Datasheet for the decision of 5 March 2020

Case Number: T 0285/16 - 3.3.10

Application Number: 09155177.0

Publication Number: 2100867

IPC: C07C17/23, C07C17/354, C07C17/38, C07C17/25, C07C21/18

Language of the proceedings: EN

Title of invention:
Process of co-producing HFC-1225YE and HFC-1234YF via 4-step reactions

Patent Proprietor:
Honeywell International Inc.

Opponents:
ARKEMA FRANCE
Mexichem Amanco Holding S.A. de C.V.

Headword:
Process of co-producing HFC-1225YE and HFC-1234YF / Honeywell

Relevant legal provisions:
EPC Art. 54(3), 56, 100(a), 100(b), 123(2), 123(3)
Keyword:
Amendments - extension beyond the content of the application as filed (no) - broadening of claim (no)
Grounds for opposition - insufficiency of disclosure (no)
Novelty - (yes)
Inventive step - (yes)

Decisions cited:

Catchword:
Case Number: T 0285/16 – 3.3.10

DECISION
of Technical Board of Appeal 3.3.10
of 5 March 2020

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Decision under appeal: Interlocutory decision of the Opposition
Division of the European Patent Office posted on
4 January 2016 concerning maintenance of the
European Patent No. 2100867 in amended form.
Composition of the Board:

Chairman: P. Gryczka
Members: J.-C. Schmid
T. Bokor
Summary of Facts and Submissions

I. The Appellant (opponent 1) lodged an appeal against the interlocutory decision of the Opposition Division which found that the European patent No. 2 100 867 in amended form met the requirements of the EPC.

II. Notice of opposition had been filed by the Appellant and the party as of right (opponent 2) requesting revocation of the patent-in-suit in its entirety on the grounds of lack of novelty and inventive step (Article 100(a) EPC), and insufficient disclosure of the invention (Article 100(b) EPC).

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

(1) WO-A-2009/084703,
(2) WO-A-2007/117391,
(3) WO-A-2008/030440 and,

According to the Opposition Division, the deletion of the term "optionally" in step b) of claim 1 limited the subject-matter to an embodiment already claimed and disclosed in the application as filed and hence did not infringe the requirements of Articles 123(2) and (3) EPC. The same applied to the amendment of the feature "one or more" to "plurality" in claim 3. The claimed method was described in a manner sufficiently clear and complete in the patent-in-suit for it to be carried out by a skilled person. The subject-matter of claim 1 of the then pending main request was novel over document (1) since there was no disclosure of a separator between the hydrogenation reactor (R11) and the
dehydrohalogenation reactor (R12) in the process according to the first embodiment of document (1). Lastly, the subject-matter of claims 1 to 3 of the then pending request involved an inventive step starting from document (3) as the closest prior art to the invention.

III. According to the Appellant, the subject-matter of claims 1 to 3 maintained by the opposition division introduced subject-matter which extended beyond the content of the application as filed (Article 123(2) EPC) and also extended the protection conferred by the patent as granted (Article 123(3) EPC). Furthermore the claimed invention was not sufficiently disclosed in the patent-in-suit to be performed by the skilled person (Article 100(b) EPC). The subject-matter of the claims lacked novelty with respect to document (1). The Appellant submitted that the claimed subject-matter lacked an inventive step starting from document (3) or (4) as the closest prior art to the invention.

IV. In reply to the statement of the grounds of appeal, the respondent filed seven auxiliary requests and contested the Appellant’s arguments.

V. With a communication pursuant to Article 15(1) RPBA to prepare the oral proceedings, the Board indicated its preliminary view that the skilled person would not be able to produce a fluorinated olefin by the claimed method from the overwhelming majority of alkenes of formula I, and hence the requirements of sufficiency of disclosure of the invention were apparently not met. Furthermore the subject-matter of claim 3 appeared to lack an inventive step in the light of document (4) (Article 56 EPC).
VI. With a letter dated 18 February 2020, the Respondent filed a new main request and two auxiliary requests. The main request corresponds to auxiliary request 6 filed with the response to the statement of grounds of appeal.

Independents claims 1 and 2 of the main request read as follows:

"1. A method for producing at least one fluorinated olefin comprising:

a. hydrogenating a starting material stream comprising at least one alkene according to Formula (I):

\[(\text{CX}_n\text{Y}_{3-n})(\text{CR}_1^a\text{R}_2^b)\text{zCX}=\text{CH}_m\text{X}_{2-m}\] (I)

by contacting said starting material with a reducing agent to produce an intermediate product stream comprising at least one alkane according to Formula (II):

\[(\text{CX}_n\text{Y}_{3-n})(\text{CR}_1^a\text{R}_2^b)\text{zCHX}=\text{CH}_{m+1}\text{X}_{2-m}\] (II)

where:
each X is independently Cl, F, I or Br, provided that at least two Xs are F;
each Y is independently H, Cl, F, I or Br;
each \text{R}_1^1 is independently H, Cl, F, I, Br or unsubstituted or halogen substituted methyl or ethyl radical;
each \text{R}_2^2 is independently H, Cl, F, I, Br or unsubstituted or halogen substituted methyl or ethyl radical;
n is 1, 2 or 3;
a and b are each 0, 1 or 2, provided that a+b = 2;
m is 0, 1 or 2; and
z is 0, 1, 2 or 3;

b. separating said intermediate product stream into a plurality of streams, said plurality of intermediate product streams comprising two or more streams selected from the group consisting of a first stream rich in at least a first alkane according to Formula II, a second stream rich in at least a second alkane according to Formula II, and an alkane recycle stream;

c. dehydrofluorinating at least a portion of said plurality of intermediate process streams of step (b) to produce an alkene product stream comprising 1,1,1,2,3-pentafluoropropene and at least one additional alkene having a lower degree of fluorine substitution compared to the degree of fluorination of the compound of formula (I);

d. separating at least a portion of said 1,1,1,2,3-pentafluoropropene from said alkene product stream to produce a recycle stream rich in 1,1,1,2,3-pentafluoropropene and a final product stream rich in said additional alkene; and

e. introducing said recycle stream at said step (a), wherein at least a portion of said 1,1,1,2,3-pentafluoropropene is hydrogenated to form a portion of said intermediate product stream;

wherein said starting material stream comprises hexafluoropropylene and said additional alkene is 1,1,1,2-tetrafluoropropene.”

“2. A system for producing at least one fluorinated olefin comprising:
a. a hydrogenation reactor;
b. a starting material feed stream fluidly connected to said hydrogenation reactor, wherein said starting material feed stream comprises hexafluoropropylene;
c. one or more intermediate streams fluidly connected to said hydrogenation reactor;
d. a first separator fluidly connected to said one or more intermediate streams;
e. a plurality of post separator streams fluidly connected to said first separator;
f. optionally, a first recycle stream fluidly connected to said first separator and said hydrogenation reactor;
g. a dehydrohalogenation reactor fluidly connected to at least one of said post separator streams;
h. a product stream fluidly connected to said dehydrohalogenation reactor;
i. a second separator fluidly connected to said product stream;
j. a second recycle stream fluidly connected to said second separator and said hydrogenation reactor; and
k. a final product stream fluidly connected to said second separator,

wherein said final product stream is rich in 1,1,1,2-tetrafluoropropene and said second recycle stream comprises 1,1,1,2,3-pentafluoropropene.”

VII. Opponent (2), which is party as of right to the appeal proceedings, did not file any requests or submissions in the appeal proceedings.

VIII. The Appellant requested that the decision under appeal be set aside and the patent be revoked. The Respondent (patent proprietor) requested that the decision under appeal be set aside and the patent be
maintained in an amended form on the basis of claims 1 and 2 of the main request, or alternatively on the basis of the 1st or 2nd auxiliary request, all filed with letter dated 18 February 2020.

IX. At the end of the oral proceedings held on 5 March 2020, the decision of the Board was announced.

Reasons for the Decision

1. The appeal is admissible.

Main request

2. Amendments

2.1 Claim 1

According to the Appellant, the subject-matter of claim 1 of the main request extended beyond the application as filed, since making step (b) mandatory in the claimed process introduced in claim 1 a technical contribution not disclosed in the application as filed, and hence contravened Article 123(2) EPC.

Claim 1 of the main request corresponds to the combination of claims 1 and 2 of the application as filed, wherein step (b) which was optional has been made mandatory. This amendment is achieved by removing the word “optionally” from step (b) and by removing the phrase “said intermediate process stream from step (a)” to maintain antecedent continuity between steps (b) and (c).

The Board cannot accept the argumentation of the Appellant based on a non-disclosed technical
contribution. In fact, given that the process including step b., although as an option, is nevertheless clearly and unambiguously disclosed in the application as filed, the subject-matter of claim 1 of the main request does not contain subject-matter which extends beyond the content of the application as filed.

Claim 1 is clearly and unambiguously derivable from originally filed claims 1 and 2, and therefore meets the requirements of Article 123(2) EPC. Furthermore, as claim 1 as granted specified already that in the process the step (b) is mandatory, claim 1 of the present main request also meets the requirement of Article 123(3) EPC.

2.2 Claim 2.

According to the Appellant, the expression “plurality of” and the term “more” in the expression “one of more” were not equivalent. Therefore, the amendment in feature e. of the system of claim 2 from “one of more” to “a plurality” extended the protection conferred by the patent as granted, contrary to Article 123(3) EPC.

Claim 2 of the main request corresponds to claim 3 of the patent as granted, or claim 10 of the application as filed wherein inter alia feature e. has been amended from “one or more post separator streams” to “a plurality of post separator streams”, with the sole consequence that a system comprising only one post separator stream is no longer covered by claim 2 of the main request. This amendment therefore represents a restriction of the scope of protection conferred by the granted patent.
Furthermore, claim 2 has been amended to specify that the recycle stream in feature k. comprises 1,1,1,2,3-pentafluoropropene (HFO-1225ye) according to page 10, lines 12 to 15 of the application as filed.

Consequently, claim 2 meets the requirements of Articles 123 (2) and (3) EPC.

3. **Sufficiency of disclosure**

3.1 The process of claim 1 of the main request has been amended to specify that the starting material stream comprises hexafluoropropylene (HFP).

The skilled person is well aware from its general knowledge and from the patent in suit that hydrogenating a mixture of hexafluoropropylene (HFP) and recycled 1,1,1,2,3-pentafluoropropene (HFO-1225ye), produces both 1,1,1,2,3,3-hexafluoropropane (HFC-236ea) and 1,1,1,2,3-pentafluoro propane (HFC-245eb), which are alkanes according to Formula (II) (see paragraph (0007), reactions set out in paragraphs [0029] and [0030] of the patent). The dehydrofluorinating of HFC-236ea and HFC-245eb produces HFO-1225ye and 1,1,1,2-tetrafluoropropene (HFO-1234yf).

The skilled person would therefore be able to produce HFO-1234yf starting from a material comprising HFP following the process of claim 1 of the main request.

3.2 According to the Appellant, step (b) of claim 1 related to the separation of the intermediate product stream into a plurality of intermediate product streams, whereas step (c) of claim 1 related to dehydrofluorinating at least a portion of said plurality of intermediate process streams of step (b).
The skilled person was therefore not capable of identifying the origin of the intermediate process streams and their composition.

However, it is clear to the skilled person that the streams to be dehydrofluorinated in step (c) are the intermediate product streams of step (b) which produce HFO-1225ye and HFO-1234yf when dehydrofluorinated.

According to the Appellant, there was no teaching in the patent how to produce an alkene product stream comprising HFO-1225ye and HFO-1234yf from a stream devoid of HFC-236ea.

However, claim 1 does not require the production of HFO-1225ye and HFO-1234yf from a stream devoid of HFC-236ea; the separation of a stream rich in HFC-236ea from an intermediate process stream does not mean that the latter is devoid of HFC-236ea.

According to the Appellant, claim 1 encompassed a method in which the “plurality of streams” has only a stream rich in HFC-236ea and an alkane recycle stream. There was no disclosure in the patent to produce a stream comprising HFO-1225ye and HFO-1234yf from these streams.

However, claim 1 requires that the plurality of intermediate product streams of claim 1 comprises “two or more streams selected from the group consisting of a first stream rich in at least a first alkane according to Formula II, a second stream rich in at least a second alkane according to Formula II, and an alkane recycle stream”. It is not required by claim 1 that the first stream only includes one of HFC-236ea and HFC-245eb. Rather, it is specified that the first
stream is rich in an alkane according to Formula II, such as HFC-236ea. Therefore, the first stream can include both HFC-236ea and HFC-245eb and can therefore be dehydrofluorinated to produce both HFO-1225ye and HFO-1234yf. Hence, the skilled person can carry out the embodiment of claim 1 where the intermediate product stream is separated in step (b) in two streams, namely a stream rich in HFC-236ea and an alkane recycle stream.

According to the Appellant, the alkane recycle stream was not sufficiently disclosed in the sense that the skilled person had no information about the compounds contained in the stream.

However, the patent provides information about the alkane recycle stream. For example, paragraph [0028] indicates that the alkane recycle stream comprises a relatively low temperature stream comprising a portion of the reaction product stream 3A after it has been cooled and separated and that the alkane recycle stream is preferably rich in HFC-236ea, HFC-245eb, or a combination of these.

According to the Appellant, it was not taught in the patent in suit how to dehydrofluorinate the alkane recycle stream to produce HFO-1225ye and HFO-1234yf.

However, the patent clearly teaches that it is the first stream rich in at least a first alkane according to Formula II and/or the second stream rich in at least a second alkane according to Formula II which is subjected to the dehydrofluorination step (c) in the process of claim 1. The alkane recycle stream, if present, is recycled back to the hydrogenation reactor, as disclosed in paragraph [0027], lines 55 to 57, where
it is stated that the feed stream 2 comprises the hydrogenation agent, preferably H₂, for the reaction step A. Flow path or stream 4, i.e. the alkane recycle stream, is a path for allowing introduction of a recycle stream into the reaction step. Figure 1 also indicates that stream 4 is recycled to the hydrogenation reactor A.

3.3 The Board therefore concludes that the skilled person can carry out the claimed process without undue burden and that the invention is thus sufficiently disclosed in the patent as required by Article 83 EPC.

4. **Novelty with respect to document (1)**

Document (1) discloses two separate processes for producing 1,1,1,2-tetrafluoropropene (HFO-1234yf), (see embodiment 1; page 4, line 14 to page 7, line 4; page 10, line 1 to page 16, line 9, claims 1 and 2, example 1, figure 1 and see embodiment 2, page 7, line 5 to page 8, line 18, pages 16, line 11 to page 21, line 16; example 2 claims 3 and 4, figure 2).

According to the Appellant, the subject-matter of claim 1 and 2 lacks novelty with respect to the process disclosed in the first embodiment of document (1).

Embodiment 1 of document (1) relates to a process for producing 1,1,1,2-tetrafluoropropene (HFO-1234yf) comprising hydrogenating hexafluoropropene (HFP) to obtain a reaction mixture containing 1,1,1,2,3,3-hexafluoropropene (HFC-236ea) and 1,1,1,2,3-pentafluoropropene (HFC-245eb), dehydrofluorinating this mixture to obtain a reaction mixture containing 1,1,1,2,3-pentafluoropropene (HFO-1225ye) and 1,1,1,2-tetrahydropropene (HFO-1234yf), separating by
distillation this mixture, recycling the fraction containing 1,1,1,2,3-pentafluoropropene (HFO-1225ye) to the first hydrogenation step (see claim 1 of document (1)).

The process of claim 1 of document (1) comprises steps a., c., d. and e. of the claimed process, but not step b. which requires the separation of the intermediate product stream obtained from step a. into a plurality of streams, said plurality of intermediate product streams comprising two or more streams selected from the group consisting of a first stream rich in at least a first alkane according to Formula II, a second stream rich in at least a second alkane according to Formula II, and an alkane recycle stream.

According to the Appellant, this requirement was disclosed at page 16, lines 2 to 4 of document (1) indicating that quite small, low-boiling and high-boiling impurities may be removed from the stream(s) (e.g. the product stream and the recycling stream) by distillation, if appropriate, in combination with the passage of page 10, lines 19 to 23 indicating that it is preferred that there is substantially no material which would not be finally converted into HFO-1234yf.

However, this passage of document (1) does not disclose that the impurities are removed from the first product stream. It also does not disclose that at least two streams containing alkanes are obtained. Example 1 and figure 1 of document (1) show that the product stream (S11) after hydrogenation contains 51% of a first alkane and 49% of a second alkane and only traces of undefined impurities. In this embodiment, the entire stream S11 is subjected to the dehydrofluorinating reaction, without any separation.
The Appellant further relies on page 16, lines 7 to 9 where it is disclosed that unreacted hydrogen may exist in the stream(s), and that it may be appropriately treated by, for example separation and removal, or recycling to the reactor R11. According the Appellant, this amounts to an implicit disclosure of step b., since it was obvious for the skilled person that unreacted hydrogen must be removed before the dehydrofluorination step and that at least some alkanes should be present in the hydrogen stream.

The Board observes that it is a generally applied principle that for concluding lack of novelty, there must be a direct and unambiguous disclosure in the state of the art which would inevitably lead the skilled person to subject-matter falling within the scope of what is claimed. In this context an "implicit disclosure" on which the novelty attack of the Appellant is based must mean a clear and unambiguous inevitable consequence of what was explicitly disclosed.

In the present case, however, the Appellant's arguments are not based on the consequences of the explicit disclosure of document (1), but on considerations relating to what the skilled person would likely have done or were likely to assume. These arguments are therefore rejected.

The Board comes therefore to the conclusion that document (1) does not directly and unambiguously disclose the subject-matter of claim 1 of the main request.

For the same reason, the subject-matter of claim 2 of the main request is novel with respect to document (1)
since the system disclosed in the first embodiment does not comprise a first separator fluidly connected to the hydrogenation reactor and a plurality of post separator streams fluidly connected to said first separator (features d. and e. of the claimed system).

Hence, the Board holds the subject-matter of claims 1 and 2 novel over document (1) (Article 54(3) EPC).

5. Inventive step

5.1 Closest prior art

During the oral proceedings before the Board, the Appellant accepted that document (4) represented the closest prior art for claims 1 and 2 of the main request.

This document discloses the following four steps for producing 1,1,1,2-tetrafluoropropene (HFO-1234yf) from hexafluoropropene (HFP), i.e.

- hydrogenation of HFP to obtain a reaction mixture containing 1,1,1,2,3,3-hexafluoropropane (HFC-236ea),

- dehydrofluorination of this compound to obtain 1,1,1,2,3-pentafluoropropene (HFO-1225ye),

- hydrogenation of HFO-1225ye to obtain HFC-245eb and,

- dehydrofluorination of HFC-245eb to produce HFO-1234yf (see the examples of document (4)).

5.2 Technical problem
According to the Respondent, the technical problem to be solved was the provision of an improved process in terms of costs to produce HFO-1234yf and a system to perform this process.

6. Solution

The proposed solution is the process of claim 1, characterized by steps b. and e., i.e.

- that the hydrogenation product stream is separated into a plurality of intermediate product streams comprising two or more streams selected from the group consisting of a first stream rich in at least a first alkane according to Formula II, a second stream rich in at least a second alkane according to Formula II, and an alkane recycle stream and,

- that at least a portion of the alkene product stream comprising 1,1,1,2,3-pentafluoropropene obtained from step c. is recycled at step a..

According to the Appellant step b. was disclosed in document (4). Paragraph [0035] discloses that at least a portion of the first effluent stream is contacted with a catalyst in the second stage, with the consequence that the first effluent stream is separated into at least two streams.

Document (4) only foresees that the hydrogenation reaction product may optionally be purified (see paragraph [0020]). With respect to paragraph [0025], using only a portion of a stream does not imply that the stream is separated into at least two streams having different composition, let alone that the streams are selected from the group consisting of a
first stream rich in at least a first alkane according to Formula II, a second stream rich in at least a second alkane according to Formula II, and an alkane recycle stream. Hence, the Appellant’s argument is rejected.

The solution to the technical problem as defined above is also reflected in the system of claim 2 in particular by the following features:
- d. a first separator fluidly connected to said one or more intermediate streams;
- e. a plurality of post separator streams fluidly connected to said first separator;
- j. a second recycle stream fluidly connected to said second separator and said hydrogenation reactor; and
- in that said second recycle stream comprises 1,1,1,2,3-pentafluoropropene.

6.1 Success

The Respondent indicated that the improvement achieved by the claimed process resided in cost savings through the sharing of the hydrogenation reactor for the first and third step of the 4-step process of producing HFO-1234ye from HFP.

According to the Appellant, this problem was not solved for the subject-matter of claim 2, since the claimed system could comprise more than one hydrogenation reactor.

However, the proposed solution in not characterized by the number of the hydrogenation reactors, but requires inter alia that at least one reactor is connected to a recycle stream comprising 1,1,1,2,3-pentafluoropropene. This Appellant’s argument is thus rejected.
In view of the above, the Board is satisfied that the technical problem has been solved by the proposed solution.

6.2 Obviousness

According to the Appellant, the proposed solution was obvious in the light of document (2), which teaches that chemical reactions can be carried out on mixtures (see claim 1). Document (4) discloses that HFP and HFO-245eb are hydrogenated under the same condition. Hence, it was obvious for the skilled person to modify the process of document (4) using the same hydrogenation reactor for both the first and the third steps of the process in order to save costs.

Document (2) discloses that a blend of HFC-236ea and HFC-245eb can be dehydrofluorinated into HFO-1234ye and HFO-1225ye. This document thus does not provides any hint to the skilled person how to modify the 4-step process disclosed in document (4), let alone in the way defined in the claimed process and apparatus.

The Appellant's argument that the skilled person led by the teaching of document (2) would have modified the process of document (4) as to recycle the HFO-1225ye at the first hydrogenation step, thus arriving at the claimed method is neither based on a prior art disclosure, nor on the general knowledge of the skilled person, and thus can only be seen as the result of an ex post facto analysis, i.e. an analysis made with the knowledge of the invention in mind and with the aim of reconstructing on purpose the claimed method and system.
Therefore, the Board comes to the conclusion that the subject-matter of claim 1 of the main request, and for the same reason, that according to claim 2 directed to a system including the recycle line comprising HFO-1225ye to the hydrogenation reactor involves an inventive step within the meaning of Article 56 EPC.

7. **First and second auxiliary requests**

Since the main request is allowable, it is not necessary to decide on the auxiliary requests.

8. In view of the fact that several embodiments in the description are no longer covered by the claims held allowable by the Board, the description needs to be adapted to these claims. The Board deems it expedient to remit the case to the Opposition Division for the adaptation of the description, and the parties agreed to this procedure.
Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the opposition division with the order to maintain the patent in an amended form on the basis of Claims 1 and 2 of the main request filed with letter dated 18 February 2020 and a description to be adapted to these claims.

The Registrar:                      The Chairman:

C. Rodriguez Rodriguez              P. Gryczka

Decision electronically authenticated