that comprises advancing the surface of crystallization of the substance into the molten substance, limiting the speed of advance of the said surface into the molten substance to a value smaller than the crystallizing velocity of the substance, and controlling the orientation of the crystal axes.

7. A machine for crystallizing a substance having, in combination, a furnace maintained at a temperature above the freezing temperature of the substance, a furnace maintained at or slightly below the freezing temperature of the substance, a mold adapted to contain a substance, means for moving the mold from the first-named furnace into the second-named furnace, and means for changing the temperature of the second-named furnace.

8. The method of the character described that comprises relatively moving a mold containing a substance from a region that is maintained at a temperature above the freezing temperature of the substance into a region that is maintained at or slightly below the freezing temperature of the substance, and changing the temperature of the second-named region.

9. A method of crystallizing a substance that comprises relatively moving a mold containing the substance from a furnace that is maintained at a temperature above the freezing temperature of the substance into a furnace that is maintained at or slightly below the freezing temperature of the substance, controlling the speed of movement of the mold, and gradually changing the temperature of the second-named furnace.

10. A method of crystallizing a substance that comprises advancing the surface of deposition of the substance, controlling the speed of advance of the surface to produce a single crystal of the substance, then redepositing the substance along new surfaces of deposition to produce a crystal of the substance of different form from the first-named crystal.

11. A method of solidifying a substance in a mold that comprises placing a seed crystal adjacent to the substance with the axis of the seed crystal oriented in a predetermined direction with respect to the mold, melting a portion of the seed crystal adjacent to the substance, and advancing the surface of solidification from the seed crystal into the substance, whereby the axis of the resulting crystal will have the same orientation with respect to the mold as the seed crystal.

12. A method of solidifying a substance that is in liquid form that comprises crystallizing a nucleus of the substance, successively subjecting successive portions of the substance while in liquid form and that are successively farther and farther removed from the nucleus to a temperature at or below the freezing temperature of the substance, and controlling the speed at which the said successive portions of the substance are subjected to the said temperature.

13. A method of crystallizing a substance the molecules of which are arranged according to a definite law of regularity into a crystal the molecules of which are arranged according to another definite law of regularity, the said method comprising crystallizing a nucleus of the substance according to the said other definite law of regularity, successively subjecting successive portions of the substance that are successively farther and farther removed from the nucleus to a temperature at or below the freezing temperature of the substance, and controlling the speed at which the said successive portions of the substance are subjected to the said temperature to maintain each of the said successive por-

tions of the substance in condition such that the molecules thereof are arranged according to the first-named definite law of regularity until after the next preceding portion of the substance has become crystallized according to the said other definite law of regularity.

14. A method of the character described that comprises successively solidifying successive surface layers of successively increasing area of a molten substance at a speed such that each of the said successive surface layers becomes solidified before the next succeeding surface layer becomes solidified.

15. A method of the character described that comprises successively solidifying successive surface layers of successively increasing area of a molten substance and maintaining each of the said successive surface layers in molten condition until after the next preceding surface layer has become solidified.

16. A method of the character described that comprises successively subjecting successive surface layers of successively increasing area of a substance to a temperature at or below the freezing temperature of the substance at a speed such that each of the said successive surface layers become solidified before the next succeeding surface layer becomes solidified.

17. A method of the character described that comprises successively subjecting successive layer of successively increasing area of a substance the molecules of which are arranged according to a definite law of regularity to a temperature at or below the freezing temperature of the substance to cause the molecules to become arranged according to another definite law of regularity and maintaining each of the said successive layers in condition such that the molecules are arranged according to the first-named definite law of regularity until after the molecules of the next preceding layer have become arranged according to the said other definite law of regularity.

18. A method of the character described that comprises relatively moving a mold containing a substance from a region that is maintained above the freezing temperature of the substance into a region that is maintained at or below the freezing temperature of the substance at a speed of movement less than the crystallizing velocity of the substance.

19. A machine for crystallizing a molten substance having, in combination, means for crystallizing a nucleus of the substance and for advancing the surface of deposition into the molten substance from the nucleus along adjacent portions of the substance that are successively farther and farther removed from the nucleus, means for controlling the speed of advance of the surface to maintain each of the said successive portions of the substance in molten condition until after the next preceding portion of the substance has become crystallized, so as to afford each successive layer of the substance time to crystallize before the next surface layer commences to crystallize, and means for maintaining the successive portions of the substance in molten condition until the next preceding portion of the substance has become crystallized.

20. A machine for crystallizing a molten substance having, in combination, means for advancing the surface of deposition into the molten substance along successive portions of the substance, means for controlling the speed of advance of the surface to permit each of the said successive portions to crystallize to the next previously crystallized portion of the crystal in the crystal arrangement already prevailing in the said portion of the crystal, so as to afford each successive layer of the substance time to crystallize before the next surface layer commences to crystallize, and means for maintaining the successive portions of the substance in molten condition until the next preceding portion of the substance has become crystallized.

21. A machine for solidifying a molten substance having, in combination, means for crystallizing a nucleus of the substance and for successively subjecting successive portions of the substance that are successively farther and farther removed from the nucleus to a temperature at or below the freezing or transition temperature of the substance, and means for controlling the speed at which the said successive portions of the substance are subjected to the said temperature.

22. A machine for crystallizing a substance having, in combination, means for crystallizing a nucleus of the substance and for successively subjecting succcessive portions of the substance that are successively farther and farther removed from the nucleus to a temperature at or below the freezing temperature of the substance, and means for controlling the speed at which the said successive portions of the substance are subjected to the said temperature to maintain each of the said successive portions of the substance in condition until after the next preceding portion of the substance has become crystallized.

23. A machine of the character described having, in combination, a mold adapted to contain a substance, means for relatively moving the mold from a region that is maintained above the freezing temperature of the substance into a region that is maintained at or below the freezing temperature of the substance, and means for limiting the movement of the mold to a speed comparable to the crystallizing velocity of the substance.

24. A machine of the character described having, in combination, a mold adapted to contain a substance, and means for relatively moving the mold from a region that is maintained above the freezing temperature of the

substance into a region that is maintained at or below the freezing temperature of the substance at a speed of movement less than the crystallizing velocity of the substance.

25. A machine of the class described having, in combination, a furnace, a vertically disposed mold adapted to contain a substance, means for relatively moving the mold out of the furnace, and means for limiting the movement of the moving means to a speed comparable to the crystallizing velocity of the substance.

26. A machine of the class described having, in combination, a furnace having an opening in its bottom, a vertically disposed mold adapted to contain a substance, means for relatively moving the mold out of the furnace through the opening, and means for limiting the movement of the moving means to a speed comparable to the crystallizing velocity of the substance.

27. A machine of the class described having, in combination, a furnace, a mold adapted to contain a substance, means for moving the mold into and out of the furnace, and means for limiting the rate of movement of the mold to a speed comparable to the crystallizing velocity of the substance.

28. A machine of the class described having, in combination, a furnace, means for controlling the temperature of the furnace, a mold adapted to contain a substance, means for moving the mold into and out of the furnace, and means for limiting the rate of movement of the mold to a speed comparable to the crystallizing velocity of the substance.

29. A method of solidifying a molten substance that comprises advancing the surface of solidification into the molten substance at a speed low enough to afford each successive surface layer of the substance time to crystallize before the next surface layer commences to crystallize, and maintaining the successive portions of the molten substance in molten condition until the next preceding portion of the substance has become crystallized.

30. A method of solidifying a molten substance that comprises crystallizing a nucleus of the substance, advancing the surface of deposition into the molten substance from the nucleus along adjacent surface layers of the substance that are successively farther and farther removed from the nucleus at a speed low enough to afford each successive surface layer of the substance time to crystallize before the next surface layer commences to crystallize, and maintaining the successive portions of the molten substance in molten condition until the next preceding portion of the substance has become crystallized.

31. A method of crystallizing a molten substance that comprises advancing the surface of crystallization of the substance into the molten substance, limiting the speed of advance of the said surface into the molten substance to a value smaller than the crystallizing velocity of the substance, and maintaining the successive portions of the molten substance in molten condition until after the next preceding portion of the substance has become crystallized.

32. A method of crystallizing a molten substance that comprises subjecting successive molecular layers of the molten substance to a temperature at or below the freezing temperature of the substance, and limiting the speed at which the said successive molecular layers are subjected to the said temperature to a value less than the crystallizing velocity of the substance.

33. A machine for purifying an impure molten substance by crystallization having, in combination, a furnace, a mold adapted to contain the substance, means for relatively moving the mold out of the furnace to advance the surface of deposition into the substance and means for controlling the speed of relative movement of the mold to maintain the respective successive portions of the substance in molten condition until the impurities contained in the substance have successively diffused away from the said respective portions.

34. A method of purifying an impure substance by crystallization that comprises relatively moving a mold containing the substance from a region that is maintained above the freezing temperature of the substance into a region that is maintained at or below the freezing temperature of the substance, and controlling the speed of relative movement of the mold to afford the impurities contained in the substance time to diffuse from the surface of deposition through the substance.

35. A method of crystallizing a molten substance that comprises relatively moving the molten substance from a region that is maintained above the freeing temperature of the substance into a region that is maintained at or below the freezing temperature of the substance at a relative speed of movement of the molten substance less than the crystallizing velocity of the substance.

36. A method of crystallizing a substance the molecules of which may, at one temperature, be arranged according to a definite law of regularity into a crystal the molecules of which may, at another temperature, be arranged according to another definite law of regularity, the said method comprising subjecting successive molecular layers of the substance to a temperature at or below the said other temperature, and limiting the speed at which the said successive molecular layers of the substance are subjected to the said other temperature to maintain each of the said successive molecular layers of the substance in condition such that the molecules thereof are arranged according to the first-

named definite law of regularity until after the next preceding molecular layer of the substance has become crystallized according to the said other definite law of regularity.

37. A machine for solidifying a molten substance having, in combination, means for advancing the surface of solidification into the molten substance, means for controlling the speed of advance of the surface so as to afford each successive layer of the substance time to crystallize before the next surface layer commences to crystallize, and means for maintaining the successive portions of the substance in molten condition until the next preceding portion of the substance has become crystallized.

38. A machine for crystallizing a molten substance having, in combination, means for advancing the surface of deposition of the substance, means for limiting the speed of advance of the surface to a value smaller than the crystallizing velocity of the substance, and means for maintaining the successive portions of the substance in molten condition until the next preceding portion of the substance has become crystallized.

39. A machine for solidifying a molten substance having, in combination, means for crystallizing a nucleus of the substance and for advancing the surface of deposition into the substance from the nucleus along adjacent surface layers of the substance that are successively farther and farther removed from the nucleus, means for limiting the speed of advance of the surface to a value low enough to afford each successive surface layer of the substance time to crystallize before the next surface layer commences to crystallize, and means for maintaining the successive portions of the substance in molten condition until the next preceding portion of the substance has become crystallized.

40. A method of purifying an impure molten substance that comprises advancing the surface of deposition into the substance, controlling the speed of advance of the surface of deposition to afford the impurities contained in the substance time to diffuse from the surface of deposition through the substance, and maintaining the successive portions of the substance in molten condition until after the impurities have diffused from the next preceding portion of the substance.

41. A machine for purifying an impure molten substance having, in combination, means for relatively moving the molten substance from a region that is maintained above the freezing temperature of the substance into a region that is maintained at or below the freezing temperature of the substance to advance the surface of deposition of the substance, and means for controlling the speed of relative movement of the substance to maintain the respective successive portions of the substance in molten condition until the impurities contained in the substance have successively diffused away from the said respective portions.

In testimony whereof, I have hereunto subscribed my name.

PERCY W. BRIDGMAN.