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**Datasheet for the decision
of 05 May 2010**

Case Number: T 0716/07 - 3.3.01

Application Number: 00309265.7

Publication Number: 1094061

IPC: C07D 207/26

Language of the proceedings: EN

Title of invention:

Process for recovering N-Vinyl-2-Pyrrolidone

Patentee:

NIPPON SHOKUBAI CO., LTD.

Opponent:

BASF SE

Headword:

Distillation of N-vinylpyrrolidone/NIPPON SHOKUBAI

Relevant legal provisions:

EPC Art. 100(a)

Relevant legal provisions (EPC 1973):

-

Keyword:

"Inventive step (yes) - the comparison between an example of the prior art with those of the patent shows an unexpected effect"

Decisions cited:

T 0197/86, T 1188/00

Catchword:

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Case Number: T 0716/07 - 3.3.01

D E C I S I O N
of the Technical Board of Appeal 3.3.01
of 05 May 2010

Appellant:
(Opponent)

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(Patent Proprietor)

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Decision under appeal:

Interlocutory decision of the Opposition
Division of the European Patent Office posted
21 February 2007 concerning maintenance of
European patent No. 1094061 in amended form.

Composition of the Board:

Chairman: P. Ranguis
Members: C. M. Radke
R. Menapace

Summary of Facts and Submissions

- I. The patent in suit relates to a process for recovering N-vinyl-2-pyrrolidone (NVP) from a liquid mixture by continuous distillation under reduced pressure.
- II. The opponent appealed against the interlocutory decision of the opposition division that the European patent no. 1 094 061 as amended according to the first auxiliary request met the requirements of the EPC.
- III. Said first auxiliary request contained five claims, the only independent claim reading as follows:

"1. A process comprising recovering N-vinyl-2-pyrrolidone by continuously distilling under a reduced pressure of 2.66×10^4 Pa or less a liquid mixture comprising N-vinyl-2-pyrrolidone, N-(2-hydroxyethyl)-2-pyrrolidone, one or more compounds having a higher boiling point than N-vinyl-2-pyrrolidone and water, using a distilling column, which process is characterised in that the temperature of the bottom liquid of the distilling column is controlled to be 180 °C or less, and the bottom residue comprising N-vinyl-2-pyrrolidone, N-(2-hydroxyethyl)-2-pyrrolidone and the compounds having a higher boiling point than N-vinyl-2-pyrrolidone is withdrawn from the distilling column and which further comprises the continuous distillation of the bottom residue withdrawn from the distilling column using a second distilling column to make N-vinyl-2-pyrrolidone distil as the distillate."

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

(D1) GB-A-717 799

(D6) W. R. A. Vauck und H. A. Müller, Grundoperationen chemischer Verfahrenstechnik, 10th edn. (1994), Deutscher Verlag für Grundstoffindustrie, Leipzig/DE, 720-725

(D7) US-A-5 951 828

(D10) Encyclopedia of Polymer Science and Engineering, revised edition, vol. 17 (1989), John Wiley & Sons, New York/US, 203-204

(D12) Kirk-Othmer, Encyclopedia of Chemical Technology, 2nd edn., vol. 16 (1968), 853-854.

V. The opponent had requested revocation of the patent in its entirety based on grounds under Article 100(a) EPC (lack of inventive step).

VI. The opposition division considered the subject-matter of the claims of the main request then on file not to be inventive in view of document (D6).

Document (D7) was considered as the closest prior art for the claims of the first auxiliary request. The problem to be solved was the provision of an alternative process for recovering N-vinyl pyrrolidone by continuous distillation. The subject-matter of the claims was deemed to be inventive as document (D7) did not hint at a two step distillation process.

VII. During the appeal proceedings the following document was additionally cited:

(D13) DE-A-1 445 737.

VIII. The claims on file are claims 1 to 3 filed as the sole request during the oral proceedings before the Board on 05 May 2010. The only independent claim reads as follows:

"1. A process comprising recovering N-vinyl-2-pyrrolidone by continuously distilling under a reduced pressure of 2.66×10^4 Pa or less a liquid mixture comprising N-vinyl-2-pyrrolidone, N-(2-hydroxyethyl)-2-pyrrolidone, one or more compounds having a higher boiling point than N-vinyl-2-pyrrolidone and water, using a distilling column, which process is characterised in that the temperature of the bottom liquid of the distilling column is controlled to be 180 °C or less, and the bottom residue comprising N-vinyl-2-pyrrolidone, N-(2-hydroxyethyl)-2-pyrrolidone and the compounds having a higher boiling point than N-vinyl-2-pyrrolidone is withdrawn from the distilling column

which further comprises continuously distilling under a reduced pressure of 1.33×10^4 Pa or less the residue withdrawn from the distilling column (the first distilling column) using a second distilling column such that N-vinyl-2-pyrrolidone is the distillate, and withdrawing the bottom residue of the second distilling column comprising N-(2-hydroxyethyl)-2-pyrrolidone and the compounds having a higher boiling point than N-vinyl-2-pyrrolidone, wherein during the distillation

the temperature of the bottom liquid of the second distilling column is controlled to be 230 °C or less."

IX. The arguments of the Appellant as far as relevant for this decision may be summarised as follows:

The mixture to be separated and its distillation was known from document (D1). Document (D7) disclosed in example 3 the separation of a similar mixture containing N-vinyl-2-pyrrolidone; for this reason it might be considered to represent the closest prior art. The process disclosed in document (D7) comprised one distillation step whereas the present claims required two. The problem to be solved was to provide an alternative process. The solution of this problem as defined in the present claims was obvious in view of

- either document (D6) which disclosed that a three component mixture might be separated by distillation in two columns
- or document (D13) which disclosed in example 5 to conduct the distillation stepwise.

It was known from document (D12) that N-vinyl-2-pyrrolidone should not be distilled at temperatures above 120°C in order to avoid polymerisation. Therefore, the effect shown in the comparative tests in the patent in suit was predictable.

As alternatives, the closest prior art for assessing inventive step could be either document (D6) or document (D13).

It was obvious to combine the teaching of document (D6) or (D13) with that of

- document (D1) which disclosed the mixture to be separated in the process of the patent in suit, and
- document (D12) which taught to distil at low temperatures in order to prevent the formation of polymers. Hence, the subject-matter of the claims did not involve an inventive step.

X. During the oral proceedings before the Board, the Respondent did not maintain its request not to admit document (D13) into the proceedings.

The Respondent deemed document (D7) to represent the closest prior art as it was the only cited document relating to the continuous distillation of a thermolabile compound.

N-vinyl-2-pyrrolidone was thermolabile as its polymerisation was accelerated by heat or moisture. Moreover, it tended to hydrolyse (see documents (D10) and (D12)). Both polymerisation and hydrolysis led to a decrease of the portion of the N-vinyl-2-pyrrolidone recovered in the course of the distillation.

The objective problem solved was to provide an improved process for the recovery of N-vinyl-2-pyrrolidone from the mixture specified in claim 1 efficiently, steadily and in high purity. A comparison of the examples in the patent in suit with example 3 of document (D7) showed

that the process claimed allowed for a much higher recovery of highly pure N-vinyl-2-pyrrolidone.

Document (D7) as such did not give the skilled person any hint to use two distillation columns.

Document (D6) did not motivate the person skilled in the art to apply the present process as it did not address the problem of distilling thermolabile monomers.

Document (D13) was less relevant as it did not deal with continuous distillation and because it only taught to distil off the alcohol if the boiling points of the vinyl compounds were close. Moreover, example 5 relates to the separation of N-vinyl-2-pyrrolidone from a mixture containing methanol and N-(2-methoxyethyl)-2-pyrrolidone; this mixture could not be considered to be similar to the water containing one to be separated in the claimed process.

The Respondent concluded that the subject-matter claimed in the patent in suit was inventive.

XI. The Appellant requested that the decision under appeal be set aside and the patent be revoked.

The Respondent requested that the decision under appeal be set aside and the patent be maintained on the basis of claims 1 to 3 of the sole request and an amended description, both filed during the oral proceedings before the Board, and figure 1 as granted.

XIII. At the end of the oral proceedings the Chairman announced the decision of the Board.

Reasons for the Decision

1. The appeal meets the requirements under Articles 106 to 108, Rules 97 and 99 EPC. Hence it is admissible.

2. *Article 123 EPC*

No objections under this Article were raised against the amended claims and description.

Claim 1 is based on original claims 1, 3 and 4, page 5, lines 15-19 and page 10, lines 26-29 of the application as originally filed. Claims 2 and 3 are based on original claims 2 and 5.

The amendments in the claims limit the scope of claim 1 as granted by the additional features of granted claims 3 and 4.

The amendments in the description merely adapt it to the amended claims.

Hence, the claims and description of the sole request do not contravene the requirements of Article 123 EPC.

3. *Novelty*

It was not disputed that the subject-matter of the claim was novel.

Present claim 1 is directed to a process comprising recovering N-vinyl-2-pyrrolidone by continuous distillation (see under point VIII above). The only document cited which discloses such a process is document (D7) where N-vinyl-2-pyrrolidone is distilled in a single distillation column.

This document discloses in example 3 the continuous distillation of N-vinyl-2-pyrrolidone, where

- N-vinyl-pyrrolidone was continuously pumped into the lower third of a packed column, and
- liquid formamide was continuously metered into the vapouriser at the top of the column.

The pressure at the top of the column was 2 mbar. In the upper quarter of the column, N-vinyl-2-pyrrolidone of a purity of more than 99 % (containing 0.06 % of formamide) was recovered as a liquid side stream. Under steady state conditions, the pressure at the bottom of the column was 10 mbar and the temperature in the column was 95°C.

The subject-matter of present claim 1 differs from the disclosure in (D7) in that it requires an additional column to distil off water. Hence, the subject-matter of claim 1 and of dependent claims 2 and 3 is novel.

4. *Inventive step*

4.1 The closest prior art

The closest state of the art is normally a prior art document disclosing subject-matter with the same objectives as the claimed invention and having the most relevant technical features in common.

The objective of the patent in suit was to recover thermally instable N-vinyl-2-pyrrolidone by continuous distillation from its reaction mixture (see paragraphs [0001], [0007] and [0008]).

Document (D6) discloses the distillation of a ternary mixture. It neither mentions N-vinyl-2-pyrrolidone nor the distillation of any other thermally instable compounds.

Document (D13) does not relate to continuous distillation.

In contrast thereto, document (D7) discloses the continuous distillation of N-vinyl-2-pyrrolidone and addresses the thermolability of this compound (see claim 7, example 3 and column 1, lines 34-36).

Therefore, document (D7) rather than document (D6) or (D13) represents the closest prior art.

4.2 The problem to be solved

The Respondent argued that a comparison between example 3 of document (D7) with the examples of the

patent in suit showed that a higher percentage of N-vinyl-2-pyrrolidone is recovered by means of the process claimed in the patent in suit (see point X above).

In particular, the Respondent pointed out that

- in example 3 of document (D7) 300 g/h of N-vinyl-2-pyrrolidone were fed into the distillation column whereas only 185 g/h of pure N-vinyl-2-pyrrolidone were withdrawn from the column under steady state condition, which meant that only 62% of the N-vinyl-2-pyrrolidone was recovered (see (D7), column 4, line 46, to column 5, line 7, in particular column 4, lines 56-58, and column 5, lines 3-6).

In contrast to this

- in examples 1 and 2 of the patent in suit, 96% and 95%, respectively, of the N-vinyl-2-pyrrolidone was recovered at a purity of 99.9% by weight (see page 8, lines 20-21, and page 10, lines 14-15).

This finding can be used to reformulate the problem to be solved only if

- said examples of document (D7) and of the patent in suit are comparable to the extent that the alleged effect is convincingly shown to have its origin in the distinguishing feature of the invention (see T 197/86, OJ EPO 1989, 371, point 6.1.3 of the reasons),

and, in the affirmative, that

- it is probable that this effect is achieved over the whole range of the claims of the patent in suit, and
- that the effect is related to the problem to be solved as disclosed in the respective application as filed

(see T 1188/00 of 30 April 2003, point 4.5 of the reasons).

4.2.1 Hence, it is first to be assessed whether or not the comparison of the example 3 of document (D7) with examples 1 and 2 of the patent in suit shows that this increase in the portion of N-vinyl-2-pyrrolidone recovered in high purity is due to the distinguishing feature.

A feature which distinguishes the subject-matter claimed from the disclosure in document (D7) as the closest prior art is that the claims of the patent in suit require that an additional column is present in which water is distilled off (see under point 3 above).

However, the process of examples 1 and 2 of the patent in suit does not differ only from that of example 3 of document (D7) in that an additional column was used but also in that

- the mixtures to be separated in the patent in suit contained water whereas no presence of water is reported in document (D7), and in that

- the distillation temperatures in the examples of the patent in suit were higher.

Consequently, the **increased** portion of high purity N-vinyl-2-pyrrolidone recovered in examples 1 and 2 of the patent in suit can only be clearly attributed to the distinguishing feature if the presence of water and the higher distillation temperature do not contribute to this effect.

As both parties pointed out, document (D12) discloses that N-vinyl-2-pyrrolidone may

- hydrolyse in acidic aqueous solutions into 2-pyrrolidone and acetaldehyde, and may
- polymerise at high temperatures so that vacuum distillation at "temperatures above 120°C should be avoided" (see page 854, under the heading "*Stability*").

Hence, the presence of water in the mixture separated in the examples of the patent in suit is rather expected to **decrease** the portion recovered due to hydrolysis of part of the N-vinyl-2-pyrrolidone.

The increase in temperature from

- 95°C in example 3 of (D7) (measured in the bottom of the column in example 3 of (D7); see column 5, lines 3-6), to

- 160°C and 195°C in the distillation columns used in example 1, and 178°C and 205°C in example 2 (see Table 2 on page 8 and Table 4 on page 9),

i.e. to temperatures well above those recommended in document (D12) in order to inhibit polymerisation of N-vinyl-2-pyrrolidone, is also expected to **decrease** the portion of N-vinyl-2-pyrrolidone recovered.

Hence, the observed **increase** in the recovered portion of N-vinyl-2-pyrrolidone can be attributed to the fact that an additional distillation column is used in examples 1 and 2 of the patent in suit, namely to the feature distinguishing the subject-matter of the present claims over the disclosure of document (D7).

- 4.2.2 Then it is to be determined whether it is probable that this effect is achieved over the whole range of the claims of the patent in suit.

The only independent claim requires

- a pressure of 2.66×10^4 Pa or less and a temperature of 180°C or less in the first column, and
- a pressure of 1.33×10^4 Pa or less and a temperature of 230°C or less in the second column (see under point VIII above).

Document (D12) recommends **vacuum** distillation of N-vinyl-2-pyrrolidone. There is no indication in this document that the application of a reduced pressure might affect the stability of N-vinyl-2-pyrrolidone. High temperatures do, however, accelerate its

polymerisation and thus lead to a decrease the recovery of N-vinyl-2-pyrrolidone (see page 854, under the heading "*Stability*").

The increased recovery of N-vinyl-2-pyrrolidone can be considered to be demonstrated over the whole range of the claims if the respective examples work under the most unfavourable conditions, namely close to the maximum temperatures indicated in the claims. This is indeed the case in example 2 where the bottom liquid temperature of the first column is 178°C in the first and 205°C in the second column (which is close to the maximum values of 180°C and 230°C indicated in present claim 1).

Hence, it has been shown that the effect is achieved over the whole range of the claims.

- 4.2.3 Finally, it is to be assessed whether the effect is related to the problem to be solved as disclosed in the respective application as filed.

According to the application as filed, the problem to be solved was to provide " ... a process ... which makes it possible to recover chemically unstable N-vinyl-2-pyrrolidone efficiently and steadily and in high purity ..." (see page 3, lines 5-11).

The demonstrated increase in the portion of high purity N-vinyl-2-pyrrolidone recovered thus is related to the problem initially disclosed.

4.2.4 Therefore, the problem posed and solved in view of document (D7) as the closest prior art may be redefined as to provide a continuous distillation of a liquid composition containing N-vinyl-2-pyrrolidone, N-(2-hydroxyethyl)-2-pyrrolidone and water, resulting in an increased recovery of highly pure chemically unstable N-vinyl-2-pyrrolidone.

4.2.5 The cause of this problem as disclosed in document (D7) is the thermolability of N-vinyl-2-pyrrolidone, namely its tendency to decompose at high temperatures (see column 1, lines 26-33 and 57-59).

4.3 Was the solution obvious?

The Appellant argued that it was obvious for the person skilled in the art to combine the disclosure of document (D7) with that of (D6) or (D13) and thus to end up with the subject-matter of the present claims (see under point IX above).

The fact that the features of a claim of a patent not disclosed in the closest prior art are preferred features of another document of the prior art does not suffice to consider the subject-matter of this claim to be obvious. Obviousness requires additionally that said other document gives an indication that said preferred features contribute to the solution of the problem solved in the patent. Such an indication can be an explicit one if the other document mentions that said preferred features solve the problem, it can be an implicit one if said other document informs how to avoid conditions which are known to be a cause of the problem.

4.3.1 Document (D7) as such gives no indication that the distillation might be performed using two distillation columns.

4.3.2 Document (D6)

Document (D6) mentions that the continuous countercurrent distillation of multicomponent mixtures containing N components requires $N-1$ columns if all the components are to be obtained in pure form. Otherwise, when a single column is used, either the top or the bottom product consist of more than one component (see the bottom paragraph on page 720). Figure 10.71 on page 721

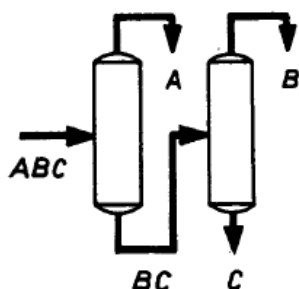


Bild 10.71. Kontinuierliche Gegenstromdestillation eines Dreistoffgemischs ABC mit den reinen Komponenten A und B als Kopfprodukt

shows a system of two columns for separating a three component mixture ABC , where the bottom product of the first column containing B and C is fed to a second column separating these components by distilling off component B .

However, document (D6) does not relate to the problem of increasing the recovery of thermolabile monomers, let alone of N -vinyl-2-pyrrolidone. Thus, it gives no explicit indication to the person skilled in the art in charge of solving this problem to modify the process disclosed in document (D7) by splitting up the

distillation into two distillation steps in two different columns.

Moreover, it does not address the conditions which are known to have an effect on the recovery of N-vinyl-2-pyrrolidone, i.e. to the presence or absence of heat, light, air or water during distillation (see document (D10), the third paragraph on page 204; see document (D12), page 854, under the heading "*Stability*").

Therefore, the person skilled in the art would not consult document (D6) when solving the problem mentioned above.

- 4.3.3 Document (D13) claims a process for making N-vinyl compounds by decomposition of the respective alpha-alkoxyalkyl compounds (see claim 1). In example 5 the catalytical elimination of methanol from N-2-methoxyethyl-2-pyrrolidone yields N-vinyl-2-pyrrolidone. Most of the methanol is distilled off during the reaction and the product is purified by fractional distillation.

The Appellant argued that document (D13) suggested to distil off the lowest boiling compound first (namely the alcohol) and thus to perform the distillation in two steps which required two columns if performed continuously (see under point IX above).

However, there is no hint in document (D13) that a two step distillation process might increase the recovery of thermolabile N-vinyl-2-pyrrolidone.

Therefore, also document (D13) does not give an indication that a modification of the process disclosed in document (D7) by splitting up the distillation into two steps might be helpful for solving the problem mentioned above.

4.3.4 Consequently, the subject-matter of claim 1 involves an inventive step. The same holds for the subject-matter of dependent claims 2 and 3.

5. *The description*

Neither did the Appellant object to the amendments in the description as submitted during the oral proceedings before the Board, nor does the Board see any reason to do so. The amendments merely adapted the description to the amended claims.

6. *Remittal to the department of first instance
(Article 111(1) EPC)*

In the present case, the Board cannot decide on the maintenance of the patent as amended because the prerequisites according to Rule 82(2), second sentence, are not yet fulfilled. Therefore, it remits the case to the department of first instance

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 with
 - claims 1 to 3 and an amended description, both filed during the oral proceedings before the Board, and

 - figure 1 as granted.

The Registrar:

The Chairman:

B. Atienza Vivancos

P. Ranguis