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**D E C I S I O N**  
**of 16 April 2002**

**Case Number:** T 0045/99 - 3.3.1

**Application Number:** 91302703.3

**Publication Number:** 0449617

**IPC:** C07C 19/08

**Language of the proceedings:** EN

**Title of invention:**

Process for the preparation of 1,1,1,2-tetrafluorethane

**Patentee:**

Ineos Flour Holdings Limited

**Opponent:**

AUSIMONT S.p.A.

**Headword:**

Tetrafluoroethane (II)/INEOS FLUOR HOLDINGS

**Relevant legal provisions:**

EPC Art. 14(1), 54(3), 56

**Keyword:**

"Novelty (yes) - combination of features not anticipated"

"Inventive step (yes) - nonobvious solution"

**Decisions cited:**

T 0939/92

**Catchword:**

-



Case Number: T 0045/99 - 3.3.1

**D E C I S I O N**  
**of the Technical Board of Appeal 3.3.1**  
**of 16 April 2002**

**Appellant:** AUSIMONT S.p.A.  
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I-20121 MILANO (IT)

**Representative:** Sama, Daniele, Dr.  
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**Respondent:** Ineos Fluor Holdings Limited  
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**Representative:** Geary, Stephen  
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**Decision under appeal:** Decision of the Opposition Division of the  
European Patent Office posted 13 November 1998  
rejecting the opposition filed against European  
patent No. 0 449 617 pursuant to Article 102(2)  
EPC.

**Composition of the Board:**

**Chairman:** A. J. Nuss  
**Members:** P. P. Bracke  
J. P. B Seitz

## Summary of Facts and Submissions

- I. The appeal lies from the Opposition Division's decision, dispatched on 13 November 1998, that granted Claims 1 to 10 of European patent No. 0 449 617 were found to meet the requirements of novelty and inventive step over the cited prior art.

The sole independent claim as granted read:

"1. A method for the manufacture of 1,1,1,2-tetrafluoroethane which comprises the steps of:

- (A) contacting a mixture of 1,1,1-trifluoro-2-chloroethane and hydrogen fluoride with a fluorination catalyst at a temperature in the range of 280 to 450°C in a first reaction zone to form a product containing 1,1,1,2-tetrafluoroethane and hydrogen chloride together with unreacted starting materials,
- (B) passing the total product of step (A) together with trichloroethylene to a second reaction zone containing a fluorination catalyst at a temperature in the range of 200 to 400°C but lower than the temperature in step (A) to form a product containing 1,1,1-trifluoro-2-chloroethane, hydrogen chloride and unreacted trichloroethylene,
- (C) treating the product of step (B) to separate 1,1,1,2-tetrafluoroethane and hydrogen chloride from 1,1,1-trifluoro-2-chloroethane, unreacted hydrogen fluoride and trichloroethylene,

- (D) feeding the 1,1,1-trifluoro-2-chloroethane mixture obtained from step (C) together with hydrogen fluoride to said first reaction zone (step (A)), and
- (E) recovering 1,1,1,2-tetrafluoroethane from the 1,1,1,2-tetrafluoroethane and hydrogen chloride separated out in step (C)."

In particular, the Opposition Division was of the opinion that the claimed method was not known from document

- (1) EP-A-0 446 869, which was cited as prior art according to Article 54(3) EPC, and

that the claimed method was not obviously derivable from the cited prior art according to Article 54(2) EPC, which existed *inter alia* of documents

- (2) WO-A-89/10341,
- (4) GB-A-1 589 924,
- (6) an English translation of JP-A-48-72105,
- (9) US-A-4 792 643 and
- (10) US-A-3 752 850.

II. Oral proceedings before the Board took place on 16 April 2002.

III. The Appellant (Opponent) submitted that all reaction steps (A) to (E) according to Claim 1 as granted were

known from document (1). In particular, the Appellant was of the opinion that steps (C) and (E) were disclosed in document (1).

As the preparation of 1,1,1-trifluoro-2-chloroethane (further referred to as R-133a) by the fluorination of trichloroethylene (further referred to as TCE) was known from documents (2) and (10), the fluorination of R-133a to 1,1,1,2-tetrafluoroethane (further referred to as R-134a) was known from documents (2) and (4) and the use of an inert gas as a diluent in the fluorination of olefins was known from document (6), the Appellant was of the opinion that the claimed method was the only way suggested by the teachings of the prior art to prepare R-134a from TCE in high yield and with low levels of 1-chloro-2,2-difluoroethylene (further referred to as R-1122).

- IV. The Respondent submitted that the claimed method was distinguished from the method disclosed in document (1) by the **simultaneous** separation of R-134a and hydrogen chloride instead of the **subsequent** separation of those components from the product stream of step (B).

Moreover, the Respondent argued that the claimed integrated method of preparing R-134a from TCE was not obviously derivable from the cited prior art documents, as it was nowhere suggested to use the effluent of step (A) as diluent in step (B) and separating R-134a only after step (B).

- V. The Appellant requested that the patent be revoked.

The Respondent requested that the appeal be dismissed.

## Reasons for the Decision

1. The appeal is admissible.
  
2. *Novelty*
  - 2.1 Document (1), which was not contested to belong to the state of the art according to Article 54(3) EPC, describes a process for preparing R-134a by reacting, in a first reaction, TCE and hydrogen fluoride in a gas phase to obtain R-133a and fluorinating, in a second reaction, R-133a with hydrogen fluoride to obtain R-134a, wherein the whole of the generated gas from the second reaction may be used as the diluent gas in the first reaction (see column 2, lines 36 to 45, and column 4, lines 12 to 15). In column 4, lines 33 to 35, it is stated that from the generated gas from the first reactor (ie the fluorination of TCE), hydrogen chloride is removed **and then** R-134a is separated.

In the presently claimed method, to the contrary, in step (C) the product of step (B) is treated to separate R-134a and hydrogen chloride from R-133a, unreacted hydrogen fluoride and TCE and in a subsequent step (E) R-134a is recovered from the mixture separated out in step (C).

- 2.2 Nevertheless, the Appellant was of the opinion that the separation of a mixture of R-134a and hydrogen chloride from the reaction mixture of step (B) and the subsequent recovery of R-134a from that mixture could not be a distinguishing feature, because on page 3, lines 20 and 21, in the patent in suit it is stated that the separation of R-134a and hydrogen chloride

from the product stream in step (C) may be effected by a distillation technique. Since hydrogen chloride has a lower boiling point than R-134a, the Appellant argued that by fractional distillation hydrogen chloride is removed before R-134 is distilled and, thus, that it is not a requirement according to step (C) of present Claim 1 that R-134a and hydrogen chloride are separated together.

2.3 In order to be novelty destroying, however, all features in the claimed combination must be directly and unambiguously derivable from the teaching of one single document. Therefore, in the present case the relevant question is whether from document (1) a method comprising the steps (A), (B) and (D), wherein the reaction mixture obtained from step (B) is treated to separate R-134a and hydrogen chloride from R-133a, unreacted hydrogen fluoride and TCE (step (C)) and wherein R-134a is subsequently recovered from the mixture separated out in step (C), is directly and unambiguously derivable from document (1).

From column 4, lines 33 to 35, of document (1) it may only be derived that hydrogen chloride is separated out from the reaction mixture obtained by fluorinating TCE and that subsequently **R-134a is separated from the remaining reaction mixture** before the then remaining mixture is conducted to the conversion reaction of R-133a into R-134a. Such teaching cannot be considered an unambiguous disclosure of the steps according to the claimed method, wherein hydrogen chloride and R-134a are both separated out from the reaction mixture obtained by fluorinating TCE (step (C)) and the remaining mixture is conducted to the conversion reaction of R-133a into R-134a (step (D)) **whereas the**

**R-134a is recovered from the separated mixture**

(step (E)).

- 2.4 The Appellant also argued that example 3 of document (1) disclosed all features of the claimed method.

Example 3 describes a method of fluorinating R-133a to form R-134a, subsequently bringing the thus obtained exit gas together with TCE into contact with a catalyst and analysing the exit gas by gas chromatography. As example 3 is completely silent about separating R-134 out at any moment, steps (C) and (E) are not disclosed therein.

- 2.5 In the absence of a direct and unambiguous disclosure of steps (C) and (E), document (1) cannot be considered to be novelty destroying for the method of Claim 1.

3. *Inventive step*

- 3.1 Document (2), 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 (2), the Appellant filed the corresponding EP-A-0 366 797, which was published after the claimed priority date and which is further referred to as document (2a), as an English translation of the Japanese document (2). As the Board does not have any reason to question that the content of document (2a) corresponds to the content of document (2), document (2a) is relied on in this decision as being a true translation of prior published document (2) (but not as being itself prior published).



3.2 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 Since 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.

As document (4), which was considered by the parties as representing the closest state of the art during the written proceedings, describes only the preparation of R-134a starting from R-133 and not the preparation of R-134a starting from TCE, it cannot 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 (9), only this document can qualify as representing a suitable starting point for assessing inventive step.

3.4 Document (9) 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 also states 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.

- 3.5 According to the patent in suit the prior art methods of fluorinating TCE into R-134a suffered from quite low yields (see page 2, lines 9 to 12) and the presently claimed method provides improved yields (see page 2, lines 9 to 14). Moreover, on page 3, lines 25 to 28, of the patent in suit the claimed method is said to have the advantage that the R-134a collected from step (B) contain a smaller amount of R-1122 than R-134a produced in step (A).

The first point to be considered in assessing inventive step is then whether it has been convincingly shown that, in view of the closest prior art, the claimed method has the mentioned advantages, namely providing R-134a from TCE in a "higher" yield and a "lower" R-1122 level, and, thus, that the problem underlying the invention has effectively been solved.

The only data available are those contained in Tables 1 and 2 of the patent in suit and obtained by comparing the claimed method with multi-step methods of preparing R-134a from TCE, differing from the claimed method by the order of reaction. There is no information available and it was never submitted that such multi-step methods correspond to some prior art in the sense of Article 54(2) EPC. Since a comparison is thereby not made with a fluorination reaction as described in document (9), it cannot be considered that with those data a credible case has been put forward that the

claimed method provides improved yields over the closest state of the art. From those data, showing that with the claimed method R-134a may be prepared from TCE in yields of 93.7% and 87.5% with R-1122 levels of only 16 ppm and 29 ppm, it may only be concluded that a credible case has been put forward that with the claimed method R-134a may be obtained from TCE in high yields and with low levels of R-1122. This has never been contested.

3.6 Therefore, it remains to be decided whether the method according to Claim 1 is an obvious solution to the solved technical problem in view of the cited prior art. In particular, the question arises whether it was suggested in the cited prior art to feed the total product of step (A) together with TCE to a second reaction zone, as defined in step (B) of Claim 1, to separate and recover R-134a only from the mixture of gases obtained from step (B) and to feed the R-133a mixture obtained from step (C) together with hydrogen fluoride to the first reaction zone (step (A)) in order to prepare R-134a with low R-1122 levels from TCE in high yield.

3.7 The Appellant submitted that, in view of the problem to be solved starting from the teaching of document (9) (see point 3.4 above), there was no other way to combine the teaching of document (2a) or (4) with the teaching of document (10) as defined in Claim 1. As support of this submission the Appellant referred to the principle described in decision T 939/92 (OJ EPO 1996, 309), that what a skilled person would have done in the light of the state of the art depended on the technical results he had set out to achieve.

3.8 It is true that document (9) 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 (9) no suggestion can be found to convert TCE into R-134a by two fluorination steps in two reaction zones, nor to feed a stream of gases still containing the desired end-product R-134a to a fluorination reaction to convert TCE into T-133a and isolating R-134a only from the stream of gases obtained from the conversion reaction of TCE into R-133a. Moreover, document (9) is completely silent about the problems arising from the presence of the toxic impurity R-1122 in preparing R-134a from R-133a, let alone, about the possibility of achieving a controlled low level of R-1122 in R-134a, ie the desired end-product.

3.9 Document (4) is concerned with the preparation of R-134a from R-133a, and it 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 (4), 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 (4) is silent about the preparation of the starting R-133a, nowhere in document (4) 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 (4) 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.

Certainly, document (4) 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 R-134a from unreacted starting material, hydrogen fluoride and by-products (eg haloethanes) and feeding the latter back to the reactor for producing R-134a and not to some other reactor for preparing R-133a.

- 3.10 Document (2a) 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 (2a) 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 processes

described therein. This document only discloses the two fluorination reactions independently of each other, without mentioning or even suggesting the direct preparation of R-134a starting from TCE. Document (2a) 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 (2a) a suggestion can be found to prepare R-134a from TCE in two reaction zones, let alone, to feed the stream of gases obtained by fluorinating R-133a into R-134a into another reaction zone for fluorinating TCE in R-133a.

- 3.11 The Appellant referred to example 5 of document (4), 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 (10), 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 reaction zone 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.

Moreover, the Appellant argued that it was known from document (2a), page 13, line 22 to page 14, line 23, that TCE may be fluorinated at 200 to 450°C to form R-133a and that R-133a may be fluorinated at 300 to 420°C to form R-134a and that it followed from example 21 of document (2a) that by fluorinating R-133a containing 2% of R-1122 at 200°C R-1122 was fluorinated while R-133a remained unreacted. Since TCE, like R-1122, also contains a double bond, there was a pointer in document (2a) to fluorinate TCE and to reduce the amount of R-1122 at the same lower temperature and, thus, to conduct the fluorination of TCE and the reduction of the level of R-1122 in the same reaction zone.

- 3.12 The Board cannot, however, follow this line of argumentation. 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 (4), are not identical and thus not to be confused 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. Moreover, from the broad temperature range of 200 to 450°C given in document (2a) for the conversion of TCE into R-133a a skilled person could not deduce that TCE could be fluorinated at the same reaction conditions as described for the reduction of the fluorination of R-1122. 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 end product R-134a, but use a known inert gas such as nitrogen, as described in the second paragraph on page 7 of document (6).

In combining the teachings of document (2a) or (4) and the teaching of document (10), 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.

- 3.13 In this respect, the Appellant alleged that a skilled person could have expected that at the lower temperatures for converting TCE to R-133a the R-134a would be inert and, consequently, that the equilibrium between R-134a and R-133a would not be negatively influenced.

As a matter of principle, however, the burden of proof is upon the party making an allegation. Since, in the present case, the Appellant made an unsubstantiated allegation, which the Respondent contested, the Board does not have any reason to accept such allegation.

- 3.14 Therefore, it is nowhere suggested in document (2a) or (4) 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 the effluent of that conversion to the fluorination reaction of TCE



into R-133a and nowhere is it suggested that by doing so R-134a could be obtained in high yield.

Additionally, nowhere in document (2a) or (4) any indication can 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.

3.15 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 according to the teachings of documents (2a), (4) and (10) a skilled person would at least also consider the possibility to use three reactors (see point 3.9 above), the Board cannot accept that in the present case the skilled person was in a "one-way-street" situation. Apart from that, he was also aware that the amount of R-1122 could be efficiently reduced, for example, by metal permanganate treatment (see document (4), page 2, lines 57 to 61).

In this respect it is to be noted that the principle described in the second paragraph of point 2.4.2 in decision T 939/92, referred to by the Appellant, stating that a person skilled in the art must be assumed to act not out of idle curiosity but with some specific technical purpose in mind, is not applicable to a "one-way-street" situation, but generally specifies that in assessing inventive step the technical purpose is to be taken into consideration.

3.16 As, thus, neither document (2a) nor document (4) provides any information how R-134a could be obtained in high yields with at the same time low R-1122 levels

and as none of those documents suggests 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 claimed method is not rendered obvious by the cited prior art documents.

3.17 Therefore, the Board comes to the conclusion that the method according to Claim 1 is not obviously derivable from the cited prior art according to Article 54(2) EPC.

Claims 2 to 10, which represent preferred embodiments of Claim 1, derive their lack of obviousness from the same inventive concept.

## **Order**

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

The appeal is dismissed.

The Registrar:

The Chairman:

N. Maslin

A. Nuss