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D E C I S I O N
of 22 June 1999

Case Number: T 0999/96 - 3.3.1

Application Number: 91310657.1

Publication Number: 0487305

IPC: C07D 303/16

Language of the proceedings: EN

Title of invention:

Method of purifying crude glycidyl (meth)acrylate

Patentee:

Mitsubishi Gas Chemical Company, Inc.

Opponent:

NOF Corporation

Headword:

Glycidyl (meth)acrylate/MITSUBISHI

Relevant legal provisions:

EPC Art. 54(1), (2), 56, 123(2), (3)

Keyword:

"Amendments - added subject-matter (no) - extension of protection (no)"

"Novelty (yes) - method features as claimed not disclosed in the prior art"

"Inventive step (yes) - claimed purification method not obvious"

Decisions cited:

-

Catchword:

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Boards of Appeal

Chambres de recours

Case Number: T 0999/96 - 3.3.1

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 22 June 1999

Appellant: NOF Corporation
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Decision under appeal: Interlocutory decision of the Opposition Division
of the European Patent Office posted 24 September
1996 concerning maintenance of European patent
No. 0 487 305 in amended form.

Composition of the Board:

Chairman: A. J. Nuss
Members: P. P. Bracke
S. C. Perryman

Summary of Facts and Submissions

I. The appeal lies from the Opposition Division's interlocutory decision, announced orally on 24 July 1996, with the reasoned decision being issued on 24 September 1996, that, account being taken of the amendments made by the Patentee during the opposition proceedings, European patent No. 0 487 305 was found to meet the requirements of novelty and inventive step over *inter alia* documents

(2) US-A-4 755 262;

(3) GB-A-2 113 222;

(5) the translation of JP-A-48-36117; and

(6) the translation of JP-A-53-90213.

II. During oral proceedings before the Board of Appeal, which took place on 22 June 1999, the Respondent (Proprietor) filed a set of 5 claims, with the only independent claim reading:

"1. A method of purifying a crude glycidyl (meth)acrylate, which comprises:

(1) subjecting a crude glycidyl (meth)acrylate containing as impurities epichlorohydrin and other chlorine compounds selected from 1,3-dichloro-2-propanol, 2,3-dichloro-1-propanol, glycerin monochlorohydrin, 2-hydroxy-3-chloropropyl (meth)acrylate and 3-hydroxy-2-chloropropyl (meth)acrylate to a stripping treatment with a mixed

gas containing oxygen gas in the presence of a quaternary ammonium salt wherein the mixed gas is used at a flow rate of 0.1 to 500 ml/minute (as a flow rate measured at 20°C under atmospheric pressure) per kilogram of the crude glycidyl (meth)acrylate, and then

(2) distilling the treated product to obtain a purified glycidyl (meth)acrylate."

III. The Appellant objected that Claim 1 filed at the oral proceedings did not meet the requirement of Article 123(2) EPC and that the claimed method was neither novel nor inventive over the cited prior art.

More particularly, he argued that Claim 1 was not novel over document (6), since in the only example therein all method features of Claim 1 were disclosed.

Furthermore, he was of the opinion that document (2) represented the closest state of the art when dealing with inventive step. Since (a) the conversion of chlorine compounds into epichlorohydrin was known from documents (2) and (5), (b) distillation was known from document (2) as a method to isolate purified glycidyl (meth)acrylate, (c) distillation is only one form of stripping and since (d) it was furthermore known from document (3) that residual epichlorohydrin in glycidyl (meth)acrylate may be reduced to very low levels by a stripping treatment, he was of the opinion that the claimed process was not inventive over the combined teaching of documents (2), (3) and (5).

In support of his submission that distillation is only one form of stripping he referred to the Condensed

Chemical Dictionary, 10th edition, 1981, page 972, giving for stripping the definition "removal of relatively volatile components from a gasoline or other liquid mixture by distillation, evaporation, or by passage of steam, air or other gas through the liquid mixture."

- IV. The Respondent submitted that by the amendments in Claim 1 no subject-matter was added which extends beyond the content of the application as filed and that document (6) disclosed neither a method of purifying a crude glycidyl (meth)acrylate nor a stripping treatment and, consequently, that the claimed method was novel over the teaching of this document.

Moreover, he agreed that for the question of inventive step document (2) represented the closest state of the art. Since document (2) was only concerned with the removal of epichlorohydrin and not with the removal of the chlorine compounds cited in Claim 1 and since neither the conversion of chlorine compounds to epichlorohydrin in order to remove the chlorine compounds nor a stripping treatment was suggested therein, he was of the opinion that the claimed method could not have been deduced from the teaching of document (2) and, consequently, that there was no incentive to combine the teaching of document (2) with any of documents (3) or (5).

- V. The Appellant requested that the decision under appeal be set aside and the European patent No. 0 487 305 be revoked.

The Respondent requested that the decision under appeal

be set aside and that the patent be maintained on the basis of the claims and amended description submitted at the oral proceedings on 22 June 1999.

Reasons for the Decision

1. The appeal is admissible.
2. *Article 123(2) and (3) EPC*
 - 2.1 Present Claim 1 differs from granted Claim 1, which is identical with Claim 1 as originally filed, by the specification that
 - (a) the other chlorine compounds are selected from 1,3-dichloro-2-propanol, 2,3-dichloro-1-propanol, glycerin monochlorohydrin, 2-hydroxy-3-chloropropyl (meth)acrylate and 3-hydroxy-2-chloropropyl (meth)acrylate and
 - (b) the mixed gas is used at a flow rate of 0.1 to 500 ml/minute (as a flow rate measured at 20°C under atmospheric pressure) per kilogram of the crude glycidyl (meth)acrylate.

It has never been contested that a basis for the mixed gas as specified under item (b) above can be found on page 6, lines 12 to 15, of the application as filed. However, the Appellant submitted that by the wording "the other chlorine compounds are **selected from**" (emphasis added) Claim 1 contained subject-matter extending beyond the application as filed, since on

page 4, lines 23 to 30, of the application as filed it was not said that the other chlorine compounds were selected from those mentioned in item (a) above but that they were **typical examples** thereof.

However, the amendment consisting in merely specifying in Claim 1 that impurities other than epichlorohydrin are "selected from" the indicated group of chlorine compounds does not introduce subject-matter extending beyond the content of the application as filed, since in the originally filed application the same group of compounds is used for defining what are "typical examples" of these impurities. Whereas in both cases a skilled person gets the information that the other chlorine compounds may consist of the ones cited, in the case of the amendment he gets the information that other possible chlorine compounds are restricted to those cited as typical examples of such impurities. The result of the amendment is thus to limit the protection conferred in comparison to that conferred by the patent as granted.

- 2.2 Claims 2 to 5 correspond with Claims 2 to 5 of the application as filed.
- 2.3 By the amendments in columns 2, 3 and 4 of the patent in suit the description is brought into conformity with the wording of the claims, which was not contested by the Appellant.
- 2.4 Therefore, the Board concludes that the requirement of Article 123(2) EPC is met by all the claims.

Since, as set out above, the amendment of the claims

result in a restriction of the protection conferred by the patent, the claims also meet the requirement of Article 123(3) EPC.

3. *Novelty*

3.1 Document (6) concerns a method of reacting almost quantitatively an unsaturated organic acid and excess epichlorohydrin in the presence of a quaternary ammonium salt and then subjecting the ester to azeotropic dehydration with epichlorohydrin in the presence of an alkali while blowing air into the reaction solution during the azeotropic dehydration reaction (see Claim 1, page 2, lines 16 to 24, and page 3, lines 6 to 12).

The only example on page 4, lines 7 to 23, describes a method of preparing glycidyl methacrylate, wherein

- 100 moles methacrylic acid and 1000 moles epichlorohydrin are reacted in the presence of triethylbenzylammonium chloride;
- thereafter air is blown into the reaction solution and aqueous caustic soda is added dropwise;
- of the epichlorohydrin and water, which are removed by azeotropic distillation, only the epichlorohydrin is returned to the reaction system; and
- by-product sodium chloride is removed by filtration from the reaction mixture, which is then subjected to distillation.

3.2 The Appellant argued that a skilled person would understand the blowing of air into the reaction solution, according to the example in document (6), as a stripping treatment with a mixed gas containing oxygen gas. Since a quaternary ammonium salt was still present in the reaction mixture at the moment of blowing air into it and since the reaction mixture was subsequently subjected to a distillation step, he was of the opinion that all method features described in Claim 1 were disclosed in that example.

However, in the judgement of the Board, since the example is completely silent about the purpose or the intended effect of the air blown into the solution, the general teaching of this document is to be taken into consideration. As it follows from the paragraph bridging pages 3 and 4 of document (6) that by blowing air into the solution a polymerisation is inhibited and that the air blowing should be controlled to an extent that epichlorohydrin is not scattered, the Board cannot accept that a skilled person would understand the blowing of air into the reaction solution as a stripping treatment.

In this respect the Appellant argued that a skilled person would interpret the passage on page 4, lines 1 to 4, saying "it is necessary to control air blowing to an extent that epichlorohydrin is not scattered off" as meaning that, although a complete stripping should be avoided, a partial stripping is possible. However, even if air is blown into the solution during the azeotropic distillation of water and epichlorohydrin, such a "partial stripping" could only be considered as a removal of either water or of an azeotropic mixture

consisting of water and epichlorohydrin, with the epichlorohydrin being subsequently separated from the water and returned to the reaction mixture, but not as the removal of epichlorohydrin and other chlorine compounds as defined in present Claim 1.

Therefore, the Board comes to the conclusion that a stripping treatment for purifying glycidyl (meth)acrylate, as described in present Claim 1, is not described in the example of document (6) and, consequently, that document (6) does not disclose all method features of Claim 1.

Having examined the other cited prior art documents, the Board has come to the conclusion that the method according to the present claims is not disclosed in any of them. The Board therefore concludes that the claimed method is novel over the cited prior art.

4. *Inventive step*

- 4.1 The Board considers document (2), which is discussed in column 1, lines 41 to 53, of the patent in suit, to represent the closest state of the art, which was not contested anymore by the Parties.

Document (2) teaches that crude glycidyl (meth)acrylate obtained by reacting an alkali metal salt of (meth)acrylic acid with epichlorohydrin in the presence of a quaternary ammonium salt, filtering the resultant reaction mixture and removing excess epichlorohydrin by distillation, has an epichlorohydrin content in the range of 0.1 to 1.0% by weight and that the quaternary ammonium salt, which remains in minute amounts in the

glycidyl (meth)acrylate, accelerates the conversion of the by-product 1,3-dichloro-2-propanol into epichlorohydrin (column 2, lines 45 to 68).

Additionally, document (2) discloses a method of purifying the above mentioned crude glycidyl (meth)acrylate to an extent to lower the epichlorohydrin content to a content of not more than 0.01% by weight (100 ppm) by treating the glycidyl (meth)acrylate with a heteropoly acid or an alkali metal salt thereof and subsequently isolating the purified glycidyl (meth)acrylate by distillation. The heteropoly acid or the alkali salt thereof is said to readily react with the quaternary ammonium salt and to form a stable complex salt therewith, thus inactivating the quaternary ammonium salt and avoiding the formation of epichlorohydrin from 1,3-dichloro-2-propanol without interfering with the effect of the polymerisation inhibitor (column 3, lines 42 to 64, and column 4, lines 35 to 41).

4.2 Starting from the disclosure of document (2) the problem underlying the invention must be seen in providing a further method of purifying crude glycidyl (meth)acrylate by effectively removing impurities of epichlorohydrin **and other chlorine compounds** without decreasing the yield of the high-purity glycidyl (meth)acrylate (see the patent in suit, column 2, lines 37 to 45).

4.3 The patent in suit claims to solve this problem by the process defined in Claim 1, more particularly, by subjecting a crude glycidyl (meth)acrylate to a stripping treatment with a mixed gas containing oxygen

gas in the presence of a quaternary ammonium salt and then distilling the thus treated product.

- 4.4 From the data presented in examples 1 to 4 of the patent in suit it follows that the chlorine concentration of crude glycidyl (meth)acrylate may be reduced with the claimed purification method from 7800 ppm to 90 ppm (example 1) or to 180 ppm (example 3) and from 4600 ppm to 420 ppm (example 2) or to 130 ppm (example 4).

The Board therefore accepts that it has been made credible that the problem underlying the invention, as defined above, is effectively solved by the claimed method, which has not been contested by the Appellant.

- 4.5 It remains to be decided, whether, in the light of the teachings of the cited documents, a skilled person seeking to solve the above mentioned problem, would have arrived at the claimed method in an obvious way.
- 4.6 The Appellant was of the opinion that a skilled person would have done so, because it was known from document (2) that by heating a mixture of glycidyl methacrylate, a polymerisation inhibitor and 1,3-dichloro-2-propanol, in the presence of a quaternary ammonium salt the 1,3-dichloro-2-propanol was partially converted into epichlorohydrin (column 3, lines 1 to 8, and Table 1) and that purified glycidyl (meth)acrylate may be isolated by distillation (column 3, lines 53 to 55), which is one form of stripping. Moreover, since it was known from document (3) that residual epichlorohydrin in glycidyl products may be reduced to very low levels by stripping with an inert gas (page 1,

lines 3 to 5 and 10 and 11), and oxygen was known to be a polymerisation inhibitor, he was of the opinion that the claimed method was obviously derivable from document (2) and (3).

4.7 However, the Board must point out that in view of the problem to be solved as set out above the relevant question in the present case is not whether all the features of the conversion of chlorine compounds into epichlorohydrin, the stripping treatment and the distillation of glycidyl (meth)acrylate were known, but whether there was any suggestion to replace the step of inactivating the quaternary ammonium salt known from document (2) by a stripping treatment with a mixed gas containing oxygen gas in the presence of a quaternary ammonium salt.

4.8 It is true that document (2) discloses that 1,3-dichloro-2-propanol may partially be converted to epichlorohydrin in the presence of a quaternary ammonium salt. However, it is the essence of document (2) that such conversion can be avoided by inactivating the quaternary ammonium salt and that by avoiding such reconversion into epichlorohydrin purified glycidyl (meth)acrylate may be obtained by a subsequent distillation, which does not contain more than 0.01% by weight of epichlorohydrin. Nothing therein suggests that such reaction could be suitable for converting the chlorine compounds cited in Claim 1 into epichlorohydrin and, subsequently, removing the epichlorohydrin.

Moreover, document (2), which is only concerned with a method of purifying glycidyl (meth)acrylate such that

less than 0.01% by weight of epichlorohydrin is obtained, is completely silent about the amount of **chlorine compounds, in general**, in the purified product and, consequently, does not provide any information whatever about possible methods of reducing the amount of chlorine compounds, in general.

- 4.9 Since the conversion of chlorine compounds, in general, into epichlorohydrin was not suggested in document (2), a skilled person did not have any incentive to combine the teaching of documents (2) with any teaching of removing epichlorohydrin, such as, by a stripping treatment, as described in document (3).

Therefore, the claimed purification method was neither suggested by the teaching of document (2) nor by the combined teaching of documents (2) and (3).

- 4.10 The Appellant also argued that the claimed purification method was obvious over the teaching of document (5), since the above mentioned conversion to epichlorohydrin is described therein, for example, on page 6, lines 20 to 25.

However, document (5) is concerned with a method of producing carboxylic acid glycidyl esters and not with a method of purifying such esters. The teaching on page 6, lines 20 to 25, concerns a method of recovering halohydrins, used as starting material, from halohydrins formed as a by-product in the method of preparing carboxylic acid glycidyl esters, and not the conversion of residual chlorine compounds into epichlorohydrin. Therefore, a skilled person looking for a method of removing residual chlorine compounds

- from crude glycidyl (meth)acrylate would not have any incentive to consider document (5).
- 4.11 As concluded in point 4.9 above, the presently claimed purification method was not suggested in document (2) or by a combination of documents (2) and (3), a skilled person would not have any incentive to combine the teaching of any of documents (2), (3) or (5). Thus, Claim 1 is not obvious in the light of the teachings of the cited prior art.
- 4.12 Claims 2 to 5, which represent preferred embodiments of Claim 1, derive their patentability from the same inventive concept.
5. Since Claims 1 to 5 and the description as submitted comply with the requirements of the EPC, the patent may be maintained on the basis of the documents specified in the Appellant's request.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The matter is remitted to the first instance with the order to maintain the patent on the basis of the claims and amended description submitted at the oral proceedings on 22 June 1999.

The Registrar:

The Chairman:

E. Görgmaier

A. Nuss