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DECISION
of 6 November 2002

Case Number: T 0067/00 - 3.4.3

Application Number: 88910210.9

Publication Number: 0389533

IPC: H01L 21/20

Language of the proceedings: EN

Title of invention:
Sublimation growth of silicon carbide single crystals

Patentee:
NORTH CAROLINA STATE UNIVERSITY

Opponent:
Siemens AG

Headword:
-

Relevant legal provisions:
EPC Art. 100(a), 100(b), 56, 83

Keyword:
"Main request: inventive step (no)"
"First auxiliary request: inventive step (no)"
"Second auxiliary request: sufficiency (yes); inventive step (yes)"

Decisions cited:
-

Catchword:
-

Case Number: T 0067/00 - 3.4.3

DECISION
of the Technical Board of Appeal 3.4.3
of 6 November 2002

Appellant: (Opponent)	Siemens AG Postfach 22 16 34 D-80506 München (DE)
Representative:	-
Respondent: (Proprietor of the patent)	NORTH CAROLINA STATE UNIVERSITY 103 Holladay Hall Campus Box 7003 Raleigh North Carolina 27695-7003 (US)
Representative:	Warren, Keith Stanley BARON & WARREN 18 South End Kensington London W8 5BU (GB)
Decision under appeal:	**Interlocutory decision of the Opposition Division of the European Patent Office posted 30 November 1999 concerning maintenance of European patent No. 0 389 533 in amended form.**

Composition of the Board:

Chairman: R. K. Shukla
Members: M. Chomentowski
M. B. Günzel

Summary of Facts of Submissions

I. An opposition was filed by Appellant 1 against the European patent No. 0 389 533, which had been granted with 21 claims on the basis of the European patent application No. 88 91 02 10.9.

Claim 1 **as granted** had the following text:

"1. A method of reproducibly controlling the growth of large single crystals of a single polytype of silicon carbide independent of the use of impurities as a primary mechanism for controlling polytype growth, and which crystals are suitable for use in producing electrical devices, the method comprising:

introducing a monocrystalline seed crystal (17, 32) of silicon carbide of desired polytype and a silicon carbide source powder (40) into a sublimation system ;

raising the temperature of the silicon carbide source powder to a temperature sufficient for the source powder to sublime; while

elevating the temperature of the growth surface of the seed crystal to a temperature approaching the temperature of the source powder but lower than the temperature of the source powder and lower than that at which silicon carbide will sublime under the gas pressure conditions of the sublimation system;

characterized by:

generating and maintaining a constant flow of vaporized Si, Si_2C, and SiC_2, per unit area per unit time from the source powder (40) to the growth surface of the seed crystal (17, 32) for a time sufficient to produce a desired amount of macroscopic growth of monocrystalline silicon carbide of the desired polytype upon the seed crystal."

The further claims were dependent claims.

The grounds of opposition were that the patent did not disclose the invention in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art (Article 100(b) EPC) and that its subject-matter did not involve an inventive step and thus was not patentable (Articles 100(a) and 56 EPC) having regard *inter alia* to the following prior art documents:

D1: JP-A-62-066000 with the corresponding Patent Abstracts of Japan, vol. 11, No. 267 (C-443), 28 August 1987 and the translations provided by the appellants;

D2: "Progress in Controlling the growth of polytypic crystals", Y.M. Tairov et al., Electrical Engineering Institute, Leningrad, P-22, 197022 USSR, pages 111-152 (submitted 24 August 1982);

D4: DE-C2-3 230 727;

D5: "Ullmanns Encyclopädie der technischen Chemie", 3rd edition, ed. W. Foerst, Urban & Schwarzenberg, München-Berlin (DE), 1958, page 828; and

D6: Chemie - Ingenieur - Technik, 28th year, 1956, Nos. 3 and 5, pages 155 and 361 - 365.

II. With the interlocutory decision dated 30 November 1999 the Opposition Division decided to maintain the patent in amended form in accordance with the patentee's first auxiliary request.

In claim 1 as maintained, in the characterizing portion, it was specified that

the steps of generating and maintaining a constant flow of vaporized Si, Si_2C, and SiC_2, per unit area per unit time from the source powder (40) to the growth surface of the seed crystal (17, 32) were **such that the relative proportions of Si, Si_2C and SiC_2 in the vapour remain constant** for a time sufficient to produce a desired amount of macroscopic growth of monocrystalline silicon carbide of the desired polytype upon the seed crystal, **said steps including increasing the thermal gradient between the seed crystal and the source powder as the crystal grows and the source powder is used up, thereby to maintain an absolute temperature difference between the source powder and the seed crystal which continues to be most favourable for crystal growth and continuously encourage further crystal growth beyond that which would be obtained by maintaining a constant thermal gradient.**"

(Amendments with respect to claim 1 as granted have been emphasised by the Board).

III. The reasoning of the Opposition Division was in substance as follows:

Main request (the patent as granted)

A process according to the preamble of claim 1 in dispute is known from document D1.

The document does not specify the features of the characterizing portion of the claim. However, it is generally known that maintaining a constant and homogeneous flow of the constituents in the vapour phase, i.e., a constant and homogeneous transport of the species, is necessary to reliably grow crystals from the vapour phase, as can be seen from documents D5 and D6. Moreover, document D2 teaches that the growth of specific polytypes of SiC in the sublimation method depends on the Si/C ratio in the gaseous phase. Therefore, for this purpose, since the proportions of Si, Si_2C, and SiC_2 are established by the thermodynamic parameters e.g. temperature and pressure of the process and parameters of the source powder such as powder grain size and polytype of the grains, it is obvious that by maintaining constant and homogeneous transport conditions in the gaseous phase during the sublimation growth process the intended purpose can be achieved. The subject-matter of claim 1 therefore does not involve an inventive step.

First auxiliary request

The request was found to satisfy the condition of sufficiency of disclosure (Article 100(b) EPC).

Inventive step

The state of the art is either silent about the thermal gradient to be maintained during the method, see e.g. document D2, or teaches that the thermal gradient is not to be modified during the sublimation method, see e.g. documents D1 and D4. Taking into account documents D5 and D6, which stress the importance of maintaining the transport parameters, the skilled person would not be incited to modify the thermal gradient, as in claim 1. Therefore, the subject-matter of this claim involves an inventive step.

IV. Appeals were lodged by the opponent and the patentee on 19 January 2000 and 4 February 2000 respectively, paying the appeal fees on the respective same day. The statements of grounds of appeal of the opponent and of the patentee were filed on 30 March and 7 April 2000 respectively.

V. During the Oral proceedings of 6 November 2002, Appellant 2 (the patentee) requested that the decision under appeal be set aside and a patent be granted on the basis of a main request which had been filed with the statement of the grounds of appeal on 7 April 2000 or of one of three auxiliary requests which had been filed during the oral proceedings.

Claim 1 has the same preamble in all the requests and is that of claim 1 as granted.

Main request

The characterizing portion of claim 1 of the **main request** reads as follows:

"generating and maintaining a constant flow of vaporized Si, Si_2C, and SiC_2, per unit area per unit time from the source powder (40) to the growth surface of the seed crystal (17, 32), **such that the relative proportions of Si, Si_2C and SiC_2 in the vapour flow remain constant,** for a time sufficient to produce a desired amount of macroscopic growth of monocrystalline silicon carbide of the desired polytype upon the seed crystal."

First auxiliary request

Claim 1 of the **first auxiliary request** differs from claim 1 maintained by the Opposition Division by substituting in the characterizing portion the words "and by the step of increasing the thermal gradient" in place of "said steps including increasing the thermal gradient", and its characterizing portion reads as follows:

"the steps of generating and maintaining a constant flow of vaporized Si, Si_2C, and SiC_2, per unit area per unit time from the source powder (40) to the growth surface of the seed crystal (17, 32) **such that the relative proportions of Si, Si_2C and SiC_2 in the vapour flow remain constant** for a time sufficient to produce a desired amount of macroscopic growth of monocrystalline silicon carbide of the desired polytype upon the seed crystal, **and by the step of increasing the thermal gradient between the seed crystal and the source powder as the crystal grows and the source powder is used up, thereby to maintain an absolute temperature difference between the source powder and the seed crystal which continues to be most favourable for crystal growth and continuously encourage further crystal growth beyond that which would be obtained by maintaining a constant thermal gradient.**"

Second auxiliary request

The characterizing portion of claim 1 of the **second auxiliary request** reads as follows:

"the steps of generating and maintaining a constant flow of vaporized Si, Si_2C, and SiC_2, per unit area per unit time from the source powder (40) to the growth surface of the seed crystal (17, 32) **such that the relative proportions of Si, Si_2C and SiC_2 in the vapour flow remain constant** for a time sufficient to produce a desired amount of macroscopic growth of monocrystalline silicon carbide of the desired polytype upon the seed crystal, **said steps including introducing a source powder having a selected polytype composition, a**

selected predetermined distribution of surface areas and a selected predetermined distribution of particle sizes, and maintaining said polytype composition, surface area and particle size distribution constant throughout the growth process".

The further claims of the second auxiliary request, i.e. the dependent claims 2 to 17, were also filed by Appellant 2 during the oral proceedings.

VI. Appellant 2 (Patentee) submitted essentially the following arguments in support of his requests:

The general principle underlying the present invention, i.e., the generation and maintenance of a constant flow of vaporized Si, Si_2C, and SiC_2 and different ways of achieving such a constant flow are sufficiently described in the patent specification in suit (see e.g. Figures 1 to 3 and page 6). In particular, it is disclosed that by increasing the thermal gradient during the sublimation, a constant flow profile of the components is obtained.

Main request

Although the process parameters are not specified in claim 1 of the request, the method as claimed is not rendered obvious by document D1, wherein there is no suggestion to increase the thermal gradient as the sublimation proceeds nor by document D2, wherein it is the ratio Si/C which is to be controlled. Documents D5 and D6 are very general and not specific as to the parameters of the method.

First auxiliary request

Modifying the temperature gradient during the sublimation process is suggested in document

D18: "Single Crystal Growth of SiC Substrate Material for Blue Light Emitting Diodes", G. Ziegler et al., IEEE Transactions on Electron Devices, vol. ED-30, No. 4, April 1983, pages 277 - 281,

only for avoiding the formation of bubbles during crystal growth, and this is a different problem to the one addressed in the patent in suit. Moreover, from Figure 3 of this document showing the temperature profile in the crucible assembly, it is not possible to determine whether, during the growth of the crystal when the source powder is being used up, the temperature gradient increases or not.

Second auxiliary request

The set of parameters and conditions i.e. a predetermined distribution of surface areas, particle sizes, and a selected polytype composition of claim 1 enable to obtain a constant flow ratio of Si, Si_2C and SiC2 in the vapour phase and thus to grow large SiC crystals of constant polytypes in a reproducible manner. This was not directly derivable from the known techniques.

VII. Appellant 1 has requested that the decision under appeal be set aside and the patent be revoked, and he provided the following arguments in particular with respect to the second auxiliary request:

It was already known, e.g. from document D4, to grow single SiC crystals of a desired polytype from the vapour phase by sublimation, and it was also known, e.g. from documents D2, D5 and D6, in different techniques of growth from the vapour phase, e.g. by sublimation, that a constant and homogeneous flux of the constituent species was necessary. It was also known from document

D17: Colloques Internationaux CNRS, No. 205 - Etudes des Transformations Cristallines à Haute Température", 1972, pages 163 to 170

that 6H SiC, a very important polytype, is the stable phase of silicon carbide. Therefore, the set of conditions and parameters in claim 1 would be obvious to any person skilled in the art of SiC crystal growth by sublimation.

Reasons for the Decision

1. The appeals are admissible.

2. *Main request*

A process according to the preamble of claim 1 in dispute is known from document **D1**. In the embodiment described in the part bridging pages 6 and 7 of the English translation, a 6H type SiC single crystal is obtained.

However, the document does not indicate that the known method for generating and maintaining a constant flow of vaporized Si, Si_2C, and SiC_2, per unit area per unit time from the source powder (40) to the growth surface of the seed crystal (17, 32) is **such that the relative proportions of Si, Si_2C and SiC_2 in the vapour remain constant** for a time sufficient to produce a desired amount of macroscopic growth of monocrystalline silicon carbide of the desired polytype upon the seed crystal. In particular, document D1 does not give any specific information about the SiC material of the source powder.

Yet, the following is to be noted in this respect:

According to the patent in suit (see page 4, lines 14 to 15 and page 5, lines 5 to 10), in one embodiment of the invention, large single crystals of silicon carbide may be grown by **controlling the source powder, e.g., such that substantially all of it has a constant polytype composition**. Other measures are mentioned in the description (see in particular page 4, lines 15 to 17), but for being used in addition or as an alternative to the substantially constant polytype composition of the source. Thus, since in claim 1 in dispute no particular process feature is indicated for arriving at the purpose mentioned in the second part of the claim, it is to be derived that controlling the source powder, for instance the polytype of said source, will already result in this purpose being achieved, i.e., that in particular the relative proportions of Si, Si_2C and SiC_2 in the vapour flow will remain constant.

However, as acknowledged in the patent in suit (cf. page 3, lines 53 to 57) with reference to document D2, the use of single crystals of particular polytypes as vapour source was already known in the art, so that such a use in the process of document D1 would be obvious to the skilled person.

The argument of Appellant 2 (Patentee) that in document D2 (see e.g. page 120, the paragraph following Figure 4) it is the ratio Si/C in the gas which is to be maintained constant, and not the ratios of Si, Si_2C and SiC_2, is not relevant since in the document (see e.g. page 120, the paragraph preceding Figure 4; Table 1, fourth line on page 117 and page 125, second paragraph) powder of a single 6H-polytype is evaporated. Incidentally, it is to be noted that a similar SiC sublimation method using mainly 6H polytype source powder is also disclosed in document D4 (see column 3, lines 27 to 35).

Therefore, the skilled person using in the sublimation crystal growth method of document D1 a SiC source powder such that substantially all of it has a constant polytype composition, made of e.g. 6H SiC, in order to obtain a satisfactory Si/C ratio in the vapour phase, will also automatically obtain constant ratios of Si, Si_2C and SiC2 as recited in claim 1. In other words, this straightforward measure leads directly to the subject-matter of claim 1.

Consequently, the subject-matter of claim 1 of the main request does not involve an inventive step in the sense of Article 56 EPC.

3. *First auxiliary request*

Contrary to the method of the first auxiliary request, in the method known from document D1, **the steps of** generating and maintaining a constant flow of vaporized Si, Si_2C, and SiC_2, per unit area per unit time from the source powder (40) to the growth surface of the seed crystal (17, 32) are not **such that the relative proportions of Si, Si_2C and SiC_2 in the vapour remain constant** for a time sufficient to produce a desired amount of macroscopic growth of monocrystalline silicon carbide of the desired polytype upon the seed crystal. Also, **the thermal gradient between the seed crystal and the source powder as the crystal grows and the source powder is used up is not increased so as to maintain an absolute temperature difference between the source powder and the seed crystal which continues to be most favourable for crystal growth and continuously encourage further crystal growth beyond that which would be obtained by maintaining a constant thermal gradient**.

The following is to be noted with respect to the thermal gradient during the growth:

Figure 3 of document D18 (see also part III. Modified Lely Process Using Seed Crystals, on pages 278 and 279) shows the crucible assembly for the growth of SiC crystals for a process of the same type as that of document D1 and the temperature profile inside the chamber. This temperature profile is not shown as a straight line and thus does not correspond to a linear relation between the temperature and the distance between the crystal seed and the surface of the powder. Indeed, since for the skilled person it is derivable that during crystal growth, in the apparatus as shown, a variation of the above-mentioned distance will occur as a result of crystal being deposited on the seed crystal on the one hand and of the source powder being used up on the other hand, it is not directly and unambiguously derivable whether the resulting change in the temperature gradient will be an increase, as in claim 1 in dispute.

However, in document D18 (see page 278, right-hand column, second paragraph, lines 6 to 12), it is stated that the temperature gradient is one of the three essential parameters for growing 6H SiC crystals on a 6H seed crystal, whereas temperature gradient and gas pressure control the transport and hence the crystal growth velocity; in particular (see page 279, right-hand column, second paragraph, lines 3 to 7), to avoid bubble formation during crystal growth, it is proposed to start crystal growth with a very small temperature gradient and subsequently increase the crystal growth velocity by moving the crystal into a region of larger temperature gradient.

Therefore, increasing the thermal gradient during crystal growth was known as helping to obtain 6H SiC crystals of good quality, so that, since in the apparatus of document D1 (see Figure 1 and the third paragraph of the English translation) the distance between the crystal seed and the powder can be adjusted, it would be obvious to the skilled person to adapt the system of document D1 so that the thermal gradient can be increased during the crystal growth.

Concerning the other features of the characterizing portion, it is to be noted that they do not differ from those in the characterizing portion of the main request, i.e., that they can be obtained in a straightforward way by selecting an SiC source powder such that substantially all of it has a constant polytype composition.

Appellant 1 (Opponent) has submitted that increasing the thermal gradient is part of the claimed method, but that there is no indication in the patent in suit that such an increase plays a role in maintaining the relative proportions of Si, Si_2C and SiC_2 in the vapour constant. The method of claim 1 of the first auxiliary request thus results from an aggregation of different features including the increase in thermal gradient, but without any synergetic effect.

Appellant 2 has argued that, starting from the method known from document D1 wherein the thermal gradient is not increased, it would not be obvious to the skilled person to adapt the system by moving the crystal into a region of larger temperature gradient, as in document D18.

However, in the Board's view, increasing the thermal gradient and selecting a source powder of a constant polytype composition are both straightforward measures known to be advantageous for crystal growth. Moreover, there is no information in the patent in suit that these measures cooperate to produce a synergetic effect, so that the argument of Appellant 2 concerning the aggregation of the features is considered as convincing.

Therefore, in the Board's judgment, the subject-matter of claim 1 of the first auxiliary request does not involve an inventive step in the sense of Article 56 EPC.

4. *Second auxiliary request*

4.1 Admissibility of the amendments and clarity

Claim 1 of the second auxiliary request results from the patent as granted, i.e., the combination of claim 1 and the dependent claims 6, 8 and 10 reciting the specific measures, e.g. the selected polytype composition and maintenance thereof, with additionally the statements in the description (see page 5, lines 29 to 32 and 49 to 54, and page 6, lines 9 to 11) stressing that during the process a consistent flux profile of the species, i.e., the relative amounts or ratios of Si, Si_2C, and SiC_2, is to be obtained. The dependent claims result from the renumbering and adapting of the dependent claims as granted.

The Board is satisfied that the amendments in the claims are admissible and that the claims are clear, with the description and drawings to be adapted for consistency. There were no objections from Appellant 1 in this respect either.

4.2 Sufficiency

In the method of claim 1 of the second auxiliary request, the steps of generating and maintaining a constant flow of vaporized Si, Si_2C, and SiC_2, per unit area per unit time from the source powder (40) to the growth surface of the seed crystal (17, 32) are such that the relative proportions of Si, Si_2C and SiC_2 in the vapour remain constant for a time sufficient to produce a desired amount of macroscopic growth of monocrystalline silicon carbide of the desired polytype upon the seed crystal; these steps include introducing a source powder having a selected polytype composition, a selected predetermined distribution of surface areas and a selected predetermined distribution of particle sizes, and maintaining said polytype composition, surface area and particle size distribution constant throughout the growth process.

Selecting a particular polytype composition for the source powder, for instance consisting of 6H silicon carbide, is disclosed in the patent in suit (see e.g. page 8, lines 25 to 38) and such selections are also known from the prior art, as see e.g. document D4 (see column 3, lines 27 to 35). Selecting a predetermined distribution of particle sizes, is also disclosed in the patent in suit (see the same text location and, additionally, page 5, line 49 to page 6, line 8), and the prior art also discloses such methods, as see e.g. document D4 (see the same text location).

Concerning the selection of a predetermined distribution of surface areas of the source powder, it is to be noted that, according to the patent in suit (see page 6, lines 9 to 11), for a given powder morphology, the exposed surface area of the source powder is proportional to the particle size, and that a consistency in exposed surface area enhances the consistency of the flux profile. Therefore, it is credible that, by adequately selecting a particular polytype composition for the source powder and a predetermined distribution of particle sizes thereof, this result can also be obtained.

The last condition in the claim, i.e., maintaining said polytype composition, surface area and particle size distribution constant throughout the growth process, can be obtained by replenishing the source powder during the sublimation process (see e.g. page 7, lines 47 to 57; claims 7, 9 and 11 of the patent as granted).

It is credible that by selecting and maintaining these conditions the skilled person will be able without undue burden to regulate the other features of the process such as the temperature so that the relative proportions of Si, Si_2C and SiC_2 in the vapour remain constant for a time sufficient to produce a desired amount of macroscopic growth of monocrystalline silicon carbide of the desired polytype upon the seed crystal.

The arguments of Appellant 1 against sufficiency of disclosure were in respect of main and auxiliary request 1.

Therefore, the Board is satisfied that the patent in suit discloses the invention in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art (Articles 83 and 100(b) EPC).

4.2 Inventive step

It is to be noted that, in the second auxiliary request, a set of three conditions and the maintenance of said set of conditions during the growth are necessary for obtaining the required result. In the main request, on the contrary, according to the description, only one of the conditions such as the selection of a particular polytype for the source powder was sufficient for obtaining the required result.

Starting from the method known from document D1, wherein in particular no indication is given as to the composition of the source powder, the skilled person intending to grow large single crystals of silicon carbide of desired polytypes in a controlled and repeatable manner can refer to the prior art for adapting its method in order to obtain large SiC crystals of a desired polytype.

However, none of the prior art documents teaches the steps of generating and maintaining a constant flow of vaporized Si, Si_2C, and SiC_2, per unit area per unit time from the source powder (40) to the growth surface of the seed crystal in a process wherein the three further conditions and their maintenance are satisfied.

Although as argued by Appellant 1 selecting a particular polytype composition for the source powder, for instance consisting of 6H silicon carbide, and a predetermined distribution of particle sizes, was known in the art, see e.g. document D4 (see column 3, lines 27 to 35), there was no indication therein of an additional control of the distribution of the surface areas and of the further method features.

Although as also argued by Appellant 1 controlling the method in such a way that the flow of species is homogenous and remains constant during the growth of single crystals from a gas phase is generally known in the art, see e.g. documents D6 (see page 362, right-hand column, antepenultimate paragraph and page 363, left-hand column, penultimate paragraph), this is however not derivable as being valid for each isolated specie in the vapour phase per unit area per unit time from the source powder to the growth surface of the seed crystal.

Document D2 (see in particular page 120, penultimate paragraph and page 141, first paragraph, 10 last lines) indeed teaches that the effect of silicon carbide powder grain size on the values of reaction rates of the equations relating the relative proportions of *inter alia* Si, Si_2C and SiC_2 in the vapour phase will in turn result in the change of vapour phase composition, Si/C ratio, in particular. The document teaches also the influence of other features on the Si/C ratio in the vapour phase. However, the arguments of Appellant 1 on this basis also cannot convince because there again there is no direct and unambiguous teaching about a necessity to control other ratios such as the relative proportions of Si, Si_2C and SiC_2 in the vapour phase, their consistency and maintenance per unit area per unit time from the source powder to the growth surface of the seed crystal.

Indeed, as argued by Appellant 1, above a temperature which is lower than the sublimation temperature, 6H SiC is the stable phase of silicon carbide, as see document D17 (see page 169, the paragraph bridging both columns). However, this is not considered as an indication toward any specific technical method measure and in particular to those of claim 1 of the second auxiliary request. It is not an indication that any measure can result in the subject-matter of claim 1 either.

The further prior art documents do not provide information more relevant than that in the documents cited here above.

As convincingly argued by Appellant 2, it was the merit of the inventors to determine that it was important to adapt the known method for obtaining large single crystals of a desired polytype in a controlled and reproducible manner and to determine the set of features necessary for this purpose. In this respect, the set of conditions and the maintenance of these conditions cannot be considered as being obvious to the skilled person starting from document D1.

Therefore, in the Board's judgment, the subject-matter of claim 1 involves an inventive step in the sense of Article 56 EPC.

5. Consequently, the patent can be maintained on the basis of claim 1 of the second auxiliary request, with the description and drawings to be adapted (Article 102(3) EPC).

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the opposition division with the order to maintain the patent with the claims 1 to 17 of the second auxiliary request filed at the oral proceedings and a description and drawings to be adapted.

The Registrar:	The Chairman:
P. Martorana	R. K. Shukla