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Datasheet for the decision of 17 October 2017

Case Number: T 2422/12 - 3.3.10

Application Number: 07811151.5

Publication Number: 2069281

IPC: C07C51/487, C07C53/08

Language of the proceedings: EN

Title of invention:
PURIFICATION OF ACETIC ACID

Applicant:
LyondellBasell Acetyls, LLC
Millennium Petrochemicals Inc.

Headword:
PURIFICATION OF ACETIC ACID / LYONDELLBASELL ACETYLS

Relevant legal provisions:
EPC Art. 56, 111(1)

Keyword:

Decisions cited:

Catchword:

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Case Number: T 2422/12 - 3.3.10

DECISION
of Technical Board of Appeal 3.3.10
of 17 October 2017

Appellant: (Applicant 1)	LyondellBasell Acetyls, LLC 1221 McKinney Street, Suite 700 Houston, TX 77010 (US)
Appellant: (Applicant 2)	Millennium Petrochemicals Inc. 1221 McKinney Houston TX 77010 (US)
Representative:	Sacco, Marco Basell Poliolefine Italia S.r.l. Intellectual Property P.le Donegani, 12 44122 Ferrara (IT)
Decision under appeal:	**Decision of the Examining Division of the European Patent Office posted on 29 June 2012 refusing European patent application No. 07811151.5 pursuant to Article 97(2) EPC.**

Composition of the Board:

Chairwoman	J. Mercey
Members:	J.-C. Schmid
	C. Schmidt

Summary of Facts and Submissions

I. The appeal lies from the decision of the Examining Division refusing European patent application No. 07811151.5 for lack of inventive step based on the following documents:

(1) US-A- 2003/0199711,
(2) EP-A-0 085 898, and
(3) EP-A-0 322 215.

II. According to the Examining Division, document (1) was the closest prior art to the invention, although document (3) could also be seen as the closest prior art. Document (1) disclosed a process for synthesizing acetic acid from methanol and carbon monoxide, wherein the reaction system comprised water, methyl iodide, a Group VIII metal catalyst and lithium iodide as catalyst stabilizer. This document further taught treating any of the streams of the process to remove the aldehyde impurities, in particular paragraph [0022] on page 3 reading "…any post reaction stream, or a portion thereof, containing aldehyde may be oxidized to convert the aldehyde…".

The subject-matter of claim 1 of the then pending main and auxiliary request differed from the process disclosed in document (1) in that aldehyde impurities were removed by forming an acetal by adding an alcohol to the acetic acid solution and distilling the mixture thus obtained. The technical problem was to provide an alternative method for removing aldehyde impurities from acetic acid. The proposed solution was obvious in the light of document (2), which related to the removal of aldehyde impurities from short chain carboxylic acids by adding 1,2-glycols to the acid, followed by distilling the acetals which were formed.

The subject-matter of claim 1 of the then pending main and auxiliary request lacked therefore an inventive step. The same conclusion applied *mutatis mutandis* to the subject-matter of independent claim 13 of the then pending main and auxiliary request, since the essential distinguishing feature was the same as in claim 1 of these requests.

III. With the statement of the grounds for appeal dated 23 October 2012, the Appellants (Applicants) filed a new main request and an auxiliary request.

Independent claims 1 and 12 (the latter currently incorrectly numbered as claim 11) of the main request read as follows:

"1. A method for producing acetic acid, said method comprising:

(a) reacting methanol and carbon monoxide in the presence of a carbonylation catalyst, a catalyst stabilizer, methyl iodide, water and methyl acetate to produce an acetic acid stream comprising acetic acid, water, methyl acetate, methyl iodide, the catalyst, the catalyst stabilizer, and an aldehyde impurity;

(b) flashing at least a portion of the acetic acid stream to produce a vapor stream comprising acetic acid, water, methyl acetate, methyl iodide and the aldehyde impurity, and a liquid stream comprising the catalyst and the catalyst stabilizer;

(c) separating the vapor stream using a distillation column into an acetic acid product stream comprising acetic acid, the aldehyde impurity, and a minor amount of water, and an overhead stream comprising methyl iodide, water, acetic acid, and methyl acetate;

(d) feeding the acetic acid product stream and a hydroxyl compound selected from the group consisting of 2-methyl-1,3-propanediol, 1,3-propylene glycol, 1,3-butanediol, 1,5 pentanediol, 1,6-hexanediol, 2,2-dimethyl-1,3-propanediol, cyclohexane-1,4-dimethanol, and mixtures thereof into a distillation column, wherein the aldehyde impurity reacts with the hydroxyl compound to form an acetal, and wherein the water is removed from a top portion of the distillation column and an essentially anhydrous acetic acid product stream comprising acetic acid and the acetal is taken from the distillation column; and

(e) separating the acetal from acetic acid by distillation.

"[12]. A method for producing acetic acid, said method comprising:

(a) reacting methanol and carbon monoxide in the presence of a carbonylation catalyst, a catalyst stabilizer, methyl iodide, water and methyl acetate to produce an acetic acid stream comprising acetic acid, water, methyl acetate, methyl iodide, the catalyst, the catalyst stabilizer, and an aldehyde impurity;

(b) flashing at least a portion of the acetic acid stream to produce a vapor stream comprising acetic acid, water, methyl acetate, methyl iodide and the aldehyde impurity, and a liquid stream comprising the catalyst and the catalyst stabilizer;

(c) feeding the vapor stream and a hydroxyl compound selected from the group consisting of 2-methyl-1,3-propanediol, 1,3-propylene glycol, 1,3-butanediol, 1,5 pentanediol, 1,6-hexanediol, 2,2-dimethyl-1,3-propanediol, cyclohexane-1,4-dimethanol, and mixtures thereof into a distillation column;

(d) withdrawing an essentially anhydrous acetic acid product stream comprising acetic acid from a middle portion of the distillation column; and

(e) withdrawing a heavy stream from a bottom portion of the distillation column;

wherein the acetal is in the essentially anhydrous acetic acid product stream, in the heavy stream, or in both."

According to the Appellants, document (1) disclosed a process to produce acetic acid in which acetaldehyde by-product was eliminated by oxidation. The goal of document (1) was to overcome the disadvantages of the prior art processes requiring elimination of the acetaldehyde waste streams. Document (2) disclosed removing traces of aldehydes from unsaturated carboxylic acids by treating them with 1,2-glycols prior to distillation. The teaching of document (2) was limited to 1,2-glycols, while the claimed invention was directed to the use of different kind of glycols. Document (3) disclosed treating acetic acid with ozone in the presence or absence of an oxidation catalyst to purify the acetic acid from impurities, including aldehydes impurities. Accordingly, the claimed subject-matter involved an inventive step over documents (1), (2) or (3), or over the the combination of documents (1) and (2) or (3) and (2).

IV. In a communication dated 11 May 2017, the Board indicated its intention to remit the case to the first instance for further prosecution.

V. With a letter dated 11 July 2017, the Appellants withdrew their request for oral proceedings provided that the case was remitted to the Examining Division for further prosecution.

VI. The Appellants requested that the decision under appeal be set aside and a patent be maintained on the basis of the main request, or subsidiarily, on the basis of the auxiliary request, both requests filed with the letter dated 23 October 2012.

Reasons for the Decision

1. The appeal is admissible.

2. Amendments

The hydroxyl compound required in step (d) of the process of claim 1 of the main request and in step (c) of process of claim 12 (currently incorrectly numbered as claim 11) of the main request has been limited to a compound selected from the group consisting of 2-methyl-1,3-propanediol, 1,3-propylene glycol, 1,3-butanediol, 1,5-pentanediol, 1,6-hexanediol, 2,2-dimethyl-1,3-propanediol, cyclohexane-1,4-dimethanol, and mixtures thereof, according to page 8, lines 19 to 22 of the application as filed.

Therefore, the amendments made to claims 1 and 12 do not generate subject-matter extending beyond the content of the application as filed and the Board concludes that the requirements of Article 123(2) EPC are satisfied.

3. *Inventive step*

The Examining Division refused the application for lack of inventive step based on the combination of documents (1) or (3) with document (2).

Claim 1 of the main request

4. *Closest prior art*

The examining division considered that document (1) was the closest prior art to the invention, although document (3) could also be seen as the closest prior art.

4.1 Document (1) discloses a process for producing acetic acid comprising steps (a), (b) and (c) of present claim 1. The vapours from the flasher are fed to a light ends or splitter column from which crude acetic product is withdrawn as a liquid side drawn or base product and sent to further purification (page 2, right-hand column, lines 8 to 12). The crude aqueous acetic liquid is withdrawn from the light ends column 9 and sent to the acetic recovering system (fig. 1, page 5, left-hand column, lines 1 to 4). At least a portion of the acetaldehyde in the light aqueous phase of the overhead of the light ends column 9 is oxidized to acetic acid, or further to CO_2 and H_2O (see paragraph [0021]).

The Examining Division considered that the removal of the aldehyde impurities from the crude acetic acid was taught in paragraph [0022] on page 3 of document (1) reading "…any post reaction stream, or a portion thereof, containing aldehyde may be oxidized to convert the aldehyde…".

Actually, this passage reads "…any post reaction stream, or a portion thereof, containing **ace**taldehyde may be oxidized to convert the **acet**aldehyde **to acetic acid**…" (emphasis added by the Board). Thus the board holds that document (1) does not aim to purify the crude aqueous acetic acid which is withdrawn from the distillation column 9 through line 10, and accordingly also does not identify any impurities comprised in this acetic acid stream to be further purified.

Document (1) identifies the streams or portions of streams containing acetaldehyde, other than the light aqueous phase, which may be oxidised as the dry organic stream from the overhead of the acetaldehyde stripper (see paragraph [0024]), the overhead stream of the acetaldehyde concentrator [see paragraph 0025], the aqueous phase separated from the heavy phase recycle (see paragraph [0026]), the heavy phase recycle removed from the decanter 13 (see paragraph [0028]), or the heavy organic phase from the flasher 6 (see bottom of paragraph [0019].

This document is not concerned with the purification of the crude aqueous acetic liquid which is withdrawn from the distillation column 9 through line 10 and which corresponds to the acetic acid product stream referred to in steps (c) and (d) of claim 1 of the main request. Document (1) discloses only that the crude acetic acid is withdrawn and sent to further purification without any further indication of how this purification is carried out (see page 2, right-hand column, lines 8 to 12; page 5, left-hand column, lines 1 to 4).

4.2 Document (3) also discloses a process for producing acetic acid comprising steps (a), (b) and (c) of claim 1 (see page 4, lines 54 and 55). The crude acetic acid is drawn off from the splitter column for final purification. Water is removed, preferably by distillation (page 5, line 10 to 23) and then the crude dry acetic acid which still contains impurities is subjected to ozonolysis, preferably in the presence of a catalyst. The carbonyl impurities present in the crude dry acetic acid are mainly crotonaldehyde, ethyl crotonaldehyde and 2-methyl-2-pentanal (see page 5, lines 18 to 23; examples 2, 4 and 5, table 1). According to example 1, acetic acid obtained from a low water carbonylation of methanol was spiked to contain 231 ppm crotonaldehyde and 224 ppm ethyl crotonaldehyde and treated with ozone. The content of these aldehydes in the treated acetic acid decreases to 9 ppm and 3 ppm, respectively (see table 1 on page 7).

4.3 Thus, document (3), which discloses steps (a) to (c) and water removal by distillation from the acetic acid stream obtained after step (a) to (c), followed by the purification of the dried acetic acid is closer to the subject-matter of claim 1 than document (1).

5. Technical problem underlying the subject-matter of claim 1

The technical problem underlying the application is seen in the provision of an alternative process for the production of acetic acid having reduced content of aldehyde impurities.

6. *Solution*

The solution is the process of claim 1 characterized by steps (d) and (e), in particular that a hydroxyl compound selected from the group consisting of 2-methyl-1,3-propanediol, 1,3-propylene glycol, 1,3-butanediol, 1,5-pentanediol, 1,6-hexanediol, 2,2-dimethyl-1,3-propanediol, cyclohexane-1,4-dimethanol, and mixtures thereof is added to the acetic acid product stream after step (c), and is fed into the distillation column for removing water (step d) and the acetal is separated from the acetic acid by distillation (step e).

7. *Obviousness*

Document (2) (see claim 1) discloses a process for removing aldehyde impurities from a carboxylic acid by adding a 1,2-glycol into the acid, reacting by heating and subsequently distilling the acid, which hence has a lower content of aldehyde impurities.

In example 1 of document (2), crude acrylic acid, having an aldehyde content of 0.98% was reacted with ethylene glycol. After distillation at a vapour temperature of 33°C, the aldehyde content of the distillate was 0.038% (380 ppm). Accordingly, document (2) does not disclose a process where a diol is added to an aqueous carboxylic acid during or before distillation to remove water.

Furthermore document (2) does not disclose the use of the specific 1,3-, 1,4-, 1,5 or 1,6-diols required by claim 1 of the main request, but only a 1,2-diol.

Under these circumstances, the Board concludes that document (2) neither discloses, nor suggests the proposed solution, in particular step (d) of the claimed process.

Claim 12 (incorrectly numbered as claim 11) of the main request

The subject-matter of claim 12 relates to the embodiment of the so-called "combined column".

8. Document (1) is closer to the subject-matter of claim 12 than document (3), since it discloses that a bottom fraction comprising some acetic acid and higher boiling components is withdrawn from the distillation column 9 (see page 5, left-hand column, lines 4 to 8, figure 1).

However, document (1) discloses that a crude aqueous acetic liquid is withdrawn from the light ends column 9 (page 5, left-hand column, lines 1 to 4), while step (d) of claim 12 requires that an essentially anhydrous acetic acid product stream comprising acetic acid is withdrawn from the middle portion of the distillation column.

9. *Technical problem underlying the subject-matter of claim 12*

The technical problem is seen in the provision of an alternative process for the production of acetic acid having reduced content of aldehyde impurities.

10. *Solution*

The solution is the process of claim 12 characterized in that a hydroxyl compound selected from the group consisting of 2-methyl-1,3-propanediol, 1,3-propylene glycol, 1,3-butanediol, 1,5-pentanediol, 1,6-hexanediol, 2,2-dimethyl-1,3-propanediol, cyclohexane-1,4-dimethanol, and mixtures thereof is added to the vapour stream containing acetic acid, water, methyl acetate, methyl iodide and the aldehyde impurities, said mixture is fed to a distillation column, and an essentially anhydrous acetic acid product stream comprising acetic acid is withdrawn from the middle portion of the distillation column.

11. *Obviousness*

Document (1) indicates that any post reaction stream, or a portion thereof, containing acetaldehyde may be oxidized to convert the acetaldehyde to acetic acid or further to CO_2 and H_2O (see paragraph [0022]). Although the vapour stream directly obtained from the flasher obviously contains acetaldehyde, document (1) does not include this stream in the list of the envisioned acetaldehyde-containing streams (see point 4.1 above).

Document (1) also does not contemplate withdrawing an essentially anhydrous acetic acid product stream comprising acetic acid from the middle portion of the distillation column.

Furthermore document (2) does not suggest that 1,2-glycols, let alone a hydroxyl compound listed in claim 12 of the main request, may be added to a vapour stream comprising acetic acid, water, methyl acetate, methyl iodide and aldehydes in order to produce acetic acid having reduced content of aldehyde impurities.

The Board concludes that the subject-matter of claim 12 of the main request is not rendered obvious by the combination of document (1) with document (2).

12. *Remittal*

The essential function of an appeal is to review the decision issued by the first-instance department.

The Board considers that the amended claims submitted by the Appellant in the appeal proceedings as the main request remove the inventive step objection based on the combination of document (1) with document (2), or document (3) with document (2), on which the decision under appeal was based.

Thus, under the present circumstances the Board finds it appropriate to remit the case to the examining division for further prosecution on the basis of the claims of the main request.

Order

For these reasons it is decided that:

1. The appeal under appeal is set aside.

2. The case is remitted to the examining division for further prosecution.

The Registrar: The Chairwoman:

Beschwerdekammern des Europäischen Patentamts
Boards of Appeal of the European Patent Office
Chambres de recours de l'Office européen des brevets

C. Rodríguez Rodríguez J. Mercey

Decision electronically authenticated