BESCHWERDEKAMMERN DES EUROPÄISCHEN PATENTAMTS

BOARDS OF APPEAL OF THE EUROPEAN PATENT OFFICE

CHAMBRES DE RECOURS DE L'OFFICE EUROPEEN DES BREVETS

Internal distribution code:
(A) [ ] Publication in OJ
(B) [X] To Chairmen and Members
(C) [ ] To Chairmen

D E C I S I O N
of 5 December 1995

Case Number: T 1077/92 - 3.2.2

Application Number: 86306561.1

Publication Number: 0219945

IPC: C23G 1/10

Language of the proceedings: EN

Title of invention:
Stabilisation of acidic hydrogen peroxide solutions

Applicant:
SOLVAY INTEROX LIMITED

Opponent:
-

Headword:
Hydrogen peroxide solutions/SOLVAY

Relevant legal provisions:
EPC Art. 56

Keyword:
"Inventive step - yes; seemingly obvious expedient"

Decisions cited:
-

Headnote:
In exceptional circumstances, technical considerations which point towards the obviousness of an alleged invention are capable of being overcome, and a reverse conclusion reached, where there is strong circumstantial evidence pointing in the opposite direction. Such a situation can arise where, in an area of active research, there is no explanation as to why a seemingly obvious proposal had not been adopted long ago.

Europäisches Patentamt | European Patent Office | Office européen des brevets

Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 1077/92 - 3.2.2

D E C I S I O N
of the Technical Board of Appeal 3.2.2
of 5 December 1995

Appellant: SOLVAY INTEROX LIMITED
Baronet Works
Baronet Road
Warrington
Cheshire WA4 6HB (GB)

Representative: Pearce, Timothy
Solvay Interox Limited
Patent Department
P.O. Box 51
Moorfield Road
Widnes
Cheshire WA8 0FE (GB)

Decision under appeal: **Decision of the Examining Division of the European Patent Office dated 5 August 1992 refusing European patent application No. 86 306 561.1 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: H. J. Seidenschwarz
Members: R. A. Lunzer
J. C. M. de Preter

Summary of Facts and Submissions

I. European patent application No. 86 306 561.1, Publication No. 219 945, filed on 26 August 1986, claiming a priority date of 5 September 1985 derived from GB Application No. 8 522 046, was refused by a decision of the Examining Division dated 5 August 1992. That decision was based on a set of 13 claims, of which Claim 1 reads as follows:

"A process for stabilising aqueous acidic hydrogen peroxide solutions in the presence of catalytic metal ions characterised by maintaining the solution in contact with a block of a solid stabiliser for hydrogen peroxide that is scarcely soluble in the acidic solution, whereby a saturated solution of stabiliser is obtained and maintained by slow dissolution to offset losses."

II. The ground for the refusal was that the alleged invention lacked any inventive step having regard to documents

(1) US-A-3 257 294,

(2) US-A-3 407 141, and

(3) DE-C-880 528.

Document (1) disclosed a method and a bath for electroplating in which self-regulation of the concentration of agents within the bath was achieved by using them in the form of sparingly soluble compounds, to achieve the effect that any depletion in concentration was made good by further solution of the sparingly soluble solid present in the bath. The

Examining Division saw no relevant distinction between the solids there used, and the "solid block" of the present alleged invention. The advantages obtained by providing the additives in the manner disclosed in document (1) (column 3, lines 42 to 53) were similar to those obtained in accordance with the process claimed in the application in suit.

Document (2) described acidified hydrogen peroxide etching solutions (e.g. for etching copper, iron, nickel etc.) containing catalytic amounts of additives for stabilising the hydrogen peroxide such as hydrobenzoic acid. As document (2) disclosed the use of sparingly soluble additives for stabilising aqueous acid hydrogen peroxide solutions in the presence of catalytic amounts of metals, and as document (1) proposed the use of sparingly soluble additives in the form of solid blocks in order to achieve self-regulation of the additive concentration in solution, it was held that it would have been evident to the skilled worker in the art to apply the method of delivery described in document (1) to the process of stabilising hydrogen peroxide described in document (2) in order to obtain sustained release of the desired additive in a solution of acidic hydrogen peroxide containing catalytic amounts of metallic ions. Accordingly it was held that the alleged invention was obvious.

In addition, the Examining Division referred to document (3), which proposed the use of a sparingly soluble solid block of anti-corrosion salts, which it interpreted as showing that providing an additive in the form of a solid block of a sparingly soluble substance was customary in the art, and without any doubt part of the common knowledge of the man skilled in the art, and

that that knowledge could be applied without difficulty to the stabilisation of aqueous acidic hydrogen peroxide solutions.

III. An appeal against that decision was filed on 5 October 1992, the appeal fee was paid on 9 October, and the statement of grounds of appeal was filed on 1 December 1992. The Appellant contended essentially that the solids described as being used in document (1) were not in the form of a solid block. The presence of the solid in the form of a block was an essential and very useful feature of the present invention, since it afforded a reliable guide to the operator of the bath of when the block was exhausted, and a fresh one needed. Document (3) concerned cooling systems for motor cars. That was not an area in which a skilled worker would search when seeking to solve a problem concerned with the operation of an acidified hydrogen peroxide bath. There was no reference in documents (1) or (2) to document (3). The age of the citations relied on by the Examining Division did not support its contention that the invention was obvious.

As indicated in the Appellant's letter of 25 September 1991, it had been manufacturing and selling hydrogen peroxide since before 1900, and with its sister companies was the largest producer in the world. Throughout all those years, the need for stabilisers in hydrogen peroxide solutions had been satisfied by the use of fully soluble additives. Never before had it been suggested that the presence of a partially soluble solid could afford better stabilisation than the liquids used hitherto.

IV. The Appellant requested that the decision under appeal be set aside, and a patent granted on the basis of Claims 1 to 13 with which the decision under appeal had

been concerned. By way of an auxiliary request it sought a modified Claim 1 which made reference to the block being replenished when no longer visible.

Reasons for the Decision

1. The appeal is admissible.

2. *Background to the alleged invention*

2.1 The use of sparingly soluble substances to achieve constant concentration in solution.

The principles governing the solubility of salts in water are so well-known that they must be regarded as part of the common general knowledge of persons skilled in the art. In particular it is part of the common general knowledge of persons dealing with the presence of salts in solutions that the concentration of an ion in solution can often be controlled at a desired level by forming a salt with a second ion, selected so that, when the saturation concentration of the salt is reached, the concentration of the ion is at the desired level. In that case, by providing a residual amount of solid salt in contact with the solution, the desired concentration of the ions in solution can be maintained, irrespective of any depletion or loss resulting from chemical or mechanical removal of the ions from solution. As soon as the concentration level of the ion tends to be reduced below the equilibrium level, the loss is made good as a result of further solution of the solid salt, until the equilibrium concentration is regained. For the sake of brevity, the use of this principle will be referred to hereafter as "self-regulation of concentration."

Although the search report produced by the EPO has revealed only two examples of the application of the above principle to the control of concentration of a given ion in solution, (documents (1) and (3)), the Board expects that many other examples of the application of this well-known principle could be found as a result of a further search.

2.2 The use of acidified hydrogen peroxide solutions.

2.2.1 The use of acidified hydrogen peroxide solutions is discussed in document (2), and also in the following documents which were referred to in the search report:

(4) US-A-3 869 401, and

(5) US-A-3 773 577.

2.2.2 Acidified hydrogen peroxide has been widely used, or contemplated for use, as an etchant for metals, more especially copper and copper base alloys (application in suit column 1, lines 5 to 8; document (2) column 1, lines 56 to 64; document (4) column 1, lines 8 to 9; document (5) column 1, lines 40 to 41). Documents (2) and (5) relate to its use as an etchant in connection with the production of printed circuit boards, an area of rapid and intensive development over the past 50 years. The problems associated with the use of acidified hydrogen peroxide reside essentially in the well-known instability of the H_2O_2 molecule, particularly in the presence of cupric ions, which are described in the application in suit at column 5, lines 40 to 41 as, "a notorious decomposition catalyst for hydrogen peroxide".

2.2.3 The problems concerning finding a suitable stabiliser for hydrogen peroxide are emphasised in the available prior art. Thus document (2) (column 1, lines 56 to 64) states:

"As an etchant for copper, aqueous hydrogen peroxide is very attractive because of its relatively low cost and ability to recover copper electrolytically from a spent peroxide solution. However, the utilization of hydrogen peroxide for a constructive purpose in metal etching is subject to numerous problems and pitfalls. Basically, potentially useful solutions combining hydrogen peroxide and acid were found subject to great deficiencies by way of etch rates and capacity."

That problem, in accordance with the proposal of document (2), is overcome by the inclusion of certain soluble stabilisers for the hydrogen peroxide (column 2, lines 30 to 36).

2.2.4 Document (4) is concerned with the stabilisation of hydrogen peroxide in the presence of chloride ions, and states as follows in its introduction (column 1, lines 8 to 13).

"H_2O_2 in acid solution is well known in the dissolution of metals in etching applications. In the presence of metal ions, however, H_2O_2 tends to decompose rapidly. It is also known that pH effects induce the degradation of H_2O_2. Stabilization of H_2O_2 against these causes of degradation has been the goal of **much research effort**." (emphasis added)

It goes on to propose the use of certain soluble stabilisers for the hydrogen peroxide (column 1, lines 24 to 41).

2.2.5 Document (5) relates to the use of hydrogen peroxide as an etchant for copper when used in printed circuit boards, and deals specifically with the problem of preventing side or lateral etching and undercutting of the resist (column 1, lines 58 to 63). Its relevance to the present decision is that it confirms that hydrogen peroxide is used as an etchant in the active area of development involved in the production of printed circuit boards.

2.3 The instability of hydrogen peroxide.

The instability of hydrogen peroxide is notorious, as already evident. So too is the need for the presence of a stabiliser. To quote from the entry for hydrogen peroxide in the Condensed Chemical Dictionary (Reinhold Publishing Corporation, 1961):

"It is fundamentally unstable; the decomposition is slow with pure material but catalysed by many impurities, especially metallic impurities. The commercial solutions commonly contain a preservative, such as acetophenetidin or acetanilide."

3. *The alleged invention*

3.1 The alleged invention, as defined in Claim 1 set out in paragraph I above, makes use of the principle of the self-regulation of concentration (defined in paragraph 2.1 above) to solve the problem of maintaining a desired concentration of stabiliser in acidic solutions of hydrogen peroxide in the presence of catalytic metallic ions.

3.2 The results given in Table 1 of the application in suit demonstrate credibly that a solution of hydrogen peroxide stabilised in accordance with the alleged

invention is capable of sustaining its activity as an etchant for copper alloys for long periods. The Board is therefore satisfied that the alleged invention overcomes the problem of keeping the proportion of stabiliser in solution constant, notwithstanding the losses which occur, such as due to adhesion of liquid to the treated articles.

4. *Novelty*

Having reviewed the cited documents (see paragraph 2 above) the Board is satisfied that none of them discloses a process having all the features defined in Claim 1. Therefore the subject-matter of Claim 1 is considered to be novel within the meaning of Article 54 EPC.

5. *Inventive step*

5.1 Document (1) concerns the field of electroplating in an aqueous solution, rather than the use of an acid etching bath, but the techniques used in these two fields overlap to such extent that a teaching in one area would be considered as potentially useful in the other. It discloses (cf. its claims) using the principle of the self-regulation of concentration, in this case as applied to the concentration of additives in an electroplating bath (cf. paragraph 2.1 above). Starting from this disclosure, it is arguable that by analogous use, the skilled worker would have foreseen no difficulty in applying the same technique, and the same principle, to the task of controlling the concentration of the stabiliser needed in an acidic hydrogen peroxide etching bath. However, the Board rejects that argument as being based on hindsight. Given a knowledge of the

invention, it was possible in conducting a search to find a disclosure of the use of the self-regulation of concentration, as exemplified by document (1).

5.2 The same objection applies equally to document (3), which is concerned with a field of activity remote from etching or plating baths, and is also a document which could be selected solely on the basis of hindsight. In addition, the Board is unable to agree with the interpretation put on this document by the Examining Division. Instead of showing that the exploitation of a block of a sparingly soluble substance is common knowledge, as was held by the Examining Division, the common knowledge identified in this document is the widespread use of readily **soluble** corrosion inhibitors in the form of salts (cf. also paragraph 2.1 above) in the water used in the cooling system of a motor car. In contrast to that general practice, its novel proposal was the replacement of soluble salts by a sparingly soluble compound present as a solid in contact with a saturated solution, i.e. the above-mention principle of self-regulation of concentration. This is not a document which would be found by a person concerned with the problem of the instability of hydrogen peroxide when in acid solution in the presence of metallic ions. Accordingly, the Board does not accept that the alleged invention was obvious in the light of its disclosure.

5.3 These documents (1) and (3) do no more than reflect the fact that the principle of self-regulation of concentration is a well-known tool in the hands of skilled persons. Accordingly it is the common general knowledge of that principle, rather than these isolated disclosures of its application in other fields, which affords the strongest basis for challenging the inventiveness of the subject-matter of Claim 1.

5.4 In refusing the application, the Examining Division interpreted document (2) as disclosing a sparingly soluble stabiliser used in an acidified hydrogen peroxide solution. That interpretation is in the Board's view mistaken. Although there is mention at column 2, lines 59 and 60 of the use of hydroxy benzoic acid, which is not readily soluble, that suggestion is qualified at line 69 by the more specific reference to "benzoic acid compounds", and these are exemplified in baths G and H of Table 1 by the soluble substance, sodium benzoate. In fact there is no suggestion in document (2) of using any stabiliser other than when taken into solution completely, and in the absence of any residual solid material. That it is concerned solely with stabilisers which are in solution is confirmed by the passage at column 6, lines 38 to 56, which discusses the supply of the stabilisers in the form of a concentrate, and their subsequent dilution prior to use. A concentrate could not be diluted reliably so as to yield solutions of consistent strength if the concentrate included a proportion of insoluble solid matter. Thus this disclosure, correctly interpreted, is no more than a further example of the numerous proposals for the inclusion of **soluble** stabilisers, such as those acknowledge at column 1, lines 18 to 23 of the application in suit, and it is not an example of the application of the principle of self-regulation of concentration, as are documents (1) and (3).

5.5 The Board has therefore to face the question: why was the principle of self-regulation of concentration never previously applied to controlling the concentration of stabilisers used in the presence of hydrogen peroxide solutions, despite the problem of stabilising hydrogen peroxide having been known for 100 years or more, as emphasised by the Appellant in its argument referred to in paragraph III above, and despite the fact that acidic

solutions of hydrogen peroxide have been the subject of intensive research in association with the development of printed circuit boards over the past decades?

5.6 The Board can find no answer to that question, other than to conclude that, for whatever the reason, it cannot have been obvious. If the step had been an obvious one, it would have been taken long ago. In the light of the factual considerations mentioned above, including the long known problem of needing to stabilise hydrogen peroxide solutions, and the active research which is likely to have been directed to solving this problem insofar as it was encountered in the rapidly developing technology involved in the etching of printed circuit boards, in the Board's judgment it must have required inventive insight to perceive that the principle of self-regulation of concentration could advantageously be applied to solve the problem.

5.7 The present case exemplifies a situation in which, although an evaluation of the technical facts, taken in isolation, points initially towards the obviousness of an invention, that preliminary view is inconsistent with the whole of the facts. In exceptional circumstances, such as those of the present case, technical considerations which point towards the obviousness of an alleged invention are capable of being overcome, and a reverse conclusion reached, where there is strong circumstantial evidence pointing in the opposite direction. As in the present case there is no evident explanation why the principle of self-regulation of concentration had not been adopted long ago, the Board is satisfied that the subject-matter of Claim 1 involves an inventive step, as is required by Article 56 EPC.

6. *Conclusion*

Claim 1 being allowable, the same applies to dependent Claims 2 to 13, which are directed to processes falling within the scope of Claim 1 and whose inventiveness is supported by that of the main Claim.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the Examining Division with the order to grant a patent on the basis of the following version:

Description: pages 1 to 8 as originally filed.

Claims: 1 to 13 as originally filed.

The Registrar: The Chairman:

S. Fabiani H. Seidenschwarz

19.12.95