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Datasheet for the decision of 5 May 2017

Case Number: T 1864/13 - 3.3.10

Application Number: 06807765.0

Publication Number: 1960346

IPC: C07C209/78

Language of the proceedings: EN

Title of invention:
PROCESS FOR PREPARING DIAMINODIPHENYLMETHANES

Patent Proprietor:
HUNTSMAN INTERNATIONAL LLC

Opponent:
BASF SE

Headword:
PROCESS FOR PREPARING DIAMINODIPHENYLMETHANES / HUNTSMAN

Relevant legal provisions:
EPC Art. 56, 84

Keyword:
Inventive step - main request and first auxiliary request (no) - second auxiliary request (yes)
Claims - clarity - second auxiliary request (yes)

Decisions cited:

Catchword:

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Case Number: T 1864/13 - 3.3.10

D E C I S I O N
of Technical Board of Appeal 3.3.10
of 5 May 2017

Appellant: (Opponent)	BASF SE 67056 Ludwigshafen (DE)
Representative:	Schuck, Alexander Isenbruck Bösl Hörschler LLP Eastsite One Seckenheimer Landstrasse 4 68163 Mannheim (DE)
Respondent: (Patent Proprietor)	HUNTSMAN INTERNATIONAL LLC 500 Huntsman Way Salt Lake City, Utah 84108 (US)
Representative:	Swinnen, Anne-Marie Huntsman (Europe) BVBA Intellectual Property Department Everslaan 45 3078 Everberg (BE)
Decision under appeal:	**Decision of the Opposition Division of the European Patent Office posted on 21 June 2013 rejecting the opposition filed against European patent No. 1960346 pursuant to Article 101(2) EPC.**

Composition of the Board:

Chairman	P. Gryczka
Members:	J.-C. Schmid
	T. Bokor

Summary of Facts and Submissions

I. The Appellant (Opponent) lodged an appeal against the decision of the Opposition Division rejecting the opposition against European patent No. 1 960 346 which was granted on the basis of four claims, claim 1 of which reading as follows:

"1. Process for preparing diamino diphenyl methane and poly-(diamino diphenyl methane) comprising the step of reacting aniline with formaldehyde in the presence of hydrogen chloride - added in the gaseous form characterised in that hydrogen chloride gas is absorbed in aniline, the aniline containing 0.1 to 7 wt%, preferably 2 to 5 wt% of a protic chemical, said protic chemical being water or methanol."

II. Notice of opposition had been filed by the Appellant requesting revocation of the patent in suit in its entirety on the grounds of lack of inventive step (Article 100(a) EPC). *Inter alia* the following documents were submitted in the opposition/appeal proceedings:

(1) GB 1 517 585 and
(2) GB 1 167 950.

In the decision under appeal, the Opposition Division held that document (2) represented the closest prior art. This document disclosed the reaction of aniline hydrochloride salt, aniline and formaldehyde wherein the presence of added water was carefully avoided. The technical problem to be solved was the provision of an alternative process for preparing diamino diphenyl methane and poly-(diamino diphenyl methane). The

proposed solution was characterized in that aniline contained 0.1 to 7 wt% of water or methanol, and that aniline was contacted with gaseous HCl. The skilled person would not have arrived at the claimed solution because document (2) taught against the presence of added water in the reaction mixture. The subject-matter of the claims of the patent as granted involved therefore an inventive step.

III. At the oral proceedings before the Board held on 5 May 2017, the Respondent (Proprietor of the patent) defended the maintenance of the patent in suit as granted (main request) and on the basis of a first auxiliary request filed with a letter dated 12 March 2014 and a second auxiliary request filed during the oral proceedings before the Board.

Claim 1 of the first auxiliary request differs from claim 1 of the main request in that the upper limit of the amount of hydrogen chloride added to aniline follows the relationship:

$$wt\%\ HCl = 1.43 \times [water] + 0.07T - 0.55$$

wherein [water] is the concentration of water in the aniline, expressed as weight per cent and T is the temperature of the aniline/water mixture, expressed in degrees Celsius.

Claim 1 of the second auxiliary request reads as follows:

"1. Process for preparing diamino diphenyl methane and poly-(diamino diphenyl methane) 3 comprising the following steps:

(i) preparation of aniline/water mixture with a water content of 0.1 to 7 wt%, preferably 2 to 5 wt%;

(ii) absorption of hydrogen chloride gas, optionally from a phosgenation process, into said aniline/water mixtures, optionally with cooling to a desired temperature wherein the upper limit of the amount of hydrogen chloride added to aniline/water mixture follows the relationship:

$$wt\% \ HCl = 1.43 \times [water] + 0.07T - 0.55$$

wherein [water] is the concentration of water in the aniline, expressed as weight per cent and T is the temperature of the aniline/water mixture, expressed in degrees Celsius; and is in the range of 40 to 75 degrees Celsius

(iii) addition, optionally with simultaneous mixing, of formaldehyde into the aniline/HCl/water mixture in one or more stages, optionally with cooling, to yield an intermediate mixture;

(iv) heating said intermediate to predetermined levels from 50 to 15O °C, preferably from 60 to 14O °C to produce the desired mixture of primary amine isomers and homologues known as DADPM;

(v) working up said mixture by neutralisation of the acid typically with sodium hydroxide solution, separation and washing of the organic and brine phases, followed by removal and recycling of excess unreacted aniline."

IV. According to the Appellant, both documents (1) and (2) could be seen as the closest prior art. These documents

disclosed a process for preparing diamino diphenyl methane by reacting aniline with formaldehyde in the presence of hydrogen chloride.

Document (2) was closer to the invention than document (1) since it addressed the problem of mobility caused by the addition of the aniline salt into aniline. Although the addition of hydrogen chloride occurred in the form of the aniline salt in example 2, the *in situ* formation of aniline salt by introducing gaseous hydrogen chloride into aniline was an obvious alternative to the step of adding already formed aniline salt. The difference between the process of claim 1 of the main and first auxiliary requests and that disclosed in example 5 of document (1) resided only in the presence of small amount of water in aniline before the condensation reaction started, i.e. water was already present during the addition of hydrogen chloride into aniline. The presence of small amounts of water in aniline had no unexpected effect, was purely arbitrary and therefore could not impart an inventive step to the claimed subject-matter. The process of claim 1 of the second auxiliary request further required to first mix a small amount of water with aniline. However, modifying the order in which the reactants are mixed in the process of document (1) was an obvious possibility for the skilled person. Accordingly, the subject-matter of claim 1 of the main, first and second auxiliary request did not involve an inventive step. In addition the subject-matter of claim 1 lacked clarity, since it was not clear whether or not the temperature range was a limiting feature for the process.

V. According to the Respondent, eihter document (1) or document (2) could be seen as the closest prior art.

Document (2) did not recognize the problem of solubility of the aniline salt in aniline. The problem addressed in example 2 of document (2) was that of poor mobility of the reaction mixture which was due to the presence of solid particles of paraformaldehyde and aniline salt. The solution proposed in document (2) to improve mobility was the addition of more aniline in order to dilute the suspension of particles. In contrast to document (2) the invention aimed to increase the solubility of the aniline salt formed in situ, thereby increasing the catalytic efficiency. Furthermore document (2) did not disclose the formation of the aniline salt with gaseous hydrogen chloride, but with aqueous hydrochloric acid. Document (1) disclosed a process for preparing diamino diphenyl methane wherein aniline, formaldehyde and hydrogen chloride were introduced into a reactor via separate lines. The claimed process differed from the process of document (1) in two respects, the first being the order in which the reactants are introduced, the second being that aniline contained small amounts of water. The embodiment disclosed on page 5, lines 54 to 56 of the patent-in-suit did not fall within the scope of claim 1 as granted and was inserted by error in the description of the patent-in-suit. The process disclosed in document (1) needed a separate conduit for the introduction of the gaseous hydrogen chloride into the reactor. The technical problem to be solved by the invention was the provision of a further process for preparing diamino diphenyl methane with simplified equipment. The solution consisted in preparing an aniline/water mixture with a water content of 0.1 to 7 wt% and then absorbing hydrogen chloride gas into said aniline/water mixture. This solution was not suggested in the prior art. Therefore the subject-matter of claim

1 of the main, first and second auxiliary requests implied an inventive step.

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

The Respondent requested that the decision under appeal be dismissed, i.e. that the patent be maintained as granted (main request), or alternatively that the decision under appeal be set aside and the patent be maintained in an amended form on the basis of the first request filed with the response to the grounds of appeal dated 12 March 2014, or on the basis of the second auxiliary request filed during the oral proceedings before the Board.

VII. At the end of the oral proceedings the decision of the Board was announced.

Reasons for the Decision

1. The appeal is admissible.

Main request

2. Inventive step

2.1 *Closest prior art*

Document (1) discloses a process for preparing diamino diphenyl methane wherein aniline, formaldehyde and hydrogen chloride are introduced into a reactor via separate lines (see example 5 on page 5, left-hand column).

According to the Respondent, the process of claim 1 of the main request differed from that of example 5 of document (1) due to the fact that it required a preliminary step wherein gaseous hydrogen chloride was absorbed in aniline. The process of example 5 of document (1) lacked this preliminary step.

However, claim 1 of the main request only requires that hydrogen chloride is absorbed in aniline without specifying at which point of time. It does therefore not require a preliminary step wherein hydrogen chloride is absorbed in aniline. On page 5, lines 54 to 56 of the patent-in-suit, it is indicated that the gaseous hydrogen chloride may also be added to the mixture formed by reaction of aniline and formaldehyde in neutral or basic conditions processes where the initial compounds formed include methylene di-aniline.

The Respondent argued that this embodiment was mentioned by error in the patent-in-suit, since it was obvious that it did not fall within the scope of claim 1. As aniline was already reacted with formaldehyde, hydrogen chloride could not be absorbed into aniline, as required by claim 1 of the main request.

However, the passage on page 5, lines 54 to 56 of the patent-in-suit, discloses an embodiment where the initial compounds formed **include** methylene di-aniline. That does not mean that the whole aniline has disappeared by reaction with formaldehyde. Thus, this embodiment falls within the scope of claim 1.

In the process disclosed in example 5 of document (1), aniline, formaldehyde and gaseous hydrogen chloride are simultaneously introduced into the reactor via separate lines. Therefore, part of the aniline will react with

formaldehyde to form methylene di-aniline while another part of the aniline will absorb the gaseous hydrogen chloride. The Board comes therefore to the conclusion that claim 1 does not require that gaseous hydrogen chloride must be absorbed in aniline before the addition of formaldehyde and that the requirement of claim 1 of the patent-in-suit that hydrogen gas is absorbed in aniline is satisfied by the continuous process of example 5 of document (1) where aniline, formaldehyde and hydrogen chloride are introduced into the reactor via separate lines.

Accordingly, the sole difference between the process of claim 1 of the main request and the process disclosed in example 5 of document (1) resides in that the aniline in which hydrogen chloride is absorbed contains from 0,1 to 7% by weight of water or methanol.

According to the Appellant, document (2) could possibly be closer to the invention, since it addressed the same technical problem as the patent-in-suit, namely the problem of mobility. However, the problem of mobility, which is only addressed in example 2 of document (2), is different from that of insufficient solubility of aniline hydrochloride salt formed in-situ addressed in the patent-in-suit, which leads to an insufficient catalytic activity. Furthermore, the process disclosed in example 2 of document (2) is not carried out with gaseous hydrogen chloride. The use of gaseous hydrogen chloride in the process for preparing diamino diphenyl methane is only mentioned on page 2, left-hand column, lines 55 to 57 of document (2) as an alternative of using hydrochloric acid, which is preferably added as a salt of a portion of the aromatic acid (also see claim 5).

Accordingly, the Board holds that document (1) which discloses a process of preparing diamino diphenyl methane using gaseous hydrogen chloride represents the prior art closest to the invention, which finding was not contested by the parties during the oral proceedings.

2.2 Technical problem

Starting from document (1) as the closest prior art, the Respondent defined the technical problem underlying the patent-in-suit as the provision of a further process for preparing diamino diphenyl methane with simplified equipment.

2.3 *Success*

According to the Respondent, the claimed process required only two entries to the reactor, while the process of document (1) required a separate entry for gaseous hydrogen chloride.

The problem relating to the simplification of the equipment is, however, not solved by the process of claim 1 of the main request, since it does not exclude the embodiment wherein gaseous hydrogen chloride is introduced via a separate line, as in the process disclosed in example 5 of document (1). The technical problem underlying the patent-in-suit must therefore be reformulated as the provision of a further process for preparing diamino diphenyl methane.

2.4 *Proposed solution*

The proposed solution is the process of claim 1 of the main request characterized in that the aniline in which

hydrogen chloride is absorbed contains from 0.1 to 7% by weight of water or methanol.

2.5 *Obviousness*

The process disclosed in document (1) is carried out in an aqueous medium, the formaldehyde being added as an aqueous solution. The present invention also envisages adding more water into the reaction medium, since formaldehyde is preferably added as an aqueous solution (see examples 1 and 3 on page 6). The amount of 0.1 to 7% by weight of water in aniline, which is required by claim 1, is not linked to any unexpected technical effect, and is therefore arbitrary. The skilled person would therefore have contemplated the use of aniline comprising water in the range of 0.1 to 7% by weight as an obvious measure. Accordingly, the subject-matter of claim 1 of the main request lacks an inventive step.

First auxiliary request

3. Claim 1 of the first auxiliary request defines an upper limit of the concentration of hydrogen chloride in aniline. The Respondent did not provide any arguments in support of inventive step linked to this additional feature. In addition, it has not been shown that this feature distinguishes the claimed process from that disclosed in document (1). Under these circumstances the Board concludes that this upper concentration limit of hydrogen chloride does not add any inventive contribution to the claimed process.

Second auxiliary request

4. Amendments, added subject-matter and clarity

Claim 1 of the second auxiliary request is based on claim 3 of the patent as granted (claim 4 of the application as filed), wherein the upper limit of the amount of hydrogen chloride is specified according to the disclosure of the page 7, lines 20 to 29 of the application as filed. This amendment restricts the protection conferred by the patent as granted. Claim 2 is identical to claim 4 of the patent as granted.

Accordingly, the requirements of Article 123(2) and (3) EPC are satisfied, which finding was not contested by the Appellant.

According to the Appellant, the subject-matter of the claim 1 as amended lacked clarity, Article 84 EPC, since it was not clear whether the temperature range indicated in claim 1 applied only to the equation setting out the upper limit of hydrogen chloride or whether this was a limiting feature of the process.

The Board however does not see any ambiguity in the subject-matter of claim 1 of the second auxiliary request, since it is clear from the wording of claim 1 that the said temperature range applies to the aniline/ water mixture in which hydrogen chloride is absorbed. The Applicant’s argument under Article 84 EPC is therefore rejected.

5. *Inventive step*

Document (1) remains the closest prior art and the technical problem underlying the patent-in-suit remains the provision of a further process for preparing diamino diphenyl methane. The proposed solution is the process of claim 1 of the second auxiliary request characterized in that

(i) an aniline/water mixture with a water content of 0.1 to 7 wt% is prepared and,
(ii) hydrogen chloride gas is absorbed into said aniline/water mixture.

This claimed solution clearly requires the preliminary step of absorbing hydrogen chloride in aniline to be performed before the main reaction.

According to the Appellant, it was obvious for the skilled person to modify the order for introducing reactants into the reactor. However, the claimed process does not only differs from that disclosed in document (1) in the order of introducing the reactants in the reactor, but also in the preparation of an aniline/water mixture with the specific water content of 0.1 to 7 wt% (step (i) of the claimed process). This technical measure avoids the precipitation of aniline chloride salt during the step of absorbing hydrogen chloride in aniline (step ii), which would have resulted from the mere inversion of the order of introducing the reactants, thus maintaining full efficiency of the catalytic activity of hydrogen chloride.

Furthermore, the Board is not aware of any documents cited in the opposition/appeal proceedings which render the proposed solution obvious.

Consequently, the Board comes to the conclusion that the subject-matter of claim 1 of the second auxiliary request is not obvious in the light of the prior art.

An inventive step can therefore be acknowledged for the subject matter of claim 1, and by the same token for

the subject-matter of dependent claim 2, of the second auxiliary request.

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 amended form on the basis of claims 1 and 2 of the second auxiliary request filed in the oral proceedings before the Board and a description yet to be adapted.

The Registrar: The Chairman:

C. Rodríguez Rodríguez P. Gryczka

Decision electronically authenticated