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A. HERCZOG ET AL

3,494,745

METHOD OF GROWING SINGLE CRYSTAL IN A HORIZONTALLY DISPOSED ROD

Filed April 6, 1967

2 Sheets-Sheet 1

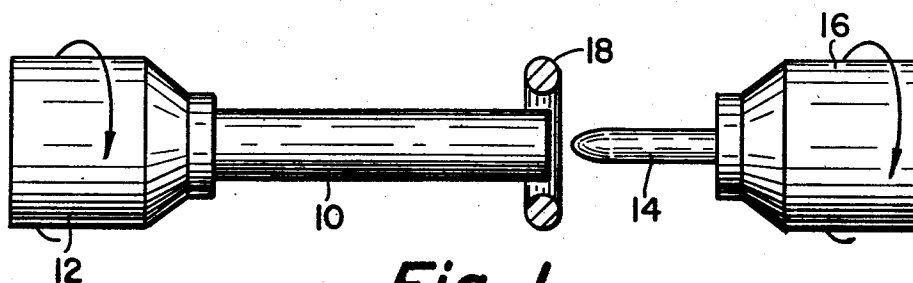


Fig. 1

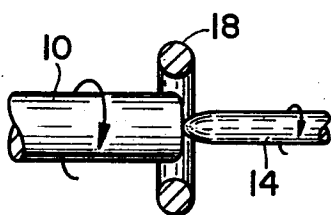


Fig. 2

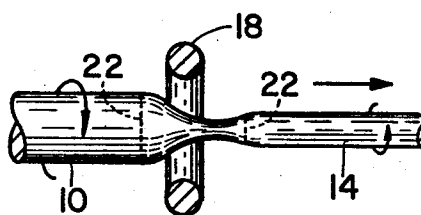


Fig. 5

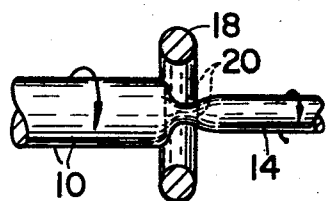


Fig. 3

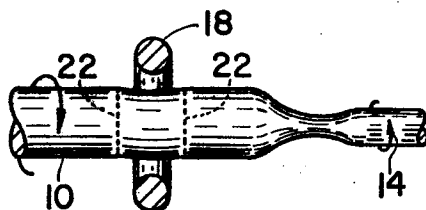


Fig. 6

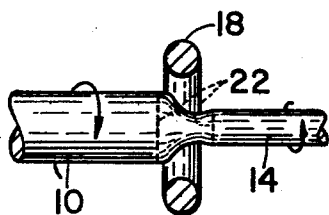


Fig. 4

INVENTORS.
ANDREW HERCZOG
DALE W. RICE
BY *Walter S. Zebrowski*
ATTORNEY

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A. HERCZOG ET AL

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2 Sheets-Sheet 2

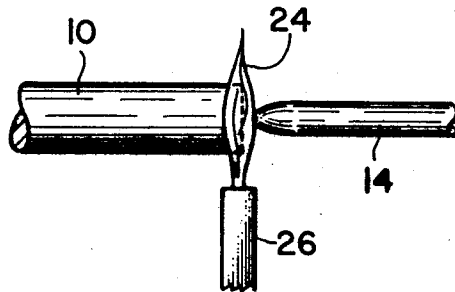


Fig. 7

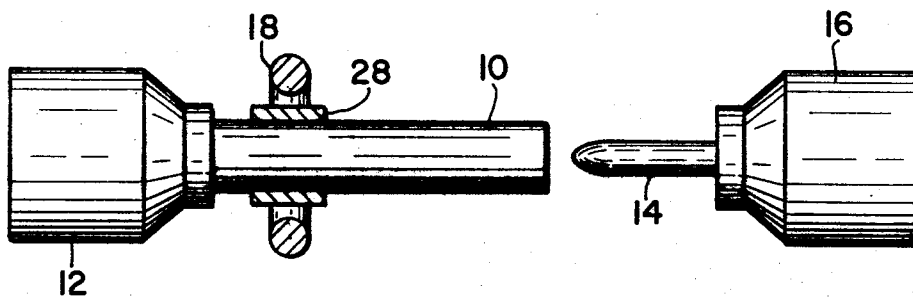


Fig. 8

INVENTORS.
ANDREW HERCZOG
DALE W. RICE
BY
Walter S. Zabrinski
ATTORNEY

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3,494,745

METHOD OF GROWING SINGLE CRYSTAL IN A HORIZONTALLY DISPOSED ROD

Andrew Herczog, Painted Post, and Dale W. Rice, Corning, N.Y., assignors to Corning Glass Works, Corning, N.Y., a corporation of New York

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Int. Cl. B01j 17/10, 17/16

U.S. Cl. 23—301

21 Claims

ABSTRACT OF THE DISCLOSURE

A method for growing single crystals from a congruently melting polycrystalline material is disclosed. A polycrystalline rod is disposed in a horizontal position and a suitably seeded, unsupported fluid zone is formed and caused to traverse at least a portion of the length of the horizontal rod whereby a single crystal is grown within the traversed portion of the rod.

BACKGROUND OF THE INVENTION

Single crystals have been grown in the past by various methods, such as for example immersing a seed crystal in a melt of material and thereafter pulling the seed causing the growth of a single crystal from the melt. Such methods, however, were limited by the use of a crucible to contain the melt.

Another method for growing single crystals involved crucibleless or floating-zone melting of a vertically disposed polycrystalline member. Some of the problems with this method are that the downward flow of the liquid in this zone was presented only by the surface tension of the liquid causing an increase in the diameter of the lower portion of the liquid zone. Such bulging would tend to cause a polycrystalline formation. Furthermore, this condition made control of the crystal diameter very difficult and when the bulging at the bottom of the liquid zone and the thinning at the top of the liquid zone became excessive, the liquid zone would spill over the edges at the bottom. Another disadvantage of this method is that convection currents in the liquid zone would contribute to heat conduction along the longitudinal axis of the zone causing a spreading out of the zone; and enhance radial temperature gradients across the zone. This resulted in highly non-planar solid-liquid interfaces. In addition, such convection currents would cause gas inclusions to be trapped in the center of the liquid zone, and ultimately in the solidified portion, since they would merely tend to rise rather than work themselves out to the surface. Also, in such a vertical method it was difficult to fuse a seed crystal to the end of the molten rod to begin crystal growing. In addition, the liquid zone could, as a practical matter, be caused to traverse the rod in only one direction, upward, since a downward traverse would cause excessive bulging at the bottom of the zone and inevitable spilling thereof. To reduce excessive bulging at the bottom of the zone and thinning at the top, it has been necessary to maintain the zone length within very critical limits when employing this method.

Forming a liquid zone within a horizontal member

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of non-crystalline material, such as glass, for the purpose of crucibleless melting, homogenization, and for bringing about a radial concentration gradient has been known as described in our co-pending application, Ser. No. 397,765, filed on Sept. 21, 1964. However, it has been generally believed that forming such a liquid zone in a crystalline horizontal member was completely unworkable and impractical in that it was believed that an unsupported horizontal liquid zone would collapse and spill out. This belief was maintained because unlike glass, which has a relatively high viscosity at its liquidus and a continuous softening range whereby its viscosity can be easily controlled by the temperature at which it is maintained, most crystalline materials simply have a melting point so that the solid transforms directly into a liquid having a very low viscosity. A liquid zone of certain selected high conductivity crystalline materials could be supported in the horizontal position by means of electro-magnetic suspension. This method, however, requires complex equipment and critical control, and further is limited to high electrical conductivity materials such as metals but is inoperative for low conductivity materials such as alumina, silicon, germanium, and the like.

SUMMARY OF THE INVENTION

The objects of the present invention are to provide an crystals from a polycrystalline rod which overcomes the heretofore noted disadvantages and which provides good dimensional stability and eliminates entrapment of gas inclusions while permitting simplified seeding of the polycrystalline material and traversing of the liquid zone in both directions.

Broadly, according to the present invention, a single crystal may be grown by providing a rod of congruently melting polycrystalline material and a seed crystal, horizontally rotating both said rod and said seed crystal about the respective longitudinal axis thereof with the axes in register with one another, heating one end each of both said rod and said seed crystal until they become molten, contacting the molten end of the rod with the molten end of the seed crystal, heating the juncture of the rod and the seed crystal to establish an unsupported fluid zone, and thereafter traversing the fluid zone through at least a portion of the length of said rod.

Additional objects, features, and advantages of the present invention will become apparent, to those skilled in the art, from the following detailed description and the attached drawings, on which, by way of example, only the preferred embodiments of this invention are illustrated.

BRIEF DESCRIPTION OF THE DRAWINGS

FIGURE 1 is a side elevation of an apparatus illustrating heating one end each of a polycrystalline rod and seed crystal.

FIGURE 2 is a fragmentary side elevation illustrating the molten ends of the crystalline rod and seed crystal being brought together.

FIGURE 3 is a fragmentary side elevation wherein the fluid zone is started.

FIGURE 4 is a fragmentary side elevation illustrating the completed fluid zone and the reversal of rotation of the seed crystal.

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FIGURE 5 is a fragmentary side elevation illustrating the initial mechanical pulling of the seed.

FIGURE 6 is a fragmentary side elevation illustrating the fluid zone traversed through a portion of the rod.

FIGURE 7 is a fragmentary side elevation illustrating another means of heating.

FIGURE 8 is a side elevation of an apparatus of the present invention illustrating still another means for heating.

DETAILED DESCRIPTION

By the term congruently melting material as used herein is meant a material which has a regular phase transformation from solid to liquid and the material does not undergo a change in composition in transforming from a solid to a liquid phase.

By the term polycrystalline material as used herein is meant a crystalline material which is not a single crystal but consists of a plurality of randomly oriented single crystallites or small crystals.

In accordance with this invention, a polycrystalline material rod 10 is illustrated in FIGURE 1 as suitably mounted in chuck 12. A seed crystal 14 is shown suitably mounted in chuck 16. Chucks 12 and 16 are arranged so that the longitudinal axis of each are in register with the other. An induction heating coil 18 is shown disposed about the unsupported ends of both rod 10 and seed 14.

Rod 10 is formed of any congruently melting material, that is a material having a regular phase transformation from solid to liquid and which does not change in composition with a change in phase. Seed 14 is a single crystal of the same material from which rod 10 is formed. Examples of suitable congruently melting polycrystalline materials are elemental metals such as nickel, iron, platinum, copper, aluminum, and the like, as are crystalline non-metals such as silicon, germanium, and the like. Compounds such as oxides, intermetallic compounds, chalcogenides, halides, salts in general, and crystalline organic compounds are also suitable materials. The present invention, however, is not limited to these materials.

As energy is applied to coil 18 to heat the unsupported ends of rod 10 and seed 14, the rod and seed are caused to rotate about their longitudinal axis in the same direction, as illustrated by the arrows adjacent the chucks. When the ends of the rod and seed become melted, they are brought together and caused to contact as illustrated in FIGURE 2. Further heating causes a fluid zone to be started as illustrated by dotted lines 20 in FIGURE 3. Continued heating causes a fully developed fluid zone to be formed as illustrated by lines 22 of FIGURE 4. At this point, the rotation of seed 14 with respect to rod 10 is reversed so that the seed is rotating in the direction opposite to that of rod 10 as illustrated by the arrows in the drawing. The purpose for the counter rotation of the elements is to obtain better mixing and homogenization of the molten material, as well as to lessen radial temperature gradients hence making the growing crystal-liquid interface more nearly planar.

At this point a single crystal may be grown by traversing the fluid zone along the length of rod 10, however, for improved quality of crystal it has been found that pulling the seed away from the rod as illustrated in FIGURE 5 is desirable. As is illustrated in the drawing, such pulling causes the fluid zone to neck down somewhat. Thereafter, the mechanical pulling is slowly decreased to allow the crystal to increase in diameter to that of the rod. As shown in FIGURE 6, when coil 18 is translated along the longitudinal axis about the periphery of rod 10, the fluid zone is traversed along the length of rod 10 whereby a single crystal which was nucleated by seed 14 is caused to grow.

Any of a variety of types of heating may be employed to produce the fluid zone of the present invention. For example, electric resistance, induction, flame, electron beam, radiation, and the like may be used. Referring to

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FIGURE 7, a flame heating means is illustrated whereby the unsupported ends of rod 10 and seed 14 are heated by means of flame 24 emitted from burner 26. Such heating means may be employed in connection with the formation of a fluid zone within a material that is not oxidizable since oxidizable materials would of necessity have a fluid zone formed therein within a vacuum or a non-oxidizing atmosphere such as an inert or reducing gas, and flame heating cannot be employed within a vacuum or such atmosphere as a practical matter. Flame heating is suitable for such materials as aluminum oxide, platinum, and the like.

Certain high electrical resistivity materials such as pure silicon and gallium phosphide are poor susceptors of inductive energy at relatively low temperatures, that is temperatures of below about 800° C., while at higher temperatures they are good susceptors of inductive energy. A fluid zone may be formed in such materials through induction heating by employing the means illustrated in FIGURE 8. A band, ring, or collar 28 of material which is a good susceptor of inductive energy is closely disposed about rod 10 at a point along the length thereof beyond that through which the fluid zone will ultimately be traversed. Suitable materials for ring 28 are molybdenum, tantalum, tungsten, and the like. In this embodiment, induction heating coil 18 is disposed about ring 28 to begin the heating process. As electrical energy is dissipated within coil 18, ring 28 is inductively heated and in turn heats rod 10 in the area adjacent ring 28 by conduction and radiation. It is seen, therefore, that in this manner rod 10 is indirectly heated by induction heating. When the temperature of rod 10 in the proximity of coil 18 reaches a temperature at which the material of rod 10 becomes a susceptor of inductive energy, coil 18 may then be traversed along the length of rod 10 continuously heating the advancing portion thereof by conduction until the unsupported end of rod 10 is reached. At this point, further inductive energy may be dissipated in the unsupported end of rod 10 until it becomes molten, whereupon, it is joined to the molten end of seed 14 and a fluid zone is established as heretofore explained. The unsupported end of seed crystal 14 is heated by radiation from the heated end of rod 10 to a temperature sufficient for direct electromagnetic coupling, and thereafter heated by inductive energy to a molten state.

The primary process conditions to be considered are the absolute and relative rate of rotation of the rod and seed crystal, zone length, zone diameter, travel rate of the zone, seed pull rate, and the like. These parameters are directly affected by the polycrystalline material employed and more specifically its density, surface tension, thermal conductivity, and the like as well as the thermal convection within the fluid zone.

The rotation rate is governed in part by the rod diameter and zone length, as well as the polycrystalline material density, surface tension, and thermal conductivity. For example, the viscous forces acting on the fluid portion of the rod and seed resulting from a high rotation rate will permit a longer fluid zone for a material of given density and surface tension. The rotation rate of each of the rod and seed may suitably range from about 50 r.p.m. to about 1000 r.p.m. or higher. Relative rotation rates will be twice these values if counter rotation is employed, that is having the direction of rotation of the seed opposite or reverse with respect to the direction of rotation of the rod. However, the present invention is not limited to such rotation rates.

The travel rate of the fluid zone may satisfactorily range from about 0.1 millimeter per minute or less to about 20 millimeters per minute or more for many polycrystalline materials. This travel rate is determined by the diameter of the rod and physical properties of the material.

The fluid zone diameter and length is affected by the polycrystalline material density, surface tension, and ther-

mal conductivity as well as the rate of travel of the zone and the relative rate of rotation of the rod and seed. The zone must be sufficiently controlled so that the force of gravity does not cause it to spill. The diameter of the zone is directly affected by the density and surface tension of the material as well as rate of rotation of the rod and seed crystal. If the zone is subjected to a pull, the pull rate and distance of the pull will also directly affect the diameter of the fluid zone. The length of the fluid zone must be limited to the extent necessary in order that the zone remains physically stable. If the zone is too long, excessive deformation due to centrifugal force or complete spillage of the zone may result. The limits of length within which the zone will remain physically stable depends upon the density and surface tension of the material, the diameter of the rod, the relative rates of rotation, the travel rate of the zone, and the pull rate. As a result, considerable variation in zone length is possible depending on these conditions. Ordinarily, ratios of zone length to rod diameter of between 2:1 and 1:4 are suitable although these ratios are not intended to be limitations upon the scope of this invention.

The pull rate which permits improved crystal growth and crystal quality, again depends on the polycrystalline material involved since an excessive pull rate could increase the length of the fluid zone causing it to deform or spill.

By growing a single crystal with the fluid zone in the horizontal position, thermal convection within the fluid zone permits a uniform radial temperature distribution and assists in causing any gaseous inclusions to be worked out to the surface. For a given temperature, thermal convection would be an inherent property of the material involved. In a horizontal zone thermal convection will tend to maintain planar or less curved solid-liquid interfaces between the fluid zone and the adjacent solidified portion of the rod.

To illustrate the invention and the manner in which it may be practiced, the following examples are provided.

EXAMPLE I

A polycrystalline rod of silicon having a diameter of $\frac{1}{2}$ inch and a seed crystal having a diameter of $\frac{1}{4}$ inch were provided and second within a pair of opposing chucks in a manner such that the longitudinal axes thereof were in register. A ring of tantalum material was disposed around the rod of silicon near its supported end. An induction heating coil was then disposed surrounding the tantalum ring and electrical energy was dissipated therein. The tantalum ring was a suitable susceptor of inductive energy and was thereby heated by the coil. The portions of the silicon rod adjacent the ring were in turn heated by conduction and radiation from the ring. When the rod reached the temperature of about 800° C. it became a susceptor for inductive energy itself and the coil was then traversed toward the unsupported end of the rod at such a rate as to keep the temperature of the rod adjacent the coil sufficiently high so that it would continue to be a susceptor.

When the coil was traversed to the unsupported end of the rod, the unsupported end of the seed crystal was maintained in close proximity to the heated end of the silicon rod so that the seed crystal temperature was raised by radiation.

At this point, the rod and seed crystal were rotated in synchronous rotation at about 200 r.p.m., then heated to at least 1410° C., the melting point of silicon, and the melted ends were brought together and caused to make contact. The induction coil was traversed along a portion of the length of the rod at a rate of about 2 millimeters per minute to establish a well developed fluid zone and the rotation of the seed crystal and rod was then changed to counter rotation, each rotating at about 250 r.p.m. thereby having a relative rotation of one to the other of about 500 r.p.m.

To improve the quality of the crystal, a mechanical pulling of the seed was started and the pull rate was gradually increased to about 19 millimeters per minute yielding a crystal growth rate of about 21 millimeters per minute through a distance of about 40 to 50 millimeters. The pull rate was then slowly decreased to zero allowing the crystal which was being grown to increase its diameter to that of the rod. The length of the fluid zone was about 1 centimeter.

The growth of the crystal was then continued at the rod diameter of $\frac{1}{2}$ inch at a rate of about 2 millimeters per minute to the desired length of about 8 inches, whereupon the grown crystal was separated or pulled away from the polycrystalline rod, the electrical energy to the coil was terminated, and the two ends permitted to solidify.

This method resulted in a high quality, single crystal of silicon having a uniform diameter for a significant and commercially usable length.

Although the energy supplied to an induction coil may be by various means well known in the art, energy was supplied to the coil in the above example by means of a commercial 15 kva., 4 megahertz power generator, the output of which was controlled by a feedback circuit. Electrical energy of about 1500 watts was dissipated within the coil while the fluid zone was traversed along the length of the rod.

EXAMPLE II

Another example of the present invention is illustrated as follows. A seed crystal and a 12 millimeter diameter germanium rod are provided and mounted in the horizontal position as heretofore described. Both the rod and seed crystal have ends tapered over a 50 millimeter length to a 4 millimeter diameter. The rod and crystal are heated in the manner described in connection with Example I. After the rod and seed are joined a fluid zone is established having a length of about 10 millimeters. The relative counter rotation between the rod and the seed is about 500 r.p.m. With a zone rate of travel of 0.8 millimeter per minute, a single crystal is grown in the germanium rod.

The induction heating in this example would be accomplished by employing a concentrator well known in the art, which allows the use of a work coil of a single turn, capable of transmitting power to the object. As in Example I, a 15 kva., 4 megahertz power generator may be used for supplying energy to the coil.

EXAMPLE III

In another example of the present invention a $\frac{1}{4}$ inch diameter rod of nickel and a suitable seed crystal are provided and mounted in a horizontal position as heretofore described. Since nickel is a suitable susceptor of inductive energy the rod and crystal is heated directly at the unsupported end. The relative counter rotation between the rod and seed crystal is about 500 r.p.m. After the unsupported ends of the rod and seed crystal are melted and joined, a fluid zone is formed having a length of about 10 millimeters. A single crystal is grown with a fluid zone rate of travel of 0.8 millimeter per minute. The induction heating in this example is also accomplished with a concentrator and work coil as described in Example II.

EXAMPLE IV

A further example is illustrated by providing a $\frac{3}{16}$ inch diameter iron rod and seed crystal, and mounting them in a horizontal position as heretofore described. A fluid zone is formed and traversed in the same manner as described in connection with Example III, except that the relative counter rotation between the rod and seed crystal is 500 r.p.m. A single crystal of iron is grown in the manner described in Example III.

EXAMPLE V

In a further example of this invention a 5 millimeter diameter rod of platinum and a seed crystal is provided and mounted as heretofore described. In this example a pair of oxyhydrogen burners are disposed so that the flame therefrom is directed to the unsupported ends of the rod and seed crystal. After the unsupported ends became molten they were joined as heretofore described at a relative counter rotation of 400 r.p.m. A fluid zone of approximately 10 millimeters in length is established and traversed along the length of the rod at a rate of 4.0 millimeters per minute, whereby a suitable single crystal is grown within the rod.

Although the present invention has been described with respect to specific details of certain embodiments thereof, it is not intended that such details be limitations upon the scope of the invention except insofar as set forth in the following claims.

We claim:

1. A method of growing a single crystal of congruently melting material comprising the steps of
 providing a rod of polycrystalline material,
 providing a seed crystal,
 rotating said rod horizontally about its longitudinal axis,
 rotating said seed crystal about its longitudinal axis arranged horizontally in register with the axis of said rod,
 disposing one end of said seed crystal adjacent one end of said rod,
 heating the adjacent ends of said rod and seed crystal until the entire cross section of the ends become molten,
 contacting the molten end of said rod with the molten end of said seed crystal.
 heating the juncture of said rod and seed crystal to establish an unsupported fluid zone,
 traversing said unsupported fluid zone through at least a portion of the length of said rod whereby a single crystal is grown within the traversed portion of said rod.

2. The method of claim 1 further comprising the step of mechanically pulling said seed away from said rod after said unsupported fluid zone has been established for a predetermined period of time, and thereafter terminating such pulling.

3. The method of claim 1 further comprising the step of rotating said seed crystal in a direction opposite to that of said rod after a fluid zone has been established.

4. The method of claim 1 wherein said heating is accomplished by dissipating inductive energy within said rod and seed crystal.

5. The method of claim 1 wherein the material of said rod and said seed crystal is silicon.

6. The method of claim 5 further comprising the step of rotating said seed crystal in a direction opposite to that of said rod after said unsupported fluid zone has been established.

7. The method of claim 6 further comprising the step of mechanically pulling said seed away from said rod after said unsupported fluid zone has been established for a predetermined period of time, and thereafter terminating such pulling.

8. The method of claim 7 further comprising the steps of

disposing a ring formed of material suitable as a susceptor of inductive energy about said rod along the length thereof, and
 thereafter heating said rod by dissipating inductive energy within said ring until said rod reaches a temperature at which it becomes a susceptor of inductive energy.

9. The method of claim 3 further comprising the step of mechanically pulling said seed away from said rod

after said unsupported fluid zone has been established for a predetermined period of time, and thereafter terminating such pulling.

10. The method of claim 1 wherein the material of said rod is selected from the group consisting essentially of nickel, platinum, germanium, and aluminum oxide.

11. The method of claim 1 wherein said rod and seed crystal is heated by means of a flame.

12. The method of claim 10 further comprising the step of rotating said seed crystal in a direction opposite to that of said rod after said unsupported fluid zone has been established.

13. The method of claim 10 wherein said rod and seed crystal is heated by means of a flame.

14. A method of growing a single crystal of silicon comprising the steps of

providing a rod of polycrystalline silicon,
 providing a silicon seed crystal,
 supporting said rod at one end in a horizontal position,
 supporting said seed crystal at one end in a horizontal position with the longitudinal axis thereof in register with the axis of said rod,
 synchronously rotating said rod and said seed crystal,
 disposing the unsupported ends of said rod and said seed crystal adjacent one another,
 heating the unsupported ends of said rod in said seed crystal until they become molten,
 contacting the molten end of said rod with the molten end of said seed crystal,
 heating the juncture of said rod and said seed crystal to establish an unsupported fluid zone,
 traversing said unsupported fluid zone through at least a portion of the length of said rod thereby growing a single crystal of silicon within the traversed portion of said rod.

15. The method of claim 14 wherein said rod and said seed crystal are heated by means of inductive energy.

16. The method of claim 15 further comprising the steps of

disposing a ring formed of material suitable as a susceptor of inductive energy about said rod along the length thereof, and
 thereafter heating said rod by dissipating inductive energy within said ring until said rod reaches a temperature at which it becomes a susceptor of inductive energy.

17. The method of claim 15 further comprising the step of mechanically pulling said seed away from said rod after said unsupported fluid zone has been established for a predetermined period of time, and thereafter terminating such pulling.

18. The method of claim 17 further comprising the step of rotating said seed crystal in a direction opposite to that of said rod after said unsupported fluid zone has been established.

19. The method of claim 18 wherein said single crystal is grown within a vacuum.

20. A method of growing a single crystal of silicon comprising the steps of

providing a rod of polycrystalline silicon,
 providing a silicon seed crystal,
 supporting one end of said rod in a horizontal position,
 supporting said seed crystal about one end thereof in a horizontal position with its longitudinal axis in register with the longitudinal axis of said rod,
 synchronously rotating said rod and said seed crystal with the unsupported end of said seed crystal adjacent the unsupported end of said rod,
 heating the unsupported ends of said rod and said seed crystal until they become molten,
 contacting the molten end of said rod with the molten end of said seed crystal,
 heating the juncture of said rod and said seed crystal to establish an unsupported fluid zone,

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reversing the direction of rotation of said rod and said seed crystal with respect to one another, traversing said unsupported fluid zone through at least a portion of the length of said rod thereby growing a single crystal of silicon within the traversed portion of said rod, 5
pulling said seed crystal away from said rod, and slowly decreasing the pull rate until it is stopped there- after allowing the diameter of said single crystal to increase to the diameter of said rod.
21. The method of claim 20 wherein the relative ro- 10 tation of the rod and the crystal seed is 500 r.p.m. and the rate at which said unsupported fluid zone is traversed along the length of said rod is about 2 millimeters per minute. 15 148—1.6

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10 NORMAN YUDKOFF, Primary Examiner
R. T. FOSTER, Assistant Examiner

U.S. Cl. X.R.

UNITED STATES PATENT OFFICE
CERTIFICATE OF CORRECTION

Patent No. 3,494,745 Dated February 10, 1970

Inventor(s) Andrew Herczog and Dale W. Rice

It is certified that error appears in the above-identified patent and that said Letters Patent are hereby corrected as shown below:

Column 2, line 26, after "an" insert --economic method and apparatus for forming single--

Column 2, line 32, "materal" should read --material--

Column 5, line 44, "second" should read --secured--

Column 5, line 54, "It" should read --it--

Column 6, line 58, "500 r.p.m." should read --550 r.p.m.--

Column 7, line 44, "puling" should read --pulling--

SIGNED AND
SEALED

JUL 28 1970

(SEAL)

Attest:

Edward M. Fletcher, Jr.

Attesting Officer

WILLIAM E. SCHUYLER, JR.
Commissioner of Patents