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Datasheet for the decision of 8 January 2019

Case Number: T 0853/15 - 3.3.10

Application Number: 07754686.9

Publication Number: 2001828

IPC: C07C17/25, C07C21/18, C07C17/38, C07C17/383

Language of the proceedings: EN

Title of invention:
COPRODUCTION OF HYDROFLUOROOLEFINS

Patent Proprietor:
The Chemours Company FC, LLC

Opponent:
Arkema France

Headword:
COPRODUCTION OF HYDROFLUOROOLEFINS/ The Chemours Company

Relevant legal provisions:
EPC Art. 56

Keyword:
Inventive step - (no)

Decisions cited:

Catchword:

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Beschwerdekammern

Boards of Appeal

Chambres de recours

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Case Number: T 0853/15 - 3.3.10

D E C I S I O N
of Technical Board of Appeal 3.3.10
of 8 January 2019

Appellant: (Opponent)	Arkema France DRD/Département Propriété Industrielle 420, rue d'Estienne d'Orves 92705 Colombes Cedex (FR)
Representative:	Dang, Doris ARKEMA France Département Propriété Industrielle 420, rue d'Estienne d'Orves 92705 Colombes Cedex (FR)
Respondent: (Patent Proprietor)	The Chemours Company FC, LLC 1007 Market Street Wilmington DE 19801 (US)
Representative:	Heinemann, Monica Abitz & Partner Patentanwälte mbB Arabellastraße 17 81925 München (DE)
Decision under appeal:	**Decision of the Opposition Division of the European Patent Office posted on 2 March 2015 rejecting the opposition filed against European patent No. 2001828 pursuant to Article 101(2) EPC.**

Composition of the Board:

Chairman	P. Gryczka
Members:	T. Bokor
	J.-C. Schmid

Summary of Facts and Submissions

I. The appellant (opponent) lodged an appeal against the decision of the Opposition Division rejecting the opposition against European patent No. 2 001 828, independent claim 1 reading as follows:

"1. A process for the co-manufacture of 1,2,3,3,3-pentafluoropropene and 2,3,3,3-tetrafluoropropene comprising:

dehydrofluorinating a blend of 1,1,1,2,3,3-hexafluoropropane and 1,1,1,2,3-pentafluoropropane in a reaction zone having a catalyst, thereby forming a product mixture comprising said 1,2,3,3,3-pentafluoropropene and 2,3,3,3-tetrafluoropropene, unreacted hydrofluorocarbons and hydrogen fluoride, and separating said blend of 1,2,3,3,3-pentafluoropropene and 2,3,3,3-tetrafluoropropene from hydrogen fluoride and said unreacted hydrofluorocarbons to produce 1,2,3,3,3-pentafluoropropene and 2,3,3,3-tetrafluroropropene.

II. Notice of opposition had been filed by the appellant requesting revocation of the patent-in-suit in its entirety on the grounds of lack of inventive step (Articles 100(a) and 56 EPC) and insufficient disclosure of the invention (Article 100(b) EPC).

Inter alia, the following documents were cited in the opposition proceedings:

(1) US-A-5 396 000,
(4) US-A-6 031 141 and

(5) Knunyants I. L., Krasuskaya, M. P., Mysov E.I., "REACTIONS OF FLUORO OLEFINS Communication 13. Catalytic hydrogenation of perfluoro olefins", Bulletin of the Academy of Sciences of the USSR, Division of Chemical Sciences, 1960, Volume 9, Issue 8, pages 1312–1317.

According to the Opposition Division, the adjustment of parameters like temperature and reaction time were routine tasks for the skilled person, especially because the patent contained concrete examples to serve as guidance. Hence, the patent-in-suit met the requirement of sufficiency of disclosure. With respect to inventive step, document (4) represented the closest prior art to the invention. The subject-matter of claim 1 of the patent-in-suit differed from the process disclosed in document (4) in that the conversion of 1,1,1,2,3,3 hexafluoropropane (HFC-236ea) was operated together with another generically disclosed hydrofluorocarbons (namely 1,1,1,2,3- pentafluoro-propane (HFC-245eb) so that a mixture of 1,2,3,3,3-pentafluoropropene (HFC-1225ye) and 2,3,3,3-tetra-fluoropropene (HFC-1234yf) was obtained. The objective technical problem was to provide a method allowing the direct production of a mixture containing both HFC-1225ye and HFC-1234yf by means of a catalyst promoted reaction from a suitable corresponding blend of starting materials. Although encompassed by the generic reaction of compounds of formula $(R^1)_2CH-CF(R^1)_2$, the conversion of HFC-245eb to HFC-1234yf was not disclosed in document (4). It was general knowledge that the dehydrofluorination reactions could be carried out on mixtures to lead to mixtures of corresponding olefins, provided that the reaction conditions of the two distinct starting materials were not too different. However, document (4) did not teach that HFC-245eb was

converted to HFC-1234yf, and that the conditions required for the dehydrofluorination of HFC-236ea were similar enough to those for the dehydrofluorination of HFC-245eb to allow the dehydrofluorination of both in mixture. Hence, the skilled person aiming at a method in which a blend of HFC-1225ye and HFC-1234yf was directly obtained from a blend comprising the corresponding starting materials did not find in the prior art sufficient indications as to the feasibility of presently claimed solution, i.e. the dehydrofluorination of a blend of HFC-236ea and HFC-245eb with the same catalyst at a certain temperature. Consequently, the subject-matter of claim 1 involved an inventive step in the sense of Article 56 EPC.

III. During the oral proceedings held on 8 January 2019 before the Board, the respondent (patent proprietor) defended the maintenance of the patent on the basis of the main request (patent as granted) and on the basis of the first to ninth auxiliary requests filed with letter dated 7 November 2018.

The process of claim 1 of the first auxiliary request differs from the process of claim 1 of the patent as granted in that it is carried out at a temperature of from 200°C to 500°C.

The process of claim 1 of the second auxiliary request differs from the process of claim 1 of the patent as granted in that it is carried out at a temperature of from 300°C to 450°C.

Claim 1 of the third auxiliary request differs from claim 1 of the main request in that the catalyst is selected from aluminum fluoride; fluorided alumina;

metals on aluminum fluoride; metals on fluorided alumina; oxides, fluorides, and oxyfluorides of magnesium, zinc and mixtures of magnesium and zinc and/or aluminum; lanthanum oxide and fluorided lanthanum oxide; chromium oxides, fluorided chromium oxides, and cubic chromium trifluoride; carbon, acid-washed carbon, activated carbon, three dimensional matrix carbonaceous materials; and metal compounds supported on carbon, wherein the metal compounds are oxides, fluorides, and oxyfluorides of at least one metal selected from the group consisting of sodium, potassium, rubidium, cesium, yttrium, lanthanum, cerium, praseodymium, neodymium, samarium, chromium, iron, cobalt, rhodium, nickel, copper, zinc, and mixtures thereof.

Claim 1 of the fourth auxiliary request differs from claim 1 of the patent as granted in that the catalyst is selected from the group consisting of aluminum fluoride, fluorided alumina and metals on fluorided alumina.

The process of claim 1 of the fifth auxiliary request differs from the process of claim 1 of the patent as granted in that it further comprises recirculating the unreacted hydrofluorocarbons back to the reaction zone.

The process of claim 1 of the sixth auxiliary request differs from the process of claim 1 of the third auxiliary request in that it is carried out at a temperature of from 200°C to 500°C.

The process of claim 1 of the seventh auxiliary request differs from the process of claim 1 of the fourth auxiliary request in that it is carried out at a temperature of from 300°C to 450°C.

The process of claim 1 of the eighth auxiliary request differs from the process of claim 1 of patent as granted in that the mole ratio of 1,1,1,2,3,3-hexafluoropropane and 1,1,1,2,3 pentafluoropropane fed to the reaction zone is from 10:1 to 80:1.

The process of claim 1 of the ninth auxiliary request differs from the process of claim 1 of patent as granted in that the process is carried out in the presence of an inert gas.

IV. According to the appellant, with regard to inventive step, document (4) represented the closest prior to the invention. This document disclosed catalytic dehydrofluorination of hydrofluorocarbon of the formula $(R^1)_2CH-CF(R^1)_2$ to produce fluoroolefins of the formula $(R^1)_2C=C(R^1)_2$. According to this generic teaching, dehydrofluorination of HFC-245eb provided HFC-1234yf and HFC-1234ze (1,3,3,3,3-tetrafluoropropene). Looking at the examples disclosed in document (4), the skilled person would understand that HFC-245eb would mainly produce HFC-1234yf. Furthermore, claim 1 did not exclude that some HFC-1234ze was produced. Document (5) taught dehydrofluorination of HFC-236ea to HFC-1225ye and HFC-245eb to HFC-1234yf in the presence of a base. As catalytic dehydrofluorination of HFC-236ea produced HFC-1225ye as shown in example 3, the skilled person would have expected that by analogy catalytic dehydrofluorination of HFC-245eb would produce HFC-1234yf. Therefore, in the light of document (4), the skilled person had a reasonable expectation of success to produce a blend of HFC-1225ye and HFC-1234yf by catalytic dehydrofluorination of a starting mixture of HFC-236ea and HFC-245eb. Thus, the subject-matter of claim 1 of the main request did not involve an inventive step. The additional characteristics present

in the auxiliary requests were common in the field of dehydrofluorination. Auxiliary request 4 specified the nature of the catalyst, which included aluminium fluoride. This catalyst was disclosed in document (1) for dehydrofluorination of HFC-236ea to HFC-1225ye. Thus, the subject-matter of auxiliary requests 1 to 9 did not involve an inventive step either.

V. According to the respondent, document (4) represented the closest prior art to the invention. The technical problem to be solved was to provide a process for the manufacture of a blend comprising as mandatory components HFC 1225ye and HFC 1234yf. The proposed solution was the process of claim 1 which was characterized by the dehydrofluorination of a blend of hydrofluorocarbons and that the hydrofluorocarbon blend comprised HFC-245eb. It was not obvious from document (4) that catalytic dehydrofluorination of HFC-245eb produced HFC-1234yf. Document (4) only foresaw starting from a single hydrofluorocarbon compound. Hydrofluorocarbons did not react in the same manner under the same reaction conditions. It was therefore not obvious to carry out the dehydrofluorination on a mixture of two hydrofluorocarbons with the same reaction conditions. The appellant's argumentation was based on hindsight. The subject-matter of claim 1 of the patent as granted involved therefore an inventive step. Claim 1 of the fourth auxiliary request was restricted to a specific catalyst, which was not disclosed in document (4). The subject-matter of claim 1 of the fifth auxiliary request involved an inventive step, since document (4) did not teach to recycle a blend of hydrofluorocarbons. The use of an inert gas provided better conversion. Therefore the subject-matter of claim 1 of the ninth auxiliary request also involved an inventive step.

VI. The appellant requested that the decision under appeal be set aside and the patent be revoked.

The respondent requested that the appeal be dismissed, or subsidiarily, that the patent be maintained on the basis of any of the first to ninth auxiliary requests filed with the letter dated 7 November 2018.

VII. At the end of the oral proceedings the decision of the Board was announced.

Reasons for the Decision

1. The appeal is admissible.

Inventive step

Main request: claim 1 of the patent as granted

2. Closest prior art

The patent-in-suit relates to the field of production and purification of fluoroolefin compounds, and more particularly to a method for the co-manufacture of 1,2,3,3,3-pentafluoropropene (HFC-1225ye) and 2,3,3,3-tetrafluoropropene (HFC-1234yf) via catalytic dehydrofluorination of 1,1,1,2,3,3-hexafluoropropane (HFC-236ea) and 1,1,1,2,3-pentafluoropropane (HFC-245eb) (see page 1, lines 7 to 11; claim 1).

Document (4) relates to a process for the production of fluoroolefins and more particularly to catalytic dehydrofluorination of hydrofluorocarbons to produce fluoroolefins (see column 1, lines 7 to 12).

Thus, document (4) discloses a process for the manufacture of fluoroolefins of the formula $(R^1)_2C=C(R^1)_2$ via catalytic dehydrofluorination of hydrofluorocarbons of the formula $(R^1)_2CH-CF(R^1)_2$, wherein each R^1 is independently selected from the group consisting of H, F, CF_3, CHF_2, CH_2F, C_2F_5, C_2HF_4 and $C_2H_2F_3$, provided that at least one R^1 is not H (see claim 1).

The process of producing of HFC-1225ye by catalytic dehydrofluorination of HFC-236ea is disclosed in example 3 (one R^1 group is CF_3, two R^1 groups are F, and one R^1 group is hydrogen) –see column 6, line 56 to column 7, line 8.

The process of producing of HFC-1234yf by catalytic dehydrofluorination of HFC-245eb is encompassed by the process disclosed in document (4) (one R^1 group is CF_3, one R^1 groups is F and two R^1 groups are hydrogen).

Hence, the process of claim 1 of the patent-in-suit differs from the process of document (4) in that two fluorocarbons fluoroolefins are simultaneously dehydrofluorinated and that it comprises specifically the catalytic dehydrofluorination of HFC-245eb and the production of HFC-1234yf.

The Board considers, in agreement with the parties, that document (4) represents the closest prior art to the invention.

3. Technical problem underlying the invention

According to the respondent, the technical problem to be solved was to provide a process for the manufacture

of a blend comprising as mandatory components HFC-1225ye and HFC-1234yf.

4. *Solution*

The proposed solution is the process of claim 1 which is characterized by the dehydrofluorination of a blend of fluorocarbons and that the fluorocarbon blend comprises HFC-245eb.

5. *Success*

It is shown in the examples of the patent that a blend comprising as mandatory components HFC-1225ye and HFC-1234yf can be obtained by dehydrofluorinating a blend of fluorocarbons comprising HFC-236ea and HFC-245eb. This finding was not contested by the appellant. The Board therefore is satisfied that the process of claim 1 of the patent as granted provides a solution to the technical problem as defined above.

6. *Obviousness*

Document (4) teaches that the two hydrofluoroolefins comprised in the blend to be prepared are obtained by the same process, i.e. by dehydrofluorination of hydrofluorocarbons of formula $(R^1)_2CH\text{-}CF(R^1)_2$ in the presence of a catalyst at temperatures of from 200° to 500°C.

The skilled man faced with the problem of providing a blend of two hydrofluoroolefins of formula $(R^1)_2C{=}C(R^1)_2$ has two obvious alternatives, either preparing separately the two hydrofluoroolefins and blending them thereafter, or else preparing concomitantly the two fluoroolefins, for example by the

process of document (4) starting from a mixture of the corresponding hydrofluorocarbons of formula $(R^1)_2CH-CF(R^1)_2$.

The claimed process therefore follows one of the two obvious options that the skilled has at his disposal to solve the technical problem of providing a process for the manufacture of a blend comprising HFC-1225ye and HFC-1234yf. Once he has chosen to start from a mixture, the step of separating out the unreacted components is an obvious step in practically any process. Consequently, the skilled person facing the problem of providing a process for preparing the blend of HFC-1234yf and FC1225ye would arrive at the subject-matter of claim 1 of the patent as granted without the exercise of inventive skill.

According to the respondent the formation of the desired isomer HFC-1234yf from HFC-245eb was not predictable, since document (4) did not indicate which isomer is obtained by catalytic dehydrofluorination of HFC-245eb.

However, claim 1 does not require that only one isomer is produced from HFC-245eb. Furthermore, the skilled person does not need an explicit teaching from document (4) to recognise from its structure that HFC-245eb is the most promising hydrofluorocarbon to produce HFC-1234yf by catalytic dehydrofluorination. The catalytic dehydrofluorination of HFC-236ea predominantly provides HFC-1225ye as described in example 3 of document (4). Thus, the skilled person identifies from that reaction that it is the fluoride, which is not vicinal to the trifluoromethyl group, which is preferably leaving. Per analogy, the skilled person will consider that HFC-245eb mainly produces

HFC-1234yf, rather than HFC-1234ze by catalytic dehydrofluorination.

In addition, the dehydrofluorination of HFC-236ea in the presence of base or a catalyst provides HFC-1225ye (document (5), page 1316, second paragraph, document (4), example 3). As the dehydrofluorination of HFC-245eb in the presence of alkali hydroxide provides HFC-1234yf (see document (5), page 1316, fourth paragraph), the skilled person has good reasons to expect, by analogy, that catalytic dehydrofluorination of HFC-245eb will produce HFC-1234yf. It is not necessary that the success of an envisaged solution of a technical problem was certain. In order to render a solution obvious it is sufficient that the skilled person would have followed the teaching of the prior art with a reasonable expectation of success. Hence, the Board cannot agree with the respondent's argument that due to some purported uncertainty as to the isomer formed, the skilled person would not have considered catalytic dehydrofluorination of HFC-245eb to produce HFC-1234yf. The skilled person has a clear incentive from document (4) to do so, it was only necessary for him to confirm it experimentally by routine work.

According to the respondent, the allegedly obvious consideration of the skilled person to take a blend of two hydrofluorocarbons of formula $(R^1)_2CH-CF(R^1)_2$ as starting material for the process of document (4) was based on hindsight, since this document only foresaw starting from a single hydrofluorocarbon.

However, operating processes by using mixtures as starting products falls within the routine practice of the skilled person, all the more when the reactants are close analogues which are disclosed to be

dehydrofluorinated under the same reaction conditions. In the present case, the skilled person is faced with the problem of providing a blend of two products which are disclosed to be obtainable under the same reaction conditions. Accordingly, starting with a blend of reactants is not based on hindsight, but is an obvious alternative for the skilled person to provide a blend of reaction products.

According to the respondent, two hydrofluorocarbons did not react in the same manner under the same reaction conditions. It could therefore not be expected that the two compounds undergo a dehydrofluorination reaction under the same reaction conditions.

However, claim 1 only requires that HFC-1225ye and HFC-1234yf are produced by the process. Document (4) indicates that the catalytic dehydrofluorination of the hydrofluorocarbons of formula $(R^1)_2CH\text{-}CF(R^1)_2$ commonly occurs at a temperature from 200°C to 500°C. Claim 1 does not require any specific reaction conditions. This argument of the respondent should therefore be rejected.

Hence, the subject-matter of claim 1 of the main request does not involve an inventive step (Article 56 EPC).

Auxiliary requests

7. Claim 1 of the first auxiliary request requires that the process is carried out at a temperature "of from 200°C to 500°C", i.e. at a temperature between 200°C to 500°C. However, this feature is known from the closest prior art document (4) which discloses that the catalytic dehydrofluorination of the hydrofluorocarbons

of formula $(R^1)_2CH-CF(R^1)_2$ takes places at exactly the same temperatures of from 200°C to 500°C (see claim 1; column 3, line 46 to 49).

8. Claim 1 of the second auxiliary request requires that the process is carried out at a temperature of from 300°C to 450°C. Document (4) discloses that the catalytic dehydrofluorination of the hydrofluorocarbons of formula $(R^1)_2CH-CF(R^1)_2$ is preferably conducted at temperatures of from 225 °C to 400°C (see column 3, line 46 to 49). Consequently, merely specifying that the process is carried out at a temperature of from 300°C to 450°C does not add any inventiveness to the claimed process.

9. Claim 1 of third auxiliary request requires that the process is carried out with specific catalysts, including cubic chromium trifluoride, which is the catalyst used in the process of document (4). Thus, this modification does not add anything new or unexpected with respect the closest prior art.

10. Claim 1 of the fourth auxiliary request requires that the catalyst is selected from the group consisting of aluminium fluoride, fluorine alumina and metals on fluorine alumina. These catalysts are usually used in dehydrofluorination of hydrofluorocarbons (see document (1), claim 1, examples 1 and 2). Furthermore, document (4) discloses that the cubic chromium trifluoride may be supported on fluorinated alumina and that it may be used in mixture with aluminium fluoride or fluorinated alumina (see column 2, line 29 to 35).

Consequently, the limitation to these specified catalysts does not impart any inventiveness to the claimed process.

11. Claim 1 of the fifth auxiliary request comprises recirculating the unreacted hydrofluorocarbons back to the reaction zone. Document (4) discloses that unreacted starting hydrofluorocarbons can be recycled to the reactor for the production of additional fluoroolefins (see column 3, lines 60 to 64).

According to the respondent, document (4) did not teach to recycle a blend of hydrofluorocarbons. However, as the starting material is a hydrofluorocarbons blend, it is obvious that the unreacted material to be recycled is the (starting) hydrofluorocarbons blend. Accordingly, this argument cannot convince the Board.

12. Claim 1 of the sixth auxiliary request requires that the process is carried out at a temperature of from 200°C to 500°C and the catalyst is selected from catalyst including cubic chromium trifluoride.

These features are disclosed in combination in claim 1 of document (4). Consequently, this further requirement cannot add any inventiveness to the claimed process.

13. Claim 1 of the seventh auxiliary request compared to claim 1 of the fourth auxiliary request requires that process is carried out at a temperature of from 200°C to 500°C. The process disclosed in document (4) is carried out at temperatures of from 200°C to 500°C. Accordingly, the addition of this feature cannot contribute to the inventiveness of the subject-matter of claim 1.

14. Claim 1 of the eighth auxiliary request requires that the ratio of HFC-236ea and HFC-245eb fed to the

reaction is from 10:1 to 80:1, i.e. in a relatively broad range.

No effect has been shown to be associated with the claimed particular ratio of HFC-236ea and HFC-245eb. It represents therefore an arbitrary choice and thus cannot impart any inventiveness to the claimed subject-matter.

15. Claim 1 of the ninth request requires that the process is carried out in the presence of an inert gas. The respondent alleged that the use of an inert gas provides better conversion. Notwithstanding that this allegation has not been corroborated, document (4) on column 3, lines 57 to 59 discloses that the reaction is done in the presence of inert gases, such as nitrogen and argon, with the consequence that this additional feature does not distinguish the solution with respect to the closest prior document and thus cannot contribute to inventive step of the claimed subject-matter.

Hence, the ninth auxiliary request also must fail.

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:

C. Rodríguez Rodríguez P. Gryczka

Decision electronically authenticated