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D E C I S I O N
of 24 October 2001

Case Number: T 0763/98 - 3.3.1

Application Number: 91103753.9

Publication Number: 0446869

IPC: C07C 19/08

Language of the proceedings: EN

Title of invention:

Process for preparing 1,1,1-trifluorochloroethane and 1,1,1,2-tetrafluoroethane

Patentee:

DAIKIN INDUSTRIES, LIMITED

Opponent:

AUSIMONT S.p.A.

Headword:

Tetrafluoroethane/DAIKIN

Relevant legal provisions:

EPC Art. 14(1), 56, 123(2)(3)

Keyword:

"Main request: support in the application as filed (yes) - extension of scope of protection (no)"

"Inventive step (yes) - problem effectively solved by whole scope of claimed process - non obvious solution"

Decisions cited:

T 0165/84, T 0552/89, T 0939/92

Catchword:

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Case Number: T 0763/98 - 3.3.1

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 24 October 2001

Appellant: AUSIMONT S.p.A.
(Opponent) 31, Foro Buonaparte
I-20121 Milano (IT)

Representative: Sama, Daniele, Dr.
Sama Patents
Via G.B. Morgagni, 2
I-20129 Milano (IT)

Respondent: DAIKIN INDUSTRIES, LIMITED
(Proprietor of the patent) Umeda Center Building
4-12 Nakazaki-nishi 2-chome
Kita-ku
Osaka-shi
Osaka-fu 530 (JP)

Representative: Lethem, David James
Hoffmann Eitle
Patent- und Rechtsanwälte
Sardinia House
52 Lincon's Inn Fields
London WC2A 3LZ (GB)

Decision under appeal: Interlocutory decision of the Opposition Division
of the European Patent Office posted 12 June 1998
concerning maintenance of European patent
No. 0 446 869 in amended form.

Composition of the Board:

Chairman: A. J. Nuss
Members: P. P. Bracke
S. C. Perryman

Summary of Facts and Submissions

I. The appeal lies from the Opposition Division's interlocutory decision, dispatched on 12 June 1998, that, account being taken of the amendments made by the Patentee during the opposition proceedings, European patent No. 0 446 869 was found to meet the requirements of the EPC.

In particular, the Opposition Division found that the claimed processes were not obviously derivable from the cited prior art, which existed *inter alia* of documents

(1) WO-A-89/10341,

(3) GB-A-1 589 924,

(5) an English translation of JP-A-48/72105,

(8) US-A-4 792 643 and

(9) US-A-3 752 850.

II. At the oral proceedings before the Board of Appeal, which took place on 24 October 2001, the Respondent (Proprietor of the patent) presented his arguments on the basis of sets of claims according to a main request and a first and second auxiliary request.

The set of claims according to the main request consisted of Claims 1 to 8 and 10 to 12 as filed on 23 March 1999 together with the statement of grounds of appeal and Claim 9 as filed at the oral proceedings before the Board of Appeal.

The two independent process Claims 1 and 9 according to the main request read as follows:

"1. A process for preparing 1,1,1,2-tetrafluoroethane comprising the steps of:

- (i) fluorinating trichloroethylene in the gas phase with hydrogen fluoride in the presence of a fluorination catalyst in a first reactor to form 1,1,1-trifluoro-2-chloroethane; and
- (ii) fluorinating the 1,1,1-trifluoro-2-chloroethane in the gas phase with hydrogen fluoride in the presence of a fluorination catalyst in a second reactor to generate a first gaseous mixture comprising 1,1,1,2-tetrafluoroethane, unreacted 1,1,1-trifluoro-2-chloroethane and 1,1-difluorochloroethylene as a by-product;

characterised in that:

- (a) at least a part of the first gaseous mixture is fed to the first reactor where it acts as a diluent for the fluorination step (i) and where the 1,1-difluorochloroethylene is fluorinated with hydrogen fluoride to 1,1,1-trifluoro-2-chloroethane; and
- (b) the 1,1,1,2-tetrafluoroethane is recovered from the mixture of gases resulting from the fluorination step (i)."

"8. A process for preparing 1,1,1,2-tetrafluoroethane which comprises the steps of:

- (1) reacting trichloroethylene with hydrogen fluoride in the gas phase in the presence of a fluorination catalyst to obtain 1,1,1-trifluoro-2-chloroethane in a first reactor,
- (2) reacting 1,1,1-trifluoro-2-chloroethane from the first reactor with hydrogen fluoride in the gas phase in the presence of a fluorination catalyst to obtain 1,1,1,2-tetrafluoroethane and by-product 1,1-difluorochloroethylene in a second reactor,
- (3) recycling the entire reaction mixture including 1,1,1,2-tetrafluoroethane and unreacted 1,1,1-trifluoro-2-chloroethane from the second reactor to the first reactor,
- (4) reacting 1,1-difluorochloroethylene produced in the second reactor with hydrogen fluoride to reduce the amount of 1,1-difluorochloroethylene in the first reactor, and
- (5) recovering 1,1,1,2-tetrafluoroethane from the reaction mixture obtained from the first reactor prior to feeding this mixture to the second reactor."

The dependent Claims 2 to 7 were directly or indirectly appended to Claim 1 and the dependent Claims 9 to 12 were dependent on Claim 8.

III. The Appellant (Opponent) contested that Claim 1 according to the main request, which required the

presence of a fluorination catalyst in both the first and second reactor, met the requirement of Article 123(3) EPC for the reason that in Claim 1 as granted the presence of a fluorination catalyst was not mandatory, with the effect that the process in its granted form was inoperative. Thus, so argued the Appellant, the scope of this granted claim is absent (zero) and the amendment therefore extended the scope of protection beyond that of the granted claims. Moreover, the Appellant submitted that, in the absence of any indication at which temperature the fluorination reactions are conducted in Claims 1 and 8, such claims are restricted to fluorination processes conducted at ambient temperature. Since it follows from the prior art that fluorination reactions only take place at higher temperatures, the desired technical effects were not obtained by the claimed processes.

For the Appellant the problem underlying the invention was the provision of a process for preparing 1,1,1,2-tetrafluoroethane (further referred to as R-134a) in high yield starting from trichloroethylene (further referred to as TCE) with a simple apparatus at a reduced cost allowing the separation of R-134a which is not complicated by the presence of 1,1-difluoroethane (further referred to as R-1122). As it was not a feature of Claim 1 according to the main request that the complete effluent of the second reactor was fed to the first reactor, the Appellant submitted that the stated technical problem had not been solved, since the problem of separating R-134a in the presence of R-1122 still remained.

The Appellant also argued that, starting from document (3) as the closest state of the art, a skilled person

would have deduced from the teaching of document (3) in combination with the teachings of documents (1), (8) and (9) that R-134a may be prepared by converting trichloroethylene into 1,1,1-trifluoro-2-chloroethane (further referred to as R-133a) in a first low temperature reactor, fluorinating R-133a into R-134a in a second high temperature reactor and reducing the amount of unwanted R-1122 in a third low temperature reactor. As it was known that both trichloroethylene and R-1122 may be converted with hydrogen fluoride into R-133a, a skilled person would have been unambiguously directed to perform both said reactions in the same low temperature reactor, thus simplifying the apparatus and reducing the cost by eliminating one reactor.

- IV. The Respondent argued that the fact that Claim 1 as granted was silent about the presence of a catalyst did not mean that the claim was restricted or limited to fluorination reactions excluding the presence of a catalyst and the fact that the wording of Claims 1 and 8 according to the main request was silent about the temperature at which the reaction was conducted did not mean that such claims were restricted to fluorination reactions at ambient temperature.

Furthermore, the Respondent submitted that, even when assuming that the technical problem underlying the invention was the least ambitious one, namely the provision of a further process for manufacturing R-134a starting from TCE, the claimed process was not obviously derivable from the cited prior art documents.

- V. The Appellant requested that the decision under appeal be set aside and that the European patent No. 0 446 869 be revoked.

The Respondent requested that the decision under appeal be set aside and that the patent be maintained on the basis of, as main request, Claims 1 to 8 and 10 to 12 filed on 23 March 1999 and Claim 9 filed at the oral proceedings on 24 October 2001, or Claims 1 to 11 of the first auxiliary request, or Claims 1 to 9 of the second auxiliary request, both auxiliary requests being filed at the oral proceedings on 24 October 2001.

Reasons for the decision

1. The appeal is admissible.

2. Document (1), published in the Japanese language, incontestably belongs to the state of the art according to Article 54(2) EPC. Instead of filing a translation in one of the official languages of the EPO according to Article 14(1) EPC of document (1), the Appellant filed the corresponding EP-A-0 366 797, which was published after the first claimed priority date and which is further referred to as document (1a), as an English translation of the Japanese document (1). As the Respondent accepted that the content of document (1a) corresponds to the content of document (1) and as the Board does not have any reason to question this, document (1a) is relied on in this decision as being a true translation of prior published document (1) (but not as being itself prior published).

3. *Main request*
 - 3.1 Article 123(2) and (3) EPC

Present Claim 9, corresponding with Claim 10 underlying

the contested decision, was modified in that the contested wording saying that the temperature in the first reactor was lower than that in the second reactor, was replaced by the requirement that the first reaction is carried out at a temperature of 180 to 300°C, and the second reaction is carried out at a temperature of 300 to 400°C. Since such reaction temperature ranges were described on page 6, lines 13 to 15, and on page 8, lines 22 and 23, of the application as filed, the Appellant did not contest any more that Claim 9 met the requirement of Article 123(2) EPC. Also Claims 1 to 12, as a whole, were not contested to meet the requirement of Article 123(2) EPC. Also the Board has reached that conclusion.

Nonetheless, the Appellant contended that the protection conferred by Claim 1 extended the protection conferred in comparison with the claims as granted, since according to present Claim 1 the fluorination must be conducted ***in the presence of a fluorination catalyst***, whereas Claim 1 as granted was silent about the presence of a catalyst (see point III, first paragraph).

This contention, however, is at variance with the normal practice of claim interpretation that if a claim places no restrictions on a feature, such as here the presence or absence of a catalyst, then the claim covers all possibilities for this feature. As Claim 1 as granted is silent about the presence or absence of a fluorination catalyst, the subject-matter of that claim in fact encompassed any process having all the features explicitly required by that claim, regardless of a fluorination catalyst being present or not. The additional requirement in present Claim 1 that the

fluorination reaction in both the first and second reactor is conducted in the presence of a fluorination catalyst thus does not result in an extension of the protection conferred by Claim 1 but in the restriction thereof.

For the point at issue here, decision T 165/84 of 29 January 1987, cited by the Appellant, is irrelevant, since that decision was concerned with a claim which as originally worded was considered to lack clarity, because essential features were lacking, which defect could not be cured because the application contained no information concerning these essential features. In the present case, however, the description contained all the necessary information on the process.

3.2 Novelty

After examination of the cited prior art documents, the Board has reached the conclusion that none of those documents describes all features of the processes as defined in Claims 1 and 8 and, consequently, that Claims 1 to 12 are novel over the cited prior art. Since this was not disputed, it is not necessary to give detailed reasons for this finding.

3.3 Inventive step

- 3.3.1 In accordance with the "problem-solution approach" applied by the Boards of Appeal to assess inventive step on an objective basis, it is necessary to establish the closest state of the art being the starting point, to determine in the light thereof the technical problem which the invention addresses and solves, and to examine the obviousness of the claimed

solution to this problem in view of the state of the art.

- 3.3.2 There was disagreement between the Parties as to which document represented the closest state of the art to be used as a starting point for assessing inventive step.

As the "closest state of the art" must be a prior art document disclosing subject-matter aiming at the same objective as the claimed invention and the objective in the present case is a process of preparing R-134a starting from TCE, only such documents could qualify as closest state of the art which also concern a process of preparing R-134a starting from TCE.

The Respondent submitted that document (1a) could qualify as the closest state of the art.

Document (1a) is related to a process for the preparation of an organic fluorine compound by reacting an organic chlorine compound or an organic unsaturated compound with hydrogen fluoride (see page 3, lines 23 to 34). Although document (1a) describes on page 13, lines 25 to 29 the fluorination of TCE to R-133a and on page 14, lines 4 to 9, the fluorination of R-133a to R-134a, it is clear from the teaching on page 13, lines 22 to 24, that such fluorination reactions are cited only as possible examples of the starting materials and reaction products in the process described therein. This document only discloses the two fluorination reactions independently of each other, without mentioning or even suggesting the preparation of R-134a starting from TCE.

Document (3), which was considered by the Appellant as

representing the closest state of the art, describes the preparation of R-134a starting from R-133 without giving any information how R-133a was obtained.

As thus neither document (1a) nor document (3) discloses the preparation of R-134a starting from TCE, none of those documents can qualify as the closest state of the art.

Since the only cited prior art document describing the preparation of R-134a starting from TCE is document (8), only this document can qualify as representing a suitable starting point for assessing inventive step.

3.3.3 Document (8) describes the fluorination of a trihaloethylene, preferably TCE, into R-134a in a vapour phase using a solid chromium-based catalyst (column 1, lines 6 to 10, column 2, lines 27 to 44, column 3, lines 53 to 59). In column 4, lines 22 to 27, it is also said that the major impurity in the product mixture emerging from the reaction is R-133a, which can be converted to R-134a by further fluorination over a catalyst.

3.3.4 Considering the Respondent's submission that in the light of the available prior art the least ambitious technical problem which the invention addresses and solves is the provision of a further process for manufacturing R-134a starting from TCE, the first point to be considered in assessing inventive step is then whether it has been convincingly shown that by the processes according to Claims 1 and 8 this problem has effectively been solved.

It has never been contested that with the data provided

in Example 1 of the patent in suit a credible case has been put forward that R-134a may be manufactured starting from TCE.

Nevertheless, the Appellant contested that the desired technical effect is obtained over the complete scope of the claimed process since Claims 1 and 8 are silent about the temperature at which the fluorination reactions are conducted and, consequently, the subject-matter of those claims is restricted to processes conducted at ambient temperature, at which temperature it is known from the prior art that fluorination does not take place.

However, the Board cannot follow this argumentation, because the processes as defined in Claims 1 and 8 only relate to such processes where in the first and second reactor fluorination with hydrogen fluoride to form or generate the specifically indicated products takes place accordingly. In view of this requirement processes wherein fluorination is excluded, due for example to unsuitable temperatures, cannot be regarded as being encompassed by the subject-matter defined in the claims.

In this respect, decision T 939/92 (OJ EPO 1996, 309), cited by the Appellant, is not relevant, since that decision concerns the principle that a technical effect which justifies the choice of the claimed compounds must be one which can be fairly assumed to be produced by substantially all the chosen compounds, whereas in the present case the technical effect of fluorination is necessarily achieved by any process falling under the claims, since the claims are restricted to those processes in which fluorination is achieved.

Additionally, since Claim 1 also embraces the possibility that only a part of the first gaseous mixture is fed to the first reactor, the Appellant argued that the problem underlying the patent in suit was not effectively solved for the complete claimed scope.

The Board can also not follow this argumentation, because the problem to be solved is only the provision of a **further** process for manufacturing R-134a starting from TCE. As it follows from the wording of Claim 1 that **at least** a part of the first gaseous mixture is fed to the first reactor and that R-134a is recovered from the mixture of gases resulting from the fluorination step (i), it is clear that R-134a is recovered only from that part of the first gaseous mixture that is fed to the first reactor. That the yield of R-134a in a process according to Claim 1 may be lower than in one according to Claim 8 (i.e. when only part of the first gaseous mixture is fed to the first reactor) is not relevant, since the problem underlying the claimed invention is only the provision of a **further** process. This means that what matters in the present case is that the desired product R-134a is also produced in a process in accordance with Claim 1 albeit possibly in a lower yield than in a process in accordance with Claim 8.

Consequently, the Board comes to the conclusion that a credible case has been put forward that R-134a may be prepared from TCE.

3.3.5 Therefore, it remains to be decided whether the process according to Claim 1 or 8 is an obvious solution to the stated technical problem in view of the cited prior

art. In particular, the question arises whether it was suggested in the cited prior art

- (i) to feed at least a part of the first gaseous mixture generated in the second reactor to the first reactor where R-1122 is fluorinated with hydrogen fluoride to R-133a thus reducing the amount of R-1122 and
- (ii) to recover R-134a from the mixture of gases resulting from the fluorination step in the first reactor prior to feeding this mixture to the second reactor.

3.3.6 It is true that document (8) teaches in column 6, lines 16 to 21, that a high content of R-133a is present in the product mixture obtained from the fluorination reaction of TCE to R-134a described therein and that it may be further fluorinated by, for example, recycling. However, for a skilled person this suggests first separating the R-134a, and then feeding the effluent gases from the separation, or at least the R-133a, back to the same reactor. In document (8) no suggestion can be found to convert TCE into R-134a by two fluorination steps in two reactors, nor to feed a stream of gases still containing the desired end-product R-134a back to any fluorination reaction or to recover R-134a in an intermediate stage before conducting a further fluorination. Moreover, document (8) is completely silent about the problems arising from the presence of R-1122 in order to obtain R-134a, let alone, about the possibility of converting R-1122 to R-133a.

3.3.7 Document (1a), which is concerned with the preparation

of R-134a from R-133a, mentions the problem of the presence of R-1122 in R-134a. Namely, it teaches in the third paragraph on page 14, that a separation of R-1122 from R-134a is very difficult. However, as solution of this problem it proposes to control the formation of R-1122 as much as possible by the molar ratio of hydrogen fluoride to R-133a. Nowhere in document (1a) is it mentioned or suggested that the problem of the presence of R-1122 in the reaction mixture, obtained by converting R-133a into R-134a, could be solved by feeding at least part of the effluent of that conversion to a fluorination reaction, let alone to the fluorination reaction of TCE into R-133a. Additionally, nowhere in document (1a) can any indication be found to recover R-134a only after the conversion of TCE into R-133a and before the further fluorination of R-133a to R-134a.

- 3.3.8 Document (3) is also concerned with the preparation of R-134a from R-133a, and it also mentions the problem of the formation of R-1122 in the fluorination of R-133a into R-134a (see page 2, lines 5 to 8). According to document (3), it was found that the content of R-1122 may be reduced by treating the mixture with hydrogen fluoride in the presence of the same catalyst used for the fluorination of R-133a into R-134a but at much lower temperature (see page 2, lines 12 to 17). On page 2, lines 34 to 54, it is taught in detail that R-133a may be converted with hydrogen fluoride over a catalyst into R-134a in a first reactor or reaction zone at 300 to 400°C and that the amount of undesired R-1122 may be reduced by conducting a further fluorination reaction in a second reactor or reaction zone at 100 to 275°C.

As document (3) is silent about the preparation of the starting R-133a, nowhere in document (3) can a suggestion be found to reduce the amount of R-1122 simultaneously in the same reactor with the preparation of R-133a from TCE and, certainly, also in document (3) nowhere could a suggestion be found to recover R-134a only after the conversion of TCE into R-133a and before the further fluorination of R-133a to R-134a.

The Appellant referred to Example 5 of document (3), describing the fluorination of R-133a over a catalyst in a first reactor at a temperature of 335 to 355°C and passing the exit gas of the first reactor over a catalyst in a second reactor at 160°C, thus reducing the amount of the undesired R-1122. As the amount of R-134a in the exit gas of the first reactor was exactly the same as in the exit gas of the second reactor, the Appellant argued that a skilled person would have concluded therefrom that R-134a was inert at fluorination conditions for converting R-1122 into R-133a. Since it was known, for example, from document (9), that TCE too can be converted into R-133a at such fluorination conditions, it was obvious to reduce the amount of R-1122 and to convert TCE into R-133a in the same reactor under the same fluorination conditions, whereby the amount of TCE is such as to replace the amount of R-134a recovered. Therefore, the lack of possible alternatives created a "one-way-street" situation, leading a skilled person to the claimed process.

The Board cannot, however, follow this line of argumentation. In combining the teachings of document (3) and (9), a skilled person would rather be led to convert TCE into R-133a in a first reactor, further

fluorinate R-133a into R-134a in a second reactor and reducing the amount of undesired R-1122 in a third reactor. Moreover, the reaction conditions when subjecting the effluents of the reactor, in which the conversion of R-133a into R-134a takes place, to a second fluorination reaction, as described in Example 5 of document (3), are not identical with the reaction conditions for converting TCE to R-133a, since in the conversion of TCE into R-133a much more hydrogen chloride is produced than in the conversion of R-1122 into R-133a. As a skilled person does not have any indication whether R-134a would be inert in such a hydrogen chloride rich medium, he does not have any pointer as to how to reduce the amount of R-1122 in an effluent containing the desired end-product R-134a simultaneously with the preparation of R-133a from TCE. Rather, a skilled person would not risk reverse reactions which might reduce the amount of the desired endproduct R-134a.

Certainly, document (3) mentions "recycling" in the sentence bridging pages 1 and 2. This is, however, clearly in the context of drawing off at least part of the mixture, separating 134a from unreacted starting material, hydrogen fluoride and by-products (e.g. haloethanes) and feeding the latter back to the reactor for producing R-134a and not to some other reactor for preparing R-133a.

Finally, a one way street situation can only be accepted as existing when a skilled person is in a situation that in view of the teaching of the prior art he does not have any alternative to the claimed solution. As the amount of R-1122 in the effluent of the second reactor may however not only be reduced by a

further fluorination reaction but also, for example, by treating the effluent with a metal permanganate, as known from page 2, lines 57 to 61, of document (3), the Board cannot accept that in the present case the skilled person was in a "one-way-street" situation.

- 3.3.9 The Appellant also argued that the claimed processes were rendered obvious by the teaching of document (5), because this document, which is related to a process of reacting halogenated olefins with hydrogen fluoride in a gaseous phase in the presence of a catalyst (see page 5, lines 2 to 5), teaches on page 7, lines 8 to 11, that the reaction gas may also comprise, besides halogenated olefin and hydrogen fluoride, lower fluorinated compounds as recycled fraction.

However, this teaching may not be taken in isolation and should be interpreted in its context. As in lines 5 to 8 on page 7 of document (5) a difference is made between lower fluorinated compounds, containing only a few fluor atoms per molecule, and higher fluorinated compounds, containing many fluor atoms per molecule, it is clear that R-134a is to be considered as a higher fluorinated compound. Therefore, document (5) only suggests to feed fluorinated compounds which do not yet have the desired degree of fluorination back to the same reactor where it was obtained from in order to increase its degree of fluorination. Since document (5) does not suggest to feed a compound which has the desired fluor substitution back to a fluorination reaction, as is the case in the claimed processes, such processes are not rendered obvious by the teaching of document (5).

- 3.3.10 The Appellant cited decision T 552/89 of 27 August

1991, which states at point 2.2 that when the objective problem established having regard to the closest state of the art as disclosed in a primary document is formed of individual problems, then the skilled person can be expected to take account of solutions to the individual problems proposed in different secondary documents in the same or neighbouring technical fields. In the present case, however, the question in assessing inventive step is whether the claimed solution can be derived as an obvious **further** process for manufacturing R-134a starting from the cited prior art documents. There is here no simple combination of known solutions to individual problems, so this line of argument for obviousness must fail.

3.3.11 Therefore, the Board comes to the conclusion that the processes according to Claims 1 and 8 are not obviously derivable from the cited prior art.

Claims 2 to 7 and Claims 9 to 12, which represent preferred embodiments of Claim 1 respectively Claim 8, derive their lack of obviousness from the same inventive concept.

3.3.12 As an inventive step can already be accepted when considering that in the light of the available prior art the least ambitious technical problem which the invention can be considered to address and solve is the provision of a further process for manufacturing R-134a starting from TCE, it is not necessary for the Board to consider whether the Claims 1 and 8 necessarily also achieve an advantage over the closest prior art for the whole scope of the claims, as a precondition for recognition that a more ambitious problem is solved by the invention.

4. *Auxiliary requests*

In the light of the above findings, there is no need to consider the auxiliary requests.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The matter is remitted to the first instance with the order to maintain the patent on the basis of Claims 1 to 8 and 10 to 12 as filed on 23 March 2001 and Claim 9 of the main request as filed at the oral proceedings on 24 October 2001 and a description to be adapted.

The Registrar:

The Chairman:

N. Maslin

A. Nuss