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Datasheet for the decision of 27 June 2014

Case Number: T 1437/12 - 3.3.10

Application Number: 07796691.9

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Title of invention:
CATALYTIC ADDITION OF HYDROFLUOROCARBONS TO FLUOROOLEFINS

Applicant:
E.I. DU PONT DE NEMOURS AND COMPANY

Headword:

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

Keyword:
Inventive step - (yes) - after amendment

Decisions cited:

Catchword:

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Case Number: T 1437/12 - 3.3.10

D E C I S I O N
of Technical Board of Appeal 3.3.10
of 27 June 2014

Appellant: (Applicant)	E.I. DU PONT DE NEMOURS AND COMPANY 1007 Market Street Wilmington, DE 19898 (US)
Representative:	Matthews, Derek Peter Dehns St Bride's House 10 Salisbury Square London EC4Y 8JD (GB)
Decision under appeal:	**Decision of the Examining Division of the European Patent Office posted on 2 January 2012 refusing European patent application No. 07796691.9 pursuant to Article 97(2) EPC.**

Composition of the Board:

Chairwoman	J. Mercey
Members:	R. Pérez Carlón
	F. Blumer

Summary of Facts and Submissions

I. The appellant (applicant) lodged an appeal against the decision of the examining division to refuse European patent application No. 07 796 691.

II. The following document was cited during examination proceedings:

D1: US 6,184,426

III. The examining division found that the subject-matter of claim 1 of the then pending main request was not novel, that claim 1 of the first auxiliary request contained added subject-matter, and that the subject-matter of auxiliary requests 2 to 6 was not inventive over document D1, which was the closest prior art. The problem of providing a process for making $CF_3CF_2CH_2F$ which minimised formation of hexafluoroethane had not been shown to be solved by the process of claim 1 of any of the then pending requests in view of the lack of fair comparative data. Thus, the problem underlying the claimed invention was reformulated as the mere provision of an alternative process for preparing $CF_3CF_2CH_2F$. The examining division considered that any pre-mixing step fell within the conceivable modifications of the closest prior art process and concluded that a process which was distinguished from D1 by a pre-mixing step was not inventive.

IV. During the oral proceedings before the board of appeal, which took place on 27 June 2014, the appellant filed a new main request, claim 1 of which reads as follows:

"*A process for making $CF_3CF_2CH_2F$, comprising:*

reacting CH_2F_2 with $CF_2=CF_2$ in the presence of SbF_5

and at least one inert solvent to produce a product mixture comprising $CF_3CF_2CH_2F$,

wherein the SbF_5, the at least one inert solvent and at least part of the CH_2F_2 are pre-mixed before contacting with the $CF_2=CF_2$,

wherein the reaction temperature is from -60°C to -10°C, and

wherein the at least one inert solvent is 1,1,1,2,2,3-hexafluoropropane."

V. The appellant argued essentially as follows:

Claim 1 of the main request found a basis in the combination of claims 1, 7, 8, 11 and 12 of the application as originally filed and the passage of the description on page 5, lines 9 to 11. Claim 2 found a basis on page 5, lines 20-21 of the application as originally filed. The requirements of Article 123(2) EPC were, thus, fulfilled.

Document D1, which failed to disclose the pre-mixing step required by claim 1, was the closest prior art. In the light of D1, the problem to be solved was to provide a process for preparing $CF_3CF_2CH_2F$ with less unwanted by-products. This problem was effectively solved in the light of the mechanism proposed and the data provided with the statement setting out the grounds of appeal, which showed that a pre-mixing step credibly reduced the amount of unwanted hexafluoroethane in the reaction product. Since document D1 was silent about this impurity and did not give any details concerning the reaction mechanism involved, the skilled person had no incentive to use a pre-mixing step, with the consequence that the subject-matter of claim 1 was inventive.

VI. The final requests of the appellant were that the decision under appeal be set aside and that a patent be granted on the basis of the main request (claims 1 and 2) as filed during the oral proceedings before the board or, subsidiarily, on the basis of any of auxiliary requests 1B, 2, 3, 3A, 3B, 4, 5, 5A and 5B, auxiliary requests 2, 3, 4 and 5 as filed with the statement setting out the grounds of appeal, auxiliary requests 1B, 3A, 3B, 5A and 5B as filed with letter dated 3 April 2014.

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

Reasons for the Decision

1. The appeal is admissible.

Amendments:

2. Claim 1 of the main request differs from claim 1 as originally filed in that the process is directed to the preparation of $CF_3CF_2CH_2F$ from difluoromethane and tetrafluoroethylene, $CF_3CF_2CH_2F$ is used as inert reaction solvent and the catalyst (SbF_5), the solvent and difluoromethane are pre-mixed before contacting with the tetrafluoroethylene.

3. Claim 1 finds a basis in the combination of claims 1, 7, 8, 11 and 12 of the application as originally filed and the passage of the description on page 5, lines 9 to 11.

Page 5, lines 9-11 of the application as originally filed discloses that at least part of RF, SbF_5 and at least one inert solvent are pre-mixed “before

contacting with $R^1R^2C=CR^1R^2$". This passage can be combined with the specific embodiment of the synthesis for preparing $CF_3CF_2CH_2F$ as disclosed in claims 1, 7, 8, 11 and 12 as originally filed, since this passage is the sole information provided in the general description of the application as filed on how to carry out the pre-mixing required by claim 8, and all Examples use this pre-mixing step for the preparation of $CF_3CF_2CH_2F$.

The general disclosure on page 5, lines 20-21 of the application as originally filed provides a basis for claim 2.

The requirements of Article 123(2) EPC are, thus, fulfilled.

Novelty

4. Claim 1 is directed to a process for preparing $CF_3CF_2CH_2F$ by reacting difluoromethane and tetrafluoroethylene over SbF_5 which includes a step of pre-mixing the SbF_5, at least part of the difluoromethane and $CF_3CF_2CH_2F$ as inert solvent before contacting with the tetrafluoroethylene.

5. Document D1 discloses a process for preparing $CF_3CF_2CH_2F$ by reacting difluoromethane and tetrafluoroethylene over SbF_5 at 50°C (see example 1 and comparative example F). However, D1 does not disclose a step of pre-mixing SbF_5, difluoromethane and $CF_3CF_2CH_2F$. The subject-matter of claim 1 is thus novel in the sense of Article 54(2) EPC.

Inventive step:

6. Closest prior art:

The board concurs with the examining division that document D1 represents the closest prior art.

As indicated above, document D1 discloses a process for preparing $CF_3CF_2CH_2F$ by reacting difluoromethane and tetrafluoroethylene over SbF_5.

D1 fails to disclose a step of pre-mixing SbF_5, at least part of difluoromethane and $CF_3CF_2CH_2F$ before contacting with the tetrafluoroethylene as required by claim 1.

7. Technical problem underlying the invention:

The technical problem underlying the claimed invention is seen in providing a process for preparing $CF_3CF_2CH_2F$ with less unwanted by-products (see page 1, lines 21 to 23 of the application).

8. Solution:

The claimed solution is a process which is characterised in that the SbF_5, $CF_3CF_2CH_2F$, and at least part of the difluoromethane are pre-mixed before contacting with the tetrafluoroethylene.

9. Success:

9.1 None of the examples/comparative examples in the application as originally filed allow a direct comparison with the closest prior art D1, since none differ from one another only by virtue of the

distinguishing feature of the claimed invention.

9.2 The appellant has, however, proposed a mechanism in the statement setting out the grounds of appeal and reproduced below, which renders it credible that the pre-mixing step required by claim 1 has an influence on the amount of hexafluoroethane obtained in the process. Said mechanism is represented by the following chemical equations:

$$F32 + SbF_5 \underset{k_{-1}}{\overset{k_1}{\rightleftarrows}} \left[CH_2{}^+SbF_6{}^-\right]$$

$$\left[CH_2{}^+SbF_6{}^-\right] + TFE \xrightarrow{k_2} CH_2FCF_2CF_3 + SbF_5$$

$$2\ TFE \xrightarrow{k_3} PTFE$$

$$TFE + SbF_5 \xrightarrow{k_4} CF_3CF_3 + SbF_3$$

$$TFE + HF \xrightarrow{k_5} CF_3CHF_2$$

whereby $[CH_2{}^+SbF_6{}^-]$ should in fact read $[CFH_2{}^+SbF_6{}^-]$, as acknowledged by the appellant at the oral proceedings before the board.

According to this mechanism, hexafluoroethane, which is an unwanted by-product, is formed by the reaction of the catalyst (SbF_5) and tetrafluoroethylene (see fourth chemical equation).

The reaction leading to the desired compound $CF_3CF_2CH_2F$ involves a first step in which difluoromethane and catalyst (SbF_5) react to form an adduct of the structure $[CFH_2{}^+SbF_6{}^-]$. This adduct then further reacts with tetrafluoroethylene leading to the desired product (first and second chemical equations above).

By forming the adduct $[CFH_2{}^+SbF_6{}^-]$, the competing reaction of SbF_5 with tetrafluoroethylene according to the fourth chemical equation and, hence, the formation

of the unwanted by-product hexafluoroethane, is minimised.

In other words, by pre-mixing the SbF_5 and at least part of the difluoromethane as required by claim 1, at least part of the SbF_5 is bound up in the form of the adduct $[CFH_2^+SbF_6^-]$ leaving less free SbF_5 to react with the subsequently added tetrafluoroethylene and form the undesired hexafluoroethane.

It is thus credible that the problem as defined above in point 7. is credibly solved by the process of claim 1.

9.3 Furthermore, with the statement setting out the grounds of appeal the appellant filed simulations based on this mechanism. From the data provided, the following conclusions can be drawn:

9.3.1 The correlation between the amount of hexafluoroethane obtained experimentally and that predicted by the simulations corroborates the proposed mechanism.

9.3.2 The data provided show that upon the basis of the proposed mechanism, carrying out the process for producing $CF_3CF_2CH_2F$ with a pre-mixing step as required by claim 1 leads to a reduction in the amount of hexafluoroethane produced.

9.4 The examining division considered that the problem as formulated in point 7 above was not effectively solved by the process of claim 1 of any of the then pending requests due to lack of experimental evidence comparing the claimed process with that disclosed in D1.

The board considers, however, that the mechanism

proposed by the appellant during the appeal proceedings, and already alluded to in the application as originally filed (page 6, lines 1 to 5), suffices to render it plausible that the problem as formulated above is indeed solved by the process of claim 1 for the reasons given in point 9.2 above.

10. Finally, it remains to be examined whether the claimed solution was obvious for the person skilled in the art:

Document D1 is silent about any impurity obtained, nor is there any explicit mention of hexafluoroethane. For this reason alone, D1 cannot provide any suggestion how to minimise the formation thereof. Furthermore, D1 does not provide any information on the reaction mechanism, let alone does it refer to the formation of an adduct between the catalyst (SbF_5) and difluoromethane. Hence, even if the skilled person had aimed to reduce the amount of the specific by-product hexafluoroethane, D1 provides no information on how it is formed, and thus no clues as to how its formation may be avoided. D1 thus provides no incentive to pre-mix the SbF_5 with at least part of the difluoromethane and $CF_3CF_2CH_2F$ as inert solvent in order to minimise the amount of hexafluoroethane relative to $CF_3CF_2CH_2F$ produced, pre-mixing also not being mentioned in D1.

For these reasons, the board concludes that the subject-matter of claim 1 is inventive in the sense of Article 56 EPC.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the examining division with the order to grant a patent on the basis of claims 1 and 2, filed as main request during the oral proceedings before the board, and a description yet to be adapted.

The Registrar: The Chairwoman:

C. Rodríguez Rodríguez J. Mercey

Decision electronically authenticated