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Datasheet for the decision of 4 September 2013

Case Number: T 0500/11 - 3.3.05

Application Number: 04748762.4

Publication Number: 1680357

IPC: C01B 33/107

Language of the proceedings: EN

Title of invention:
Method for production of trichlorosilane

Patent Proprietor:
Elkem ASA

Opponent:
Evonik Degussa GmbH

Headword:
Trichlorosilane/ELKEM

Relevant legal provisions:
EPC Art. 54(1)(2), 56, 83, 123(2)

Keyword:
"Disclosure of the invention: sufficient; no gaps in information"
"Main request: Novelty (no) - end value of a prior art range takes away the novelty of the claimed range"
"Auxiliary request: amendment - admissible intermediate generalisation - other features not inextricably linked to the feature picked out from a working example; novelty (yes); inventive step (yes) - improvement (yes); obviousness (no)"

Decisions cited:
T 0240/95, T 0962/98, T 0520/06, T 0273/10

Catchword:
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Case Number: T 0500/11 - 3.3.05

D E C I S I O N
of the Technical Board of Appeal 3.3.05
of 4 September 2013

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Decision under appeal: **Interlocutory decision of the Opposition Division of the European Patent Office posted 12 January 2011 concerning maintenance of European patent No. 1680357 in amended form.**

Composition of the Board:

Chairman: G. Raths
Members: J.-M. Schwaller
C. Vallet

Summary of Facts and Submissions

I. The present appeals lie from the decision of the opposition division concerning maintenance of European patent No. 1 680 357 on the basis of the claims of the auxiliary request filed with letter dated 2 March 2010, claim 1 thereof reading:

"*1. Method for the production of trichlorosiliane by reaction of silicon with HCl gas at a temperature between 250° and 1100°C, and an absolute pressure of 0.5 — 30 atm in a fluidized bed reactor, in a stirred bed reactor or in a solid bed reactor, characterised in that the silicon supplied to the reactor contains between 550 and 10 000 ppm chromium.*"

Claim 1 of the main request (also claim 1 as granted) reads as follows:

"*1. Method for the production of trichlorosiliane by reaction of silicon with HCl gas at a temperature between 250° and 1100°C, and an absolute pressure of 0.5 — 30 atm in a fluidized bed reactor, in a stirred bed reactor or in a solid bed reactor, characterised in that the silicon supplied to the reactor contains between* ***50*** *and 10 000 ppm chromium.*"

II. Among the documents cited in the first-instance proceedings, the following are of relevance for the present decision:

D1: B. Kanner et al., "*Commercial Production of Silanes by the direct Synthesis*", pages 1 to 48,

Catalyzed Direct Reactions of Silicon, K.M. Lewis and D.G. Rethwisch (Editors), 1993.

D2: DE 3 640 172 C1

D4: J. Acker et al. "*Formation of Silicides in the System Metal-Silicon-Chlorine-Hydrogen: Consequences for the Synthesis of Trichlorosilane from Silicon and Hydrogen chloride*", Silicon for the Chemical Industry, Tromsø, Norway, May 29 to June 2, 2000, pages 121 to 133.

D5: WO 03/018207

D8: DE 3 230 590

D9: Ullmanns Encyclopedia of Industrial Chemistry, 6th Edition, 1998, Electronic Release: part 4.1 "*Metallurgical Silicon*", 2 pages.

III. According to the contested decision, the patent disclosed sufficient details as regards the way of adding chromium and silicon to the reactor, and so - in accordance with the requirements of Article 83 EPC – it disclosed sufficient information for the skilled person to carry out the claimed method for producing trichlorosilane (hereinafter "TCS").

Claim 1 of the main request was novel in the light of document D1, as there was no "unmistakable evidence" that a silicon exhibiting an amount of Cr in the range claimed was ever used for the preparation of TCS.

Claim 1 lacked inventive step when starting from document D1, because in Table, the skilled person found the teaching that the silicon used for the preparation of chlorosilanes (TCS in the technical context of D1) was of metallurgical grade and contained between 5 and 200 ppm Cr.

Claim 1 of the auxiliary request fulfilled the requirements of Article 123(2) EPC, because the amount of 550 ppm Cr disclosed in example 3 of the patent in suit was neither linked to the conditions of temperature nor to the type of reactor used in this example. This claim also fulfilled the requirements of Article 56 EPC as none of the documents suggested that the selectivity of the reaction at issue was linked to the amount of Cr contained in the silicon.

IV. With its statement of grounds of appeal, the patentee (hereinafter "appellant I") submitted that the subject-matter of claim 1 of the main request was inventive in the light of D1.

V. With its statement of grounds of appeal, the opponent (hereinafter "appellant II") submitted two further documents

D10: Ullmanns Enzyklopädie der technischen Chemie, 4., neubearbeitete und erweiterte Auflage, volume 21, pages 417 to 419, 478, 479 and 483 (1982), and

D11: EP 0 658 359 A2.

Further, it contested the allowability under Article 123(2) EPC of amended claim 1 of the auxiliary request

and held that its subject-matter lacked an inventive step in the light of the teachings of documents D1 and D10.

VI. With a letter dated 27 July 2011, appellant II further challenged the sufficiency of disclosure of the invention and the novelty of claim 1 of the main request in the light of the disclosure of document D1.

VII. With a letter dated 3 October 2011, appellant I submitted a declaration of Dr Freissmuth stating that document D10 contained a typographical error and that "Cr" should read "Ca".

Appellant I further contested the Article 123(2) EPC objection. It argued that the general teaching of the contested patent was that an elevated Cr content in the silicon feedstock had a beneficial effect on the selectivity of TCS, independently of other factors. Further, Example 3 taught that the selectivity to TCS was higher when using pure Si alloyed with 550 ppm of Cr than when using pure Si alone, and this teaching had nothing to do with the effect of temperature on this reaction.

VIII. At the oral proceedings, which took place on 4 September 2013, the discussion concerned in essence the novelty of the main request and Article 123(2) and 56 EPC issues regarding the auxiliary request.

IX. After closing the debate, the board established the parties' requests to be as follows:

Appellant I requested that the decision under appeal be set aside and the patent be maintained as granted.

Appellant II requested that the decision be set aside and the patent be revoked.

Reasons for the Decision

1. *Disclosure of the invention*

1.1 According to Article 83 EPC and its counterpart in Article 100(b) EPC, an invention must be disclosed in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art. According to the case law, in order to prove insufficiency of disclosure, gaps in information have to be ascertained.

1.2 In the case at issue, a detailed description of the invention - which concerns the production of TCS by reaction of silicon and HCl gas - is given in Examples 1 to 4 of the patent in suit. Further, as regards the manner in which chromium is added to the silicon supplied to the reactor, the examples and the description, in particular paragraphs [0013] to [0019], disclose several ways of adding chromium to silicon and thus sufficient information is given to the skilled person for carrying out the claimed invention.

1.3 Appellant II - which bears the burden of proof – has furthermore not provided any evidence, for instance a reproduction of at least one of the examples, showing that the preparation details disclosed in the patent in

suit were insufficient to arrive at the claimed subject-matter.

1.4 Under these circumstances, with appellant II unable to identify any gap in information in the performance of the claimed invention, the board concludes that the requirements of Article 83 EPC and its counterpart in Article 100(b) EPC are satisfied.

2. *Main request – Novelty*

2.1 D1 (Chapter 3. "*Manufacture of chlorosilanes by the direct synthesis*", pages 5 and 6) discloses the reaction of HCl with silicon at temperatures greater than 180°C to produce a mixture of silanes, with TCS being the main reaction product. The composition of the reaction mixture depends e.g. on the reaction temperature and pressure, the presence of metallic impurities in the silicon and the type of reactor used. In laboratory fluidised beds, optimum TCS yields span 90-95 wt.% in the temperature range of from 300 to 330°C at atmospheric pressure. This yield decreases with increasing temperature and pressure and silicon tetrachloride yield increases beyond 2-5 wt%. at temperatures greater than 350°C and pressures greater than 4 atm. Commercial synthesis is customarily performed at 320 to 370°C and 1 to 4 atm, with the reaction mixture having the approximate composition: 75 to 85 wt.% $HSiCl_3$ (TCS), 8 to 10 wt.% $SiCl_4$ and 7 to 15 wt.% others.

According to paragraph 4.1.1 at page 8 of D1, the silicon employed in the direct synthesis of chlorosilanes and methylchlorosilanes (hereinafter MCS)

is commonly a technical grade material containing about 98 to 99 wt% Si. Further, according to Table 3 (D1, page 10), the silicon is of metallurgical grade which comprises 98 to 99 wt.% silicon and, among other metallic impurities, 5 to 200 ppm of chromium.

2.2 Appellant I argued that even though Table 3 reported that metallurgical silicon used for the preparation of TCS or MCS might contain chromium in an amount up to 200 ppm, the range disclosed in Table 3 was the result of a compilation of more than 450 individual technical scientific publications concerned with the synthesis of silanes. It was thus not necessarily true that the silicon used for the preparation of TCS was the same as the one for the preparation of MCS, let alone that the silicon used for the preparation of TCS exhibited such a high amount of Cr. Therefore, the assertion that Dl disclosed the use of a silicon exhibiting a high amount of Cr to produce TCS was to be regarded as a selection from two lists of possible features.

2.3 The board cannot accept these arguments. It is true that Table 3 of D1 concerns the preparation of two different types of silanes, however, there is no indication in D1 that the metallurgical silicon used for preparing chlorosilanes is different from the one used in the preparation of methylchlorosilanes. As to the presence of impurities in the metallurgical silicon, D1 does not disclose that chromium - in contrast to iron, lead or aluminium - has a particular effect on the synthesis of the above silanes, let alone on the specific preparation of the one or the other of these two types of silanes.

It follows that the skilled reader of the chapters concerned with the preparation of chlorosilanes and methylchlorosilanes, and in particular of Table 3, directly and unambiguously understands that the metallurgical silicon used for the synthesis of chlorosilanes can contain the same amount of chromium as the one used for the synthesis of methylchlorosilanes and so, the range of from 5 to 200 ppm Cr disclosed in D1 applies to the synthesis of both types of silanes. It follows, contrary to appellant I's argument, that the skilled person concerned with the direct synthesis of a particular type of chlorosilane does not have to make any choice as regards the chromium content of the metallurgical silicon to be used.

In the board's view, the skilled reader of D1 directly and unambiguously understands from chapter 4, in particular from the paragraph bridging pages 5 and 6 and from the first and third full paragraphs of page 6, that the preparation of chlorosilanes inevitably includes the preparation of TCS, in particular in the preferred ranges of temperatures and pressure identified in point 1.4 above, which ranges entirely fall in the corresponding ranges defined in claim 1 at issue, and in which ranges TCS is the main reaction product.

It follows from the above considerations that all the features of present claim 1 are disclosed in combination in D1, with the exception of the chromium range of 50 to 10000 ppm, which overlaps with the range of 5 to 200 ppm disclosed in D1, table 3.

2.4 In accordance with the case law of the boards of appeal (see in particular T 0240/95, point 4.2 of the reasons or T 0520/06, point 15.1.1 of the reasons), the disclosure of a range is an explicit disclosure of the end values. The disclosure of a specially disclosed value in the prior art taking away the novelty of a claimed range, it follows in the present case that the upper end value of the range disclosed in table 3 of D1, namely 200 ppm, anticipates the claimed range of 50 to 10000 ppm.

2.5 For the above reasons, the subject-matter of claim 1 as granted is no longer novel in the light of the disclosure of document D1. Claim 1 therefore does not meet the requirements of Article 54(1) and (2) EPC.

3. *Auxiliary request – Allowability of the amendments*

3.1 The amendment in claim 1 at issue corresponds to the restriction of the range "*between 50 to 10000 ppm of chromium*" by the lower end value of **550** ppm. It is uncontested that there is a literal basis for this specific value in example 3 of the patent in suit, however not in combination with the other features of amended claim 1 under dispute.

3.2 Appellant II argued that the amendment consisted of an inadmissible intermediate generalisation in the sense of T 0962/98, because the feature 550 ppm Cr was inextricably linked to the other features of the process according to example 3 - namely the type of reactor, the pressure and temperature, the particle size and the contact time - and thus this specific value could not be arbitrarily extracted from its

context. Appellant II referred also in this respect to paragraph [0003] of the patent which disclosed that the "*selectivity and reactivity will depend strongly on the process temperature when silicon and HCl is reacted*". This would make clear that the temperature was a feature which was inextricably linked to the selectivity to TCS.

3.3 The question to be answered now is whether the feature "550 ppm Cr" is inextricably linked to the other features defined in Example 3 of the patent in suit.

The board observes in this respect that there is no doubt that not only the concentration of chromium has an influence on the selectivity of TCS but also other parameters too, such as e.g. the particle size, the pressure, the temperature, and the contact time in the reactor. All these features however are parameters which may be varied individually and each of these variations would necessarily lead to a different result on the selectivity to TCS.

If the feature "550 ppm chromium" was inextricably linked to specific other parameters of Example 3, this would mean that the selectivity to TCS would only be achieved in the definite combination of "550 ppm chromium" with the specific other parameters of example 3. This however is manifestly not the case here.

Moreover, according to paragraph [0020] of the patent in suit, the addition of chromium to silicon is described very generally as improving the selectivity to TCS in the reaction of silicon with gaseous HCl.

It follows from the above considerations that the different parameters, and in particular those in Example 4, can be varied independently one from another and that there is no reciprocal relationship, which would imply that the concentration of chromium is part of an entanglement in which a variation of the other parameters is not allowed.

3.4 The question at issue was already addressed in several decisions, in particular in T 0273/10 or T 0962/98, to which the board refers in the following paragraphs.

3.4.1 According to T 0962/98 (point 2.5 of the reasons), an intermediate generalisation would be admissible if the skilled person could recognise without any doubt from the application as filed that the characteristics taken from a working example were not closely related to the other characteristics of the working example and applied directly and unambiguously to the more general context.

3.4.2 In decision T 0273/10 (points 14.2 and 14.3), a claim amended by inclusion of a bundle of features extracted from a specific embodiment had been found allowable because the bundle of features proposed as an amendment comprised all the features essential for the performance of the invention. Further, according to T 0273/10, those features of the embodiment which did not contribute to solving the problem underlying the invention did not have to be part of the claimed subject-matter resulting from the amendment.

3.4.3 In the case at issue, as explained in point 3.3 above the aim of the invention – namely increasing the

selectivity to TCS – is merely achieved by addition of selected amounts of chromium to silicon and thus this feature is not "closely related to the other characteristics of the working example and applies directly and unambiguously to the more general context" as required in T 0962/98.

3.4.4 The present decision is also in line with decision T 0273/10 since the proposed amendment comprises the feature essential for the performance of the invention, namely a certain amount of chromium, and moreover those features of the embodiment which do not contribute to solve the problem underlying the invention are not part of the claimed subject-matter resulting from the amendment.

3.5 It follows from the above considerations that the gain of selectivity is solely due to the presence of chromium in the silicon feedstock, and that the picking out of the value 550 ppm from the working example 3 is therefore plainly acceptable in the case at issue, with the consequence that claim 1 of this request meets the requirements of Article 123(2) EPC.

Dependent claims 2 to 6, which have a basis in the corresponding dependent claims 2 to 6 of the application as filed, also meet the requirements of Article 123(2) EPC.

4. *Auxiliary request - Novelty*

The board is satisfied that the claimed subject-matter is novel, because none of the documents cited in the

present proceedings discloses an amount of chromium which falls within the terms of claim 1 at issue.

The board is in particular convinced that document D10 does not anticipate the subject-matter of claim 1 at issue, because the declaration of Mr Freissmuth - author of D10 - and the disclosure of much lower amounts of chromium in documents D1 and D9 (D1: 5 to 200 ppm; D9: 100 to 400 ppm) renders Mr Freissmuth's conclusion credible that the range "Cr 0,20 – 0,30%" at page 419 of D10 contains a typographical error. Said range should read "**Ca** 0,20 – 0,30%".

It follows from the above considerations that claim 1 of this request, and by the same token dependent claims 2 to 6, which include all the features of claim 1, meet the requirements of Article 54(1)(2) EPC.

5. *Auxiliary request - inventive step*

By applying the problem-solution approach, the board came to the following conclusions.

5.1 The invention concerns a method for the production of trichlorosilane by reaction of silicon with HCl gas (paragraph [0001] of the contested patent).

5.2 Such a process is already known from document D1 which according to the parties represented the closest state of the art to the claimed subject-matter, and which thus is to be taken as starting point for the assessment of the inventive step of claim 1 (see details of the disclosure of document D1 under point 2.1 above).

5.3 As to the technical problem underlying the contested patent, this is defined in paragraph [0010] of the patent in suit as consisting in the provision of a higher selectivity in the production of TCS by reaction of silicon with HCl.

5.4 As a solution to this problem, the invention proposes the method for the production of TCS according to claim 1 of the auxiliary request, characterised in particular in that the silicon supplied to the reactor contains between 550 and 10000 ppm chromium.

5.5 As to the question whether the problem underlying the patent in suit has been solved, appellant II stated that it was not solved, so that the technical problem boiled down to the provision of a mere alternative process to the one known in D1. It argued in this respect that a comparison between example 3 of the patent and the additional experiment filed on 3 December 2007 by the proprietor showed that experimental errors were so high in the measurement of the selectivity that it was doubtful whether a selectivity gain was actually achieved. And even if any improvement in terms of selectivity had been achieved at all, it was doubtful whether this effect covered the whole breadth of the claimed subject-matter. In particular, example 4 of the patent in suit showed that the selectivity (70 to 48% according to the amount of Si converted; T = 545°C) was lower than in a similar process in document D8 (page 3, lines 10 to 15) wherein the selectivity to TCS was 95% (T = 260°C). D8 (page 3, line 15) further described the selectivity to TCS to be very low (20%) at higher temperatures (800°C); thus it was not credible that an effect still existed at the

upper limit of the range claimed, i.e. at temperatures close to 1100°C.

The board cannot accept these arguments, because the examples in the patent clearly show that an increase of selectivity could be achieved as well at 365°C (examples 1 to 3) as at 545°C (example 4) and so it is credible that even at higher temperatures this effect still exists. Appellant II, on which the burden of proof rests, in any case did not provide any piece of evidence to the contrary.

Regarding the alleged experimental errors, appellant II did not reproduce one of the examples of the patent in suit at least, and so it is difficult to follow its argument since the figures of the patent and the figure dated 3 December 2007 filed during the examination proceedings clearly show an increase of selectivity when chromium is added to silicon.
Concerning the argument that D8 disclosed selectivities which were higher than those in Example 4, this is true; however, these values are not comparable since the process temperatures are substantially different (example 4: 545°C; D8: 260°C).

It follows that the examples of the patent clearly show that the increase of selectivity to TCS is due to the sole presence of chromium in the silicon feed, and so the problem as defined in the patent is successfully solved.

5.6 It remains to be decided whether the proposed solution is obvious in view of the other documents of the cited prior art.

The board observes that none of the known state of the art documents discloses that chromium improved the selectivity to TCS in the reaction between silicon and HCl gas. It follows that the skilled person faced with the problem of increasing the selectivity to TCS in the above reaction would not find any hint to the solution to this specific problem in anyone of these documents, and so he would also not arrive at the subject-matter of claim 1 of the auxiliary request in an obvious way.

Contrary to appellant II's allegations, the board did not find any indication in documents D4 and D11 that chromium would improve the selectivity to TCS. Furthermore, regarding the indications in paragraph [0003] of the patent and in document D8 that the selectivity to TCS was hardly influenced by the temperature, these indications have no incidence on the inventive step of the claimed subject-matter since - as shown by the examples - chromium has a beneficial influence on the selectivity to TCS not only in the lower temperature range but also in the higher range of temperatures, for instance at 545°C, and so, even if the selectivity decreases with an increase of temperature, there is evidence in the patent that at higher temperatures the effect still exists, and so inventiveness is to be conceded on the whole scope of claim 1 at issue.

It follows that claim 1, and by the same token claims 2 to 6, which include all the features of claim 1, meet the requirements of Article 56 EPC.

6. For the above reasons, the board holds both appeals to be unfounded.

Order

For these reasons it is decided that:

The appeals are dismissed.

The Registrar: The Chairman:

C. Vodz G. Raths