Dec. 2, 1969 MITSUHIRO MARUYAMA 3,481,711

CRYSTAL GROWTH APPARATUS

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Fig. 1.

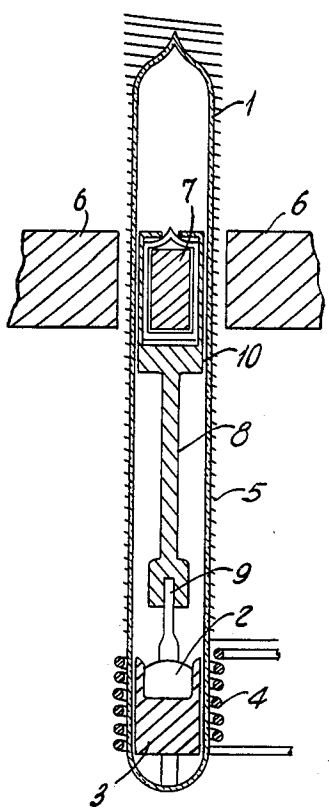
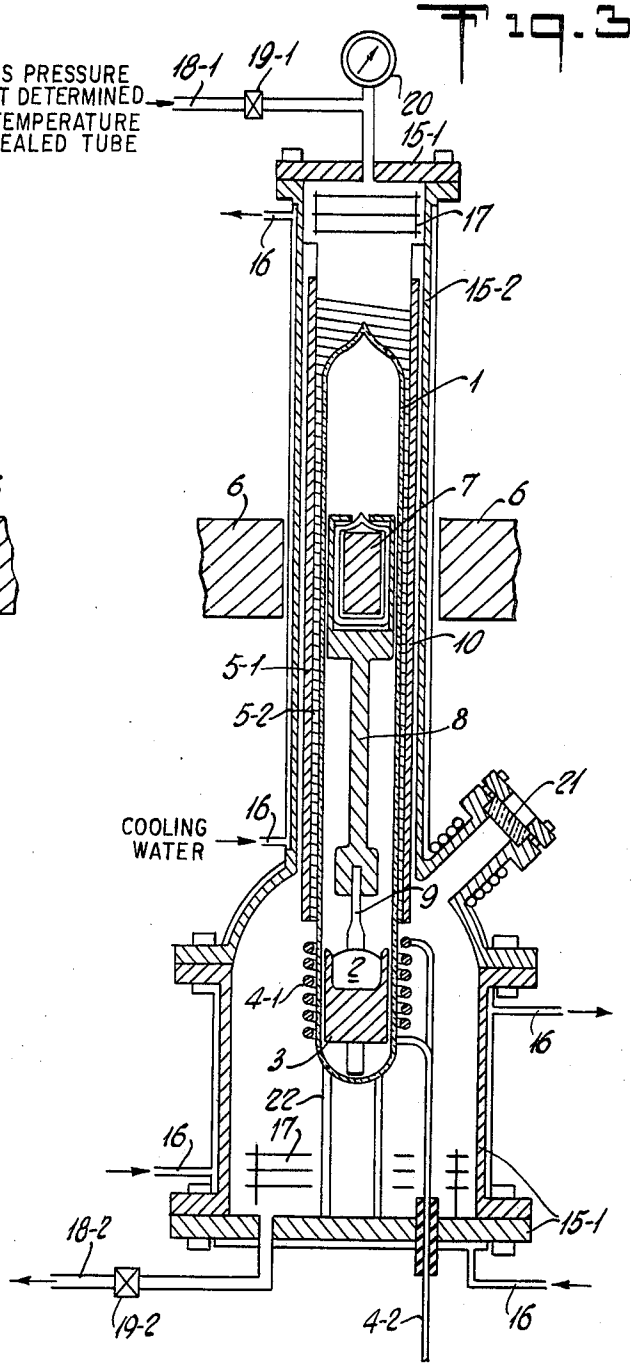


Fig. 3.



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Fig. 2.

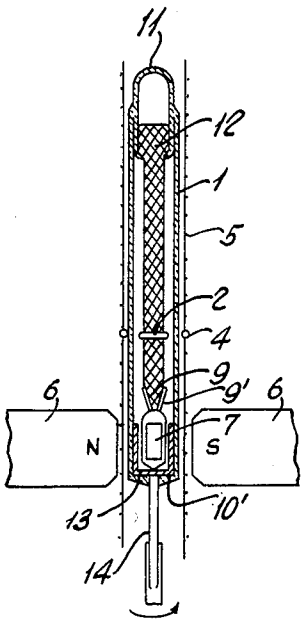
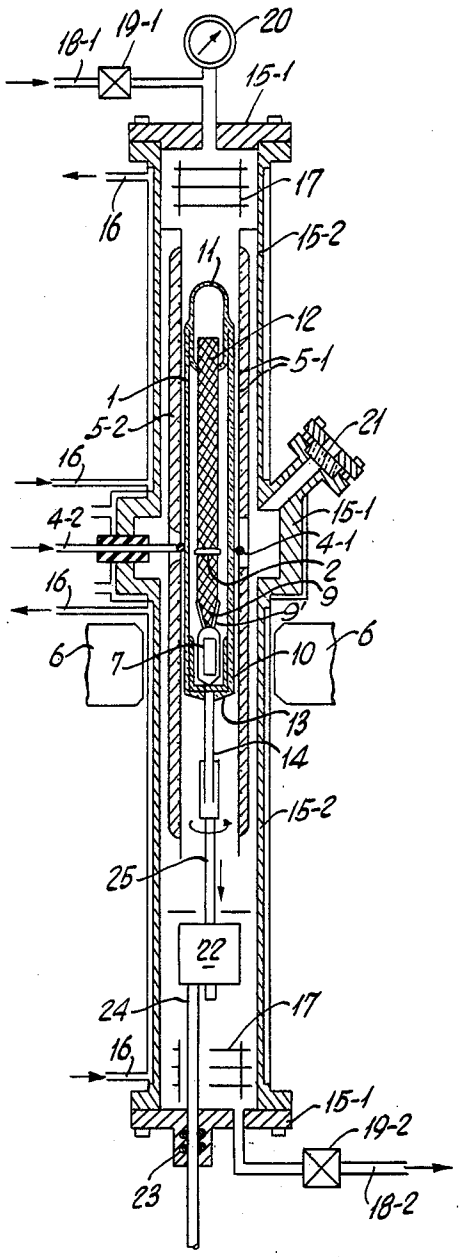


Fig. 4.



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Fig. 9A.

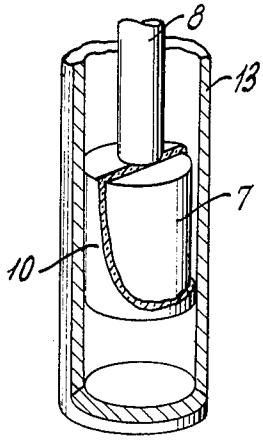


Fig. 9B.

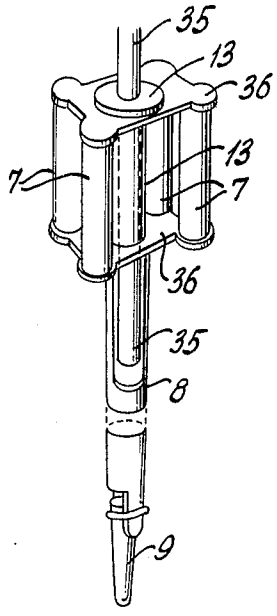
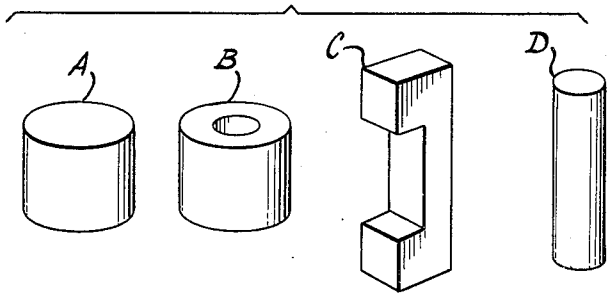


Fig. 10.



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FIG. 5.

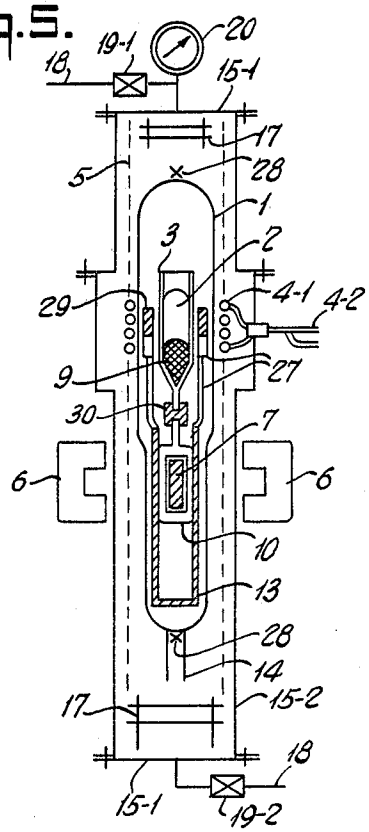
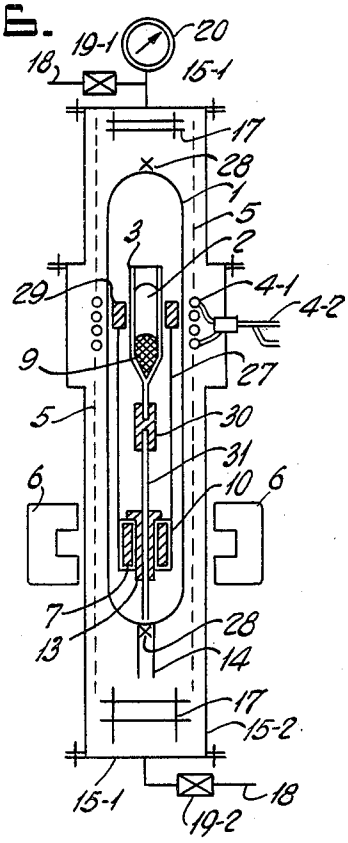


FIG. 6.



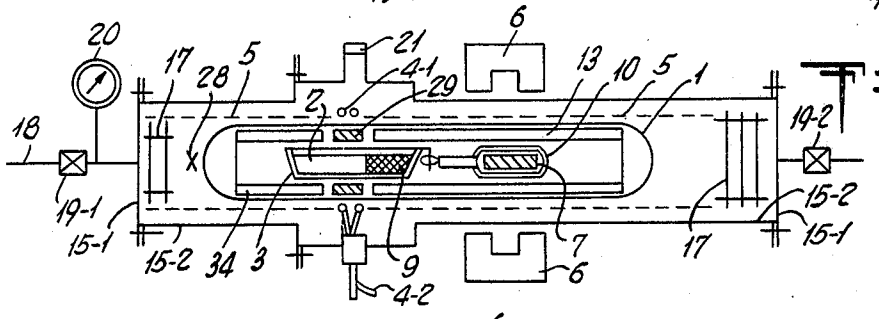


FIG. 7.

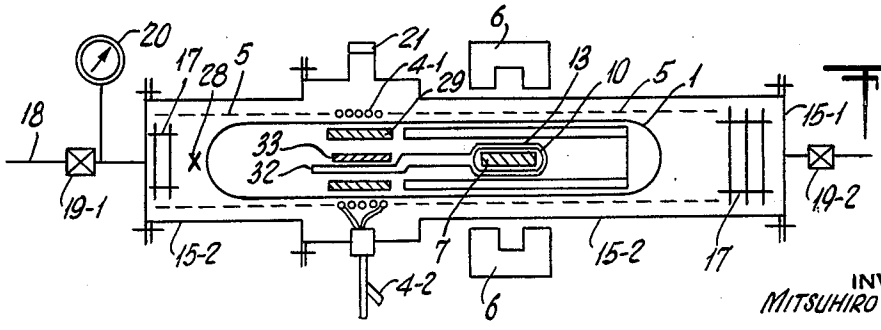


FIG. 8.

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CRYSTAL GROWTH APPARATUS

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Claims priority, application Japan, Aug. 4, 1964, 39/44,528
Int. Cl. B01j 17/18
U.S. Cl. 23—273 8 Claims

ABSTRACT OF THE DISCLOSURE

Apparatus and method for growing crystals which includes placing a sealed tube containing the crystal raw material into an outer pressure chamber, applying pressure to the pressure chamber to reduce the outward pressure on the tube walls during crystal growth, varying the outer chamber pressure in response to changes in the magnitude of the pressure within the sealed tube to maintain the outward differential pressure on the walls of the sealed tube within a predetermined range, to thereby improve the crystal and prevent tube explosions.

This invention relates to the manufacture of crystals.

It is an object of this invention to provide improved apparatus for crystal manufacture and purification.

All of the objects, features and advantages of this invention and the manner of attaining them will become more apparent and the invention itself will be best understood by reference to the following description of an embodiment of the invention taken in conjunction with the accompanying drawing, in which:

FIGS. 1 and 2 show longitudinal cross sectional views of conventional crystal growth apparatus according to the Czochralski and the Bridgeman techniques, respectively,

FIGS. 3 and 4 show longitudinal cross sectional views of crystal growth apparatus for use according to the Czochralski and the floating zone techniques respectively, which have been modified in accordance with the teachings of this invention,

FIGS. 5 and 6 show longitudinal cross sectional views respectively, of a conventional vertical type Bridgeman crystal growth apparatus and a modification thereof in accordance with the teachings of this invention,

FIGS. 7 and 8 show longitudinal cross sectional views respectively, of a horizontal type Bridgeman crystal growth apparatus and horizontal type crystal processing apparatus which have been modified in accordance with the teachings of this invention,

FIGS. 9A and 9B show schematic diagrams illustrating two different embodiments of the movable element, in partial section and in perspective view respectively, for use as an integral part of the apparatus according to this invention, and

FIG. 10 shows a variety of geometrical configurations that may be employed for the ferromagnetic piece as a component part of the movable element.

Crystal growth and purification have hitherto been effected by various methods including the Bridgeman method, the temperature gradient method, the Czochralski method, the zone melting method, the floating zone method and by modifications and combinations thereof.

According to these methods, crystals are allowed to grow or are purified usually in a controlled atmosphere at a pressure of from about 1 to 5 atmospheres. Compounds, for example those of Groups III–V and II–VI of the Periodic Table, which contain constituents having high vapor pressures and which are easily vaporized, e.g. phosphorus, arsenic, oxygen, sulfur, selenium, tellurium, zinc, and nitrogen, are so easily decomposed and are so reactive that the methods referred to above can barely crystallize the compounds with stoichiometric compositions. To overcome this difficulty, it has generally been the practice to seed the materials, such as for example, quartz in bar or wafer single crystal form, in sealed tubes, and heat the tubes to a suitable temperature to cause crystal growth by the temperature gradient method, the Bridgeman method, or by coupling the supporters of seed crystal inside the tubes magnetically from the outside. FIG. 1 shows a conventional crystal growth apparatus devised by Gremmelmaier (Zeitschrift für Naturforschung, 11A, S11–13 (1956)), which is also known as the low-pressure magnetic coupling Czochralski type crystal growth apparatus. Improvements have since been made on the apparatus by Weisberg and other workers.

The apparatus of FIG. 1 includes a sealed tube of quartz **1**, the upper end portion (low-temperature region) of which is kept at about 605° C. (arsensic vapor pressure of 0.9 atmosphere). A molten metal **2**, e.g. GaAs is maintained at about 1237° C. as the graphite crucible **3** is heated by a high-frequency coil **4**. A seed crystal **9** is supported by a holder **8**. On top of the holder **8**, a magnetic member **7**, e.g. of Permendur, covered by quartz, is fitted, which can be vertically moved or revolved by the magnetic force of the magnet **6**. The numeral **10** indicates a graphite guide precisely finished in order to provide smooth motion of the magnetic member **7** and the holder **8**. A heater **5** is so adjusted as to maintain the minimum temperature inside the sealed tube **1** at 605° C. As the seed crystal **9** is urged to rotate and moved vertically by the magnet **6**, the molten metal GaAs **2** in contact therewith is cooled by the seed crystal **9** and reduced to a single crystal. A single crystal is thus grown. According to this method, the procedure may be reversed by fixing the seed crystal and moving the crucible with respect to the crystal (vid. Miller et al., Fall Meeting of the Electrochemical Society, Houston, Tex., October 1960).

FIG. 2 shows an improvement of the ordinary low-pressure floating refining type apparatus devised by Cunnell et al. (vid. Solid State Electronics, 1, 97 (1960)). The improvement was made by W. P. Allred et al. (vid. "Compound Semiconductors," vol. 1, Preparation of III–V Compounds, p. 271, New York, Reinhold Publishing Corporation (1962)).

The apparatus is operated in such a manner that the sealed quartz tube **1** is equipped with a heater **5** and the temperature is controlled so that the minimum temperature is kept at 605° C. A polycrystal ingot **12**, e.g. of GaAs, is supported by a quartz holder **11**, and a seed crystal **9** is supported by a quartz holder **9'**, which in turn is connected to a quartz jacket **10'** enclosing a Permendur member **7**. The jacket **10'** and the graphite **13** are precisely finished and the graphite **13** serves as a guide. The Permendur member **7** is magnetically coupled to the magnet **6**, and the seed **9** can be revolved and moved vertically by maneuvering the magnet **6**. On the interface between the ingot **12** and the seed **9**, a high frequency wave from a high-frequency coil **4** is applied, causing fusion, with the result that a fused zone **2** is maintained by surface tension. The width of this fused zone is controlled by suitably adjusting the high-frequency output and the position of the magnet **6**. The sealed tube **1** is held in place by a quartz bar **14**, which is interlocked with the magnet **6**.

With such arrangement, the ingot inside the sealed tube is moved relative to the coil **4** by means of the quartz supporter **14**, whereby floating zone purification and single crystallization are carried out.

As described hereinbefore, the ordinary methods depend on magnetic coupling of movable elements inside

sealed tubes for crystal growth, and, in order to satisfy the conditions that the inside of the sealed tubes should be visible from the outside, and that the tubes should be prevented from contamination as far as possible, quartz of high purity is usually employed as the material for the sealed tubes. Since quartz has a high melting point and hence poor weldability and workability, the quartz tubes employed are usually from 10 to 50 mm. in diameter and from about 1 to 3 mm. in wall thickness. For that reason, the tubes have poor pressure resistance at elevated temperatures, in most cases ranging from 5 to 10 atmospheres. However, compounds containing phosphorus, sulfur, or the like often have a stoichiometric equilibrium pressure of more than 10 atmospheres, and the conventional methods when applied to those compounds are hazardous and involve much difficulty in operation because they are likely to cause explosions.

The present invention provides an apparatus which obviates all the foregoing difficulties and has far broader applications. In brief, the method according to the invention consists of placing a heater and sealed tube in a pressure chamber (autoclave), applying a pressure between the sealed tube and the pressure chamber for balancing with the pressure inside the sealed tube, thereby to decrease the pressure difference applicable to the walls of the sealed tube to a degree below the withstand pressure of the tube walls, and at the same time effecting magnetic coupling from the outside of the pressure chamber to drive the movable element having a seed inside the sealed tube in order thereby to effect crystal growth.

In FIG. 3 there is shown an example of the high-pressure magnetic-coupling Czochralski type crystal growth apparatus according to the present invention. The numerals **1** to **10** denote like parts as in FIG. 1. The designation **4–1** indicates a high-frequency coil, and **4–2** a water-cooled inlet terminal for high-frequency current. **5–1** is a heater, **5–2** is a heat-insulating material, e.g. asbestos or quartz sheet, **15–1** and **15–2** are pressure metal chambers or autoclaves, **15–2** being formed of a less magnetic material, e.g. stainless steel, and both of the component parts of the autoclave are cooled with water. Feeding and discharging ports for cooling water are indicated generally at **16**. Numeral **17** denotes a heat insulating board, e.g. of stainless steel, molybdenum, or steatite, **18–1** a gas inlet port, **18–2** a gas discharge port, **19–1** a gas inlet valve, **19–2** a gas discharge valve, **20** a pressure gauge, **21** a transparent quartz window, and **22** a sealed tube base of a heat insulating material.

The apparatus is operated in the following way. An electric power is applied to the heater **5–1**, the vapor pressure of the vaporizable material inside the sealed tube is increased. By opening the valve **19–1** in response to an increase of said vapor pressure, a suitable atmospheric gas, e.g. nitrogen or argon, kept at a pressure equivalent to said vapor pressure is fed into the chamber through the inlet port **18–1** which is connected to a suitable compressor or reducing valve, not shown, for pressure control. As the pressure outside the sealed tube is equalized to the pressure inside, the pressure difference is reduced below the withstand pressure of the sealed tube. As a result, a much higher pressure is produced inside the tube, which has previously been impracticable. At the same time, the magnetic force from the magnet placed outside the chamber is directly coupled to the core of Permendur (an alloy having a high Curie point) inside the tube, because the magnetism of the pressure chamber is negligible. If the pressure inside the pressure chamber rises excessively due to temperature variation inside, the gas is discharged through the gas discharge valve **19–2** and the gas discharge port **18–2**, thereby to reduce the internal pressure to a predetermined level. This may be accomplished, for example, by a discharge valve which is not shown. To be more precise, a reducing valve and a discharge valve are combined, for example, to regulate the external pressure of the sealed tube in proportion to

the variation (program) of the internal pressure (as determined by temperature) of the tube, so that the pressure conditions which satisfy a certain stoichiometric composition are maintained inside the tube.

Under the above conditions, a float, composed of the members **7**, **8**, and **9** holding a seed crystal, is magnetically coupled with the magnet **6** for rotation and vertical motion, with oscillations if desired, until a single crystal is allowed to grow out of the solution **2** by the Czochralski method, in the same manner as in the case of FIG. 1.

Description will now be made of several embodiments of the mehod of effecting the growth of an indium phosphide (InP) single crystal by means of the apparatus shown in FIG. 3.

EMBODIMENT 1

Referring now specifically to FIG. 3, a transparent quartz tube 2 mm. in wall thickness and 450 mm. in length is used as the evacuated envelope **1** in which is enclosed argon gas at about 100 mm. of Hg together with an excess of phosphor. Crucible **3** made of high-purity carbon is supported within tube **1** and the crucible temperature is controlled with the aid of an optical pyrometer. Where an external crucible type structure differing from that illustrated is employed, the bottom part of the tube **1** will constitute the inner surface of the crucible, enabling temperature control with the aid of a thermocouple. By heating the tube with RF heater **3** and heater **5–1**, the excess of phosphor within the tube is vaporized, causing the tube pressure to increase. With a pressure equal to 5 atmospheres maintained in the high pressure chamber through gas inlet **18–1** prior to the heating process, the gas pressure within the chamber should be controlled so that the difference between the pressures exerted on the opposite sides of the tube walls may be within a pressure of 5 atmospheres upon exceeding a tube temperature of the order of 450°–480° C.

When the minimum temperature of 550° C. in the tube is reached (corresponding to internal gas pressure of about 20 atm.), fine temperature control must be conducted by heater **5–1**. If the crucible temperature is further increased under this condition, the polycrystalline material in the crucible initiates melting at a temperature between 1065° C. and 1070° C.

Upon reaching a temperature-and-pressure equilibrium state within the apparatus after the material has been perfectly melted, holder **8** is lowered by magnet **6** so that the tip end of the seed crystal **9** may be brought in contact with molten indium phosphide **2**. By viewing the interior through window **21**, the seed is then dipped into the melt and after wetting it is slowly raised to see that the wetted state is suitable, and then magnet **6** is rotated and pulled upward. Then the seed crystal **9** is moved upward by maintaining rotational motion.

By lowering the melt temeprature, the growth of an InP single crystal takes place from the tip end of the seed crystal. The shape of the growing crystal is governed by the withdrawing speed of the crystal **9** and the temperature programming for the crucible.

After completion of the crystal growth, the crucible temperature is gradually lowered to approximately 1000° C.–1030° C. to solidify the residual melt and the temperature is then further decreased to about 800° C.

As the second step, with the gas pressure within the quartz tube maintined at a reduced value by decreasing the temperature of the heater **5–1**, the gas pressure in the high pressure chamber is lowered by controlling the discharge rate of the gas from the discharge pipe **18–2** so that the difference between the gas pressure acting on the opposite sides of the tube walls is within approximately 5 atmospheres. When the temperature in the sealed tube has reached below 450° C. (the corresponding inner pressure should then be approximately from 1 to 3 atmospheres), the power sources for radio-frequency coil **4–1**

and heater 5–1 are turned "off" and the grown single crystal is then removed.

EMBODIMENT 2

The crucible temperature in an equilibrium state of the single crystal growth of gallium phosphide (GaP) and the temperature of heater 5–1 should be 1467° C. and approximately 600° C. respectively, at an internal pressure of about 35 atmospheres. The operating processes are substantially the same as in the previous embodiment.

EMBODIMENT 3

To cause the reaction and the single crystal growth to initiate directly from the raw material introduced into the crucible without using indium phosphide in polycrystalline form, metallic indium and impurities in suitable amounts are put into the crucible and heated for melting. The crucible temperature should be between 1000° C. and 1070° C.

Next, a piece of phosphor, not illustrated, which has been put into the space below the crucible 3 is subjected to a heating process at a temperature 450° C.– 500° C. for approximately 3–4 hours and to another heating process at 550° C. for approximately 2 hours so that the phosphor may be absorbed by the indium. During this time interval, a suitable gas pressure should be maintained within the high pressure chamber relative to the inner pressure of the sealed tube so that the difference between the two pressures exerted on the opposite sides of the sealed tube walls may be kept within approximately 5 atmospheres. At the conclusion of the absorption process, crystal growth should be initiated in the same manner as described in embodiment 1.

As a modification of the method illustrated in FIG. 3, it is also possible to fix the seed crystal in a part of the sealed tube and to magnetically couple the support for the melt-containing crucible, thereby to effect crystal growth.

FIG. 4 shows a preferred embodiment of a high-pressure magnetic-coupling floating-zone crystal purifying (single crystal growth) apparatus of the invention. The numerals 1 to 14 indicate the same parts as in FIG. 2, and 15 to 21 indicate the same parts as in FIG. 3. Numeral 22 is a gear box, 23 a seal, e.g. of a metal or Teflon resin, 24 a shaft for rotation or axial movement, and 25 a shaft for receiving a quartz supporter, being capable of rotation or axial movement while carrying the member 14 thereon, if desired, in direct coupling with the seal 23.

The apparatus of FIG. 4 is operated in the following manner. As electric power is applied to the heater 5–1, the vapor pressure of the vaporizable material inside the sealed tube is increased, and with the increase of the internal vapor pressure, a suitable atmospheric gas maintained at a pressure equivalent to the vapor pressure is fed into the pressure chamber through the reducing valve and discharge valve, in the same manner as described in connection with the apparatus of FIG. 3, so that the pressure can be regulated. Thus, the internal pressure of the sealed tube and the external pressure (i.e., the pressure inside the pressure chamber) are balanced, and the pressure difference upon the sealed tube is reduced below the withstand pressure of the sealed tube and the inside of the tube can be kept under a higher pressure condition. Now that the required pressure conditions have been satisfied, a high-frequency current from 4–2 is applied to 4–1, and by induction the interface between the ingot 12 and seed crystal 9 is heated and melted together to form a molten zone 2. In this state, regulation of the width of the molten zone is accomplished in the same manner as with the apparatus of FIG. 2 by controlling the high-frequency current and the relative distance between the ingot 12 and the seed crystal 9.

Because of the magnetic member 7 in its supporter, the seed 9 can be magnetically coupled to the magnet 6 through the less magnetic walls of the pressure chamber and is able to make rotation and vertical movement. Also because it can be fixed in position in a certain relative direction by the magnetic force of the magnet 6, the seed can rotate relative to the ingot fixed in the sealed tube by the shaft 25, and thus a homogenizing agitation of the input can be attained. While the vertical motion of the shaft 25 and relative position of the magnet 6 are kept constant, the ingot formed of the materials 9 and 12 is moved with respect to the high-frequency coil 4. This permits the melt 2 to pass through the ingot for floating-zone purification and single crystallization. The conditions of the molten zone can be observed through the quartz window 21.

FIGS. 5 and 6 show an embodiment of the underpressure magnetic coupling type Bridgeman crystal growth apparatus according to this invention, and a modification thereof, respectively. In these figures, the numerals 1 through 22 indicate like parts as in FIGS. 1 through 4, numeral 27 denotes a support member for the heating unit 29 (for instance, a carbon ring), 28 the position of a thermocouple, 30 a holder for coupling the crucible 3 to the movable element, and numeral 31 denotes a precision-ground guide rod for the movable element.

Operation of the apparatus of FIGS. 5 and 6 will now be described. When an equilibrium state is reached with the apparatus of FIG. 5 by balancing the inner pressure of the sealed tube accompanied by the temperature rise as described in connection with FIGS. 3 and 4, with the inner pressure of the pressure chamber, heating unit 29 is heated by a suitable radio-frequency source. Satisfactory heating and melting of a crystal or metal can easily be achieved by the use of a radio-frequency source by utilizing the principle of the induction furnace in lieu of the heating unit 29. While the boat 3 is gradually lowered by the movable element, the metal inside the boat is melted. As the crucible 3 is further lowered through the position of the heating unit 29, the molten zone is cooled and crystal growth is initiated from the bottom of the crucible.

With the apparatus of FIG. 6 the heating unit 29 is supported by the magnet 6 and heated by a suitable radio-frequency source.

Since the crucible 3 is fixed in the sealed tube 1, solidification of the metal in the crucible into a single crystal takes place from the bottom of the vessel by moving the sealed tube away from the heating unit.

The apparatus shown in FIGS. 5 and 6 affords a number of advantages. One advantage is that no seed crystal is required. Another is that no unnecessary temperature rise of the sealed tube walls can occur and also the heating unit may be protected from heat dissipation by a heat-insulating spacer disposed outside the unit because it is incorporated in the sealed tube. A further advantage is that the reaction and the crystal growth may be performed simultaneously from the raw material.

FIG. 7 shows a longitudinal cross sectional view of an embodiment of the underpressure magnetic coupling type horizontal Bridgeman crystal growth apparatus, wherein the numerals 1 through 31 show like component parts indicated by the same numerals in FIGS. 5 and 6. The mechanism of crystal growth with this apparatus is the same as with the apparatus of FIGS. 5 and 6, except that the horizontal type boat is designed to move magnetically with the movement of the movable element in a zone in which the temperature gradients exist, whereby the crystal growth is initiated. Cylinder 34 in combination with bushing 13 serves the function of a guide for the boat. The growth of a seed crystal placed in the boat occurs approximately at the position predetermined by the boat size and the temperature gradients or the temperature distribution in the vessel. This holds true for the apparatus shown in FIGS. 5 and 6.

FIG. 8 shows a longitudinal cross sectional view of an embodiment of apparatus specifically designed for the purpose of subjecting a single crystal placed in a vessel

constituting a part of the movable element sealed in the tube, to a heating process, an alloying process, an impurity diffusion process, or an epitaxial growth process from a gas or liquid phase. As one example, in diffusing zinc into indium phosphide (InP), a gas pressure of more than 20 atmospheres within the sealed tube is needed in order to prevent decomposition of the compound and to obtain a uniform diffusion layer. To make the diffusion time accurate, it is desirable that the single crystal be previously placed in a low-temperature zone and be moved to a high-temperature zone by magnetic action of the movable element after an equilibrium state has been reached.

In FIG. 8 the numerals **1** through **31** show like parts indicated by the same numerals in FIGS. 1 through 7, numeral **32** denotes a vessel such as a quartz plate integral with the movable element, and numeral **33** denotes a single crystal to be subjected to a heating and diffusion process. At the conclusion of the diffusion process, the single crystal should be transferred to a low-temperature zone and then externally removed. This apparatus has the advantages of providing feasibility of diffusion, alloying, or epitaxial crystal growth under unprecedented high temperatures and pressures and can also be employed for many other applications.

Two different embodiments of the movable element are illustrated by FIGS. 9A and 9B, wherein the numeral **7** denotes a ferromagnetic member, **10** a jacket made of quartz or a suitable metal for enclosing or fixing the ferromagnetic member, **8** a seed crystal holder connected to the jacket, **13** a precision-ground guide, made, for example of graphite, **35** denotes a guide shaft made of quartz or graphite for example, designed to slide over the interior surface of the sealed tube or to be directly connected to the sealed tube or the movable element to serve the function of a guide in combination with the bushing.

FIG. 9A illustrates a detailed arrangement of the movable element for use in the apparatus shown in FIG. 4 while FIG. 9B illustrates an arrangement of the movable element comprising a plurality of ferromagnetic members or pieces **7** disposed symmetrically with respect to the rotating shaft. In FIG. 9B, the numeral **36** denotes a holding plate for rigidly assembling the holder **8**, ferromagnetic pieces **7** and carbon bushing **13** together and **9** denotes a seed crystal rigidly held in the holder **8**.

Several different geometrical configurations for the ferromagnetic piece which constitutes an element of the movable assembly are exemplified in FIG. 10, see the members A through D. The ferromagnetic pieces should generally be made of materials such as iron, Permendur, etc. and assume the configurations adapted for use in the movable element. They may be solid bodies or made of laminated iron plate. They may be used without a coating or covering thereon, unless unaffected by a gas or vapor in the tube. If a deleterious effect of such a gas or vapor will be encountered, they should be sealed in quartz enclosures. These pieces should be disposed symmetrically with respect to the axis of the guide shaft and rigidly bonded to the holder.

Thus according to the invention, an external pressure is applied to a sealed tube maintained within a suitable temperature distribution range inside a pressure chamber, and simultaneously magnetic coupling is effected from the outside of the pressure chamber thereby to cause relative rotation and movement of a movable element attached to the seed crystal inside the sealed tube. With this method, crystal growth under high pressure conditions, which has hitherto been impracticable, is now achieved in a practical manner. In addition, the wall thickness of the sealed tube of quartz to be used can be minimized. Thus, the method is not only efficient and economical, but enables highly pure materials to be used in forming the sealed tube and crucible. The pressure chamber is thoroughly cooled with water, and therefore is highly resistant to pressure and ensures considerable safety. The magnet is positioned outside the chamber so that it is not affected by the heat and it enables the chamber to be simplified in structure and permits the whole apparatus to be compact and small in size. Moreover, assembling, disassembling, and cleaning of the apparatus can be carried out with great efficiency.

While the foregoing description sets forth the principles of the invention in connection with specific apparatus, it is to be understood that the description is made only by way of example and not as a limitation of the scope of the invention as set forth in the objects thereof and in the accompanying claims.

What is claimed is:

1. Apparatus for growing crystals comprising

a hermetically sealed tube containing therein a material to be grown into a crystal,

at least a portion of the walls of said sealed tube comprising a transparent material whose main constituent is silicon dioxide one to three millimeters in wall thickness,

a ferromagnetic body,

a movable element seed holder mechanically coupled to said ferromagnetic body, the ferromagnetic body and seed holder being freely mounted in the sealed tube,

heating means for said sealed tube at a peripheral portion thereof,

a water-cooled pressure chamber enclosing said sealed tube and said heating means, with said pressure chamber provided with a viewing window which allows visibility of said crystals during growth thereof,

at least a part of said pressure chamber surrounding said ferromagnetic body and consisting of a non-ferromagnetic stainless steel substance,

magnetic field generating means disposed externally of said pressure chamber and in the vicinity of said ferromagnetic body and capable of moving and rotating said movable element by a magnetic force transmitted from said magnetic field generating means,

means for providing a gas pressure externally of said sealed tube in said pressure chamber,

and means for causing said gas pressure to vary in response to and in proportion to the magnitude of the pressure within said sealed tube determined by the temperature within the tube so as to maintain the difference between the two pressures exerted on the opposite sides of the walls of said sealed tube within a predetermined range of values whereby explosions of said sealed tube are prevented.

2. Crystal growth apparatus according to claim **1** in which said sealed tube further contains a seed crystal to be grown into a larger crystal.

3. Crystal growth apparatus according to claim **2** in which said seed crystal is held in said movable element and is capable of displacement and rotation relative to said material.

4. Crystal growth apparatus according to claim **1** in which said sealed tube further contains a vessel for accommodating said material and a heating unit disposed in the vicinity of said vessel and heated by said heating means.

5. Crystal growth apparatus according to claim **4** wherein

said sealed tube further contains a vessel holder for mechanically coupling said vessel to said movable element,

said heating unit being fixed in position within said sealed tube, and wherein said material in said vessel is permitted to move and rotate relative to said heating unit.

6. Crystal growth apparatus according to claim **1** wherein

said pressure chamber is equipped with gas supply means including a gas inlet integral therewith,

and wherein external gas pressure can be applied to said sealed tube by introducing a gas into the space between said sealed tube and said pressure chamber through said gas inlet.

7. Crystal growth apparatus according to claim 6 wherein

said pressure chamber is equipped with a gas outlet for said gas,

a valve installed at said gas outlet and a valve installed at said gas inlet,

and wherein the external pressure applied to said sealed tube can be varied by opening and closing said latter valves in response to changes in the pressure within said sealed tube.

8. Crystal growth apparatus according to claim 6 wherein further comprises

means for measuring the minimum temperature within said sealed tube,

a valve installed at said gas outlet,

said valve installed at said gas inlet being connected to said pressure chamber,

and said means for causing gas pressure variation comprising means for opening and closing said valves in response to the minimum temperature within said sealed tube measured by said temperature measuring means in order to control the external gas pressure applied to said sealed tube.

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