

Publication in the Official Journal ~~Yes~~ / No

File Number: T 63/90 - 3.3.1
Application No.: 84 200 660.3
Publication No.: 0 125 726
Title of invention: Improved process for preparing alkyl isocyanates

Classification: C07C 119/042

D E C I S I O N
of 30 September 1991

Proprietor of the patent: ENICHEM SINTESI S.p.A.
Opponent: Bayer AG, Leverkusen Konzernverwaltung RP
Patente Konzern

Headword: Alkyl isocyanates/ENICHEM SINTESI
EPC Articles 56 and 83
Keyword: "Sufficiency (yes)" - "Inventive step (yes)"

Headnote



Case Number : T 63/90 - 3.3.1

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 30 September 1991

Appellant :
(Opponent)

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(Proprietor of the patent)

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Decision under appeal :

Interlocutory decision of the Opposition Division
of the European Patent Office delivered orally on
14 September 1989, with written reasons posted on
12 December 1989, concerning maintenance of
European patent No. 0 125 726 in amended form.

Composition of the Board :

Chairman : K.J.A. Jahn
Members : J.M. Jonk
J.-C. Saisset

Summary of Facts and Submissions

I. The grant of European patent No. 0 125 726 in respect of European patent application No. 84 200 660.3 was announced on 16 July 1986 (cf. Bulletin 86/29).

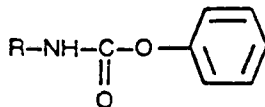
II. A Notice of Opposition was filed on 25 November 1986 requesting revocation of the patent on the grounds of lack of novelty and inventive step, and insufficient disclosure. The opposition was supported by:

- (1) US-A-3 366 662,
- (2) DE-A-2 756 928, and
- (7) US-A-4 123 450

which are relevant to the present decision.

III. By a decision delivered orally on 14 September 1989, with written reasons posted on 12 December 1989, the Opposition Division maintained the patent in amended form on the basis of Claim 1 submitted on 14 September 1989 and Claims 2 and 3 of the patent in suit, Claim 1 reading as follows:

"Process for the preparation of alkyl isocyanates having the general formula R-N=C=O, wherein R is an alkyl radical having from 1 to 4 carbon atoms, by thermal decomposition of phenyl N-alkylurethans having the general formula:



wherein R is as hereinabove defined, characterized in that said thermal decomposition is carried out, without using any substance capable of catalyzing the decomposition reaction, by heating a mixture of phenol and a phenyl N-

alkylurethan, in a molar ratio of the former to the latter of from 1:1 to 3:1, to a temperature of from 180°C to 210°C and under the atmospherical pressure or slightly below same, evaporating off out of the reaction environment both the phenol and the alkyl isocyanate concerned as they are being formed, their vapours being thereafter separately condensed and recovered, and further characterized in that a molar ratio of phenol to the phenyl N-alkylurethan concerned never below 1:1 is maintained in the reaction environment virtually throughout the whole reaction decomposition time."

The Opposition Division held that the subject-matter of the amended patent was novel. The process according to Claim 1 differed from that of document (1) in that it concerns the decomposition of phenyl N-alkylurethans wherein the alkyl group has 1 to 4 carbon atoms and that the molar ratio of the phenol to the phenyl N-alkylurethan is between 1:1 to 1:3 for the entire duration of the thermal decomposition.

The claimed process also involved an inventive step, because it was not obvious to the skilled person in the light of the prior art disclosed in documents (1), (2) and (7) that the decomposition of the particular urethan by heating a mixture of phenol and the urethan in a molar ratio of from 1:1 to 3:1 and preventing this ratio from dropping below 1:1 would provide the desired isocyanates in surprisingly high yields and selectivity.

The comparative examples filed by the Opponent on 25 November 1986 purporting to demonstrate that the improvements were not obtained did not fulfil the condition that the molar ratio phenol to urethan must never drop below 1:1 and actually supported the presence of an inventive step.

The objection that Claim 1 would not meet Article 100(b) EPC was rejected because the Examples in the patent showed that the reaction flask was surmounted by a distillation device, so that the skilled person would recognise that the specific molar ratio could be adjusted by regulating the reflux ratio of the distillation device.

- IV. A Notice of Appeal was filed against this decision on 23 January 1990 and the appeal fee was paid on the same date.

A Statement of Grounds of Appeal was submitted on 8 February 1990.

- V. The Appellant argued that, in relation to the requirements of Article 100(b) EPC, a comparison of Example 4 of the filed comparative examples with the substantially identical Example 2 of the patent in suit showed that the example of the patent did not disclose all the essential features. Also in the drawing of the patent in suit - as was admitted by the Respondent - no means were indicated to regulate the separation of the phenol. In this connection the Appellant pointed out that the claimed ratio could not be responsible for the alleged improvement of the yield because this would be in contradiction to the law of mass action. The filed comparative Examples 3 and 4 showed that without the addition of phenol the yield was higher.

It was also argued by the Appellant that the claimed process lacked an inventive step in view of the combined disclosures of documents (7) and (1). Document (7) disclosed the decomposition of phenyl N-methylurethan at temperatures of 190 to 208°C and the separate condensation of the resulting vapours, and the use of a starting mixture containing equimolar amounts of phenyl N-alkylurethan and phenol was already known from document

(1). An improvement in the yields of isocyanate in comparison with a process, wherein phenol was recycled in accordance with Example 3 of document (7) or comparative Example 1 of the disputed patent, would not be surprising in the light of the law of mass action.

Referring to his letter filed on 25 November 1986 the Appellant also maintained his objection in relation to the inventive step based on the disclosure of document (2), because this disclosure also comprised the use of a starting mixture containing urethan and phenol in a ratio of 1:1.

VI. The Respondent refuted the objection of insufficiency. The conditions to be adopted were clearly stated and the steps to be taken to ensure such conditions were within the common general knowledge of the skilled person. The wording of Example 2 actually gave a strong hint that a partial reflux of phenol was necessary to ensure that the molar ratio of phenol to the phenyl N-alkylurethan was never below 1:1. Also the figure in the patent showing a vertical condenser implied that reflux of phenol was envisaged.

Moreover, the claimed process was not obvious to the skilled person, because none of the cited documents gave any hint with regard to the basic requirement of the claimed process, i.e. the maintenance of the claimed ratio. Also, there was no incentive to combine the teachings of these documents.

Furthermore, according to the claimed process much higher yields were obtained. Apart from comparative Example 2 filed by the Opponent, all his comparative examples were mere fictive examples. On the other hand, comparative

Example 1 of the patent truly represented the state of the art, namely Example 3 of document (7). Comparative Example 2 of the Opponent also corresponded essentially to Example 3 of document (7).

VII. During the oral proceedings held on 30 September 1991 the Respondent filed new Claims 1 to 3 in order to meet the requirements of Article 123(2) EPC and the Board's objection that the amended claims accepted by the Opposition Division did not meet the well established jurisprudence of the Boards that all amendments submitted in the course of opposition must be directed to meeting the grounds of opposition and nothing else. Claim 1 differed from that as maintained by the Opposition Division in that the ratio "from 1:1 to 3:1" was amended to "about 1:1", the expression "and further characterised in that" was deleted, the condition "never below 1:1 is" was replaced by "from 1:1 up to 3:1 being", and the term "environment" was amended to "mixture".

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

The Respondent requested that the appeal be dismissed and that the patent be maintained on the basis of the amended set of Claims 1 to 3, an amended description and the drawing submitted during the oral proceedings.

IX. At the conclusion of the oral proceedings, the Board's decision to maintain the patent in the amended form was announced.

Reasons for the Decision

1. The appeal complies with Articles 106 to 108 and Rule 64 EPC and is, therefore, admissible.

2. There are no objections to the claims under Article 123 EPC. The amendment in Claim 1 of the molar ratio of the starting mixture, namely "from 1:1 to 3:1" to "about 1:1", is based on Claim 1 of the application as originally filed and column 2, lines 41 to 43 and all examples, save the comparative example, of the patent in suit. The amendment of the molar ratio maintained during the reaction to "from 1:1 up to 3:1" finds a basis in page 5, lines 11 to 25, particularly lines 22 and 23, and Claim 3 of the original application and column 3, lines 8 to 23, particularly line 19, of the opposed patent. The replacement of the term "environment" by the more precise term "mixture" is supported by Claim 1 of the original application and column 2, line 47 of the patent specification. Having regard to the restriction of ratios of the reactants both at the start and during the reaction the amended Claim 1 does not extend the protection conferred.

3. After examination of the cited documents, the Board has reached the conclusion that the claimed subject-matter is novel. Since novelty was finally not in dispute, it is not necessary to give detailed reasons for this finding.

4. In the Board's judgment, the Appellant's objection to the sufficiency of disclosure fails since the requirement of Claim 1 of the disputed patent that the molar ratio of phenol to the phenyl N-alkylurethan during the decomposition reaction is maintained at a value of from 1:1 up to 3:1 can be put into practice by a skilled person on the basis of his common general knowledge. Thus, the skilled person knows that this ratio can be monitored and controlled by adjusting the reflux ratio of the phenol

using a distillation device in accordance with the drawing, which, of course, must be slightly modified in a well-known way so that the reflux of phenol is possible.

5. The remaining issue to be dealt with is whether the subject-matter of the claims involves an inventive step.

5.1 After consideration of the cited prior art documents the Board is of the opinion that citation (7) represents the closest state of the art.

Document (7) relates to a process for the preparation of lower alkyl isocyanates by reacting a suitable phenol, phosgene and an alkylamine to produce the corresponding phenyl N-alkylurethan and pyrolyzing the obtained urethan (cf. column 1, lines 5 to 10). Example 3 discloses the preparation of methyl isocyanate by thermal decomposition of phenyl N-methylurethan at temperatures of 190 to 208°C in the absence of a catalyst; the co-produced phenol being condensed and returned to the flask.

However, it was considered by the Respondent that the yields of isocyanate obtained by this process was not satisfactory (cf. under section VI. above, third paragraph).

5.2 Therefore, in the light of this closest prior art, the technical problem underlying the patent in suit is to be seen in providing a process for preparing lower alkyl isocyanates in higher yields and selectivities (cf. also column 1, line 60 to column 2, line 21 of the opposed patent).

According to Claim 1, this technical problem is essentially solved by heating a starting mixture of phenol and a phenyl N-alkylurethan in a molar ratio of about 1:1, evaporating off phenol and alkyl isocyanate as they are

formed and maintaining a molar ratio of phenol to the phenyl N-alkylurethan from 1:1 to 3:1 in the reaction mixture virtually throughout the whole reaction time.

The Appellant disputed that the achievement of higher yields of alkyl isocyanate could be due to the claimed presence of phenol in view of the law of mass action and the comparative Examples 1 to 4 filed on 25 November 1986.

However, the Appellant's argumentation that higher yields of alkyl isocyanate in a process starting from a mixture of phenol and urethan would be in contradiction to the law of mass action fails, because, at the temperatures within the claimed range of 180 to 210°C, both decomposition products, namely the lower alkyl isocyanate and phenol, are in the vapour phase and removed from the reaction system as they are formed. In the case of the low boiling isocyanates - methyl isocyanate has a boiling point of 38°C - this removal will even proceed in the form of flash distillation. It is commonly known that the law of mass action in its strict sense can only be applied if ideal conditions, including equilibrium conditions in a one-phase system, prevail. It can hardly be said that the above-mentioned conditions correspond to ideal ones.

The Appellant's argumentation that the comparative Examples 1 to 4 filed on 25 November 1986 confirm that the law of mass action is applicable in the present case and that the improvement in the yields cannot be the result of the claimed presence of phenol also cannot be accepted. It is true that the Appellant's comparative Examples 1, 2, 3 and 4 with yields of 32%, 65.5%, 84.1% and 81.2% respectively indicate that, by using a starting mixture of urethan and phenol in a molar ratio of 1:1 (comparative Examples 1 and 4) instead of urethan alone (comparative Examples 2 and 3), lower yields of the isocyanate product

are obtained. This may be the result of a higher phenol concentration during the decomposition reaction. However, in the Board's judgment, this finding does not provide any evidence for the above allegation that the improved yields of isocyanate obtained in the examples of the disputed patent cannot be the result of the claimed conditions for the phenol concentration, because none of these comparative examples meet both mandatory requirements of Claim 1 of the disputed patent, i.e. firstly, a phenol to urethan starting ratio of 1:1 and, secondly, the control of the molar ratio of these two components within the indicated range virtually throughout the whole reaction time. Thus, comparative Example 1, in which the initial reaction mixture contains phenyl N-methylurethan and phenol in a molar ratio of 1:1 with total reflux of the phenol, does not meet the second mandatory requirement because the upper limit of this molar ratio will already be reached after decomposition of 50% of the urethan. Furthermore, comparative Example 2, which essentially differs from comparative Example 1 in that the initial reaction mixture only contains the urethan, does not meet either of the mandatory requirements. Neither of these requirements are fulfilled by comparative Example 3, in which the urethan is heated and phenol and methyl isocyanate are both removed from the reaction vessel. Finally, comparative Example 4, in which at the beginning of the reaction phenol and the urethan are present in a molar ratio of 1:1 and phenol and methyl isocyanate are both distilled off, does not comply with the second requirement.

Having regard to the disclosure in document (7), in which it is indicated that during the pyrolysis of phenyl N-methylurethan the evolution of methyl isocyanate ceases if the pot composition reaches a molar ratio of about 1 part of the urethan compound to about 3 parts phenol (cf. column 3, lines 36 to 41), the Board had some misgivings

that improved yields would be obtained in the upper end of claimed range. However, since the Appellant, who has the burden of proof (cf. for instance T 219/83, OJ EPO 1986, 211, Section 12, 4th paragraph), did not provide any convincing evidence in this respect, the Board gives the benefit of doubt in this respect to the Respondent. Moreover, in view of the results of the examples in the opposed patent, in which comparative Example 1, which is essentially in accordance with Example 3 of document (7), results in a conversion of the urethan of 74%, with a selectivity of the isocyanate product of 93% relative to the converted urethan, whereas Examples 2 to 4 result in conversions and selectivities of about 98% and 99% respectively, the Board is satisfied that the above technical problem is solved.

5.3 It remains to be decided whether the claimed process involves an inventive step.

5.4 As already indicated in section 5.1 above, paragraph 2, document (7) discloses a process for the preparation of lower alkyl isocyanates by pyrolysis of the corresponding phenyl N-alkylurethan, in which the alkyl isocyanate is selectively condensed by being taken off overhead by means of a primary condenser maintained at a temperature above the condensation temperature of the alkyl isocyanate but below the condensation temperature of the co-produced hydroxybenzene and the hydroxybenzene is condensed and either returned to the pyrolysis vessel or separately collected (cf. column 3, lines 22 to 31). The process wherein the hydroxybenzene co-product is totally refluxed - as disclosed in Example 3 - is apparently preferred. Since it is stated in this document that the evolution of methyl isocyanate ceases if the pot composition reaches a molar ratio of about 1 part phenyl N-methylurethan to about 3 parts phenol (cf. column 3, lines 36 to 41 and Example 3, lines 42 to 44), it could be deduced that an

upper limit of the ratio phenol to urethan of about 3:1 should not be exceeded. However, in the Board's judgment, this document does not hold out any prospect that the use of a molar ratio of phenol to the urethan at the beginning of the decomposition reaction of about 1:1 and the maintenance of this ratio between 1:1 and 3:1 throughout the whole reaction time would provide better yields of alkyl isocyanates and, hence, solve the present problem. In the Board's view, it could even be inferred from this document that a phenol to urethan ratio of 1:1 at the beginning of the reaction would be disadvantageous, because the ratio at which the decomposition ceases would be reached earlier. Moreover, it is clearly indicated that, after the pyrolysis has ceased the phenol is stripped off completely before the next cycle of the process is started (cf. column 3, lines 41 to 52 and Example 3, lines 42 to 49).

- 5.5 Document (1) discloses the preparation of isocyanates by reacting diphenyl carbonate with an amine (cf. Claim 1). Suitable amines are aromatic amines, aliphatic monoamines and aliphatic diamines (cf. Claim 1 and column 2, lines 24 to 36). The aliphatic monoamines comprise, inter alia, lower alkylamines such as ethyl-, n-propyl, isopropyl- and tert. butylamine (cf. column 2, lines 27 to 29). The reaction may be carried out by adding the amine to molten diphenyl carbonate at a temperature of about 50° to 350°C, preferably about 60° to 250°C (cf. column 2, lines 13 to 17). According to the examples, after the addition of the amine, the various products are distilled off at increasing temperatures, whereby the isocyanates (none of them belonging to the relatively difficultly available lower alkyl isocyanates) are obtained in amounts of 55.5%, 68% and 59.3% respectively. Although the process is represented by an equation which shows that two consecutive reactions take place, so that after the first reaction a urethan and phenol are obtained in a molar

ratio of 1:1, there is, in the Board's judgment, no indication in this document that the yields of isocyanates might be improved by maintaining the claimed phenol to urethan ratio over the whole reaction time. In this context it is observed that the distillation of the various products at increasing temperatures disclosed in the examples of this document exclude the maintenance of the critical phenol to urethan ratio in the reaction medium. Also the extraction recovery method, referred to in column 2, lines 11 and 12, means in the case of lower boiling isocyanates, that the higher boiling phenol necessarily has not been distilled off, so that the maintenance of the claimed phenol to urethan ratio is not possible.

Moreover, the skilled person would, in the Board's judgment, have had no reason to combine the teaching of this document with that of document (7), because - as set out in section 5.4 above - this more recent prior art clearly suggests that a phenol to urethan ratio of 1:1 at the beginning of the pyrolysis reaction would be disadvantageous.

- 5.6 Document (2) relates to the preparation of a lower alkyl isocyanate by pyrolysis of a phenyl N-alkylurethan in the presence of an inert solvent at temperatures of 190 to 250°C, whereby all the vapours produced are passed into a distillation column from which the phenol co-product and at least the major amount of the solvent are recovered as a common stream or as several side streams and the alkyl isocyanate, optionally with a portion of the solvent, is obtained at the head of the column (cf. Claim 1). According to this document the dilution of the mixture of reactive lower alkyl isocyanate and co-produced phenol by the solvent vapour prevents their recombination (cf. page 7, first complete paragraph; page 12, from line 5; and and page 19, first complete paragraph). In the Board's

judgment, this teaching leads away from the present process, which provides additional phenol at the beginning of the reaction in an amount about equal to that of the starting urethan and also requires that a phenol to urethan ratio of 1:1 to 3:1 be maintained throughout the whole conversion.

- 5.7 Consequently, in the Board's judgment, the proposed solution to the technical problem underlying the patent in suit is inventive. Thus Claim 1 is allowable.


Dependent Claims 2 and 3, which relate to preferred embodiments of the process according to Claim 1, are likewise allowable.

Order

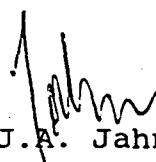
For these reasons, it is decided that:

1. The contested decision is set aside.
2. The case is remitted to the first instance with the order to maintain the patent on the basis of the complete specification as submitted at the oral proceedings.

The Registrar:


E. Gorgmaier

The Chairman:


K.J.A. Jahn