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Datasheet for the decision of 16 May 2017

Case Number: T 0529/15 - 3.3.10

Application Number: 09158166.0

Publication Number: 2112131

IPC: C07C17/25, C07C21/18

Language of the proceedings: EN

Title of invention:
Process for dehydrofluorination of 3-chloro-1,1,1,3-tetrafluoropropane to 1-chloro-3,3,3-trifluoropropene

Patent Proprietor:
Honeywell International Inc.

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

Headword:

Relevant legal provisions:
EPC Art. 100(a), 123(2), 54, 56

Keyword:

Grounds for opposition - lack of patentability (yes)
Inventive step - (no) - 1st to 4th auxiliary requests - (yes) - 5th auxiliary request
Amendments - allowable (yes) - 5th auxiliary request
Novelty - (yes) - 5th auxiliary request

Decisions cited:

Catchword:

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Case Number: T 0529/15 - 3.3.10

D E C I S I O N
of Technical Board of Appeal 3.3.10
of 16 May 2017

Appellant: (Opponent 1)	ARKEMA FRANCE Département Propriété Industrielle 420, rue d'Estienne d'Orves 92700 Colombes (FR)
Representative:	Dang, Doris ARKEMA FRANCE Département Propriété Industrielle 420, rue d'Estienne d'Orves 92705 Colombes Cedex (FR)
Appellant: (Opponent 2)	Mexichem Amanco Holding S.A. de C.V. Río San Javier No. 10 Fraccionamiento Viveros del Río Tlalnepantla, Estado de México C.P. 54060 (MX)
Representative:	Potter Clarkson LLP The Belgrave Centre Talbot Street Nottingham NG1 5GG (GB)
Respondent: (Patent Proprietor)	Honeywell International Inc. 101 Columbia Road Morristown, NJ 07962 (US)
Representative:	Crooks, Elizabeth Caroline Kilburn & Strode LLP 20 Red Lion Street London WC1R 4PJ (GB)
Decision under appeal:	**Interlocutory decision of the Opposition Division of the European Patent Office posted on 27 January 2015 concerning maintenance of the European Patent No. 2112131 in amended form.**

Composition of the Board:

Chairman	P. Gryczka
Members:	R. Pérez Carlón
	C. Schmidt

Summary of Facts and Submissions

I. The appeal lies from the decision of the opposition division to maintain European patent No. 2 112 131 in the form of the main request then pending. Both opponents appealed the decision.

II. Two notices of opposition had been filed, on the grounds of added subject-matter (Article 100(c) EPC), insufficiency of disclosure (Article 100(b) EPC), and lack of novelty and inventive step (Article 100(a) EPC).

III. The documents filed during the opposition proceedings included the following:

D4: EP 0 974 571 A2
D11: US 2007/0129579 A1
D13: EP 1 916 231 A2
E18: Asbury Carbons, Amorphous Graphite, 2013 http://asbury.com/technical-presentations-papers/materials-in-depth/amorphous-graphite/

The documents filed during the appeal proceedings included the following:

D19: Types of carbon adsorbents and their production, J. A. Menéndez-Díaz and I. Martín-Gullón, Activated carbon surfaces in environmental remediation (Interface science and technology series 7) T. Bandosz Ed. Elsevier 2006, 1-48
D20: Desotec Activated Carbon https://www.desotec.com/fr/carbonology/carbonology-academy/chemical-structure-activated-carbon
D21: Activated carbon principles. Information Bulletin Calgon Carbon Corporation.

IV. The opposition division concluded with respect to the issue of inventive step that document D11, which related to a process for obtaining the same product from the same starting material, was the closest prior art. The problem underlying the claimed invention was to provide an alternative process for the dehydrofluorination of 244fa, and the solution, which was characterised by the catalyst employed, was inventive having regard to the prior art.

V. Claim 1 of the main request, which corresponds to the main request in the opposition proceedings, reads as follows:

"A process for making 1-chloro-3,3,3-trifluoropropene, comprising dehydrofluorinating 3-chloro-1,1,1,3-tetrafluoropropane under conditions sufficient to effect dehydrofluorination in the presence of a catalyst selected from the group consisting of (i) one or more halogenated trivalent or higher valent metal oxides, (ii) one or more natural or synthetic graphites, and (iii) combinations thereof, wherein the dehydrofluorinating is carried out in the vapor phase."

Claim 1 of the first auxiliary request relates only to embodiment (i) of claim 1 of the main request.

Claim 1 of the second auxiliary request contains all the features of claim 1 of the first auxiliary request, and, in addition,

"*wherein the metal ion of the catalyst is selected from the group consisting of Al^{3+}, Ga^{3+}, In^{3+}, Sc^{3+}, Y^{3+}, La^{3+}, Cr^{3+}, Fe^{3+}, Co^{3+}, Ti^{4+}, Zr^{4+}, Ce^{4+}, Sn^{4+}, Mn^{4+}, Nb^{5+}, W^{6+}, and combinations thereof, wherein the*

dehydrofluorinating is carried out at a temperature from 100°C to 600°C, wherein the dehydrofluorinating is carried out at a pressure from 101 kPa to 2027 kPa (1 atm to 20 atm), and wherein the dehydrofluorinating is carried out for a residence time from 0.5 seconds to 600 seconds."

Claim 1 of the third auxiliary request contains all the features of claim 1 of the first auxiliary request, and, in addition,

"wherein the metal ion of the catalyst is Cr^{3+} and the halogen of the catalyst is F."

Claim 1 of the fourth auxiliary request contains all the features of claim 1 of the third auxiliary request and, in addition,

"*wherein the dehydrofluorinating is carried out at a temperature from 100°C to 600°C, wherein the dehydrofluorinating is carried out at a pressure from 101 kPa to 2027 kPa (1 atm to 20 atm), and wherein the dehydrofluorinating is carried out for a residence time from 0.5 seconds to 600 seconds."*

Lastly, claim 1 of the fifth auxiliary request is directed to embodiment (ii) of claim 1 of the main request, and reads as follows:

"A process for making 1-chloro-3,3,3-trifluoropropene, comprising dehydrofluorinating 3-chloro-1,1,1,3-tetrafluoropropane under conditions sufficient to effect dehydrofluorination in the presence of a catalyst selected from one or more natural or synthetic graphites, wherein the dehydrofluorinating is carried

out in the vapor phase."

VI. The arguments of appellants 1 and 2 (opponents 1 and 2) relevant for the present decision were the following:

Concerning the main request and auxiliary requests 1 to 4:

Examples 2 to 4 of document D11, which disclosed a process for preparing HCFC-1233zd from 244fa, over metal halide catalysts, were the closest prior art for claim 1 of the main request and of the first to the fourth auxiliary requests. The technical problem underlying the claimed invention was to provide a further process for preparing HCFC-1233zd by dehydrofluorination of 244fa. The solution, which was characterised by using a catalyst selected from one or more halogenated trivalent or higher valent metal oxides, such as fluorinated Cr^{3+} oxides, was obvious having regard to D4, which also disclosed the reactions conditions required by claim 1 of the second and fourth auxiliary requests. For these reasons, the process of claim 1 of the main request and of the first to fourth auxiliary requests was not inventive.

Concerning the fifth auxiliary request:

Claims 2, 3 and 4 of the fifth auxiliary request contained added subject-matter, as the application as originally filed did not disclose the combination of temperature, pressure and residence time required by said claims.

Documents D19 to D21 were filed with the statement setting out the grounds of appeal in response to the arguments of the opposition division in the contested

decision and should therefore be admitted into the proceedings.

Documents D11 and D13 disclosed a process for making HCFC-1233zd by dehydrofluorination of 244fa over activated carbon, which was a form of graphite, as shown by D19. For this reason, the process of claim 1 of the fifth auxiliary request was not novel.

If, nevertheless, it were considered that activated carbon was not a form of graphite, example 1 of document D11 would be the closest prior art for the assessment of inventive step, and the technical problem underlying the claimed invention would be to provide a further process for preparing HCFC-1233zd by dehydrofluorination of 244fa. The claimed solution was obvious, as document D11 disclosed that graphite could be used as an additive or a support in the same process, with the consequence that the process of claim 1 of the fifth auxiliary request was not inventive.

VII. The respondent (patent proprietor) did not dispute that document D11 was the closest prior art, or that the technical problem underlying the claimed invention was to provide a further process for preparing HCFC-1233zd by dehydrofluorination of 244fa. It considered, however, that the skilled person would not have modified the process of D11 by changing the catalyst, which D11 disclosed as essential. For this reason, he would not have combined the teaching of D4 with that of D11. The process of claim 1 of the main request and of the first to fourth auxiliary requests was thus inventive.

There was no reason for not filing documents D19 to D21 during the opposition proceedings. Their filing was not

a reaction to arguments of the opposition division which could have caught the appellants by surprise. For this reason, these documents should not be admitted into the proceedings.

Claims 2, 3 and 4 of the fifth auxiliary request found a basis in claims 7, 8 and 9 as originally filed and, for this reason, the fifth auxiliary request did not contain added subject-matter.

As documents D11 and D13 disclosed a process carried out in the presence of activated carbon, which was not graphite, claim 1 of the fifth auxiliary request was novel.

Example 1 of document D11 was the closest prior art for the process of claim 1 of the fifth auxiliary request. The problem underlying the claimed invention was to provide a further process for preparing HCFC-1233zd by dehydrofluorination of 244fa. The solution, which was characterised by using graphite as a catalyst, was inventive as none of the documents opposed to the patent in suit related to graphite as a catalyst, let alone in the context of a similar reaction.

VIII. Oral proceedings before the board of appeal took place on 16 May 2017.

IX. The final requests of the parties were the following:

- Appellants 1 and 2 requested that the decision under appeal be set aside and that the patent be revoked.

- The respondent requested that the appeal be dismissed (main request) or subsidiarily, that the

patent be maintained in the form of one of the auxiliary requests 1 to 8, all requests having been filed with letter dated 26 October 2015.

X. At the end of the oral proceedings, the decision was announced.

Reasons for the Decision

1. The appeal is admissible.

Fourth auxiliary request

2. Claim 1 of the fourth auxiliary request is directed to a process for making 1-chloro-3,3,3-trifluoropropene (HCFC-1233zd) comprising dehydrofluorinating 3-chloro-1,1,1,3-tetrafluoropropane (244fa) in the vapour phase, in the presence of fluorinated Cr^{3+} oxides at 100°C to 600°C, 1 atm to 20 atm, and during 0.5 seconds to 600 seconds.

3. Inventive step

3.1 Closest prior art

The opposition division and the parties considered that document D11 was the closest prior art, and the board sees no reason to differ.

It has not been disputed that examples 2 to 4 of document D11 disclose the preparation of HCFC-1233zd by dehydrofluorination of 244fa over metal halides as catalysts. The process of D11 differs from the process of claim 1 of the fourth auxiliary request by virtue of the nature of the catalyst and the temperature,

pressure and residence time conditions.

3.2 Technical problem underlying the invention

It has not been disputed that the technical problem underlying the claimed invention is to provide an alternative process for making HCFC-1233zd by dehydrofluorination of 244fa.

3.3 Solution

The solution to this technical problem is the process of claim 1, characterised in that it requires a catalyst selected from one or more fluorinated Cr^{3+} oxides at a temperature from 100°C to 600°C, at a pressure from 1 atm to 20 atm, and for a residence time from 0.5 seconds to 600 seconds.

3.4 Success

The board agrees with the opposition division and the parties that, having regard to the data provided in table 1 of the patent in suit, the problem formulated in point 3.2 above is credibly solved by the process of claim 1.

3.5 It thus remains to be decided whether or not the proposed solution to the objective problem defined above is obvious in view of the state of the art.

3.5.1 Trying to obtain an alternative process for preparing HCFC-1233zd from 244fa, the skilled person would turn to processes which could be successfully carried out over similar starting materials and involve the same reaction type, such as document D4, which describes the vapour phase dehydrofluorination of 1,1,1,3,3-

pentafluoropropene (245fa), which differs from 244fa (3-chloro-1,1,1,3-trifluoropropene) merely by having a 3-fluorine instead of a 3-chlorine substituent. In D4, the dehydrofluorination reaction is carried out under the same conditions and over the same catalyst as required by claim 1, i.e. over fluorinated chromia, at 100°C to 600°C, atmospheric pressure and for a contact time of 1 second to 60 seconds [0004]. Therefore, the claimed process is obvious for the skilled person from the combination of the teaching of D11 with D4.

For this reason, it is concluded that the process of claim 1 is not inventive, as required by Article 56 EPC, with the consequence that the fourth auxiliary request is not allowable.

3.5.2 The respondent argued that the skilled person would not have taken into consideration the teaching of D4, as it was directed to a different reaction over a different starting material, and thus aimed at a different objective. The respondent further argued that the skilled person could modify the process of D11 in multiple ways, and that document D11 disclosed the nature of the catalyst as essential. The skilled person would have considered proven chemistry, such as that disclosed in D9, and use 240fa as starting material, but would not have been directed towards D4, as he would not contemplate changing the catalyst of D11.

3.5.3 However, the skilled person would turn to processes which could be successfully carried out over a similar starting material (245a) by an analogous mechanism (dehydrofluorination). Notwithstanding that halogenated chromium oxide could catalyse both dehydrochlorination and dehydrofluorination, known to be competing (examples of D11), the skilled person would expect

dehydrofluorination to take place, at least to some extent.

In addition, the fact that the skilled person could modify a process in various ways, by changing the starting material, the catalyst, temperature, pressure, etc., does not render each of these options inventive.

3.5.4 Lastly, the respondent argued that documents D11 and D4 had been published almost 10 years before the filing of the patent in suit. If it were obvious to combine their teaching, it should have been disclosed before.

However, there could be many reasons why such a disclosure was not made before, on which it is only possible to speculate.

3.5.5 For these reason, the respondent's arguments cannot succeed.

Main request, first to third auxiliary request

4. As the process of claim 1 of the fourth auxiliary request represents an embodiment of claim 1 of the main request and of the first, second and third auxiliary requests, the reasoning with respect to inventive step in point 3. above applies *mutatis mutandis*, with the consequence that neither the main request nor the first, second or third auxiliary requests are allowable.

Fifth auxiliary request

5. Claim 1 of the fifth auxiliary request is directed to a process for making HCFC-1233zd by dehydrofluorinating 244fa in the vapour phase, and in the presence of a

catalyst selected from one or more natural or synthetic graphites.

6. Amendments

6.1 It has not been disputed that claim 1 of the fifth auxiliary request finds a basis in the combination of claims 1 and 3, and embodiment (iii) of claim 2 as originally filed.

Claims 2, 3 and 4 of the fifth auxiliary request relate to temperature, pressure and residence time of the process of claim 1. It has not been disputed either that these features can be found in claims 7, 8 and 9 as originally filed or that each of these claims only refers back to the process either of claim 1 or of claim 2 as originally filed, but not to claim 3, whose features have been included in claim 1 of the fifth auxiliary request, or to each other.

6.2 Appellant 1 argues that claims 2 to 4 of the fifth auxiliary request refer to "any preceding claim" and that, in doing so, they are now directed to combinations which were not disclosed in the application as originally filed.

However, temperature, pressure and residence time are not optional features, but parameters required by every reaction. For this reason alone, the subject-matter of original claims 7 to 9, which disclose the broadest possible range for each of these variables in the application as originally filed, would be considered as combined by the skilled person, and provide the required basis for claims 2, 3 and 4 of the fifth auxiliary request.

6.3 Appellant 1 further argued that claim 1 related to a selection within a "Markush formula" defined in claim 2 as originally filed, and that the conditions of temperature, pressure and time of claims 2 to 4 of the fifth auxiliary request were not explicitly combined with embodiment (iii) of said "Markush formula".

However, it is not disputed that claim 2, to which claims 7 to 9 as originally filed refer back, explicitly discloses the embodiment that the catalyst is "one or more natural or synthetic graphites" required by claim 1 of the fifth auxiliary request. There is no hint in the application as originally filed indicating that different catalysts need different reaction conditions, and all the examples thereof are carried out at the same temperature, in reactors of the same size and with the same flow rate (these parameters determine the same pressure and residence time), irrespectively of the catalyst used. Thus, there is no reason why these requirements would not be combined with each type of catalyst in claim 2 as originally filed, including one or more natural or synthetic graphites.

6.4 It is thus concluded that claims 1 to 4 of the fifth auxiliary request find a basis in the application as originally filed, as required by Article 123(2) EPC.

6.5 Claim 1 of the fifth auxiliary request is restricted only to embodiment (ii) of claim 1 as granted and further includes the feature "under conditions sufficient to effect dehydrofluorination". Thus, claim 1 of the fifth auxiliary request does not extend the scope of protection conferred by the patent as granted (Article 123(3) EPC).

7. Admissibility of late-filed documents

The respondent requested that documents D19 to D21, filed by appellant 1 under cover of a letter dated 21 March 2017, not be admitted into the proceedings.

Of these documents, only D19 was discussed at the oral proceedings before the board. It aims at supporting appellant 1's argument that activated carbon is a form of graphite, which had been part of the opposition proceedings on which the opposition division had decided and which had been maintained in the statement setting out the grounds of appeal. This document had been filed approximately six weeks before the oral proceedings, the respondent had had sufficient time to familiarise itself with its content, and in fact also relied on it for supporting its arguments.

For these reasons, document D19 is admitted into the proceedings (Article 13(1) RPBA).

As none of documents D20 and D21 is relevant for the present decision, it is not necessary to decide whether they should be admitted into the proceedings.

8. Novelty

8.1 Both appellants considered that documents D11 and D13, which describe in their example 1 a process over activated carbon, disclosed all the features of claim 1, as activated carbon was a type of graphite.

Document D19 discloses that activated carbon is a "non-graphitic", "non-graphitizable carbon" (page 3, third full paragraph; section 4.1, first paragraph). Graphite, which is crystalline, is an allotropic form

of carbon (see figure 1 of D19), i.e. contains only carbon atoms. In contrast, activated carbon is amorphous and contains elements other than carbon (figures 3 and 5 of D19). Hence, it cannot be concluded, as alleged by the appellants, that activated carbon is a form of graphite.

For this reason, the processes on activated carbon disclosed in documents D11 and D13 do not fall under the claimed process.

8.2 Appellant 1 argued that the description of the patent in suit referred to "amorphous graphite" as a type of graphite suitable for the process of claim 1.

However, the term "amorphous graphite" refers to microcrystalline graphite, which appears amorphous to the naked eye but which still maintains the crystalline structure of graphite (D18, first paragraph).

8.3 It is thus concluded that the process of claim 1, which requires a catalyst selected from one or more natural or synthetic graphites, is novel over those of D11 and D13, which are carried out over activated carbon (Article 54(2) EPC).

9. Inventive step

9.1 Closest prior art

Document D11 is the closest prior art, and discloses the preparation of HCFC-1233zd by dehydrofluorination of 244fa over activated carbon (example 1). The subject-matter of claim 1 of the fifth auxiliary request differs from the process disclosed in D11 by

virtue of the catalyst employed for the reaction.

9.2 Technical problem underlying the invention

The technical problem underlying the claimed invention as defined by the respondent is to provide an alternative process for making HCFC-1233zd by dehydrofluorination of 244fa.

9.3 Solution

The solution to this technical problem is the claimed process, characterised in that it requires a catalyst selected from one or more natural or synthetic graphites.

9.4 Success

The board agrees with the opposition division and the parties that, having regard to the data provided in example 3 of the patent in suit, the problem formulated in point 9.2 above is credibly solved by the process of claim 1.

9.5 It thus remains to be decided whether or not the proposed solution to the objective problem defined above is obvious in view of the state of the art.

There is no document on file disclosing graphite as catalyst, let alone as catalyst in a dehydrofluorination process. For this reason alone, it is concluded that the subject-matter of claim 1 is inventive, as required by Article 56 EPC.

The appellant argued that the skilled person would add graphite to the catalysts of the examples of D11 and

arrive at the claimed invention, as document D11 disclosed that graphite could be used as an additive [0050] and as a support [0051].

However, in order to arrive at claim 1 starting from example 1 of D11, the skilled person would have had to add graphite as a support to carbon black [0051] or as an additive to help granulate and shape of the catalyst [0050], which carbon black does not require.

In addition, claim 1 requires "a catalyst selected from one or more natural or synthetic graphites", whereas D11 discloses graphite as a support or an additive. Thus, even if the skilled person had considered the teaching of [0050] and [0051] of D11, he would not have arrived at the claimed invention, as claim 1 requires graphite to be present as a catalyst.

For these reasons, the appellant's arguments cannot succeed.

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 on the basis of the fifth auxiliary request filed with letter dated 26 October 2015 and a description to be adapted.

The Registrar: The Chairman:

C. Rodríguez Rodríguez P. Gryczka

Decision electronically authenticated