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(54) Titre : PROCEDE POUR AMELIORER LA STABILITE A L'HYDROLYSE DANS DES SYSTEMES A BASE DE POLYURETHANE (PU)

(54) Title: PROCESS FOR IMPROVING HYDROLYSIS RESISTANCE IN POLYURETHANE (PU) BASED SYSTEMS

(57) Abrégé/Abstract:
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# PROCESS FOR IMPROVING HYDROLYSIS RESISTANCE IN POLYURETHANE (PU) BASED SYSTEMS

## Abstract

The invention relates to novel processes for improving hydrolysis resistance in polyurethane (PU) based systems, preferably PU adhesives, PU casting resins, PU elastomers or PU foams.

# PROCESS FOR IMPROVING HYDROLYSIS RESISTANCE IN POLYURETHANE (PU) BASED SYSTEMS

The invention relates to novel processes for improving hydrolysis resistance in polyurethane (PU) based systems, preferably PU adhesives, PU casting resins, PU elastomers or PU foams.

Polyurethanes are formed, almost quantitatively, by polyaddition reaction of polyisocyanates with polyhydric alcohols, i.e. polyols. Linking ensues by the reaction of an isocyanate group (–N=C=O) of one molecule with a hydroxyl group (-OH) of another molecule to form a urethane group (-NH-CO-O-).

The course of the reaction between the diisocyanate and the polyol depends on the molar ratio between the components. Intermediate products having a desirable average molecular weight and desirable end groups may well be formed. These intermediate products can then be chain extended later by reaction with a diol or diamine to form the desired polyurethane or polyurethane-polyurea hybrid. These intermediate products are generally known as prepolymers.

Suitable polyols for forming prepolymers include not only diols but also polyalkylene glycol ethers, polyether esters or polyesters having terminal hydroxyl groups (polyester polyols).

Polyester polyols are preferably used to form polyurethanes designed to have high mechanical or dynamical fatigue resistance.

The polyether esters or polyesters with terminal hydroxyl groups that are formed by polycondensation of simple diols and carboxylic acids still contain free carboxylic acids. These catalyse the reaction between the ester groups in the polymer and water, and this leads to a low level of hydrolysis resistance.

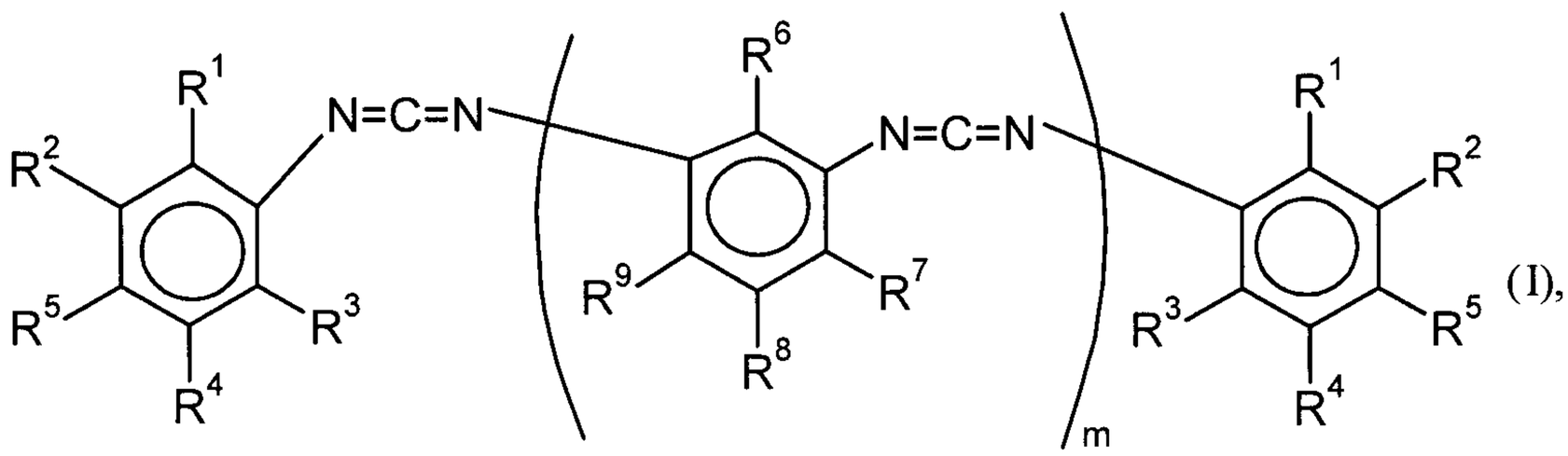
Currently commercially available carbodiimides, as described in EP-A 0799843, are too sluggish for rapid acid removal within the time in which the prepolymers are prepared and made available for further processing to form the cured polymer, or insufficiently soluble to be practical and economical.

The problem addressed by the present invention was therefore that of providing processes for improving the hydrolysis resistance of polyurethane (PU) based systems, that are useful in particular for the production of PU adhesives, PU casting resins, PU elastomers or PU foams, while eschewing materials that are costly and inconvenient to produce.

The problem was surprisingly solved by the process of the present invention in which specified carbodiimides are added to the polyol.

The present invention accordingly provides a process for improving hydrolysis resistance in polyurethane (PU) based systems, in which

- at least one carbodiimide of formula (I)



(I),

where m is 0 – 10,

$R^1$, $R^3$ and $R^5$ are each independently H or methyl,

$R^2$ and $R^4$ are each independently H, methyl, NH-C(O)-$OR^{10}$, where

$R^{10}$ is $C_1$-$C_4$-alkyl or

-$(CH_2)_h$-O-[$(CH_2)_k$-O]$_g$-$R^{11}$,

where h is 1-3, k is 1-3, g is 0-12 and $R^{11}$ is H or $C_1$-$C_4$-alkyl,

and $R^6$, $R^7$, $R^8$ and $R^9$ are each independently H or methyl, and

at least one diisocyanate and optionally a diamine and/or a diol are stirred into

- at least one polyol selected from the group of polyester polyols and/or polyetherester polyols

at temperatures in the range from 0°C to +130°C, preferably +10°C to +60°C, more preferably +15°C to +30°C.

In a preferred embodiment of the invention, m is 0 and

$R^1$, $R^3$ and $R^5$ are each independently H or methyl,

$R^2$ and $R^4$ are each independently H, methyl or -NH-C(O)-$OR^{10}$, where $R^{10}$ is $C_1$-$C_4$-alkyl or

-$(CH_2)_h$-O-[$(CH_2)_k$-O]$_g$-$R^{11}$,

where h is 1-3, k is 1-3, g is 0-12 and $R^{11}$ is H or $C_1$-$C_4$-alkyl,

preferably $R^1$, $R^3$, $R^4$ and $R^5$ are each H or methyl, more preferably $R^1$, $R^3$ and $R^5$ are each methyl and $R^4$ is H,

$R^2$ is -NH-C(O)-$OR^{10}$, where $R^{10}$ is -$C_1$-$C_4$-alkyl or

-$(CH_2)_h$-O-[$(CH_2)_k$-O]$_g$-$R^{11}$,

where h is 1-3, k is 1-3, g is 0-12 and $R^{11}$ is H or $C_1$-$C_4$-alkyl.

It is very particularly preferable in the case of m = 0 when $R^1$ is methyl and $R^2$, $R^3$, $R^4$ and $R^5$ are each H.

It is likewise very particularly preferable in the case of m = 0 when

$R^3$ or $R^5$ is methyl or H,

$R^2$ is -NH-C(O)-$OR^{10}$, where $R^{10}$ is -$C_1$-$C_4$-alkyl or -$(CH_2)_h$-(O-$(CH_2)_k$-O)$_g$-$R^{11}$,

where h is 1-3, k is 1-3, g is 0-12 and $R^{11}$ is H or $C_1$-$C_4$-alkyl

and $R^1$ and $R^4$ are each H.

In a further likewise preferred embodiment of the invention, m is > 0, more preferably m is 1, with $R^1$, $R^3$ and $R^5$ each independently being H or methyl, and

$R^2$ and $R^4$ are each H, methyl or -NH-C(O)-$OR^{10}$, where $R^{10}$ is $C_1$-$C_4$-alkyl or -$(CH_2)_h$-O-[$(CH_2)_k$-O]$_g$-$R^{11}$,

where h is 1-3, k is 1-3, g is 0-12 and $R^{11}$ is H or $C_1$-$C_4$-alkyl, provided one or more of $R^6$, $R^7$, $R^8$ and $R^9$ are each independently H or methyl,

preferably $R^1$, $R^3$, $R^4$ and $R^5$ are each H or methyl, more preferably $R^1$, $R^3$ and $R^5$ are each methyl and $R^4$ is H,

$R^2$ and $R^4$ are each H, methyl or -NH-C(O)-$OR^{10}$, where $R^{10}$ is $C_1$-$C_4$-alkyl or -$(CH_2)_h$-O-[$(CH_2)_k$-O]$_g$-$R^{11}$,

where h is 1-3, k is 1-3, g is 0-12 and $R^{11}$ is H or $C_1$-$C_4$-alkyl, provided one or more of $R^6$, $R^7$, $R^8$ and $R^9$ are each independently H or methyl,

It is further preferable when $R^1$, $R^3$, $R^4$, $R^5$ are each H or methyl, more preferably methyl,

$R^2$ is -NH-C(O)-$OR^{10}$, where $R^{10}$ is -$C_1$-$C_4$-alkyl or

-$(CH_2)_h$-O-[$(CH_2)_k$-O]$_g$-$R^{11}$,

where h is 1-3, k is 1-3, g is 0-12 and $R^{11}$ is H or $C_1$-$C_4$-alkyl,

$R^6$, $R^7$, $R^8$ and $R^9$ are each independently H or methyl and more preferably at least one of $R^6$, $R^7$ and $R^9$ is methyl.

It is likewise highly preferable in the case of m = 1 when

$R^3$ or $R^5$ is methyl or H,

$R^2$ is -NH-C(O)-$OR^{10}$, where $R^{10}$ is -$C_1$-$C_4$-alkyl or

-$(CH_2)_h$-O-[$(CH_2)_k$-O]$_g$-$R^{11}$,

where h is 1-3, k is 1-3, g is 0-12 and $R^{11}$ is H or $C_1$-$C_4$-alkyl,

and $R^1$ and $R^4$ are each H and

$R^6$, $R^7$, $R^8$ and $R^9$ are each independently H or methyl, preferably at least one of $R^6$, $R^7$ and $R^9$ is methyl.

In a preferred embodiment of the invention, at least one of $R^7$ and $R^9$ is methyl.

The compounds of formula (I) are commercially available substances in that they are available from Rhein Chemie Rheinau GmbH, for example, under the trade names Stabaxol® and Hycasyl® for example.

Preference is likewise given to mixtures of two or more carbodiimides, at least one of which corresponds to formula (I). In the case of a mixture, mean m can also be a fractional number.

Polyols for the purposes of the invention are selected from the group of polyester polyols and/or polyetherester polyols.

Polyester polyols for the purposes of the invention are compounds with a molecular weight in g/mol of preferably up to 2000, more preferably in the range from 500 to 2000 and yet more preferably in the range from 500 to 1000.

The term polyesterpolyols is to be understood as meaning for the purposes of the present invention not only compounds having two or three hydroxyl groups per molecule but also compounds having more than three hydroxyl groups per molecule.

Polyester polyols are preferred polyols. It is advantageous for the polyester polyols and/or polyetherester polyols to have an OH number of up to 200, preferably between 20 and 150 and more preferably between 50 and 115.

Particularly suitable polyester polyols are reaction products of various diols with aromatic or aliphatic dicarboxylic acids and/or polymers of lactones.

Preference here is given to aromatic dicarboxylic acids useful for forming suitable polyester polyols. Particular preference is given here to terephthalic acid, isophthalic acid, phthalic acid, phthalic anhydride and also substituted dicarboxylic acid compounds having a benzene ring.

Useful aliphatic dicarboxylic acids are preferably those aliphatic dicarboxylic acids useful for forming suitable polyester polyols, more preferably sebacic acid, adipic acid and glutaric acid.

Preferred polymers of lactones are useful for forming suitable polyester polyols, more preferably polycaprolactone.

The dicarboxylic acids and the polymers of lactones are commercially available substances.

Particular preference is also given to those diols useful for forming suitable polyester polyols, most preferably ethylene glycol, butanediol, neopentyl glycol, hexanediol, propylene glycol, dipropylene glycol, diethylene glycol and cyclohexanedimethanol.

In a further preferred embodiment of the invention, at least one polyetherester polyol is used.

Preference for this is given to the reaction products of various aforementioned polyols with aromatic or aliphatic dicarboxylic acids and/or polymers of lactones (e.g. polycaprolactone).

The polyester polyols and/or polyetherester polyols used for the purposes of the inventions are commercially available compounds in that they are available from Bayer MaterialScience AG under the trade name of Baycoll® or Desmophen®.

Aromatic and aliphatic diisocyanates are preferred. Tolylene 2,4-diisocyanate, tolylene 2,6-diisocyanate, phenylene diisocyanate, 4,4-diphenylmethane diisocyanate, methylene bis(4-phenyl isocyanate), naphthalene 1,5-diisocyanate, tetramethylene 1,4-diisocyanate and/or hexamethylene 1,6-diisocyanate are particularly preferred and tolylene 2,4-diisocyanate and tolylene 2,6-diisocyanate are very particularly preferred.

The diisocyanates used for the purposes of the inventions are commercially available compounds in that they are available from Bayer MaterialScience AG under the trade name of Desmodur®.

In a further embodiment of the invention, the composition additionally contains at least one diamine and/or diol.

Diamines, which are used for chain extension, are preferably 2-methylpropyl 3,5-diamino-4-chlorobenzoate, bis(4,4′-amino-3-chlorophenyl)methane, 3,5-dimethylthio-2,4-tolylenediamine, 3,5-dimethylthio-2,4-tolylenediamine, 3,5-diethyl-2,4-tolylenediamine, 3,5-diethyl-2,6-tolylenediamine, 4,4′-methylenebis(3-chloro-2,6-diethylaniline) and 1,3-propanediol bis(4-aminobenzoate).

Preference for use as diols is given to butanediol, neopentyl glycol, hexanediol, propylene glycol, dipropylene glycol, diethylene glycol and/or cyclohexanedimethanol. 1,3-butanediol or 1,6-hexanediol are particularly preferred.

The diamines or diols used for chain extension within the meaning of the invention are commercially available compounds in that they are available from Rhein Chemie Rheinau GmbH under the trade name of Addolink®.

Catalysts used are preferably dibutyltin dilaurates or triethylenediamine in dipropylene glycol.

Catalysts used for the purposes of the inventions are commercially available compounds in that they are available from Rhein Chemie Rheinau GmbH under the trade name of Addocat®.

The ratio of carbodiimide to polyol is preferably 0.1-5, more preferably 1-3 parts by weight per 100 parts by weight of polyol.

The ratio of diisocyanate to polyol is preferably 20-50:100 parts by weight, more preferably 30:100 parts by weight.

In those cases where the composition contains at least one diamine and/or diol in addition to the polyester polyol and/or polyetherester polyol and the carbodiimide and also the diisocyanate, the amount of diamine and/or diol is 5-30 wt%, based on the composition.

In those cases where the composition contains at least one catalyst in addition to the polyol and the carbodiimide and also the diisocyanate, the amount of catalyst is 0.01-1 wt%, based on the composition.

The polyurethane (PU) based systems obtained by this process have increased hydrolysis resistance.

The purview of the invention encompasses all the moiety definitions, indices, parameters and explications recited hereinabove and hereinbelow in general terms or in preferred ranges in combination with one another, including that is in any desired combination of the respective ranges and preferred ranges.

The examples which follow are offered by way of elucidation not limitation of the invention.

**Working examples**

The following substances were used in the examples which follow:

Baycoll® AV 2113: a branched polyester polyol having an OH number of 110 mg KOH/g and an acid number of 0.83 mg KOH/g, from Bayer MaterialScience AG.

Stabaxol® I TC: a carbodiimide of formula (I) where m = 0 and $R^1$=$CH_3$, $R^2$=H, $R^3$=H, $R^4$=H and $R^5$=H.

CDI 1: carbodiimide of formula (I) where m = 0 and $R^1$ = $CH_3$, $R^4$ = H, $R^3$ = H, $R^2$ = NH-C(O)-OR and $5^R$ = H.

Stabaxol® P 200: a polymeric aromatic carbodiimide based on tetramethylxylylene diisocyanate from Rhein Chemie Rheinau GmbH.

Stabaxol® I: a monomeric carbodiimide based on 2,6-diisopropylphenyl isocyanate from Rhein Chemie Rheinau GmbH.

Desmodur® PU 0129: a 2,4/4,4 diphenylmethane diisocyanate isomer mixture.

Addolink® B: a 1,4-butanediol from Rhein Chemie Rheinau GmbH as diol component.

Addocat® 201: a dibutyltin dilaurate from Rhein Chemie Rheinau GmbH, as catalyst.

Carbodilite® HMV-8 CA: a polymeric aliphatic carbodiimide from Nisshinbo Industries, INC.

One portion of the formulation further contains a molecular sieve for moisture adsorption.

Desmocoll® 140: a substantially linear hydroxyl polyurethane having a hydroxyl content < 0.1 from Bayer MaterialScience AG.

Baycoll® AS 2060: a lightly crosslinked polyester polyol having a hydroxyl number of 60 ± 3 mg KOH/g and an acid number of ≤ 2.0 mg KOH/g from Bayer MaterialScience AG.

Desmodur® RFE: a solution of tris(p-isocyanatophenyl) thiophosphate in ethyl acetate having an NCO content of 7.2 ± 0.2% as isocyanate curative from Bayer MaterialScience AG.

**Example 1**

The following mixtures were produced as follows:

Mixture A (comparator): 100 g of the room temperature liquid Baycoll® AV 2113.

Mixture B (for preparing mixture II of present invention): 100 g of the room temperature liquid Baycoll® AV 2113 were admixed with 0.6 g of the carbodiimide of formula (I) where m = 0 and $R^1$=$CH_3$, $R^2$=H, $R^3$=H, $R^4$=H and $R^5$=H (Stabaxal® I TC) and stored at 30°C for 24 h.

Mixture C (comparator): 100 g of the room temperature liquid Baycoll® AV 2113 were admixed with 0.6 g of Stabaxol® I and stored at 30°C for 24 h.

Mixture D (comparator): 100 g of the room temperature liquid Baycoll® AV 2113 were admixed with 0.6 g of Stabaxol® P 200 and stored at 30°C for 24 h.

Mixture E (comparator): 100 g of the room temperature liquid Baycoll® AV 2113 were admixed with 0.6 g of Carbodilite® HMV-8 CA and stored at 30°C for 24 h. The two substances cannot be mixed. So this mixture was not employable for further tests.

The acid number of the mixtures was measured in accordance with DIN 53402 after storage. The results are shown in table 1:

**Table 1:**

| | acid number mg KOH/g |
|---|---|
| mixture A (comparator) | 0.83 |
| mixture B | 0.30 |
| mixture C (comparator) | 0.80 |
| mixture D (comparator) | 0.60 |

The particular composition of the elastomers obtained is apparent from table 2. All particulars are in parts by weight, unless otherwise stated.

**Table 2:**

| | mixture A (comparator) | mixture B | mixture C (comparator) | mixture D (comparator) | Addocat® 201 | Addolink® B | Desmodur® PU 0129 |
|---|---|---|---|---|---|---|---|
| I (c) | 100 | | | | 0.06 | 10 | 56 |
| II(i) | | 100 | | | 0.06 | 10 | 56 |
| III (c) | | | 100 | | 0.06 | 10 | 56 |
| IV (c) | | | | 100 | 0.06 | 10 | 56 |

c = comparative example; i = inventive example

All the mixtures additionally contained 5 parts by weight of molecular sieve for moisture absorption.

The mixtures were processed by the one-shot method, i.e. premixed with molecular sieve, Addocat® 201 and Addolink® B and reacted with the diisocyanate (Desmodur® PU 0129). The mixture which was initially liquid and reacted to form a solid elastomer after a few minutes was poured into a warm test mould at 30°C, demoulded after 1 h and conditioned at 100°C for 16 h.

Standard test specimens were die-cut out of the test sheets thus obtained for mixtures I to IV, after they had been stored at 22°C for 7 days.

The hydrolysis resistance of mixtures I to IV was determined as follows:

The die-cut standard test specimens were stored in water at a temperature of 80°C for 4 days. The tensile strength of the test specimens stored in water was measured after every 24 h.

Table 3 shows the percentage relative tensile strength starting at day 0 with 100%.

**Table 3:**

| | day 0 | day 1 | day 2 | day 3 | day 4 |
|---|---|---|---|---|---|
| I (c) | 100 | 80 | 70 | 60 | 50 |
| II (i) | 100 | 90 | 90 | 90 | 80 |
| III (c) | 100 | 90 | 80 | 70 | 60 |
| IV (c) | 100 | 90 | 85 | 75 | 65 |

c = comparative example; i = inventive example

**Example 2:**

The following mixtures were produced as follows:

Mixture A2 (comparator): 14 g of Desmocoll® 140 were dissolved in 75 g of ethyl acetate at 85°C. 7 g of Baycoll® AS 2060 were added during cooling.

Mixture B2 (comparator): 14 g of Desmocoll® 140 were dissolved in 75 g of ethyl acetate at 85°C. 7 g of Baycoll® AS 2060 were added during cooling. 0.32 g of Stabaxol® I was dissolved in the cold mixture with stirring, followed by storage at room temperature for five days.

Mixture C2 (to produce the inventive mixture): 14 g of Desmocoll® 140 were dissolved in 75 g of ethyl acetate at 85°C. 7 g of Baycoll® AS 2060 were added during cooling. 0.2 g of the carbodiimide of formula (I) where $m = 0$ and $R^1 = CH_3$, $R^2 = H$, $R^3 = H$, $R^4 = H$ and $R^5 = H$ (Stabaxol® I TC) was dissolved in the cold mixture with stirring, followed by storage at room temperature for five days.

Mixture D2 (to produce the inventive mixture): 14 g of Desmocoll® 140 were dissolved in 75 g of ethyl acetate at 85°C. 7 g of Baycoll® AS 2060 were added during cooling. 0.31 g of CDI 1 was dissolved in the cold mixture with stirring, followed by storage at room temperature for five days.

**Table 4:**

| adhesive number | mixture A2 | mixture B2 | mixture C2 | mixture D2 | Desmodur® RFE |
|---|---|---|---|---|---|
| I (comparator) | 96 g | | | | 4 g |
| II(comparator) | | 96.32 g | | | 4 g |
| III | | | 96.2 g | | 4 g |
| IV | | | | 96.31 g | 4 g |

Mixtures A2, B2, C2 and D2 were each admixed at room temperature with 4 g of Desmodur® RFE isocyanate curative. Adhesives I, II, III and IV thus obtained were applied by hand with a wire-wound blade with 10 µm size to a commercially available, 23 µm thick, unpretreated DIN A4-sized PET sheet, although the topmost 50 mm of the sheet were not coated with adhesive owing to a protective strip to be removed later. The solvent of the adhesive (ethyl acetate) was subsequently flashed off at room temperature for five minutes. Then, a commercially available, 25 µm thick, unpretreated aluminium foil was laminated in place by hand. The laminate thus formed was cured at 50°C under a moulding pressure of 10 kg for one hour. Thereafter, the samples were stored under standard conditions for at least seven days, cut to size in accordance with ISO 11339 and subjected to the hydrolysis resistance test. In this test, the stored samples were stored individually freely suspended in an autoclave at 121°C (250°F) and 100% relative humidity for 30 min. The measurements were subsequently carried out at an extension rate of 100 mm/min.

The results of the hydrolysis resistance tests are reported in table 5.

**Table 5:**

| adhesive number | 0 min storage [N/100 mm] | 30 min storage [N/100 mm] |
|---|---|---|
| I (comparator) | 1.73 | 0.31 |
| II (comparator) | 1.74 | 0.29 |
| III | 1.53 | 1.33 |
| IV | 1.39 | 1.34 |

Interpretation of experimental results:

The aromatic carbodiimides from the prior art exhibit no positive effect with respect to the tensile shear strength compared to the carbodiimide-free samples. In the examples according to the invention, by contrast, adhesives III and IV exhibit a markedly increased hydrolysis resistance.

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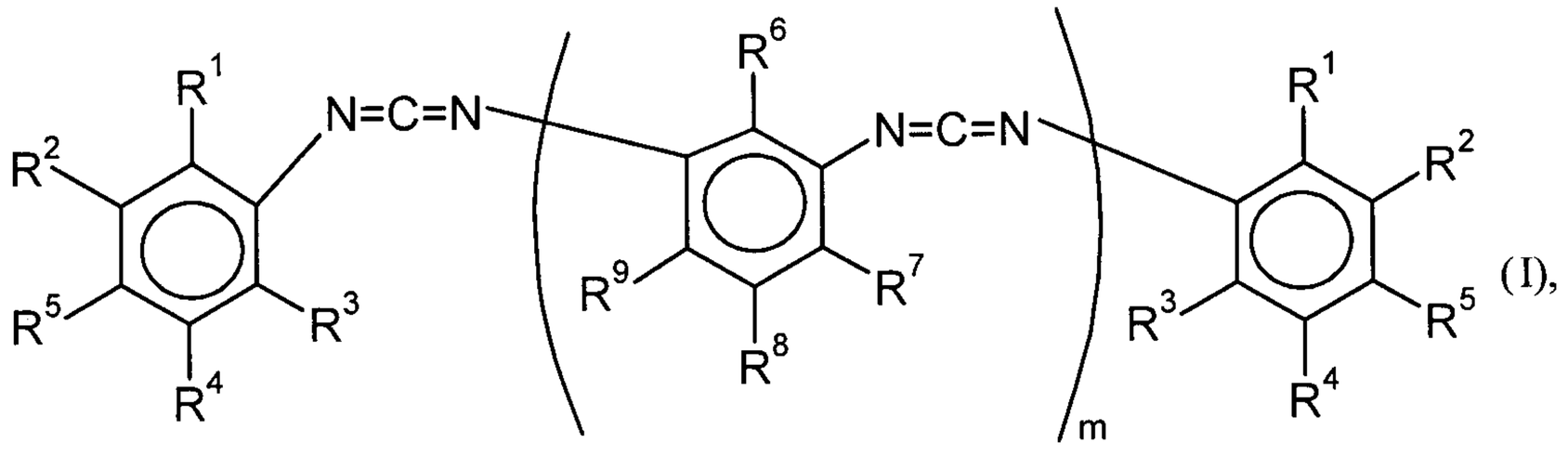
**Claims**

1. Process for improving the hydrolysis resistance of polyurethane (PU) based systems, characterized in that

- at least one carbodiimide of formula (I)



(I),

where m is 0 – 10,

$R^1$, $R^3$ and $R^5$ are each independently H or methyl,

$R^2$ and $R^4$ are each independently H, methyl, NH-C(O)-$OR^{10}$, where

$R^{10}$ is $C_1$-$C_4$-alkyl, or

$-(CH_2)_h-O-[(CH_2)_k-O]_g-R^{11}$,

where h is 1-3, k is 1-3, g is 0-12 and $R^{11}$ is H or $C_1$-$C_4$-alkyl,

and $R^6$, $R^7$, $R^8$ and $R^9$ are each independently H or methyl, and

at least one diisocyanate and optionally a diamine and/or a diol are stirred into

- at least one polyol selected from the group of polyester polyols and/or polyetherester polyols

at temperatures in the range from 0°C to +130°C, preferably +10°C to +60°C, more preferably +15°C to +30°C.

2. Process according to Claim 1, characterized in that in the carbodiimide of formula (I) m is 0 and

$R^1$, $R^3$ and $R^5$ are each independently H or methyl,

$R^2$ and $R^4$ are each independently H, methyl or -NH-C(O)-$OR^{10}$, where $R^{10}$ is $C_1$-$C_4$-alkyl

or

$-(CH_2)_h-O-[(CH_2)_k-O]_g-R^{11}$,

where h is 1-3, k is 1-3, g is 0-12 and $R^{11}$ is H or $C_1$-$C_4$-alkyl,

preferably $R^1$, $R^3$, $R^4$ and $R^5$ are each H or methyl, more preferably $R^1$, $R^3$ and $R^5$ are each methyl and $R^4$ is H,

$R^2$ is -NH-C(O)-$OR^{10}$, where $R^{10}$ is -$C_1$-$C_4$-alkyl or
-$(CH_2)_h$-O-[$(CH_2)_k$-O]$_g$-$R^{11}$,

where h is 1-3, k is 1-3, g is 0-12 and $R^{11}$ is H or $C_1$-$C_4$-alkyl.

3. Process according to Claim 1, characterized in that in the carbodiimide of formula (I) m is > 0,

with $R^1$, $R^3$ and $R^5$ each independently being H or methyl, and

$R^2$ and $R^4$ are each H, methyl or -NH-C(O)-$OR^{10}$, where $R^{10}$ is $C_1$-$C_4$-alkyl or -$(CH_2)_h$-O-[$(CH_2)_k$-O]$_g$-$R^{11}$,

where h is 1-3, k is 1-3, g is 0-12 and $R^{11}$ is H or $C_1$-$C_4$-alkyl, provided one or more of $R^6$, $R^7$, $R^8$ and $R^9$ are each independently H or methyl,

preferably $R^1$, $R^3$, $R^4$ and $R^5$ are each H or methyl, more preferably $R^1$, $R^3$ and $R^5$ are each methyl and $R^4$ is H,

$R^2$ is -NH-C(O)-$OR^{10}$, where $R^{10}$ is -$C_1$-$C_4$-alkyl or
-$(CH_2)_h$-O-[$(CH_2)_k$-O]$_g$-$R^{11}$,

where h is 1-3, k is 1-3, g is 0-12 and $R^{11}$ is H or $C_1$-$C_4$-alkyl,

$R^6$, $R^7$, $R^8$ and $R^9$ are each independently H or methyl and more preferably at least one of $R^6$, $R^7$ and $R^9$ is methyl.

4. Process according to one or more of Claims 1 to 3, characterized in that the carbodiimides are mixtures of two or more carbodiimides, at least one of which corresponds to formula (I).

5. Process according to one or more of Claims 1 to 4, characterized in that it comprises at least one catalyst.

6. Process according to one or more of Claims 1 to 5, characterized in that at least one diamine and/or diol are used.

7. Process according to one or more of Claims 1 to 4, characterized in that, prior to the addition of the diisocyanate, at least one catalyst, preferably dibutyltin dilaurate, and at least one diol, preferably 1,4-butanediol, and optionally molecular sieve are added.