

A		B		C	X
---	--	---	--	---	---

File Number: T 640/89 - 3.3.3

Application No.: 84 105 642.7

Publication No.: 0 131 714

Title of invention: Polyurethane plastics with improved impact resistance

Classification: C08L 75/04

D E C I S I O N
of 5 October 1992

Applicant: THE DOW CHEMICAL COMPANY

Headword:

EPC Article 56

Keyword: "Inventive step - yes after amendment"



Case Number : T 640/89 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 5 October 1992

Appellant : THE DOW CHEMICAL COMPANY
2030 Dow Center
Abbott Road
PO BOX 1967
Midland
Michigan 48640-1967 (US)

Representative : Henkel, Feiler, Hänzel & Partner
Möhlstrasse 37
W - 8000 München 80 (DE)

Decision under appeal : Decision of Examining Division of the European
Patent Office dated 10 May 1989 refusing European
patent application No. 84 105 642.7 pursuant to
Article 97(1) EPC.

Composition of the Board :

Chairman : F. Antony
Members : C. Gérardin
M. Aúz Castro

Summary of Facts and Submissions

I. European patent application No. 84 105 642.7 filed on 17 May 1984, claiming priority of 18 July 1983 from an earlier application in the United States and published under the publication No. 0 131 714, was refused by a decision of the Examining Division dated 10 May 1989. That decision was based on a set of eight claims for the Contracting States BE, CH, DE, FR, GB, IT, LI, NL and SE, of which Claim 1 filed on 2 January 1989 reads as follows:

"A polymer blend characterized by high impact resistance, high flexural modulus and a heat deflection temperature of at least 50°C at 264 psi which comprises a blend of:

- (a) from 3 to 30 parts by weight, per 100 parts by weight of said blend, of a polymeric impact modifier which is incompatible with the major component of the blend; and, as the balance of said blend,
- (b) a clear polyurethane glass which is the product of reaction of:
 - (i) an organic polyisocyanate and
 - (ii) at least one chain extender having a functionality from 2 to 3 and a molecular weight from about 50 to about 400;

the proportions of said components (i) and (ii) being such that the overall ratio of isocyanate groups to active hydrogen groups is in the range of 0.95:1 to about 1.05:1."

The dependent Claims 2 to 8, which are directed to preferred embodiments of the main claim, are the original claims.

For the Contracting State AT there was a similar set of eight claims wherein, however, the main claim filed on 2 January 1989 and the seven dependent claims filed originally were drafted as method claims.

- II. The ground for that decision was lack of inventive step with regard to the teaching of EP-A1-12 416 (document (2)). That citation described blends with excellent mechanical properties consisting of 3 to 25 weight percent of a polymer made from ethylenically unsaturated monomers and 97 to 75 weight percent of a thermoplastic polyurethane made from a polyisocyanate, a long chain polyol and a chain extender. A high ratio (chain extender): (long chain polyol) was said to improve the impact resistance of these blends. The total omission of the long chain polyol in order to further improve the impact resistance would have been considered by the skilled man; it followed that the claimed subject-matter did not involve an inventive step.
- III. On 22 June 1989 a Notice of Appeal was lodged against that decision with payment of the prescribed fee.
- (i) In the Statement of Grounds of Appeal filed on 8 September 1989 the Appellant (Applicant) first presented arguments in favour of the patentability of the claims rejected by the Examining Division which were maintained unamended as main request. Although a high ratio (chain extender): (long chain polyol) was preferred to improve the impact resistance of the blends, the long chain polyol remained an essential component of the thermoplastic polyurethane according to document (2); moreover, nothing was said about the

possible effects which might result from simply omitting that component.

- (ii) Simultaneously, an alternative set of six claims to be considered as an auxiliary request was filed; in the main claim thereof component (a) of the blend was defined as follows:

"from 3 to 30 parts by weight, per 100 parts by weight of said blend, of a polymeric impact modifier which is incompatible with the major component of the blend and is a multiphase composite interpolymer based on a polyacrylate".

The Appellant referred to the comparative experiments submitted on 2 January 1989 which demonstrated that this specific multiphase composite interpolymer conferred particularly good impact strength to the blends.

- IV. The Appellant requested that the decision under appeal be set aside and that a patent be granted on the basis of Claim 1 filed on 2 January 1989 and Claims 2 to 8 filed originally as main request, or on the basis of Claims 1 to 6 filed on 8 September 1989 as auxiliary request.

Reasons for the Decision

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

Main request

2. The current wording of the claims does not give rise to any objections under Article 123(2) EPC.

Claim 1 differs basically from the main claim originally filed by the deletion of component (iii) in the preparation of the polyurethane glass (b); since that component was optional, the claimed subject-matter simply corresponds to a specific embodiment originally disclosed which does not extend beyond the content of the application as filed.

As to the dependent Claims 2 to 8, they have been maintained unamended, thus as originally filed.

3. The application in suit concerns polyurethane plastics with improved impact resistance. Such polyurethane compositions are described in document (2). More specifically, these prior art compositions consist of (A) 75 to 97 weight percent of a thermoplastic polyurethane and (B) 25 to 3 weight percent of a graft polymer obtained by polymerising (a) 5 to 35 weight percent of one or several monomer(s) on (b) 95 to 65 weight percent of a rubbery polymer having a glass transition temperature lower than -30°C , the total content of styrene, α -methylstyrene and acrylonitrile in component (B) being lower than 50% (Claim 1). The polyurethane component (A) is described as being the reaction product of a diisocyanate, a low molecular weight chain extender and a long chain polyol having a molecular weight between 400 and 10 000, the amounts of these reactants being such that the overall ratio of isocyanate groups to active hydrogen atoms is preferably between 0.95 and 1.05 (page 6, line 27 to page 7, line 7). These blends of flexibilised polyurethane and graft polymer exhibit a desirable combination of properties over a wide range of temperatures, in particular excellent impact resistance properties (page 3, line 26 to page 4, line 8; Examples 2a, 2b and 3). This applies in particular to the

blends prepared from hard thermoplastic polyurethanes wherein the molar ratio (chain extender): (flexibilising polyol) is higher than 4:1, preferably higher than 6:1.

In the light of this prior art teaching and the data submitted, the technical problem underlying the application in suit can thus be seen in providing further polyurethane compositions having the same desirable combination of properties, in particular essentially the same impact resistance properties.

According to the application in suit this problem is solved by blends of a polyurethane prepared from a diisocyanate and a low molecular weight chain extender, and of a conventional impact modifier. In view of the experimental results in the application in suit, which show that blends based on non-flexibilised polyurethanes (Table II) and blends based on flexibilised polyurethanes (Tables III to VIII) exhibit comparable properties, in particular in terms of impact resistance, flexural strength, flexural modulus and heat deflection temperature, the Board is satisfied that the above-defined technical problem has been effectively solved.

4. As acknowledged by the Examining Division, this solution is not disclosed in document (2); since the objection under Article 54(3) EPC with regard to the teaching of EP-A1-156 941 (document (1)) raised initially during examination procedure was no longer maintained in the decision under appeal, it is not necessary to consider the issue of novelty in further detail.
5. It still remains to be decided whether the claimed subject-matter involves an inventive step with regard to document (2).

Although this citation is basically concerned with the improvement of the properties of flexibilised polyurethanes, it specifies that the same advantages would be obtained for hard thermoplastic polyurethanes, i.e. polyurethanes having a molar ratio (chain extender): (long chain polyol) higher than 4, preferably higher than 6 (page 9, line 23 to page 10, line 2). This definition clearly also encompasses polyurethanes obtained from mixtures of chain extender and long chain polyol used in much higher molar ratios.

In the Board's view, however, the question is not so much which upper limit should be attributed to that ratio because even a very high ratio, say 100:1, would still correspond to a polyurethane outside the scope of Claim 1, but what kind of polyurethanes could be qualified as extremely hard, in particular have a Shore Hardness above 50, preferably above 60. On the basis of his common general knowledge the skilled person would certainly expect polyurethanes which do not contain any soft segments to belong to that category; this skilled person would naturally try in the framework of a routine test to extend to such non-flexibilised polyurethanes the advantages which can be obtained for hard polyurethanes in general, as taught in document (2). From that point of view, the total omission of the long chain polyol in the preparation of polyurethanes to be blended with graft polymers must be regarded as nothing but normal experimentation; as it appears from the experimental data in the application in suit, the resulting blends do not exhibit other properties than those disclosed in document (2). For these various reasons the solution according to Claim 1 of the main request does not involve any inventive step.

6. Claim 1 not being allowable, the same applies to the dependent Claims 2 to 8 which are directed to preferred embodiments of the main claim and thus fall with it.

Auxiliary request

7. As noted above in point III(ii), the wording of Claim 1 according to the auxiliary request differs from that of Claim 1 according to the main request by the additional feature that the polymeric impact modifier (a) is characterised as being "a multiphase composite interpolymer based on a polyacrylate".

This structural feature, however, is not disclosed as such in the original documents and the list of impact modifiers in the description only mentions vinyl chloride-ethylene-vinyl acetate graft polymers beside homopolymers, copolymers and terpolymers (page 5, line 33 to page 6, line 14); among the specific impact modifiers used in the examples, the polymeric additive KM-330 is identified as being a poly(butylacrylate). To justify the introduction of the general concept of multiphase composite interpolymer in Claim 1, the Appellant has referred to US-A-4 404 161, wherein it is specified (column 9, lines 22 to 26) that KM-330 is a multiphase composite interpolymer available from Rohm and Haas based on n-butyl acrylate, 1,3-butylene diacrylate, diallyl maleate and methyl methacrylate produced according to Example 26 of US-A-4 096 202. The latter document indicates the exact composition of that impact modifier and identifies impact modifiers based on acrylic rubbers as being multiphase composite interpolymers (column 1, lines 10 to 27; column 2, line 12 to column 3, line 46). Since rubbery impact modifiers are said to be the preferred additives in the application in suit (page 6, lines 2 to 4), the Board takes the view that this structural definition provides

adequate support for the general concept of "multiphase composite interpolymer based on a polyacrylate" in the main claim.

Further, the dependent Claims 2 to 4 are identical with the version of these claims as originally filed. Claim 5 (after correction of an obvious error, viz. insertion of "(a)" after "component" in its second line) corresponds to original Claim 5 after deletion of the feature that component (a) is a particulate rubber incompatible with component (b); since that feature is now implicitly contained in Claim 1, the deletion thereof in a dependent claim is not objectionable. As to Claim 6, it additionally contains the structural feature "multiphase composite interpolymer" which is acceptable for the reasons given above.

It follows, thus, that no objection arises having regard to Article 123(2) EPC.

8. In view of the presence in the main claim of the additional structural feature discussed above, there are even less reasons to consider the issue of novelty than in the case of the main request.
9. It still remains to be decided whether the claimed subject-matter involves an inventive step with regard to document (2).

For that purpose the main point to examine is the compositional definition and the structure of the graft polymers described in that citation. The first phase is said to be elastomeric with a glass transition temperature lower than -30°C , preferably lower than -50°C ; it is advantageously a diene rubber containing up to 30 weight percent of styrene and/or acrylonitrile or an acrylic

rubber, especially a copolymer of butadiene and an acrylate ester (page 5, lines 16 to 24). For the preparation of the second phase the grafting monomers are basically selected among styrene, α -methylstyrene and acrylonitrile; in order to achieve specific properties, however, these monomers can be replaced partially or totally by methacrylonitrile, acrylates, methacrylates or by halogenated derivatives of styrene and α -methylstyrene (page 6, lines 8 to 16). In Example 1c, which corresponds to a typical embodiment, 5.6 parts of acrylonitrile and 14.4 parts of styrene are grafted onto 137 parts of a polybutadiene latex having a glass transition temperature of -80°C ; the latter feature is essential to confer good mechanical properties to the blends with thermoplastic polyurethanes. As it appears from a comparison with Example 1b, wherein a regular ABS polymer had been used, a too high glass transition temperature of the rubbery component tends to affect the mechanical properties of the blend.

In the Board's view, this teaching would not provide an incentive to the skilled man to use as component (a) in the blends a multiphase composite interpolymer based on a polyacrylate with a core having an unspecified glass transition temperature. In view of the experimental data submitted by the Appellant on 2 January 1989, which show that blends containing 10 weight percent of the multiphase composite interpolymer Acryloid KM-330 (Blends 1 and 2) have better impact resistance than blends containing the same amount of a conventional polymer additive, such as the methacrylate/butadiene/styrene interpolymer Acryloid KM-611 (Blends 3 and 4) and a regular ABS resin (Blends 5 and 6), an inventive step can be acknowledged.

10. Claim 1 being allowable, the same applies to the dependent Claims 2 to 6, which are directed to preferred embodiments of the subject-matter of Claim 1 and whose inventiveness is supported by that of the main claim.

11. Although the claims according to the auxiliary request meet the requirements of Article 56 EPC, a patent cannot yet be granted, since it remains to adapt the description in order to bring it in line with the claims as amended, in particular with the more specific definition of the polymeric impact modifier. Further, it is noted that the Examining Division's invitation that "S.I. units should be used throughout the application (Rule 35(12) EPC)" is still outstanding (communication of 8 February 1988, point 3); this applies in the first line to Claim 1 as it stands as well as to the description. In addition, it might be appropriate to have a corresponding set of claims directed to a method for preparing the same polymer blend, since Austria is a designated Contracting State. For these various reasons the case is remitted to the first instance.

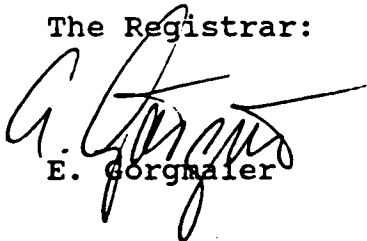
Order

For these reasons, it is decided that:

1. The decision under appeal is set aside.

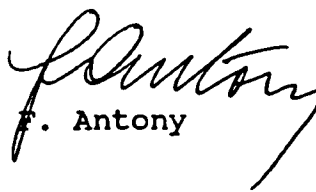
2. The case is remitted to the Examining Division for further prosecution on the basis of Claims 1 to 6 filed on 8 September 1989 as auxiliary request.

The Registrar:



E. Gorgmaier

The Chairman:



F. Antony