BESCHWERDEKAMMERN DES EUROPÄISCHEN PATENTAMTS

BOARDS OF APPEAL OF THE EUROPEAN PATENT OFFICE

CHAMBRES DE RECOURS DE L'OFFICE EUROPEEN DES BREVETS

Publication in the Official Journal ~~Yes~~ / No

File Number: T 121/91 - 3.3.1

Application No.: 84 305 503.9

Publication No.: 0 146 203

Title of invention: Preparation of nitro compounds by vapor phase nitration of oxygenated hydrocarbons

Classification: C07C 76/02

D E C I S I O N
of 2 July 1992

Applicant: W.R. Grace & Co.-Conn. (a Connecticut corp.)

Headword: Nitro compounds/GRACE

EPC Articles 56, 111(1)

Keyword: "Significance of scientific literature for a technical process - solution of the technical problem over the claimed range" - "remittal for further prosecution"

Europäisches Patentamt | European Patent Office | Office européen des brevets

Case Number : T 121/91 - 3.3.1

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 2 July 1992

Appellant : W.R. Grace & Co.-Conn.
Grace Plaza
1114 Avenue of the Americas
New York
NY 10036 (US)

Representative : Collier, Jeremy Austin Grey
J.A. Kemp & Co.
14, South Square
Gray's Inn
London WC1R 5EU (US)

Decision under appeal : Decision of Examining Division of the European Patent Office dated 30 August 1990 refusing European patent application No. 84 305 503.9 pursuant to Article 97(1) EPC.

Composition of the Board :

Chairman : K.J.A. Jahn
Members : R. Spangenberg
J.-C. Saisset

Summary of Facts and Submissions

I. The Appellant is the applicant of European patent application No. 84 305 503.9, corresponding to EP-A-146 203, claiming priority of 19 December 1983 from two earlier US applications.

II. The appeal was filed on 22 October 1990, accompanied by the payment of the appropriate fee, and lies from the decision of the Examining Division of the EPO dated 30 August 1990 refusing the application.

III. The decision under appeal was based upon Claims 1 to 7 received on 16 May 1989 relating to the formation of nitroalkanes from certain oxygenated hydrocarbons. The stated ground of refusal was that the claimed process did not involve an inventive step in view of the following documents:

D1: H.B. Hass, J.A.C.S 76(1954), 2692 - 2694
D2: EP-A-0 085 328
D3: EP-A-0 004 812.

The only relevant difference between the process according to Claim 1 and the known process of D1 was seen to be that the claimed process was performed at a pressure of 5 - 20 bars. However, since the person skilled in the art knew, according to D2 and D3, that direct nitration in the vapour phase could be improved by contacting a hydrocarbon feed with nitrogen dioxide in the presence of oxygen at elevated pressure, it was obvious to also apply elevated pressure to the process according to D1, because this process, too, comprised the formation of nitroalkanes by the fission of carbon-carbon bonds.

IV. In the Statement of Grounds of Appeal, received on 9 January 1991, as well as during the oral proceedings held on 2 July 1992, the Appellant maintained his opinion that a person skilled in the art would not have considered D1 when looking for an improved technical process for the selective manufacture of nitroalkanes, since this document only described a scientific investigation which was performed with nitric acid at atmospheric pressure, and which resulted in low yields and poor selectivity. In view of the structural differences between the starting materials of the claimed process and the alkanes used in the technical processes of D2 and D3, there was no reason why the person skilled in the art would have expected that the use of the reaction conditions taught in these documents for improving the yield and selectivity of nitromethane formation from mainly propane would be equally suitable for a technical process for obtaining nitroalkanes from quite different starting materials.

V. During the oral proceedings some formal matters arising from the wording of the claims were discussed. In addition, the Board indicated that a person skilled in the art, once having decided to abandon the most readily available alkanes as starting materials and to look for promising starting compounds for a selective technical production of nitromethane, would search the scientific literature and would, therefore, consider D1, thereby obtaining a hint towards at least ethanol as a suitable starting material. In response to these objections, the Appellant filed two sets of amended claims as his main and auxiliary requests. Claim 1 of the main request reads as follows:

"A process for forming nitroalkanes and nitroaromatics which comprises contacting a C_3-C_{10} alcohol, a C_2-C_{10}

aldehyde, a C_2-C_{10} ketone or a C_4-C_{10} ether having the formula:

$R\text{-}C(R')_2\text{-}OH$, R-CHO, $R''\text{-}CH_2\text{-}O\text{-}CH_2\text{-}R'''$ or $R''\text{-}CO\text{-}R'''$

(wherein R represents an alkyl, alkaryl or aryl group, each R' separately represents hydrogen or R when R is an alkyl, and R' separately represents hydrogen when R is aryl, and R" and R"' each represents an alkyl group) with nitrogen dioxide or its precursors in a homogeneous gas phase reaction zone at an elevated pressure of 5 to 20 bars and at a temperature of from 100°C to 500°C and recovering the formed nitro compounds having the formula RNO_2, $R''NO_2$ and $R'''NO_2$ wherein R, R" and R"' are as hereinbefore defined."

In Claim 1 of the auxiliary request the starting materials did no longer comprise alcohols.

VI. The Appellant requested that the decision under appeal be set aside and that the case be remitted to the first instance for further prosecution on the basis of Claims 1 to 7 of the main or auxiliary request, both submitted during oral proceedings.

At the end of the oral proceedings the Board's decision to remit the case to the first instance for further prosecution was announced.

Reasons for the Decision

1. The appeal is admissible.

2. Main request

2.1 The present Claim 1 incorporates the subject-matter of original Claims 1 to 3. The pressure of 5 to 20 bars is disclosed on page 7, line 1 of the description as filed. The limitation of the number of carbon atoms of the alcohol starting materials finds its basis in the description as filed, page 5, lines 19 to 21. The minimum number of carbon atoms in the ether of the formula indicated must be four. The respective amendment therefore removes an obvious error. Claim 2 is supported by the disclosure on page 5, lines 1 to 3 of the description as filed. Claims 3 to 7 are based on Claims 4 to 8 as filed. Thus the requirement of Article 123(2) EPC is met.

2.2 The only question at issue is that of inventive step.

2.2.1 In the present case, it becomes clear from the introductory part of the description that the main object of the patent application is to provide a technical process for obtaining nitroalkanes. The Board can therefore accept the Appellant's point of view that it would be inappropriate, in these particular circumstances, to consider a scientific paper, which is in no way concerned with a technical process, as the closest state of the art only for the reason that it describes the use of the same starting materials as the patent application in suit. Accordingly, the Board considers that the state of the art relating to the technical production of these compounds, of which nitromethane is the technically most important, is the closest state of the art in the light of which the technical problem underlying the claimed process should be defined. These considerations lead to the conclusion that D2 and D3 are to be regarded as representing the closest state of the art. The technical processes according to these documents started

from propane or ethane. The use of the said starting materials inevitably led to a mixture of nitroalkanes and it was a long-standing problem to improve the selectivity of this process with respect to the production of in particular nitromethane (see the patent application, page 2, last paragraph).

According to the description, in particular page 3, paragraph 1 and the paragraph bridging pages 4 and 5, "selectivity" means that the process aims at producing one single or dominant nitro compound.

Thus the Board sees the technical problem underlying the application in improving selectivity in the technical production of nitroalkanes.

The patent application proposes to solve this problem essentially by using certain oxygen containing starting materials, in particular acetaldehyde and acetone, instead of propane or ethane and to react these compounds with NO_2 or its precursors, optionally in the presence of oxygen and/or water, substantially under the reaction conditions hitherto used for reacting propane or ethane. It follows from the Examples II and III that nitromethane is the only nitro compound produced from acetaldehyde and acetone and that the yields are technically acceptable. In addition, in a letter received on 1 June 1987, further test results were presented showing that with propionaldehyde as the starting material nitroethane is formed as the dominant nitroalkane product. Thus the above technical problem may be regarded as being effectively solved insofar as these types of starting materials are concerned. However, considering that

- nitric acid is a well known precursor of NO_2 (see the description, page 6, lines 8 to 12) and that therefore

the reaction conditions used in D1 differ from those used according to the application only insofar as elevated pressure is used according to the latter,

- it is disclosed in D1 (see Table I) that a mixture of products is formed from diethyl ether, 2-methyl-1-propanol and di-n-propylketone which are all suitable starting materials according to the present Claim 1, and

- no evidence is available from which the Board could infer that, contrary to what would have been expected from the disclosure in D1, only one dominant nitroalkane product is obtained from the above starting materials,

the Board is not in the position to decide the extent to which the existing technical problem has been effectively solved.

Since that matter has not yet been finally considered by the examining division, it is appropriate, in the Board's judgment, to remit the case to the examining division according to Article 111(1) EPC in order to give the Appellant the opportunity to have that matter considered by two instances.

2.2.2 However, such a remittal is only possible if the Board is satisfied that the grounds for refusal stated in the decision under appeal no longer apply to the claimed subject-matter. In the Board's judgment, this is the case here, insofar as those starting materials are concerned the use of which solves the present technical problem.

2.2.3 It has already been indicated in point V above that, in the Board's judgment, a person skilled in the art would, in principle, consider the content of D1 when looking for

suitable starting materials for the manufacturing of nitroalkanes on a technical scale, despite the fact that it is a scientific paper mainly concerned with investigating whether or not nitro derivatives of e.g. ethers, alcohols and ketones can be obtained by direct nitration of the respective starting materials (see the summary, the introductory paragraph and the experimental part). However, attention would only be paid to the information in this document which could be regarded as a promising starting point for solving the present technical problem, i.e. information relating to the selective formation of nitroalkanes. In this respect only three of the starting materials mentioned in Table I are relevant, namely dimethyl ether, methyl tert.butyl ether, and ethanol, since, as indicated in Table I, all other compounds investigated in D1 yield mixtures of products. After the exclusion of ethanol, none of these compounds is a starting material according to the process as claimed.

2.2.4 A further compound, acetone, is said to yield nitromethane and probably nitroacetone. However, in the experimental section on the top of the right-hand column of page 2694, it is explained that any nitroacetone so formed decomposed during its attempted separation. Thus, in the sense of the definition of selectivity given in the patent application (see point 2.2.1 above) acetone may be also regarded as "selectively" yielding nitromethane. Nevertheless, in the Board's judgment, the expectation of selectivity alone is not sufficient to suggest to a person skilled in the art using a starting material for the technical production of nitromethane, since for such a suggestion to be seriously considered a reasonable expectation of a technically acceptable yield must also have existed. In this respect the document contains the information that "yields for the most part were low, but no attempts were made to improve them. Yields of nitro compounds were about 20% in the case

of ethers and ketones (except acetone) when calculated upon the compound being nitrated. Acetone, the alcohols and propionic acid gave yields of 5% or less." Although, in the Board's judgment, a yield in the order of magnitude of 5%, calculated on the product to be nitrated lies within the range of yields obtainable by the method of the application in suit and is therefore to be regarded as being technically acceptable, the Board is not satisfied that the information of D1 as a whole, taking into account that not only nitromethane is formed but also an unspecified amount of nitroacetone which decomposed to compounds other than nitromethane, provided a reasonable expectation that acetone would be suitable as a starting material for a technical process for obtaining nitromethane.

2.2.5 With respect to the use of diethyl ether and acetaldehyde as starting materials the decision under appeal was based on the assumption that a person skilled in the art would infer from the disclosure of D1, in particular from the alleged fact that ethanol and acetone yielded exclusively nitromethane, that other oxygenated C_2-derivatives would also yield exclusively nitromethane. However, diethyl ether, which is also an "oxygenated C_2-derivative" according to the definition underlying the decision under appeal, is said in Table I of D1 to yield not only nitromethane but also nitroethane and ß-nitroethyl ethyl ether. Thus, the above assumption is in the Board's judgment not supported by the state of the art.

2.2.6 For those starting materials which contain carbon chains longer than two carbon atoms, e.g. propionaldehyde, it follows immediately from what is stated in point 2.2.1 above that the formation of only one single dominant nitroalkane product could not be expected, since otherwise

no doubt would have existed as to whether the stated problem has been solved.

2.2.7 Thus the Board is satisfied that the reasons for refusal are no longer applicable to the subject-matter of the claims according to the main request to the extent indicated in point 2.2.2 above.

3. In these circumstances, there is no need to consider the auxiliary request. However, since the allowable scope of the statement of claim cannot at present be established (see item 2.2.1. above), in the Board's judgment, it is not appropriate to indicate a particular set of claims for further prosecution.

Order

For these reasons, it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the first instance for further prosecution.

The Registrar: The Chairman:

E. Görgmaier K. Jahn