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**D E C I S I O N**  
**of 15 March 2005**

**Case Number:** T 0302/01 - 3.3.1

**Application Number:** 94922379.6

**Publication Number:** 0665210

**IPC:** C07C 53/08

**Language of the proceedings:** EN

**Title of invention:**

Process for producing acetic anhydride alone or both of acetic anhydride and acetic acid

**Patentee:**

DAICEL CHEMICAL INDUSTRIES, LTD.

**Opponent:**

BP Chemicals Ltd

**Headword:**

Acetic anhydride production/DAICEL

**Relevant legal provisions:**

EPC Art. 56, 113(2)

**Keyword:**

"Admissibility of a late-filed request (yes)"

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

**Decisions cited:**

-

**Catchword:**

-



Case Number: T 0302/01 - 3.3.1

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.1  
of 15 March 2005

**Appellant:**  
(Opponent)

BP Chemicals Ltd  
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London EC2M 7BA (GB)

**Representative:**

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**Respondent:**  
(Proprietor of the patent)

DAICEL CHEMICAL INDUSTRIES, LTD.  
1, Teppo-cho  
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Osaka (JP)

**Representative:**

Grünecker, Kinkeldey,  
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**Decision under appeal:**

Interlocutory decision of the Opposition  
Division of the European Patent Office posted  
23 January 2001 concerning maintenance of  
European patent No. 0665210 in amended form.

**Composition of the Board:**

**Chairman:**

A. J. Nuss

**Members:**

J. M. Jonk

R. T. Menapace

## Summary of Facts and Submissions

- I. The Appellant (Opponent) lodged an appeal against the interlocutory decision of the Opposition Division in which it was found that the subject-matter of the patent in suit No. 0 665 210 (European patent application No. 94 922 397.6) as amended meets the requirements of the EPC.
- II. The decision was based on Claims 1 to 3 filed on 22 May 2000, independent Claim 1 reading as follows:
- "A process for producing acetic anhydride alone or acetic anhydride and acetic acid by
- (a) continuously reacting dimethyl ether and/or methyl acetate and optionally water and/or methanol with carbon monoxide alone or carbon monoxide and hydrogen in the presence of a rhodium compound and methyl iodide as the principle catalysts in a carbonylation reactor,
  - (b) introducing the resulting liquid reaction mixture into an evaporator having a pressure lower than that of the reactor to separate said reaction mixture into a volatile phase containing the product, unreacted dimethyl ether and/or methyl acetate, and methyl iodide, and a nonvolatile phase containing said rhodium compound,
  - (c) distilling said volatile phase from step (b) to obtain the product and a distillate containing the unreacted dimethyl ether and/or methyl acetate and methyl iodide, and

- (d) recirculating said liquid distillate from step (c) to said reactor while recirculating said nonvolatile phase from step (b) containing said rhodium compound to said reactor,

characterized in that

- the distillate of step (c) is introduced into a further distillation column, wherein acetaldehyde contained in said distillate is distilled and removed, while methyl iodide, methyl acetate and dimethyl ether are withdrawn as bottom products from said further distillation column and recirculated to said carbonylation reactor,

and/or

- the bottoms obtained in step (c) containing the product and vinylacetate are introduced into a next distillation column, from the top of which the vinylacetate is separated and removed."

III. The opposition was filed against the patent as a whole, and based on the ground of lack of inventive step as indicated in Article 100(a) EPC. It was supported by several documents including:

- (1) EP-A-0 487 284, and
- (3) US-A-4 252 748.

IV. Oral proceedings before the Board were held on 15 March 2005. The Appellant, who was duly summoned, did not

attend the oral proceedings as announced in his facsimile dated 9 February 2005.

- V. The Appellant argued in writing that the subject-matter of Claim 1 of the set of claims filed on 22 May 2000 forming the basis for the decision of the Opposition Division lacked inventive step in view of documents (1) and (3).

In this context, he submitted in particular that document (1) disclosed the reduction of tar formation by removing carbonyl impurities, such as acetaldehyde, from a methyl iodide-rich recycle stream to the reactor, and that document (3) disclosed the separation of methyl acetate and methyl iodide by distillation. Moreover, he submitted that document (3) also disclosed that fractional distillation was effective to separate acetaldehyde, vinyl acetate, acetic anhydride, ethylidene diacetate, methyl iodide and methyl acetate from each other.

- VI. During oral proceedings, the Respondent (Patentee) defended the patentability of the subject-matter of the patent in suit on the basis of a new set of Claims 1 to 3, which corresponded to those forming the basis for the decision of the Opposition Division, except that the distillation step (c) of Claim 1 specified under point II above had been amended as follows:

"(c) distilling said volatile phase from step (b) to obtain bottoms containing the product and vinyl acetate, and a distillate containing the unreacted dimethyl ether and/or methyl acetate and methyl iodide".

VII. The Respondent argued that the subject-matter of the present claims involved an inventive step, since the cited state of the art did not provide any incentive to the skilled person that the forming of tars in the reaction mixture could be diminished by reducing acetaldehyde and/or vinyl acetate in the methyl iodide-rich recycle stream by separating in an evaporator a volatile phase from a non-volatile phase containing the rhodium compound, distilling the volatile phase to separate a distillate containing the unreacted starting compound(s) and methyl iodide from bottoms containing the product(s) and vinyl acetate, and separating acetaldehyde from the methyl iodide-rich distillate by a further distillation before recycling said distillate to the reactor and/or separating vinyl acetate from the product(s) by another further distillation.

In this context, he emphasised that according to document (1) the reduction of acetaldehyde concentration in the methyl iodide containing recycle stream involved a mandatory conversion of the carbonyl impurities including acetaldehyde with an amine and, subsequently, the separation of the achieved oximation products by decantation and distillation, and that the recirculation of vinylacetate to the reactor would not be avoided.

Moreover, he submitted with respect to the teaching of document (3), that this document related to a totally different technical problem, namely the separation of acetone from the reaction mixture as a valuable by-product, and in fact did not disclose a distillation of the overhead from an evaporator in a distillation

column to obtain a distillate containing acetaldehyde and methyl iodide and bottoms containing vinyl acetate and a product rendering it possible to reduce the content of acetaldehyde and/or vinyl acetate in the methyl iodide-rich recycle stream to the reactor in order to diminish the forming of tars therein.

VIII. The Appellant requested in writing that the decision of the Opposition Division be set aside and the patent be revoked in its entirety.

The Respondent requested that the patent be maintained with Claims 1 to 3 and pages 2 and 3 of the description as submitted during the oral proceedings, description pages 4 and 5 and Figure 1 as granted.

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

### **Reasons for the Decision**

1. The appeal is admissible.
2. *Admissibility of the late-filed request*
  - 2.1 The Respondent filed his present request consisting of a new Claim 1 and dependent Claims 2 and 3 corresponding to the dependent Claims 2 and 3 as granted at a very late stage, namely during the oral proceedings before the Board and in the absence of the Appellant who did not attend these proceedings as previously announced. One of the issues to be decided is, therefore, whether or not admitting the new request

into the proceedings violates the Appellant's right to be heard (Article 113(2) EPC).

2.2 Present Claim 1 was filed by the Respondent after a discussion with the Board of the objections raised by the Appellant concerning inventive step and in particular after having heard the Board's opinion on this issue. Moreover, the amendment of claim 1 as granted with respect to the distillation step (c) functionally restricts the distillation conditions in that the bottoms obtained in this step contain vinylacetate which can be separated from the product as indicated in the characterising part of Claim 1 as granted. This embodiment of the process of the patent in suit was discussed during the opposition proceedings and also dealt with by the Appellant in his Statement of the Grounds of Appeal. By such a restriction of the subject-matter of Claim 1 as granted the Appellant could not be taken by surprise and a decision could be based on the claims of the present request without getting into conflict with what has been explained in the opinion G 4/92 (OJ EPO 1994/149) in respect of Article 113(1) EPC.

2.3 In these circumstances, the Board considers it appropriate to exercise its discretion to admit the present request into the proceedings.

### 3. *Amendments (Article 123(2) and (3) EPC)*

3.1 Present Claim 1 differs from the subject-matter of Claim 1 of the patent in suit by the feature that the bottoms obtained in the distillation step (c) contain vinylacetate. This amendment finds its support in



Claim 4 and on page 12, lines 1 to 5, of the application as filed, as well as in Claim 1 as granted indicating that the bottoms obtained in this step can be distilled in order to separate vinylacetate from the product.

Present Claims 2 and 3 correspond to Claims 2 and 3 as granted, and are supported by page 10, lines 2 to 5, and Claim 5 of the application as filed.

3.2 Therefore, the Board finds that the subject-matter of the present claims meets the requirements of Article 123(2) and (3).

4. *Inventive step*

4.1 For deciding whether subject-matter claimed involves an inventive step, the Boards of Appeal consistently apply the problem and solution approach, which essentially consists of identifying the closest prior art, determining in the light thereof the technical problem which the claimed invention addresses and successfully solves, and examining whether or not the claimed solution to this problem is obvious for the skilled person in view of the state of the art.

4.2 The Board considers, in agreement with the parties to the proceedings, that the closest state of the art with respect to the claimed subject-matter of the patent in suit is document (1).

4.3 Document (1) relates to a process for producing acetic acid and/or acetic anhydride by continuously reacting methanol or methyl acetate with carbon monoxide in the

presence of a rhodium catalyst, an iodide salt, and methyl iodide, which comprises a treatment of the methyl iodide recycle stream to the carbonylation reactor to reduce the amount of carbonyl impurities in the methyl iodide recycle stream, and consequently to diminish the forming of tars in the reaction fluid which have a detrimental effect on the catalyst activity (see page 3, lines 21 to 44).

The treatment of the methyl iodide recycle stream is carried out by:

- (a) contacting the recycle stream which contains the carbonyl impurities including aldehydes, such as acetaldehyde, crotonaldehyde and butyraldehyde, with an amine compound, preferably an aqueous hydroxylamine salt, and a base, so that the carbonyl impurities are reacted with hydroxylamine to form oximation products which are soluble in the aqueous phase (see page 5, lines 37 to 45, and page 6, lines 24 to 33),
- (b) directing the reaction products to a decanter for separating the organic phase from the aqueous phase containing unreacted hydroxylamine salt as well as most of the oximation products (see page 6, lines 34 to 41),
- (c) directing the separated organic phase containing methyl iodide-rich recycle, minor amounts of water as well as trace amounts of hydroxylamine compound, oximes and other impurities which do not separate with the aqueous phase withdrawn from the decanter, to a distillation tower for removal of these

impurities from the recycle (see page 6, lines 41 to 44), and

- (d) distilling the organic phase in the presence of added water in order to obtain a purified methyl iodide recycle stream which can be recycled to the carbonylation reactor and a bottom stream comprising the separated aqueous oximes as well as other impurities such as alkanes (see page 6, lines 44 to 56).

4.4 Starting from the teaching of this closest state of the art, the Board considers, in agreement with the parties to the proceedings, that the technical problem underlying the patent in suit consists in providing a further process for preparing acetic anhydride and/or acetic acid, in which the forming of tars in the reaction fluid is reduced (see also page 2, lines 52 to 57, of the patent in suit).

4.5 According to Claim 1 of the patent in suit this technical problem is solved by reducing, as a first alternative, the concentration of acetaldehyde and vinylacetate in the methyl iodide-rich recycle stream to the reactor applying the following steps:

- a distillation step as indicated under (c) to obtain bottoms containing the product and vinyl acetate, and a distillate containing the unreacted dimethyl ether and/or methyl acetate, methyl iodide and acetaldehyde due to its low boiling point,

- a further distillation step in order to separate and remove acetaldehyde from said distillate, and recycling the so purified distillate to the reactor, and
  
- removing said bottoms containing vinylacetate and product and, optionally, leading the bottoms to a further distillation column to separate the vinylacetate from the product as indicated in the second characterising step,

or,

by reducing, as a second alternative, the concentration of vinylacetate alone in the methyl iodide-rich recycle stream to the reactor applying the same steps, except that the separation and removal of acetaldehyde is omitted, so that the distillate obtained in the distillation step (c) is recycled to the reactor as such.

Furthermore, in view of the examples of the patent in suit, the Board is satisfied that the technical problem as defined above has been successfully solved within the whole area claimed. This has not been disputed by the Appellant.

- 4.6 The question now is whether the solution of the technical problem underlying the patent in suit involves an inventive step in view of the cited prior art.
  
- 4.7 In challenging the inventive step, the Appellant submitted that document (1) discloses the reduction of

tar formation by removing carbonyl impurities, such as acetaldehyde, from a methyl iodide-rich recycle stream to the reactor, that document (3) discloses a process in which acetaldehyde and methyl iodide are separated by distillation, and that this last mentioned document also teaches that fractional distillation would be effective to separate acetaldehyde, vinyl acetate, acetic anhydride, ethylidene diacetate, methyl iodide and methyl acetate from each other.

- 4.8 However, document (1) discloses that it was difficult to remove the minor amounts of carbonyl impurities by conventional means such as distillation, since the impurities have boiling points close to that of acetic acid and acetic anhydride products (see page 3, lines 6 to 8), and it proposes therefore the process as set out under point 4.3 above, which essentially comprises a conversion of the carbonyl impurities with an amine compound and a separation of the oximation products obtained thereby. Thus, document (1) rather leads away from a distillation step for removing carbonyl impurities including acetaldehyde as such.

Moreover, document (1) does not comprise any suggestion that the recycling of vinyl acetate to the reactor should be reduced in order to avoid the forming of tars in the reaction liquid, let alone a pointer to a separation of this by-product by a distillation step corresponding to step (c) of present Claim 1, making it possible to separate acetaldehyde and vinyl acetate from the methyl iodide-rich recycle stream to the reactor.

Therefore, document (1) cannot render the claimed solution of the technical problem underlying the patent in suit obvious to the skilled person by itself

4.9 Document (3) does not address the technical problem underlying the patent in suit as defined above, so that already for this reason the skilled person would not have any reason to take this document into consideration for the solution of the present technical problem.

In fact, this document relates to a process for recovering acetone produced as by-product in the reaction of methyl acetate with carbon monoxide and hydrogen in the presence of a Group VIII noble metal catalyst and methyl iodide from the reaction mixture by

- supplying acetone to provide an effluent from the reactor having an acetone to methyl iodide molar ratio of at least 1:10;
- distilling said effluent to obtain a distillate containing essentially all of the methyl iodide and some of the acetone and methyl acetate, and a first bottoms product;
- recycling said distillate to the reactor;
- distilling the first bottoms product containing acetone, methyl acetate and the other components of said effluent to obtain a distillate containing the acetone and the methyl acetate, and a second bottoms product;

- subjecting this distillate to an azeotropic distillation in the presence of an alkane or alkene having 5 carbon atoms in order to separate the acetone from the methyl acetate; and
- recovering the desired product(s) from said second bottoms product containing the higher boiling components of said effluent in a convenient manner, e.g. by ordinary distillation (see column 2, lines 11 to 59).

Therefore, like document (1), this document (3) does not suggest to the skilled person a process for preparing acetic anhydride or acetic anhydride and acetic acid comprising a distillation step corresponding to step (c) of present Claim 1, in which a vinylacetate containing fraction is separated from an acetaldehyde fraction rendering it possible to reduce the acetaldehyde and vinyl acetate concentration in the methyl iodide-rich recycle stream.

4.10 It is true that document (3) discloses the separation of acetaldehyde from the reactor effluent (see column 2, lines 35 to 40, column 3, lines 20 to 27, and column 9, lines 9 to 17) and, as in the prior art referred to, that acetaldehyde, vinyl acetate, acetic anhydride, ethylidene diacetate, methyl iodide and methyl acetate could be separated from each other by fractional distillation (see column 1, lines 51 to 54).

However, the acetaldehyde is separated for a different purpose, namely to provide a feed material to be treated for acetone removal (see column 2, lines 37 to 40) instead of a reduction of the tar forming in the

reaction liquid, and its separation is carried out directly after the effluent has left the reactor, i.e. before the distillation step (b) of present Claim 1.

Furthermore, the possibility of separating the specified compounds by fractional distillation as indicated in said document solely represents a general statement concerning prior art at that time without any information about the nature of the compositions to be distilled and the distillation efficiency in separating one or more of said compounds.

- 4.11 Therefore, document (1) alone, or document (1) in combination with document (3), does not provide any suggestion to the skilled person to arrive at the claimed solution of the above defined technical problem underlying the application in suit.
- 4.12 In conclusion, the subject-matter of present Claim 1, and by the same token, that of the dependent Claims 2 and 3, involves an inventive step within the meaning of Articles 52(1) and 56 EPC.



**Order**

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

1. The decision under appeal is set aside.
  
2. The case is remitted to the department of first instance with the order to maintain the patent in the following version:
  - description pages 2 and 3 as submitted during the oral proceedings, and pages 4 and 5 as granted,
  
  - Claims 1 to 3 as submitted during the oral proceedings, and
  
  - drawings Fig. 1/1 as granted.

The Registrar:

The Chairman:

N. Maslin

A. Nuss