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D E C I S I O N
of 18 July 2001

Case Number: T 1142/97 - 3.3.1

Application Number: 86308369.7

Publication Number: 0223447

IPC: C07F 7/16

Language of the proceedings: EN

Title of invention:
A method of manufacturing alkylhalosilanes

Patentee:
Dow Corning Corporation

Opponent:
GE Bayer Silicones GmbH & Co. KG

Headword:
Alkylhalosilanes/DOW CORNING CORPORATION

Relevant legal provisions:
EPC Art. 56, 83, 100(b)
EPC R. 71(2)

Keyword:
"Main request - sufficiency of disclosure (no)"
"Auxiliary request - inventive step (no) - obvious improvement"

Decisions cited:
-

Catchword:
-

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Case Number: T 1142/97 - 3.3.1

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 18 July 2001

Appellant I: (Opponent)
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Representative:
-

Appellant II: (Proprietor of the patent)
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Representative:
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Decision under appeal:
Interlocutory decision of the Opposition Division of the European Patent Office posted 15 September 1997 concerning maintenance of European patent No. 0 223 447 in amended form.

Composition of the Board:

Chairman: P. P. Bracke
Members: P. F. Ranguis
S. C. Perryman

Summary of Facts and Submissions

I. Appellant I (Opponent) and Appellant II (Proprietor of the patent) lodged an appeal against the interlocutory decision of the Opposition Division to maintain the European patent No. 0 223 447 (application No. 86 308 369.7) in the form as amended pursuant to Article 102(3) EPC.

II. The patent as granted comprised ten claims, independent Claims 1 and 6, reading as follows:

"1. A method of controlling a process for the manufacture of alkylhalosilanes which comprises contacting an alkylhalide with metallurgical grade silicon, at a temperature of 250°C to 350°C, in the presence of tin or tin compounds, and copper or copper compounds, characterised in that there are added to the silicon containing contact mass amounts by weight based on the silicon and calculated as elemental metal of 0.2 to 10 weight percent of copper or copper compounds and 5 to 200 parts per million of tin or a tin compound as co-catalyst, and in that there is also added to said contact mass, in addition to the amount of phosphorus normally present in metallurgical grade silicon, an amount based on the silicon present and calculated as elemental phosphorus, of 25 to 931 parts per million of a phosphorus promoter selected from the group consisting of:

(I) elemental phosphorus;

(II) metal phosphides; and

(III) compounds capable of forming metal phosphides in

the reaction mass of the process."

"6. A composition of matter for use as a silicon-containing contact mass in the manufacture of alkylhalosilanes from alkylhalides by reaction with silicon in the presence of copper and tin as co-catalysts, characterised in that said composition contains metallurgical grade silicon, amounts based on the silicon present and calculated as elemental metal of 0.2 to 10 weight percent of copper or a copper compound and 5 to 200 parts per million of tin or a tin compound and, in addition to the amount of phosphorus normally present in said metallurgical grade silicon, an amount, based on the silicon present and calculated as elemental phosphorus, of 25 to 931 parts per million of a phosphorus promoter selected from the group consisting of:

(I) elemental phosphorus;

(II) metal phosphides; and

(III) compounds capable of forming metal phosphides in the reaction between the alkylhalide and the silicon-containing contact mass."

III. The opposition sought revocation of the patent in suit in its entirety on the ground that the subject-matter of the patent in suit was not patentable under Article 100(b) EPC and under Article 100(a) EPC (lack of inventive step). *Inter alia* the following documents were considered in the contested decision:

(2) N.P. Lobusevich et al., translated from Zhurnal Obshchei Khimii, Vol. 34, No. 8, **1964**, pages 2706

to 2708,

(5) US-A- 4 520 130,

(6) DE-A-3 425 424,

(7) GB-A-2 153 697,

(8a) Ullmanns Encyklopädie der technischen Chemie, 4. Auflage, 21. Band, **1982**, page 418,

(10) M.E. Rubénovitch in C.r. Hebd. Sciences Acad. Sci. Vol. 128, **1989**, 1398-1401,

(12) Affidavit of Dr. Kuivila dated 26 October 1995 filed by the Appellant (Proprietor of the patent),

(13) Affidavit of Dr. Halm dated 24 October 1995 filed by the Appellant (Proprietor of the patent),

(14) The Chemistry of Phosphorus, Pergamon Press, **1973**, pages 406 to 407

(15) Table I "Gibbs free energies of Cu_3P-forming reactions at 600 K" and Table II "Gibbs free energies of formation for copper and phosphorus compounds at 600 K", filed with letter dated 1 July 1997.

IV. The Opposition Division was of the opinion that the European 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 insofar as Claims 1 and 6 related to promoter (III) "compounds capable of forming metal phosphides in the reaction

mass of the process" were concerned (Article 100(b) EPC).

The Opposition Division held, furthermore, that the auxiliary request wherein Claims 1 and 6 as granted were amended by deletion of the feature related to the group (III) promoter could be maintained pursuant to Article 102 (3) EPC.

V. In its decision, the Opposition Division found that the patent in suit did not provide the person skilled in the art with adequate instructions to select the suitable phosphorus compounds capable of forming metal phosphides in the reaction mass under the conditions of the claimed process. Nor could rely the person skilled in the art upon the documents (10), (14) or Table of Gibbs free energies (15) as common general knowledge given those documents were entirely unrelated to the reaction conditions of the claimed process. On the other hand, the Opposition Division found that the claims of the auxiliary request (see point IV above) were not obvious over documents (5) to (7), in particular document (6) as the closest state of the art, in combination with the teaching of document (2) given that it could not be derived therefrom that a copper-tin catalyst system with phosphorus would enhance the overall yield, increase the conversion of raw materials to usable products and would increase the selectivity in favour of Me_2SiCl_2 in the method of manufacturing alkylhalosilanes as claimed.

VI. Appellant I was originally BAYER AG, Germany. The Board was informed on 2 July 1998 by BAYER AG that they had transferred their silicon business to a joint venture with General Electric, and that accordingly with effect

from 1 July 1998 they transferred the opposition to this joint venture, namely GE Bayer Silicones GmbH and Co. KG., to whom future correspondence should be addressed.

VII. Oral proceedings took place on 18 July 2001. The Appellant I informed the Board by letter of 8 June 2001 that it would not be represented at these oral proceedings and requested that a decision be taken on the basis of its written submissions. These Oral proceedings thus took place in the absence of Appellant I (Rule 71(2) EPC).

VIII. Appellant I's submissions in the written proceedings can be summarised as follows:

- Regarding the patent in suit (main request), the decision of the Opposition Division was to be approved given that the patent in suit did not give any guidance to perform the claimed embodiment wherein phosphorus promoters (III) selected from the group consisting of "compounds capable of forming metal phosphides in the reaction mass of the process" were added. Furthermore, document (10) related to the synthesis of PCu_3 by reaction of pure substances which did not correspond to the reaction conditions of the claimed process and document (15) disclosed nothing regarding the conditions of the reaction and the activation energy of each reaction.

- Regarding inventive step of both the main and auxiliary request, the claimed process was obvious in view, on the one hand, of document (6), as the

closest state of the art, and also documents (5) and (7) which were closely related documents, and, on the other hand, document (2). Document (6) disclosed a process of manufacturing methylchlorosilane by effecting the reaction between an alkylhalide and powdered silicon in the presence of a copper zinc catalyst, selectivity and reaction rate being improved by the addition of a "pinch" of tin. Document (2) taught that the addition of phosphorus in an amount of 200 ppm, in addition to promoters like zinc, considerably improved the catalytic properties of silicon-copper alloys. Seeking to improve the process according to document (6) involving a copper zinc tin catalyst, a person skilled in the art would have added with a reasonable expectation of success phosphorus in an amount of 200 ppm in order to arrive at the claimed solution.

IX. The Appellant II's submissions both in the written proceedings and at the oral proceedings can be summarised as follows:

- Regarding the alleged insufficiency of disclosure of the main request, it was conceded that the patent in suit did not give any specific information concerning the compounds capable of forming metal phosphides in the reaction mass of the process. However, the person skilled in the art on the basis of his common general knowledge could have found out without undue burden the appropriate compounds. In particular, document (15) showed that the Gibbs free energy of the reaction at 600°K of copper with phosphorus compounds such as P, PH_3, PCl_3, PCl_5, PBr_3, PI_3 was

negative indicating that the reaction was possible. Documents (10) and (14) confirmed that metal phosphides could be obtained by reaction of metal such as Cu with phosphorus hydrides.

- Regarding inventive step of both main and auxiliary request, the technical problem to be solved in view of document (6) as the closest prior art was to provide a process for the manufacture of alkylhalosilanes involving the reaction of alkylhalide with metallurgical grade silicon providing enhanced overall yields and increased selectivity towards formation of $(CH_3)_2SiCl_2$ or at least an increased selectivity towards formation of $(CH_3)_2SiCl_2$. It would not have been obvious for achieving those improvements to combine the teaching of document (6) with that of document (2) for, on the one hand, not only document (6) but also documents (5) and (7) did not mention the addition of phosphorus and, on the other hand, the person skilled in the art would not have paid attention to document (2) because this document taught that phosphorus was a poison for the reaction and because the disclosed improvement due to the addition of phosphorus to promoters such as zinc, arsenic or antimony was so vague, let alone the fact that tin was not mentioned as a promoter, that the person skilled in the art could not derive from that any practical teaching to solve the above technical problem.

X. The Appellant I requested that the decision be set aside and that the European patent No. 0 223 447 be revoked

The Appellant II requested as main request that the decision under appeal be set aside and that the patent be maintained as granted and as auxiliary request that the appeal of the opponent be dismissed.

XI. At the end of the Oral proceedings the decision was announced orally.

Reasons for the Decision

1. The appeals are admissible.

2. *Identity of the Appellant I/Opponent*

On the basis of the submissions made by the then Appellant I (see point VI above), the Board is satisfied that GE Bayer Silicones GmbH and Co. KG is to be treated as their successor. This was not contested by the Appellant II.

Main request

3. *Article 100(b) EPC - Sufficiency of disclosure*

3.1 The claimed invention relates to three different and individualized ways to perform the addition of 25 to 931 ppm of phosphorus (see point II above).

3.2 The sole question to decide here is whether the European patent discloses the invention in a manner sufficiently clear and complete to be carried out insofar as the claimed process involves the use of a promoter consisting of "compounds capable of forming

metal phosphides in the reaction mass of the process" (alternative III according to the patent in suit).

3.3 The guiding principle is that the person skilled in the art should, after reading the description, and on the basis of the common general knowledge, be able to perform the said alternative III without undue burden.

3.4 It is not disputed by the Appellant II that the patent contains no specific instruction enabling the person skilled in the art to determine from which compounds and in which conditions metal phosphides can be formed in the reaction mass of the process. Appellant II argued nevertheless that the person skilled in the art could find the relevant instructions on the basis of his common general knowledge such as set out in documents (10), (14) and (15).

3.5 However, the Board does not share this opinion for the following reasons:

- Document (10) is a scientific publication which normally does not form part of the common general knowledge. Even if one would accept for the sake of argument that this document was common general knowledge, it would not provide the required instructions, given that the disclosed reaction between phosphine and copper is carried out in presence of pure phosphine and in absolutely air free atmosphere. The conditions of the reactions are clearly different from that which prevail in a mixture comprising alkylhalide, metallurgical grade silicon and tin in addition to copper, let alone the temperature which is different, as recognised by Appellant II.

- Document (14) relates to the preparation of phosphides by the heating of the metal or metal halide with phosphorus without detailing the conditions of the reaction. The fact that this reaction is illustrated by the preparation of boron phosphide by the heating of B_2S_3 with Phosphine at 1200°C to 1400°C cannot give to the person skilled in the art the relevant instructions enabling him to perform without undue burden a reaction between "compounds" in the conditions of the claimed process.

- Document (15) is a list of reactions between phosphorus compounds and copper showing that at 327°C (600°K) the Gibbs free energy of reaction is negative. Those data were not contested by the Appellant I. The Board does not deny that document (15) is of relevance because it shows that copper phosphide can theoretically be obtained by the reaction of copper with phosphorus halides or P or PH_3 given that the Gibbs free energy of reaction is negative. However, as recognised by the Appellant II, a negative free energy of reaction does not guarantee that a reaction necessarily will take place, only that there is sufficient chemical potential for it to occur. As pointed out by the Appellant I the Gibbs free energy of reaction says nothing about the real possibility for the reaction to take place which depends *inter alia* on the activation energy, let alone the fact that it does not take into account the specificity of the contact mass. To summarize, even with the Gibbs free energy data, the person skilled in the art would without any guidance have to devise for himself experiments in the conditions of the

claimed process with all the compounds listed in the Table in order to be able to determine those which possibly might form metal phosphides. In the Board's judgment, embarking on such research without any guidance goes far beyond the routine type of experimentation that can be required from the person skilled in the art when trying to put into practice a claimed invention.

3.6 In view of the above considerations, the Board comes to the conclusion that the European patent does not enable a person skilled in the art on basis of his common general knowledge to achieve without undue burden the claimed process. The present request therefore cannot be allowed.

Auxiliary request

4. *Article 123(2) and (3) EPC*

4.1 The amendments made with respect to the set of claims as granted concern the deletion in Claims 1 and 6 of the feature related to the use of a promoter consisting of "compounds capable of forming metal phosphides in the reaction mass of the process" (see point IV, second paragraph above).

4.2 The Board is satisfied that Claims 1 to 6 are not amended in such a way that they contain subject-matter which extends beyond the content of the application as filed. Those claims are not amended as to extend the protection conferred, either. Those findings were not contested by Appellant I.

5. A*rticle 56 EPC - Inventive step*

5.1 The Board considers, in agreement with the parties, that the closest state of the art is represented by the disclosure of document (6). Indeed, this document aims at the same objective and has the most relevant technical features in common with the claimed subject matter.

5.2 Document (6) relates to a process for the manufacture of alkylhalosilanes involving the reaction of alkylhalide with powdered silicon of at least 98 % purity at a temperature of 250°C to 350°C, in the presence of a catalyst containing copper, zinc and tin, the amounts of those elements being 0.5% to 10% weight of copper based on the silicon, 0.01 to 0.5 parts per million of zinc based on copper and 200 to 3000 parts per million of tin based on copper (see hand-numbered page 10, lines 2 to 13 and hand-numbered page 11, lines 28 to 31).

5.3 In the light of this closest state of the art, the technical problem underlying the patent in suit may be seen, as submitted by the Appellant II and not contested by the Appellant I, in providing a process having enhanced overall yiels from the raw materials and increased selectivity towards formation of $(CH_3)_2SiCl_2$ or at least an increased selectivity towards formation of $(CH_3)_2SiCl_2$ (see page 2, lines 5 to 11 and 29 to 33 of the patent in suit).

5.4 According to the patent in suit this problem is solved by adding to the contact mass, in addition to the amount of phosphorus normally present in metallurgical grade silicon, an amount, based on the silicon present and calculated as elemental phosphorus, of 25 to 931

parts per million of a phosphorus promoter selected from the group consisting of:

(I) elemental phosphorus; and

(II) metal phosphides.

In view of the tests reported in the declaration of Dr. Kuivila (document (12)), a credible case has been put forward that the adding to the contact mass of an amount of phosphorus as defined in Claim 1 provides enhanced overall yiels and increased selectivity towards formation of $(CH_3)_2SiCl_2$. This finding was not challenged by Appellant I. Therefore, the Board accepts that the process as defined in Claim 1 solves the above stated technical problem (see paragraph 5.3 above).

5.5 The remaining question is thus whether the prior art as a whole would have suggested to the person skilled in the art solving the technical problem indicated above in the proposed way.

5.6 Document (6) teaches in the part summarizing the prior art that zinc or tin are **valuable** promoters for copper catalysts or copper-silicon-contact mass (emphasis added by the Board), but that either the formation rate of methylchlorosilane (K_p) or the T/D ratio (methyltrichlorosilane / dimethyldichlorosilane) are often unfavourable. Thus, in the reaction with methylchloride, a K_p of 16 can be obtained when a mixture of powdered silicon with 5 weight percent copper and 0.5 weight percent zinc is used, while a K_p of 45 can be obtained when a mixture of powdered silicon with 5 weight percent copper and 0.005 weight percent tin is used. However, it was observed that the selectivity (T/D ratio) is lower with a tin activated copper catalyst (see hand-numbered page 8, line 21 to hand-numbered page 9, line 15). It was found that the direct reaction between powdered silicon and methylchloride in presence of copper-zinc-tin-catalysts doubled the value of K_p compared to a tin activated copper catalyst and improved the selectivity over tin activated copper catalyst and over zinc activated copper catalyst (see hand-numbered page 9, line 28 to hand-numbered page 10, line 2).

5.7 Document (5) discloses a copper catalyst containing 400 to 3000 ppm of tin for producing an alkyl or arylhalosilane (such as dimethyldichlorosilane from methyl chloride and silicon) at elevated temperature (see column 1, lines 12 to 16 and 37 to 40). The catalyst can also contain a promoter such as zinc or antimony (see column 1, line 67 to column 2, line 1).

5.8 Document (7) discloses a method for making organosilanes which comprises effecting reaction between an organohalogen compound such as methyl

chloride and powdered silicon metal at a purity of at least 98% in a reactor, at a temperature in the range of 250-350°C, in the presence of an effective amount of catalyst composition consisting essentially of:

(a) a mixture of Cu°, Cu_2O and CuO,

(b) from about 200 to 5000 ppm tin, relative to copper and

(c) from about 50 to 5000 ppm aluminium relative to copper.

In addition to the tin and aluminium promoters, the copper catalyst can include iron and zinc (see page 3, lines 5 to 13 and page 4, lines 29 to 30).

5.9 It is true that those documents do not mention the presence of phosphorus. The Board observes, nevertheless, that Appellant II acknowledged that silicon used in documents (5) to (7) was metallurgical grade silicon and that such a silicon contained 20-50 ppm phosphorus as set out in document (8a), on page 418, right-hand column under the paragraph 1.3 "manufacture".

5.10 Document (2), published in 1964, namely 21 years before the priority filing date of the patent in suit, describes the influence of additions of some elements to silicon-copper alloys on their activity in the reaction with methyl chloride. Arsenic, zinc, like antimony, are reported to be active promoters, raising the total and selective activities of the alloys in the synthesis of dimethyldichlorosilane at a concentration of arsenic of 0.05-0.1% and of zinc of 0.5-1.5% (see

page 2727, lines 5 to 8). Furthermore, it is stated that "in the presence of a promoter, phosphorus and beryllium can also act as promoters, making it possible to reduce the synthesis temperature by 20-40°C with a simultaneous increase in the content of dimethyldichlorosilane in the reaction products up to 75%" (see page 2729, second paragraph). Figure 6, on page 2728, shows that the addition of an amount of phosphorus below about 0.09% (900 ppm), in the presence of a promoter, increases the activity of silicon-copper alloys at 360°C regarding the total activity, the content of dimethyldichlorosilane and the productivity with respect to dimethyldichlorosilane (g/h from 1 kg), the maximum being reached when about 0.02% (200 ppm) of phosphorus are present. In summary, the authors conclude that **phosphorus** or beryllium, added to alloys **in addition to promoters** (emphasis added by the Board), considerably improve the catalytic properties of silicon-copper alloys (see page 2729 "Summary" part. 2).

5.11 Appellant II argued first that the documents (5) to (7) represented the teaching applicable on an industrial scale and that none of them mentioned the adding of phosphorus. From this prior art as a whole no incentive to use phosphorus in order to solve the technical problem above defined could be derived. Moreover, the person skilled in the art would not have combined this teaching with that of document (2) for the following reasons: Documents (5) to (7) were published in 1985, while document (2) was published in 1964. The person skilled in the art would not have had, therefore, any reason to consider such a document. Furthermore, the description of document (2) was vague given that Figure 6 did not specify the nature of the promoter and

finally the total activity did not exceed 75%, while this activity was higher using the claimed process.

5.12 The Board cannot accept those arguments for the following reasons:

- Document (6) and also documents (5) and (7) do not mention the adding of phosphorus to the contact mass in addition to the amount of phosphorus normally present in metallurgical grade silicon. However, it was undisputed that metallurgical grade silicon used according to that documents contains by itself 20-50 ppm phosphorus as stated by document (8a). In that context, the Board observes that starting, according to the patent in suit, from a metallurgical grade silicon containing 20 ppm of phosphorus and adding, as claimed, 25 ppm of phosphorus would lead to a contact mass comprising 45 ppm of phosphorus, value which is quite conceivable within the disclosure of documents (5), (6) or (7). As the sole distinguishing feature related to the "adding" of elemental phosphorus was not stated as essential, the Board concludes that those documents do not teach away from the presence of phosphorus in the contact mass.

- It is true that document (2) is old. However, although a period of 21 years is of a significant importance, the Board observes that the starting point (document (6)) relied upon to define the problem to be solved, along with documents (5) and (7) which are closely related to document (6), were published in 1985, the priority filing date year of the patent in suit. In such a case it

cannot be said that a need for the solution of an unsolved problem had existed for a long time, since the new problem arose only with the publication of any of documents (5), (6) or (7). This situation is quite different from that which arose in the decisions cited in Case Law of the Boards of Appeal of the European Patent Office, 3rd edition 1998, I. D. 7.3.

- Starting from document (6) and trying to solve the above defined technical problem, the person skilled in the art would have sought relevant technical information in the same technical field. He would have noted from document (2) that phosphorus enhances the activity of promoters such as zinc, arsenic and antimony in an amount which is below 900 ppm with a maximum of activity at 200 ppm and he would have immediately related this finding to the fact that tin like zinc was considered as a valuable promoter. It would have been, therefore, obvious for a man skilled in the art to add an amount of phosphorus within the defined range to improve with a reasonable expectation of success the process according to document (6) in terms of conversion and selectivity and then get to the claimed invention. It is thereby immaterial that the content of dimethyldichlorosilane in the reaction products is said in document (2) to be only up to 75%. The only relevant information a skilled person gets from document (2) is that by adding phosphorus to alloys in addition to promoters the catalytic activities of silicon-copper alloys in the reaction with methylchloride are considerably improved. From this information a skilled person

could have expected that by adding phosphorus to the metallurgical grade silicon described in document (6) the catalytic activity in the reaction with alkylhalides would be improved.

5.13 For these reasons the Board holds that the subject matter of Claim 1 of the auxiliary request does not involve an inventive step within the meaning of Article 56 EPC so that the patent cannot be maintained in the form as amended in the auxiliary request.

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:

N. Maslin P. P. Bracke