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D E C I S I O N
of 4 April 2001

Case Number: T 0228/98 - 3.3.3

Application Number: 91904246.5

Publication Number: 0514459

IPC: C08G 18/06

Language of the proceedings: EN

Title of invention:

Thermally-reversible isocyanate polymers

Applicant:

BATTELLE MEMORIAL INSTITUTE

Opponent:

-

Headword:

-

Relevant legal provisions:

EPC Art. 84, 123(2)

Keyword:

"Claims - functional feature"

"Amendments - added subject-matter (no)"

Decisions cited:

T 0066/85

Catchword:

-



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Boards of Appeal

Chambres de recours

Case Number: T 0228/98 - 3.3.3

D E C I S I O N
of the Technical Board of Appeal 3.3.3
of 4 April 2001

Appellant: BATTELLE MEMORIAL INSTITUTE
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Decision under appeal: Decision of the Examining Division of the
European Patent Office dated 16 September 1997
and issued in writing on 23 October 1997 refusing
European patent application No. 91 904 246.5
pursuant to Article 97(1) EPC.

Composition of the Board:

Chairman: R. Young
Members: A. Däweritz
V. Di Cerbo

Summary of Facts and Submissions

I. European patent application No. 91 904 246.5, based on International application No. PCT/US91/00742, filed on 4 February 1991, claiming the priority of 5 February 1990 of an earlier application in the United States of America (475112) and published under No. WO-A-91/11476 (EP-A-0 514 459) on 8 August 1991, was refused by a decision of the Examining Division, announced orally on 16 September 1997 and issued in writing on 23 October 1997, for the reason of non-compliance of Claim 1 with the requirements of Article 123(2) EPC.

II. The decision was based on a set of 12 claims submitted with a letter dated 13 August 1997, received on 18 August 1997. Claim 1 read as follows:

"1. A stable, melt-processable, high-temperature polyurethane polymer composition, comprising:

(A) a labile-hydrogen functionality segment selected from the group consisting of

oligomeric aromatic carbonates and oligomeric aromatic esters with phenolic hydroxyl end group functionalities having labile hydrogen end groups;
and

(B) an isocyanate functionality segment selected from the group consisting of:

oligomeric aromatic, aliphatic cycloaliphatic or aralkyl polyisocyanate containing from 6 to 100 carbon atoms having isocyanate reactive end groups;

wherein said labile-hydrogen segment and said isocyanate segment are linked by a thermally-reversible, isocyanate-labile urethane bond hydrogen backbone linkage, and wherein said linkage has the characteristic of dissociating only above 150°C into said labile-hydrogen segment and said isocyanate segment as a melt with a viscosity of less than about 100,000 poise above the melt temperature and at 1 Hertz as calculated from Figures 1 to 5."

Claims 7 and 10 concerned processes for forming such compositions and Claim 8 related to a method for using these compositions.

Dependent Claims 2 to 6, 9, 11 and 12 related to preferred embodiments of the composition according to Claim 1, the method according to Claim 8 and the process according to Claim 10, respectively.

- (i) In the decision, the Examining Division considered the passage "... at 1 Hertz as calculated from Figures 1 to 5" referring to the melt of the composition claimed at temperature above the melt temperature not to be originally disclosed. The Examining Division did not accept the applicant's calculation submitted by letter on 18 October 1995, because it held that a value of 1 Hertz could not unambiguously be derived from the values of storage modulus G' and loss modulus G" taken from logarithmic scales in Figures 1 to 5, and because the calculation referred to complex viscosity rather than to dynamic viscosity as originally disclosed, which is the real part of complex viscosity.

- (ii) Additionally, the Examining Division mentioned further objections against the claims for lack of clarity under Article 84 EPC, which were *expressis verbis* referred to as not being a ground of refusal. It was thus criticised that the measuring time was not defined although the viscosity apparently depended thereon as indicated in the description. Moreover, terms such as "oligomeric", "low" or "high" melting, "high temperature", "toughening" and "stable" were deemed vague and not to define clearly the matter for which protection was sought.

III. On 12 December 1997, a Notice of Appeal against the above decision was lodged by the Appellant (Applicant). The prescribed fee was paid on the same date.

In the Statement of Grounds of Appeal, submitted on 2 March 1998, the Appellant

- (i) requested that the decision be set aside and that a patent be granted on the basis of a new set of Claims 1 to 12 which differed from the above version in that Claims 1, 7 and 10 were further amended. Thus, the oligomers in segment A were limited to a degree of polymerisation of 1 to 20 and the viscosity mentioned was defined to be "as measured by dynamic oscillatory mechanical spectrometry"; and

- (ii) argued essentially as follows:

1. It could be seen by a person skilled in the art of rheology from the original disclosure that the viscosity measurements had been

carried out at a frequency of 1 Hz, in particular from Figures 1 to 5. The calculation enclosed to prove this was said to be based on "Rheology in Polymer Processing", C. D. Han, Academic Press, New York (1976), Section on "Oscillatory Flow Measurement", pages 353 to 356. Moreover, the highest frequency possible in the Rheometrics Model RMS-605, specified at page 6 of the application, was 100 rad/s or 15.92 Hz. The polymers which disassociated into small molecules above 140°C would be below their critical molecular weight for entanglement induced non-Newtonian behaviour above the disassociation temperature.

2. The viscosity of the material was not time dependent, because the reaction between the isocyanate functional groups and the hydroxyl groups forming long chain linkages was not a time dependent degradation reaction, but it was at its equilibrium at a given temperature within the range investigated.
3. Novelty and inventive step had not been put in question by the Examining Division.

(iii) Additional data, explanations and calculation results were submitted in an Affidavit by John D. Clay, received on 25 February 1999, to support the above arguments.

IV. An annex to a summons, issued on 20 November 2000, to oral proceedings included a brief discussion of the points raised in the decision under appeal and

addressed some further points of lack of clarity. It informed the Appellant that the Board would presumably not deal with the questions of novelty and inventive step as raised by the Examining Division in previous communications only, but that it would concentrate on the wording of the claims with respect to Article 84 and 123(2) EPC objections.

V. In reply to this annex, the Appellant submitted a new set of Claims 1 to 11, which were received on 5 March 2001 and were accompanied by additional comments on the points raised in the said annex.

- (i) The new claims differed from the previous version in that in Claim 4, a passage further defining the "labile-hydrogen segment" was added: "which provides ambient or low temperature flexibility and toughness to the final polymer composition"; former Claim 7 was deleted and former Claims 8 to 12 were renumbered accordingly. In renumbered Claim 7 (former Claim 8), the wording "as an adhesive" was added after the reference to Claims 1 to 6 in order to define a functional feature of the claimed use of the composition.
- (ii) The Appellant argued that the wording of some claims which had been objected to were "more or less clearly defined in the specification" and referred to Article 69(1) EPC which offered a basis to interpret the claims.
- (iii) It gave additional frequency calculations and further information about the way the calculations had been carried out.

- VI. The oral proceedings took place on 4 April 2001.
- VII. The discussion in the oral proceedings focused mainly on the question whether the frequency of 1 Hz which had been inserted into the independent claims could be derived in a clear and unambiguous way from the application as originally filed.
- (i) Initially, the Appellant gave a short explanation for the meaning of 100 000 poise. Thus, asphalt was said to have a viscosity in the range of about 10^7 poise, syrup in the range of about 10^5 poise, water 10^{-2} poise. The intention of the wording in the claims was to define the composition as forming a free-flowing melt, as opposed to a solid, the viscosity of which would achieve a magnitude of about 10^{10} poise, or a semi-melt including solid and liquid phases.
- (ii) With respect to the passage of the independent claims which had been objected to, the Appellant argued that the values disclosed in Figures 1 to 5 gave the clear indication that 1 Hz had been used in the measurements of the viscosity ζ^* , storage and loss modules G' and G'' . The variations of the respective values measured at a frequency within a range as derived from Figures 1 to 5 would be within a normal range of error.

However, it was conceded that 1 Hz (equal to 6.28 rad/s, i.e. radians per second) was not a standard, but that values of 0.01 to 1000 rad/s could be used in such measurements. It was accepted by the technical expert of the

Appellant, who was present in the oral proceedings, that the values actually used depended e.g. on the scale available on the apparatus. He indicated that, in his experiments, he used in general 1 rad/s.

- (iii) When the Board indicated that it might not be in a position to allow the definition of the measuring conditions in the independent claims under Article 123(2) EPC and upon further discussion with respect to clarity (Article 84 EPC) of Claims 7 to 10 as submitted on 5 March 2001, the Appellant repeatedly proposed further amended claims to overcome these objections.

- (iv) The finally submitted version of the claims forming the basis for the sole request of the Appellant reads as follows (after correction of a punctuation error at the end of Claim 6):
 - "1. A stable, melt-processable, high-temperature polyurethane polymer composition, comprising:
 - (A) a labile-hydrogen functionality segment selected from the group consisting of
 - oligomeric aromatic carbonates and
 - oligomeric aromatic esters with phenolic hydroxyl end group functionalities having labile hydrogen end groups and with a degree of polymerization of 1-20 and

 - (B) an isocyanate functionality segment selected from the group consisting of:

oligomeric aromatic, aliphatic, cycloaliphatic or aralkyl polyisocyanate containing from 6 to 100 carbon atoms having isocyanate reactive end groups;

wherein said labile-hydrogen segment and said isocyanate segment are linked by a thermally-reversible, isocyanate-labile urethane bond hydrogen backbone linkage, and wherein said linkage has the characteristic of dissociating only above 150 °C into said labile-hydrogen segment and said isocyanate segment as a free flowing melt.

2. The melt-processable polymer composition of claim 1 further comprising said thermally-reversible isocyanate-labile urethane bond hydrogen cross-links between neighboring polymer chains.
3. The melt-processable polymer composition of claim 1 characterized in that said isocyanate-labile urethane bond hydrogen backbone linkage is bonded to an aryl group, an alkyl group, an aryl and an alkyl group, or only aryl groups.
4. The melt-processable polymer composition of claim 1 further comprising:
 - (C) a toughening aliphatic prepolymer backbone group having a labile-hydrogen segment which provides ambient or low-temperature flexibility and toughness to the final polymer composition characterized as a

polyol, a polycaprolactone diol, a polytetramethylene ether glycol, a polyaliphatic carbonate diol, a hydroxy-ended aliphatic polyester, or a saturated hydroxy-ended phthalic acid-based polyester.

5. The melt-processable polymer composition of claim 1 wherein said isocyanate segment is characterized as a toluene diisocyanate, a naphthalene diisocyanate, a phenyl diisocyanate, or a triisocyanate including a triisocyanate formed in situ from a diisocyanate and a triol such as trimethylolpropane.
6. The melt-processable polymer composition of claim 1 characterized in that said aromatic polyester is a bishydroquinone ester of isophthalic acid with phenolic end groups or a bis-paraacetoxy-phenyl ester of isophthalic acid with the acetate groups replaced by parahydroxybenzoate groups; and said polycarbonate oligomer is formed from Bisphenol A and phosgene and has phenolic hydroxyl end groups.
7. Use of said polymer composition of any of claims 1-6 as a hot melt adhesive.
8. A process for forming a composition according to claim 1 comprising reacting a first compound with isocyanate functionality as defined under (B) in claim 1 with a second compound with labile-hydrogen functionality as defined under (A) in

claim 1.

9. The process according to claim 8 further characterized by the step of adding, prior to the addition of said compound with isocyanate functionality, an additional labile-hydrogen compound characterized in having ionic functionality capable of forming thermally-reversible ionic bonds."

VIII. The Appellant requested that the decision under appeal be set aside and that the case be remitted to the first instance for further examination.

Reasons for the Decision

1. The appeal is admissible.
2. *Procedural matter*

As indicated in the annex to the summons and in accordance with the sole request of the Appellant, this decision deals only with the reason for the refusal in the decision under appeal and some further questions of clarity under Article 84 EPC.

3. *Article 123(2) EPC*
 - 3.1 The decision under appeal is based on the sole reason that Claim 1 did not meet the requirements of Article 123(2) EPC.
 - 3.2 The Board sees no reason to diverge from the statements of the Examining Division that (i) the viscosity of the

melt in Claim 1 is an essential feature of the subject-matter claimed and that (ii) the viscosity of polymeric compounds depends on the method and conditions of its measurement (see the decision under appeal, points 7.1 and 7.2). Aspect (i) has been confirmed by the Appellant during the oral proceedings who explained the meaning of the value of 100 000 poise (see section VII(i)).

3.3 It has not been disputed by the Appellant, that the application does not disclose *expressis verbis* a reference to a frequency of 1 Hz. Whilst it may be true that Figures 1 to 5 of the application provide an indication to a person skilled in the art of viscosity measurements that the Rheometrics Dynamic Mechanical Spectrometer RMS 605 was used at a frequency in the range of about 1 Hz, this does not amount to an implicit disclosure of 1 Hertz, for the following reasons.

3.3.1 As admitted by the Appellant, such a frequency is not a standard value, but depends e.g. on the scale of the apparatus used which may be graduated e.g. in radians per time unit or Hertz. Thus, a value of 6.28 rad/s equals 1 Hz (see formula 10 in the Affidavit by Dr Clay submitted on 25 February 1999).

3.3.2 The calculations presented have been based on certain assumptions, that e.g. the variation of eyeball readings from the graphs was "about 6% to 20% higher", and that 1 Hz was one of the standard frequencies used for viscosity tests in the school attended by Appellant's technical expert, which led him to the conclusion that this frequency had also been used in the application (Statement of Grounds of Appeal,

page 6, last paragraph).

- 3.3.3 An amendment which is allowable under Article 123(2) EPC must be directly and unambiguously be derivable from the application as originally filed. The only basis for deriving the frequency can, according to the Appellant, be found in Figures 1 to 5. As indicated above, it is evident that the precision of the measurements displayed in these figures having logarithmic scales (6 decades equal to about 127 mm; each symbol of a measured value having a size of about 2 mm) does not allow to derive values on which an exact calculation can be based. The variation of the different values read from these figures by the Appellant clearly support this view (see the Statement of Grounds of Appeal, pages 2 to 4, point 2; the table at the bottom of page 3 of the above Affidavit; sheet "Frequency calculation for reversible polyurethane patent", submitted on 5 March 2001).
- 3.3.4 It follows that a frequency of 1 Hz can neither be derived from the data provided by Figures 1 to 5 in a clear and unambiguous manner nor from any part of the application as originally filed with the precision that would be necessary (cf. Case Law, 3rd edition, 1999, chapter III. A, 1.3 and 3 to 3.3).
- 3.4 In fact, in the set of claims on which the final request of the Appellant is based, the feature of "a melt with a viscosity ... of less than about 100000 poise ..." has been replaced by the expression "a free-flowing melt" explicitly in Claim 1 or by reference to that claim. Therefore, the further observations need be focused only on Claim 1.

- 3.4.1 It has to be decided whether the original term in the independent claims can be replaced by the other definition without contravening Article 123(2) EPC.

According to the wording of present Claim 1 as well as original Claims 1 and 41 (cf. point 4.4, below), the melt is composed of "labile-hydrogen segments" and "isocyanate segments" which are the exclusive result of a dissociation of the product of components (A) and (B) at temperatures above 150°C. Hence, the composition of the melt in present Claim 1 does not differ from that in previous Claim 1. The only difference resides in the definition of the functional limiting feature which refers to a certain flowability of the melt.

- 3.4.2 Whilst the previous wording of the feature in question limited the flowability in terms of a maximum viscosity at a minimum temperature, the new definition is worded as a functional limitation requiring free-flowability of the melt resulting from the dissociation of the polyurethane polymer composition at a temperature above 150°C. The basis for this new definition of the feature can be found in the description of the application as originally filed (page 4, lines 10 to 15; page 14, lines 9/10, 14; page 15, lines 5 to 17; page 16, lines 1 to 7). It is evident that the solubility of the melt, mentioned there, need not be considered in this context.

- 3.4.3 From these passages in the original version of the description, it is evident that it is not the exact quantitative value of the upper limit of the viscosity at a minimum temperature which is the essential feature of the invention, but, to the contrary, it rather implies only a range of acceptable viscosity values to

illustrate the free flowability of the melt.

This view is supported e.g. by Figures 1 to 5 which show that an increase in the temperature results in a decrease of the viscosity, i.e. in an increase in flowability.

- 3.4.4 Thus, the purpose of the original limitation of a maximum viscosity of the melt of 100 000 poise (method of measurement not further defined) was not to provide a precise mathematical measure of viscosity but rather to express the relevant functional relationship within the claim.
- 3.4.5 The present situation is in this respect similar to that in decision T 66/85 (OJ EPO 1989, 167, in particular point 2 of the reasons) where the Board agreed to a decision of the Examining Division that Article 123(2) EPC has to be interpreted as meaning that where a feature is entirely omitted from a claim, such excision is not permissible, unless there is a basis for the broadened claim in the original application. Such a basis need not be presented in express terms but it must be sufficiently clear to a skilled person to be unambiguously recognisable as such. As pointed out above, in the present case the feature in question describing the flowability has not been deleted but has been replaced by a more functional expression which is supported by the original disclosure. Under these circumstances, it is not necessary for the Board further to scrutinise whether Claim 1 has actually been broadened by the new wording.
- 3.5 Consequently, the amendment finds a basis in the disclosure of the application as originally filed, and

it follows that the new wording of Claim 1 complies with the requirement of Article 123(2) EPC.

3.6 As is evident from the wording of the further claims all of which contain a reference to Claim 1 (see section VII(iv)), this finding applies to them as well.

4. *Article 84 EPC*

In the decision under appeal, some objections as to lack of clarity were raised which were not a reason for the refusal (points 7 to 7.3).

4.1 In order to meet the objection to the characterisation of component (A) as being oligomeric the component has been further defined on the basis of page 9, lines 6 to 35 of the application as filed).

4.2 Claim 4 was amended on the basis of page 10, line 36 to page 11, line 1 to provide a definition of "toughening".

4.3 Claim 7 was reworded as a use claim in accordance with page 13, line 29.

4.4 Claim 8 relating to a process for forming the claimed composition was formulated on the basis of original Claim 41 and the definitions of the reactants in Claim 1.

4.5 For the reasons already given, the term "free flowing melt" is regarded as sufficiently clear to be unambiguously understandable by the skilled reader of the application.

- 4.6 In view of these amendments, the Board is satisfied that the claims comply with Article 84 EPC.
5. Consequently, the sole request of the Appellant is successful.

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 for further prosecution on the basis of Claims 1 to 9 filed during the oral proceedings on 4 April 2001.

The Registrar:

The Chairman:

E. Görgmaier

R. Young