

Internal distribution code:

- (A) [] Publication in OJ
(B) [] To Chairmen and Members
(C) [X] To Chairmen

D E C I S I O N
of 22 April 1996

Case Number: T 0884/92 - 3.3.3

Application Number: 88110369.1

Publication Number: 0297536

IPC: C08G 65/32

Language of the proceedings: EN

Title of invention:
N-(polyoxyalkyl)-N-(alkyl)amines

Applicant:
UNION CARBIDE CORPORATION

Opponent:
-

Headword:
-

Relevant legal provisions:
EPC Art. 56

Keyword:
"Inventive step (affirmed)"
"Choice of closest state of the art - definition of technical problem"

Decisions cited:
T 0026/85; T 0246/91; T 0495/91

Catchword:
-



Case Number: T 0884/92 - 3.3.3

DECISION
of the Technical Board of Appeal 3.3.3
of 22 April 1996

Appellant: UNION CARBIDE CORPORATION
39 Old Ridgebury Road
Danbury
Connecticut 06817 (US)

Representative: Wuesthoff, Franz, Dr.-Ing.
Wuesthoff & Wuesthoff
Patent- und Rechtsanwälte
Schweigerstrasse 2
D-81541 München (DE)

Decision under appeal: Decision of the Examining Division of the European
Patent Office dated 13 May 1992 refusing European
patent application No. 88 110 369.1 pursuant to
Article 97(1) EPC.

Composition of the Board:

Chairman: C. Gérardin
Members: R. Young
J. A. Stephens-Ofner

Summary of Facts and Submissions

I. European application No. 88 110 369.1, relating to "N-(polyoxyalkyl)-N-(alkyl)amines", filed on 29 June 1988 and published under No. 0 297 536 was refused by a decision of the Examining Division dated 13 May 1992, for lack of inventive step having regard to the disclosure of the document:

D1: WO-A-86/05795.

II. According to the decision, the secondary amine terminated polyethers claimed in the application in suit were distinguished from those of D1, which was considered to be the closest state of the art, essentially in that they had a specific equivalent molecular weight in combination with a specific amine content. Whilst this combination was not expressis verbis disclosed in D1, the latter disclosed secondary amine terminated polyethers with a degree of amination from 25 to 100% used in the manufacture of polyurea products. The experimental data which had been submitted in the form of an Affidavit were not considered relevant for substantiating an inventive step, since they were based on a comparison with primary amine terminated polyethers (rather than secondary amine terminated polyethers) and could not therefore be considered to have been made with the closest state of the art. Since there was no evidence that the claimed secondary amine terminated polyethers led to unexpected properties in the polyurea products, compared with those obtained from the secondary amine terminated polyethers disclosed in D1, the subject-matter claimed in the application was an arbitrary selection from the prior art and as such lacked an inventive step.

III. On 13 July 1992, a Notice of Appeal against the above decision was filed, the prescribed fee being paid on the same day.

:

In the Statement of Grounds of Appeal filed on 11 September 1992, the Appellant argued essentially as follows:

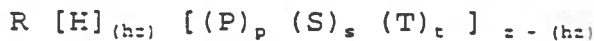
- (a) The claimed compounds were N-(polyoxyalkyl)-N-(alkyl)amines containing at least 3 polyether chains each containing at least 20 residues of propylene oxide and which were at least 70% capped by amino functionalities, at least 50% of the latter being secondary amino groups. This combination of features was not disclosed in D1, nor was there the slightest incentive to provide such compounds.
- (b) It was not accepted that the compounds of D1 in general were used for the manufacture of polyurea products, since any mention of secondary amine terminated polyethers in D1 referred only to a first step, in which an amine functional compound was reacted with an excess of a polyisocyanate to form an isocyanate-terminated prepolymer or quasi-prepolymer. There was no hint of their use in a method for producing polyurea foams.
- (c) A comparison between D1 and the claimed subject-matter could only be made concerning the Zerewitinoff active compounds actually employed in the process of producing polyurea foams. In this connection, D1 mentioned secondary amines only in general terms, the remaining relevant compounds not being comparable with the claimed compounds.

(d) The compounds referred to in the decision under appeal were intended for a different kind of use, which had to be regarded as a functional feature, and therefore could not serve as a comparative basis. The correct comparison was with primary amine terminated polyethers as provided by the Affidavit, which showed that the claimed compounds met certain needs in the production of polyurea foams.

IV. A communication was issued on 10 October 1995, setting out the result of an interview held between the Representative of the Appellant and the Rapporteur of the Board on 5 October 1995, in which a number of preliminary issues concerning the wording of the claims had been discussed.

V. On 22 November 1995, the Appellant filed a revised set of Claims 1 to 7 to be considered as his sole request. This was followed, on 6 December 1995, 11 December 1995 and 10 April 1996, by further revised sets of Claims 1 to 7 in replacement thereof. Claim 1 of the final set reads as follows:

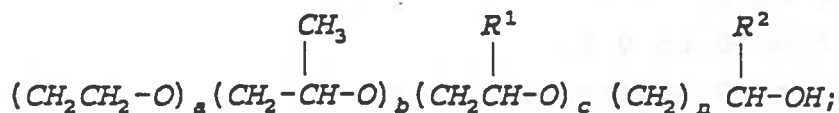
"N-(polyoxyalkyl)-N-(alkyl)amines of the formula:



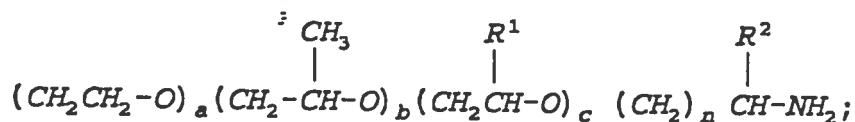
wherein:

R is an initiator radical based on a compound containing Zerewitinoff active hydrogen atoms;

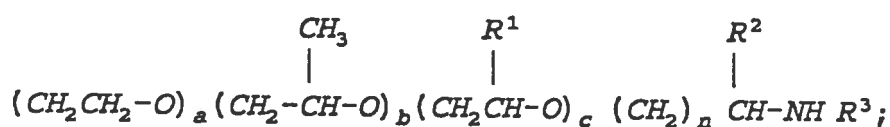
H is the group represented by the formula:



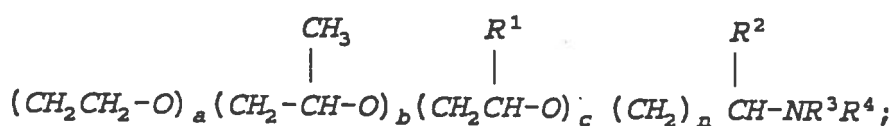
P is the group represented by the formula:



S is the group represented by the formula:



T is the group represented by the formula:



R¹ is an alkyl group containing from 2 carbon atoms to 18 carbon atoms;

R² is hydrogen or an alkyl group containing up to 18 carbon atoms;

R³ and R⁴ are independently alkyl groups containing from 2 to 12 carbon atoms;

a is 0 to 175;

b is 20 to 115;

c is 0 to 30;

n is 1 to 3;

h is from 0 to 0.3;

p is from 0 to less than 0.5;

s is from 0.5 to 1.0;

t is from 0 to 0.15; the sum of p, s and t must equal 1;

and

z is an integer derived from the number of Zerewitinoff active hydrogens on the initiator, z being 3-6."

Dependent Claims: 2 to 7 relate to elaborations of the amines according to Claim 1.

- VI. The Appellant requested that the decision under appeal be set aside, and that a patent be granted on the basis of the set of Claims 1 to 7 filed on 10 April 1996. In the alternative, he requested oral proceedings.

Reasons for the Decision

1. The appeal is admissible.

2. *Allowability of amendments*

The text of the patent application forming the subject of the appeal consists of Claims 1 to 7 filed on 10 April 1996 and the description as originally filed.

Claim 1 differs from the original version by a narrower definition of the parameters b, h and z, which are now restricted to respective preferred ranges, as well as an explicit statement that the sum of p, s and t be equal to 1. These amendments are supported by the description on page 4, line 52 and page 5, lines 20, 21, 30 and 32 of the application as published (page 8, second complete paragraph, page 9, fourth paragraph, and page 10, first two paragraphs of the description as originally filed). Claims 2, 3, 4, 6 and 7 have been amended to remove redundant features and provide consistency with Claim 1. The remaining Claim 5 is unchanged compared with the version as originally filed.

Consequently, there are no objections under Article 123(2) EPC to the amended claims.

3. *The closest state of the art: the technical problem*

The application in suit is concerned with N-(polyoxyalkyl)-N-(alkyl)amines, which have utility in the preparation of polyurea and polyurethane/urea elastomers and foams (page 2, lines 9 to 11).

According to the application, a class of such compounds having the potential to meet the requirements imposed by more rigorous applications such as high performance reaction injection moulding (RIM) and rapid demould foams and elastomers is taught in D1A: US-A-3 654 370. These materials are amine terminated polyethers where the hydroxyl group of a conventional polyol has been replaced with a primary amino group (page 2, lines 31 to 40).

These primary amines are, however, extremely reactive with isocyanates and their utility is correspondingly limited (page 2, lines 47 to 50).

Thus, in the reaction of these primary amines with isocyanates to form polyurea and polyurethane/urea elastomers and foams, a disadvantage is the very rapidity of the reaction, which gives rise to difficulties in process control, which it is an object of the application to overcome (page 2, lines 47 to 50; page 3, lines 30 to 34).

3.1 A key issue to be decided in this appeal is whether the decision under appeal was correct in holding that D1 was the closest state of the art with respect to which any relevant comparison should be made, and in particular whether it represented a closer state of the art than that already acknowledged in the application, based on D1A (section 3, above), in respect of which the Appellant had already filed a comparative test (section III (d), above).

3.2 The Boards of Appeal have held on more than one occasion that an objective definition of the technical problem to be solved should normally start from the technical problem actually described by the Applicant. Only if it turns out that an incorrect state of the art was used to define the technical problem or that the technical problem disclosed has in fact not been solved, can an inquiry be made as to which other technical problem objectively existed (see T 0246/91 of 14 September 1993, No. 4.4 of the Reasons for the Decision, and T 0495/91 of 20 July 1993, No. 4.2 of the Reasons for the decision, neither published in the OJ EPO).

Whilst both of these decisions concern granted patents, their legal principles are clearly not limited to post-grant proceedings.

3.3 Consequently, the questions which need to be addressed in this appeal are (a) whether the technical problem described in the application is solved and (b) whether this was the correct problem to consider. The latter question boils down to whether D1 represents a closer piece of the state of the art than that already acknowledged in the application in suit.

- 3.4 Compared with the state of the art acknowledged in the application in suit, the technical problem objectively arising is to provide amines which can be employed, with improved process control, in reactions with isocyanates directly to form polyurea and polyurethane/urea elastomers and foams.
- 3.5 The solution proposed according to Claim 1 of the application in suit is to replace the primary amine terminated polyoxyalkylene ethers of D1A by the defined N-(polyoxyalkyl)-N-(alkyl)amines, which are characterised by containing at least 3 polyether chains (z is at least 3) each containing at least 20 residues of propylene oxide (b is at least 20), at least 70% of which are capped by amino functionalities (h is not more than 0.3), at least 50% of the latter being secondary amino groups (s is at least 0.5).
- 3.6 It can be seen from the experimental data in the application in suit, especially the relative gel times for the reaction, with isocyanates, of the amine terminated polyether of Example 1 (molecular weight 5 000; derived by catalytic amination of a glycerine-based polyoxyalkylene triol to 93% amine conversion) having 50% primary and 50% secondary amine termination, compared with a commercially available amine species of similar molecular weight and having 100% primary amine termination (Jeffamine™ T-5000), that a slower and more controlled reaction is achieved with the former than with the latter (page 12; Table).
- 3.7 These results are corroborated by the comparative test report contained in the Affidavit of R. M. Gerkin, filed on 22 May 1991 with the submission dated 21 May 1991, in which a secondary amine terminated polyether (containing 73% secondary amine) reacted slowly enough that the composition could be mixed by hand and the mould cavity

was fully filled and produced a high quality product, whereas the corresponding species with essentially 100% primary amine termination (Jeffamine T-5000) reacted so quickly that it gelled and failed completely to fill the mould cavity and thus produced a product which was poorly mixed.

Thus, it is credible that the proposed measures provide an effective solution of the technical problem identified and presented in the application in suit.

3.8 Since condition (a) in section 3.3, above, is evidently fulfilled, it only remains to investigate condition (b), i.e. to assess whether D1 represents a closer state of the art than the one acknowledged in the application.

3.9 According to D1, which relates to a process for preparing polyurea and/or polyurea-polyurethane polymers, particularly non-cellular or microcellular elastomeric and structural polyureas, an amine functional compound having an equivalent weight of at least 400 is reacted with an excess of a polyisocyanate to form an isocyanate-terminated prepolymer or quasi-prepolymer which has an isocyanate content of from 3 to 12 percent by weight. This prepolymer is then reacted with an isocyanate reactive material to form a polyurea and/or polyurethane polymer (page 1, opening paragraph, and Claim 1).

3.9.1 Preferred amine functional compounds are primary or secondary amine-terminated polyethers containing an average of from 1 to 4 amine groups per molecule (page 6, lines 27 to 31). The production of secondary amine-terminated polyethers is stated to be described in particular in US-A-4 152 353 and US-A-4 153 581 (page 7, lines 14 to 20). According to the latter documents (hereinafter D1C and D1D, respectively), an alcohol,

aldehyde or ketone or mixture thereof, for example polypropylene glycol, is contacted, in the presence of a catalyst composition, for instance of Ni, Cu and Fe (D1C) or Co, Cu and Fe (D1D), with an aminating agent, which may be ammonia, or a primary or secondary amine, to obtain primary, secondary or tertiary amines respectively, the use of ammonia being exemplified.

3.9.2 Further according to D1, due to the great reactivity of the amine functional compound, it tends to form polymers of significant molecular weight upon contact with the polyisocyanate, which broadens the molecular weight distribution and affects the properties of the polymer. The reaction, which is usually almost instantaneous, is conducted until substantially all the amine groups have reacted with the polyisocyanate (page 10, lines 7 to 16; page 11, lines 14 to 19).

3.9.3 Stability of the resulting prepolymer is generally favoured by the use of less reactive polyisocyanates and less than quantitatively aminated amine functional compounds. Secondary amine-terminated amine functional compounds form more stable prepolymers than do primary amine terminated materials (page 12, lines 1 to 18).

Highly reactive isocyanates typically form stable prepolymers with primary amine terminated compounds having a degree of amination of 25 to 85%, and with secondary amine terminated materials having a degree of amination of 25 to 100% (page 13, lines 13 to 21).

3.9.4 According to Example 1, an aminated polyoxypropylene diol containing 50 percent primary amine groups, 37 percent methylamine (secondary amine) groups and 3 percent hydroxyl groups, prepared according to the process described in D1C is added to hexamethylene diisocyanate (HMDI) to yield a prepolymer having a -NCO

content of 8.9 percent. This prepolymer has a gel time of 30 sec. when reacted with diethyltoluenediamine (DETDA), the resulting elastomer having a glass transition temperature of -55°C , compared to an expected value of -40°C (pages 18, 19).

- 3.9.5 Hence, although secondary amine terminated polyethers having a degree of amination of up to 100% (section 3.9.3, last sentence, above) are referred to in D1, only those having a measure of secondary amine termination of up to about 37% (Example 1) are explicitly disclosed in individualised form suitable for forming the basis of a practical comparison (page 13, lines 18 to 21).
- 3.9.6 Furthermore, even the references to the methods of preparation of such secondary amine terminated polyethers in D1C and D1D, referred to in D1, although mentioning both primary and secondary amine terminated polyethers, exemplify only the use of ammonia as aminating agent, which would result in essentially only primary amine terminated species being obtained.
- 3.9.7, Indeed, according to a submission of the Appellant, filed on 22 May 1991, attempts to apply the methods of D1C and D1D to obtain higher proportions of secondary amine terminated polyethers had not been successful in achieving more than the approximately 37% secondary amine groups reported in Example 1 of D1 (cf. submission, page 4, last paragraph).

The latter submission, which is of considerable significance to the outcome of the appeal, whilst not being supported by concrete experimental evidence nevertheless has not been contested, or indeed commented upon in the decision under appeal, and is in any case consistent with the complete absence, from the cited

documents, of any individualised disclosure of an amine terminated polyether having a proportion of secondary amine termination substantially more than this level. Accordingly, the Board has no reason to doubt the accuracy of the submission.

Consequently, on the available evidence, there is no reason to conclude that D1 contains any teaching enabling the skilled person to obtain secondary amine terminated polyethers having a proportion of secondary amine termination of more than about 37%.

3.9.8 In any case, these secondary amine terminated polyethers are not disclosed in D1 for direct use in the formation of a polyurea or polyurethane/urea product, but only for the formation of an isocyanate terminated prepolymer, in which all the active hydrogen atoms present in the amine groups have been consumed by reaction with isocyanate.

Thus, there is no disclosure in D1 of the relevant effect with which the application in suit is concerned, namely the speed of reaction, in direct polymer formation techniques such as RIM and rapid demold foams and elastomers, of these active hydrogen atoms with isocyanate, which is the factor limiting process control (section 3, above).

On the contrary, an important aim of the prepolymer process according to D1 is to avoid the problems arising in RIM processes (page 3, lines 3 to 15).

In other words, those amine terminated polyethers which, according to D1, have structural approximation to those defined in Claim 1 of the application in suit (i.e. those containing about 37% secondary amine groups), are not disclosed for a comparable use.

3.9.9 In this connection, the only amine chain extender which is exemplified in D1 for use in a reaction which yields a final polymer product (i.e. for reaction with the prepolymer) is DETDA (Examples 1 to 5). This is, however, a primary amine which, furthermore, being aromatic, lies further from the claimed subject matter than the aliphatic primary amine polyethers belonging to the state of the art acknowledged in the application.

Consequently, D1 does not represent a closer state of the art than that acknowledged in the application in suit itself.

3.10 To read D1 as effectively disclosing the use of such secondary amine terminated polyethers as reactants directly in such a process would represent a radical departure from, not to say a direct contradiction of, what is actually stated in D1.

To insist on using such a modification of the disclosure of D1 as a starting point in assessing patentability, as was done in the decision under appeal (Reasons, paragraph (2)(b)), is, moreover, tantamount to imposing an arbitrarily generated variant lying substantially closer to the claimed subject-matter than D1, as the basis for comparison.

Such a variant does not, however, belong to the state of the art within the meaning of Article 54 EPC, since it has not been made available to the public before the relevant filing date (T 0026/85, OJ EPO 1990, 022). Logically, therefore, it cannot constitute "the closest state of the art" in the sense the term is used in the application of the problem and solution approach.

Hence, the requirement for a comparison test of the kind demanded in the decision under appeal was not justified.

- 3.11 The further finding of the decision under appeal, that the essential difference from D1 is the selection of specific secondary amine terminated polyethers from those disclosed in D1 is inappropriate, because the level of secondary amine termination evidently achievable according to D1 is not high enough to embrace that characterising the solution of the stated problem (section 3.9.7, above).

Consequently, the secondary amine terminated polyethers used in the process according to the application in suit are not to be understood as a selection from those disclosed in D1.

- 3.12 In summary, the comparative test provided by the Appellant (section 3.7, above) constitutes, in the Board's view, a fair comparison with the closest state of the art, and the effect demonstrated may properly be taken into account in the assessment of inventive step.

4. *Novelty*

The decision under appeal does not allege lack of novelty in respect of the amines of Claims 1 to 7. Nor does the Board see any ground for raising such an objection.

Consequently the subject-matter of Claims 1 to 7 is held to be novel.

5. *Inventive step*

In view of the conclusions reached in section 3 etc., above, the question which has to be answered is whether the skilled person would have expected that modification of the primary amine terminated polyethers disclosed in D1A by replacing the essentially 100% primary amine

terminated polyethers by such polyethers in which at least 50% of the terminal amine groups are secondary amine groups in a high molecular weight trifunctional polyether as defined in the solution to the stated problem, would lead to improved process control in the direct (one-shot) preparation of polyurea foams or elastomers.

5.1 There is, of course, no suggestion to make such a replacement in the state of the art acknowledged in the application in suit. This applies in particular to the use of polyoxyalkylene amines produced according to the process of D1A, in which the reductive amination of polyoxyalkylene alcohols is done using ammonia, and therefore results in the formation of primary amines (section 3, above).

5.2 As to D1, the relevant behaviour of both primary and secondary amine terminated polyethers is covered by a single statement, according to which, "Due to the great reactivity of the amine functional compound it tends to form polymers of significant molecular weight upon contact with the polyisocyanate" (page 10, lines 7 to 10; section 3.9.2, above).

Consequently, the skilled person would have had no reason from the disclosure of D1 to expect that amines having a high proportion of secondary amine groups, let alone the other characteristics defined, would provide a solution of the technical problem.

5.3 In summary, the solution of the stated problem does not arise in an obvious way starting from the state of the art acknowledged in the application.

- 5.4 Even if one were to start, as did the decision under appeal, from the disclosure of D1 as closest state of the art, the result would not be different, for the following reasons.
- 5.4.1 D1 does not suggest the utility of the secondary amine terminated polyethers for the purpose with which the application in suit is concerned (section 3.9.8, above). Consequently, the stated problem is not derivable from its disclosure, let alone its solution (sections 3.4, 3.5, above).
- 5.4.2 Even ignoring the terms of the stated problem and considering the disclosure of D1 in isolation, the only incentive for using secondary amine terminated polyethers is that they form more stable prepolymers (D1, page 12, lines 9 to 18: section 3.9.3, above).
- 5.4.3 Quite apart from the fact that the secondary amine terminated polyethers exemplified in D1 are based on diols and not triols as required by Claim 1 of the application in suit, the disclosure of D1 evidently does not in itself enable such secondary amine terminated polyethers to be obtained with more than about 37% secondary amine termination (sections 3.9.5 to 3.9.7, above).

Consequently, there is an obstacle to obtaining secondary amine terminated polyethers according to Claim 1 of the application in suit.

- 5.4.4 Since, furthermore, the teaching of D1 does not attach any particular importance to achieving a level of secondary amine termination above that already exemplified (approximately 37%), it cannot be regarded as obvious for the skilled person to search for, let alone attempt extra measures to go beyond this level.

5.5 In other words, the subject-matter of Claim 1 does not arise in an obvious way from the state of the art. Hence, it involves an inventive step. By the same token, the subject-matter of Claims 2 to 7, which are dependent on Claim 1, also involves an inventive step.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The case is remitted to the first instance with the order to grant a patent on Claims 1 to 7 filed on 10 April 1996, and a description yet to be adapted.

The Registrar:


E. Görgmaier

The Chairman:


C. Gérardin

