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# Datasheet for the decision of 30 August 2011

**Case Number:** T 1761/08 - 3.3.01

**Application Number:** 03775165.8

**Publication Number:** 1556366

**IPC:** C07D 301/12

**Language of the proceedings:** EN

**Title of invention:**
Process for the epoxidation of olefins

**Patentees:**
Evonik Degussa GmbH
Uhde GmbH

**Opponents:**
BASF SE
THE DOW CHEMICAL COMPANY
Solvay (Société Anonyme)

**Headword:**
Epoxidation of olefins/EVONIK

**Relevant legal provisions:**
EPC Art. 100(a)(b)

**Relevant legal provisions (EPC 1973):**
-

**Keyword:**
"Interpretation of the claims"
"Invention sufficiently disclosed"
"Main request: novelty (yes): no particular effect required"
"Inventive step (yes): solution of the problem non obvious"

**Decisions cited:**
T 1204/06, T 0230/07

**Catchword:**
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**Case Number:** T 1761/08 - 3.3.01

# D E C I S I O N
## of the Technical Board of Appeal 3.3.01
## of 30 August 2011

**Appellants I**:
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**Decision under appeal:**
**Interlocutory decision of the Opposition Division of the European Patent Office posted 18 July 2008 concerning maintenance of the European patent No. 1556366 in amended form.**

**Composition of the Board:**

**Chairman:** P. Ranguis
**Members:** C. M. Radke
C.-P. Brandt

## Summary of Facts and Submissions

I. The patent in suit relates to a continuous process for the epoxidation of olefins using a heterogeneous catalyst.

II. In its interlocutory decision posted on 18 July 2008, the Opposition Division decided that the patent amended according to the first auxiliary request then on file met the requirements of the EPC.

III. This decision was appealed by the patentees (appellants I) and by the opponents BASF SE (appellant II) and The Dow Chemical Company (appellant III). The remaining opponent, Solvay S.A., did not appeal and is party to the appeal proceedings as of right.

IV. The oppositions were directed against the patent in its entirety and were based on grounds under Article 100(a) EPC (lack of novelty and of inventive step), and, as far as the opponent Solvay S.A. was concerned, on Article 100(b) EPC.

V. The following documents were inter alia cited during the opposition proceedings:

(D1) EP-A-1 072 600

(D2) F. Bellinger et al., Industrial and Engineering Chemistry, vol. 38, no. 3 (1946), 310-320

(D3) WO-A-00/76 989

(D21) Aldrich Catalogue Handbook of Fine Chemicals, Aldrich Chemie Bruxelles/BE 1990-1991, 712-713.

VI. As their main request the patentees asked for the maintenance of the patent as granted. Claim 1 as granted reads as follows:

"1. A continuos process for the epoxidation of olefins with hydrogen peroxide in presence of a heterogeneous catalyst promoting the epoxidation reaction, whereby the aqueous reaction mixture comprises:

i) an olefin;
ii) hydrogen peroxide;
iii) less than 100 wppm of alkali metals, earth alkali metals, both irrespective whether in ionic or complex form, bases or cations of bases having a $pk_B$ of less than 4.5, or combinations thereof; and
iv) at least 100 wppm of bases or cations of bases having a $pk_B$ of at least 4.5 or combinations thereof,

whereby the wppm are based on the total weight of hydrogen peroxide in the reaction mixture."

VII. The opposition division decided that

- the subject-matter requiring "bases or cations of bases having a $pk_B$ of at least 4.5" as claimed in the main request did not meet the requirement of Article 83 EPC;
- the subject-matter claimed in the first auxiliary request met the requirements of Article 83 EPC; it was novel as document (D1) did not disclose feature (iii);
- document (D1) was considered to represent the closest prior art. The problem solved was to provide an alternative continuous process.

Document (D1) did not indicate that the amount of component (iii) should be limited. Nor could the combination of the disclosure of document (D1) with that of (D2) or (D3) render the claimed subject-matter obvious.

VIII. During the appeal proceedings *inter alia* the following were additionally cited:

(D22) H. R. Christen, Grundlagen der allgemeinen und anorganischen Chemie, Otto Salle Verlag, Frankfurt/Main/DE, sixth edn. 1980, 278-279 and 358-359

(D23) H. R. Christen, Grundlagen der organischen Chemie, Verlag Sauerländer AG, Aarau/CH, 1st edn. (1970), 341-352

(D24) Test report "Experiments Supporting EP-B-1,556,366 Appeal", submitted under cover of appellant III's letter dated 28 November 2008, 3 pages.

IX. The claims on file are
Claims 1 to 20 as granted (main request);
claims 1 to 19 of the first auxiliary request;
claims 1 to 19 of the second auxiliary request;
claims 1 to 18 of the third auxiliary request;
where the claims of all the auxiliary requests were filed with the letter dated 11 May 2007.

Claim 1 of the main request is cited under point VI above.

X. **Appellants II and III** held that grounds under Article 100(b) EPC prejudiced the maintenance of the patent as

- the total amount of hydrogen peroxide in the reaction mixture was not defined in a continuous process; therefore, the patent did not disclose how to keep the concentration of component (iii) within the given range;
- anions having a $pK_B$ of less than 4.5, such as the $PO_4^{3-}$ ion used to stabilise the hydrogen peroxide, were to be considered as components (iii) according to present claim 1, so that none of the examples of the patent was covered by the present claims, and
- the definition of components (iii) and (iv) overlapped as the $pK_B$ was temperature dependent.

Appellant III argued that the subject-matter of the claims was not novel in view of document (D1). Appellants II and III considered document (D1) as the closest prior art when assessing inventive step. The objective problem solved was the provision of an alternative continuous process having a good long term hydrogen peroxide conversion and selectivity. They held that the experiments on file did not show a surprising effect over the whole breadth of the claims, so that the ranges indicated in the present claims were arbitrary. The solution as defined in the claims was obvious in view of documents (D2), (D3) and (D21).

XI. **Appellants I** argued that the only meaningful interpretation of the expression "the total weight of hydrogen peroxide in the reaction mixture" was that the "reaction mixture" was the mixture of reactants fed into the reactor. This was also in line with the description. The expression "bases or cations of bases having a $pk_B$ of less than 4.5" clearly related to

cations of bases and to the respective neutral bases. It did not include anions having a $pk_B$ of less than 4.5 as this would be in contradiction with claim 6 as granted and the description.

It was evident from document (D22) that $pK_B$ values were to be determined at standard conditions.

For these reasons and in view of the examples of the patent in suit, it was easy for the skilled person to choose the starting materials and their concentrations such that the process conditions of claim 1 were met.

The subject-matter of the claims was novel as document (D1) did not disclose feature (iii) of present claim 1. Document (D1) represented the closest prior art. The problem posed was to provide a continuous process for the epoxidation of olefins, said process ensuring an improved long term activity and selectivity of the heterogeneous catalyst in an economic manner without additional process steps. The comparative tests showed that this problem was solved. There was no motivation in the prior art to modify the reaction mixture disclosed in document (D1) so that it met the requirement (iii) of present claim 1.

XII. The party as of right, **Solvay S.A.**, neither submitted any arguments nor filed any requests during the appeal proceedings.

XIII. Appellants I requested that the decision under appeal be set aside and the patent be maintained unamended (main request) or on the basis of the claims of any of

the first to third auxiliary requests (see point IX above).

Appellants II and III requested that the decision under appeal be set aside and the patent be revoked.

XIV. Oral proceedings were held on 30 August 2011. The party as of right was duly summoned but did not attend the oral proceedings as announced in its letter dated 25 July 2011.

The proceedings were thus continued in the absence of the duly summoned party as of right in accordance with Rule 115(2) EPC.

At the end of the oral proceedings, the chairman announced the decision of the Board.

## Reasons for the Decision

1. The appeals are admissible.

*Main request*

2. Interpretation of the claims

2.1 Feature (iii) of present claim 1 reads as follows:

" iii) less than 100 wppm of alkali metals, earth alkali metals, both irrespective whether in ionic or complex form, bases or cations of bases having a $pk_B$ of less than 4.5, or combinations thereof; ...

whereby the wppm are based on the total weight of hydrogen peroxide in the reaction mixture."

2.2 It was disputed how the expressions

- "the total weight of hydrogen peroxide in the reaction mixture" and
- "bases or cations of bases having a $pk_B$ of less than 4.5"

in said feature (iii) were to be interpreted (see above under points X and XI).

2.3 "The claims are ... directed to the person skilled in the art who will rule out interpretations which are illogical or do not make technical sense" (T 1204/06 of 8 April 2008, point 3.4 of the reasons).

2.4 Appellants I held that the term "reaction mixture" in the first expression referred to the mixture fed into the reactor, while appellants II and III deemed that this term was not defined and could, e.g., be read as a mixture in which the reaction had already partly or totally taken place.

2.4.1 Feature (iii) sets an upper limit to the content of certain classes of compounds in the reaction mixture (hereinafter called compounds (iii)). In order to avoid corrosion, reactors and tubings are usually made of materials inert under the reaction conditions. So, the only considerable sources of the compounds (iii) are the starting materials as fed into the reactor. Hence, it makes sense to base the amount of these compounds on the reaction mixture as fed into the reactor. In order to keep the amount of compounds (iii) within the limits

required by feature (iii) in claim 1, the person skilled in the art may use purer starting materials.

2.4.2 So, it remains to be decided whether basing the amount of compounds (iii) on the total weight of hydrogen peroxide in the reaction mixture within the reactor or leaving the reactor also makes technical sense.

As essentially the only source of compounds (iii) is the feed stream into the reactor, the total amount of these compounds is constant at constant velocity of the feed stream (see under point 2.4.1 above).

Present claim 1 relates to a "continuous process for the epoxidation of olefins with hydrogen peroxide". This means that hydrogen peroxide is consumed as the reaction proceeds.

As the total amount of compounds (iii) remains constant during the reaction while the total amount of hydrogen peroxide decreases, the amount of compounds (iii) based on the weight of hydrogen peroxide increases. To base the amount of compounds (iii) on the total weight of hydrogen peroxide within the reactor would require to determine said weight by monitoring the hydrogen peroxide concentration along the reaction path. On the one hand this is so complicated that it makes no technical sense.

On the other hand basing the amount of compounds (iii) on the total weight of hydrogen peroxide in the reaction mixture within the reactor or leaving the reactor means that one could meet the requirement (iii) of claim 1 simply by keeping the hydrogen peroxide at a

high level, i.e. by keeping its conversion at a minimum. This does not make sense as chemical technology aims at reaching high conversions of the reactants.

2.4.3 Therefore, the only interpretation that makes sense is that "the total weight of hydrogen peroxide in the reaction mixture" is based on the reaction mixture fed into the reactor.

2.5 Appellants I argued that the term "bases and cations of bases" clearly related to cations of bases and to the respective neutral bases, whereas appellants II and III held that this term also comprised anionic bases.

Appellants I deemed that this interpretation of appellants II and III was in contradiction with claim 6 as granted which allowed the addition of unlimited amounts of anions.

The Board cannot detect such a contradiction between claims 1 and 6. The reaction mixture may well contain unlimited amounts of anions as long as the anions having a $pK_B$ of less than 4.5 are limited as required in claim 1. Hence, the interpretation that the bases mentioned in feature (iii) of claim 1 may comprise anions is not illogical. It also makes technical sense as many of the most common bases are anions, such as the hydroxyl ion.

2.6 For these reasons, the Board concludes that

– the compounds the content of which is limited in feature (iii) of claim 1 include anions having a $pK_B$ of less than 4.5; and

– "the wppm ... based on the total weight of hydrogen peroxide in the reaction mixture" refers to the reaction mixture as fed into the reactor.

3. Article 100(b) EPC

3.1 Appellants II and III argued that the patent did not disclose the invention in a way such that the person skilled in the art knew how to meet requirement (iii) of claim 1 (see point X above).

3.2 Their argument that "the total weight of hydrogen peroxide in the reaction mixture" was not defined is not shared by the Board for the reasons given under point 2.4 above.

3.3 They further argued that the $pK_B$ was temperature dependent and thus not well defined. Appellants' I counterargument that it was evident the $pK_B$ had to be determined at standard conditions was based on document (D22). This document (D22) consists of pages from a textbook of general chemistry. The $pk_S$ values given there were determined at 25 °C (see the first line on page 358). The document also states that the standard conditions of a compound were a temperature of 25 °C and a pressure of one bar (see page 279, the first sentence under the heading "8.2 Thermochemie"). As the $pK_S$ (also denoted as $pK_A$) is linked to the $pK_B$ ($pK_A + pK_B = 14$; see document (D23), the first equation on page 342), it is apparent that the $pK_B$ values are also determined at a standard temperature of 25 °C.

3.4 For these reasons the Board does not share the view of Appellants II and III that the features "the total

weight of hydrogen peroxide in the reaction mixture" and "a $pk_B$ of less than 4.5" in present claim 1 are ambiguous to the person skilled in the art. Therefore it need not be decided whether or not the respective objection should be subsumed under Article 84 rather than under Article 100(b) EPC.

3.5 Hence, no ground under Article 100(b) EPC prejudices the maintenance of the patent unamended.

4. Novelty

4.1 Appellant III considered the subject-matter of the claims not to be novel in view of the disclosure of document (D1) (see under point X above). Said document disclosed all the features of present claim 1 except feature (iii). It argued that its comparative tests (D24) showed that the limitation of components (iii) to less than 100 wppm was arbitrary and could not render the subject-matter claimed novel.

4.2 Document (D1) relates to "A continuous process for the preparation of olefin oxides by the direct epoxidation of an olefin with hydrogen peroxide, ..., 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" (see claim 1).

In the examples, ammonia was used as the nitrogenated base. Ammonia (i.e. $NH_3$) has a $pK_B$ of 4.76 (see Table 2 on page 10 of the patent in suit) and thus is no base component (iii) according to present claim 1.

The fact that document (D1) does not mention the addition of compounds (iii) according to present claim 1 does not necessarily mean that no such compound is present in the starting materials used in the examples. Document (D1) is silent on the origin of the hydrogen peroxide used in the process. Commercially available hydrogen peroxide is usually stabilised with salts which may or may not be considered as compounds (iii) according to present claim 1 and in quantities which may exceed 100 wppm (see document (D3), page 3, lines 9-13). Present claim 1, however, requires that compounds (iii) only be present in an amount of less than 100 wppm based on the weight of the hydrogen peroxide.

4.3 Hence, the Board agrees with all the parties that document (D1) does not disclose a process including the requirement (iii) of present claim 1. A particular effect caused by this differing feature is no prerequisite for novelty (see T 0230/07 of 5 May 2010, point 4.1.6 of the reasons). Hence, the disclosure of document (D1) does not deprive the subject-matter of the present claims of novelty. Neither have the parties based a novelty objection on any other document nor is the Board aware of a document relevant for the assessment of novelty of the subject-matter claimed.

For these reasons, the subject-matter of the claims is novel.

5. Inventive step

5.1 The Board agrees with the parties that document (D1) represents the closest prior art for the assessment of inventive step.

5.2 In accordance with the "problem-solution" approach consistently applied by the Boards of Appeal, it is necessary to determine in the light of the closest prior art the technical problem which the invention addresses and successfully solves.

5.2.1 The problem addressed in the patent application on which the patent in suit was granted was "to provide a continuous process for the epoxidation of olefins with hydrogen peroxide in presence of a heterogeneous catalyst promoting the epoxidation reaction wherein an improvement in long term activity and selectivity of the catalyst ... is achieved without adding addition process steps in an economic way" (see page 6, lines 16-21 of the application as filed).

5.2.2 Appellants I argued that example E6 when compared with example CE8 of the patent in suit showed that this problem was solved in view of the disclosure of document (D1). Appellants II and III argued that the comparative tests (D24) showed that this problem was not solved.

5.2.3 The continuous epoxydation of propylene with hydrogen peroxide described in document (D24) consists of three periods of time,

– a first, 584 hour period intended to reproduce example E6 of the patent in suit (where the alkali

level was 30 ppm based on the total weight of hydrogen peroxide);

- a second, 550 hour period in which the alkali level was increased to 110 ppm based on the total weight of hydrogen peroxide, and, finally,
- a third, 312 hour period in which the alkali level was switched back to that of the first period.

Appellant III argued that this test showed that increasing the alkali content above the threshold of 100 ppm indicated in present claim 1 had no effect on the hydrogen peroxide conversion and the selectivity of the catalyst.

When interpreting the data presented in document (D24), it has to be taken into account that the temperature of the hot oil used as the heating medium was modified throughout the experiment (see page 2, runtime 48 h: hot oil intro: 30 °C; hot oil reflux: 31,3 °C; runtime 1446 h: hot oil intro: 37.0 °C; hot oil reflux: 40.5 °C). This was apparently done in order to maintain a constant conversion (see the last sentence on page 1 of document (D24)).

This rise in heating temperature may not only have an effect on the conversion of hydrogen peroxide but also on the selectivity, as the rates of the desired and the side reactions may differ in temperature dependence due to different activation energies.

For these reasons, the experiment described in document (D24) is not suitable for supporting the arguments of Appellant III.

5.2.4 The experiments E6 and CE8 listed in Table 1 on page 10 of the patent in suit were carried out using the same temperature of the cooling or heating medium (see page 9, lines 45-46). They differ only in that in experiment CE8

- the running time was slightly lower, and
- 100 wppm of methylamine based on the total weight of hydrogen peroxide in the feed stream was added.

In these examples, components (iii) are

- in both experiments:
  30 ppm of sodium (which is an alkali metal; see paragraph [0071] of the patent in suit); and
- in experiment CE8: additionally 100 wppm of methylamine (which a base having a $pK_B$ of 3.36; see Table 2 on page 10).

Hence, experiment CE8 does not meet the requirement that the concentration of compounds (iii) is to be less than 100 wppm based on the total weight of hydrogen peroxide.

A comparison of experiment E6 with CE8 shows that the addition of 100 wppm of methylamine leads to a decrease in the conversion of hydrogen peroxide from 94 to 21 % and in the selectivity from 90 to 82 %.

5.2.5 Appellants II and III argued that appellant I had not shown that an effect was achieved over the whole breadth of the claims. Furthermore, they deemed experiment E6 not to be according to the present claims due to the presence of large amounts of phosphates in the hydrogen peroxide.

In opposition appeal proceedings it is up to the party raising an argument to provide evidence in its support.

The latter argument is based on the fact that

- the orthophosphate anion ($PO_4^{3-}$) has a $pK_B$ below 4.5 and thus is a compound (iii) according to present claim 1; and
- the hydrogen peroxide used in the experiments "contained 250 mg/kg $H_2O_2$ phosphates" (see page 9 lines 29-30 of the patent in suit).

However, Appellants II and III have provided no evidence showing that in experiment E6 the content of orthophosphate anions $PO_4^{3-}$ in the phosphates is such that it exceeds, together with the 30 ppm of sodium, the limit of less than 100 ppm of components (iii).

Neither did appellants II and III argue that any phosphate species other than $PO_4^{3-}$ that could be formed in the reaction mixture had a $pK_B$ of less than 4.5 and thus could be considered as a component (iii) according to present claim 1, nor could the Board find any evidence in this respect. In fact, the patent in suit teaches that the "phosphates" addded are 200 ppm sodium **pyro**phosphate and 50 ppm orthosphosphoric **acid** $H_3PO_4$ (see page 9, lines 24 and 27). Therefore, there is no reason to believe that considerable amounts of "the phosphates" in the reaction mixture are in the form of $PO_4^{3-}$. This even holds when one takes into account that 1000 ppm of the base ammonia is added, as ammonia is a weaker base than $PO_4^{3-}$.

For this reason, the Board proceeds from the fact that experiment E6 is an example according to present claim 1.

Likewise, appellants II and III did not provide any evidence showing that the effect mentioned under point 5.2.4 above was not achieved over the whole breadth of the claims.

5.2.6 Hence, the Board proceeds from the fact that the problem posed according to point 5.2.1 above was solved by the claimed subject-matter over the whole breadth of the claims.

5.3 Finally it has to be assessed whether or not the solution of this problem as defined in the present claims was obvious in view of the disclosure of the closest prior art document (D1) alone or in view of its combination with any other documents of the prior art. As such other documents, appellants II and III cited documents (D2), (D3) and (D21).

5.3.1 Document (D1) neither suggests limiting the concentration of components (iii) nor indicates that their presence could have a negative effect on hydrogen peroxide conversion or selectivity (see examples 8 and 9 as compared with example 2, where no sodium ions were added). Therefore, this document alone cannot render the subject-matter claimed obvious.

5.3.2 Documents (D2) and (D21) disclose compositions of commercially available stabilised brands of hydrogen peroxide. These documents, however, neither relate to an epoxydation reaction nor to catalysts used therein.

Hence, they could not give any indication to the person skilled in the art how to solve the problem posed.

5.3.3 Document (D3) relates to a process for reacting an olefin with a hydrogen peroxide in which the concentrations of each of the dissolved anions and cations is below 100 ppm (see claims 1 and 6). The document aims at increasing the selectivity of the reaction (see page 3, lines 25-28). It mentions that this can be achieved without the pretreatment of the catalyst with a basic compound and without adding any basic salt to the reaction mixture (see page 3, line 30, to page 4, line 7). The person skilled in the art applying this teaching to the process disclosed in document (D1) would have avoided adding any basic compound, contrary to feature (iv) of present claim 1.

5.3.4 The Board is not aware of any other cited document which could render the subject-matter of the present claims obvious.

5.4 Hence, the subject-matter of the claims of the main request is based on an inventive step.

6. As the main request is allowable, there is no need to deal with the auxiliary requests.

## Order

**For these reasons it is decided that:**

1. The appeals of appellants II and III are dismissed.

2. The decision under appeal is set aside.

3. European patent No. 1 556 366 is maintained unamended.

The Registrar: The Chairman:

M. Schalow P. Ranguis