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D E C I S I O N
of 19 June 2002

Case Number: T 0313/99 - 3.3.1

Application Number: 93202018.3

Publication Number: 0579323

IPC: C07C 409/14

Language of the proceedings: EN

Title of invention:

Process for the preparation of cyclohexyl hydroperoxide

Patentee:

DSM N.V.

Opponent:

RHODIA POLYAMIDE INTERMEDIATES

Headword:

Cyclohexyl hydroperoxide/RHODIA POLYAMIDE INTERMEDIATES

Relevant legal provisions:

EPC Art. 54(1), 56

Keyword:

"Novelty (yes)"

"Inventive step (yes) - non-obvious solution"

Decisions cited:

-

Catchword:

-



Case Number: T 0313/99 - 3.3.1

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 19 June 2002

Appellant: RHODIA POLYAMIDE INTERMEDIATES
(Opponent) Avenue Ramboz
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Representative: ESSON, Jean-Pierre
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Respondent: DSM N.V.
(Proprietor of the patent) Het Overloon 1
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Representative: -

Decision under appeal: Decision of the Opposition Division of the
European Patent Office posted 26 January 1999
rejecting the opposition filed against European
patent No. 0 579 323 pursuant to Article 102(2)
EPC.

Composition of the Board:

Chairman: J. M. Jonk
Members: P. P. Bracke
S. C. Perryman

Summary of Facts and Submissions

- I. The appeal lies from the Opposition Division's decision, dispatched on 26 January 1999, rejecting the opposition against European patent No. 0 579 323, which was granted on the basis of Claims 1 to 14.

The sole independent claim as granted read:

"1.Process for the preparation of cyclohexyl hydroperoxide by converting cyclohexane into a mixture consisting substantially of 0.5-8 wt.% of cyclohexyl hydroperoxide and 0.1-4 wt.% of cyclohexanol and cyclohexanone in cyclohexane, this being effected using an oxygen-containing gas at a temperature between 130 and 200°C and a pressure between 4 and 50 bar during 0.05 to 14 hours in the absence of catalysts, and optionally subjecting the mixture after the reaction to partial expansion, this process being characterized in that 0.1 to 3 wt.% of oxidic products with linear or cyclic alkyl chains with 1-6 carbon atoms are present in the cyclohexane at the start of the oxidation reaction."

- II. The Opposition Division held in particular that the claimed process essentially differed from the known processes by the presence at the start of the oxidation reaction of 0.1 to 3 wt.% of oxidic products as defined in the characterizing part of Claim 1. Moreover, it held that the claimed process involved an inventive step, since it was not suggested in the prior art that the presence of 0.1 to 3 wt.% of such oxidic products at the start of the oxidation reaction would result into a reaction accelerating effect.

III. Oral proceedings before the Board took place on 19 June 2002.

IV. The Appellant submitted that document

(6) US-A-2 223 494,

which was filed with the statement setting out the grounds of appeal, was novelty destroying for the claimed process.

Furthermore, the Appellant was of the opinion that the claimed process was obviously derivable from the teaching of document

(3) FR-A-2 119 397

in combination with documents (6) and

(2) Journal of Organic Chemistry, 41, pages 1 to 10, 1976,

since it was known from document (3) that hydrocarbons, such as cyclohexane, may be converted into their hydroperoxides at the temperature and pressure as defined in present Claim 1, since it was known from document (2) that cyclohexanone accelerates the oxidation of cyclohexane and since it was known from document (6) that the start of the oxidation was promoted by using 0.1 to 0.5 wt-% of an initiator such as cyclohexanone.

V. The Respondent contested that document (6) would be novelty destroying for present Claim 1, since it neither disclosed a process for preparing cyclohexyl

hydroperoxide nor an oxidation process in the absence of a catalyst.

Concerning inventive step, the Respondent argued that a skilled person would not combine the teaching of document (6) with the teaching of document (3) because document (6) concerned the preparation of cyclic alcohols and cyclic ketones by conducting the oxidation in the presence of a catalyst whereas document (3) concerned the preparation of hydroperoxides in the absence of a catalyst. Moreover, he submitted that document (2) was concerned with the oxidation of cyclohexane into cyclohexyl hydroperoxide in the presence of specific initiators, which differed from those of present Claim 1, and that it neither suggested the reaction temperature nor the amounts of initiator as claimed in present Claim 1.

VI. The Appellant requested that the decision be set aside and that the patent be revoked.

The Respondent requested that the appeal be dismissed and that the patent be maintained.

VII. At the conclusion of the oral proceedings the Board's decision was pronounced.

Reasons for the Decision

1. The appeal is admissible.

2. *Novelty*

After examination of the cited prior art documents, the

Board has reached the conclusion that the subject-matter of Claims 1 to 14 is novel over the teachings of the cited prior art documents, since none of those documents discloses a process of oxidizing cyclohexane in the absence of a catalyst wherein 0.1 to 3 wt.% of oxidic products with linear or cyclic alkyl chains with 1 to 6 carbon atoms are present in the cyclohexane at the start of the oxidation reaction.

In this context, the Board observes that Claim 1 of the patent in suit concerns an oxidation process for preparing hydroperoxides **in the absence of a catalyst**, whereas document (6) merely describes an oxidation process **in the presence of a catalyst**. Thus, already on the basis of this difference the claimed subject-matter must be considered as being novel.

3. *Inventive step*

3.1 In accordance with the "problem-solution approach" applied by the Boards of Appeal to assess inventive step on an objective basis, it is necessary to establish the closest state of the art being the starting point, to determine in the light thereof the technical problem which the invention addresses and solves, and to examine the obviousness of the claimed solution to this problem in view of the state of the art.

3.2 According to the established jurisprudence of the Boards of Appeal the "closest state of the art" is normally a prior art document disclosing subject-matter aiming at the same objective as the claimed invention and having the most relevant technical features in common.

Consequently, the Board considers in agreement with both Parties that document (3) represents the closest prior art.

3.3 Document (3) discloses the uncatalysed oxidation of hydrocarbons, such as cyclohexane, in high selectivity and high conversion rates in the presence of both a tertiary alcohol as stabiliser and a tertiary hydroperoxide as initiator. The oxidation is carried out at 80 to 180°C and at 1 to 20.7 bar (1 to 20.4 atm) in a molar range of tertiary alcohol to hydrocarbon of 0.05:1 to 1.5:1 and a molar ratio of tertiary hydroperoxide to hydrocarbon of 0.01:1 to 0.3:1 (page 2, line 27 to 33, page 3, lines 6 to 8, page 4, lines 13 to 18, and example 1).

3.4 Regarding this closest state of the art, the Respondent submitted that by applying the claimed process a high selectivity for the hydroperoxide product combined with a high conversion rate of the cyclohexane starting compound is achieved, while the use of a tertiary alcohol and a tertiary hydroperoxide can be avoided.

3.5 Thus, in view of these submissions, which have not been contested by the Appellant, it is the Board's position that the technical problem underlying the patent in suit is the provision of a further process for preparing cyclohexane hydroperoxide in high yields and selectivity at relatively high conversion rates, without the need of a tertiary alcohol and a tertiary hydroperoxide (see also column 2, line 55 to column 3, line 1 of the patent in suit).

3.6 According to present Claim 1 this technical problem is solved by providing a process which is essentially

characterised by the use of 0.1 to 3 wt.% oxidic products with linear or cyclic alkyl chains with 1 to 6 carbon atoms at the start of the oxidation reaction, ie as an initiator.

- 3.7 Having regard to the technical information provided in the patent in suit, in particular in the examples, the Board considers it plausible that the technical problem as defined above has been solved. This was not disputed by the Appellant.
- 3.8 In assessing inventive step the question now is whether a skilled person starting from document (3), and having knowledge of documents (2) and (6), would arrive at the solution of the above defined technical problem as claimed.
- 3.9 The Appellant argued that it would be obvious to the skilled person to replace the initiator used in document (3) by cyclohexanone in view of documents (2) and (6).
- 3.10 Document (2) discloses the oxidation of cyclohexane and its oxidation products. In its introduction concerning earlier research in this technical field it indicates the difference between an uncatalysed oxidation giving the hydroperoxide as the major product and a catalysed process, such as caused by the steel vessel wall of the reactor, where peroxide is apparently decomposed and cyclohexanone is achieved as the major product (see page 1, left-hand column, last paragraph to the right-hand column, line 1). In this context it also discloses that "Adding cyclohexyl hydroperoxide initially to a cyclohexane oxidation at 145° in steel had no significant effect on the reaction; however, if

cyclohexanone is added in amounts found normally after about 10 hr of reaction, the reaction begins at the maximum rate that normally would be delayed for several hours" (page 1, right-hand column, lines 1 to 6). This teaching therefore concerns an oxidation reaction in a steel vessel functioning as a catalyst, wherein consequently cyclohexanone is achieved as the main oxidation product. It does not provide any information to the skilled person that the addition of cyclohexanone at the start of the reaction in an uncatalysed oxidation reaction would solve the technical problem as defined above.

Furthermore, it discloses the results of investigations relating to the oxidation of cyclohexane in a glass vessel, ie in the absence of a catalyst, in the presence of di-tert.butyl peroxide, N,N'-azobis(isobutyronitrile) or N,N'-azobis(cyanocyclohexane) as initiator. This teaching does not provide any incentive to the use of an initiator as claimed in Claim 1 of the patent in suit.

It is true that it is indicated by referring to Table I that in an experiment using N,N'-azobis(1-cyanocyclohexane) as initiator the addition of cyclohexanone in an amount of about 2% accelerates the conversion rate of cyclohexane by a factor of 3 (see under "Preliminary Oxidations" in the right-hand column of page 2). However, this does not suggest to the skilled person that cyclohexanone could be used as an initiator as such, let alone that it would lead to the forming of cyclohexane hydroperoxide as the major product. Actually, it follows from Table I, last experiment, that said addition of cyclohexanone results in the forming of cyclohexanone in an amount of about

five times that of cyclohexane hydroperoxide. Moreover, the statements in the right-hand column on page 9: "The accumulating ketone also catalyzes the decomposition of hydroperoxide into radicals, increasing the rate of oxidation, regenerating ketone, and producing cyclohexanol, which is readily oxidized to more ketone. Cyclohexanol also accelerates decompositions of hydroperoxide" actually lead away from the use of cyclohexanone and cyclohexanol for the solution of the above defined technical problem.

3.11 Document (6) concerns the production of cyclic alcohols and cyclic ketones in good yields by oxidation of cyclic saturated hydrocarbons in the presence of a catalyst (see page 1, left column, lines 6 to 31). Moreover, it discloses that at temperatures of 170°C, or below, the yields and efficiency of the process, ie the production of cyclic alcohols and cyclic ketones, may be even further improved by performing the oxidation in the presence of an initiator, such as cyclohexanone, in amounts of about 0.3 % by weight (see page 1, left column, line 53 to the right column, line 53, as well as Example 1 relating to the production of cyclohexanol and cyclohexanone at a temperature of 145°C and an elevated pressure). Since this document does not relate to the preparation of cyclohexyl hydroperoxide at all, and actually concerns a different technical problem, in the Board's judgment, the skilled person would not have any reason to modify the process of document (3) by replacing the initiators used therein.

3.12 In this context, the Board observes that the skilled person in view of the fact that - as submitted by the Appellant and not contested by the Respondent - it was

known that the oxidation of cyclohexane to cyclohexanol and cyclohexanone involves the forming of cyclohexyl hydroperoxide as an intermediate compound in a first step **could** have taken the use of an initiator as disclosed in document (6) into consideration for preparing cyclohexyl hydroperoxide. However, according to the established case law of the Boards of Appeal for determining lack of inventive step, it is necessary to show that considering the teaching of the prior art as a whole, without using hindsight based on the knowledge of the claimed invention, the skilled person **would** have arrived at the claimed solution of **the technical problem to be solved**. However, as indicated above, a skilled person, when trying to solve the present technical problem underlying the patent in suit, would not have had any reason to replace the initiators of document (3) by those as defined in present Claim 1 in order to provide a process for preparing cyclohexyl hydroperoxide in high yields and selectivity at relatively high conversion rates.

3.13 Therefore, the Board comes to the conclusion that the process according to Claim 1 is not obviously derivable from the cited prior art.

3.14 Claims 2 to 14, which represent preferred embodiments of Claim 1, derive their patentability from the same inventive concept.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:

N. Maslin

J. Jonk