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(54) **METHOD AND APPARATUS FOR GROWING HIGH QUALITY SINGLE CRYSTAL**  
VERFAHREN UND VORRICHTUNG ZUR HERSTELLUNG VON HOCHQUALITATIVEN  
EINKRISTALLEN  
PROCEDE ET APPAREIL DE FABRICATION DE MONOCRISTAUX DE GRANDE QUALITE

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- **PATENT ABSTRACTS OF JAPAN vol. 017, no. 700 (C-1145), 21 December 1993 (1993-12-21) & JP 05 238875 A (MITSUBISHI MATERIALS CORP;OTHERS: 01), 17 September 1993 (1993-09-17)**
- **FURUYA H. ET. AL.: 'Development of New Nonlinear Optical Crystal GdYCOB with tunable Birefringence' NIPPON KESSHO SEICHO GAKKAISHI vol. 25, no. 5, 1998, pages 193 - 199, XP002929051**

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## Description

**[0001]** The present invention relates to a method and an apparatus for growing a high quality single crystal. Particularly, the present invention relates to a new method for growing a high quality single crystal, the method being capable of growing a high quality single crystal using even highly viscous liquid raw materials and to an apparatus for practicing the method .

**[0002]** Conventionally, as a method for growing a single crystal of an oxide or the like, a method is known in which raw materials are melted under heating in a crucible, thereafter a seed crystal is brought into contact with the raw material melt and a round bar-like single crystal is pulled up whilst rotating the seed crystal to grow a single crystal. This pulling method is used for the growth of various single crystals as a method which can grow a large-diameter crystal in an efficient manner.

**[0003]** Also, for example, a method (Kyropoulos method) is known in which raw materials are melted under heating in a crucible, thereafter a seed crystal is brought into contact with the raw material melt and the temperature below liquid level is slowly lowered to precipitate and grow a crystal.

**[0004]** EP-A-732427 describes a method for the growth of a single crystal which involves contacting the lower end of a column crystal with a raw material melt in the presence of a cylindrical body acting as a baffle arrangement.

**[0005]** However, in the conventional methods of growing a single crystal by bringing a seed crystal into contact with a raw material melt, there is the problem that when the viscosity of the raw material melt during growth at the required temperature is high, the flow of the raw material melt in the crucible is retarded, causing nonuniformity in temperature, the degree of supersaturation or the like, leading to the result that the quality of the crystal tends to be lowered.

**[0006]** For example, CsLiB<sub>6</sub>O<sub>10</sub> (CLBO) and the like as non-linear optical crystals attract attention as those for the generation of high output ultraviolet laser light and are desired to have excellent performances and qualities such as very high resistance to damages by lasers, very high optical loss and high uniformity. However, since it is a borate type crystal, its melt has a high viscosity, which makes it difficult to grow a high quality and high performance single crystal. It has been confirmed that in actual measurement, the viscosity of a CLBO solution of a self flux composition is as high as about 1000 cs (centistokes) at a growth temperature, around 840°C.

**[0007]** For example, in the growth of a single crystal by means of the rotation of a seed bar in the CLBO cooling method, the distribution of temperature in the raw material melt is not good as shown in FIG. 7 and the growth of a crystal is rapid, and therefore there is a restriction inevitably on the growth of a high quality and high performance crystal.

**[0008]** In view of this situation, it is an object of the present invention to solve the problems in prior art as mentioned above and to provide an improved new method which can grow a high quality and high performance single crystal in the case of using even a highly viscous raw material melt and also to provide an apparatus for practicing the method.

**[0009]** The present invention has been made to solve the aforementioned problem and, first, provides a method for growing a high quality single crystal, said method comprising bringing a seed crystal into contact with a raw material melt which is heated and melted within a crucible, wherein a blade member having a screw form is arranged in the raw material melt in the crucible and the single crystal is grown by rotating the crucible without rotating the blade member and slowly cooling the raw material melt with which the seed crystal makes contact below liquid level to precipitate a single crystal on the surface of the seed crystal. Also, the present invention provides a method for growing by slowly pulling up the seed crystal which is brought into contact with the raw material melt. In the method the seed crystal may also be rotated while rotating the crucible. A single crystal of an oxide may be grown, the single crystal of an oxide preferably being a single crystal of a borate type oxide. The borate type oxide may be CsLiB<sub>6</sub>O<sub>10</sub> or an oxide obtained by partially substituting at least one of Cs and Li of CsLiB<sub>6</sub>O<sub>10</sub> with at least one type among other alkali metal elements and alkali earth metal elements. The oxide may be an oxide doped with at least one of Al and Ga elements. The borate type oxide may be represented by Gd<sub>x</sub>Y<sub>1-x</sub>Ca<sub>4</sub>O(BO<sub>3</sub>)<sub>3</sub> (0<x<1) and the crystal may be grown by a pulling method. The single crystal of an oxide may be LiNbO<sub>3</sub>, LiTaO<sub>3</sub>, a high-temperature superconductive oxide material or a heat-electricity-conversion oxide material.

**[0010]** Further, the present invention provides a growing apparatus for growing a single crystal by bringing a seed crystal into contact with a raw material melt which is heated and melted within a crucible, comprising a blade member having a screw form arranged in the raw material melt in the crucible and a rotating means for rotating the crucible and a cooling mechanism for slowly cooling the raw material melt, with which the seed crystal makes contact, below liquid level. The growing apparatus may comprise a pulling mechanism for slowly pulling up the seed crystal which is brought into contact with the raw material melt. The apparatus may also comprise a mechanism for rotating the seed crystal. Furthermore, there is provided an apparatus for growing a single crystal of an oxide according to any one of the aforementioned growing apparatuses and an apparatus for growing a single crystal of a borate type oxide.

**[0011]** In the accompanying drawings:

FIG. 1 is a structural view showing the outline of a method and an apparatus according to this invention.

FIG. 2 is a sectional view showing a growing apparatus as an example.

FIG. 3 is a plan view illustrating a blade member.

FIG. 4 is a side view of a blade member.

FIG. 5 is a view showing the hysteresis of crystal growth.

FIG. 6 is a view showing the distribution of temperature in a raw material melt.

5 FIG. 7 is a view showing the distribution of temperature in a melt in the case of a conventional method.

**[0012]** The symbols in these figures represent the following parts.

- 1 Crucible
- 10 2 Raw material melt
- 3 Seed bar
- 4 Seed crystal
- 5 Blade member
- 6 Rotating member
- 15 7 Support bar

**[0013]** The present invention has the characteristics as aforementioned and embodiments of the invention will be hereinafter explained.

20 **[0014]** First, a method for growing a single crystal according to the present invention is based upon the bringing of a seed crystal into contact with a melt of raw materials melted under heating in a crucible to grow a single crystal. In the growing method of this invention, various modes such as a conventionally known pulling method and a cooling method (Kyropoulos method) using gradual cooling are optionally adopted. The point that the seed crystal is brought into contact with the melt (including the case of a melt) produced by melting raw materials to grow a single crystal is common to all of these modes.

25 **[0015]** When practicing the method of this invention, a growing apparatus basically comprises a crucible, a heating means for heating and melting raw materials placed in the crucible, a means for detecting and controlling heating temperature and a crystal support means for bringing a seed crystal into contact with a melt (including a melt) of the raw materials which are melted under heating. As shown in FIG. 1 showing the typical view of the apparatus, the present invention has the characteristics that when a seed crystal (4) supported by a crystal support means such as  
30 a seed bar (3) is brought into contact with a melt (2) of raw materials melted under heating in a crucible (1) to grow a single crystal, a blade member (5) is disposed in the raw material melt (2) in the crucible (1) and the crucible is rotated to grow the single crystal. For the rotation of the crucible (1), the growing apparatus is provided with a rotating means (6) which rotates the crucible (1) in the condition that the crucible (1) is mounted thereon.

35 **[0016]** In the case of a pulling method, the seed bar (3) is pulled up towards above whilst rotating the seed bar (3) or in such a stationary state that the seed bar (3) is not rotated. On the other hand, in the case of the cooling method, for example, a method in which using a hollow seed bar (3), cooling gas is supplied to the hollow portion and the raw material melt below liquid level is cooled slowly to precipitate a single crystal on the surface of the seed crystal (4), thereby growing a single crystal or a method in which the temperature of whole heaters in a furnace is slowly lowered to cool the temperature of the raw material melt below liquid level may be adopted. The former method is used to  
40 prevent the seed crystal from being melted. In these cases, the support means such as the seed bar (3) may also be either rotated or be in such a stationary state that the seed bar (3) is not rotated. It is to be noted that the present invention has, as its basic thought, the characteristics that the rotations of the seed bar (3) as aforementioned and seed crystal (4) which are supported by the seed bar (3) are made to be unessential and the crucible (1) is rotated. Whether the seed bar (3) and the seed crystal (4) are rotated or not is determined according to need as a relative  
45 motion to the rotation of the crucible (1).

**[0017]** In the present invention, the blade member (5) itself may be in a stationary state whereas the crucible (1) is designed to be rotated by the rotating material (6) in the raw material melt (2) as shown in, for example, FIG. 1.

50 **[0018]** The presence of the blade member (5) having such characteristics and the rotation of the crucible (1) enhance the effect of stirring the raw material melt, which makes it possible to make the diffusion boundary layer thin, to increase the amount of the raw materials to be supplied to the growth surface and to make the degree of supersaturation uniform. This enables the growth of a high quality and high performance single crystal even if the raw material melt is highly viscous at the growth temperature.

55 **[0019]** The blade member (5) as those having various shapes may be arranged in consideration of the composition and type of single crystal which is a subject of growth and raw material, the composition and viscosity of the raw material melt, further the depth of the above member inserted into the raw material melt (2), a distance from the rotation center of the crucible (1), the flow direction and flow speed of the raw material melt (2) and the like. As preferable examples of the blade member (5), plural blades are arranged radially and fixed at the center of them. Namely, those made to have a screw form are exemplified.

**[0020]** Although these blade members (5) are not rotated but placed in a stationary state, vibrations and motions such as reciprocating motions in at least one of a vertical direction and a horizontal direction may be imparted to these members. Also, as shown in FIG. 1, the blade member (5) may be inserted into the crucible (1) from the above and pulled up by the support bar (7), may be disposed such that, for example, the depth of the member to be arranged in the raw material melt (2) can be adjusted and may be fitted and fixed to the bottom or the like of the crucible (1). More preferably, it is considered to make the member have the former structure.

**[0021]** As to the rotation of the crucible (1) by means of the rotating means (6), it is considered that the crucible (1) is made to be able to rotate in a direction positive with respect to the direction of the rotation of the seed crystal (4) or in both directions reversibly. Also, regarding the rotation of the crucible (1), it is considered to change the rate of rotation under control during the course of the growth. It is also considered that the direction of rotation and the rate of rotation are changed under control in relation to the optical detection of, for example, the flow and temperature of the melt and growth size of the single crystal or to pressure sensitive and heat-sensitive detection of, for example, the flow and temperature of the melt at the blade member (5) or the support bar (7) in the crucible (1).

**[0022]** The single crystal which is an object of this invention may be of various types and this invention is more effective when the raw material melt is highly viscous at growth temperatures. As examples of the single crystal, various oxides are given. Particularly, this invention is suitable for the growth of highly viscous borate type crystals such as  $\text{CsLiB}_6\text{O}_{10}$  (CLBO) for which a high quality and high performance single crystal is expected, compositions prepared by partially substituting at least one of Cs and Li of  $\text{CsLiB}_6\text{O}_{10}$  with one of other alkali metal elements or alkali earth metal atoms or those doped with an atom such as Al or Ga. In an example explained later, explanations are offered taking, as an example, the case where the above CLBO is grown by a cooling method, though the invention is not limited to this case.

**[0023]** Also in the growth of a crystal of  $\text{GdYCOB}$ , namely,  $\text{Gd}_x\text{Y}_{1-x}\text{Ca}_4\text{O}(\text{BO}_3)_3$  by a pulling method (Czochralski method: Cz method), a high quality crystal is obtained by the method of this invention. In the pulling method in particular, it is also effective to dispose the baffle member.

**[0024]** Also, as the oxides to be grown,  $\text{LiNbO}_3$ ,  $\text{LiTaO}_3$ , high-temperature superconductive oxide materials and heat-electricity conversion oxide materials such as  $\text{Na}_x\text{CO}_2\text{O}_4$  (x is about 1) are also exemplified.

**[0025]** Now, the present invention will be explained in more detail by way of examples shown below.

#### EXAMPLES

(Growing apparatus)

**[0026]** As a growing apparatus, the whole was designed to have the structure of FIG. 2. Using a platinum crucible, the crucible was designed to be able to be rotated by a motor. Also, in this apparatus, a hollow one was used as a seed bar (3), a seed crystal (4) was supported by the lower end of the seed bar(3) and the seed crystal (4) was designed to be capable of being cooled by supplying seed-cooling gas to the inside of the hollow seed bar (3). This prevented the seed crystal (4) from melting and falling. This apparatus enables growth using a melt composition which conventionally involves a difficulty in the growth of a crystal because the seed crystal melts.

**[0027]** In the platinum crucible, a screw type blade member (5) made of platinum shown in FIG. 3 and FIG. 4 was disposed by fitting it to the support bar (7). The blade member (5) had six blades which were arranged at a blade angle of 40 degrees. The blade member (5) was arranged such that the center (A) of the blade was disposed at a plane position corresponding to the center of rotation of the crucible. The distance (H) from the inside bottom of the crucible to the center (A) of the blade was designed to be capable of being controlled. It is to be noted that as to the distance (H), the center (A) of the blade was designed to be positioned as close as possible to the inside bottom of the crucible.

**[0028]** It is to be noted that both of the Alsynto pipe and FKS pipe shown in FIG. 4 were purchased from FURUYA-METAL CO., LTD.). The Alsynto pipe was made of alumina ( $\text{Al}_2\text{O}_3$ ) as its primary raw material and the FKS pipe was constituted of a material comprising platinum (Pt) containing  $\text{ZrO}_2$ .

(Single crystal growth)

**[0029]** Using the aforementioned growing apparatus, the crucible was rotated to grow a CLBO single crystal by a cooling method.

**[0030]** The seed bar was not rotated and similarly the blade member was not rotated to make growth in a stationary state. The raw material melt was made to have a CLBO self flux composition. The percentage composition of this self flux was as follows: Cs : Li : B : O = 1 : 1 : 5.5 : 9.2. Also, it has been confirmed that this composition is preferably a stoichiometric composition (melt composition).

**[0031]** The maximum heating temperature of the raw material melt was set to 900°C.

**[0032]** Each condition of temperature descending rate and the rotation of the crucible was as follows.

Temperature descending rate

0.1°C/day

Rotation of the crucible

30 rpm

5 **[0033]** The position where the temperature descending rate was measured was the level of the melt as the first standard. Thereafter, the temperature of the melt was made to descend at a rate of 0.1°C/day from the temperature at the level. The temperature at this time was measured by control sensors shown in FIG. 2 and the temperature of the whole melt was made to descend equally at a rate of 0.1°C/day.

10 **[0034]** FIG. 5 shows the hysteresis of the crystal when compared with that of a conventional usual method and FIG. 6 shows the distribution of temperature in the raw material melt. It is understood from FIG. 6 that the distribution of temperature of the melt in the crucible is made more uniform in the direction of the height than that of a conventional method and the crystal is grown uniformly.

15 **[0035]** Further studies as to the results of the distribution of temperature in the raw material melt in FIG. 6 proved it desirable for the growth of a high quality crystal that a difference ( $\Delta t$ ) in temperature between positions extending from the liquid level up to a height (depth) of about 10 cm was in a range of up to -0.5°C, namely between -0.5°C to 0°C.

20 **[0036]** Also, as shown in FIG. 5, in the conventional growing method, although a growth at the first start is slow, the rate of growth rises on the way and the final rate of growth becomes considerably high. This shows that when a crystal is small, a poor stirring effect is produced even if a seed bar is rotated whereas when a crystal is large, the crystal itself stirs a melt to thereby accelerate the growth instantly.

25 **[0037]** On the contrary, in the growth according to the method of this invention in which the blade member is inserted and the crucible is rotated, a growth at the first start is faster than in the case of the conventional growth using the rotation of a seed bar. This is because the melt is sufficiently stirred by the rotation of the crucible and therefore a layer called the diffusion boundary layer which determines the rate of growth becomes thin. Further, this is because the degree of supersaturation is made uniform.

(Evaluation of crystals)

30 **[0038]** In order to evaluate the quality of the grown crystal, samples prepared by cutting the crystal into wafer-like shapes 1.5 cm in thickness and polishing the three-planes of the cut crystal were subjected to a test using a He-Ne laser to observe the internal diffusion in the crystal. In the crystals having high qualities, diffusion arises in the inside and the inside is bright with red light, so that the points of diffusion were seen. In deficient places, passes were viewed.

**[0039]** As a result of the observation, it was confirmed that the crystal grown according to the method of this invention in which the blade member was inserted and the crucible was rotated had good qualities and a few passes were viewed at the lower portion of the seed crystal.

35 **[0040]** On the other hand, in the crystals grown by the conventional method, passes were viewed as a whole, giving rise to a problem concerning the qualities of the crystal.

40 **[0041]** Also, as samples for the evaluation of resistance to a laser, the same samples as above and crystals 10 mm×10 mm×15 mm in size which were grown by the conventional method were used. The measurement of damage threshold value was made for the (001) plane. As the laser light source, a longitudinal and lateral single mode Q switch Nd:YAG laser was used. The evaluation was made using an oscillation wavelength of 266 nm which was fourth harmonics of the Nd:YAG laser. The pulse width was 0.75 ns.

45 **[0042]** Light having a diameter of 8 mm was converged using a lens with a focal distance of 100 mm. Here, the position of the crystal was adjusted such that the focus portion was located at a distance of 5 mm from the plane of incidence and the crystal was moved every shot. It has been confirmed that no damage is produced on the plane of incidence in the light conversion condition in this case. Continuous light of a He-Ne laser was allowed to pass on the same axis of the Nd:YAG laser to confirm whether a diffusion point was present or not on the portion irradiated with the laser light every movement and to examine visually whether or not diffusion points newly occurred after the shot to thereby determine whether damages were present or not. When the incident energy is higher than the damage threshold value, plasma was observed at the light convergence portion. In the vicinity of the threshold value, the occurrence of diffusion points was only confirmed. The intensity of the laser pulse was changed by a combination of a  $\lambda/2$  plate (polarization rotor) and a polarizer. The incident energy was monitored by a biplanaphoto tube and an oscilloscope which were calibrated by a calorimeter. Fused quartz (10.4 GW/cm<sup>2</sup>) was used as a reference example.

50 **[0043]** The internal laser damage threshold value was measured using the fourth harmonics (266 nm) of an Nd:YAG laser in such procedures. The internal laser damage threshold value of the crystal grown according to this invention in which the blade member was inserted and the crucible was rotated and the internal laser damage threshold values of the crystal grown by the conventional method and fused quartz are shown in Table 1.

Table 1

Method	Damage threshold value (GW/cm <sup>2</sup> )
Fused quartz	10.4
Conventional method	8.8-8.9
This invention	10.4-20.8

**[0044]** As shown in Table 1, it was confirmed that the internal laser damage threshold value of the crystal grown by the conventional growing method was lower than that of fused quartz whereas the internal laser damage threshold value of the crystal grown according to the method of this invention in which the blade member was inserted and the crucible was rotated was higher than that of fused quartz even when the threshold value was lower and was about two times that of fused quartz when the threshold value was the highest.

**[0045]** As mentioned above, when comparing the internal laser damage threshold value of the crystal grown by the conventional method with that of the crystal grown by the method of this invention, it is understood that the crystal of this invention is considerably higher than the conventional crystal in the internal laser damage threshold value. This means that the crystallinity is significantly improved.

**[0046]** As mentioned above in detail, the present invention ensures that various single crystals including CLBO are grown as high quality and high performance crystals from a highly viscous raw material melt.

### Claims

1. A method for growing a single crystal, said method comprising bringing a seed crystal into contact with a raw material melt which is heated and melted within a crucible, wherein a blade member having a screw form is arranged in said raw material melt in said crucible and said single crystal is grown by rotating said crucible without rotating said blade member and slowly cooling said raw material melt with which said seed crystal makes contact below liquid level to precipitate a single crystal on the surface of said seed crystal.
2. A method as claimed in claim 1 wherein said blade member has a plurality of blades arranged radially.
3. A method as claimed in claim 1 or claim 2, wherein said seed crystal is also rotated while rotating the crucible.
4. A method as claimed in any one of claims 1 to 3, wherein a single crystal of an oxide is grown.
5. A method as claimed in claim 4, wherein the single crystal of an oxide is a single crystal of a borate type oxide.
6. A method as claimed in claim 5, wherein the borate type oxide is CsLiB<sub>6</sub>O<sub>10</sub> or an oxide obtained by partially substituting at least one of Cs and Li of CsLiB<sub>6</sub>O<sub>10</sub> with at least one type among other alkali metal elements and alkali earth metal elements.
7. A method as claimed in claim 6, wherein the oxide is an oxide doped with at least one of Al and Ga elements.
8. A method as claimed in claim 5, wherein the borate type oxide is represented by Gd<sub>x</sub>Y<sub>1-x</sub>Ca<sub>4</sub>O(BO<sub>3</sub>)<sub>3</sub> (0<x<1) and the crystal is grown by a pulling method.
9. A method according to claim 4, wherein the single crystal of an oxide is LiNbO<sub>3</sub>, LiTaO<sub>3</sub>, a high-temperature superconductive oxide material or a heat-electricity-conversion oxide material.
10. A growing apparatus for growing a single crystal by bringing a seed crystal into contact with a raw material melt which is heated and melted within a crucible, comprising a blade member having a screw form arranged in the raw material melt in the crucible and a rotating means for rotating the crucible and a cooling mechanism for slowly cooling the raw material melt, with which the seed crystal makes contact, below liquid level.
11. A growing apparatus as claimed in claim 10 comprising a mechanism for rotating said seed crystal.
12. An apparatus for growing a single crystal of an oxide comprising the growing apparatus as claimed in claims 10

or 11.

13. A growing apparatus as claimed in claim 12 for growing a single crystal of a borate type oxide.

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**Patentansprüche**

1. Verfahren zur Züchtung eines Einkristalles, wobei man einen Impfkristall in Kontakt bringt mit einer Rohmaterialschmelze, die in einem Tiegel erhitzt und geschmolzen wird, wobei eine schraubenförmige Schaufelvorrichtung in der Rohmaterialschmelze in dem Tiegel angeordnet ist und der Einkristall gezüchtet wird, indem man den Tiegel dreht ohne die Schaufelvorrichtung zu drehen und langsam die Rohmaterialschmelze abkühlt, mit welcher der Impfkristall unter dem Flüssigkeitsspiegel in Kontakt steht, so dass ein Einkristall auf der Oberfläche des Impfkristalls sich niederschlägt.
2. Verfahren nach Anspruch 1, worin die Schaufelvorrichtung mehrerer radial angeordnete Schaufeln aufweist.
3. Verfahren nach Anspruch 1 oder 2, worin während der Rotation des Tiegels der Impfkristall ebenfalls gedreht wird.
4. Verfahren nach einem der Ansprüche 1 bis 3, worin ein Einkristall eines Oxids gezüchtet wird.
5. Verfahren nach Anspruch 4, worin der Einkristall eines Oxids ein Einkristall eines Oxids des Borat-Typs ist.
6. Verfahren nach Anspruch 5, worin das Oxid des Borat-Typs das  $CsLiB_6O_{10}$  ist oder ein Oxid, das durch partielle Substitution wenigstens eines der Elemente Cs und Li des  $CsLiB_6O_{10}$  durch wenigstens einen Typ anderer Alkalimetallelemente und Erdalkalimetallelemente erhalten worden ist.
7. Verfahren nach Anspruch 6, worin das Oxid ein mit wenigstens einem der Elemente Al und Ga dotiertes Oxid ist.
8. Verfahren nach Anspruch 5, worin das Oxid des Borat-Typs durch  $Gd_xY_{1-x}Ca_4O(BO_3)_3$  ( $0 < x < 1$ ) repräsentiert wird und der Kristall mit Hilfe eines Ziehverfahrens gezüchtet wird.
9. Verfahren nach Anspruch 4, worin der Einkristall eines Oxides das  $LiNbO_3$ ,  $LiTaO_3$ , ein hochtemperatursupraleitendes Oxidmaterial oder ein Wärme-Elektrizitäts-Umwandlungs-Oxidmaterial ist.
10. Züchtungsvorrichtung zum Züchten eines Einkristalles, in dem man einen Impfkristall in Kontakt mit einer Rohmaterialschmelze bringt, die in einem Tiegel erhitzt und geschmolzen wird, mit einer schraubenförmigen Schaufelvorrichtung, die in der Rohmaterialschmelze im Tiegel angeordnet ist und Drehmitteln zum Drehen des Tiegels und einem Kühlmechanismus zum langsamen Abkühlen der Rohmaterialschmelze, mit welcher der Impfkristall unter dem Flüssigkeitsspiegel in Kontakt steht.
11. Züchtungsvorrichtung nach Anspruch 10 mit einem Mechanismus zum Drehen des Impfkristalles.
12. Vorrichtung zum Züchten eines Einkristalles eines Oxids, welche die Züchtungsvorrichtung nach den Ansprüchen 10 oder 11 umfasst.
13. Züchtungsvorrichtung nach Anspruch 12 zum Züchten eines Einkristalles eines Oxids des Borat-Typs.

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**Revendications**

1. Procédé de fabrication (ou de croissance) d'un monocristal, ledit procédé comprenant la mise en contact d'un germe cristallin avec une matière première fondue qui est chauffée et fondue à l'intérieur d'un creuset, dans lequel un élément de coupe ayant une forme d'hélice est disposé dans ladite matière première fondue dans ledit creuset et ledit monocristal est fabriqué (ou mis en croissance) en faisant tourner ledit creuset sans faire tourner ledit élément de coupe et en faisant doucement refroidir ladite matière première fondue avec laquelle ledit germe cristallin est en contact en dessous du niveau du liquide afin de précipiter un monocristal sur la surface dudit germe cristallin.

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2. Procédé selon la revendication 1, dans lequel ledit élément de coupe présente une pluralité de lames disposées radialement.
- 5 3. Procédé selon la revendication 1 ou 2, dans lequel ledit germe cristallin tourne également pendant que le creuset tourne.
4. Procédé selon l'une quelconque des revendications 1 à 3, dans lequel un monocristal d'un oxyde est fabriqué.
- 10 5. Procédé selon la revendication 4, dans lequel le monocristal d'un oxyde est un monocristal d'un oxyde de type borate.
- 15 6. Procédé selon la revendication 5, dans lequel l'oxyde de type borate est le  $\text{CsLiB}_6\text{O}_{10}$  ou un oxyde obtenu en substituant partiellement au moins un des Cs et Li du  $\text{CsLiB}_6\text{O}_{10}$  avec au moins un type parmi d'autres éléments de métal alcalin et éléments de métal alcalino-terreux.
- 20 7. Procédé selon la revendication 6, dans lequel l'oxyde est un oxyde dopé avec au moins un des éléments Al et Ga.
8. Procédé selon la revendication 5, dans lequel l'oxyde de type borate est représenté par  $\text{Gd}_x\text{Y}_{1-x}\text{Ca}_4\text{O}(\text{BO}_3)_3$  ( $0 < x < 1$ ) et dans lequel le cristal est fabriqué par une méthode de tirage.
- 25 9. Procédé selon la revendication 4, dans lequel le monocristal d'un oxyde est  $\text{LiNbO}_3$ ,  $\text{LiTaO}_3$ , un matériau d'oxyde superconducteur à haute température, ou un matériau d'oxyde de conversion électricité-chaleur.
- 30 10. Appareil de fabrication pour fabriquer un monocristal en mettant en contact un germe cristallin avec une matière première fondue qui est chauffée et fondue à l'intérieur d'un creuset, comprenant un élément de coupe ayant une forme d'hélice disposé dans la matière première fondue dans le creuset et un moyen de rotation pour faire tourner le creuset et un mécanisme de refroidissement pour faire doucement refroidir la matière première fondue, avec laquelle le germe cristallin est en contact, en dessous du niveau du liquide.
- 35 11. Appareil de fabrication selon la revendication 10, comprenant un mécanisme pour faire tourner ledit germe cristallin.
12. Appareil de fabrication d'un monocristal d'un oxyde, comprenant l'appareil de fabrication selon la revendication 10 ou 11.
- 40 13. Appareil de fabrication selon la revendication 12 pour fabriquer un monocristal d'un oxyde de type borate.



Fig. 1

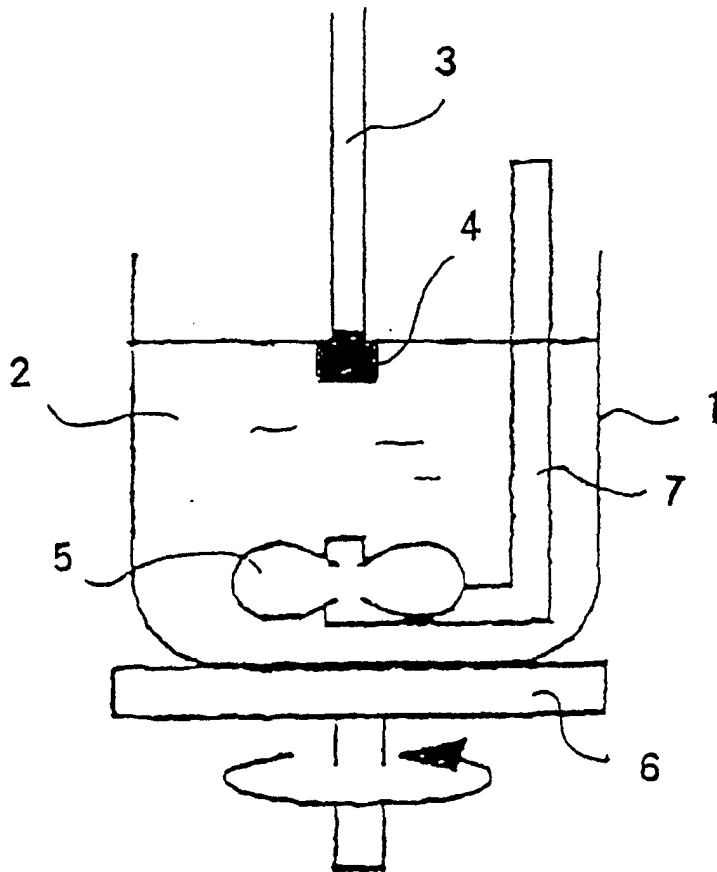


Fig.2

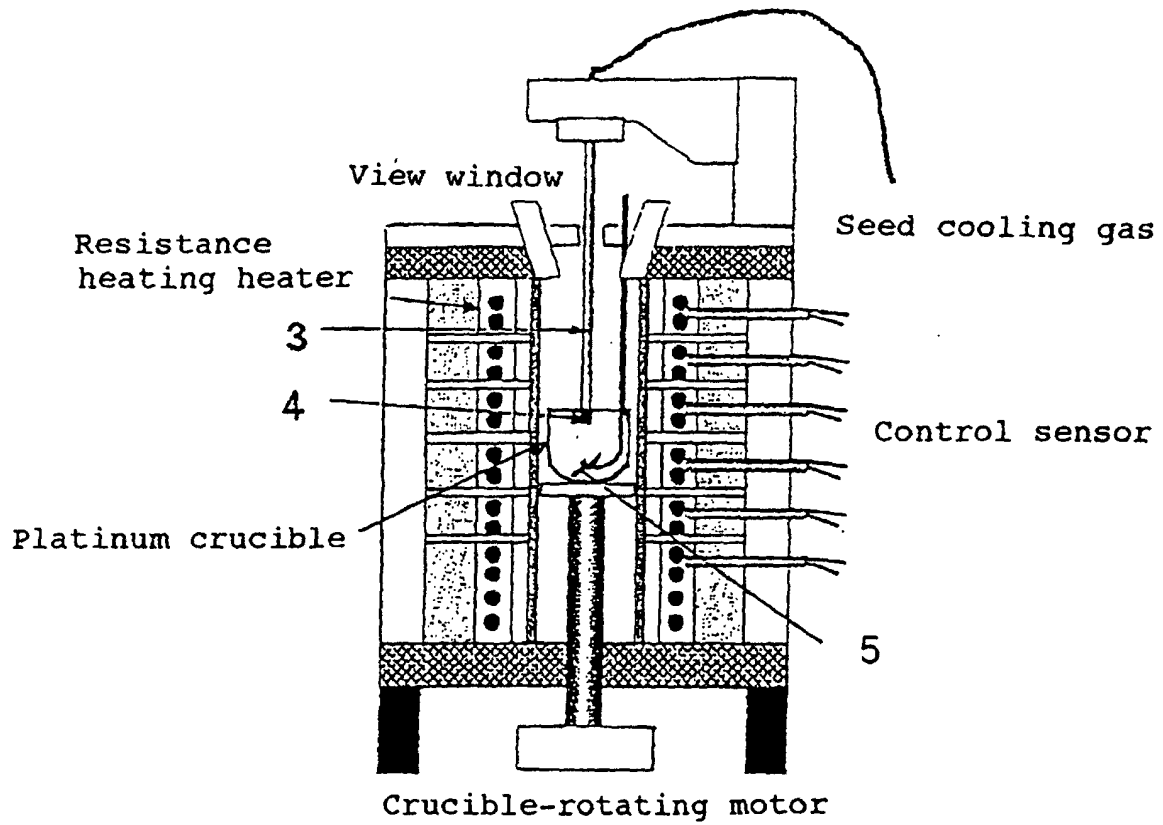


Fig.3

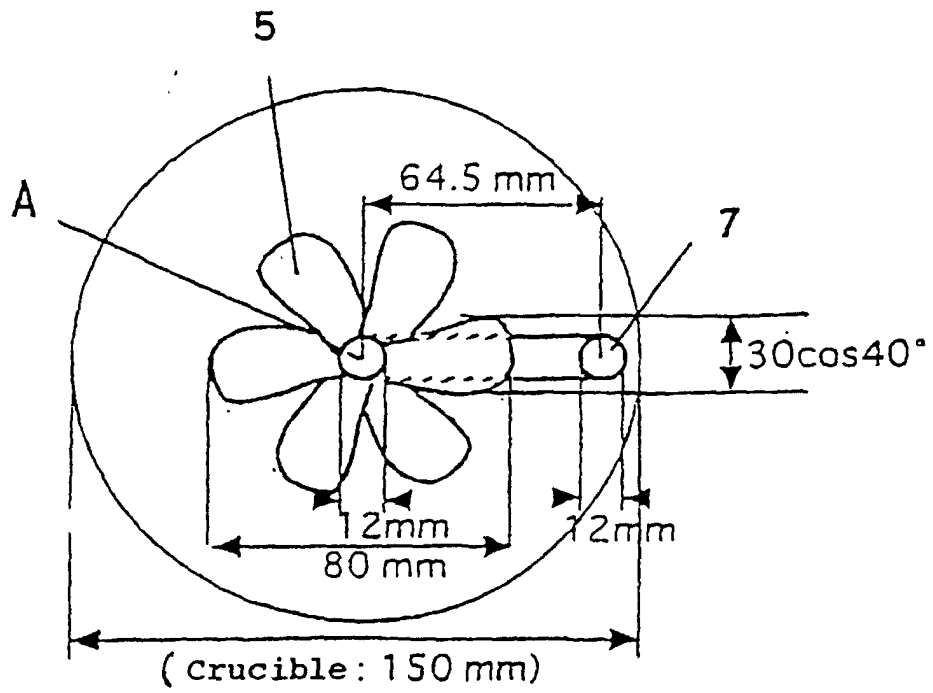


Fig.4

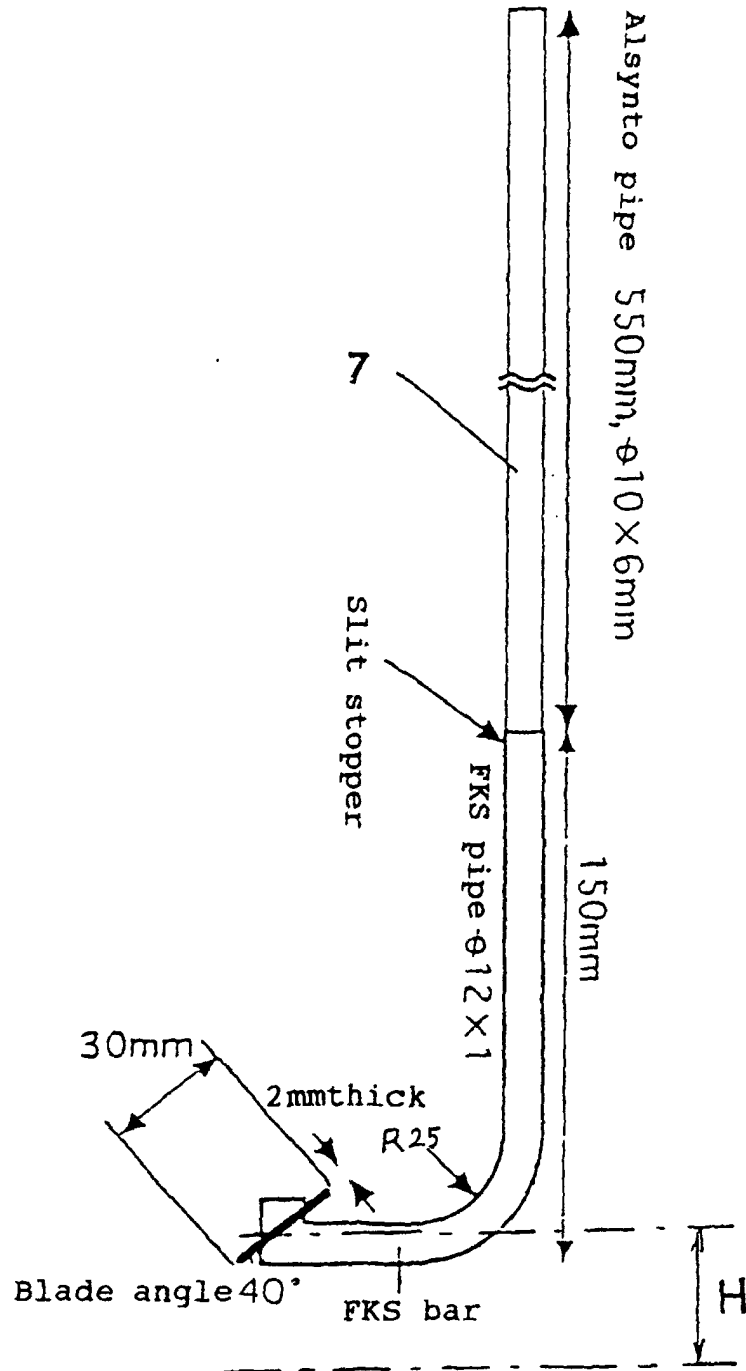


Fig.5

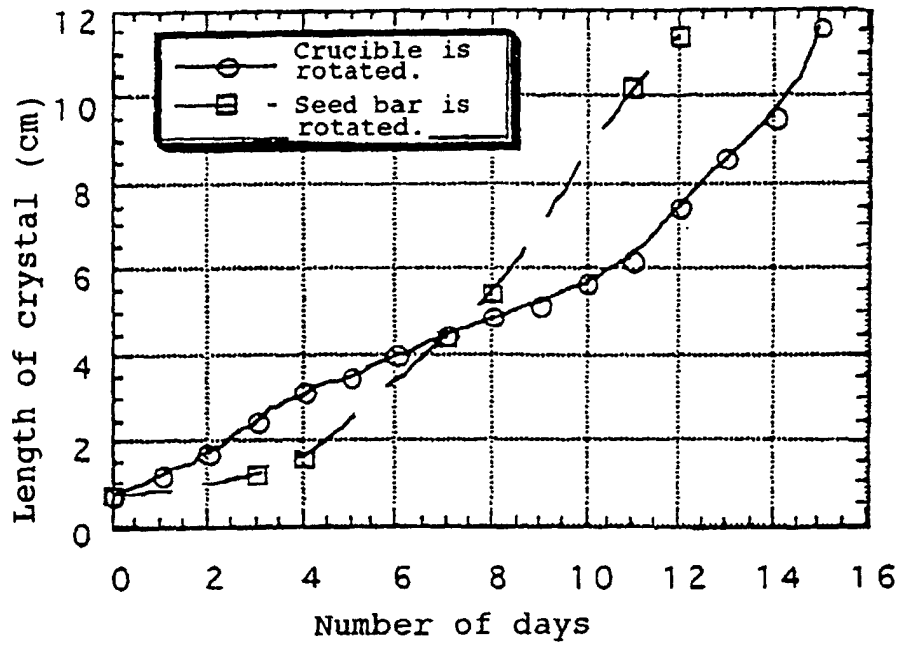


Fig.6

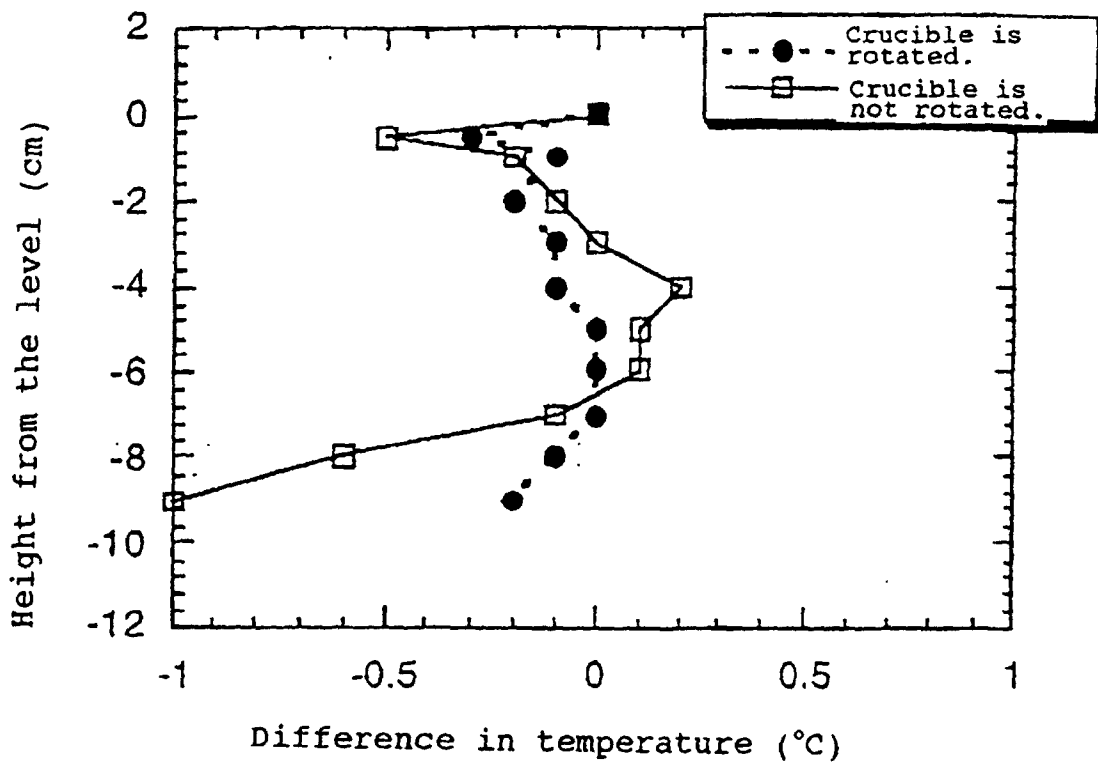


Fig. 7

