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**Datasheet for the decision  
of 14 November 2013**

**Case Number:** T 1216/11 - 3.2.08  
**Application Number:** 99103540.3  
**Publication Number:** 939146  
**IPC:** C30B 11/00, C30B 29/06  
**Language of the proceedings:** EN

**Title of invention:**

Method for producing silicon ingot having directional solidification structure and apparatus for producing the same

**Patent Proprietor:**

mitsubishi materials corporation

**Opponent:**

ALD Vacuum Technologies GmbH

**Headword:**

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**Relevant legal provisions:**

EPC Art. 56

**Keyword:**

"Inventive step (no)"

**Decisions cited:**

-

**Catchword:**

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Case Number: T 1216/11 - 3.2.08

**D E C I S I O N**  
of the Technical Board of Appeal 3.2.08  
of 14 November 2013

**Appellant:**  
(Opponent)

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**Decision under appeal:**

Interlocutory decision of the Opposition  
Division of the European Patent Office posted  
21 March 2011 concerning maintenance of  
European patent No. 939146 in amended form.

**Composition of the Board:**

**Chairman:** T. Kriner  
**Members:** R. Ries  
D. T. Keeling

## Summary of Facts and Submissions

- I. By its interlocutory decision dispatched on 21 March 2011, the opposition division held that the subject matter of the claims according to the auxiliary request then on file met the requirements of the EPC and that the European patent No 0 939 146 could be maintained in amended form on the basis of this request.
- II. The appellant (opponent) lodged an appeal against this decision on 31 May 2011, paying the appeal fee on the same day. The statement setting out the grounds of appeal was filed on 29 July 2011.
- III. On appeal, the parties referred to the following documents:
- D1: DE-A-33 23 896,  
D2: US-A-4 175 610,  
D6: EP-A-0 810 305.
- IV. Oral proceedings took place before the Board on 14 November 2013. The following requests were made:
- The appellant requested that the decision under appeal be set aside and European patent No 0 939 146 be revoked.
  - The respondent (patent proprietor) requested that the decision that the appeal be dismissed.
- V. Independent claim 1 upheld by the opposition division reads as follows:

"1. Method for producing a silicon ingot having a directional solidification structure, especially for producing silicon substrates for use in photovoltaic solar cells, the method, comprising the steps of: placing a silicon raw material (2) into a silica crucible (14) of a melting device constructed by mounting a chill plate (15) capable of chilling with a refrigerant on an underfloor heater (13), mounting the crucible on the chill plate, providing an overhead heater (16) over the crucible (14), and surrounding the circumference of the crucible with a heat insulator (17) with carbon in the heat insulator, heat-melting the silicon raw material (2) by flowing an electrical current through the underfloor heater (13) and the overhead heater (16) while halting or decreasing the electric current through the underfloor heater after the silicon raw material has been completely melted, the molten silicon (8) being chilled from the bottom of the crucible by chilling the chill plate by feeding the refrigerant; and intermittently or continuously lowering the temperature of the overhead heater (16) by intermittently or continuously decreasing the electric current through the overhead heater (16) along with halting the electric current or decreasing the electric power through the underfloor heater (13), wherein the silicon raw material in the crucible (14) is maintained under an inert gas atmosphere during heat melting by inert gas being directed towards the surface of the molten silicon in the crucible by the inert gas feed device (23)."

VI. The appellant's arguments relevant to the present decision can be summarized as follows:

Document D1 as the closest prior art disclosed all the technical features of the claimed process except for the step of directing inert gas into the crucible towards the surface of the molten silicon during heat melting. Using a quartz crucible in the claimed process to melt and hold liquid silicon was conventional practice in the art, as mentioned for instance in documents D6 and D2.

The essential problem underlying the patent resided therefore in preventing contamination of the molten Si by SiC which formed when carbon (comprised in the heat insulation material) reacted with silicon monoxide  $\text{SiO}_{(\text{gas})}$ . In the first place this problem originated from using a quartz crucible because  $\text{SiO}_{\text{gas}}$  was only formed when  $\text{SiO}_2$  constituting the crucible reacted with Si. This interaction was disclosed for instance in document D6, page 2, lines 31 to 51.

The skilled person, faced with the problem of avoiding undesirable reactions between the silicon melt in the quartz crucible and  $\text{SiO}_{\text{gas}}$  and  $\text{CO}_{\text{gas}}$  contained in the furnace atmosphere, would protect the molten silicon by feeding an inert gas into the crucible on the surface of the melt, as depicted in D6, Figures 1 and 2 and described on page 2, lines 16 to 22 and page 4, lines 51 to 54. For the same reason, the casting station shown in the Figure of D2 was blanketed continuously with an inert gas, such as argon (D2, column 2, lines 45 to 48). Acting in that way to cope with the technical problem addressed in the patent amounted to

nothing more than what was well known to and carried out by those skilled in the art. The process set out in claim 1 was, therefore, obvious for the skilled person and did not involve an inventive step.

VII. The respondent's arguments relevant to the present decision can be summarized as follows:

Document D1 neither disclosed the material of the crucible nor the step of maintaining the crucible under an inert gas atmosphere during heat melting by directing inert gas towards the surface of the molten silicon in the crucible.

Contrary to the appellant's arguments, it was not conventional practice in the art to use a quartz ( $\text{SiO}_2$ ) crucible as required for the claimed process. Rather, a crucible made of graphite could also be selected, as described in the process of D2 where molten silicon was poured into a mould made of graphite.

Starting from D1 as the closest prior art, the problem underlying the patent was to avoid the formation of SiC in the Si-melt. The reason for doing so was to improve the photovoltaic conversion efficiency of solar cells, which was adversely affected by using silicon ingots contaminated with SiC. Moreover, the orientation of the directionally solidified structure of the Si-ingots was improved when SiC was absent, as shown in Table 1 of the patent. This problem was solved by feeding inert gas towards the surface of the molten silicon held in the quartz crucible.

Although in the process of D2 the casting station comprising a melting vessel with molten silicon was blanketed continuously with an inert gas in order to prevent the deposition of SiO, there was no hint anywhere in this document that the formation of SiC in the molten silicon could be prevented by this step. Thus, document D2 was not concerned with the technical problem addressed in the patent.

Document D6 dealt with the Czochralski-process and an apparatus for pulling silicon single crystals. Hence, D6 was directed to a different crystal growth technique. It was true that D6 described the chemical reactions which occurred between the crucible material (SiO<sub>2</sub>), the Si-melt and carbon existing in the heat-isolating material of the furnace and, more particularly, how SiC was formed inside the furnace. However, this document was essentially concerned with preventing the formation of SiC in the upper regions of the temperature keeping cylindrical body and the crucible due to the carbon existing in these regions because the resulting SiC could create cracks in the cylindrical body or the crucible protection part when the apparatus was cooled. Specifically, the reaction of Si<sub>vapour</sub> and SiO<sub>gas</sub> with the carbonaceous substrate of the heat isolating material was prevented by a layer of thermally decomposed carbon (D6, page 4, line 55 to page 5, line 7). D6 did not however provide relevant technical information with respect to preventing the formation of SiC in the silicon melt. Hence, D6 did not address the problem underlying the patent either. Consequently, the technical teaching of D1 combined neither with that of D2 nor with that of D6 led in an obvious manner to the claimed process.

The subject matter of claim 1 therefore involved an inventive step.

### **Reasons for the Decision**

1. The appeal is admissible.
2. The closest prior art
  - 2.1 It was common ground to the parties and the Board that document D1 qualified as being the closest prior art. Like the patent at issue, D1 is concerned with a process for producing silicon ingots having a directionally solidified structure (D1, page 4, single paragraph; page 8, lines 4 to 6).
  - 2.2 The process disclosed in D1 is carried out in the apparatus depicted in Figure 2 and includes the use of crucibles (3) mounted on a graphite chill plate (5), overhead and underfloor heaters (11) and (11a) provided under and over the crucibles, and a heat insulator (18) made of a graphite felt layer surrounding the crucibles and the heaters (D1, page 12, first line to page 13, line 8; page 14, line 18 to page 15, line 2). The process further comprises the steps of placing silicon raw material in the crucibles, mounting the crucibles on the chill plate, melting the raw material by flowing electric current through the overhead and underfloor heaters, decreasing the electrical current through the underfloor heater when the raw material is completely melted and chilling the molten silicon by the chill plate which is cooled by flowing a cooling medium



through the chill plate (D1, page 15, line 15 to line 21).

Although the process of document D1 remains silent on the material for the crucibles for melting the silicon raw material, it is clear for the skilled person that the only practical material for this purpose is quartz. It is common practice in the technical field of producing silicon castings and ingots to heat-melt and maintain the molten silicon in a quartz or silica ( $\text{SiO}_2$ ) crucible. Contrary to the respondent's view, this general finding is corroborated e.g. by document D6 disclosing on page 2, lines 26 to 33 that in conventional apparatuses for producing silicon single crystals the crucible's inner portion, which comes into direct contact with the silicon melt, is made of quartz ( $\text{SiO}_2$ ) in order to prevent the inclusion of impurities such as metals or the like into the melt. The remaining outer portion of the crucible is generally made of graphite or carbon-bonded carbon fibre composite (C/C-composite) which exhibit excellent heat resistance and prevent the quartz crucible from softening and deforming when it is heated to above  $1400^\circ\text{C}$  for forming the silicon melt.

The respondent's argument that D6 is concerned with a method and apparatus for pulling silicon single crystals and therefore relates to a different technical field is unconvincing because in either process of D6 and the patent at issue silicon is, in a first step, melted and held in a quartz crucible and in a second step, the melt is cooled to form an ingot having a directional solidification structure.

Likewise document D2, which is concerned with a process and apparatus for the semi-continuous production of silicon castings having a columnar structure and consisting of single crystal regions of crystals, discloses in column 2, lines 19 to 21 that the melting vessel containing the molten silicon is preferably made of quartz.

The respondent's argument that in the process of D2 the molten silicon is cast into a mould preferably made of graphite, is misconceived because the requirements on a casting mould for rapidly solidifying the silicon melt cannot be compared with the high demands which are made on the material for the melting vessel wherein the silicon raw material is heat-melted and maintained at a high temperature in the molten state for a long period of time.

In conclusion, using a quartz crucible for melting and maintaining molten silicon is common practice and therefore implicitly disclosed in document D1.

2.3 However, document D1 remains silent on the gas atmosphere inside the apparatus and possible interactions and chemical reactions between the gas atmosphere in the furnace, the crucible and the silicon melt which could adversely affect the purity of the molten silicon.

### 3. Problem and solution

Starting from the technical disclosure of document D1, the problem underlying the patent in suit is, therefore, seen to reside in preventing contamination of the

molten silicon through unwanted by-products originating from chemical reactions between the molten silicon, the crucible, the surrounding atmosphere and the carbonaceous material, e.g. the graphite felt of the heat insulator.

The problem is solved by feeding inert gas into the crucible towards the surface of the molten silicon during melting.

4. Inventive step; Article 56 EPC

The solution of the problem is, however, obvious to the skilled person, as is shown in the following.

- 4.1 It is generally known in metallurgy that unwanted chemical reactions between a molten metal and the surrounding atmosphere, which lead to deleterious by-products contaminating the molten metal, can be impeded by providing a protective blanket of inert gas over the surface of the melt. Reference is made in this context, by way of example, to the inert-gas-shielded metal welding technique which uses an inert gas blanket to prevent contamination of the molten metal formed in the welding pool. The purpose of the inert gas is to protect the weld area from coming into contact with atmospheric gases, such as oxygen, nitrogen and water vapour and their influence on the materials being welded. These atmospheric gases can reduce the quality of the weld or make the welding more difficult.

Protecting the surface of a liquid metal from interacting with the (furnace) atmosphere by an inert gas blanket is therefore well known.

4.2 Specifically when forming a silicon melt in conventional apparatuses, it is necessary to eliminate unwanted reaction products and to prevent the reaction of gases inside the furnace with the liquid surface of the melt and the single crystal. To this end an inert gas such as argon is fed from an inert gas source through a pipe system to the upper portion of the furnace vessel (D6, page 2, lines 16 to 22; Figure 1: prior art).

It is further known from document D6 that the  $\text{SiO}_2$  constituting the quartz crucible and the Si-melt in the crucible react to form SiO gas, as described by the formula  $\text{SiO}_2 + \text{Si} \rightarrow 2\text{SiO}_{(\text{gas})}$  (D6, page 2, lines 31 to 36). The detailed statement given on page 2, lines 37 to 45 of D6, which complies with the explanations reflected in paragraph [0019] of the patent at issue, describes that  $\text{SiO}_{\text{gas}}$  dispersed from the quartz crucible and the Si-melt reacts with carbon, e.g. the graphite of the heat-insulation material, to form carbon monoxide (CO) and silicon carbide (SiC) which deposits on the upper region of the furnace or the crucible protection parts and results in cracks. If cracks are created in these parts, pieces of SiC and of the carbonaceous material are peeled off to contaminate the inside of the apparatus. These scales could contaminate the molten silicon and result in crystal defects of the silicon single crystal so that the formation of SiC should be controlled safely (D6, page 2, lines 48 to 51). Contrary to the respondents position, the process of D6 therefore aims at avoiding the formation of silicon carbide inside the apparatus and the contamination of the molten silicon through exfoliations of SiC scaled

off from the furnace walls by floating an inert gas through an inlet port into the crucible towards the surface of the molten silicon, as depicted in Figure 2 of D6 (see also D6, page 2, line 57 to page 3, first line; page 3, line 57 to page 4, line 2; page 4, lines 51 to 54).

The general technique of protecting the surface of a molten material from contamination by feeding inert gas onto its surface is, therefore, realized also by the process disclosed in document D6.

4.3 Likewise, document D2 discloses that the casting station (9), where molten silicon is maintained in a quartz melting vessel (5), is blanketed continuously with an inert gas, for example argon, through a gas feed (16), as shown in the drawing. The preferential feeding of inert gas at the upper rim of the casting station (9) prevents in particular the deposition of SiO (D2, column 2, lines 45 to 48 and 54 to 57). Also this document confirms the general practice referred to above that undesired chemical reactions between the molten silicon and the surrounding atmosphere, in particular with carbon monoxide, can be avoided by feeding an inert gas into the crucible and directing it towards the surface of the molten silicon.

4.4 In conclusion, the provision of the step of feeding inert gas into the crucible towards the surface of the molten silicon during melting in the process according to D1 amounts to nothing more than what is conventionally carried out in the art by a skilled person who is confronted with the problem cited above and tries to solve it.

The subject matter of claim 1 therefore does not involve an inventive step.

**Order**

**For these reasons it is decided that:**

1. The decision under appeal is set aside.
2. The patent is revoked.

The Registrar:

The Chairman:

V. Commare

T. Kriner