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(54) Title: PROCESS FOR PRODUCING MULTICRYSTALLINE SILICON INGOTS BY THE INDUCTION METHOD AND APPARATUS FOR CARRYING OUT THE SAME

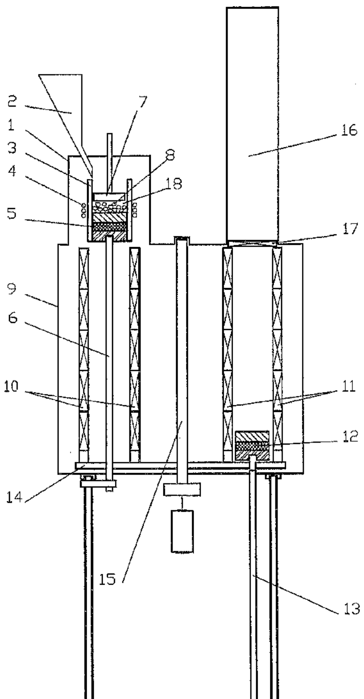

Fig. 1

(57) Abstract: This invention is related to obtaining multicrystalline silicon using induction method. The method comprises melting and casting of a pool in the form of a melting space, crystallization of a multicrystalline silicon ingot, and its controlled cooling by means of a heating equipment set. After the pool melting and casting is terminated, crystallization of the remaining part of the multicrystalline silicon ingot is finished along with the controlled cooling of the whole ingot; the ingot is then removed together with a movable bottom and the heating equipment set; and its controlled cooling continues. At the same time, another heating equipment set is supplied to the vacated place with another movable bottom; then the new movable bottom is moved into the water-cooled crucible; and the process steps are repeated in order to produce the next ingot. The method is implemented using an apparatus that additionally includes a platform installed in the controlled cooling compartment and designed to revolve on its axis. This platform holds at least two heating equipment sets. The proposed invention ensures increased output of multicrystalline silicon suitable for production of solar cells.

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PROCESS FOR PRODUCING MULTICRYSTALLINE SILICON INGOTS BY THE INDUCTION METHOD AND APPARATUS FOR CARRYING OUT THE SAME

Technical field

The invention relates to producing polycrystalline silicon, particularly to producing multicrystalline silicon by the induction method, and can be used in manufacture of solar cells from multicrystalline silicon.

Crystal silicon is used for producing solar cells to converse solar energy into electrical energy. Single-crystal silicon is usually used for this purpose.

Background Art

Recently completed research has demonstrated that polycrystalline silicon formed by the large crystals, so called multicrystalline silicon, allowed to reach the efficiency of converting solar energy into electric energy close to that of single-crystal silicon. Production capacity of equipment for producing multicrystalline silicon is several times higher than for single-crystal silicon, and its technology is easier than the technology for obtaining single-crystal silicon. The use of multicrystalline silicon enables to reduce the cost of solar panels and to start their production on the industrial level.

Currently in use is a process for producing multicrystalline silicon ingots by the induction method comprising a continuous supplying , and an induction melting of polycrystalline lump silicon batch material in a silicon melt-pool on a movable bed of a water-cooled crucible, casting the molten silicon to the shape of the melting space, and subsequent crystallizing a multicrystalline silicon ingot (US, 4572812). The melt-pool is contained within a skull formed using the water-cooled crucible consisting of the vertical copper tube sections cooled with water. The copper sections are separated by gaps and form a melting volume enclosed on the perimeter. Gaps between the sections permit inductor electromagnetic field to penetrate into the crucible melting volume. The melting volume can be shaped as a circle, a square, or a rectangle. During melting, a melt-pool fills up the entire transverse area of the crucible, thus resulting both in melting and casting of the silicon pool as an ingot of specific transverse size and shape. As the silicon batch melts, and the crucible movable bottom moves downward, the pool in the bottom part of the melt-pool is crystallizing. The speed of ingot movement corresponds to the speed of the lump batch melting in the upper part of the melt-pool. As a result of this known method, a long multicrystalline silicon ingot with a specified cross-section is produced and later on used in the production of solar cell plates.

A disadvantage of the above process for producing multicrystalline silicon ingots by induction method is the appearance of thermal stress in the ingot resulting in lower quality degradation of plates produced using such ingot. Thermal stresses in the ingot and in the plates made of such ingot results in decreased efficiency of energy conversion by solar cells formed of these plates. In addition, the output factor of good plates also decreases due to their rupture caused by thermal stresses.

The identified problems are resolved by the process of producing multicrystalline silicon ingots by induction method that is described in EP,1254861. According to this known process it is provided the additional heating of a silicon ingot obtained in the process of continuous casting by using the heaters located underneath the water-cooled crucible, and additional heating of the ingot by plasma discharge of a plasmatron located above the water-cooled crucible. At the same time, the plasma discharge is scanning over the pool surface. By using this process it is able to provide controlled cooling of the obtained ingot within the predetermined temperature gradient at length. Electric circuit for the plasmatron is looped at the silicon ingot through a special contact that is arranged underneath the place where the ingot exits the processing chamber. The method provides reducing the temperature gradient over the silicon ingot radius down to 9...7 °C/m, and thus results in achievement of a high efficiency (14.2...14.5%) of converting solar energy into electric energy for the plates made of this ingot .

However, during continuous melting and production of long ingots of multicrystalline silicon with a permanent supply of lump batch to the melt-pool, only at the beginning of the process. concentration of impurities in the melt-pool is fitting with the concentration of impurities in the loaded batch . Concentration of impurities in an ingot is defined by the segregation factor of each impurity. Inasmuch as the segregation factor for typical impurities in the source material is lower than 1, concentration of each impurity in the ingot is lower than its concentration in melt. As much as the ingot grows longer, concentration of impurities in the melt-pool increases due to their accumulation, and consequently their concentration in the produced multicrystalline ingot also increases. When concentration of impurities in the pool exceeds its limit established for each specific impurity, multicrystalline silicon becomes unsuitable for producing solar cells. The parts of the ingot with the concentration of impurities higher than the prescribed limit cannot be used for producing solar cells and are rejected, thereby significantly decreasing a fraction of produced solar cells with a high conversion efficiency.

The most relevant process to the claimed invention is a process of producing multicrystalline silicon ingots by the induction method described in EP, 1754806. This method comprises charging and start-up heating a lump silicon charge material in a controlled atmosphere on a movable bottom within the melting space of a water-cooled crucible, creating a bath of molten silicon, and subsequently melting and casting the molten silicon to the shape of the melting space, crystallizing a multicrystalline silicon ingot, and controllably cooling the silicon ingot using a heating equipment set. As the multicrystalline silicon ingot cools down, it is removed from the processing chamber via a gas seal which prevents atmospheric air from penetrating into the chamber, and is cut down into cut-to-length sections by a cutter. In order to increase the efficiency of the method, after a permissible limit of impurities concentration is reached in the pool, the melting process is stopped, the melt-pool is crystallized, and a separation device is drawn down into the crystallized ingot in the melting volume of the water-cooled crucible to block the melting volume and prevent impure silicon from the bottom surface of the separation device from entering the upper surface. At the same time, the initial lump batch of silicon is supplied onto the upper surface of the separation device, and the operations are repeated starting from supply and start heating of silicon lump batch.

The prior art method has the following drawbacks.

When induction melting and casting are stopped, if impurities reach their critical content in the pool in the process of obtaining a long ingot (e.g., 14 m long), the entire upper part of the ingot - approximately 2.5 m long - located inside the heating equipment and processing chamber (above the gas seal) should run the step of controlled cooling. For this purpose the controlled cooling is carrying out in the mode similar to the one used for the whole ingot and this step takes approximately 30 hours. In addition, each step of inserting the separation device into the furnace melting volume and resuming the process of melting and casting takes about 7.2 hours. During this period, induction melting and casting of the pool is not provided.

The step of inserting a separation device into a melting volume requires high precision, since even a small misalignment error during the installation of the separation device may result in its jamming and damage of the water-cooled crucible, consequently resulting in the compulsory termination of melting and casting.

Besides, inserting of a separation device made of foreign material - specifically silicon nitride, or graphite - into the melt-pool gives rise to contamination of the lower part of the ingot becomes impure, and as a result to reduce quality and output factor of a good silicon . The need to resume melting on the top of already produced silicon ingot leads to the need of stopping its movement into the heating equipment and keeping it inside the water-cooled crucible for a long time. This results in uncontrolled cooling of this part of the ingot, appearance of thermal stresses and microcracks in that area, and consequently, in the need of rejecting the upper part of the ingot.

EP,1754806 describes a system for producing multicrystalline silicon ingots comprising a chamber with a built-in water-cooled crucible with a movable bottom, and a heating equipment set for controlled cooling of the ingot . The water-cooled crucible includes the isolated sections made of electricity- and heat-conducting material – typically, copper - cooled by a water flow. The water-cooled crucible is enveloped by an inductor and connected to a batch bin. The movable bottom is designed to move up and down along the heating equipment set. Besides, the system is provided with a separation device (divider) that can be installed on a crystallized ingot in the melting volume of the water-cooled crucible. Subsequent heating of a silicon lump batch, melting and casting can take place above the upper surface of the separation device.

A drawback of the current system is a low production capacity, especially when a batch with a high content of impurities is used.

The most relevant to the claimed invention is an apparatus for producing multicrystalline silicon ingots by the induction method which is disclosed in EP, 0349904. In this apparatus , a batch bin is connected with a chamber inside which the following equipment is installed: a water-cooled crucible enveloped by an inductor, a device for start-up heating of silicon lump batch, a movable bottom with a rod connected to moving equipment, and a controlled cooling compartment with a heating equipment set located underneath the water-cooled crucible. The movable bottom is arranged to move up and down along the heating equipment set.

The availability of heating equipment enables to control a speed of cooling the produced ingot as it moves continuously inside the controlled cooling compartment, thus achieving decrease of temperature gradients over the ingot length within 5 to 10 ^{0}C/cm.

A drawback of the prior art apparatus is degradation of ingot quality and decrease of production output when using silicon lump batch with a high content of impurities, for example, metallurgical-grade silicon characterized by an increased content of iron (Fe) and aluminum (Al) admixtures. Operational capabilities of solar cells decreases when a content of Fe exceeds 0.01 ppmw, and Al - 0.1 ppmw. Due to segregating the identified impurities, satisfactory quality of silicon is maintained within a limited length of the produced multicrystalline silicon ingot, not more than 2-4 m, depending on the amount of impurities. However, upon producing the ingots of the identified length time period required to remove the ingot from the water-cooled crucible and the controlled cooling compartment increases relative to the time of induction melting and casting, and the equipment capacity decreases.

Summary of the invention

An object of the invention is to improve the process for producing multicrystalline silicon ingots by the induction which would lead to increased output of multicrystalline silicon suitable for producing solar cells due to the proposed relocation of an ingot and heating equipment with a movable bottom during controlled cooling.

Another object of the invention is to upgrade the apparatus for producing multicrystalline silicon which due to the proposed design would increase production capacity of obtaining multicrystalline silicon ingots suitable for producing solar cells.

The proposed method of obtaining multicrystalline silicon ingots using induction comprises the steps of charging and start-up heating a lump silicon charge material in a controlled atmosphere on a movable bottom within the melting space of a water-cooled crucible, creating a bath of molten silicon, and subsequently melting and casting the molten silicon to the shape of the melting space, crystallizing a multicrystalline silicon ingot, and controllably cooling the silicon ingot using a heating equipment set, terminating the steps of melting and casting when the content of impurities in the molten silicon becomes critical, and repeating the_process steps starting from the steps of charging and start-up heating the lump silicon charge material. According to the invention, upon terminating the steps of melting and casting, the step of crystallizing the remaining part of the multicrystalline silicon ingot is finished as the whole ingot is controllably cooled; upon completion of crystallization, the multicrystalline silicon ingot is removed together with the movable bottom and the heating equipment set and further cooled in a controllable manner; and at the same time, another heating equipment set including another movable bottom is supplied to the vacated space; then said another movable bottom is moved into the water-cooled crucible; and the process steps are repeated in order to produce the next ingot.

A simultaneous removal of the multicrystalline silicon ingot with the heating equipment set and the supply of another heating equipment set together with another movable bottom is done by a 180-degree rotary turn is the preferred embodiment of the claimed process.

In the another aspect of the claimed invention it is proposed an apparatus for carrying out the process for producing multicrystalline silicon ingots by the induction method comprising a chamber connected to a charge bin, the chamber including a water-cooled crucible enveloped by an inductor, a device for start-up heating a lump silicon charge material, and a movable bottom with a rod connected to a movement means, as well as a controlled cooling compartment arranged underneath the water-cooled crucible and including a heating equipment set, the movable bottom being movable up and down along the heating equipment set. According to invention the claimed apparatus further comprises a platform installed in the controlled cooling compartment and capable of rotation about the axis whereon said heating equipment set is mounted; and the apparatus further comprising at least one more heating equipment set installed on the platform and another movable bottom arranged in said another heating equipment set together with a rod connected to a suitable movement means therefor.

In one of the preffered embodiment of the invention the apparatus may contain two heating equipment sets installed on the platform symmetrical around an axis of_rotation. At the same time, each of the heating equipment sets has an algorithm for changing temperature vertically to ensure an established temperature gradient is maintained in the produced multicrystalline silicon ingot.

It has been determined experimentally that by simultaneous implementation of induction melting and casting first, together with the controlled cooling, and then, after stopping of the induction melting, ingot casting and crystallization as well as by continuation of the controlled cooling of the ingot in the process of preparing and beginning the production of the next ingot, it is provided the production of ingots with a controlled content of impurities.

Besides, the proposed staging of the controlled cooling of a multicrystalline silicon ingot which can be implemented without interrupting the overall cooling process allows to be flexible in regulating the length of the produced ingot depending on the amount of impurities in the starting batch.

Hence, the claimed process is effective process, characterized by a high production output of ingots suitable for producing solar batteries, and can be used for batches with a high content of impurities.

A reduced period of induction melting downtime, and lack of dependency of a part of the controlled cooling process on melting and casting processes permit to increase capacity of the system for producing multicrystalline silicon.

Brief Description of Drawings

The invention is demonstrated but not limited by the accompanying drawings. The drawings illustrate the apparatus for producing multicrystalline silicon ingots by the induction method as having two heating equipment sets for controlled cooling of multicrystalline silicon, in which:

Fig. 1 is an embodiment of the apparatus at a start-up heating step:

Fig. 2 is the apparatus of Fig.1 at the step of induction melting and casting of a multicrystalline silicon ingot;

Fig. 3 illustrates the position of the multicrystalline silicon ingot before movement;

Fig. 4 illustrates the position of the multicrystalline silicon ingot after movement;

Fig. 5 is the apparatus of Fig.1 at the step of induction melting and casting of the next multicrystalline silicon ingot, and removal of the previous multicrystalline silicon ingot

Fig. 6 illustrates a chart demonstrating changes of iron concentration over the length of the multicrystalline silicon ingot with a square cross-section (a side is 337 mm long) for the starting material specified in Table .

Best Mode for Carrying Out the Invention

The apparatus for producing multicrystalline silicon ingots by the using induction method as demonstrated on Fig.1 comprises a chamber 1 connected to a charging bin 2, a water-cooled crucible 3 enveloped by an inductor 4. A movable bottom 5 with a rod 6 connected to the movement devices as well as a start-up heating device 7 of lump silicon charge material 8 are installed inside the chamber 1. The start-up heating device 7 is made of conductive material, e.g., graphite. The water-cooled crucible 3 is made of copper sections cooled with water. In the chamber 1, underneath the water-cooled crucible, a controlled cooling compartment 9 including a a heating equipment set 10 and a similar heating equipment set 11 is located. The movable bottom 5 is arranged to move vertically along the heating equipment set 10. The heating equipment set 11 consists of a movable bottom 12 with a rod 13 connected to the corresponding movement devices. The movable bottom 12 can move vertically along the heating equipment set 11. The heating equipment sets 10 and 11 are installed on a platform 14. The platform 14 is placed in the controlled cooling compartment 9, and is able to rotate about axis 15. The chamber 1 is connected to a discharge device 16 through a gas seal 17.

The apparatus works as follows.

In the chamber 1, under controlled atmosphere, the movable bottom 5 is moved toward the water-cooled crucible 3, and a high-frequency electromagnetic field is created by inductor 4. The lump silicon charge material 8 is downloaded from charge bin 2 into a melting space 18 created by the water-cooled crucible 3 and the movable bottom 5. Then the start-up heating device 7 is inserted into the melting space 18 located inside a high-frequency electromagnetic field created by the inductor 4. The start-up heating device 7 gets heated up, and the lump silicon charge material 8 warms up and begins melting under the influence of radiated heat and electromagnetic field created by the inductor 4 (Fig. 1).

The start-up heating device 7 is removed from the electromagnetic field created by the inductor 4, while in the melting space 18 a melt-pool 19 is produced in the form of its lateral section. As a result of heat emission along the periphery of melt-pool 19, the pool is crystallized, and skull is formed which prevents the pool from spilling from the melting space 18. After forming of the melt-pool 19 , a lump silicon charge material 8 is permanently supplied onto its surface from the charging bin 2. The lump silicon charge material 8 melts down, and at the same time, the movable bottom 5 together with the melt-pool 19 with skull is steadily moved downward. The movement is at the speed which allows the melt-pool 19 to remain at the unchanged level relative to the inductor 4 and the water-cooled crucible 3, crystallization of the multicrystalline silicon ingot 20 being effected continuously in the lower part of the melt-pool. Thereby formed multicrystalline silicon ingot 20 is steadily moved down into the controlled cooling compartment 9 toward the heating equipment set 10, where its controlled cooling is performed and thermal stresses are removed (Fig. 2).

If the content of impurities in the pool becomes critical, the supply of the lump silicon charge material 8 is stopped, the electromagnetic field of the inductor 4 is removed, and the induction melting and pool casting are stopped. The produced multicrystalline silicon ingot 20 is removed from the water-cooled crucible 3, and placed into the heating equipment set 10 to terminate crystallization (Fig. 3). Then the multicrystalline silicon ingot 20 is transferred together with the movable bottom 5 and the heating equipment set 10 (fig. 4) by rotating the platform 14 and the controlled cooling of the ingot continues. Such movement can be accomplished, for example, by a 180-degrees rotary turn of the platform 14 around the axis 15.

Simultaneously with relocating the multicrystalline silicon ingot 20 together with the movable bottom 5 and the heating equipment set 10, the heating equipment set 11 with the movable bottom 12 is transferred to the vacated space (Fig. 4). The heating equipment sets 10 and 11 are electrically switched over and the algorithm of temperature regulation is changed accordingly.

The movable bottom 12 is transferred to the water-cooled crucible 3, defining a new melting space 21, and the process steps are repeated starting from the supply and start-up heating of lump silicon charge material , in order to obtain a next ingot 22 (Fig. 5). The cooled down multicrystalline silicon ingot 20 located inside the heating equipment set 10 is moved upward on the movable bottom 5 into the discharge device 16.

An example of how multicrystalline silicon ingots can be obtained by the induction method according to the process of the invention with the use of the apparatus described above follows.

Example.

Multicrystalline silicon ingots were obtained using the apparatus with a melting space of a square cross-section and a side length of 340 mm. This apparatus allows to produce multicrystalline silicon ingots with a square cross-section and a side length of 337 mm.

As a starting material for the production of multicrystalline silicon ingots , metallurgical-grade bulk silicon was used l with the following usual admixtures such as boron (B), phosphorus (P), iron (Fe), and aluminum (Al). The content of impurities in the starting material is shown in the Table. Additionally, an alloy was used to maintain specific resistance within the range of 0.8-1.2 Ohm×cm.

Table

Content of usual impurities in metallurgical-grade lump silicon batch

Impurity	Concentration at/cm^3
B	$5.46*10^{16}$
P	$7.07*10^{16}$
Al	$2.29*10^{16}$
Fe	$1.30*10^{17}$

Critical content of impurities in a pool depending on a composition of the starting material was determined by the experimental calculations. Based on this data, dependence between concentration of iron admixtures (the impurity determining quality of multicrystalline silicon) and the length of multicrystalline silicon ingot of square cross-section and the side length of 337 mm was found for the selected starting material (Fig. 6).

For the starting material with the content of impurities listed in the Table, obtained multicrystalline silicon ingots of square cross-section and the side length of 337 mm, the critical content of impurities is reached when the ingot gets 2.0 ... 2.5 m long, an optimal length of the ingot being 2.0 ... 2.3 m.

Inside an air-tight chamber, a water-cooled crucible is installed and enveloped by 3- turn inductor of 140 mm in height connected to a source of power with an operating frequency of 10 kHz, and a movable bottom made in the form of a square silicon plate of 40 mm thick with a side length of 338 mm which is placed using a thermal insulation disk on a heat-resistant steel platform, and is attached to the rod via a plug-type connection. In the controlled cooling compartment under the water-cooled crucible on the platform the first heating equipment set is installed having a heating space of a square cross-section and the side length of 380 mm. Symmetrically, the second heating equipment set is installed on the platform, having a heating space of a square cross-section and the side length of 380 mm. The heating space of each heating equipment set is of 2.4 m in height . Each heating equipment set is provided with graphite heater elements connected to the industrial frequency transformer through the current controllers, thus allowing to ensure controlled changes of the temperature field vertically. The second heating equipment set has another movable bottom installed using a thermal insulation disk on a heat-resistant steel platform, and attached to the rod via a plug-type connection.

The chamber was vacuumized and filled with argon. In the melting space of the water-cooled crucible, high-frequency electromagnetic field was created using the inductor. Lump silicon charge material was loaded into the melting space from a charging bin, and a start-up heating device in the form of a graphite disk was inserted. The start-up heating device was heated up, and under the influence of heat emission and electromagnetic field, lump silicon starting material was heated to a temperature of 800-1100 °C, and started melting. The start-up heating device was removed from the inductor electromagnetic field. A melt-pool was formed inside the melting space , which expanded and reached the walls of a cold crucible, i.e. the pool was cast in the form of the water-cooled crucible. At the same time, a skull was formed and prevented the pool from spilling and contained the melt-pool. Lump silicon starting material was then supplied continuously onto the pool surface from the charging bin. The lump silicon starting material was melted, the movable bottom was steadily moved down, and a multicrystalline silicon ingot was formed.

As the multicrystalline silicon ingot was melted, cast and formed, it was moved down into the heating space of the first heating equipment set, where the controlled cooling and removal of thermal stresses was carried out by warming up the multicrystalline silicon ingot - already crystallized and chilled – mainly in the corners of the ingot. The temperature was raised up to 1200°C with the following decrease of a temperature with the gradient of no more than 7°C over the ingot length.

Once the content of impurities in the pool becomes critical when the ingot was approximately 2.2 m long (Fig. 6), the supply of lump silicon starting material was stopped, the inductor was shut down, and induction melting and pool casting were ceased. As a result, the melt-pool was crystallized. The obtained multicrystalline silicon ingot, 2.2 m long, was removed from the water-cooled crucible and completely inserted into the heating space of the first heating equipment set. Then the first heating equipment set was disconnected from the current controllers, and by 180-degree turn of the platform round its axis, the multicrystalline silicon ingot was shifted together with the movable bottom and the first heating equipment set.

Upon completion of the platform turning, the heating equipment sets were exchanged - the second heating equipment set together with the other movable bottom turned out to be underneath the water-cooled crucible, whereas the first heating equipment set with the multicrystalline silicon ingot located inside its heating space occurred in the place where the second heating equipment set used to be. Then both heating equipment sets were connected to the current controllers- the second heating equipment set located under the water-cooled crucible was connected using the melting algorithm, and the first heating equipment set located out of contact with the water-cooled crucible – using the algorithm for the controlled cooling of the multicrystalline ingot. After the multicrystalline ingot had been cooled down to a temperature below 250°C, it was lifted on the movable bottom into the discharge device.

The movable bottom located inside the heating space of the second heating equipment set, after completion of the platform turning , was transferred to the water-cooled crucible, and the process steps were repeated starting from supplying and start-up heating of the lump charge until obtaining the next ingot.

Testing of the pilot apparatus in accordance with the claimed invention resulted in an average output of 18 kg per hour of multicrystalline silicon in the form of an ingot with a cross-section of 337 x 337 mm^2. At the same time, it was established that neither the upper, nor the lower part of the produced ingots has any microcracks or additional impurities, and they are suitable for the production of solar cell plates, with the exception of the residual melt-pool covering the upper part of each ingot over the length of about 160 mm. The output of a good product was 93% of the obtained ingot.

Experimental melting operations based on the method described in EP,1754806 using a graphite separation device allowed to reach the equipment capacity of 16.2 kg/h for a similar starting material and a similar size ingot with three operations of inserting the separation device. At the same time, in addition to the residual melt-pool, about 50 to 70 mm of an ingot were rejected due to microcracks. Graphite impurities were found in the lower part of each ingot. Such areas were up to 50 mm long and were rejected. As a result, the output of a good product was 88% of the obtained ingot.

The proposed invention ensures increased output of multicrystalline silicon suitable for solar cell fabrication.

CLAIMS

1. A process for producing multicrystalline silicon ingots by the induction method, comprising charging and start-up heating a lump silicon charge material in a controlled atmosphere on a movable bottom within the melting space of a water-cooled crucible, creating a bath of molten silicon, and subsequently melting and casting the molten silicon to the shape of the melting space, crystallizing a multicrystalline silicon ingot, and controllably cooling the silicon ingot using a heating equipment set, terminating the steps of melting and casting when the content of impurities in the molten silicon becomes critical, and repeating the process steps starting from the steps of charging and start-up heating the lump silicon charge material, **characterized in that** upon terminating the steps of melting and casting, the step of crystallizing the remaining part of the multicrystalline silicon ingot is finished as the whole ingot is controllably cooled; upon completion of crystallization, the multicrystalline silicon ingot is removed together with the movable bottom and the heating equipment set and further cooled in a controllable manner; and at the same time, another heating equipment set including another movable bottom is supplied to the vacated space; then said another movable bottom is moved into the water-cooled crucible; and the process steps are repeated in order to produce the next ingot.

2. The process according to Claim 1, **characterized in that** simultaneous removal of the multicrystalline silicon ingot with the heating equipment set and the supply of another heating equipment set together with another movable bottom is done by a 180-degree rotary turn.

3. An apparatus for producing multicrystalline silicon ingots by the induction method, comprising a chamber connected to a charge bin, the chamber including a water-cooled crucible enveloped by an inductor, a device for start-up heating a lump silicon charge material, and a movable bottom with a rod connected to a movement means, as well as a controlled cooling compartment arranged underneath the water-cooled crucible and including a heating equipment set, the movable bottom being movable up and down along the heating equipment set, **characterized by** further comprising a platform installed in the controlled cooling compartment and capable of rotation about the axis whereon said heating equipment set is mounted; and the apparatus further comprising at least one more heating equipment set installed on the platform and another movable

bottom arranged in said another heating equipment set together with a rod connected to a suitable movement means therefor.

4. The apparatus according to Claim 3, **characterized in that** it contains two heating equipment sets installed on the platform symmetrical around an axis of_rotation.

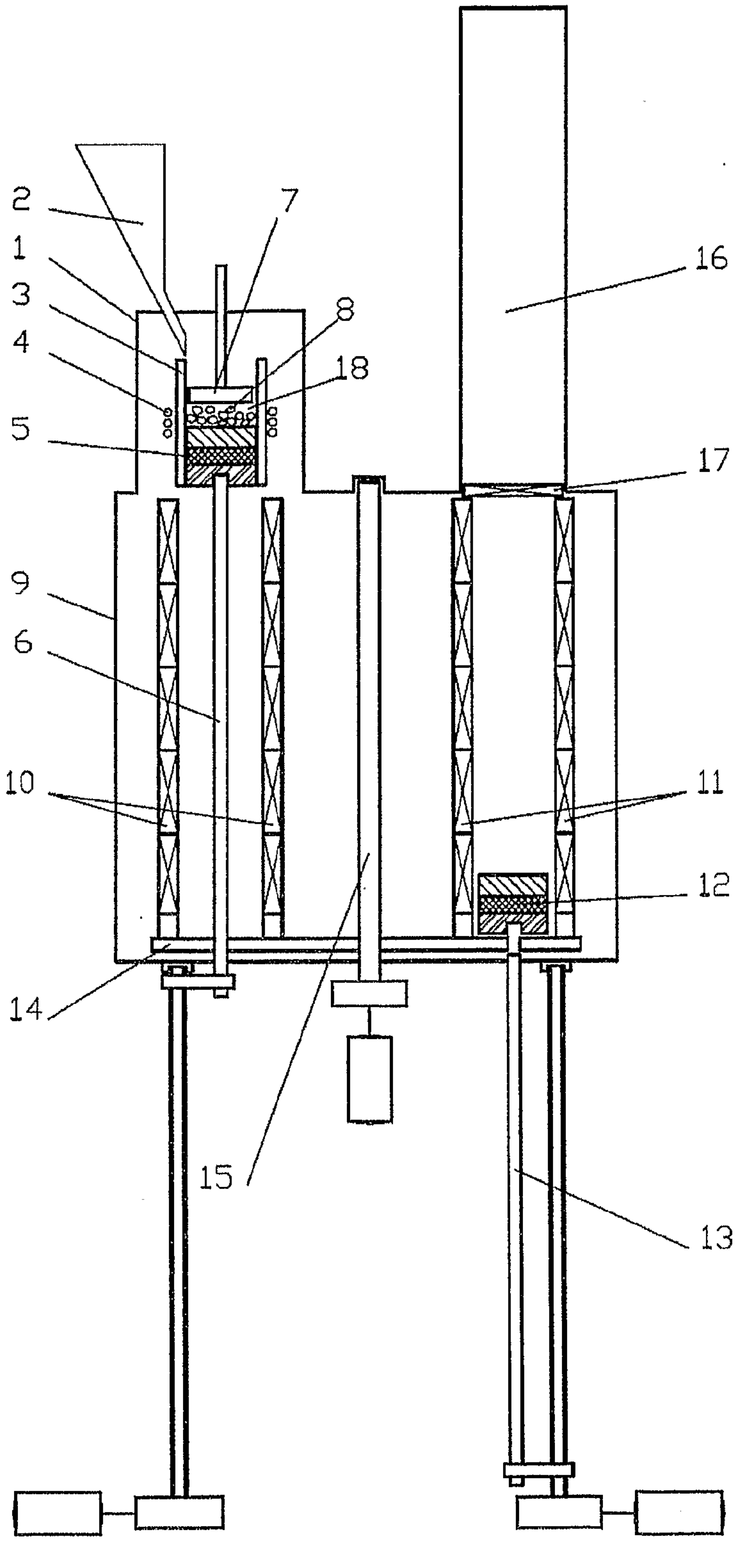

Fig. 1

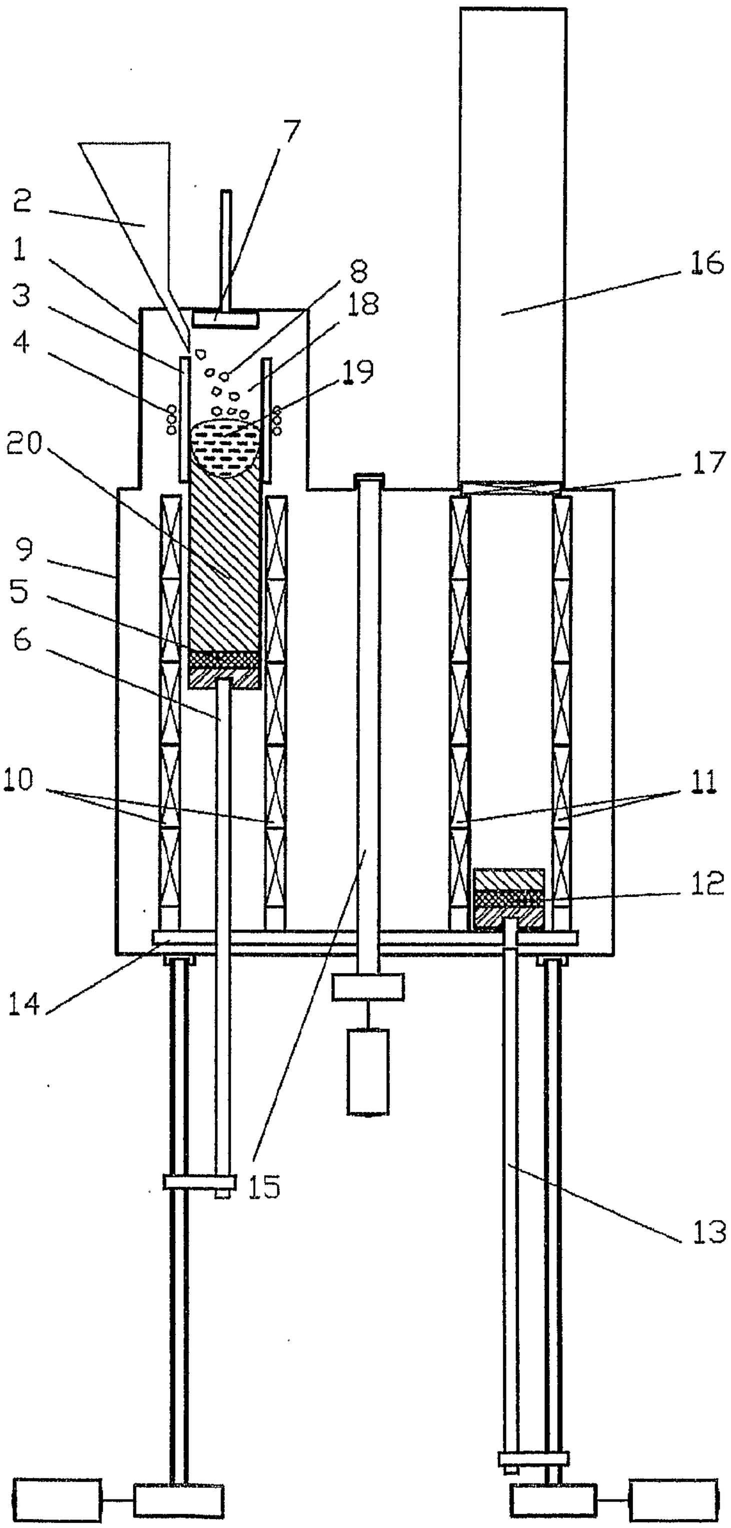

Fig. 2

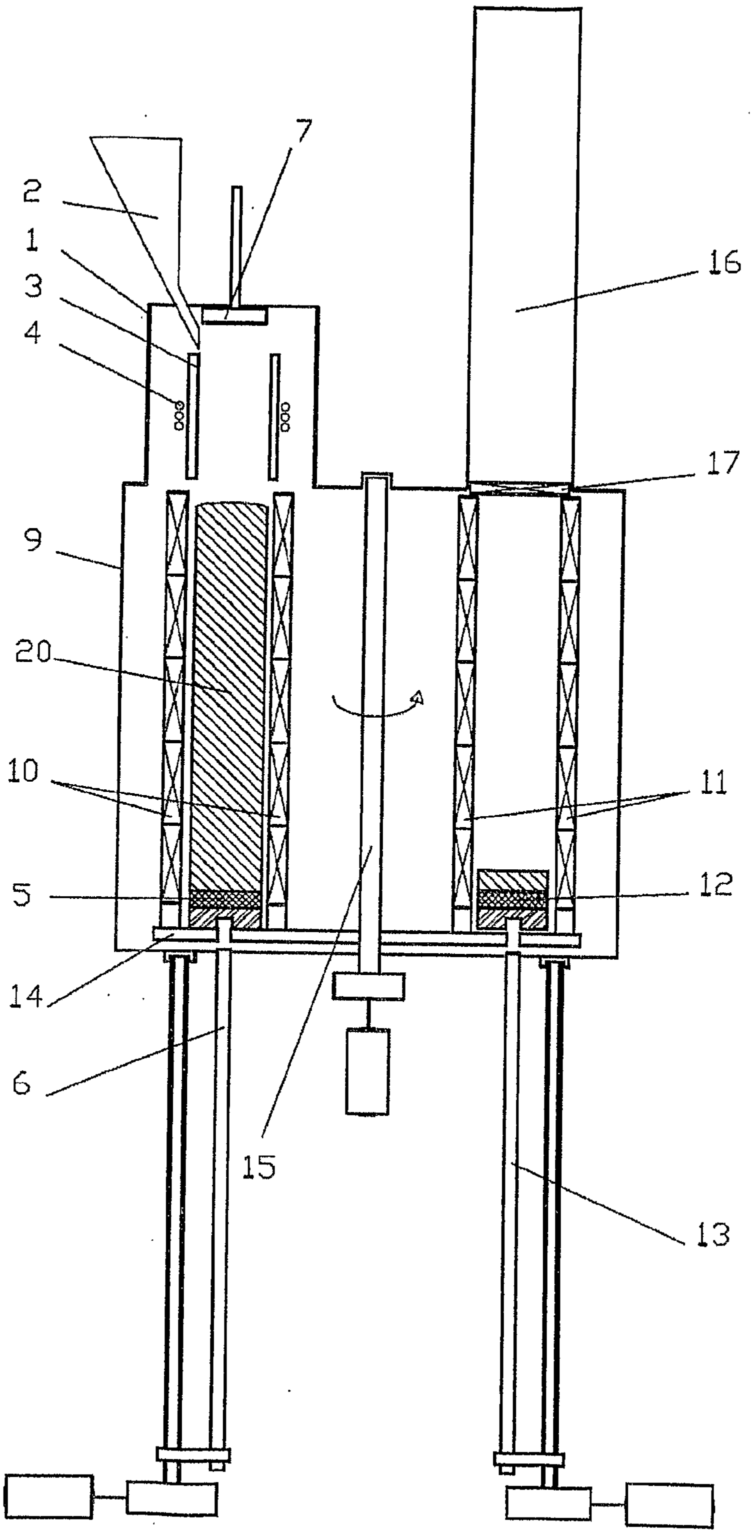

Fig. 3

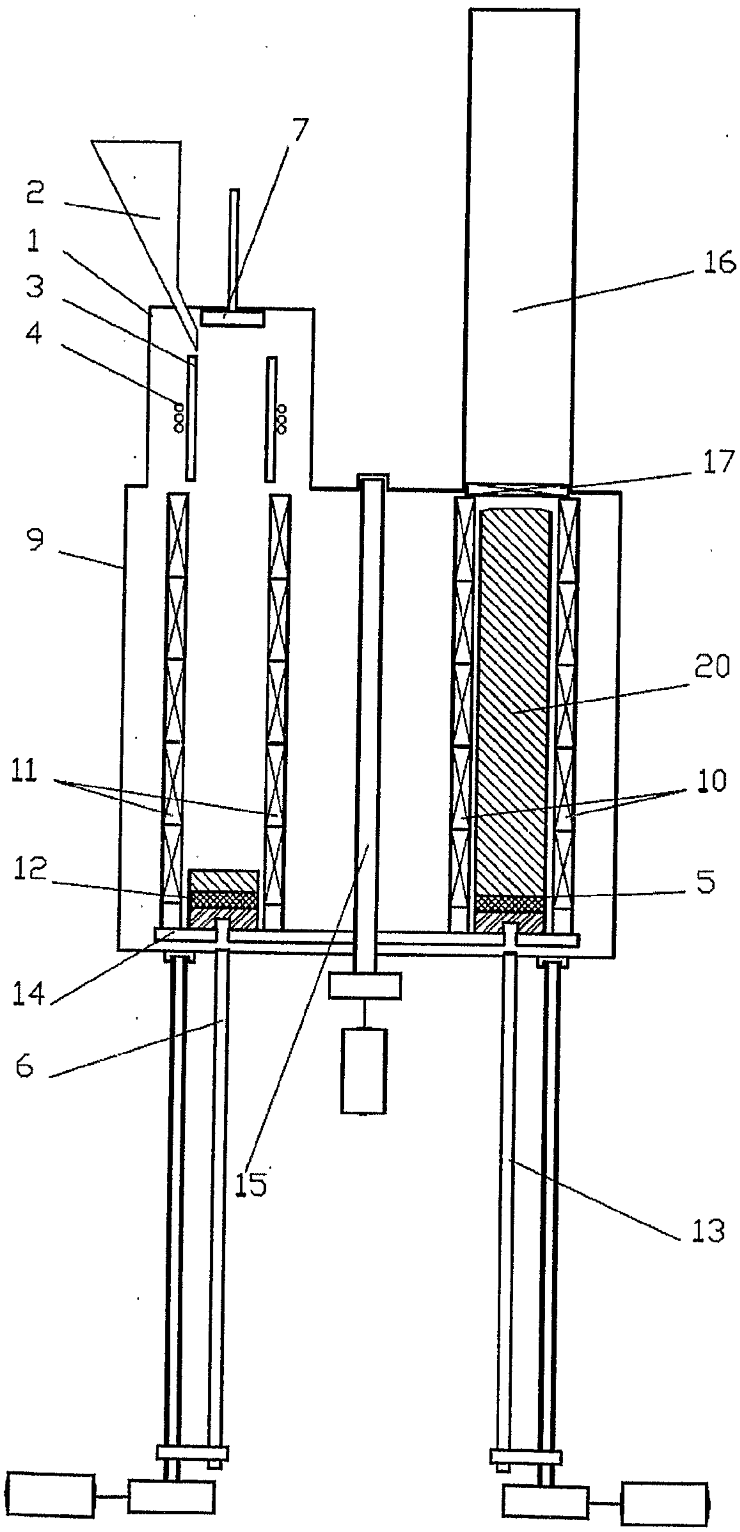

Fig. 4

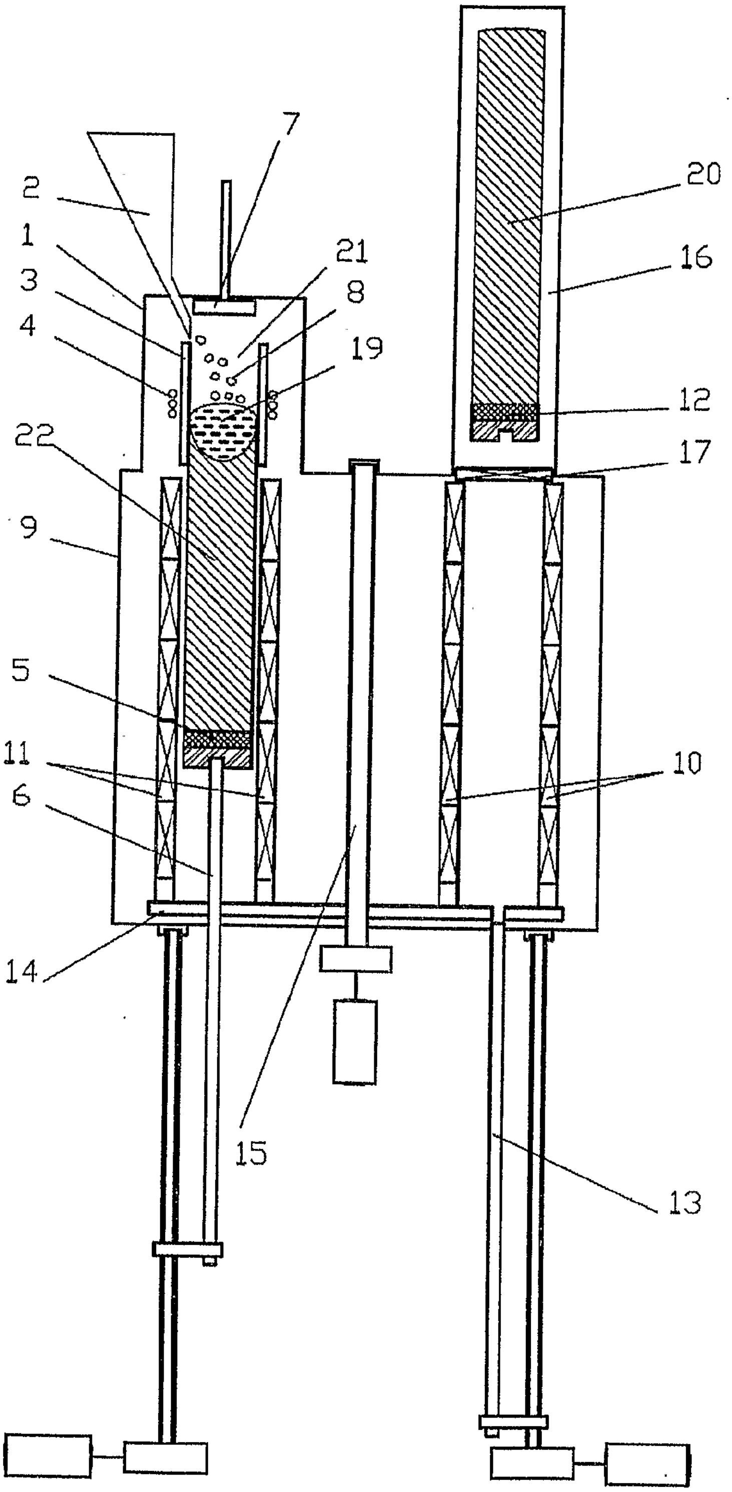

Fig. 5

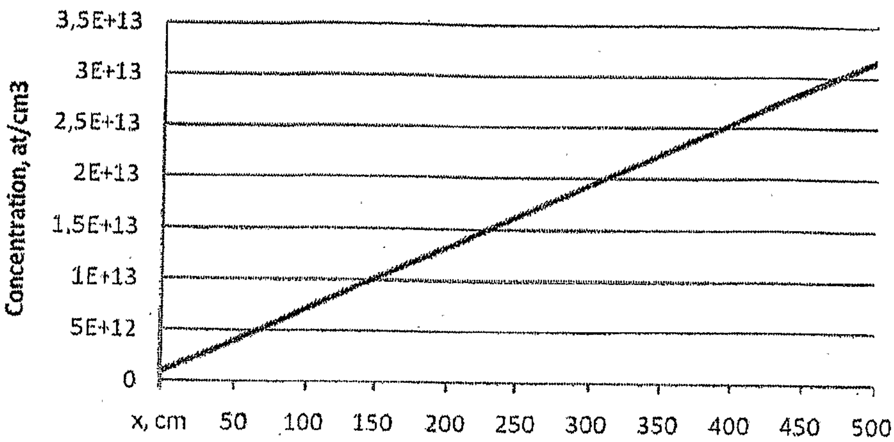
3,5E+13
3E+13
2,5E+13
2E+13
1,5E+13
1E+13
5E+12
0
Concentration, at/cm3
x, cm
50
100
150
200
250
300
350
400
450
500

Fig.6

INTERNATIONAL SEARCH REPORT

International application No
PCT/UA2009/000067

A. CLASSIFICATION OF SUBJECT MATTER
INV. B22D27/02 C01B33/037 C30B29/06
ADD.

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
B22D C01B C30B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, CHEM ABS Data, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	EP 1 754 806 A1 (SUMCO SOLAR CORP [JP]) 21 February 2007 (2007-02-21) cited in the application paragraphs [0027] - [0031], [0034] - [0038]; figures 1,2	1-4
A	US 2008/179037 A1 (YOSHIHARA MITSUO [JP] ET AL) 31 July 2008 (2008-07-31) paragraphs [0048] - [0051], [0054]; figure 4	1-4
A	US 2008/283211 A1 (HONG YONG-QIANG [CN]) 20 November 2008 (2008-11-20) paragraphs [0007] - [0009], [0013] - [0023]; figures 1,2	1-4

☐ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

* Special categories of cited documents :

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"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

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"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
27 April 2010	06/05/2010

Name and mailing address of the ISA/	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL – 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Werner, Håkan

INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
PCT/UA2009/000067

Patent document cited in search report		Publication date	Patent family member(s)			Publication date
EP 1754806	A1	21-02-2007	JP	2007051026	A	01-03-2007
			US	2007039544	A1	22-02-2007
US 2008179037	A1	31-07-2008	JP	2008156166	A	10-07-2008
US 2008283211	A1	20-11-2008	BR	PI0801172	A2	06-01-2009
			CA	2630724	A1	16-11-2008
			CN	101307487	A	19-11-2008
			FR	2916206	A1	21-11-2008