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D E C I S I O N
of 21 October 1996

Case Number: T 0078/93 - 3.3.3

Application Number: 82107817.7

Publication Number: 0080031

IPC: C08G 18/65

Language of the proceedings: EN

Title of invention:
Polyurethanes useful as engineering plastics

Patentee:
THE DOW CHEMICAL COMPANY

Opponent:
Bayer AG, Leverkusen Konzernverwaltung RP Patente Konzern
Montedipe S.p.A.

Headword:
-

Relevant legal provisions:
EPC Art. 56, 84, 114(1), 114(2), 123(2), 123(3)

Keyword:
"Inventive step (yes); improvement contrary to expected trend"

Decisions cited:
-

Catchword:
-

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Case Number: T 0078/93 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 21 October 1996

Other party:
(Opponent 01)

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Decision under appeal:

Interlocutory decision of the Opposition Division of the European Patent Office dated 12 May 1992 and issued in writing on 24 November 1992 concerning maintenance of European patent No. 0 080 031 in amended form.

Composition of the Board:

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

Summary of Facts and Submissions

I. The mention of the grant of European patent No. 0 080 031 in respect of European patent application No. 82 107 817.7, filed on 25 August 1982 and claiming a US priority of 14 October 1981 (US 311198) was announced on 7 January 1988 (Bulletin 88/01).

II. Two Notices of Opposition were filed, both on 6 October 1988, on the grounds of lack of novelty (Opponent II) and inventive step (Opponents I and II). The oppositions were supported inter alia by the following documents:

D3: US-A-3 356 650 and

D4: Journal of Applied Polymer Science, Vol. 24, pages 2041 to 2050 (1979).

An objection of lack of sufficiency was also raised by Opponent II in a later submission.

III. By an interlocutory decision which was given at the end of oral proceedings held on 12 May 1992 and issued in writing on 24 November 1992, the Opposition Division found that the patent could be maintained in amended form on the basis of the sets of Claims 1 to 5 for all Contracting states (except AT); and Claims 1 to 5 for AT, respectively, filed on 6 April 1992.

Claim 1 of the set of claims for all Contracting states (except AT) reads as follows:

"A thermoplastic polyurethane characterized by high impact resistance at ambient conditions of at least 5.44 mKg/m (ASTM D 256-56) of notch as measured by the notched Izod test, high flexural modulus of at least

1 034.5 Mpa (ASTM-D 790) and a heat deflection temperature of at least 50°C at 1820 kPa (ASTM D 648-56) which comprises the product of a one-shot reaction of

(a) an organic polyisocyanate comprising a diisocyanate selected from 4,4'-methylenebis(phenyl isocyanate) and mixtures of 4,4'-methylenebis(phenyl isocyanate) with the corresponding 2,4'-isomer in an amount up to 70 percent by weight based on the total diisocyanate mixture, or an organic polyisocyanate corresponding to said diisocyanate wherein said 4,4'-methylenebis(phenyl isocyanate) or the mixture thereof with the 2,4'-isomer is in the form of a liquid at ambient temperature and which has been obtained by (a) converting a minor proportion thereof to the corresponding carbodiimide or (b) reacting the 4,4'-methylenebis(phenyl isocyanate) or its admixture with the 2,4'-isomer with a minor amount of at least one aliphatic diol,

(b) an isocyanate-reactive material having an average functionality of at least 1.9 and not exceeding 4, a glass transition temperature (Tg) of less than 20°C, and a molecular weight in the range of 500 to 20,000 and

(c) at least one aliphatic diol chain extender having a molecular weight from 50 to 400;

said polyurethane being further characterized in that the proportion by weight of said isocyanate-reactive material (b) in said polyurethane is from 4 to 15 percent and the overall ratio of isocyanate groups to active hydrogen groups in said reactants employed to prepare said polyurethane is in the range of 0.95:1 to 1.05:1."

Claims 2 to 5 are directed to elaborations of the polyurethane of Claim 1.

Claim 1 of the set of claims for AT reads as follows:

"A process for the preparation of a thermoplastic polyurethane characterized by high impact resistance at ambient conditions of at least 5.44 mKg/m (ASTM-D 256-56) of notch as measured by the notched Izod test, high flexural modulus of at least 1 034.5 Mpa (ASTM-D 790) and a heat deflection temperature of at least 50°C at 1820 kPa (ASTM-D 648-56) which process comprises reacting in a one-shot reaction

(a) [wording identical with sub-paragraph (a) of Claim 1 for all Contracting states (except AT)],

(b) [wording identical with sub-paragraph (b) of Claim 1 for all Contracting states (except AT)] and

(c) [wording identical with sub-paragraph (c) of Claim 1 for all Contracting states (except AT)];

said process being further characterized in that the said isocyanate-reactive material (b) is employed in a proportion by weight of from 4 to 15 percent of the polyurethane weight and the overall ratio of isocyanate groups to active hydrogen groups in which said reactants are employed is in the range of 0.95:1 to 1.05:1."

Claims 2 to 5 of this set are directed to elaborations of the process of Claim 1.

The decision held that the objection of insufficiency had not originally been a ground of opposition and in any case had not been substantiated. Furthermore, the claimed subject-matter was novel. Finally, as to inventive step, whilst D3 had been suggested as being the closest state of the art, no convincing arguments had been provided that the problem it solved was the same as in the patent in suit. Hindsight had been used (i) to regard document D3 as actually relevant with regard to Article 56 EPC, (ii) to modify Example 8 thereof by correcting the diisocyanate used and (iii) to suggest a combination with document D4. Indeed, none of the cited documents referred specifically to the same problem to be solved, namely the production of polyurethane resins having improved structural properties, which excluded any combination of these documents. Consequently, the claimed subject-matter involved an inventive step.

IV. On 18 January 1993 a Notice of Appeal against the above decision was filed by Opponent II (the Appellant), together with payment of the prescribed fee.

In the Statement of Grounds of Appeal, filed on 26 March 1993, the Appellant argued in essence as follows against both sets of claims:

(a) The requirements in Claim 1 for the polyurethane to exhibit (1) an impact resistance at ambient conditions of at least 5.44 mkg/m of notch, (2) a flexural modulus of at least 1034.5 Mpa and (3) a heat deflection temperature of at least 50°C at 1820 Kpa were either fulfilled automatically and therefore superfluous, or further conditions had to be observed in which case the disclosure was insufficient.

(b) A different approach, starting from D4 as closest prior art was justified. The only true feature distinguishing the claimed subject-matter from the process of D4 was that the proportion by weight of the isocyanate-reactive material in the polyurethane was 4 to 15 per cent. However, it was generally stated that an increase in the portion of hard segments resulted in an improvement of various properties, inter alia of the flexural modulus (page 2045, paragraph 3). Whilst no data about Izod values and the heat deflection temperature were given, an improvement of these values would automatically occur when the measure for improving the flexural modulus was taken. They were thus "bonus effects" of an obvious operation.

(c) A number of batches in each of Tables 1, 2 and 3 no longer fell under the claims of the patent in suit.

V. The Respondent (Patentee) on the other hand argued in essence that:

(a) The values of good impact resistance, high flexural modulus and heat deflection temperature shown by the product resulted from the preparation conditions set out in the claim. They were mentioned in order to reflect the outstanding quality of the material.

(b) In the investigation carried out according to D4 to determine the influence of the hard segment concentration on the physical-mechanical properties of thermoplastic polyurethane, its concentration had been varied from 20 to 80% in exactly 10% increments. Whilst the hardness, tensile strength, tear strength and flexural modulus increased when the hard segment concentration increased, nevertheless the ultimate elongation decreased especially above 60 wt% hard segment, which resulted in a change of the nature of the urethane from a tough elastomeric material to a more brittle, high modulus plastic. Consequently, the skilled person would conclude that the increase in hardness, tensile strength, tear strength and flexural modulus brought about by increasing the hard segment proportion would result in brittleness and lowered impact strength.

(c) The statement that an improvement in Izod values and the heat deflection temperature would concurrently and automatically occur when the measure for improving the flexural modulus was taken was incorrect, since typically in segmented polyurethanes the Izod impact toughness property decreased as the soft segment proportion declined in the formulation.

(d) There was no hint in D3 to select the process features of the patent in suit in the knowledge that the properties of a product as defined in the patent in suit would be achieved.

Consequently, it was not obvious for the skilled person to increase the hard segment concentration above 80%.

VI. Oral proceedings were appointed by the Board, in accordance with the auxiliary requests of both parties, for 24 October 1996.

VII. With a letter filed on 5 August 1996, Opponent I, which had not taken any active part in the appeal proceedings but, as a party to these proceedings, had been duly summoned, informed the EPO that it would not attend the hearing and requested a decision on the basis of the written submissions.

VIII. With a letter filed on 6 September 1996, the Appellant withdrew its request for oral proceedings and requested instead a decision according to the current state of the file.

IX. The Board accordingly informed the parties, in a communication issued on 14 October 1996, that the oral proceedings had been cancelled.

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

The Respondent requested that the appeal be dismissed, or, in the alternative, oral proceedings.

Reasons for the Decision

1. The appeal is admissible.

2. *Amendments*

No objection has been raised under Article 123(2) or (3) EPC in the appeal to the amended text of the patent in suit forming the basis of the interlocutory decision of the Opposition Division and corresponding to the present request of the Respondent. Nor does the Board see any reason to raise such an objection itself . (Article 114(1) EPC).

Consequently, the amendments are held to be allowable under Articles 123(2) and (3) EPC.

3. *Sufficiency; conciseness; clarity*

3.1 The objection of the Appellant concerning the mention in Claim 1 of threshold requirements of the relevant mechanical and thermal parameters (1), (2) and (3) of the thermoplastic polyurethane resins amounts to an objection of lack of sufficiency of disclosure, or, in the alternative, of lack of conciseness (Section IV (a), above).

In relation to the issue of lack of sufficiency, this ground had already been excluded from the proceedings, as belated and lacking in substantiation, in the decision under appeal. The Board sees no reason to take a different view of the latest objection, which is in any case unsupported by any concrete evidence whatsoever. Accordingly, the first aspect of the objection is excluded from the proceedings under Article 114(2) EPC.

3.2 The alternative aspect of the objection, that the parameter features are superfluous, is also unjustified, since the combination of properties represented by the parameters corresponds, in the Board's view, to a reasonable summary of the essential character of the polyurethanes forming the subject-matter of the patent in suit. It cannot, therefore, be construed as resulting in an objectionable lack of conciseness (Article 84 EPC).

3.3 The objection of the Appellant, that certain of the examples fell outside the scope of the claims, was not elaborated further. Nor has it been disputed that the wording of the claims was perfectly clear (Article 84 EPC).

The Board sees no reason, therefore, to pursue this matter *ex officio*, particularly in view of the request by the Appellant for a decision based on the current state of the file and nothing else.

4. *The technical problem*

The patent in suit relates to thermoplastic polyurethane resins having improved structural strength properties, inter alia a high flexural modulus of at least 1 034.5 MPa and a high heat deflection temperature, and their preparation. The latter is effected by reacting, in a one-shot process:

(a) a specific methylenebis(phenyl isocyanate);

(b) an isocyanate-reactive material having an average functionality of at least 1.9 and not exceeding 4, a glass transition temperature (Tg) of less than 20°C, and a molecular weight in the range of 500 to 20,000, preferably a polyether polyol; and

(c) at least one aliphatic diol chain extender having a molecular weight from 50 to 400,

the overall ratio of isocyanate groups to active hydrogen groups in the reactants employed being in the range of 0.95:1 to 1.05:1 (Claims 1, 2).

According to the patent in suit, the relative proportions of the polymeric polyol to the low molecular weight chain extender greatly influences the properties of the polyurethane which is obtained, the polymer chain units derived from the extender being relatively rigid ("hard segments"), and the units derived from the polymeric polyols exhibiting low moduli of elasticity ("soft segments"). In the case, for example, of a relatively linear polyurethane, prepared from a diisocyanate, a polymeric glycol and a chain extender, increasing the proportion of extender to polymeric diol gives a progressively more rigid polyurethane and, beyond a certain point, the polymer becomes relatively brittle and shows very low impact resistance (patent in suit, page 2, line 57 to page 3, line 2).

4.1 This state of the art, which is considered to represent the closest state of the art, is illustrated by the cited document D4.

According to D4, physical mechanical, thermal and dynamic-mechanical properies, such as hardness, tensile strength, flexural modulus, heat sag and ultimate elongation, of a series of polyether-based thermoplastic polyurethane resins were studied as a function of hard segment content. The concentration of the hard segments, which were based on 4, 4'-diphenylmethane diisocyanate (MDI) and 1, 4-butanediol (BDO), was varied from 20 to 80 wt.% in 10% intervals. A 2 000 molecular weight poly(oxypropylene-oxyethylene)

diol was used as the soft segment (page 2041, last paragraph in combination with page 2044, Table I).

The polyether diol, which contained 30.4 wt% of oxyethylene as an end block on polypropylene, contained 83% primary hydroxyls and had a functionality of 1.964 (page 2042, first paragraph).

The thermoplastic polyurethane samples were prepared by a hand casting technique, in which the MDI was added, in 4% excess, to a mixture of polyether diol, BDO and catalyst (0.02%) which had been stirred under vacuum for 30 min. After further stirring under vacuum for 30 sec., the reacting mixture was poured on to a moulding surface and allowed to cure in an oven at 100°C, then demolded and allowed to "age" for a week at ambient temperatures prior to testing (page 2042, second paragraph).

The results of the tests show that, as the hard-segment concentration increases, the properties of hardness, tensile strength and flexural modulus also increase. The ultimate elongation decreases, however, especially above 60 wt% "hard segment". A graph showing the dependence of tensile strength on the hard segment concentration shows an inflexion point at approximately 60% "hard segment" (page 2042, Figure 1), which could represent a marked change in domain morphology related either to inversion of the soft and hard phases or mixing thereof. This marks a change in the nature of the polymer from a tough, elastomeric material to a more brittle, high modulus plastic (pages 2045, "Polymer Physical-Mechanical Properties", and 2049, "Conclusions").

4.2 Compared with this state of the art, the technical problem is to be seen in the search for a thermoplastic polyurethane resin having a more favourable balance of

physical-mechanical and dynamic-mechanical properties, in particular avoiding brittleness at high tensile strengths.

The solution proposed according to Claim 1 of the patent in suit is to increase the hard segment content of the thermoplastic polyurethane relative to the soft segment content beyond the maximum (80%) disclosed in D4, so that the proportion of soft segment material (isocyanate reactive polymeric polyol) is from 4 to 15 wt%, the resulting polyurethane exhibiting, at ambient conditions, an impact resistance of at least 5.44 mKg/m of notch as measured by the notched Izod test (ASTM D 256-56), a flexural modulus of at least 1 034 MPa (ASTM-D 790) and a heat deflection temperature of at least 50°C at 1820 kPa (ASTM D 648-56).

4.3 It can be seen from the results of the large number of examples in the patent in suit, that the reported values of the relevant mechanical and thermal parameters all fulfil, and, in many cases, greatly exceed, the thresholds required by the solution of the stated problem.

4.4 Thus, it is credible that the claimed measures provide an effective solution of the stated problem.

5. *Novelty*

It was not disputed that the subject-matter claimed was novel. On the contrary, the Appellant explicitly acknowledged, in relation to D4, that the feature of the proportion by weight of the isocyanate-reactive material in the polyurethane being 4 to 15 percent was able formally to "restrict" (i.e. distinguish) the claimed subject-matter over the known process (Statement of Grounds of Appeal, page 4, reference to "feature 9" and reference to this feature on page 2).

The Board sees no reason to take a different view of the matter and hence finds the claimed subject-matter to be novel.

6. *Inventive step*

To assess the question of inventive step it is necessary to consider whether the skilled person, starting from D4, would have expected that a higher Izod impact strength without loss of physical-mechanical properties such as tensile strength and flexural modulus, would be obtained by increasing the "hard segment" content of the polyurethane beyond the limit of 80% disclosed in D4, i.e. using a level of "soft segment" in the range 4 to 15% of the polyurethane.

6.1 Whilst it is true that D4 deals in general terms with physical-mechanical, thermal and dynamic mechanical properties of thermoplastic polyurethane resins, it does not mention impact strength specifically at all, let alone the particular measure represented by the Izod notched impact test (ASTM D 256-56), which is the crucial area in which the improvement provided by the claimed subject-matter is obtained.

Consequently, the person skilled in the art looking for a relative improvement in this parameter would not find any direct pointer to the solution in the disclosure of D4.

6.2 If, however, the skilled person were nevertheless to consider the teaching of D4, for instance to see if its teaching concerning other physical-mechanical and and dynamic-mechanical properties of thermoplastic polyurethane resins could give an indirect insight into their impact behaviour, it is evident that the most relevant indication in this connection is the reference

to the brittleness of the thermoplastic polyurethane resins. This is stated to occur at 60-65% of "hard segment", which results in the nature of the urethane changing from a tough, elastic material to a more brittle, high-modulus plastic (D4, page 2049, "Conclusions").

Brittleness is, however, itself an indication of lack of impact resistance, and, being associated with an increased proportion of "hard segment", this characteristic would be expected to become more marked, rather than less, as the proportion of "hard segment" was further increased beyond the maximum of 80% reported in D4.

Consequently, the skilled person would have concluded that, with a further increase of the "hard segment" content of the polyurethane resin, the Izod notched impact strength, if measured, would show a corresponding decrease.

6.3 The argument of the Appellant, that the Izod impact strength values would inevitably increase together with the flexural modulus, is beside the point. The relevant question at issue is not what would in fact have been inevitable, but what would have been expected to be inevitable. For the reasons given above, the skilled reader of D4 would not have expected the impact strength values to increase in the manner argued, but, on the contrary, would have expected them to decrease.

In other words, the solution of the stated problem does not arise in an obvious way from the disclosure of D4.

6.4 As regards the other documents in the proceedings, especially D3, no arguments have been developed in the submissions of the Appellant, beyond those already considered in the decision under appeal, in particular those regarding the relevance of Example 8 and the choice of MDI as the isocyanate, from which the Board sees no reason to differ.

6.5 Consequently, the subject-matter of Claim 1 of both sets of claims, and that of the claims dependent thereon involves an inventive step in the light of the documents cited, whether taken individually or in combination.

Order

for these reasons it is decided that:

The appeal is dismissed.

The Registrar:

E. Görgmaier

The Chairman:

C. Gérardin