

Publication in the Official Journal	Yes / No
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File Number: T 112/90 - 3.3.3

Application No.: 83 112 931.7

Publication No.: 0 112 557

Title of invention: Process for producing a polyurethane elastomer by reaction injection molding.

Classification: C08G 18/48

D E C I S I O N
of 4 October 1991

Applicant: ASAHI GLASS COMPANY LTD

Headword:

EPC Article 56

Keyword: "Inventive step (yes) - evidence provided of technical problem effectively solved"

Headnote



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Boards of Appeal

Chambres de recours

Case Number : T 112/90 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 4 October 1991

Appellant :

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Representative :

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Decision under appeal :

Decision of Examining Division 012 of the
European Patent Office dated 20 September 1989
refusing European patent application
No. 83 112 931.7 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. 83 112 931.7 filed on 21 December 1983, claiming priorities of 24 December 1982 and 8 April 1983 from two earlier applications in Japan and published under the publication No. 0 112 557, was rejected by a decision of the Examining Division dated 20 September 1989.

That decision was based on a set of 18 claims filed on 16 June 1988, of which Claim 1 reads as follows:

"A process for producing a shaped article of a non-cellular or microcellular polyurethane elastomer by reaction injection molding by using at least two components of (A) a polyol component comprising a high molecular weight polyol and a chain extender as essential ingredients and (B) an isocyanate component comprising a polyisocyanate compound as essential ingredient, whereby substantially all or at least 80% by weight of said high molecular weight polyol is composed of a polyoxyalkylene polyol mixture,

c h a r a c t e r i z e d in that the polyoxyalkylene polyol mixture comprises

(1) at least 70% by weight of a polyoxypropylene-oxyethylene triol (a) wherein substantially all of the oxyethylene groups are present at the terminal portions of the oxyalkylene chain, having a hydroxyl value of from 15 to 60, an oxyethylene group content of less than 15% by weight and an unsaturated monool content of at the most 0.085 meq/g, and

(2) up to 30% by weight of other polyoxyalkylene polyol (c),

provided that the polyoxyalkylene polyol mixture has an average hydroxyl functionality of no more than 2.8 and an average hydroxyl value of 15 to 60 and contains at least

5% by weight, in an average, of oxyethylene groups at the terminal portions of the oxyalkylene chain and in which the total oxyethylene group content is, in an average, less than 21% by weight and the average unsaturated monool content is at the most 0.085 meq/g."

Claim 9 is another independent process claim mainly differing from Claim 1 by the fact that the polyoxyalkylene polyol mixture comprises (1) from 60 to 95% by weight of a polyoxypropylene-oxyethylene diol, and (2) from 5 to 40% by weight of other polyoxyalkylene polyol, and further by comparatively minor variations in the amounts and/or ranges of various structural and compositional features.

Claims 2 to 8 and 10 to 18 are dependent claims directed to preferred embodiments of respectively Claims 1 and 9.

II. Following objections of lack of novelty as well as inventive step with regard to the teaching of mainly the following document

(2) EP-A-44 481,

oral proceedings were held on 16 June 1988, during which the above claims were filed. In order to demonstrate that the features which confer novelty to the claimed subject-matter also contributed to an inventive step, the Appellant was invited to submit two comparative tests showing that the compositional features according to Claim 1 led to both shorter moulding time and lower dimensional change upon absorption of water (Minutes of the oral proceedings).

On 6 February 1989 the Applicant filed a single comparative test (Experiment 13) in which Experiment 3 of Table 1 of the application was repeated with a polyol identified as being essentially similar to polyether polyol (e) on page 30 of document (2).

On 20 September 1989 the Examining Division issued the decision to refuse the application on the ground that the requirement of inventive step was not met. More specifically, it was stated in that decision that those comparative data were not sufficient to provide evidence that the claimed advantages were actually achieved.

III. On 20 November 1989 a Notice of Appeal was lodged against that decision with payment of the prescribed fee. Together with the Statement of Grounds of Appeal filed on 30 January 1990 the Appellant (Applicant) submitted another comparative test (Experiment 14), from which the advantages, in terms of moulding time and dimensional stability upon absorption of water, of a mixture of polyether triol and diol over polyether triol alone appeared clearly. Those effects together with the results of Experiment 13 were regarded as surprising.

IV. The Appellant requested that the decision under appeal be set aside and a patent be granted on the basis of the set of Claims 1 to 18 filed on 16 June 1988.

Reasons for the Decision

1. The appeal complies with Articles 106 to 108 and Rule 64 EPC and is, therefore, admissible.
2. The amendments to the claims do not give rise to any objections having regard to Article 123(2) EPC.

2.1 The present wording of Claim 1 differs essentially from that of Claim 1 as originally filed by the fact that (i) the polyoxyalkylene polyol comprises a mixture of at least 70% by weight of triol and up to 30% by weight of another polyol, (ii) substantially all of the oxyethylene groups of the triol are present at the terminal portions of the oxyalkylene chain, (iii) this triol has an oxyethylene group content of less than 15% by weight, and (iv) the polyol mixture has an average hydroxyl functionality of no more than 2.8. Feature (i) is supported by original Claim 3, feature (ii) by original Claim 9, feature (iii) by part (a) of original Claim 2, and feature (iv) by original Claim 6. The other amendments are merely of editorial nature and as such have no impact on the scope of the protection sought.

As far as the dependent Claims 2 to 8 are concerned, they correspond respectively to Claims 7, 8 and 10 to 14 as originally filed with their numbers and appendancies adjusted.

2.2 Claim 9 is another independent claim which essentially differs from Claim 1 as originally filed, read in conjunction with part (b) of original Claim 2, by the fact that (v) the polyoxyalkylene polyol mixture comprises from 60 to 95% by weight of a diol and from 5 to 40% by weight of another polyol, (vi) substantially all of the oxyethylene groups of the diol are present at the terminal portions of the oxyalkylene chain, and (vii) the polyol mixture has an average hydroxyl functionality of 2.1 to 2.49. Feature (v) is supported by original Claim 15, feature (vi) by original Claim 21, and feature (vii) by original Claim 18.

As to the dependent Claims 10 to 18, they correspond to respectively Claims 16, 17, 19, 20 and 22 to 26 as originally filed with their numbers and appendancies adjusted.

3. The application in suit concerns a process for producing a polyurethane elastomer by reaction injection moulding (RIM). Such a process is described in document (2), which the Board, like the Examining Division, regards as the closest state of the art. That citation describes a process for producing a shaped article of a cellular or non-cellular polyurethane elastomer by the RIM technique from a reaction mixture comprising (A) a polyisocyanate, (B) an oxyethylene-tipped polyoxyalkylene polyol, (C) an oxypropylene-tipped polyoxyalkylene polyol, (D) catalysts, and (E) amine chain extenders (Claim 1). The amounts of components (B) and (C) are such that the moulded products contain less than 12% of oxyethylene units, which corresponds to a total amount of these units in the starting polyols of about 16 to 20% by weight (page 19, lines 1 to 6; page 30, footnote (c)). In practice, component (B) contains more than 50%, preferably 80 to 100%, of terminal oxyethylene groups with primary hydroxyl groups (page 18, lines 13 to 19 in combination with the first formula of the same page), an average functionality of 2 to 4, preferably 3 (page 13, lines 23 to 25), and an oxyethylene content of 12 to 22% by weight (Claim 1 and page 12, lines 27/28), in particular 13% (page 30, footnote (e)). Although those polyurethane elastomers exhibit an overall desirable combination of properties (page 19, line 14 to page 20, line 2), their mouldability as well as their dimensional change upon absorption of water cannot be regarded as entirely satisfactory.

In the light of this prior art teaching the problem underlying the application in suit may thus be seen in

providing polyurethane elastomers which have a shorter moulding time and an improved dimensional stability upon absorption of water:

According to the application in suit this problem is solved, according to both present Claims 1 and 9, by using a polyoxyalkylene polyol mixture wherein (i) the polyol compounds must meet specific requirements regarding the hydroxyl value as well as the amount and the position of the oxyethylene units, and (ii) the unsaturated monool content of the triol is limited to a certain maximum.

4. As it appears from point II above, the rejection occurred on the ground that the Appellant had not provided evidence that the two parts of the above-defined technical problem were effectively solved.

4.1 In accordance with one of the two conclusions of the oral proceedings of 16 August 1988 (cf. Minutes issued on 25 July 1988, Form 2009.2 and point 4), the Appellant submitted a comparative test designated "Example 13" on 6 February 1989, wherein Comparative Example 3 of Table 1 of the application in suit was repeated with a polyetherpolyol similar to that defined under (e) on page 30 of document (2). That polyol, called "polyol X", was identified as a polyoxypropylene oxyethylene polyol, i.e. a triol obtained by adding propylene oxide to glycerol, and then adding ethylene oxide thereto, with the following properties:

- hydroxyl number: 3
- oxyethylene group content (% by weight): 13
- unsaturated value (meq/g): 0.11
- hydroxyl value: 28.

This compound can be regarded as fairly close to polyol A used in Example 3 of the application in suit (compare Table 1 of page 33 and the Table on page 30), thus appropriate for the comparison requested by the Examining Division. The comparative data show that, whilst the moulding time of Example 13, using "Polyol X" is the same as in the case of Comparative Example 3 of the application in suit, viz. 120 seconds, the dimensional change upon absorption of water is significantly higher in the case of Example 13, which is evidence of inferior properties.

4.2 In the second comparative test, which was filed as Example 14 in annex to the Statement of Grounds of Appeal, there has been used a mixture of polyol X and polyol Y, the latter being characterised as follows:

- hydroxyl number: 2
- oxyethylene group content (% by weight): 20
- unsaturated value (meq/g): 0.10
- hydroxyl value: 28.

From the experimental data it clearly appears that the use of a mixture of polyols X and Y leads to both longer moulding time (140 s) and higher dimensional change upon absorption of water (0.99%), compared to use of polyol X alone (120 s and 0.92%). These results, which are all based on the simultaneous use of 17 parts by weight of ethylene glycol, provide thus a reliable counter-evidence to the arguments raised in point 3.2.2 of the decision under appeal.

By contrast, a comparison between Comparative Example 3 and Example 4 in Table 1 of the application in suit reveals the opposite trend with regard to the moulding time. Whilst in the former Example the trifunctional

polyol A is used alone, in the latter there is used a mixture of said polyol A with the difunctional polyol G in a weight ratio of 76:24 (see Tables on pages 30 and 31). The data of Table 1 show that use of such a mixture of polyols leads to a much shorter moulding time (45 s) than use of the triol alone (120 s), which finding must be regarded as unexpected.

4.3 A further point not considered hitherto concerns the two physical properties mentioned last in Table 1 of the application in suit, namely tensile strength and elongation. The figures in that table when compared with those in Examples 13 and 14 demonstrate that the use of polyols (Examples 2, 3 and 13) and polyol combinations (Examples 1 and 14) outside the scope of Claim 1 lead, on an average, to both lower tensile strength (190 kg/cm²) and elongation (187%) than polyol mixtures according to Reference Examples 4 to 11 (221 kg/cm² respectively 204%). For that comparison the results of Reference Example 12 have not been taken into account, since two different values are given for each property, one in the direction parallel to the flow of the liquid, and the other in the direction perpendicular to the flow of the liquid (see footnote *b).

Whether one considers these improved mechanical properties as further beneficial technical effects, or incorporates them into the definition of the technical problem to solve, which would then be more ambitious, is irrelevant for the purpose of the present decision. In the Board's view, they provide further evidence that the combination of features (i) and (ii) as defined in point 3 above is crucial in order to achieve simultaneously a shorter moulding time and a lower dimensional change upon absorption of water.

- 4.4 In conclusion, the comparative data provided by the Appellant make it credible that the above-defined technical problem is effectively solved, thus that the subject-matter of Claim 1, whose novelty is not under dispute, involves an inventive step.
5. Claim 1 being allowable, the same applies to dependent Claims 2 to 8, which are directed to preferred embodiments of the subject-matter of Claim 1 and whose inventiveness is supported by that of the main claim.
6. Referring to points III/4 and II/5 of the decision under appeal, the Board is satisfied, on the basis of the experimental results discussed above, that the hypothesis of said point II/4 is no longer tenable; in other words, that there are no "potential Article 84 problems" left. Nor is there, in the Board's view, any problem concerning lack of unity between Claims 1 to 8 on the one hand and Claims 9 to 18 on the other hand; for, both formally independent proposals aim at solving the same problem (see point 3, paragraph 2 of present decision), following the same principle of solution (see above point 3, paragraph 3).

Still unanswered, however, is the problem alluded to in the last sentence of the decision under appeal, because convincing experimental evidence concerning the effect achieved by the proposal of Claim 9 (and the appendant Claims 10 to 18) is so far missing. The Board therefore considers it appropriate to remit the case to the Examining Division for examination of the merits of Claims 9 to 18.

Order

For these reasons, it is decided that:

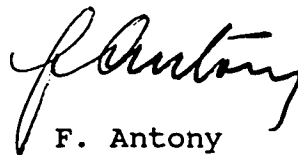
1. The decision under appeal is set aside.
2. The case is remitted to the Examining Division for further prosecution.

The Registrar:



E. Gorgmaier

The Chairman:



F. Antony