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D E C I S I O N
of 30 October 2001

Case Number: T 0693/97 - 3.3.1

Application Number: 91302699.3

Publication Number: 0449614

IPC: C07C 19/08

Language of the proceedings: EN

Title of invention:

Process for the preparation of 1,1,1,2-tetra-fluoroethane

Patentee:

Ineos Fluor Holdings Limited

Opponent:

AUSIMONT S.p.A.

Headword:

Tetrafluoroethane/INEOS FLUOR HOLDINGS

Relevant legal provisions:

EPC Art. 54, 56, 87(1)

Keyword:

"Priority right (no) - not the same invention"

"Novelty (yes)"

"Inventive step (no) - obvious alternative process"

Decisions cited:

G 0002/98

Headnote/Catchword:

-



Case Number: T 0693/97 - 3.3.1

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 30 October 2001

Appellant:
(Opponent)

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Respondent:
(Proprietor of the patent)

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Decision under appeal:

Decision of the Opposition Division of the
European Patent Office posted 25 April 1997
rejecting the opposition filed against European
patent No. 0 449 614 pursuant to Article 102(2)
EPC.

Composition of the Board:

Chairman: A. J. Nuss
Members: J. M. Jonk
S. C. Perryman

Summary of Facts and Submissions

I. The Appellant (Opponent) lodged an appeal against the decision of the Opposition Division by which the opposition based on the grounds of lack of novelty and lack of inventive step as indicated in Article 100(a) EPC, which had been filed against the European patent No. 0 449 614 (granted in respect of European patent application No. 91 302 699.3) as a whole, was rejected.

II. The opposition was supported by several documents, including:

- (3) GB-A-819 849,
- (4) WO-90/08755,
- (7) US-A-3 752 850,
- (8) GB-A-1 000 485,
- (12) EP-A-0 331 991,
- (13) GB-A-2 030 981, and
- (17) EP-A-0 408 005.

III. Independent Claim 1 as granted read as follows:

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

- (A) contacting a mixture of trichloroethylene and hydrogen fluoride with a fluorination catalyst under superatmospheric pressure of at least 2 bars at a temperature in the range of about 200 to 400°C in a first reaction zone to form a product containing 1,1,1-trifluoro-2-chloroethane and hydrogen chloride together with unreacted starting materials,

- (B) passing the total product of step (A) together with hydrogen fluoride to a second reaction zone containing a fluorination catalyst at a temperature in the range of about 280-450°C but higher than the temperature in step (A) to form a product containing 1,1,1,2-tetrafluoroethane, 1,1,1-trifluoro-2-chloroethane and hydrogen chloride,
- (C) treating the product of step (B) to separate 1,1,1,2-tetrafluoroethane and hydrogen chloride from 1,1,1-trifluoro-2-chloroethane and unreacted hydrogen fluoride,
- (D) feeding the 1,1,1-trifluoro-2-chloroethane mixture obtained from step (C) together with trichloroethylene and 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)."

IV. The Opposition Division held that the subject-matter of the claims was novel and also involved an inventive step.

In this context, it firstly decided that the claimed priority right based on GB 9007029 could not be acknowledged, and that therefore documents (4) and (17) represented state of the art under Article 54(1) and (2) EPC.

Concerning novelty, it considered that the subject-matter as claimed in the present patent was novel, since the process for preparing 1,1,1,2-tetrafluoroethane (R134a) in accordance with document (4) was carried out in a single reaction zone without the need

of the particular pressure and temperature conditions indicated in Claim 1 of the present patent with respect to the reaction steps A and B, and because document (17) did not provide more relevant information than document (4).

Regarding inventive step, it held that document (4) represented the closest prior art. Having regard to this closest prior art, the technical problem underlying the present patent was the provision of a process for preparing R134a, wherein the catalyst lifetime was increased and the reaction temperature had not significantly to be raised to maintain the original conversion rate. Furthermore, it held that the solution of this technical problem as claimed in the present patent was not obvious to the skilled person in the light of the cited documents.

V. Oral proceedings before the Board were held on 30 October 2001.

VI. The Appellant maintained his point of view that the subject-matter of the patent in suit lacked novelty in view of documents (4) and (17). In this context, he emphasised that the process for preparing R134a as disclosed in said documents was actually performed in two reaction zones. Moreover, the temperature in the second reaction zone was inevitably higher than in the first reaction zone because of the heat of reaction developed in the first reaction zone. Furthermore, a superatmospheric pressure could be applied.

Concerning inventive step, he submitted that the technical problem underlying the patent in suit was the provision of a process for preparing R134a with improved yields at increased catalyst lifetime, and that this problem was not solved if the catalyst of document (17) were used. In support of this contention

he referred in particular to the test-report in Annex 3 submitted with the written statement setting out the grounds of appeal dated 26 August 1997. Furthermore, he submitted that the production of R134a in separate reaction steps by preparing 1,1,1-trifluoro-2-chloroethane (R133a) starting from trichloroethylene (TCE) and hydrogen fluoride and by converting said compound R133a with hydrogen fluoride to achieve the desired R134a was well known in the art as indicated in document (4), and that the reaction conditions for these two steps as well as the recycling and separation steps as claimed in the patent in suit were obvious to the skilled person in view of the cited documents.

- VII. The Respondent argued that the claimed priority based on GB 9007029 had to be acknowledged, since the skilled person could directly and unambiguously derive the claimed subject-matter of the patent in suit from said previous application as a whole. This point of view would be in conformity with the Opinion of the Enlarged Board of Appeal dated 31 May 2001 (G 2/98). He concluded that the documents (4) and (17) therefore represented state of the art within the meaning of Article 54(3) and (4) EPC and consequently could not be considered in deciding the issue of inventive step.

Furthermore, he argued that the claimed subject-matter was novel, since the process as claimed in the patent in suit differed essentially from those of documents (4) and (17) in that according to the patent in suit the preparation of R134a was performed in two reaction zones at different temperatures and a particular super-atmospheric pressure as indicated in Claim 1, whereas according to said documents the preparation of R134a was carried out in only one reaction zone at a fixed reaction temperature and without the need of a superatmospheric pressure.

He also submitted that none of the cited documents, alone or in combination, rendered the claimed subject-matter of the patent in suit obvious, since it was surprisingly found that the use of the two-step process as claimed led to an increased catalyst lifetime, which effect was independent from the applied catalyst and therefore led to more flexibility with respect to the choice of the catalyst. In this context, he referred to the declaration of John David Scott submitted on 28 August 2001, which comprised a test-report showing the effects of the claimed process by using a less effective catalyst in order to mimic the effects of catalyst aging and deactivation.

VIII. The Appellant requested that the decision under appeal be set aside and that the European patent No. 0 449 614 be revoked.

The Respondent requested that the appeal be dismissed and that the patent be maintained.

IX. At the conclusion of the oral proceedings the Board's decision was pronounced.

Reasons for the decision

1. The appeal is admissible.

2. *Priority right (Article 87(1) EPC)*

2.1 The Appellant submitted that the subject-matter of Claim 1 of the patent in suit lacked novelty in view of documents (4) and (17). Both documents are, however, intermediate documents having priority dates between the priority date claimed by the patent in suit and the filing date thereof. Therefore, the first question to

be decided by the Board is whether or not Claim 1 of the patent in suit is entitled to the claimed priority.

2.2 Pursuant to Article 87(1) EPC, a right of priority may only be enjoyed in respect of the same invention.

Therefore, in deciding whether Claim 1 of the patent in suit is entitled to the claimed priority, it needs to be decided whether in the priority document GB 9007029 the same invention is disclosed as in present Claim 1 (see also the Opinion of the Enlarged Board of Appeal G 2/98, OJ EPO 2001, 413, in particular point 9 of the reasons).

2.3 The priority document aims at providing a process for preparing R134a in improved yields and with a reduced level of toxic unsaturated by-products (see page 1, lines 21 to 23 and page 5, lines 4 to 12).

In order to achieve these objects, the priority document teaches on page 1, line 24 to page 2, line 20, the preparation of R134a by a method which comprises the steps of:

- (A) contacting a mixture of R133a and hydrogen fluoride with a fluorination catalyst 300 to 450°C in a first reaction zone to form a product containing R134a and hydrogen chloride together with unreacted starting materials,
- (B) passing the total product of step (A) together with TCE to a second reaction zone containing a fluorination catalyst at 200-400°C to form a product containing R133a, R134a and hydrogen chloride,

- (C) treating the product of step (B) to separate R133a and unreacted hydrogen fluoride from a mixture containing R134a and hydrogen chloride,
- (D) feeding the R133a obtained from step (C) together with hydrogen fluoride to said first reaction zone (step (A)), and
- (E) recovering R134a from the R134a/hydrogen chloride mixture obtained from step (C).

2.4 It follows from this disclosure that the process for preparing R134a as claimed in Claim 1 of the patent in suit essentially differs from the process disclosed in the priority document in that the reaction steps A and B as indicated in the priority document are reversed, and in that in the patent in suit the TCE starting compound is introduced in the first reaction zone (step A), whereas according to the priority document this starting compound is introduced in the second reaction zone (step B). In these circumstances, and in view of the strict interpretation of the concept of "the same invention" equating the concept of "the same subject-matter" in said Opinion of the Enlarged Board of Appeal G 2/98 (see point 9 of the reasons), the Board concludes that the invention as defined in Claim 1 of the patent in suit is not the same as that disclosed the priority document.

2.5 The Appellant's submission that the subject-matter of Claim 1 of the patent in suit can directly and unambiguously be derived from the comparative example in the priority document (see page 4, lines 17 to 31) already fails in view of the fact that the use of a pressure of at least 2 bars in step A representing a mandatory feature of the process of Claim 1 of the patent in suit can neither be derived from this example nor from the description of the priority document.

Moreover, the Board observes that the process for preparing R134a comprising the steps A to E specified under point 2.3 above is disclosed in the priority document as the sole invention, and that therefore the ~~subject-matter of the comparative example cannot be~~ considered as another invention upon which a priority right could be based.

2.6 For these reasons, in the Board's judgment, Claim 1 of the patent in suit is not entitled to the claimed priority right.

2.7 Furthermore, the Board observes that in view of the fact that the claimed priority cannot be acknowledged the effective date of the subject-matter of the patent in suit is the filing date of the corresponding patent application, and that therefore documents (4) and (17) represent state of the art to be considered under Articles 54(1)(2) and 56 EPC.

3. *Novelty*

3.1 The next issue to be dealt with is whether the subject-matter of Claim 1 of the patent in suit is novel in view of documents (4) and (17).

3.2 Document (4) discloses an improved process for preparing R134a by reacting HF with TCE and/or R133a in the presence of a catalyst at an elevated temperature of 300 to 500°C, most preferably 370 to 410°C, in which the improvement resides in conducting the reaction in a single reaction zone (see page 5, line 1 to page 6, line 7 and page 9, lines 3 to 9). According to all the examples only fixed reaction temperatures adjusted by means of a sand bath are used (see concerning the temperature adjustment page 10, lines 26 to 29 and

page 11, first paragraph). Moreover, it discloses that pressure is not critical, and that therefore atmospheric and superatmospheric pressure are convenient (see page 10, lines 14 to 16).

3.3 Document (17) discloses a process for preparing R134a at a high selectivity and with industrially acceptable conversions by reacting a mixture of TCE and R133a in TCE/R133a molar ratios of from 5/95 to 50/50, preferably of about 15/85, with HF in the presence of a catalyst comprising Cr_2O_3 carried on aluminium trifluoride (see page 2, lines 39 to 43). It also discloses the use of a single reactor using a fixed temperature of preferably of from 300 to 400°C, particularly of from 330 to 380°C, at atmospheric pressure or at higher pressures up to 15 atmospheres (see page 2, last two paragraphs, and the examples).

3.4 The Appellant's novelty objection was essentially based on his contention that the process disclosed in both document (4) and (17) was actually performed in two reaction zones, since the temperature in the second reaction zone was inevitably higher than in the first reaction zone because of the heat of reaction developed in the first reaction zone. In this context he only relied on thermodynamic data indicated in Annex 1 as submitted by the Appellant with his Statement of Grounds of Appeal showing that the reaction of TCE with HF providing the intermediate compound R133a was highly exothermic and that the conversion of said intermediate compound to R134a was slightly exothermic.

However, this contention is in contradiction with the facts, since - as indicated above - both documents only disclose the use of a fixed reaction temperature in a single reactor. Moreover, in the Board's judgment, the reaction temperature strongly depends on (i) the nature of the reactor, (ii) the means for adjusting the

reaction temperature therein, and (iii) the reaction conditions including the molar ratios of the reactants, the catalyst and the contact time. Consequently, the existence of two reaction zones in the reactor, in which the temperature in the second zone is higher than in the first one, could only be proved by a precise reproduction of one of the examples. Therefore, in the absence of such proof, the Appellant's submission in this respect cannot be accepted by the Board.

3.5 In these circumstances, the Board concludes that the claimed subject-matter is novel, since documents (4) and (17) do not directly and unambiguously disclose as a technical teaching the production of R134a in two reaction zones, let alone the use therein of the temperature conditions as defined in Claim 1 and the application of a pressure of at least 2 bars in the first reaction zone.

4. *Inventive step*

4.1 The remaining issue to be dealt with is whether the subject-matter of the claims as granted involves an inventive step.

4.2 For deciding whether or not a claimed invention meets this criterion, the Boards of Appeal consistently apply the problem and solution approach, which involves essentially identifying the closest prior art, determining in the light thereof the technical problem which the claimed invention addresses and successfully solves, and examining whether or not the claimed solution to this problem is obvious for the skilled person in view of the state of the art.

According to the established jurisprudence of the Boards of Appeal, the closest prior art is normally a prior document disclosing subject-matter conceived for

the same purpose as the claimed invention and having the most relevant technical features in common.

Furthermore, if the technical results of the claimed invention provide some improvement over the closest prior art, the problem can be seen as providing such improvement, provided this improvement necessarily results from the claimed features for all that is claimed. If, however, there is no improvement, but the means of implementation are different, the technical problem can be defined as the provision of an alternative to the closest prior art.

- 4.3 The Board considers, in line with the above and in agreement with the parties, that the closest state of the art with respect to the claimed subject-matter of the patent in suit is the disclosure of document (17).

This document relates to a process for preparing R134a which is conceived for the same purpose as the claimed invention, i.e. to overcome the problem of a rapid decrease of the catalyst activity (see page 2, lines 33 to 35 and 44 to 46, as well as the patent in suit, page 2, lines 14 to 29). Moreover, the essential features of this prior art process, i.e. the use of a feed into the reactor comprising a mixture of TCE and R133a in a molar ratio of 5/95 to 50/50 and the application of the defined catalyst, fall under the scope of Claim 1 of the patent in suit. In this context, the Board notes that Example 1 of the patent in suit concerns a process in which, in the steady state of the process, the feed to the first reaction zone contains 15 mol% of TCE based on the organics which substantially consist of TCE and recycled R133a.

- 4.4 Regarding this closest state of the art, the Respondent contended essentially that the process as claimed led to an increased catalyst lifetime, which effect was

independant from the applied catalyst and therefore led to more flexibility with respect to the choice of the catalyst. In support, he referred to the test-report provided by John David Scott showing the effects of the claimed process.

On the other hand, the Appellant disputed this contention. In this context, he referred to the outstanding lifetime of the catalyst of document (17) amounting to periods of time of the order of hundreds of hours, and to Test II of the test-report in Annex 3 submitted with his statement setting out the grounds of appeal. This Test II demonstrated that by reacting a mixture of HF, R133a and TCE having a molar ratio HF/(R133a+TCE) of 3.7 and a molar ratio HF/TCE of 6.4 in the presence of a catalyst prepared according to Example 1 of document (17) at a temperature of 365°C and a pressure of about 1.2 bar higher selectivities and a lower content of by-products compared with Example 1 of the patent in suit were achieved and that, in contradiction to the comparative example of the patent in suit in which the process has been carried out at atmospheric pressure instead of at least 2 bars (actually 13.5 bars) as claimed (see under Example 1 and Table 1), after 24 hours and also after 50 hours the activity of the catalyst remained essentially the same.

The Respondent neither disputed the technical information given in document (17) nor the results of said Test II. He submitted in this respect that according to Test II a high activity catalyst was used, which evidently had not reached the stage of beginning to suffer deactivation over the test period, i.e. under circumstances in which the benefit of the claimed process was masked by the catalyst performance. On the other hand, the claimed invention of the patent in suit concerned the problem of catalyst deactivation

inevitably occurring in a continuously operated process. Thus, in order to show the effect of the claimed process under circumstances where the catalyst activity diminishes, some experiments were carried out under simulated deactivation conditions, namely by using a catalyst having a lower activity than the catalyst tested by the Appellant in said Test II, and the results of these experiments were enclosed in the affidavit of John David Scott.

The catalyst used in these experiments was prepared by impregnating an alumina, which was crushed and sieved to generate particles of about 1 mm diameter, with a chromium chloride solution to form an alumina based fluorination catalyst containing 5% by weight of chromium. The catalyst was then dried in nitrogen at 250°C.

On the other hand, the catalyst of document (17), which was tested by the Appellant in Test II, was prepared as indicated in Example 1 by impregnating a carrier consisting of AlF_3 , prevailing in the gamma form with a chromium chloride solution to obtain a catalyst containing 10% by weight of chromium, whereby the chromium chloride solution was added in three almost equal portions and after each addition the catalyst was dried for 4 hours at 120°C. Thereafter, the catalyst was fluidised with a nitrogen stream for 10 hours at 400°C.

Hence, the Respondent's experimental report enclosed in the affidavit of John David Scott making use of a catalyst, which differs from the catalyst of document (17) with respect to its chromium content and its support, and which has been prepared in a quite different way, does not reflect the closest state of the art represented by document (17). Moreover, it follows from the experimental report that the tests

have been performed without any aging of the catalyst, and in fact only show the effect of the different reaction conditions in the presence of a catalyst having a low activity.

Therefore, the Board does not consider it credible that the improvement as alleged by the Respondent, namely the achievement of an increased catalyst lifetime independently from the applied catalyst, could be obtained by substantially all the embodiments encompassed within the scope of Claim 1 of the patent in suit.

- 4.5 Thus, in view of these considerations, the Board finds that in the light of the closest state of the art the technical problem underlying the patent in suit can only be seen in the provision of a further process for preparing R134a resulting in a long catalyst life.
- 4.6 According to present Claim 1 this technical problem is solved by providing a process in which the reaction is carried out in two reaction zones (which may consist in two reactors as indicated on page 3, line 46, and in Example 1 of the patent in suit) in which the temperature in the second zone is higher than in the first one, and the pressure in the first reaction zone is at least 2 bars.
- 4.7 Having regard to the technical information provided in the patent in suit, the Board considers it plausible that the technical problem as defined above has been solved. This was also not disputed by the Appellant.
- 4.8 In assessing inventive step the question thus is whether a skilled person starting from document (17) and having knowledge of the other cited documents would arrive at the solution of the above defined technical problem as claimed.

4.9 In this context, the Board notes that, as submitted by the Appellant, it was well known in the prior art that the catalysed reaction of HF with TCE to achieve R134a is a sequential reaction, in which at first the intermediate R133a is produced in an almost quantitative yield and in the second reaction R133a is converted to the final product R134a. This was never denied by the Respondent and follows, for instance, from the discussion of the prior art in document (4) (see page 4, line 29 to page 5, line 7).

Concerning the reaction of HF with TCE for preparing the intermediate compound R133a said document (4) refers to documents (8) and (7) (see page 2, lines 14 to 36), whereas the Appellant, in addition to these two documents, also mentioned document (3). From these documents it can be derived that, using different catalysts, essentially quantitative conversions of TCE and yields of R133a of about 85 to 95% on the basis of the converted TCE can be obtained (see document (3), page 1, lines 54 to 69, and Example 1; document (7), column 1, lines 59 to 67, and Examples 1, 3 and 8; and document (8), page 2, lines 28 to 35, and Example 1, tests 1 to 5 indicated in Table 1). Moreover, it can be derived from the examples in these documents that the preferred reaction temperatures are between 250°C and 360°C (see the examples mentioned above, in particular Example 8 in document (7) and Example 1, test 3, in document (8)). Furthermore, it is indicated in document (3) that by increasing the temperature, the amount of fluorination can be increased.

With respect to the fluorination of R133a to obtain R134a document (4) refers to document (13) (see page 3, line 25 to page 4, line 4), whereas the appellant, in addition, also mentioned document (12). From these documents it follows that this reaction, independently of the catalysts used therein, is preferably conducted

at temperatures of about 350 to 450°C, and gives good conversions of R133a to R134a at small amounts of impurities rendering it possible to recycle the unreacted R133a and to achieve a high selectivity regarding R134a (see document (12), page 3, lines 56 to 58, and the examples, in particular those relating to reaction temperatures of at least 400°C; and document (13), page 1, line 57 to page 2, line 3, and the examples, all using a reaction temperature of 400°C).

Therefore, the Board finds that it can be derived from this prior art, that the preparation of R134a, starting from TCE and involving the forming of the intermediate compound R133a, can be carried out in two reaction zones (or reactors), and that in doing so optimum results regarding the selectivity to R133a in the first step and to R134a in the second step can be achieved at temperature conditions corresponding to those indicated in Claim 1 of the patent in suit, i.e. at a temperature about 200 to 400°C in a first reaction zone (or reactor) to form the intermediate product R133a, and at a higher temperature of about 280 to 450°C in the second reaction zone to convert R133a to the desired R134a.

Furthermore, documents (17) and (4), both disclose that the one-pot reaction of TCE with HF to R134a, which - as follows from the considerations above - involves a sequential reaction including the conversion of TCE to the intermediate compound R133a, can be carried out at superatmospheric pressures (see document (17), page 2, last two lines, indicating higher pressures up to about 15 bars; and document (4), page 10, lines 14 to 16). In addition, document (7), which has been discussed above in relation to the conversion of TCE with HF to form the intermediate compound R133a, explicitly discloses the possibility to apply superatmospheric pressures in performing this conversion (see column 3, lines 17 to 21). Therefore, it follows from these documents that

the conversion of TCE to R133a corresponding to step (A) of Claim 1 of the patent in suit can be performed at superatmospheric pressures such as 2 bars or higher. In any case, in the Board's judgment, it would be obvious to the skilled person in the light of the cited documents to find an optimum pressure suitable to the other reaction parameters, such as the catalyst, the reaction temperature and the HF/TCE ratio, which finding can therefore be carried out without any inventive skill.

4.10 Thus in view of these considerations, the Board concludes that the solution of the above defined technical problem as claimed in Claim 1 of the patent in suit does not involve an inventive step in the sense of Article 56 EPC.


The further claims fall with Claim 1, since the Board can only decide on the Appellant's request as a whole.

Order

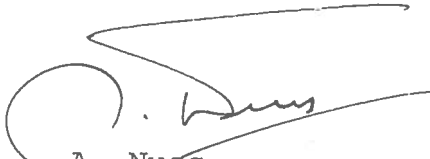
For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The patent is revoked.

The Registrar:


N. Maslin

The Chairman:


A. Nuss

