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Bezeichnung der Erfindung:
Title of invention:
Titre de l'invention : Polyol compositions and polyurethane foams manufactured therefrom

Klassifikation / Classification / Classement : C08G 18/40

ENTSCHEIDUNG / DECISION

vom / of / du 22 June 1990

Anmelder / Applicant / Demandeur : Imperial Chemical Industries PLC

Patentinhaber / Proprietor of the patent /
Titulaire du brevet :

Einsprechender / Opponent / Opposant :

Stichwort / Headword / Référence :

EPÜ / EPC / CBE Article 56

Schlagwort / Keyword / Mot clé : "Inventive step (yes) - non-obvious combination of structural features improving a specific property"

Leitsatz / Headnote / Sommaire

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Case Number : T 390/89 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 22 June 1990

Appellant : IMPERIAL CHEMICAL INDUSTRIES PLC
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Decision under appeal : Decision of Examining Division 012 of the European Patent Office dated 4 January 1989 refusing European patent application No. 85 300 654.2 pursuant to Article 97(1) EPC

Composition of the Board :

Chairman : F. Antony
Members : C. Gérardin
J. Stephens-Ofner

Summary of Facts and Submissions

I. European patent application No. 85 300 654.2 filed on 31 January 1985, claiming priority of 8 March 1984 from an earlier application GB-8 406 050 and published under the publication number 161 039, was rejected by a decision of the Examining Division dated 4 January 1989.

That decision was based on a set of three claims as filed originally and amended according to the request of 1 July 1988, of which Claim 1 reads as follows:

"A polyol composition comprising:

(A) A polyester polyol containing residues derived from succinic, glutaric and adipic acids having a hydroxyl number in the range 150 to 450 and 0.5 to 2.0 branch points per 1000 units of molecular weight, and

(B) a non-polymeric or polyoxyalkylene polyol having a functionality of from 3 to 4 and a hydroxyl number of at least 1120, the relative proportions of A and B being such that the composition has a hydroxyl number in the range 400 to 600."

II. The only ground for this decision was non-compliance with the requirements of Article 56 EPC with regard to the teaching of FR-A-2 379 557 (document (1)) and GB-A-1 475 541 (document (2)). More specifically, it was stated in the decision that the preparation of rigid polyurethane foams from polyol blends comprising a polyester polyol derived from a technical mixture of succinic, glutaric and adipic acids and a polyether polyol with a hydroxyl number of 25 to 800 was generally known from document (1).

Similarly, the use of polyester polyols having hydroxyl numbers in the range 200 to 800 together with triethanolamine for the preparation of rigid polyurethane foams could be regarded as being implicitly disclosed in document (2). The comparative data provided by the Applicant (Appellant) in order to demonstrate that the hydroxyl number of the polyether component was a critical parameter as far as shrinkage was concerned, could not be regarded as conclusive. In the absence of any convincing evidence to the contrary, the claimed subject-matter was thus merely judged to be an arbitrary selection from the prior art teaching not involving any inventive step.

III. A notice of appeal was lodged against that decision on 1 March 1989 with payment of the prescribed fee. The arguments presented in the Statement of Grounds of Appeal filed on 22 March 1989 were, in the first place, based on the result of the comparative example previously filed during the examination procedure on 1 July 1988. This example showed that, other compositional features being equal, hydroxyl numbers of the polyether polyol of at least 1120 had a beneficial effect on shrinkage. In the second place, additional experimental data filed with the Statement of Grounds of Appeal demonstrated that the use of such polyether polyol resulted in improved dimensional stability of the polyurethane foam at low temperature. These advantages were unexpected in view of the broad disclosure of document (1) which encompassed many alternatives; none of these suggested the present range for the hydroxyl number of the polyether polyol and, therefore, the present combination of polyols.

IV. In accordance with these arguments, the Appellant filed a new Claim 1 on 15 June 1990, wherein component (B) was defined as follows: "as the other organic polyol a polyoxyalkylene polyol having a hydroxyl number of at least

1120 obtained by reacting propylene oxide and/or ethylene oxide with an initiator having a functionality of from 3 to 4".

V. The Appellant requested that the decision under appeal be set aside and a patent be granted on the basis of Claim 1 filed on 15 June 1990 and Claims 2 and 3 as filed on 23 September 1987.

Reasons for the Decision

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

2. The wording of Claim 1 does not give rise to objections under Article 123(2) EPC.

The only amendments with regard to original Claim 1 concern the definition of component (B) which is now limited to a polyoxyalkylene polyol having a hydroxyl number of at least 1120 obtained by reacting propylene oxide and/or ethylene oxide with an initiator having a functionality of from 3 to 4. As mentioned on page 5 of the Statement of Grounds of Appeal, the expression "as the other organic polyol" before the proper definition seeks to specify that the polyol composition contains only two polyol components, i.e. a polyester polyol as component (A) and a polyether polyol as component (B); this is a clear limitation with regard to original Claim 1, wherein not only non-polymeric polyols were included as component (B), but also further unspecified polyols, besides the two components (A) and (B), as the result of the presence of the non-limiting wording "comprising". The method of preparation ("obtained by..."), based on the reaction between the alkylene oxide and an initiator, is supported by the original description (page 3, lines 16 to 19), whilst the lower limit of 1120

for the hydroxyl number corresponds to the value of this parameter for the oxypropylated glycerol used as polyether in Formulation 2 given on page 6, lines 1 to 3 of the original application. The Board is aware that this value was only disclosed in the application as filed in the context of a particular polyol composition, not as a general feature in isolation; however, since the other major component of the composition is not a specific polyester, but a class of polyester polyols having between 0.5 and 2.0 branch points per 1000 units of molecular weight and derived from a technical mixture of acid esters in variable proportions, Formulation 2 can be regarded as a broad definition encompassing various polyol compositions having the same polyether polyol (B) in common. For this reason, given the specific context of this case, the Board is willing to follow the line of the Examining Division and accept the generalisation of 1120 as lower limit of the hydroxyl number to any polyoxyalkylene polyol.

As to Claims 2 and 3, they correspond to the original version of these claims, except for the correction of an inconsistency noted in the Examining Division's communication of 25 March 1987, bottom of page 2.

3. The application in suit concerns polyol compositions and a method for the preparation of a rigid polyurethane foam using these compositions. Such polyol compositions and their application are described in document (1). According to this disclosure, rigid polyurethane foams are obtained by reacting organic polyisocyanates with polyhydroxy compositions in the presence of conventional additives, the polyhydroxy compositions comprising combinations of polyester polyols and polyether polyols. These polyester polyols are obtained by reacting a dicarboxylic acid mixture which contains from 20 to 35% by weight of succinic

acid, from 30% to 50% by weight of glutaric acid and from 20% to 32% by weight of adipic acid, based on the total weight of the said dicarboxylic acids, with an isopropanolamine mixture and/or hexanetriol isomer mixture (Claim 1); the resulting polyesters have hydroxyl numbers of from 180 to 650 and functionalities of from 2.5 to 6 (page 5, lines 32 to 36), which corresponds to from 0.64 to 7.7 branch points per 1000 units of molecular weight (Applicant's letter received on 23 September 1987, page 3, paragraphs 1 and 2). The polyether polyols, which have molecular weights of from 200 to 6500 and hydroxyl numbers of from 25 to 800, are prepared by condensing one or more alkylene oxides with a starter molecule which contains at least two active hydrogen atoms in the molecule (page 6, lines 12 to 18). The polyhydroxy compositions containing such polyester polyols and polyether polyols are particularly suitable as polyol components for the preparation of rigid polyurethane foams; although these foams exhibit a desirable combination of physical and mechanical properties, especially a fine cell structure and a high compressive strength, they do tend to shrink shortly after preparation or in subsequent testing under cold storage conditions.

In the light of this prior art teaching, the technical problem underlying the application in suit can be seen in providing a polyol composition giving rise to polyurethane foams with improved dimensional stability, without impairing their high compressive strength properties.

According to the application in suit, this problem is solved schematically by selecting a narrow range of from 0.5 to 2 branch points per 1000 units of molecular weight in the polyester polyol, and by choosing a specific polyether polyol having a hydroxyl number of at least 1120.

In view of the examples in the application in suit as well as the comparative examples filed during the examination procedure and together with the Statement of Grounds of Appeal, the Board is satisfied that the above defined technical problem has been effectively solved.

4. After examination of documents (1) and (2), the Board has reached the conclusion that the claimed solution is not disclosed therein and that the subject-matter of the application in suit is, therefore, novel. Since the issue of novelty was not raised in the decision under appeal, it is not necessary to consider this matter in detail.

5. It still remains to be examined whether the claimed subject-matter involves an inventive step.

5.1 The first feature to be considered is the structure of the hydroxy reactants to be used with the technical mixture of dicarboxylic acids for the manufacture of the polyester polyols described in document (1). These reactants, which are defined as isopropanolamine mixtures containing 5 to 40% by weight of diisopropanolamine and 95 to 60% by weight of triisopropanolamine and/or hexanetriol isomer mixtures containing 40 to 99.8% by weight of 2,4-dihydroxy-3-methylolpentane as major component, are in fact very specific compounds (page 3, lines 5 to 36). It clearly appears from Table II on page 14 that their combination with the technical mixture of dicarboxylic acids is essential to obtain polyurethane foams with good mechanical properties; the comparison of deflection and compressive strength of polyurethane foams in Examples 8 to 14, wherein polyesters derived from these specific isopropanolamine and/or hexanetriol isomer mixtures are used, and in Comparative Examples A to C, wherein a conventional polyester is used, shows that these properties are on

average better for the particular reactants than for the commercial product. For the skilled man, faced with the problem of at least not impairing the high compressive strength properties referred to in document (1), there was thus no reason to depart from the advantageous compositional combinations disclosed in this prior art.

As the Appellant showed in his submission filed on 23 September 1987 during the examination procedure (page 2, paragraph 3 to page 4), the branch points per 1000 units of molecular weight, which can be calculated from the hydroxyl numbers and the functionalities of the polyester polyols described in document (1), correspond to a range of from 0.64 to 7.7. The actual values of this parameter in the seven examples of that document vary between 2.52 (Example 4) and 4.27 (Example 6). Neither the general range, nor these specific values, would point to the selection made by the Appellant, which is a comparatively narrow range at the lower end of that disclosed by the prior art.

5.2 Unlike the polyether polyols according to document (1), which have hydroxyl numbers of from 25 to 800 (page 6, lines 12 to 15), the hydroxyl number of component (B) according to Claim 1 of the application in suit is at least 1120. As the Appellant demonstrated, this is a critical parameter for the dimensional stability of the polyurethane foams.

For the purpose of comparison, the Reference Example with Formulation 2 according to the application in suit was repeated by modifying only the hydroxyl number of the polyether polyol in the polyol composition (comparative example filed on 1 July 1988). Formulation 2 in the application in suit designates a composition containing, besides a diphenylmethane diisocyanate composition and

various conventional ingredients, a polyol composition which is the mixture of a polyester having a hydroxyl number of 250 and between 0.5 and 2 branch points per 1000 units of molecular weight and a polyether polyol which is an oxypropylated glycerol having a hydroxyl number of 1120 (page 5, lines 2 to 4; page 5, line 28 to page 6, line 3); visual control shows that the rigid polyurethane foam made from Formulation 2 does not shrink. By contrast, when the hydroxyl number of the oxypropylated glycerol is 540, thus within the range envisaged in the prior art, the foam shows shrinkage under the same conditions.

Example A, filed with the Statement of Grounds of Appeal, provides evidence that the influence of the hydroxyl number of the polyester polyol employed on the properties of the polyurethane foam is even more pronounced at low temperatures. A polyol composition based on the polyester polyol having a hydroxyl value of 338 according to Example 1 of document (1) and an oxypropylated glycerol having a hydroxyl value of 540 - thus lying within the scope of that document as well - as polyether polyol was used to prepare a polyurethane foam. The cold storage stability of that foam was determined by keeping a piece of foam at -18°C for 24 hours, measuring the decrease in length and width, and calculating the stability figure from the formula (proportional change in length + proportional change in width):2. Whereas the stability figure of the prior art foam was -4.5, which is totally unacceptable from a commercial point of view, it was only -0.1 for the foam prepared from the polyol composition according to Formulation 2.

Besides the hydroxyl number of the polyether polyol, it is essential to consider the chemical composition of this component. Although ethylene oxide and propylene oxide are mentioned in document (1) as preferred alkylene oxides to

be reacted with the initiator for the preparation of the polyether polyol, compounds which are structurally quite different, such as styrene oxide and tetrahydrofuran, would be suitable as well (page 6, lines 18 to 23); likewise, compounds having a high hydroxy functionality, such as sorbitol and saccharose, would be appropriate initiators according to the prior art (page 7, line 2), but would be outside the scope of the application in suit. The broad definitions of the alkylene oxide and the initiator in the prior art thus contrast with the restrictive definition of component (B) in Claim 1 of the application in suit which has to be the reaction product of propylene oxide and/or ethylene oxide with an initiator having a functionality of from 3 to 4.

For the above reasons, in the Board's judgement, the requirements in terms of functionality of the initiator and structure of the alkylene oxide, together with the range of the hydroxyl number of component (B) could not be derived from the teaching of document (1) in order to achieve improved dimensional stability of the polyurethane foams (cf. application in suit, page 4, lines 19 to 21).

5.3 In view of the above and in the absence of any reference in document (1) to the low temperature behaviour of the polyurethane foams, the skilled man would have had no indication whatsoever about the parameters and features likely to influence the dimensional stability of the foams, especially at low temperature, and thus no incentive to carry out routine investigations.

However, even if the skilled man embarked upon such systematic experiments in order to improve a particular property of the polyurethane foams, in the Board's view, he would first consider alternative embodiments within the broad scope of the prior art teaching, not outside.

Following the prior art suggestions, he would thus use low molecular weight polyols in addition to the basic isopropanolamine mixture and/or hexanetriol isomer mixture (page 4, lines 9 to 16), or replace part of the technical mixture of dicarboxylic acids by aromatic and/or ethylenically unsaturated dicarboxylic acids (page 4, line 36 to page 5, line 12) in the preparation of the polyester polyol (A), or further employ chain extenders of various functionalities and/or molecular weights in addition to the high molecular weight polyhydroxy components (A) and (B) (page 8, lines 9 to 12), or even incorporate various assistants and additives into the reaction mixture (page 9, lines 15 to 21), all such steps being known to modify the properties of the final polymer. In the Board's view, such modifications within the scope of document (1) were more obvious than, for instance, the adoption of 1120 as lower limit for the hydroxyl number of the polyether polyol which is outside the scope of this document.

Furthermore, as shown above, the polyol compositions according to the application in suit differ from those described in document (1) not by one feature only, but in several respects. To arrive at the claimed polyol compositions from the general disclosure of document (1), it was necessary, first, to select a degree of branching at the lower end of the permitted range in the polyester polyol known from document (1) and to forgo the contribution of specific trifunctional compounds to the mechanical properties of the polyurethane foams, then, to adopt a range of high values for the hydroxyl numbers of the polyether polyol outside the scope of the prior art disclosure and to define specific structural requirements for this compound. In the Board's view, this extensive combination of numerous modifications with regard to the prior art polyol composition described in document (1) cannot be the result of mere routine experiments.

5.4 As to document (2), the mere fact that it mentions the preparation of rigid polyurethane foams from polyester polyols having a hydroxyl number in the range 200 to 800 and derived from an acid mixture of from 50 to 60% of glutaric acid, from 15 to 25% of adipic acid and from 20 to 30% of succinic acid, on a weight basis, cannot be regarded as providing an incentive to use mixtures of such polyester polyols and polyether polyols for this reaction, let alone select polyether polyols having a hydroxyl number of at least 1120. Therefore, in the Board's view, this document, either in isolation or in combination with document (1), cannot suggest the claimed subject-matter.

5.5 In conclusion, for the above reasons, the subject-matter of the application in suit cannot be derived from documents (1) and (2) in an obvious manner and, in view of the improved dimensional stability, especially at low temperature, thus involves an inventive step.

6. From a formal point of view, the present wording of Claim 1 overcomes the objection of ambiguity repeatedly put forward by the Examining Division regarding the actual scope of the claim. As pointed out in paragraph 2 above, it is now clear that the claimed compositions contain only two organic polyols, a polyester polyol (A) and a polyether polyol (B); as to the word "comprising", it is justified by the fact that the polyol compositions may be used in the manufacture of polyurethane foams as one component containing the various assistants and additives, the other component being the organic polyisocyanate (application in suit, page 4, lines 11 to 14). Furthermore, the present drafting of Claim 1 removes an inconsistency between the description and Claim 1 as rejected in the definition of the polyether polyol.

7. Claim 1 being allowable, the same applies to Claim 2, which is directed to a preferred polyol composition, and to Claim 3 which concerns a method for the preparation of a rigid polyurethane foam using the polyol composition according to Claim 1 and whose patentability is supported by that of the main claim.

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 the basis of Claim 1 filed on 15 June 1990, Claims 2 and 3 filed on 23 September 1987 and a description yet to be adapted.

The Registrar: The Chairman:

M. Beer F. Antony

CG