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Datasheet for the decision of 18 September 2007

T 1044/05 - 3.3.01 Case Number:

Application Number: 00202658.1

Publication Number: 1072600

IPC: C07D 301/12

Language of the proceedings: EN

Title of invention:

Process for the preparation of Epoxides

Patentee:

DOW GLOBAL TECHNOLOGIES INC.

Opponent:

Evonic Degussa GmbH

Headword:

Olefin epoxidation/DOW GLOBAL TECHNOLOGIES

Relevant legal provisions:

EPC Art. 54, 56, 100(a)(b), 114(2), 123(2)(3)

Keyword:

"Main request: sufficiency of disclosure (no) - no possibility to determine an essential parameter of the claimed process" "First auxiliary request: objection under Article 100(b) EPC not admitted as extending the scope of the opposition amendments supported by the application as filed (yes) novelty (yes) - inventive step (yes) - non obvious solution"

Decisions cited:

T 0409/91, T 0435/91, T 1018/02, T 1140/02

Catchword:



Europäisches Patentamt

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Beschwerdekammern

Boards of Appeal

Chambres de recours

Case Number: T 1044/05 - 3.3.01

DECISION
of the Technical Board of Appeal 3.3.01
of 18 September 2007

Appellant: DOW GLOBAL TECHNOLOGIES INC.

(Patent Proprietor) Washington Stret, 1790 Building

Midland, MI 48674 (US)

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Respondent: Evonic Degussa GmbH (Opponent) Rodenbacher Chaussee 4 D-63457 Hanau (DE)

Representative: -

Decision under appeal: Decision of the Opposition Division of the

European Patent Office posted 7 June 2005 revoking European patent No. 1072600 pursuant

to Article 102(1) EPC.

Composition of the Board:

Chairman: A. Nuss Members: P. Ranguis

D. Rogers

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Summary of Facts and Submissions

- I. The appeal lies from the decision of the Opposition Division to revoke the European patent No. 1 072 600 (European patent application No. 00202658.1).
- II. Notice of opposition had been filed by the Respondent (Opponent) requesting revocation of the patent as granted in its entirety on the ground of insufficiency of disclosure (Article 100(b) EPC), or lack of novelty or inventive step (Article 100(a) EPC). The following documents were cited in the opposition proceedings.
 - (1) "Grundlagen der allgemeinen und anorganischenChemie", Verlag Sauerländer, 1977, pages 355-376,
 - (2) EP-B-0 712 852
 - (3) EP-B-0 757 045
 - (4) US-A-5 374 747
 - (5) WO-A-99/48882, state of the art under Article 54(3)(4) EPC
 - (6) EP-A-0 795 537
 - (7) Aldrich Katalog, 1996, page 109
- III. The patent in suit comprises twenty five claims.
 Claim 1, the sole independent claim, reads as follows:
 - "1. A continuous process for the preparation of olefin oxides by the direct epoxidation of an olefin with hydrogen peroxide, or compounds capable of producing hydrogen peroxide under the reaction conditions, in a solvent medium, in the presence of a catalytic system consisting of a zeolite containing titanium atoms and a buffer system with a pH controlled within values

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ranging from 5.5 to 8.0, consisting of a nitrogenated base and a salt thereof with an organic or inorganic acid".

Dependent Claims 19 and 22 read as follows:

- "19. The process according to Claim 1, wherein the solvent medium is selected from alcohols, hydro-alcohol mixtures, ketones, ethers, aliphatic and aromatic hydrocarbons, halogenated hydrocarbons, esters or glycols."
- "22. The process according to Claim 19, wherein the ethers are selected from tetrahydrofuran and butyl ether."
- IV. In its decision, the Opposition Division held that Claim 1 interpreted in combination with the description required the use of a buffer system such that the pH of the entire reaction system is in the range of 5.5 to 8.0. Regarding the objected lack of sufficiency of disclosure, the Opponent had not shown that from the wording of Claim 1 it was not possible to reach a pH controlled within the values ranging from 5.5 to 8.0. Sufficient examples were present in that respect in the description for enabling the person skilled in the art to reproduce the invention.

None of the documents (2), (3) or (5) could anticipate the subject-matter of Claim 1. Document (2) did not disclose the specific pH range of Claim 1. Example 11 might seem to describe a buffer system but the pH reached was not defined. Document (3) did not mention any specific pH range. No mention could be found in

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document (5) that the reaction system might be controlled with a buffer system.

Regarding inventive step, in view of document (3) as the closest prior art, the technical problem to be solved might be seen in the provision of an improved process for the preparation of olefin oxides by the direct epoxidation of an olefin with hydrogen peroxide, so that the catalytic activity is kept stable over a certain period of time and the epoxides are prepared with high yields and selectivities. From the examples disclosed in document (3), it could be concluded that the chelating agent had no influence on the selectivity of the catalyst. Furthermore, since acetic acid, formic acid and ammonium hydroxide were added to the reaction mixture, a buffer system was formed. From the examples, it was also apparent that the higher the amount of ammonia used, the better was the propylene oxide selectivity achieved. Thus, the teaching of document (3) in combination with that of document (6) dealing with the same kind of process wherein the pH had to be kept between 5 and 7, led obviously to the claimed process.

- V. In the course of the appeal proceedings, the Appellant (Proprietor of the patent) submitted with a letter received on 3 July 2006 a further set of twenty three claims as first auxiliary request. Claim 1, the sole independent claim reads as follows:
 - "1. A continuous process for the preparation of olefin oxides by the direct epoxidation of an olefin with hydrogen peroxide, or compounds capable of producing hydrogen peroxide under the reaction conditions, in a

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solvent medium selected from alcohols, hydro-alcohol mixtures, ketones, esters and glycols, in the presence of a catalytic system consisting of a zeolite containing titanium atoms and a buffer system with a pH controlled within values ranging from 5.5 to 8.0, consisting of a nitrogenated base and a salt thereof with an organic or inorganic acid".

- VI. In addition to documents (1) to (7), the parties have submitted in the course of the appeal proceedings the following documents as evidence
 - (8) Fundamentals of Analytical Chemistry, 2nd edition, 1969, pages 253 to 256 and 285-286,
 - (9) Supplemental data related to Example 5 of the patent in suit submitted by the Appellant (Proprietor of the patent),
 - (10) Criteria for standardization of pH measurements,

 Pure and Appl. Chem., 57 (1985), pages 865 to 876,
 - (11) Analysis and evaluation of the examples of document (3), i.e. EP-A-757 045, submitted by the Respondent (Opponent),
 - (12) Calculations of pH of the solutions set up in document (3), submitted by the Appellant.
- VII. Oral proceedings before the Board took place on 18 September 2007. The Appellant submitted at the hearing new pages 2 to 7 of the description aiming at adapting the description as granted to the amended set of claims submitted in the written proceedings (see point V above).
- VIII. The arguments of the Appellant may be summarized as follows:

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As established by the Opposition Division, the pH range defined in Claim 1, i.e. 5.5 to 8.0, referred to the pH of the reaction medium, not the pH of the nitrogenated buffer system.

The patent in suit disclosed the invention in a manner sufficiently clear and complete. Contrary to the view expressed by the Respondent, document (10) showed that standard buffer systems were available for standardizing a pH probe in non-aqueous solvents. Furthermore, the pH representing the acid strength, in aqueous systems, this was the same as the negative logarithm of the H⁺ ion concentration and the pH could simply be measured using a pH meter. For systems not containing water, the pH or the acid strength could be measured using similar techniques, well-known to the skilled person, for instance other potentiometric measurements using electrodes. A pH meter was also a potentiometer with which the potential was displayed directly on a scale in pH units. For non-aqueous solvents, the person skilled in the art had all the necessary knowledge to calculate the acid strength on the basis of the potentiometric measurement carried out. While the person skilled in the art might expect some error in the pH measurement due to the non-aqueous system (the art generally refers to an "apparent" pH), the measurement was nevertheless simple and practically effective, as evidenced in the results set out in the patent in suit.

The patent in suit was solely opposed under

Article 100(b) EPC insofar as Claims 19 and 22 related
to a solvent medium selected from ethers, aliphatic and

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aromatic hydrocarbons and halogenated hydrocarbons. As a consequence of the amended claims filed by the Appellant with the letter dated 3 July 2006, the subject-matter of the first auxiliary request overcame this objection.

With regard to novelty, contrary to the finding of the Opposition Division, document (2) did not relate to buffered systems. As evidenced by document (8), solutions of ammonium acetate were classified in the acid-base category of "Salt of a Weak Acid and a Weak Base", which differed from the category of buffer solutions. Document (5) did not directly and unambiguously described a continuous process wherein a buffer was used consisting of a nitrogenated base and a salt thereof with an organic or inorganic acid.

Regarding inventive step, document (3) taught the epoxidation reaction in continuous operation, in the presence of a basic salt (ammonium hydroxide) and a chelating agent for the purpose of retarding catalyst deterioration as measured by non-selective hydrogen peroxide decomposition. The pH of the epoxidation process was not mentioned nor was used a buffer system. The calculations submitted as evidence, i.e. document (12) showed that the pH of the solution set up in Example 3 ranged from 2.7 to 4.0 and that no free ammonia was present.

Contrary to the finding of the Opposition Division that the improved propylene selectivity only resulted from an increased quantity of ammonium hydroxide, the chelating agent was not without influence in that respect. At the same time as ammonium hydroxide was

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increased, time-on-stream was significantly decreased from 350 hours in Example 1 to 230 hours in Example 3. Decreasing time-on-stream could account for improved selectivity.

It had been unexpectedly found that by using the claimed nitrogenated buffer system to control the pH of the entire reaction system between 5.5-8.0 one could obtain simultaneously high hydrogen peroxide conversion, high propylene oxide selectivity and extended catalyst lifetime. Document (3) taught to use a chelating agent to solve the aging problem. Such chelating agent was not required by the present invention.

Furthermore, the experimental results submitted, i.e, document (9), showed that in a test performed in the conditions disclosed in Example 5 of the patent in suit, the conversion and propylene oxide selectivity remained very high at 95.0 percent and 98.0 percent respectively up to 1,000 hours run time at pH 6.3. By contrast, in Example 4 of document (3), at the longest time-on-stream (630 hours), the referenced process had degraded to a propylene conversion of 69.0 percent and a propylene oxide selectivity of 86.0 percent.

Neither was a combination of document (3) with documents (6) and (2) suitable for bringing the person skilled in the art to the claimed invention.

Document (6) did not disclose the same kind of process and would not have been considered by the person skilled in the art. In particular, document (6) did not address the aging problem and did not relate to a

continuous process. It only taught batch processes wherein the pH was adjusted by successive addition of bases and acids. Document (2) did not relate to a continuous process, did not refer to the aging problem and did not teach to use a buffer either.

IX. The arguments of the Respondent may be summarized as follows:

Contrary to the Opposition Division's finding, the pH range of 5.5 to 8.0 was clearly and unambiguously related to the "buffer system" and not to the entire reaction system on a proper understanding of Claim 1 as granted. It could not be referred to the description for giving another meaning to a clear feature. It was not correct to refer to document (8) to allege that the molar ratio of the nitrogenated base and the salt thereof with an acid ranged from 1:10 to 10:1 since in that case the subject-matter of Claims 2 and 3 would have been outside the scope of Claim 1.

The subject-matter of the patent in suit as reflected by Claims 19 and 22, wherein the process is performed in a solvent medium such as ethers, aliphatic and aromatic hydrocarbons and halogenated hydrocarbons, gave rise to objection under Article 100(b) EPC.

Neither the general description nor the examples gave the person skilled in the art the relevant information to determine a pH value in such solvents where the solubility of water was lower than 10%. Document (10) in that respect showed that direct pH measurement in organic solvents was immeasurable and had to be defined operationally, namely, in terms of the operation or method used to determine it. However, the patent in

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suit did not provide any information about the process used for measuring the pH and, therefore, no calibration was possible. The person skilled in the art could not determine the pH in the solvent medium defined above and encompassed by Claims 19 and 22, so that the process according to these claims which required that the pH be maintained between 5.5 and 8.0 could not be reproduced.

Furthermore, the Appellant admitted that the measure of pH in the examples of the patent in suit had been made with an electrode which had not been calibrated in an hydro-alcoholic medium but in an aqueous medium. Such a method was not according to the common rules in this art. Indeed, it led to a deviation with respect to the pH measured by an electrode having been calibrated in an hydro-alcoholic medium. With a mixture methanol/water of 84/16, in a buffer system of 0.05 mol/kg of potassium hydrogenophthalate, the pH value in document (10), Table I, was 6.236, whereas the pH value was 6.11 when the electrode was calibrated in aqueous medium. Moreover, the pH value of the methanol/water 84/16 medium in a buffer system 0.05 mol/kg ammonia and 0.05 mol/kg ammonium acetate was 8.88 when the electrode was calibrated in an aqueous medium. This was clearly beyond the upper limit of pH as defined in Claim 1 of the first auxiliary request. The Appellant contended that the examples were an empiric evidence that it was possible to produce a buffer system consisting of a nitrogenated base and its salt thereof for obtaining a reaction mixture with a pH between 5.5 and 8.0. However, that contention relied on the fact that the buffer system remained unchanged in the course of the process whereas acidic by-products

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were produced which reacted with the nitrogenated base so that no conclusion regarding the amount of base could be drawn. Moreover, in the case of a buffer consisting of ammonia and ammonium acetate, i.e.

Example 4 of the patent in suit, the ratio of nitrogenated base to salt thereof, for a pH of 6.4 to 6.5, was 1:240 to 1:300. Given that the epoxidation reaction produced carboxylic acids, all the nitrogenated base was consumed resulting in a buffer consisting of carboxylic acids and ammonium salts thereof and not the buffer defined in Claim 1. One of the examples referred, therefore, to a buffer consisting of acetic acid and acetate salt thereof in a pH range from 6 to 6.5.

Regarding novelty, document (2) disclosed a process for preparing propylene oxide by epoxidation of propylene wherein a titanium silicalite is utilized in the presence of hydrogen peroxide. The epoxidation reaction could be carried out in a continuous manner. The selectivity of the epoxidation reaction could be improved through adding a non basic salt such as ammonium acetate, as set out in Example 11. From document (7) it was known that aqueous solutions of ammonium acetate had a pH of 7. It was also clear from document (1) that an aqueous solution of ammonium acetate contained a low amount of ammonia and acetic acid of 0.55 weight% or so. For the person skilled in the art it emerged unambiguously from Example 11 of document (2), wherein an aqueous solution of ammonium acetate was used at pH 7, that free ammonia was also present, anticipating, therefore the claimed subjectmatter.

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Document (5) disclosed a process for the preparation of olefine oxide by epoxydation of olefins with hydrogen peroxide in a solvent medium in the presence of zeolite containing titanium atoms, wherein the pH of the reaction mixture was within the range from 4.8 to 6.5. In a preferred form the pH value was maintained in the said range through addition of a weak base. As a weak base, ammonia (and not NH₄OH as wrongly cited in this document) was mentioned. For the person skilled in the art, the addition of NH₃ implied necessarily that a salt of ammonium was formed due to the acids present at the surface of the catalyst. It derived therefrom that document (5) involved the same buffer system as the patent in suit when the nitrogenated base was NH₃.

Regarding inventive step, document (3) disclosed a continuous process for the preparation of propylene oxide by direct epoxidation of propylene with hydrogen peroxide in an hydro-alcohol solvent in the presence of a zeolite containing titanium. The mixture comprised in addition ammonia, acetic acid and amino acids, which together formed a buffer system comprising a nitrogenated base and a salt thereof with an organic acid and which corresponded to the buffer defined in the patent in suit. The addition of a chelating agent allowed to avoid the non-selective hydrogen peroxide decomposition to oxygen, liable to gradually deteriorate the performance of the titanium-containing molecular sieve catalyst. The sole feature not disclosed in this document was the pH value range within 5.5 to 8.0.

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This document represented the closest prior art. In view of document (3) as the closest prior art, no improvement could be acknowledged in the absence of a proper comparison between the claimed subject-matter and the process disclosed in document (3).

In order to define properly the technical problem to be solved, it was crucial to point out that document (3) as properly understood by the Opposition Division disclosed two independent not interrelated technical effects. On the one hand, the improved selectivities and high yields were solely achieved by the increasing amount of ammonium hydroxide. Contrary to the Appellant's contention, the chelating agent played no role in that respect. The adding of chelating agents only aimed to avoid the non-selective hydrogen peroxide decomposition to oxygen. It was, therefore, not proper to mix up the two effects.

The technical problem, therefore, could only be seen in the provision of a further process allowing the preparation of epoxides with high yields and selectivities.

As recognized by the Opposition Division, Table I of document (3) showed that it was known that increased amounts of ammonia improved the propylene oxide selectivity.

The person skilled in the art would have been, therefore, directed to bring the pH in the range as defined in Claim 1, by simple routine experiments, in order to obtain a better selectivity. This finding was confirmed by the teaching of document (6) which

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recommended to keep the pH value range within 5 and 7 by addition of ammonia to the reaction mixture for preparing with high yields and selectivity 2-butene-1,4-diesters by epoxidation with hydrogen peroxide in the presence of zeolite containing titanium.

Regarding the first auxiliary request, amended Claim 1 extended beyond the content of the application as filed in contravention with the requirements of Article 123(2) EPC. The description as originally filed did not contain glycol in the list of solvents. Furthermore, Claim 19 as originally filed defined at the same level a list of solvents selected from alcohols, hydro-alcohol mixtures, ketones, ethers, aliphatic and aromatic hydrocarbons, halogenated hydrocarbons, esters and glycols. The selection among those solvents of preferred solvents represented an additional choice not directly and unambiguously derivable from the application as filed.

X. The Appellant requested that the decision under appeal be set aside and the patent be maintained as granted or on the basis of the set of twenty three claims submitted as a first auxiliary request with a letter received on 3 July 2006 and amended page(s) 2-7 of the description submitted at the oral proceedings on 18 September 2007.

The Respondent requested that the appeal be dismissed.

XI. At the end of the oral proceedings the decision of the Board was announced.

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Reasons for the Decision

1. The appeal is admissible.

Main request

2. With respect to Claim 1 as granted, the parties have conflicting views concerning what the pH range 5.5 to 8.0 refers to. The Appellant contended that the pH range referred to the pH of the reaction medium. The Respondent argued that this pH range referred to the buffer system itself.

On the one hand, accepting the Respondent's view that the pH range of 5.5 to 8.0 was clearly and unambiguously related to the "buffer system" and not to the entire reaction system, makes little, if any sense, as admitted by him in the written submissions. A buffer system cannot indeed consist of a nitrogenated base and a salt thereof, excluding thereby any other component, whereas a pH value is generally defined for aqueous solutions. Since a claim cannot be read in a way which does not make sense, the meaning given by the Respondent to the pH value is to be rejected (see T 1018/02, point 3.8).

On the other hand, the Board observes that in Claim 1 the term "controlled" following the expression "a buffer system with a pH" (see point III above) can only refer to the buffered **solution** which undergoes the reaction process and the pH of which is "controlled" by the means of a buffer system. The Board concludes, therefore, that on a proper reading Claim 1 requires a buffer system such that the pH of the entire reaction

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system is in the range of 5.5 to 8.0. Since Claim 1 is clear there is no need in that respect to refer to the description.

- 3. Article 100(b) EPC
- 3.1 The Respondent submitted that the subject-matter of the patent in suit as embodied by Claims 19 and 22, wherein the process is performed in a solvent medium selected from ethers, aliphatic and aromatic hydrocarbons and halogenated hydrocarbons, gave rise to objection under Article 100(b) EPC on the ground that the solubility in water of those solvents being lower than 10%, no method was disclosed for measuring the pH.
- An European patent gives rise to objection under Article 100(b) EPC if it does not disclose the invention in a manner sufficiently clear and complete for it to be carried out by the person skilled in the art. It is established jurisprudence of the Boards of Appeal that the requirements of sufficiency of disclosure are only met if the invention as defined in the independent claim can be performed by a person skilled in the art without undue burden, having regard to further information given in the patent in suit and using common general knowledge (see decisions T 409/91, OJ EPO 1994, 653, point 3.5 and T 435/91, OJ EPO 1995, 188, point 2.2.1).
- 3.3 Since the process for the preparation of olefin oxides as defined in Claim 1 requires that the pH be controlled within values ranging from 5.5 to 8.0, it becomes clear that the implementation of the process presupposes that the value of the pH can be determined.

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This implies that the person skilled in the art can find in the patent in suit or with the help of his common general knowledge the method and the conditions necessary for determining this parameter since he would otherwise be at a loss when trying to choose the process conditions in order to obtain the desired result.

- Although the patent in suit is silent regarding the method used for measuring the pH, in particular in methanol/water media present in all the examples, the Board accepts that the person skilled in the art is acquainted with standardized methods for measuring the pH based on the activity of the H⁺ ion in aqueous medium. For systems not containing water, the Appellant contended that the pH or the acid strength could be measured using similar techniques, well-known to the skilled person, for instance other potentiometric measurements using electrodes.
- 3.5 However, document (10) relating to "Criteria for standardization of pH measurements" points out that it is well known that the response of the usual H⁺-sensing electrodes often becomes problematic, if it exists at all, when the last traces of water are removed from solvent mixtures with certain co-solvents where, inter alia, it would be rather difficult to asses protolytic equilibria susceptible of pH-metric standardisation (see paragraph 2.2, page 867).
- 3.6 Already for this reason, it is rather unlikely that the person skilled in the art could use, for the above cited solvent medium (see point 3.1 above), the

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standardized methods for measuring the pH in aqueous medium.

- 3.7 Since the patent in suit moreover is totally silent with regard to a possible method of measuring the pH in the above cited solvents, it is to be concluded that the person skilled in the art is not in a position to determine without undue burden the pH prevailing in the medium, when the solvents are selected from ethers, aliphatic or aromatic hydrocarbons and halogenated hydrocarbons, in the course of the process defined in Claims 19 and 22.
- 3.8 The Board concludes from the above that the Respondent's objections under Article 100(b) EPC are founded and for this reason the present request is to be rejected.

First auxiliary request

4. Amendments

- 4.1 With respect to Claim 1 as granted, Claim 1 of this request differs in that the solvent medium was restricted to a list of solvents selected from alcohols, hydro-alcohol mixtures, ketones, esters and glycols.

 Dependent claims 19 and 22 as granted were deleted.
- 4.2 The Respondent submitted that the choice of particular solvents resulted from a purposive selection not directly and unambiguously derivable from the content of the application as originally filed.

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- 4.3 However, the solvents recited in Claim 1 are disclosed in Claim 19 of the application as originally filed among a list comprising, in addition to the solvents mentioned in point 4.1 above, also ethers, aliphatic and aromatic hydrocarbons, halogenated hydrocarbons. Claim 19 originally filed is so drafted that each of the indicated solvents represents a concrete embodiment of the "solvent medium". Therefore, the deletion of some of originally disclosed alternatives from that list is a simple limitation in respect of the possibilities for concretely defining the solvent medium. Since the alternatives kept in the claim are all solvents explicitly indicated in Claim 19 as originally filed, there is no extension of the content of the application as originally filed. For this reason, Claim 1 of this request does not give rise to objections under Article 123(2) EPC.
- 4.4 With respect to the amended pages of the description submitted by the Appellant in the course of the oral proceedings, the Board is satisfied that the amendments are necessary and appropriate to render the description consistent with the claimed subject-matter according to the first auxiliary request. This was not disputed by the Respondent.
- 4.5 Since the amendment brings about a restriction of the scope of protection conferred by the patent in suit, it is also in conformity with the requirements of Article 123(3) EPC.

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- 5. Insufficiency of disclosure Admissibility
- The Respondent had opposed the patent in suit under Article 100(b) EPC to the extent that Claims 19 and 22 as granted related to a process involving solvents selected from ethers, aliphatic and aromatic hydrocarbons and halogenated hydrocarbons (see page 14, paragraph 4.1 of the notice of opposition). Since the attack was based on the fact that the pH could not be determined without undue burden for solvents which had a low miscibility with water, it cannot extend to the other solvents listed in Claim 19.
- After the oral proceedings before the Board were arranged, in a letter dated 5 July 2007, the Respondent opposed Claim 1 of the auxiliary request which excluded the solvents in dispute (see point V above), on the ground that a buffer system could not be produced within the defined pH range in methanol/water solvent, in the presence of ammonia/ammonium acetate. In particular, the buffer system ammonia/ammonium acetate was not present in the solution of Example 4 of the patent in suit.
- 5.3 This objection raises a completely new issue which does not find any substantiation in the facts and arguments put forward against the possibility of measuring pH in the above cited solvents, i.e. ethers, aliphatic and aromatic hydrocarbons and halogenated hydrocarbons. On the one hand, the Respondent did not provide any justification for such late filing. On the other hand, the Appellant objected to the admissibility of this objection arguing that, given the facts and arguments submitted by the Respondent, the amended set of claims

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submitted as auxiliary request had for sole aim to avoid the objection under 100(b) EPC.

- In the Board's judgment, the issue relating to the presence or not in Example 4 of the patent in suit of a buffer system as defined in Claim 1 would require further detailed examination. Under these circumstances, the other party, i.e. the Appellant, and the deciding Board cannot reasonably be expected to deal with that new issue without accepting significant procedural delay, such as cancelling the arranged oral proceedings and continuing the appeal procedure in writing or remitting the case to the first instance for further prosecution.
- 5.5 In view of the above, the Board finds appropriate to exercise its discretion not to admit into the procedure the new issue in accordance with the provision of Article 114(2) EPC so that the case can be expected to be ready for a final decision at the conclusion of the oral proceedings (see Article 11(6) of the Rules of Procedure of the Boards of Appeal, OJ EPO 2003, 89).
- 6. Novelty
- At the oral proceedings before the Board, the
 Respondent challenged the novelty of the claimed
 subject-matter solely with regard to documents (2) and
 (5). In the circumstances of this case, the Board
 limits its considerations with respect to novelty to
 these documents in the absence of other documents
 relevant for that issue.

- 6.2 The Board observes that it has long been a generally applied principle by the Boards of appeal of the EPO that for concluding lack of novelty, there must be an unambiguous disclosure in the state of the art of subject-matter falling within the scope of what is claimed.
- 6.3 Document (2) discloses a process for the epoxidation of propylene carried out in a batch, continuous or semicontinuous manner wherein a titanium silicalite is used in the presence of hydrogen peroxide in a suitable solvent such as water, alcohols, ketones and mixtures of such solvents and a low concentration of a nonbasic salt to selectively catalyse the formation of the epoxide (see page 1, lines 5 to 8, page 3, lines 8 to 10 and page 4, lines 43-44; 48 to 51). "Nonbasic" in the context of this disclosure means a substance which when dissolved at a concentration of 0.1N in water at 25°C, provides a solution having a pH of less than 8 and preferably greater than 4 (see page 4, lines 7 to 10). Example 11, in particular, describes to this end the use of ammonium acetate in a batch process. First, since Example 11 discloses a batch process contrary to the process claimed which is a continuous process, it cannot be novelty destroying. Furthermore, both the general disclosure of document (2) as well as Example 11 are silent with regard to the possible presence of ammonia, in addition to ammonium acetate, and more generally with regard to the presence in the reaction medium of a buffer system as defined in Claim 1. As set out in document (8), an aqueous solution of ammonium acetate salt does not fall into the category of "Buffer Solutions" (see pages 253 and 285).

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- 6.4 The Respondent argued that from document (7), which is a sales catalogue of chemical substances submitted as evidence, it was known that the aqueous solution of ammonium acetate 99.99+% had a pH of 7 and, therefore, contained inherently 0.55% or so of ammonia and acetic acid. However, the Board is not convinced that this approach based on a pure aqueous solution can be unmistakably applied to the complex reaction medium disclosed in document (2) as set out in point 6.3 above. In the absence of evidence showing that such ammonium acetate nonbasic salt contains in addition ammonia and may act as a buffer system able to maintain the pH between 5.5 and 8.0, the argument of the Respondent remains unsubstantiated and for this reason the claimed subject-matter is also not unambiguously disclosed in document (2).
- Document (5) discloses a process for the preparation of olefine oxide by epoxydation of olefins with hydrogen peroxide in a solvent medium having a significant miscibility with water, in the presence of zeolite containing titanium atoms, wherein the pH of the reaction mixture is within the range from 4.8 to 6.5 (see page 1, lines 1 to 5; 15 to 19; page 3, lines 7 to 10 and lines 27 to 30). In a preferred form the pH value is maintained in the said range through addition of a weak base. As a weak base, NH₄OH is mentioned (see page 2, lines 4 to 10).

The Respondent argued that due to the well-known acidity of the zeolite, a salt of the weak base with said zeolite was formed, disclosing, therefore, the buffer system defined in Claim 1. However, in the

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absence of verifiable evidence in support thereof, this allegation is not convincing and for this reason, the claimed subject-matter is not unambiguously disclosed in document (5).

- 6.6 Therefore, the Respondent's novelty objections (Article 54 EPC) are unfounded.
- 7. Inventive step
- 7.1 The patent in suit according to the subject-matter of the claims of the first auxiliary request relates to a continuous process for the preparation of olefin oxides.
- 7.2 In agreement with the Opposition Division and the parties, the Board considers that whereas both documents (2) and (3) aim at the same objective, namely the epoxidation of olefinic compounds using titanium-containing molecular sieves as catalysts, document (3) is the closest prior art for defining the technical problem to be solved given that the examples of document (3) are concerned with a continuous process, whereas the examples of document (2) refer to a batch process.
- 7.3 Thus, starting from document (3), the technical results or effects successfully achieved by the claimed subject-matter are to be determined for defining the objective technical problem to be solved.
- 7.3.1 Document (3) teaches that the formation of by-products by non-selective ring-opening reactions can be reduced and, therefore, the selectivity for epoxides increased, when the catalyst is treated prior to the reaction or

during the reaction with a suitable acid neutralizing agent. Neutralization may be accomplished with water soluble basic substances chosen inter alia among weak bases such as ammonium hydroxide (see page 2, lines 11 to 21). However, it was found that by the use of such acid neutralizing agents, non-selective hydrogen peroxide decomposition to oxygen and water tended to gradually increase as the titanium silicalite catalyst aged (see page 2, lines 26 to 30). According to this document this detrimental tendency could be ameliorated by performing the epoxidation in the presence of a chelating agent (see page 2, lines 39 to 45). As shown by Table I of document (3), the addition of efficient amounts of NH₄OH and ATPM (aminotrimethylene phosphonic acid) in continuous propylene epoxidation runs provided a good yield, i.e. 69%, a good propylene oxide selectivity, i.e. 84%, whereas the catalytic activity was kept stable over a period of time of 630 hours (see Example 4).

7.3.2 The Respondent argued that the process according to document (3) relied on two independent technical effects, namely yields and propylene oxide selectivity on the one hand and stability of the catalyst on the other hand as set out in the diagram submitted as evidence (11) and, therefore, the technical problem could not be defined from those two separate effects.

However, the Board does not share the Respondent's view. Even though the technical result of this process is based on two independent technical effects, it remains that this is the **result** actually achieved which must be taken into account to define the technical problem to be solved.

- 7.3.3 The Appellant argued that in view of the additional data submitted in the course of the appeal proceedings, i.e. document (9), a run performed according to Example 5 of the patent in suit showed that even after 1,000 hours, H₂O₂ conversion and propylene oxide selectivity remained very high (95.0% and 98.0% respectively). Therefore, the technical problem to be solved vis-à-vis document (3) so argued the Appellant could be seen in an **improved** process for epoxidizing olefinic compounds in terms of yield, selectivity and stability of the catalyst (catalyst aging).
- However, in accordance with the established jurisprudence of the Boards of appeal, defining the technical problem to be solved in terms of an improvement vis-à-vis the closest prior art would require to verify whether or not the comparison is such that the effect is convincingly shown to have its origin in the distinguishing feature of the claimed subject-matter. The Board observes in that context that the composition of the base feed involved in both processes, namely the working examples of document (3) and the working examples of the patent in suit, comprise substantial differences, i.e. 2.5 weight% H₂O₂, 73 weight% isopropanol, 24 weight% water with respect to the experiments according to document (3) and 3.5 weight% H_2O_2 , 17 weight% water and 79.5 weight% MeOH in the examples of the patent in suit. Under these circumstances, the improvement alleged by the Appellant is not to be retained for establishing the problem to be solved and a less ambitious technical problem which the claimed invention addresses is to be defined (see T 1140/02, point 4.3).

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- 7.3.5 In view of document (3), the technical problem to be solved can be seen in the provision of a **further** process for preparation of epoxides with high yields and selectivity, whereas keeping stable the catalytic activity over a certain period of time. This objective technical problem is in line with that defined in the patent in suit (see page 2, line 39ff of the patent in suit).
- 7.4 The next step is to verify whether or not the technical problem defined above is solved by the claimed solution.
- 7.4.1 The Respondent argued that in a buffer consisting of ammonia and ammonium acetate, i.e. Example 4, the ratio of nitrogenated base to salt thereof was for a pH of 6.4 to 6.5 of 1:240 to 1:300. Given that the epoxidation reaction produced carboxylic acids, all the nitrogenated base was consumed resulting in a buffer consisting of carboxylic acids and ammonium salts thereof and not of the buffer as defined in Claim 1. The Appellant contended, by contrast, that support for a buffering system controlling the pH according to the claimed invention is found in the results of that example.
- 7.4.2 In the Board's judgment, the objection of the Respondent, raises a complex technical question, a proper response to which would require an experiment that seeks to reproduce Example 4 of the patent in suit. In the absence of such evidence, the Respondent's allegation is unsupported and, therefore, not convincing (see also point 5.4 above).

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- 7.4.3 Having regard to the technical information provided in the patent in suit, in particular Examples 3, 4 and 5, which show for each of them a yield of 94% or higher, a propylene selectivity of 96% or higher and a stability of the catalyst over a period of time longer than 100 hours, it can be considered that the claimed process according to this request successfully solves the technical problem defined above.
- 7.5 It remains to be decided whether or not the claimed solution is obvious in view of the cited prior art.
- 7.5.1 First, from the comparison of Examples 1, 3 of document (3), it becomes apparent that an increasing amount of ammonium hydroxide as cation source increases the propylene oxide selectivity (see table I, line "Propylene oxide selectivity"). However, the use of a cation source such as ammonium hydroxide tends to gradually increase the non-selective hydrogen peroxide decomposition to oxygen and water as the titanium silicalite catalyst ages (see page 2, lines 26 to 32; lines 39-41; Table I, Examples 1 and 3, line "Oxygen selectivity" and page 7, last line to page 8, line 3).
- 7.5.2 In the Board's judgment, document (3) establishes a clear link between the increase of selectivity due to the increasing amount of the source of cation such as ammonium hydroxide and the tendency of the catalyst to gradually deteriorate (see in particular, page 2, lines 39 to 41).
- 7.5.3 Furthermore, according to the uncontested calculations provided by the Appellant, i.e. document (12), the solution set up in Example 3 of document (3) has a pH

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ranging from 2.7 to 4.0 and no free ammonia is present. Therefore, no buffer system as defined in Claim 1 is present in the solution.

- 7.5.4 In view of this teaching, document (3) gives no incentive to the person skilled in the art to raise pH values beyond 4 by adding more ammonium hydroxide since he would have expected a still more detrimental aging effect on catalyst performance as measured by non-selective hydrogen peroxide decomposition to oxygen. This document gives, if anything, even less incentive to use a buffer system according to Claim 1.
- 7.5.5 The fact to use a buffer system as defined in Claim 1 so that the pH of the process is controlled between 5.5 and 8.0 as a solution to the technical problem defined above is for this reason unexpected.
- 7.5.6 The other prior art cited cannot rebut this finding. Even though document (6) teaches that the epoxidation of butadiene is achieved in maintaining the pH between 5 and 7 by adding alkaline aqueous solutions such as ammonium hydroxide or acid aqueous solutions (see page 2, lines 57 to 59), no teaching regarding the catalyst aging can be deduced since in view of all the working examples related to this epoxidation reaction, i.e. 1 to 19, the process disclosed is a batch process and lasts no more than a few hours, generally between 0.5 and 10 hours (see page 3, lines 2 to 4). In view of the teaching of document (3), the person skilled in the art would have rather thought that in the long run the catalyst would have gradually deteriorated. He would not have, therefore, taken into consideration the teaching of document (6) for solving the problem

defined above. Furthermore, the reaction medium disclosed in document (6) is clearly not buffered given that in all the examples, KOH and sulphuric acid must regularly be added to maintain the pH at 6.5. Such a document also for this reason cannot in combination with document (3) lead to the present claimed subjectmatter.

- 7.5.7 Likewise document (2), not cited by the Respondent in support of his attack for lack of inventive step, relates in all the examples to batch processes (one hour run time) and cannot address the problem of catalyst aging and for the same reasons as set out with regards to document (6) does not direct in combination with document (3) the person skilled in the art to the proposed solution.
- 7.5.8 Likewise document (4), not cited by the Respondent in support of his attack for lack of inventive step, does not help the person skilled in the art towards the claimed solution since the pH of the reaction is not mentioned and the buffer system referred to is a weak acid and a salt thereof with an inorganic base, i.e. NaOAc/HOAc (see col. 4, lines 56 to 60).
- 7.5.9 Since the cited documents do not lead the person skilled in the art to the claimed subject-matter for solving the technical problem defined above, Claim 1 of the first auxiliary request involves an inventive step in the sense of Article 56 EPC. The same applies to dependent Claims 2 to 23 which represent particular embodiments of Claim 1.

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Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

The case is remitted to the first instance with the order to maintain the patent as amended in the following version:

Description:

Pages 2-7 received during the oral proceedings of 18 September 2007;

Claims:

No. 1-23 filed with the letter dated 3 July 2006.

The Registrar

The Chairman

A. Townend

A. Nuss