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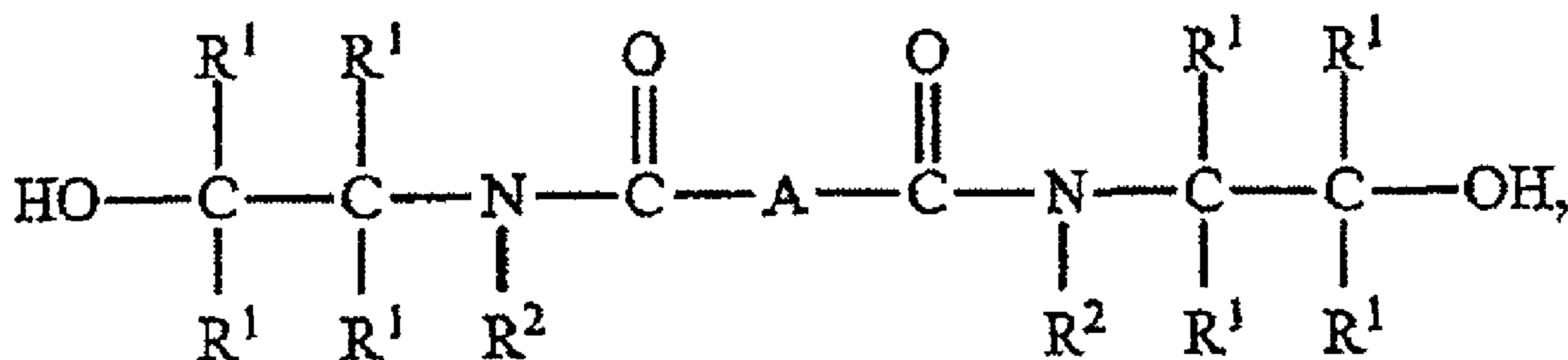
(72) **Inventeurs/Inventors:**
GREND, WERNER, DE;
SPYROU, EMMANOUIL, DE;
WEIHRAUCH, THOMAS, DE;
LAMMERS, CHRISTOPH, DE;
LOESCH, HOLGER, DE;
BEHRENDT, KLAUS, DE

(73) **Propriétaire/Owner:**
EVONIK DEGUSSA GMBH, DE

(74) **Agent:** FETHERSTONHAUGH & CO.

(54) **Titre :** β -HYDROXYALKYLAMIDES, PROCEDE POUR LES PRODUIRE ET LEUR UTILISATION

(54) **Title:** β -HYDROXYALKYLAMIDES, METHOD FOR THEIR PRODUCTION AND USE THEREOF



(57) **Abrégé/Abstract:**

The invention relates to novel β -hydroxyalkylamides, to a method for their production and to the use thereof. In an embodiment, the invention relates to a β -hydroxyalkylamide, comprising two or three or four β -hydroxyalkylamide groups per molecule of the formula I: (see formula I) wherein: R^1 , R^2 and A are as defined herein.

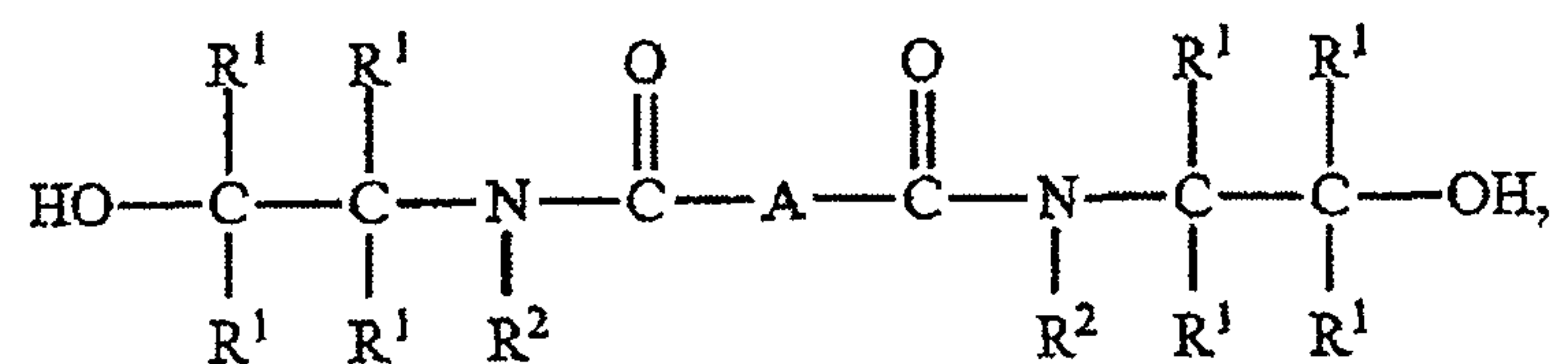


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Abstract

The invention relates to novel β -hydroxyalkylamides, to a method for their production and to the use thereof. In an embodiment, the invention relates to a β -hydroxyalkylamide, comprising two or three or four β -hydroxyalkylamide groups per

5 molecule of the formula I:



wherein: R^1 , R^2 and A are as defined herein.

β -Hydroxyalkylamides, method for their production and use thereof

The invention relates to new β -hydroxyalkylamides, to a method for preparing them, and to the use.

5

β -Hydroxyalkylamides are valuable intermediates in organic synthesis.

For ten years, β -hydroxyalkylamides have found technical applications in powder coating materials, as curing agents (also referred to as crosslinkers).

10

β -Hydroxyalkylamides and methods for preparing them are also known from the following patent documents: DE 25 09 237, DE 198 23 925, EP 0 473 380, EP 0 960 878, WO 2000050384, WO 200055266.

15

Powder coating materials based on triglycidyl isocyanurate (TGIC) as crosslinker (curing agent) and on acid-functional polyesters produce corrosion-resistant and weather-stable powder coatings. TGIC, however, is classed as mutagenic and toxic.

20

Toxicologically unobjectionable and at the same time also more reactive are β -hydroxyalkylamide crosslinkers. Patent specifications US 4,076,917 and US 4,101,606 combine β -hydroxyalkylamides with polymers having at least one carboxylate or anhydride function, more particularly with polyacrylates, to give powder coating materials. EP 0 322 834 describes thermosetting powder coating materials composed of β -hydroxyalkylamides and polyesters containing acid groups.

25

Considerable interest attaches to coating systems which give a substrate a uniformly even and matt surface. The reason for this is primarily practical. Glossy surfaces require a far higher degree of cleaning than matt surfaces. Furthermore, it may be desirable on safety grounds to avoid strongly reflecting surfaces. Across broad areas of application in the powder coatings industry, such as the architectural, automotive, and metal furniture segments, etc., there is increasing demand for matt (10-30 units) and semimatt (30-50 units) surfaces, measured as reflectometer values according to DIN 67530 / ISO 2813 at an incident angle of 60°.

30

35

The simplest principle for obtaining a matt surface is to admix the powder coating

material with smaller or larger amounts – depending on the extent of the desired matt effect – of fillers, such as chalk, finely divided silicon dioxide or barium sulfate, for example. These additions, however, produce a deterioration in the technical film properties, such as adhesion, flexibility, impact strength, and chemicals resistance.

5

The addition of substances incompatible with the coating material, such as waxes or cellulose derivatives, for example, does indeed result in matting, distinctly, but slight changes during the course of extrusion lead to fluctuations in the surface gloss and to fade-out in dark shades. The reproducibility of the matt effect is not ensured.

10

EP 0698645 describes producing matt powder coating materials by dry-blending at least two separately fabricated hydroxyalkylamide powder coating materials.

15

For semimatt and matt (< 50 gloss units) powder coatings with hydroxyalkylamides, then, the state of the art comprises dry blends – in other words, it is necessary to produce, separately, two hydroxyalkylamide powder coating materials, with different acid numbers of the binder components, which are then supplied for grinding as a dry blend. This involves considerable extra cost and effort and, in the event of deviation in a binder component, leads to gloss deviations, which take considerable additional cost and effort to correct. Furthermore, these dry blends suffer separation, including at the premises of the end customer, with a resulting shift in gloss if the powder coating material is – as is usual – to be recycled.

20

The laid-open specification KR 10-2009-0111720 (application number 10-2008-0037454), translated title “CYCLOALKANE DICARBOXAMIDE COMPOUNDS, THEIR PREPARATION AND APPLICATION” (see also J. Korean Ind. Eng. Chem., Vol. 20, No. 2, April 2009, 195-200), discloses, in particular in example 1, the there-named compound N¹, N¹, N⁴, N⁴-tetrakis(2-hydroxyethyl)cyclohexane-1,4-dicarboxamide (formula 3). This compound according to figure 2 has only one peak by DSC analysis, with a maximum peak at approximately 190°C. A cis/trans content for the compound is not stated. Furthermore, polyesters containing carboxyl groups, which are not precisely defined but are indicated only by broad ranges for certain parameters (polyesters not unambiguously characterized and unknown on the market with this viscosity), are crosslinked with this compound and compared with the known β-hydroxyalkylamide, here identified in example 3 as [N¹,N¹,N⁶,N⁶-tetrakis(2-hydroxyethyl)adipamide]

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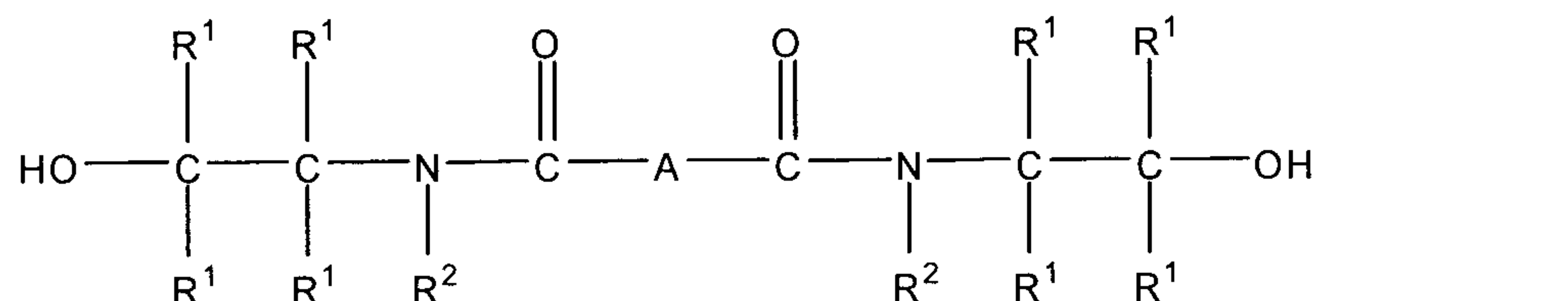
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(available as VESTAGON HAA 320 or PRIMID XL 552), in other words with curing agents from the prior art and with long-established market products known to lead to glossy surfaces on the coatings produced. Figures 3 and 4 show the metal panels. No description is given to the effect that the coatings in question are matt coatings. Nor is this possible, since glossy coatings are obtained with the conventional curing agent.

It was an object of the present invention to find new β -hydroxyalkylamides which can be used as intermediates and curing agents. A particular object of the invention was to find new β -hydroxyalkylamides which in powder coating materials, after curing, lead to matt surfaces, and which do not require dry blending in the production of the powder coating materials.

This object has been achieved by the new β -hydroxyalkylamides of the invention.

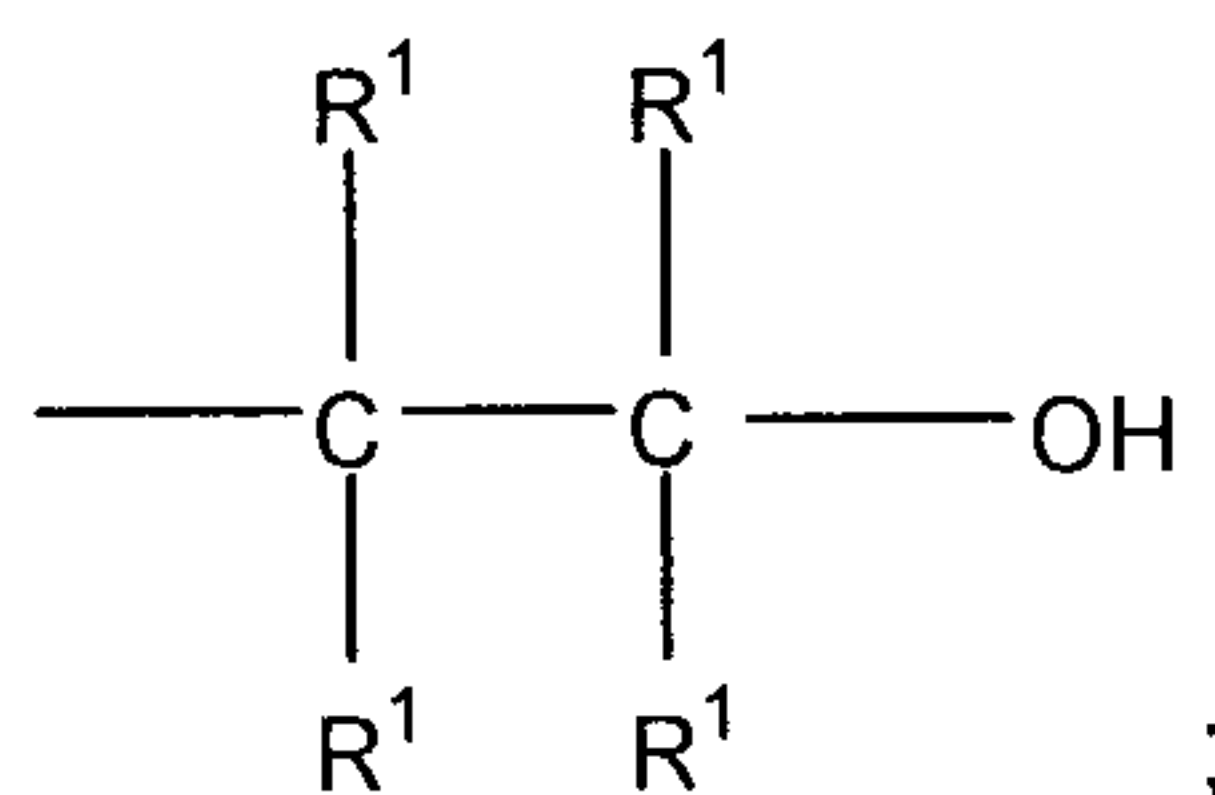
The invention provides β -hydroxyalkylamides having two or three or four β -hydroxyalkylamide groups per molecule of the formula I



where

R^1 and R^2 independently of one another are identical or different radicals selected from alkyl radical, cycloalkyl radical, aryl radical, aralkyl radical or alkenyl radical having 1-24 carbon atoms, it also being possible for the radicals to contain heteroatoms and/or functional groups, and it also being possible for R^1 to be hydrogen,

and it also being possible for R^2 to be

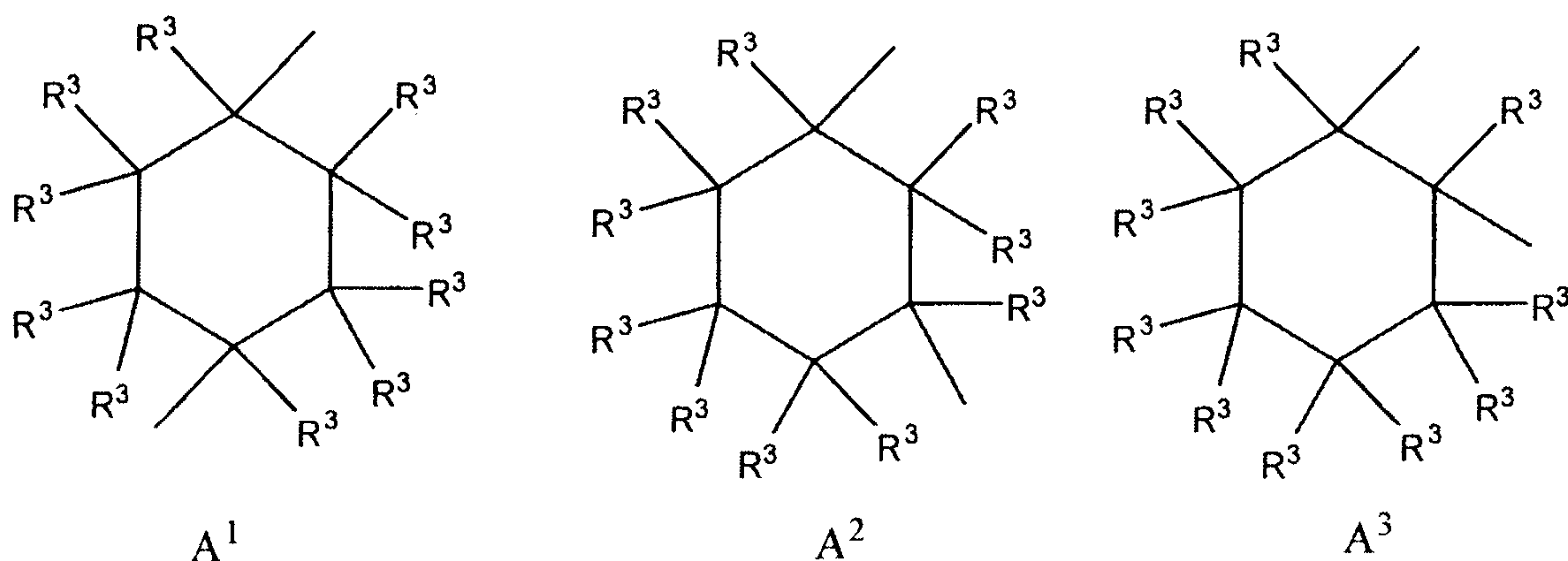


and

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4

A is



where radicals R^3 independently of one another are identical or different radicals selected from hydrogen, alkyl radical, cycloalkyl radical, aryl radical, aralkyl radical or alkenyl radical having 1-24 carbon atoms, it also being possible for the radicals to contain heteroatoms and/or functional groups, and it being possible for two or more substituents R^3 to be linked with one another to form rings;

where the β -hydroxyalkylamides are present in solid form below 150°C .

It may be that the invention is represented by the formula I above, wherein the β -hydroxyalkylamide has a trans content of greater than or equal to 70 mol % based on the total amount of all of the isomers of the β -hydroxyalkylamide; wherein the β -hydroxyalkylamide has two endothermic peaks according to DSC analysis (differential scanning calorimetry), where peak 1 is situated in the region of 140 - 170°C ., with a maximum of 155 - 165°C ., and peak 2 is situated in the region of 170 - 210°C ., with a maximum of 175 - 207°C .; and wherein a ratio of the enthalpies of endothermic peak 1 to endothermic peak 2 is 1:1 to 1:5.

It may be that the invention is represented by the formula I above, wherein the β -hydroxyalkylamide has a trans content of greater than or equal to 70 mol % based on the total amount of all of the isomers of the β -hydroxyalkylamide; two endothermic peaks according to DSC analysis (differential scanning calorimetry), where peak 1 is

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situated in the region of 140-170° C., with a maximum of 155-165° C., and peak 2 is situated in the region of 170-210° C., with a maximum of 175-207° C.; a ratio of the enthalpies of endothermic peak 1 to endothermic peak 2 is 1:1 to 1:5; and a XRPD spectrum of the powder sample in an x-ray diffractometer, measured with Cu K α radiation (1.541 Å) has the following peaks:

Peak #	Degrees 2theta \pm 0.2 degree 2theta	d (Å)
1	14.90	5.94
2	16.70	5.31
3	17.40	5.09
4	21.20	4.19
5	21.60	4.11
6	26.00	3.43.

Surprisingly it has been found that β -hydroxyalkylamides having a cyclohexane ring in the framework, and where the β -hydroxyalkylamides are present in solid form below 150° C, lead in powder coating materials, after curing, to matt surfaces.

10 Furthermore, the β -hydroxyalkylamides of the invention represent new intermediates.

The β -hydroxyalkylamides can be prepared from various starting materials. A known reaction is that of β -hydroxyalkylamines with esters of carboxylic acids, the latter generating the parent structure (A). Depending on the selection of the starting materials, it is possible in this way to generate the β -hydroxyalkylamides of the
15 invention.

Alternative but less preferred methods are based on other carboxylic acid derivatives, such as carboxylic acids, carbonyl chlorides, carboxylic anhydrides or other activated carboxylic acid derivatives as starting materials, which are reacted with β -hydroxyalkylamines.

20 Suitable β -hydroxyalkylamines are those which have alkyl groups having at least 2 to 10 carbon atoms in the hydrocarbon framework. The alkyl groups may be linear, branched or else cyclic. It is likewise possible for the alkyl groups to be substituted by

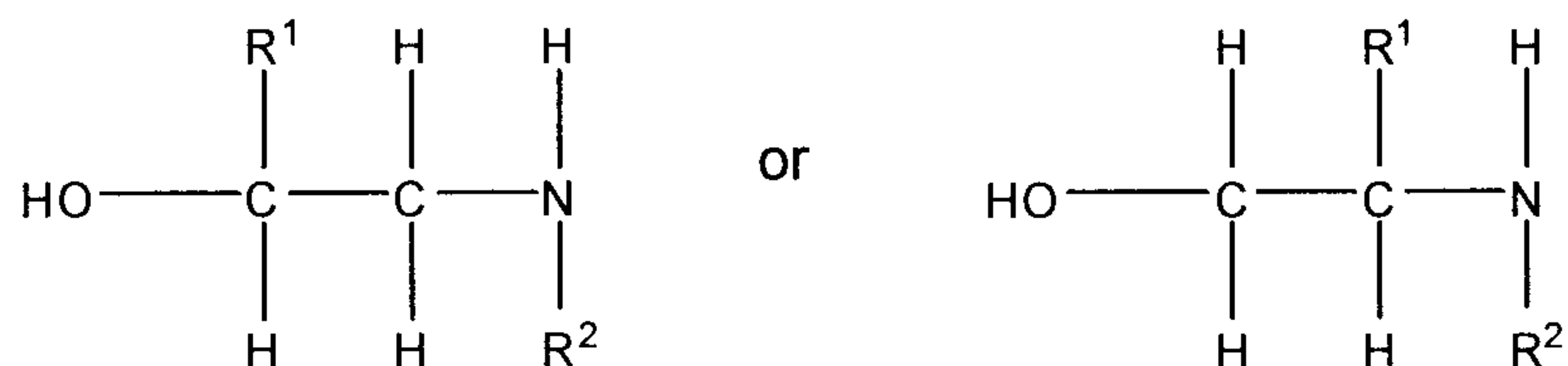
heteroatoms, preferably oxygen, nitrogen. Furthermore, these alkyl groups may also contain functional groups, preferably carbonyl groups, carboxyl groups, amino groups, amide groups, urethane groups, and may carry an additional alkyl radical on the nitrogen.

5

In this invention the β -hydroxyalkylamides are preferably prepared from N-alkyl-1,2-alkanolamines and/or from N,N-bis-2-hydroxyalkylamines and esters of cyclohexanedicarboxylic acids.

- 10 Particular preference is given to using β -hydroxyalkylamines of the formula II and/or III:

formulae II



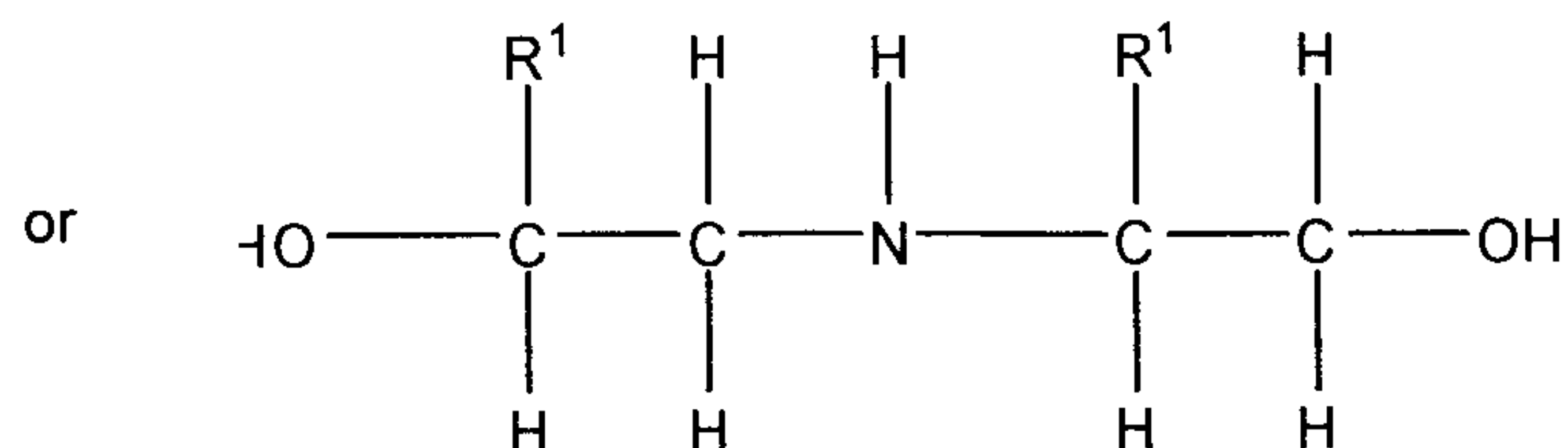
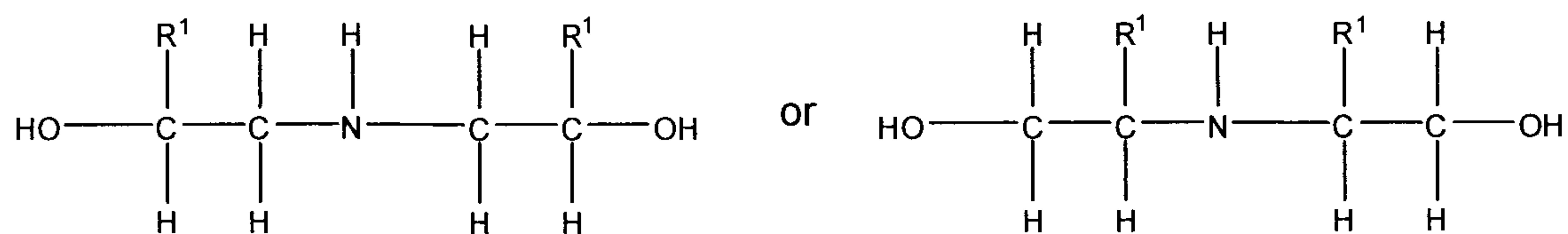
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where

R^1 is hydrogen, methyl, ethyl, propyl,

R^2 is methyl;

- 20 formulae III



25

where radicals R^1 simultaneously or independently of one another are hydrogen, methyl, ethyl, propyl.

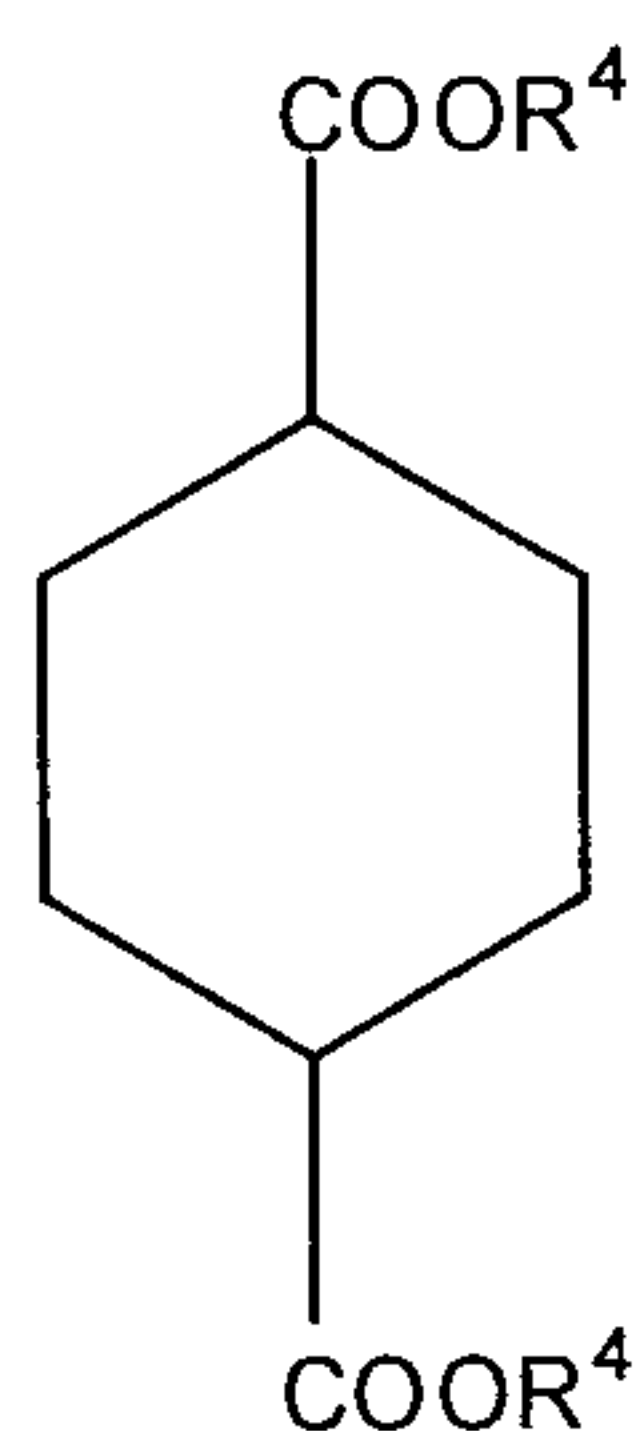
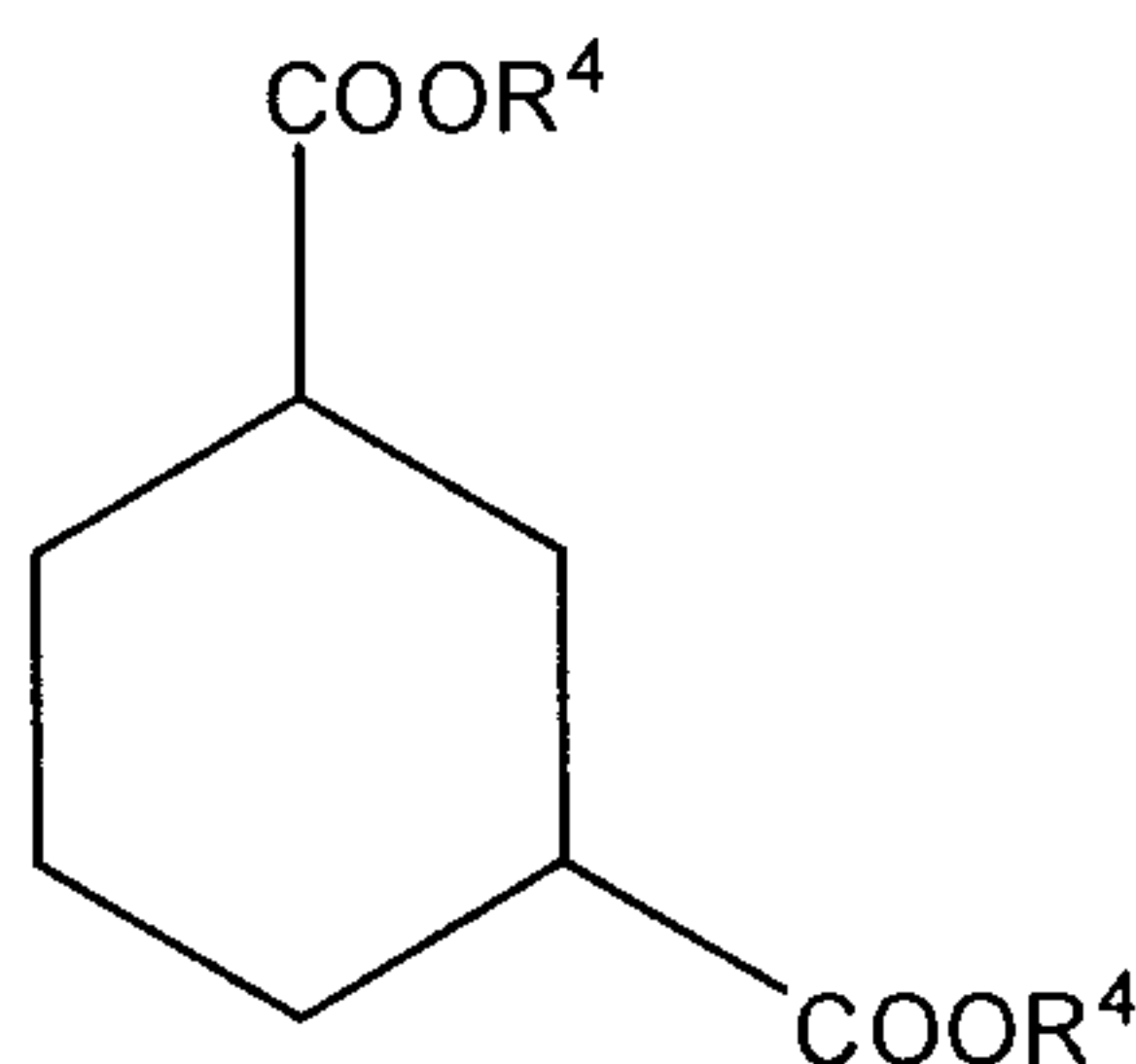
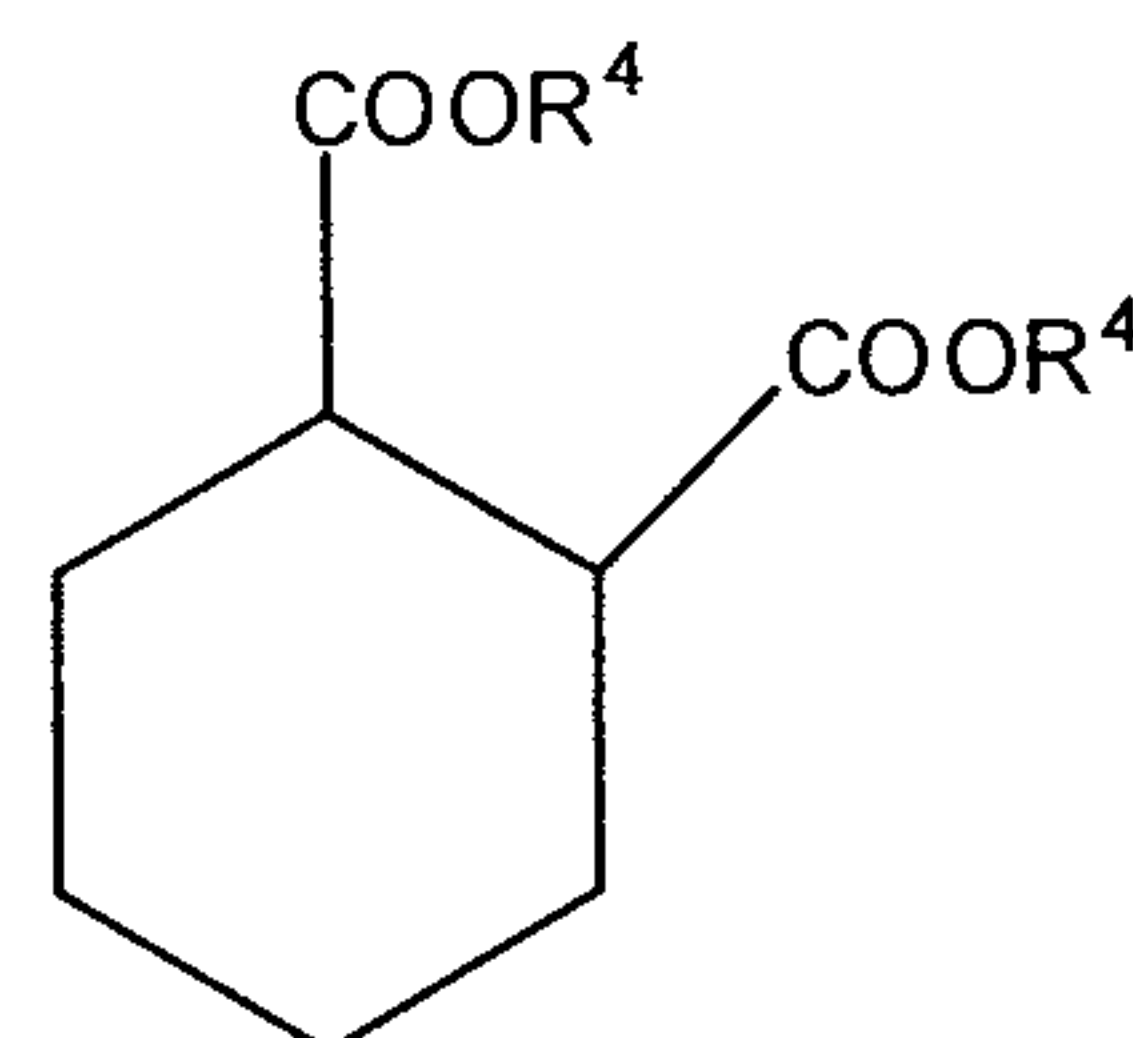
Particular preference is given in accordance with the invention to using the following compounds as starting materials for preparing the β -hydroxyalkylamides:

5 diethanolamine (DEA), di-isopropanolamine (DIPA), di-sec-butanolamine, N-methylethanolamine, N-methylisopropanolamine.

Suitable starting compounds for the substituent A in the β -hydroxyalkylamides of the invention are 1,2-substituted, 1,3-substituted, and 1,4-substituted

10 cyclohexanedicarboxylic acid derivatives, more particularly dialkyl esters of cyclohexanedicarboxylic acids. The starting compounds here may have any desired cis/trans content.

15 Preference is given to using compounds of the formula IV:

 A^4  A^5  A^6

where radicals R^4 simultaneously or independently of one another are methyl, ethyl,

20 propyl, butyl.

Particular preference is given to using 1,4-substituted cyclohexanedicarboxylic esters, very preferably dimethyl 1,4-cyclohexyldicarboxylate.

25 The β -hydroxyalkylamides that are particularly preferred in accordance with the invention, formed from dialkyl 1,4-cyclohexyldicarboxylates, preferably from dimethyl 1,4-cyclohexyldicarboxylate, have a trans content, based on the position of the

carboxyl groups on the cyclohexyl ring, of greater than or equal to 70 mol%, preferably greater than 80 mol%, and more preferably of greater than 85 mol%. It is possible here to use dialkyl 1,4-cyclohexyldicarboxylates which have any desired trans content.

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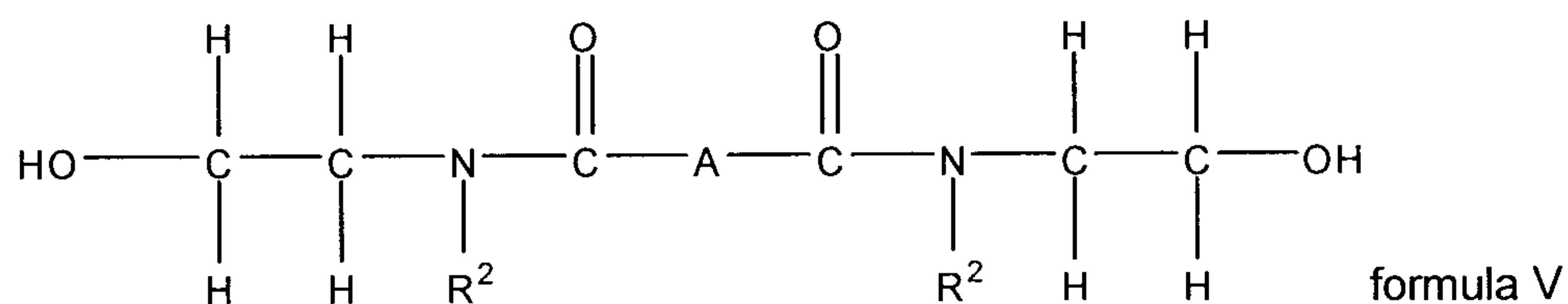
The β -hydroxyalkylamides (I) of the invention are present in solid form below 150°C, preferably below 170°C, more preferably below 180°C.

As byproducts the β -hydroxyalkylamides also contain, to a small extent, dimers, trimers, oligomers, and other condensation products of the target product.

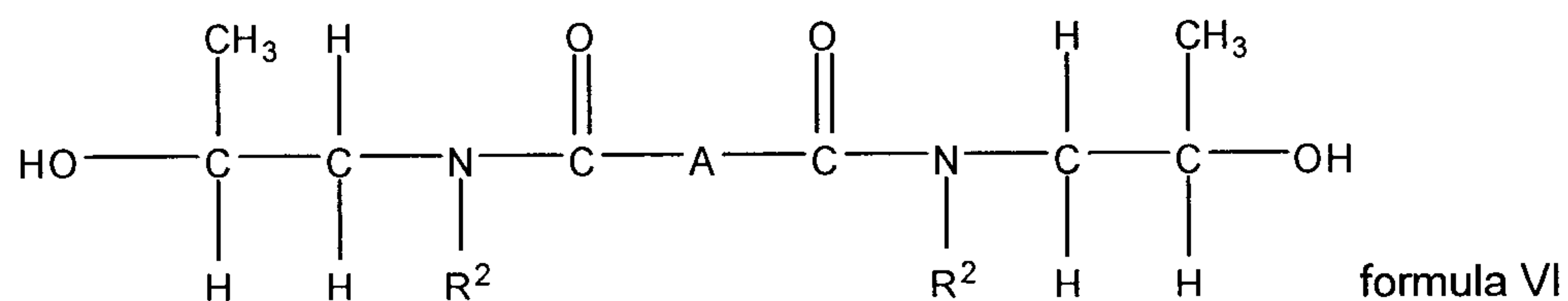
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Particularly preferred β -hydroxyalkylamides of the invention have the following formulae:

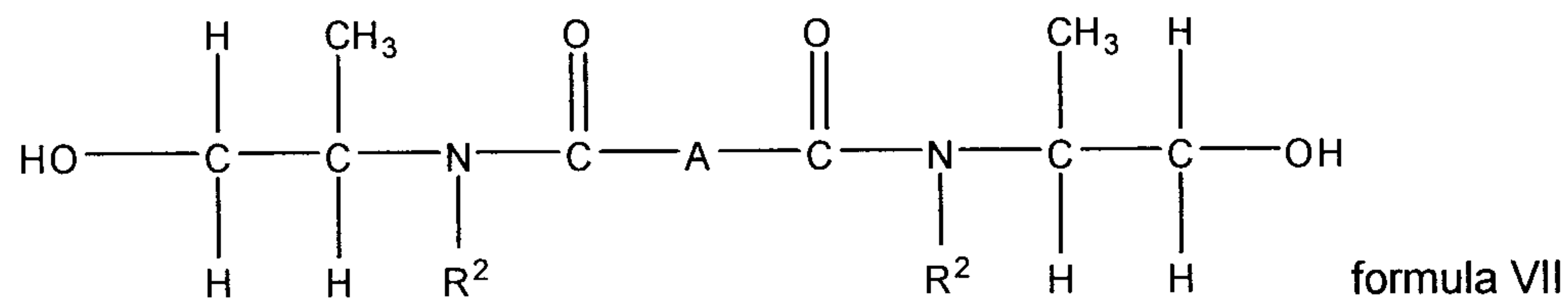
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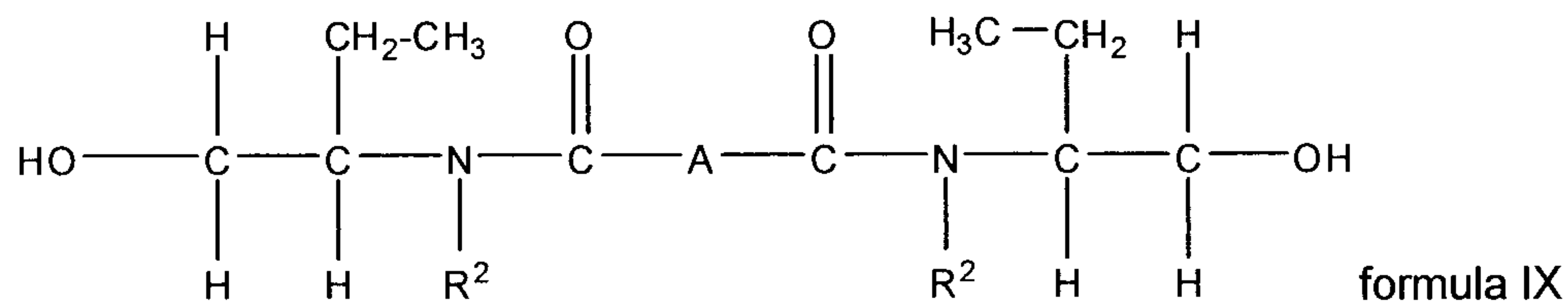
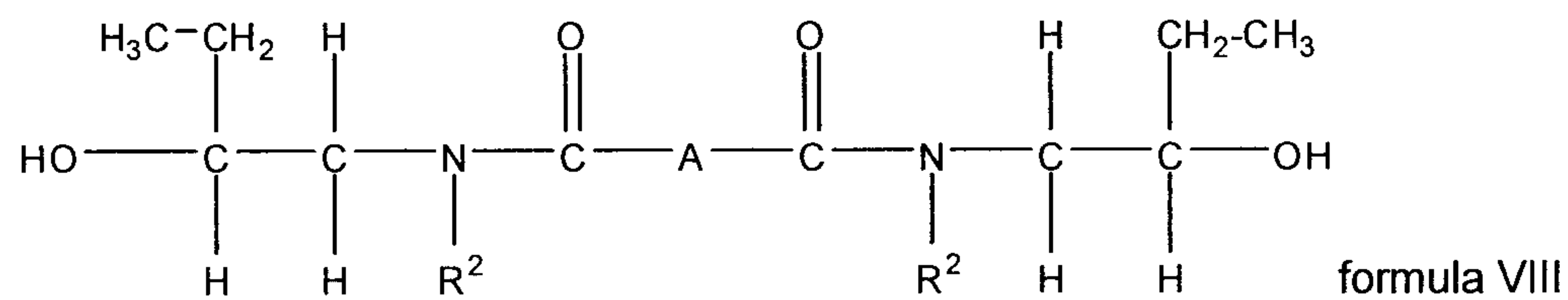
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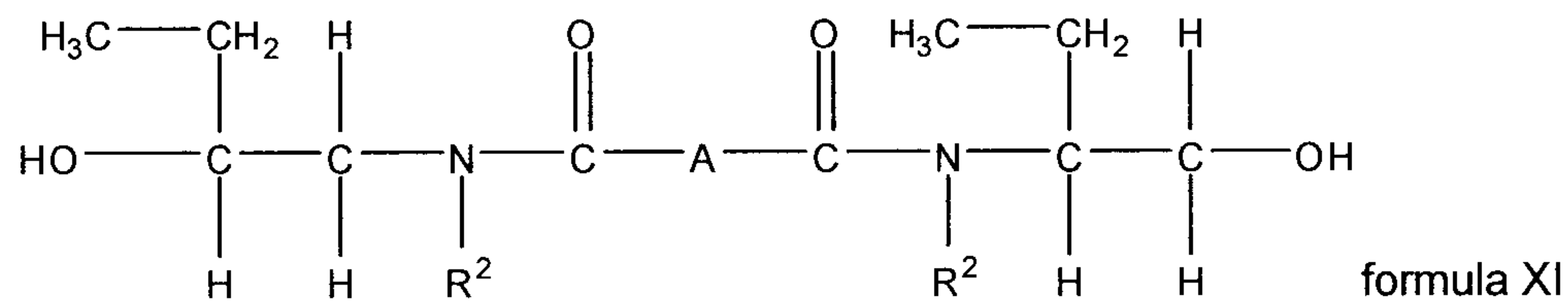
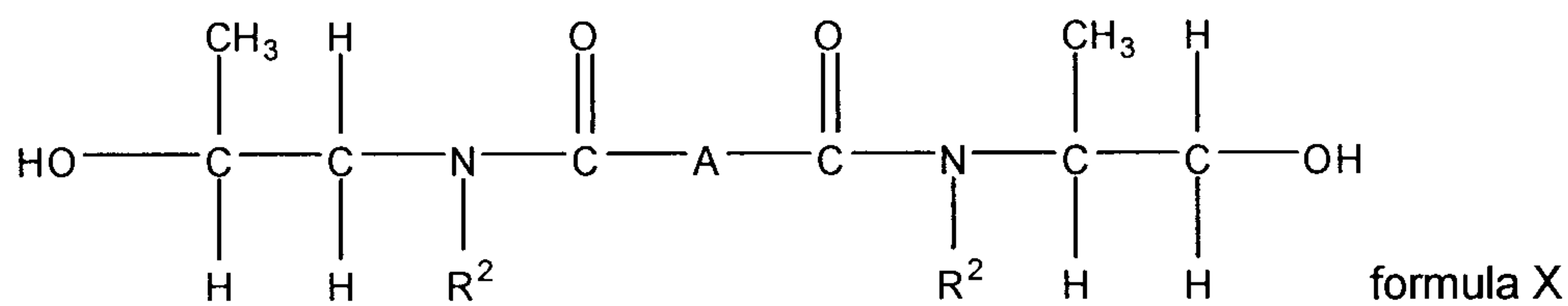
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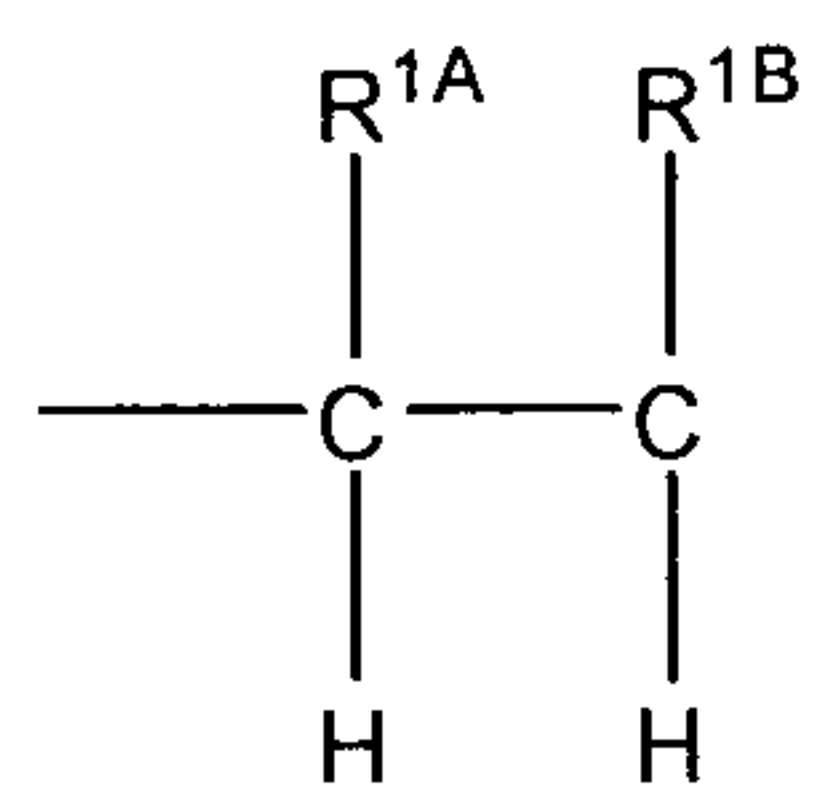
10

where

 R^2 is methyl,

15

or

where $\text{R}^{1\text{A}}$ is hydrogen and $\text{R}^{1\text{B}}$ is methyl, ethyl, propyl,

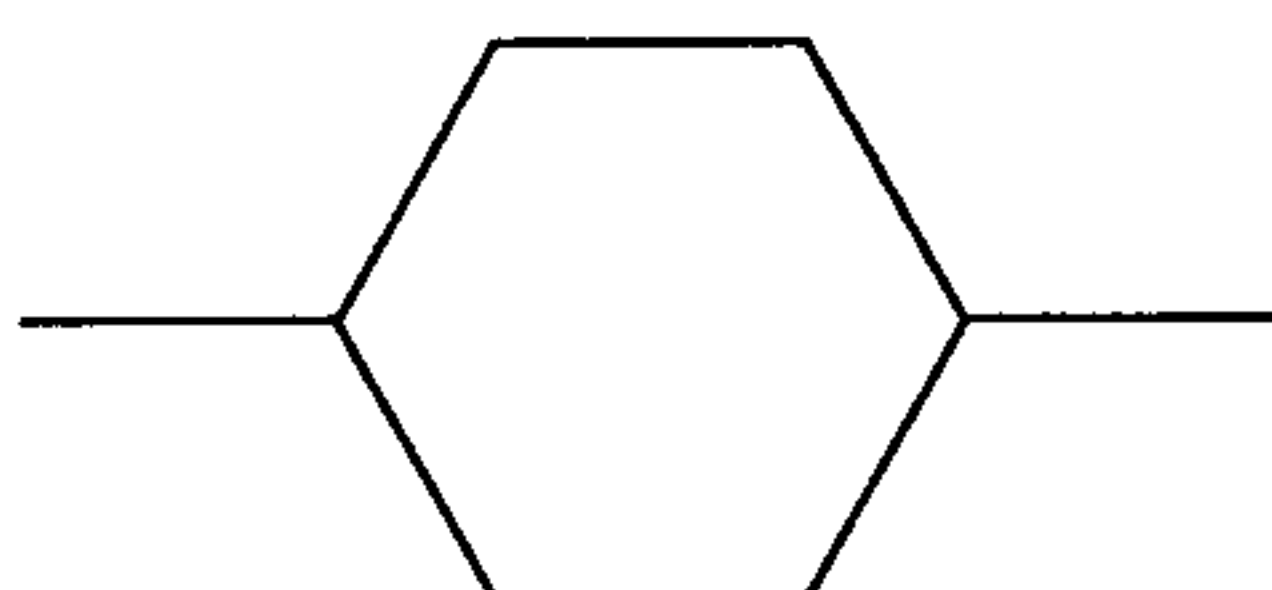
or

20

 $\text{R}^{1\text{A}}$ is methyl, ethyl, propyl and $\text{R}^{1\text{B}}$ is hydrogen;

and

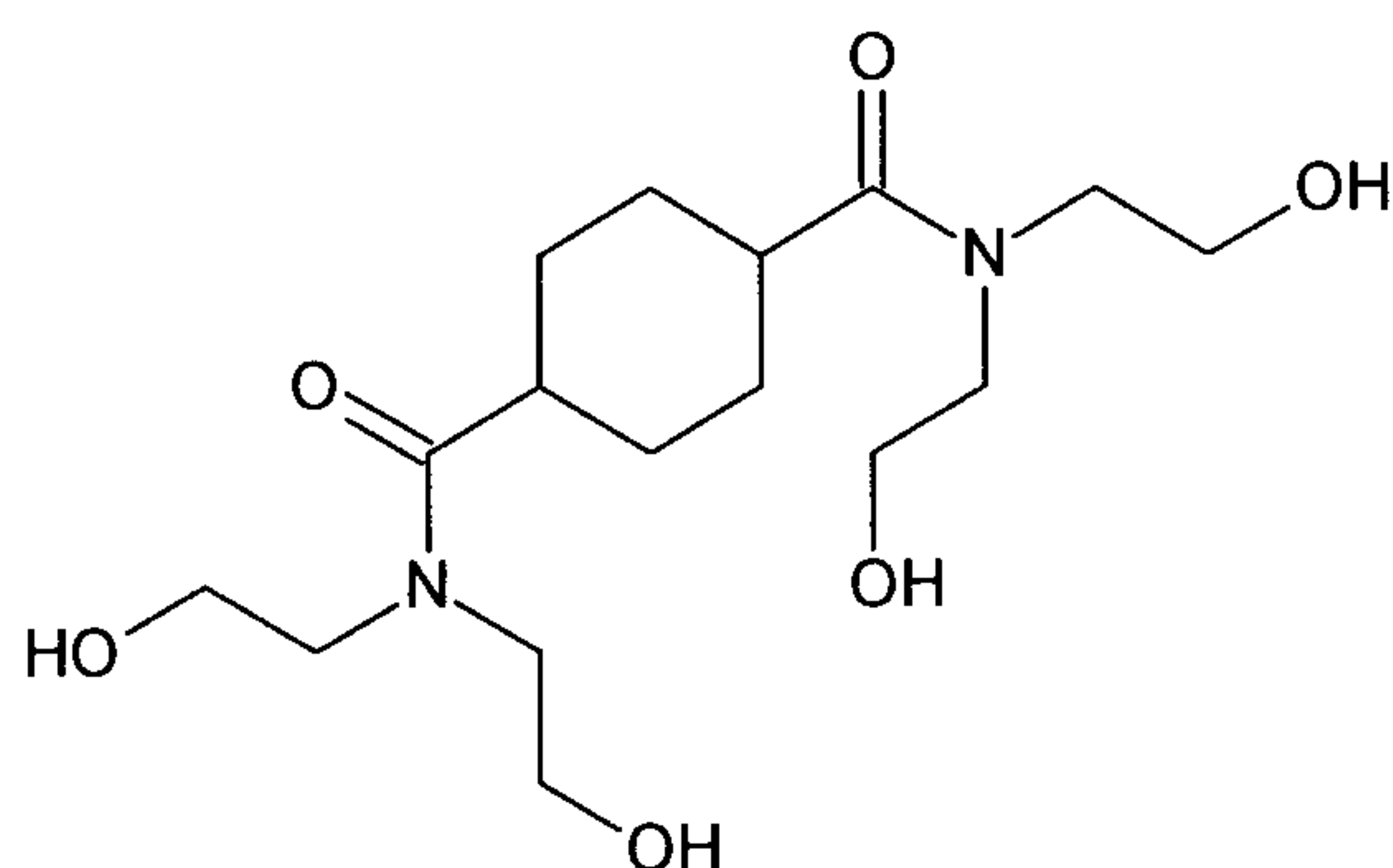
A is a 1,4-disubstituted cyclohexane ring of the formula



where the trans content of A is ≥ 70 mol%;

and where the β -hydroxyalkylamides are present in solid form below 150°C.

The β -hydroxyalkylamide which is particularly preferred in accordance with the invention, formed from dimethyl 1,4-cyclohexyldicarboxylate and diethanolamine, with four β -hydroxyalkylamide groups per molecule of the formula XII



formula XII

has a trans content on the cyclohexyl ring of greater than or equal to 70 mol%, preferably greater than 80 mol%, and more preferably of greater than 85 mol%.

The β -hydroxyalkylamides of the invention can be prepared in principle by known methods, as for example in accordance with DE 25 09 237, DE 198 23 925, EP 473 380, EP 960 878, WO 2000050384, WO 200055266. The method may be carried out continuously, semicontinuously or discontinuously, such as by a batch method, for example.

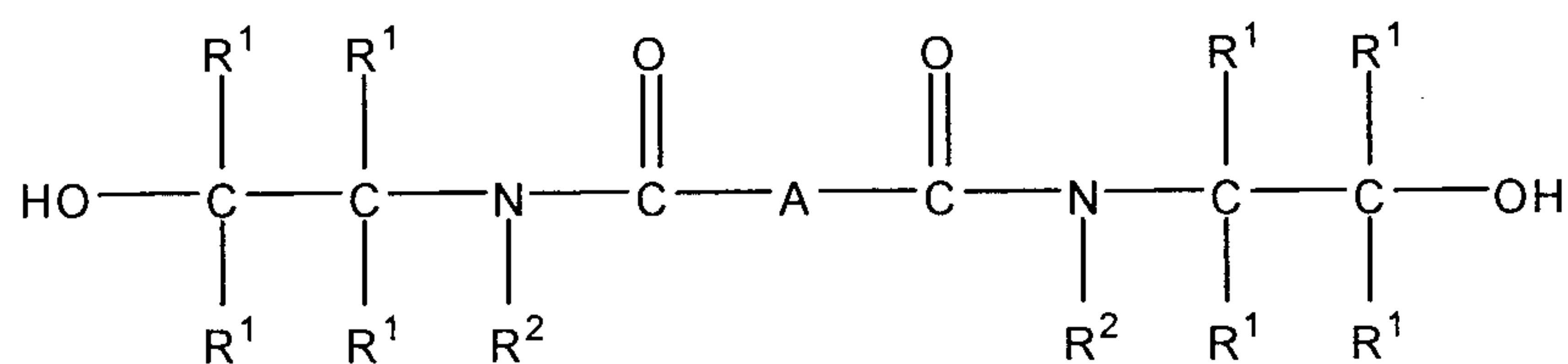
Preferred, however, is the continuous method, described in more detail below, for preparing the β -hydroxyalkylamides from dialkyl 1,4-cyclohexyldicarboxylates.

The invention also relates to a method for the solvent-free, continuous preparation of the preferred β -hydroxyalkylamides of the invention from dialkyl 1,4-cyclohexyldicarboxylates, more particularly from dimethyl 1,4-cyclohexyldicarboxylate, having a trans content on the cyclohexyl ring of greater than or equal to 70 mol%, preferably greater than 80 mol%, and more preferably of greater than 85 mol%, and present in solid form below 150°C, in an extruder, intensive compounder, intensive mixer or static mixer.

It has been found that an accumulation of the trans form to 70 mol% trans on the 1,4-disubstituted cyclohexane ring, or more, takes place, surprisingly, very simply in the preparation of the β -hydroxyalkylamides, by means of a continuous method in an extruder, intensive compounder, intensive mixer or static mixer. It is possible here to use dialkyl 1,4-cyclohexyldicarboxylates which have any desired trans content.

In the dialkyl 1,4-cyclohexyldicarboxylate starting product of the invention that is used, depending on the source of the raw material, the trans configuration is usually between 15 and 35 mol%. However, any desired isomer composition can be used.

The invention accordingly provides a method for the solvent-free and continuous preparation of β -hydroxyalkylamides having at least two or three or four β -hydroxyalkylamide groups per molecule of the formula I

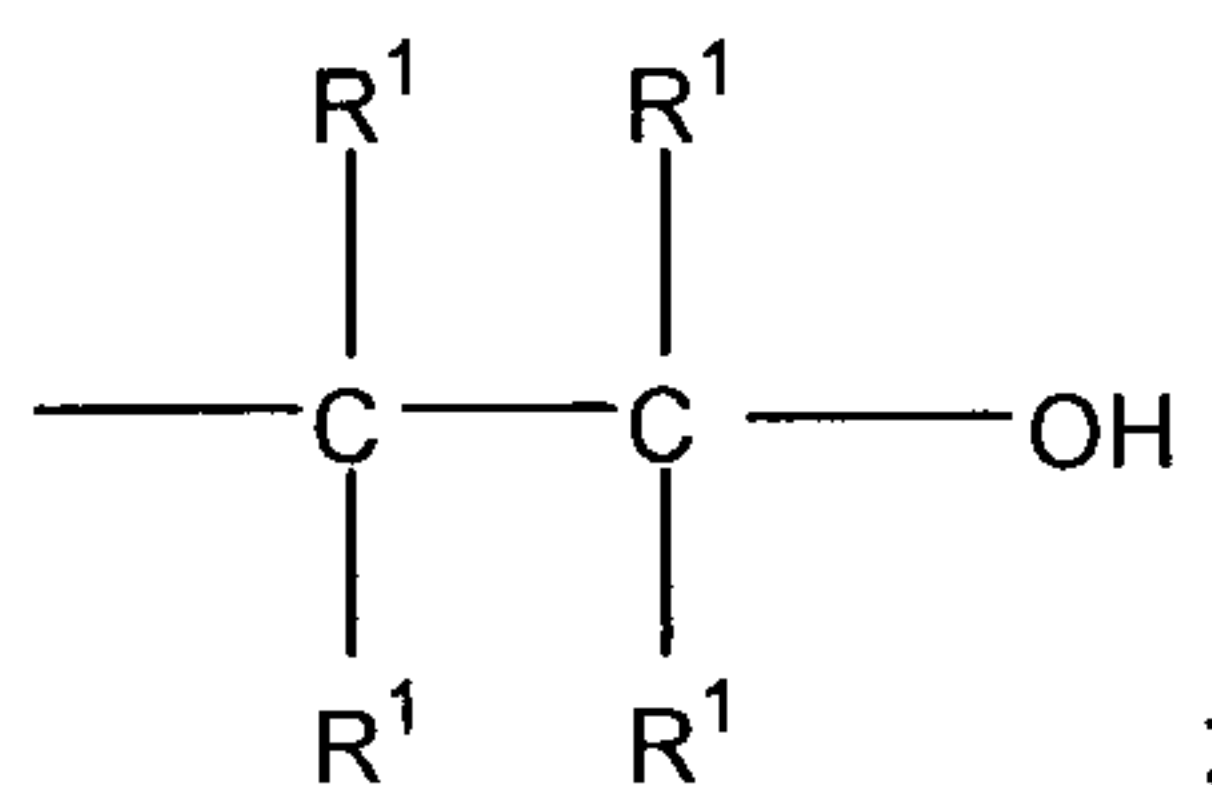


formula I

where

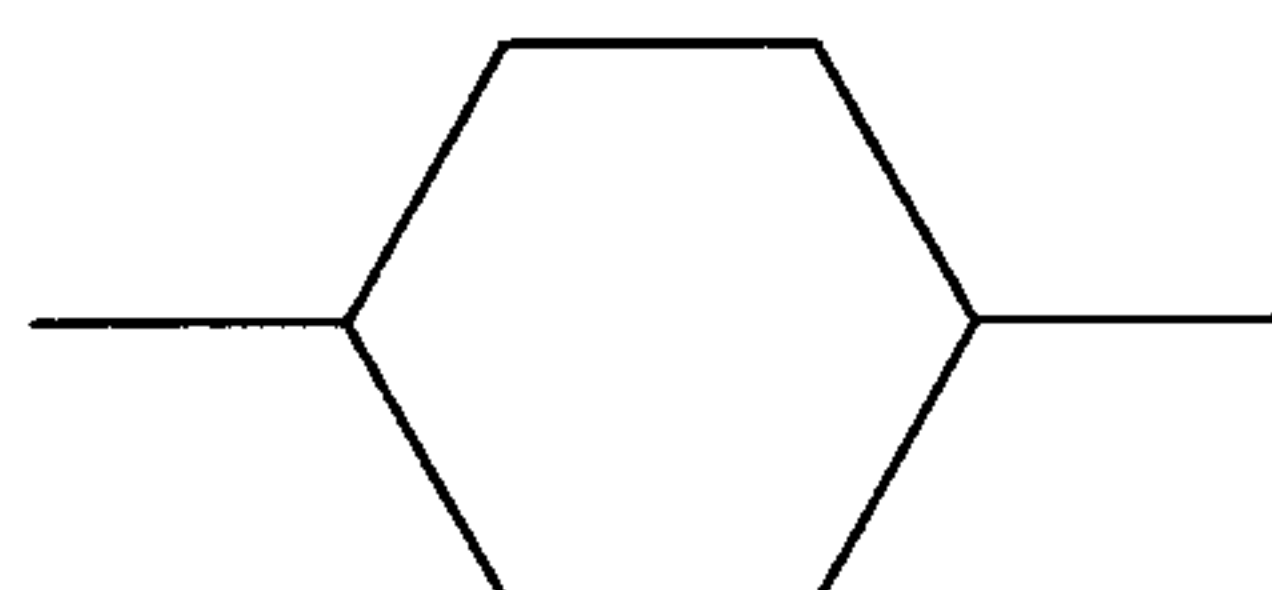
R^1 and R^2 independently of one another are identical or different radicals selected from alkyl radical, cycloalkyl radical, aryl radical, aralkyl radical or alkenyl radical having 1-24 carbon atoms, it also being possible for the radicals to contain heteroatoms and/or functional groups, and it also being possible for R^1 to be hydrogen,

and it also being possible for R² to be



5 and

A is a 1,4-disubstituted cyclohexane ring of the formula



10

where the trans content of A is ≥ 70 mol%;

and

where the β -hydroxyalkylamides (I) are present in solid form below 150°C,
in an extruder, intensive compounder, intensive mixer or static mixer.

15

The principle of the method is that the reaction of the reactants takes place
continuously in an extruder, intensive compounder, intensive mixer or static mixer by
intensive mixing and brief reaction, preferably with supply of heat.

20

In the method it is possible to employ temperatures of 50 to 325°C, with the
temperature varying according to the product, as shown by the examples.

Intensive mixing and brief reaction with supply of heat means that the residence time
of the reactants in the abovementioned assemblies is typically 3 seconds to 15

25

minutes, preferably 3 seconds to 5 minutes, more preferably 5 to 180 seconds.

During this time, the reactants are briefly brought to reaction with supply of heat at
temperatures from 50°C to 325°C, preferably from 50 to 225°C, very preferably from
70 to 200°C. Depending on the nature of the reactants and of the end products,
however, these figures for residence time and temperature may also occupy other
30 preferred ranges.

Optionally, a continuous after-reaction is added on.

The completeness of the reaction is ensured by removal of the alcohols formed in the amidation. This removal is accomplished preferably by the alcohol as being stripped
5 off using reduced pressure via openings in the housings of the extruder or intensive compounder or intensive mixer or static mixer, and/or by passing a gas stream over the intensively combined reaction mixture, with the alcohols, which are more volatile, being carried out by the gas stream.

10 The reaction may be accelerated by catalysts. Suitability is possessed by hydroxides and/or alcoholates of alkali metals, such as sodium or potassium hydroxide, sodium or potassium methanolate, quaternary ammonium hydroxides, alkoxides and/or other strong bases. The concentration is 0.01% to 5%, preferably 0.1% to 3%, based on the total mass employed.

15

The arrangement of reduced-pressure domes and/or gas overpassage locations may be varied and is guided by the nature of the reactants and of the alcohols which are formed. Also possible is an additional location, located downstream of the actual reaction section, for the removal of residual amounts of alcohol.

20

Through subsequent rapid cooling the end product is then obtained.

Assemblies particularly suitable, and preferably used, for the method of the invention are extruders such as single-screw or multiscrew extruders, more particularly twin-
25 screw extruders, planetary roller extruders or annular extruders, (flow tube, intensive compounders, intensive mixers, or static mixers). Particularly preferred are twin-screw or multiscrew extruders, more particularly twin-screw extruders.

It was surprising that the reaction, which in a continuous method takes several hours,
30 proceeds completely within a few seconds in the stated assemblies, and, given suitable catalysis, that the conversion of the cis form to the trans form is also accomplished. A fundamental fact is that brief thermal exposure in conjunction with the mixing effect of the intensive compounder is sufficient for complete or very substantial reaction of the reactants. As a result of appropriate equipping of the
35 mixing chambers and/or compilation of the screw geometries, the intensive compounders allow intensive, rapid mixing with intensive heat exchange at the same

time. Moreover, a uniform flow in the longitudinal direction, with an extremely uniform residence time, is also ensured. Also possible must be different thermal conditioning in the individual barrels or sections of the apparatus.

- 5 The starting products are metered to the assemblies generally in separate product streams. When more than two product streams, they may also be supplied in bundled form. It is also possible to admix this product stream, additionally, with catalysts and/or adjuvants such as flow control agents, stabilizers or adhesion promoters. The streams may also be divided and thus supplied in different fractions
10 at different locations to the assemblies. In this way, concentration gradients are set up in a specific way, allowing the reaction to be taken to completion. The entry location of the product streams can be variable in sequence and staggered in time.

- For preliminary reaction and/or for completion of the reaction it is also possible for
15 two or more assemblies to be combined.

- The assemblies used for the reaction are equipped with reduced-pressure domes, for removal during the reaction of the alcohols formed during the reaction (depending on the carboxylic ester used). This measure serves for completion of the reaction, by
20 shifting the chemical equilibrium in the direction of the desired β -hydroxyalkylamide.

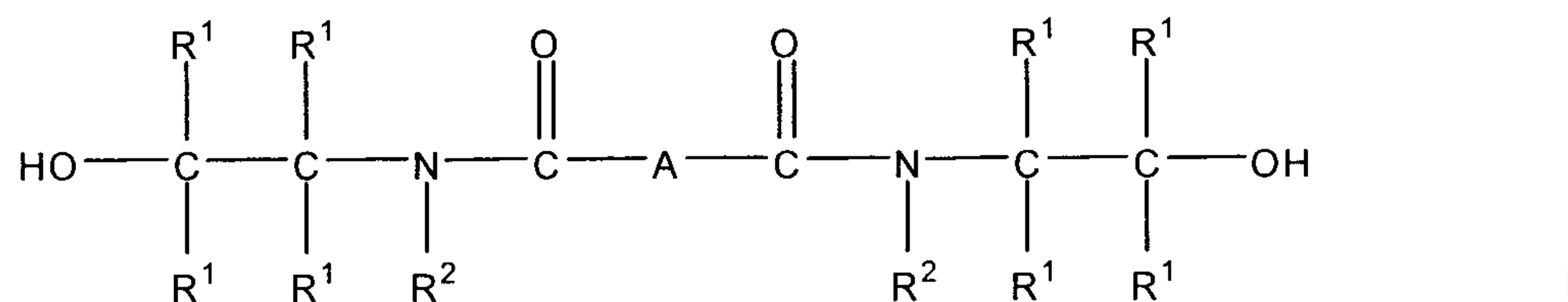
Storage at temperatures above 40°C for between 1 h and four weeks and/or by recrystallization of the product, may improve the product quality.

- 25 Final processing, depending on the viscosity of the product leaving the assembly or the after-reaction zone, is first brought to an appropriate temperature by further cooling using suitable appliances. This is followed by pelletizing or else by comminution to a desired particle size, using roll crushers, hammer mills, cutting mills, classifier mills, pinned-disk mills, flaking rolls or the like.

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The invention also provides the use of the β -hydroxyalkylamides having two or three or four β -hydroxyalkylamide groups per molecule of the formula I

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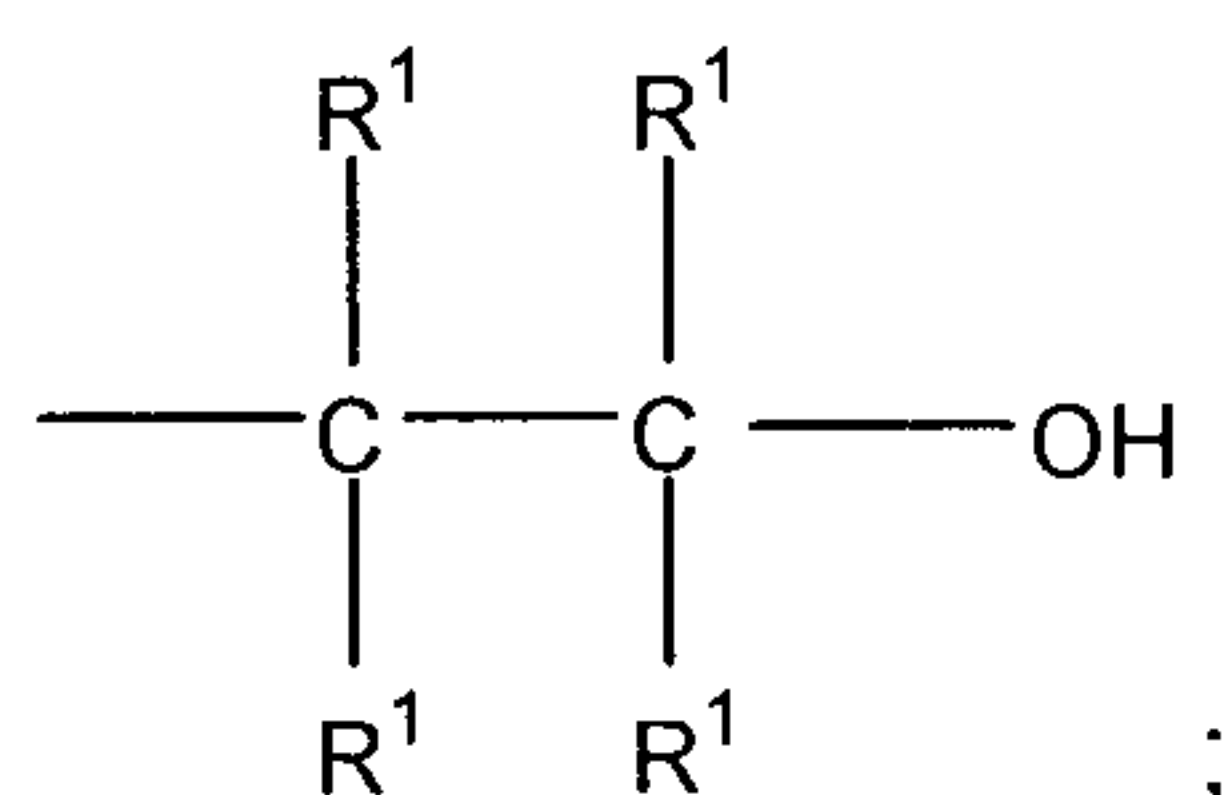
where

R^1 and R^2 independently of one another are identical or different radicals

selected from alkyl radical, cycloalkyl radical, aryl radical, aralkyl radical or alkenyl

5 radical having 1-24 carbon atoms, it also being possible for the radicals to contain heteroatoms and/or functional groups, and it also being possible for R^1 to be hydrogen,

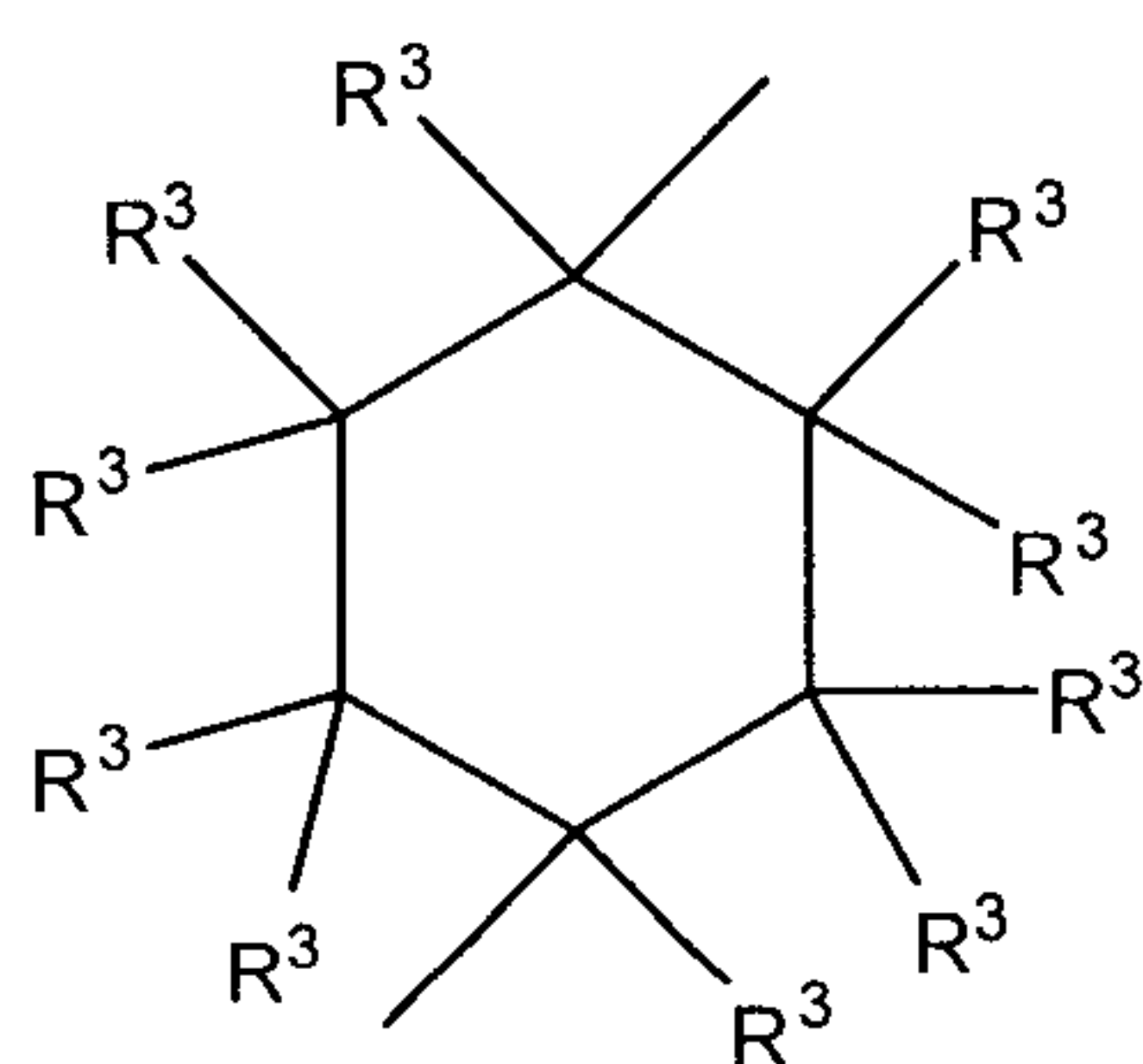
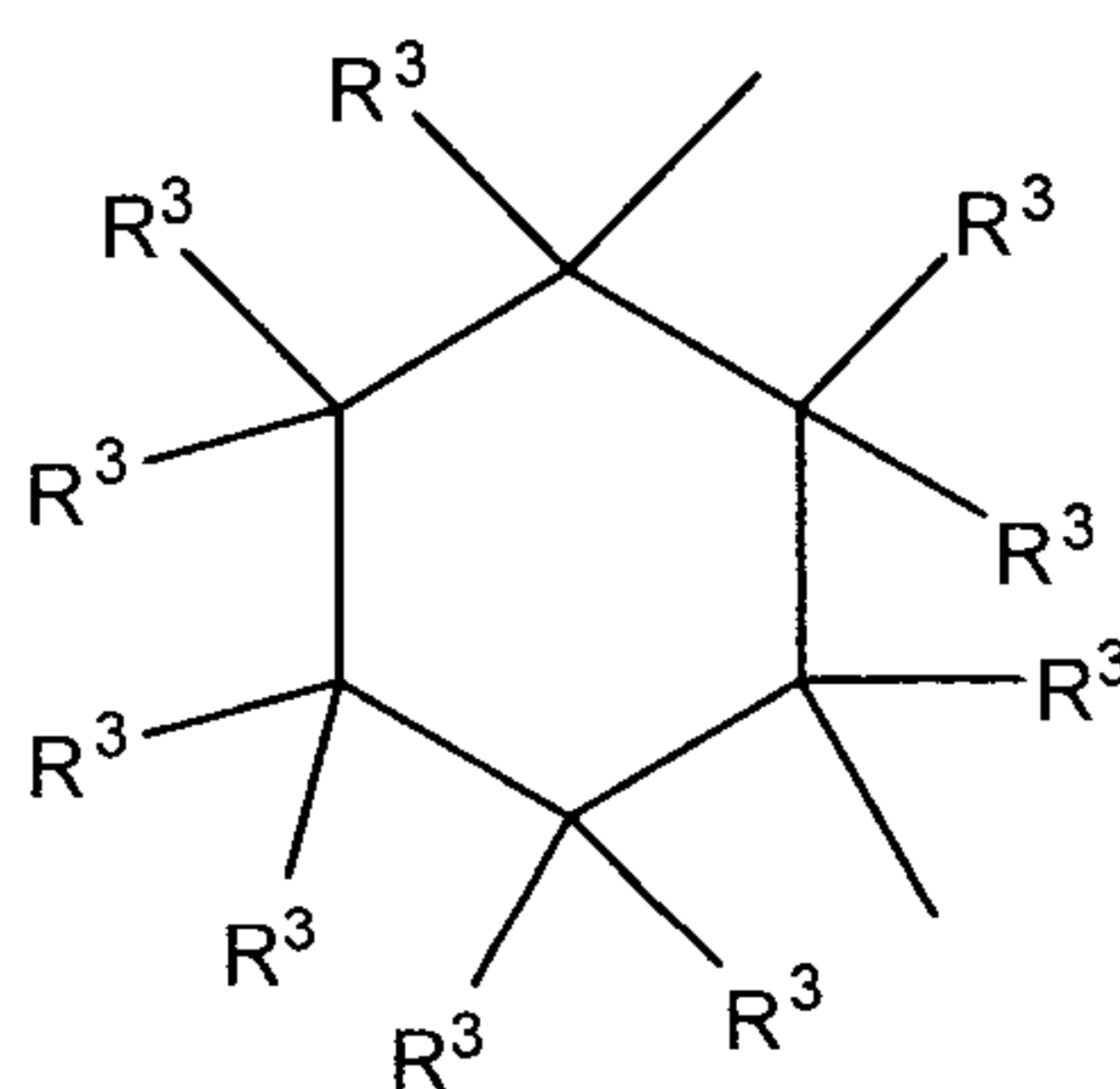
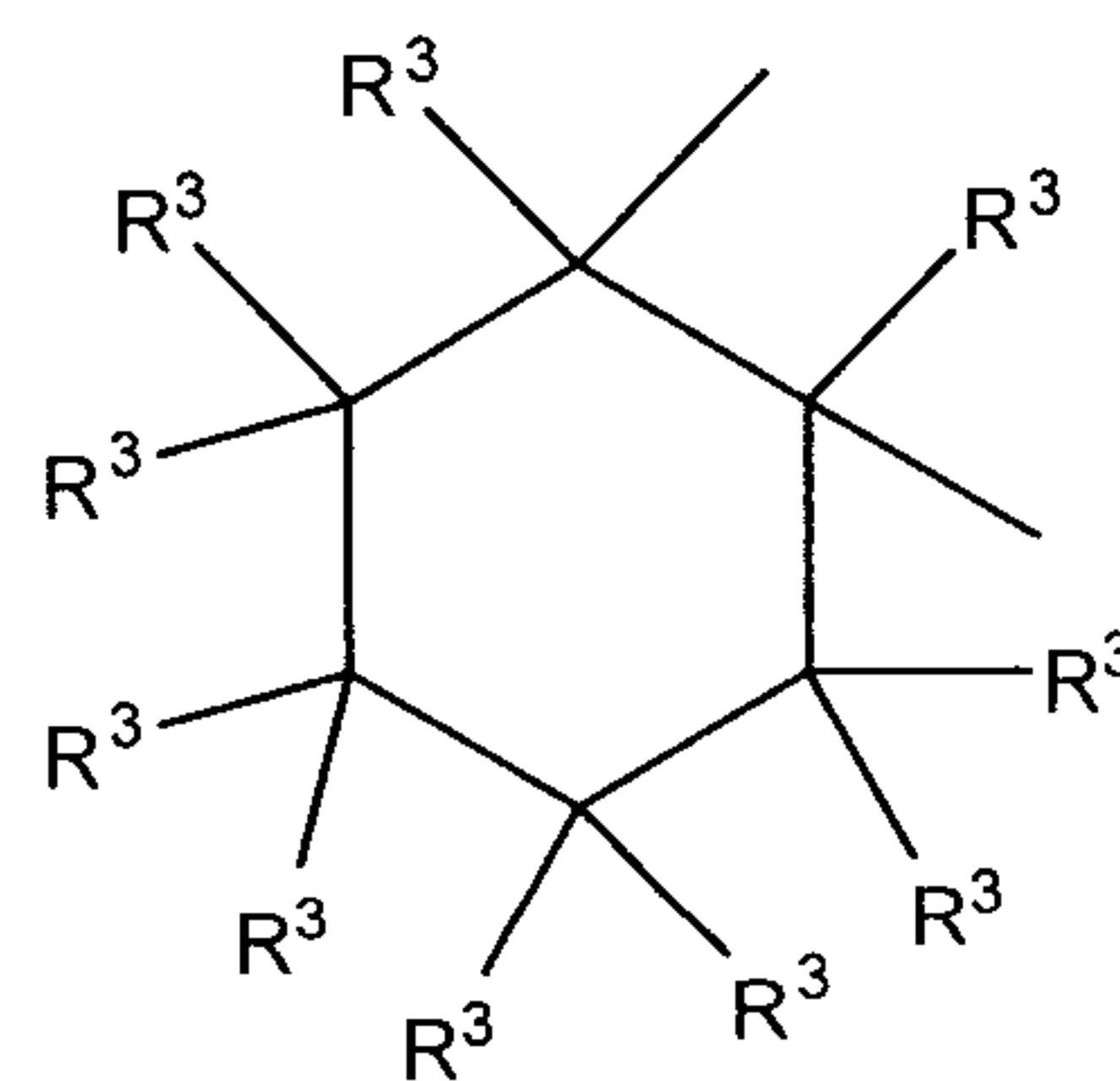
and it also being possible for R^2 to be



10

and

A is

 A^1  A^2  A^3

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where radicals R^3 independently of one another are identical or different radicals selected from hydrogen, alkyl radical, cycloalkyl radical, aryl radical, aralkyl radical or alkenyl radical having 1-24 carbon atoms, it also being possible for the radicals to contain heteroatoms and/or functional groups, and it being possible for two or more substituents R^3 to be linked with one another to form rings;

20

where the β -hydroxyalkylamides are present in solid form below 150°C,

as crosslinkers for polymers containing carboxyl groups, preferably for polyesters containing carboxyl groups.

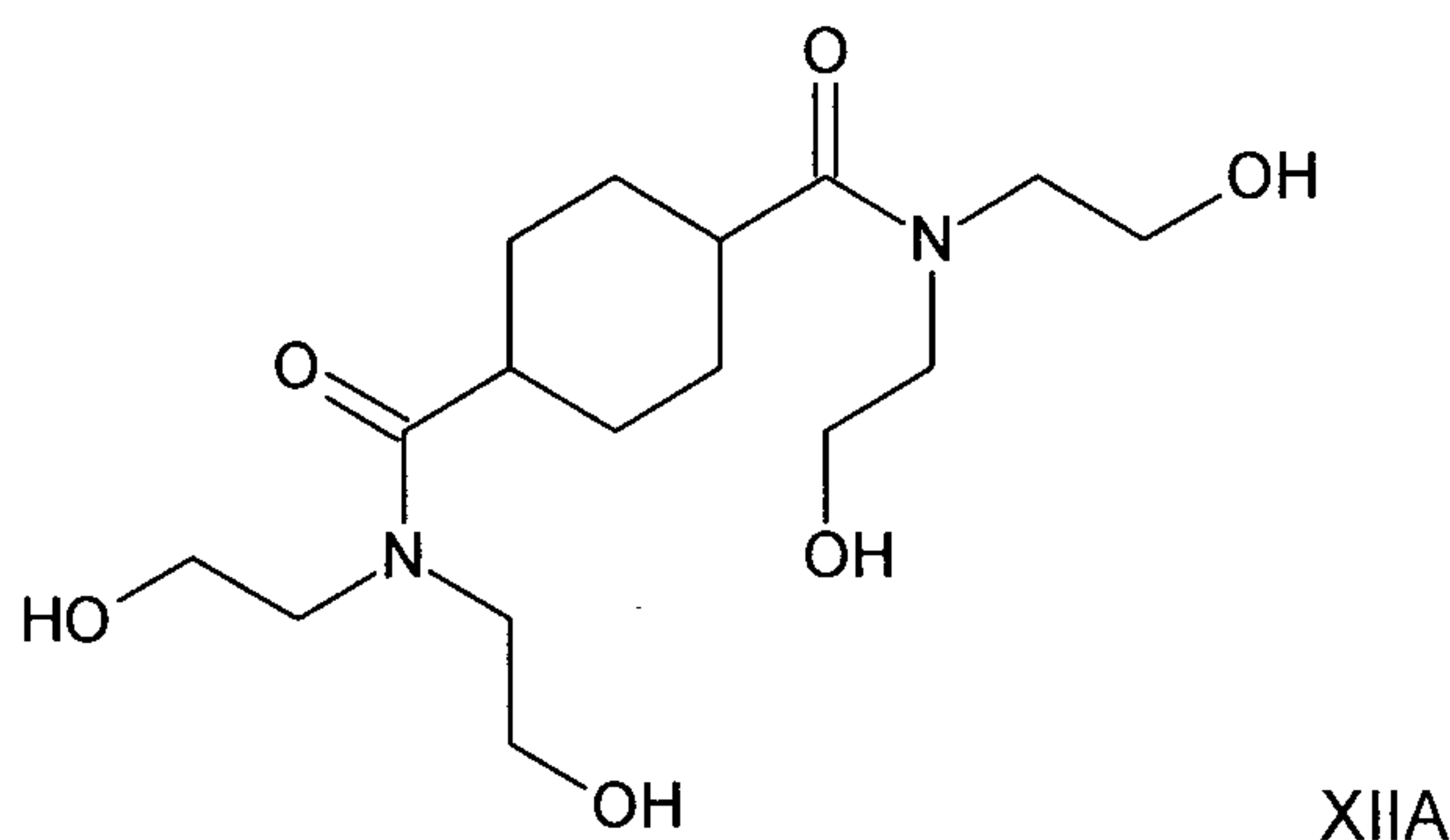
The invention also provides the use of the β -hydroxyalkylamides of the invention in powder coating materials, preferably in carboxyl-containing polyester powder coating materials.

5

The invention also provides the use of the β -hydroxyalkylamides of the invention in powder coating materials which after curing have matt surfaces, with < 50 gloss units, measured as reflectometer values according to DIN 67530 / ISO 2813 at an incident angle of 60°.

10

Provided preferably by the invention is the compound N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to the formula XIIA



15

which has the following parameters:

1. a trans content on the cyclohexyl ring of greater than or equal to 70 mol%, based on the total amount of all of the isomers of N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide that are present,

20

and

2. two endothermic peaks according to DSC analysis (differential scanning calorimetry), where peak 1 is situated in the region of 140 - 170°C, with a maximum of 155 - 165°C, and peak 2 is situated in the region of 170 - 210°C, with a maximum of 175 - 207°C,

25

and

3. the ratio of the enthalpies of endothermic peak 1 to endothermic peak 2 is 1:1 to 1:5,

and

4. the XRPD spectrum of the powder sample in an x-ray diffractometer, measured with Cu K α radiation (1.541 Å) has the following peaks:

30

	Peak #	Degrees 2theta	d (Å)
		± 0.2 degree 2theta	
5	1	14.90	5.94
	2	16.70	5.31
	3	17.40	5.09
	4	21.20	4.19
	5	21.60	4.11
	6	26.00	3.43

10

Provided more preferably by the invention is the β -hydroxyalkylamide N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to the formula XIIA, having a trans content on the cyclohexyl ring of greater than or equal to 70 mol%, preferably greater than 80 mol%, and more preferably of greater than 85 mol%, based on the total amount of all of the isomers of N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide that are present.

Additionally, this β -hydroxyalkylamide of the invention, N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to the formula XIIA, has two endothermic peaks by DSC analysis (differential scanning calorimetry): first a peak having a maximum (peak 1) of about 160°C, and a further, second peak having a maximum (peak 2) at about 190°C – see the figures for the examples. Preferably, the first peak is located in the region of 140 - 170°C, with a maximum of 155 - 165°C, and the second peak is located in the region of 170 - 210°C, with a maximum of 175 - 207°C. More preferably, the first peak is located in the region of 155 - 170°C, with a maximum of 158 - 165°C, and the second peak is located in the region of 170 - 210°C, with a maximum of 180 - 205°C.

The ratio of the enthalpies of endothermic peak 1 (~160°C) to endothermic peak 2 (~190°C) may be 1:1 to 1:5, preferably 1:1 to 1:3.

The DSC measurements were carried out in accordance with DIN EN ISO 11357-1 from March 2010. A heat flow difference calorimeter from the manufacturer Mettler-Toledo, model DSC 821, was used. The samples are run once from -30°C to 250°C at 10 K/min.

The XRPD measurements on powder samples were carried out in an x-ray diffractometer using Cu K α radiation (1.541 Å). In accordance with figure 9, the

following significant and characteristic 6 peaks were found for the β -hydroxyalkylamide N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to the formula XIIA:

5 10	Peak #	Degrees 2theta ± 0.2 degree 2theta	d (Å)
	1	14.90	5.94
	2	16.70	5.31
	3	17.40	5.09
	4	21.20	4.19
	5	21.60	4.11
	6	26.00	3.43

Especially preferred is the β -hydroxyalkylamide N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to the formula XIIA having a trans content on the cyclohexyl ring of greater than or equal to 92 mol%, preferably greater than 94 mol%, and very preferably of greater than 96 mol%, and especially preferably of greater than 98 mol%, based on the total amount of all of the isomers of N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide that are present.

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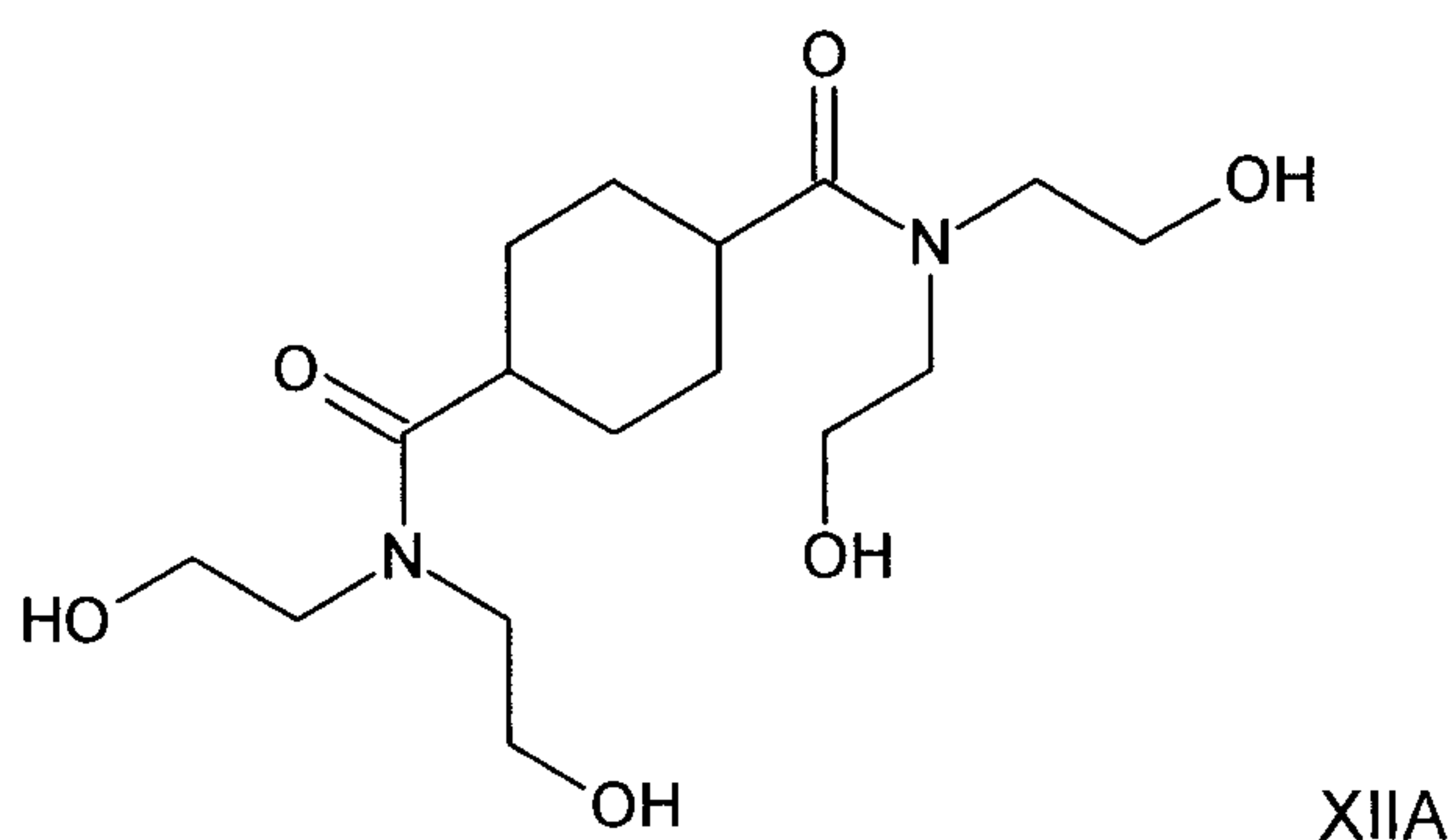
The β -hydroxyalkylamide of the invention, of the formula XIIA, is present in solid form below 175°C, preferably below 180°C, and more preferably of below 185°C.

The β -hydroxyalkylamide of the invention, of the formula XIIA, having features 1. to 4. was investigated by x-ray structural analysis of a single crystal. Comprehensive details relating to the measurement are summarized in annex 1. The x-ray structural analysis of a single crystal gave the following result for the structure:

30	Crystal system:	Orthorhombic	
	Space group:	Pbca	
35	Unit cell dimensions:	a = 10.06350(10) Å	$\alpha = 90^\circ$.
		b = 11.85290(10) Å	$\beta = 90^\circ$.
		c = 14.6275(2) Å	$\gamma = 90^\circ$.
		Volume:	1744.79(3) Å ³

The figures in the brackets indicate the measurement accuracy in plus and minus in each case for the corresponding last digit or last two digits.

- 5 The invention also provides the compound N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to the formula XIIA



- 10 which has the following parameters:
1. a trans content on the cyclohexyl ring of greater than or equal to 70 mol%, based on the total amount of all of the isomers of N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide that are present,
- and
- 15 2. two endothermic peaks according to DSC analysis (differential scanning calorimetry), where peak 1 is situated in the region of 140 - 170°C, with a maximum of 155 - 165°C, and peak 2 is situated in the region of 170 - 210°C, with a maximum of 175 - 207°C,
- and
- 20 3. the ratio of the enthalpies of endothermic peak 1 to endothermic peak 2 is 1:1 to 1:5,
- and
4. the XRPD spectrum of the powder sample in an x-ray diffractometer, measured with Cu K α radiation (1.541 Å) has the following peaks:

25

Peak #	Degrees 2theta ± 0.2 degree 2theta	d (Å)
1	14.90	5.94
2	16.70	5.31

3	17.40	5.09
4	21.20	4.19
5	21.60	4.11
6	26.00	3.43

5

5. and which, according to x-ray structural analysis of a single crystal, has the following parameters:

	Crystal system:	Orthorhombic	
10	Space group:	Pbca	
	Unit cell dimensions:	a = 10.06350(10) Å	$\alpha = 90^\circ$.
		b = 11.85290(10) Å	$\beta = 90^\circ$.
		c = 14.6275(2) Å	$\gamma = 90^\circ$.
15	Volume:	1744.79(3) Å ³	

Preparation

The particularly preferred N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to the formula XIIA is obtainable by various methods:

- 20 First of all, as described precisely earlier on above, the N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to the formula XIIA is prepared, preferably solventlessly, in an extruder, intensive compounder, intensive mixer or static mixer, preferably in an extruder. Temperatures of 100 to 180°C are employed here. This is followed by recrystallization from a suitable solvent, preferably water.
- 25 Following dissolution at temperatures of 20 – 100°C and crystallization, the N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to the formula XIIA with the above-stated parameters is obtained. It can then, subsequently, be washed with alcohols, preferably methanol, and dried. Drying takes place preferably at temperatures of 20-90°C, and may also take place under reduced pressure.

30

- A further variant of the preparation takes place as described precisely earlier on above, by the N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to the formula XIIA being prepared, preferably solventlessly, in an extruder, intensive compounder, intensive mixer or static mixer, preferably in an extruder. In this case,
- 35 temperatures of 100 to 180°C are employed. This is followed by thermal conditioning at temperatures of 50-100°C, preferably at temperatures of 70-85°C. The time is

more than 6 hours, preferably more than 12 hours. Thermal conditioning may take place under reduced pressure.

Also provided by the invention, accordingly, is a method for the solvent-free, continuous preparation of N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to the formula XIIA from dimethyl 1,4-cyclohexyldicarboxylate and diethanolamine, having four β -hydroxyalkylamide groups per molecule, which has the following parameters:

1. a trans content on the cyclohexyl ring of greater than or equal to 70 mol%, based on the total amount of all of the isomers of N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide that are present, and
2. two endothermic peaks according to DSC analysis (differential scanning calorimetry), where peak 1 is situated in the region of 140 - 170°C, with a maximum of 155 - 165°C, and peak 2 is situated in the region of 170 - 210°C, with a maximum of 175 - 207°C, and
3. the ratio of the enthalpies of endothermic peak 1 to endothermic peak 2 is 1:1 to 1:5, and
4. the XRPD spectrum of the powder sample in an x-ray diffractometer, measured with Cu K α radiation (1.541 Å) has the following peaks:

Peak #	Degrees 2theta	
	± 0.2 degree 2theta	d (Å)
1	14.90	5.94
2	16.70	5.31
3	17.40	5.09
4	21.20	4.19
5	21.60	4.11
6	26.00	3.43

in an extruder, intensive compounder, intensive mixer or static mixer;

- a) and recrystallization

b) or thermal conditioning at temperatures of 50 – 100°C, the time being more than 6 hours,
of the resultant product.

- 5 The particularly preferred N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to the formula XIIA may also take place discontinuously in solvent, in other words in a batch method.

The reaction is carried out in customary reactors. Operation here may be
10 unpressurized, using a reflux condenser, or under pressure, with a closed reactor. The synthesis is carried out in a solvent, preferably in alcohols, preferably methanol. The amount of solvent added is greater than 10% by weight, preferably greater than 15% by weight, based on the total amount of all of the reactants (starting materials) employed. It is possible here to operate under reflux, or else at lower temperatures,
15 and also higher temperatures, under pressure. Preparation takes place at temperatures of 20 to 120°C, preferably at 60 to 90°C, more preferably at 70 to 85°C. Crystallization gives the N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to the formula XIIA having the parameters stated above.

- 20 The invention also relates to a method for the discontinuous preparation of N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to the formula XIIA from dimethyl 1,4-cyclohexyldicarboxylate and diethanolamine, having four β -hydroxyalkylamide groups per molecule,

25 which has the following parameters:

1. a trans content on the cyclohexyl ring of greater than or equal to 70 mol%, based on the total amount of all of the isomers of N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide that are present,
30 and
2. two endothermic peaks according to DSC analysis (differential scanning calorimetry), where peak 1 is situated in the region of 140 - 170°C, with a maximum of 155 – 165°C, and peak 2 is situated in the region of 170 – 210°C, with a maximum of 175 – 207°C,
35 and

3. the ratio of the enthalpies of endothermic peak 1 to endothermic peak 2 is 1:1 to 1:5,

and

4. the XRPD spectrum of the powder sample in an x-ray diffractometer, measured with Cu K α radiation (1.541 Å) has the following peaks:

	Peak #	Degrees 2theta	d (Å)
		± 0.2 degree 2theta	
10	1	14.90	5.94
	2	16.70	5.31
	3	17.40	5.09
	4	21.20	4.19
	5	21.60	4.11
	6	26.00	3.43

15

in solvent.

The preparation of the N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to the formula XIIA may also take place in closed apparatus under pressure at temperatures of 60 to 140°C, without addition of solvents, and this is likewise provided by the invention.

The N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to the formula XIIA that is thus prepared in a batch method can be recrystallized from suitable solvents, preferably from water or alcohols, preferably from methanol.

Moreover, the preparation of the N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to the formula XIIA may also take place discontinuously without solvents.

The reaction is carried out in customary reactors. It is possible here to operate using a reflux condenser. The preparation takes place preferably at temperatures of 20 to 140°C, preferably 60 to 90°C, more preferably at 70 to 85°C. The β -hydroxyalkylamide obtained thus in a batch method is then recrystallized from suitable solvents, preferably from water or alcohols, preferably from methanol.

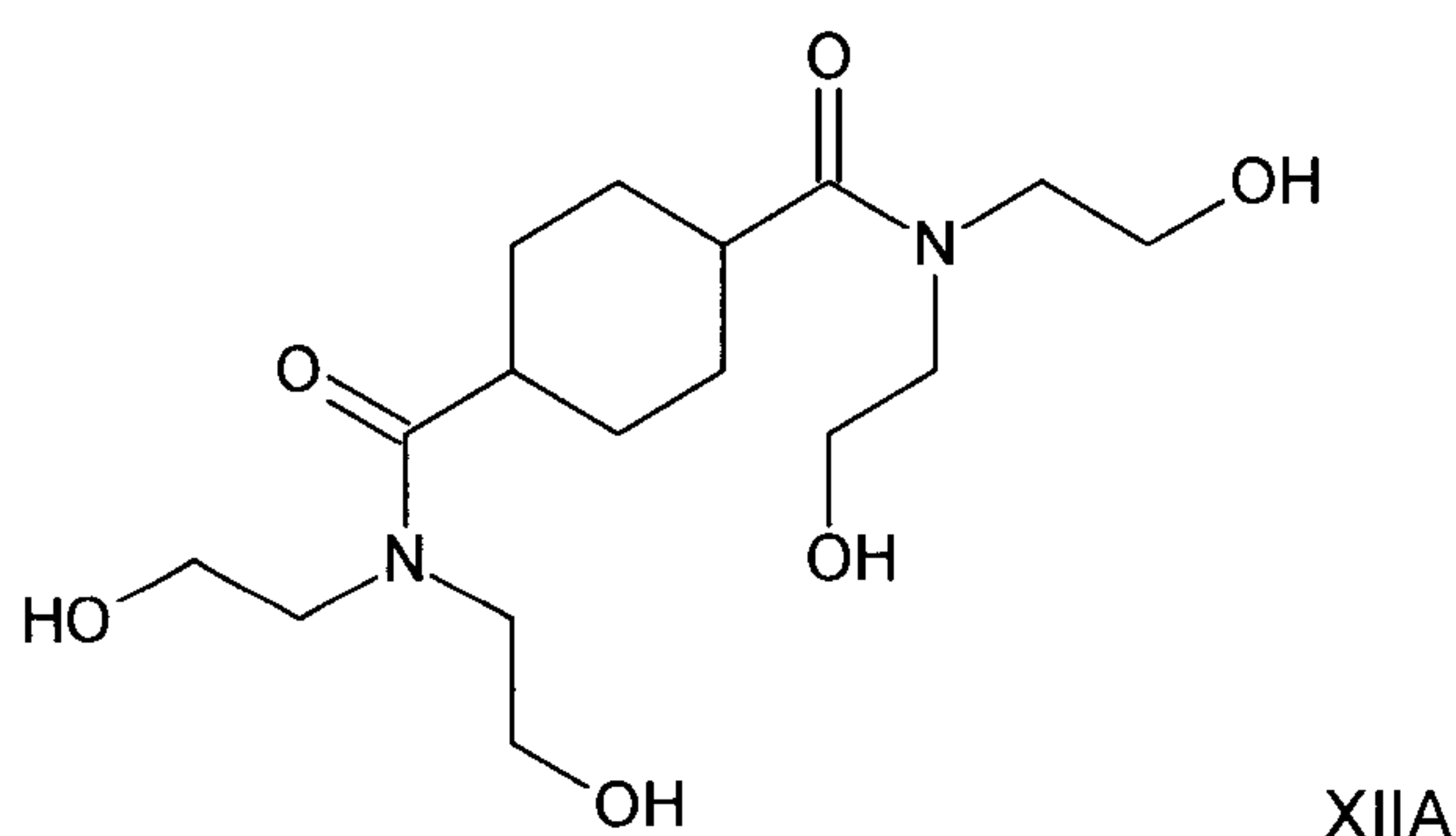
Crystallization gives the N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide

according to the formula XIIA with the parameters specified above. This method is likewise provided by the invention.

The concentration of all of the isomers of N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide in the end product after its preparation is 75% by mass, preferably 80% by mass, and more preferably 85% by mass.

This β -hydroxyalkylamide as described and characterized here, N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide of the formula XIIA, produces far-reaching matting in powder coating materials, with a gloss of less than 50 scale divisions at the 60° angle, as has been shown in the examples. This product of the formula XIIA therefore differs clearly from the disclosed β -hydroxyalkylamide according to laid-open specification KR 10-2009-0111720 (and that from β -hydroxyalkylamide Korean Ind. Eng. Chem., vol. 20, No. 2, April 2009, 195-200), as demonstrated there in fig. 2 on page 15, having only one peak, according to DSC analysis, at about 190°C, and, as shown by comparative example 4c, not leading to coatings having matt surfaces.

The invention also provides the use of N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to the formula XIIA



which has the following parameters:

1. a trans content on the cyclohexyl ring of greater than or equal to 70 mol%, based on the total amount of all of the isomers of N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide that are present, and
2. two endothermic peaks according to DSC analysis (differential scanning calorimetry), where peak 1 is situated in the region of 140 - 170°C, with a

maximum of 155 – 165°C, and peak 2 is situated in the region of 170 – 210°C, with a maximum of 175 – 207°C,

and

3. the ratio of the enthalpies of endothermic peak 1 to endothermic peak 2 is 1:1

5 to 1:5,

and

4. the XRPD spectrum of the powder sample in an x-ray diffractometer, measured with Cu K α radiation (1.541 Å) has the following peaks:

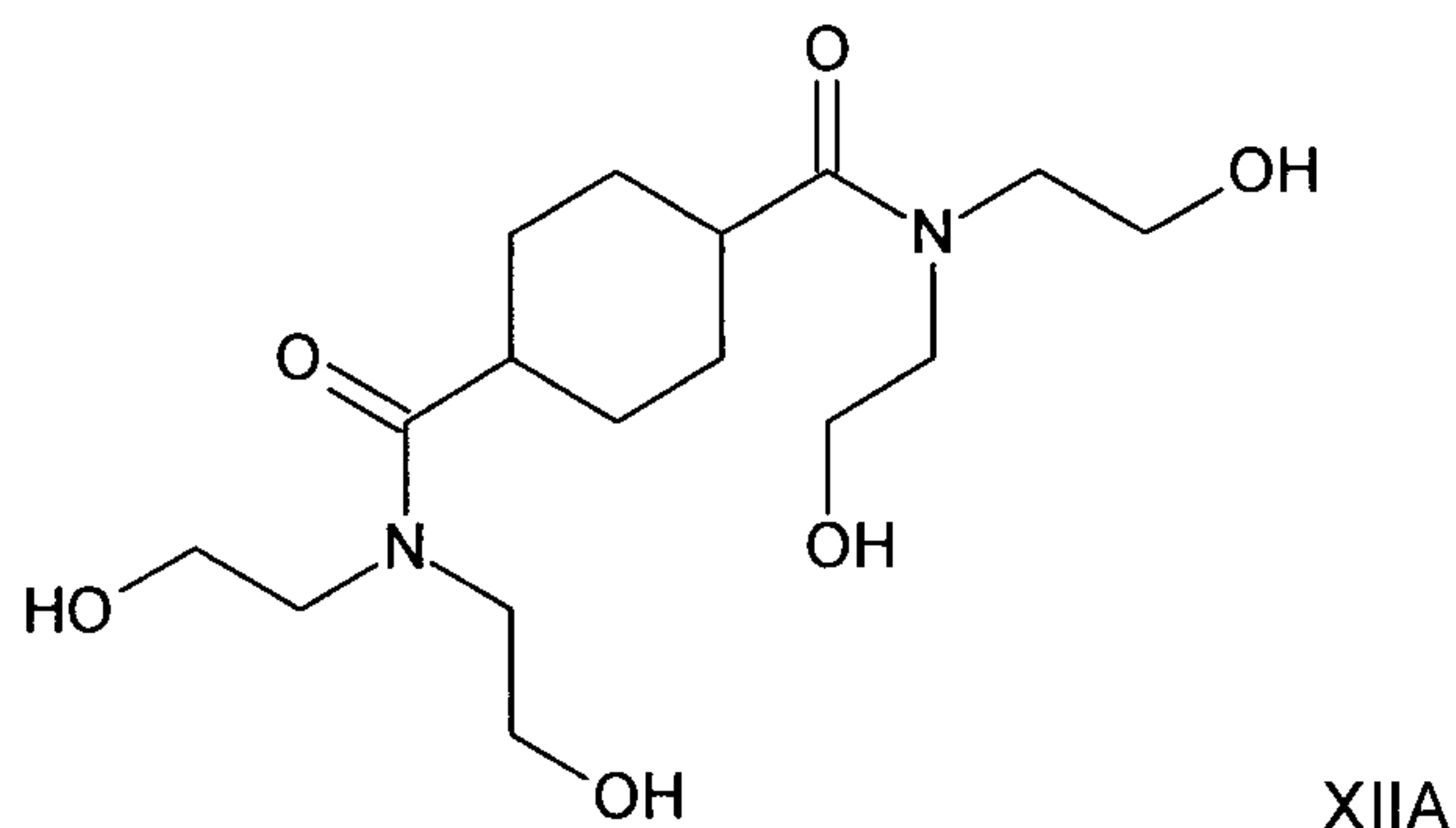
10	Peak #	Degrees 2theta	d (Å)
		± 0.2 degree 2theta	
	1	14.90	5.94
	2	16.70	5.31
	3	17.40	5.09
15	4	21.20	4.19
	5	21.60	4.11
	6	26.00	3.43

for producing coatings having matt surfaces, more particularly in powder coating
20 materials, preferably in carboxyl-containing polyester powder coating materials.

The invention also provides the use of N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to the formula XIIA for producing coatings having matt surfaces with < 50 gloss units, measured as reflectometer values according to
25 DIN 67530 / ISO 2813 at an incident angle of 60°.

The invention also provides the use of N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to the formula XIIA

25



which has the following parameters:

1. a trans content on the cyclohexyl ring of greater than or equal to 70 mol%,
based on the total amount of all of the isomers of N,N,N',N'-tetrakis(2-
hydroxyethyl)cyclohexyl-1,4-diamide that are present,
and
2. two endothermic peaks according to DSC analysis (differential scanning
calorimetry), where peak 1 is situated in the region of 140 - 170°C, with a
maximum of 155 – 165°C, and peak 2 is situated in the region of 170 –
210°C, with a maximum of 175 – 207°C,
and
3. the ratio of the enthalpies of endothermic peak 1 to endothermic peak 2 is 1:1
to 1:5,
and
4. the XRPD spectrum of the powder sample in an x-ray diffractometer,
measured with Cu K α radiation (1.541 Å) has the following peaks:

Peak #	Degrees 2theta \pm 0.2 degree 2theta	d (Å)
1	14.90	5.94
2	16.70	5.31
3	17.40	5.09
4	21.20	4.19
5	21.60	4.11
6	26.00	3.43

5. and which, according to x-ray structural analysis of a single crystal, has the
following parameters:

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26

Crystal system:

Orthorhombic

Space group:

Pbca

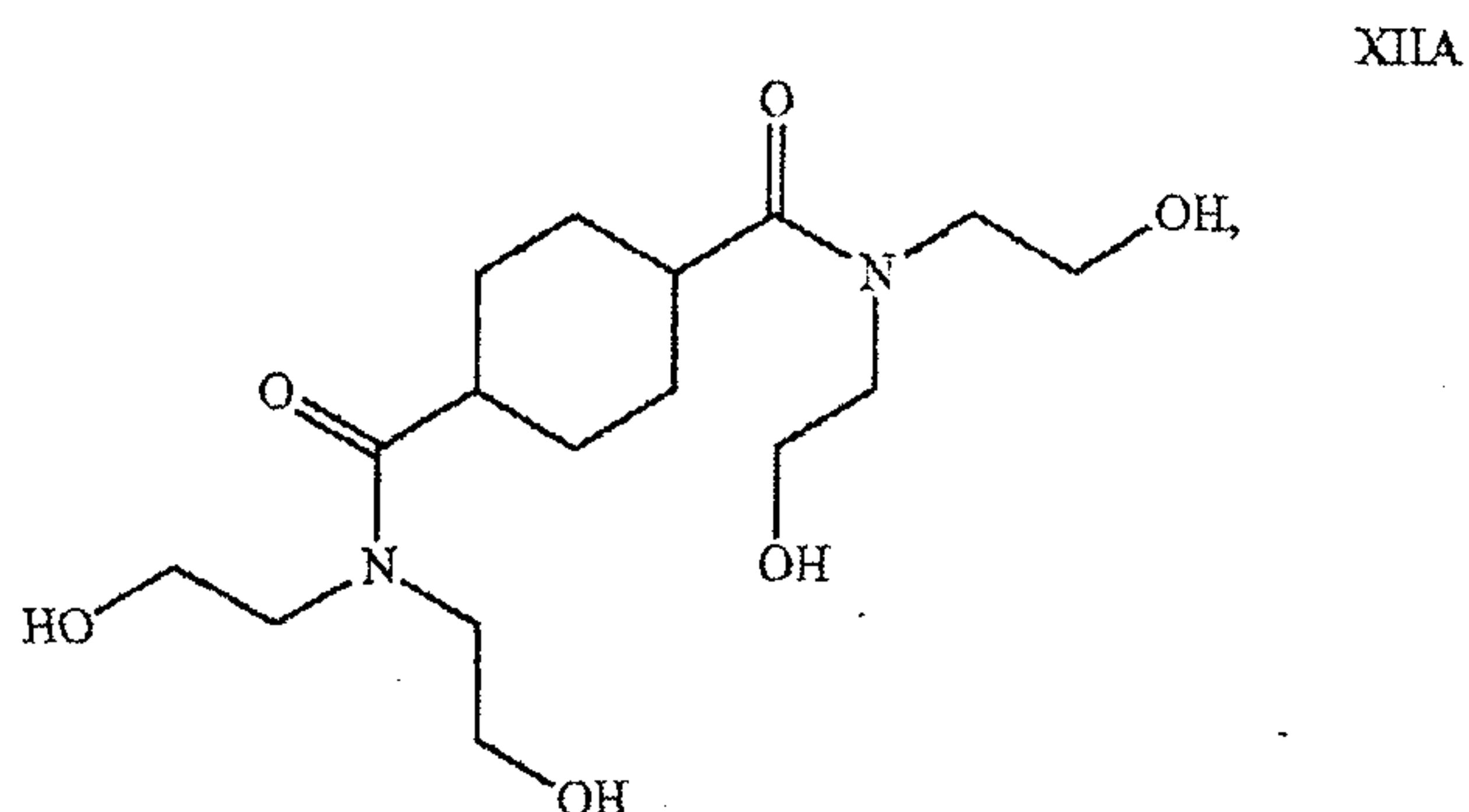
Unit cell dimensions:

 $a = 10.06350(10) \text{ \AA} \quad \alpha = 90^\circ$ $b = 11.85290(10) \text{ \AA} \quad \beta = 90^\circ$ $c = 14.6275(2) \text{ \AA} \quad \gamma = 90^\circ$

Volume:

 $1744.79(3) \text{ \AA}^3$

A further aspect of the invention is a method for the discontinuous preparation of an N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to formula XIA:



- 5 the method comprising reacting dimethyl 1,4-cyclohexyldicarboxylate and diethanolamine in a solvent in an extruder, intensive compounder, intensive mixer or static mixer, wherein the N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide has the following parameters: a trans content on the cyclohexyl ring of greater than or equal to 70 mol %, based on the total amount of all of the isomers of
- 10 N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide; two endothermic peaks according to DSC analysis (differential scanning calorimetry), where peak 1 is situated in the region of 140-170° C., with a maximum of 155-165° C., and peak 2 is situated in the region of 170-210° C., with a maximum of 175-207° C.; a ratio of the enthalpies of endothermic peak 1 to endothermic peak 2 is 1:1 to 1:5; and an XRPD

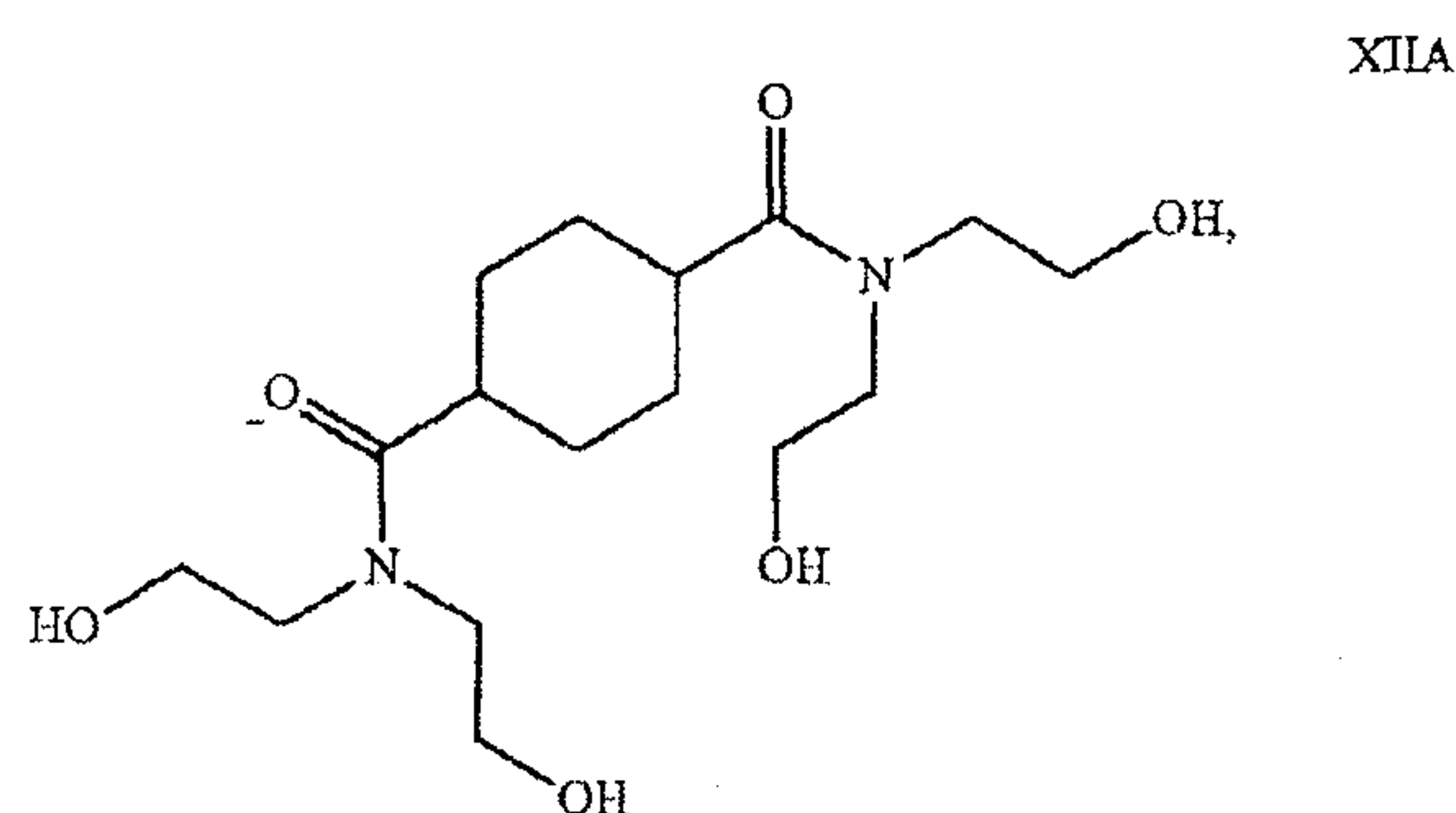
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26a

spectrum of the powder sample in an x-ray diffractometer, measured with Cu K α radiation (1.541 Å) has the following peaks:

Peak #	Degrees 2theta \pm 0.2 degree 2theta	d (Å)
1	14.90	5.94
2	16.70	5.31
3	17.40	5.09
4	21.20	4.19
5	21.60	4.11
6	26.00	3.43

A further aspect of the invention is a method for the discontinuous preparation of an
 5 N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to formula XIA:



the method comprising reacting dimethyl 1,4-cyclohexyldicarboxylate and diethanolamine in closed apparatus under pressure at temperatures from 60 to 140° C. without addition of solvents, wherein the N,N,N',N'-tetrakis(2-
 10 hydroxyethyl)cyclohexyl-1,4-diamide has the following parameters: a trans content on the cyclohexyl ring of greater than or equal to 70 mol %, based on the total amount of all of the isomers of N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide that are present; two endothermic peaks according to DSC analysis (differential scanning calorimetry), where peak 1 is situated in the region of 140-170° C., with a maximum
 15 of 155-165° C., and peak 2 is situated in the region of 170-210° C., with a maximum of 175-207° C.; a ratio of the enthalpies of endothermic peak 1 to endothermic peak 2

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26b

is 1:1 to 1:5; and an XRPD spectrum of the powder sample in an x-ray diffractometer, measured with Cu K α radiation (1.541 Å) has the following peaks:

Peak #	Degrees 2theta \pm 0.2 degree 2theta	d (Å)
1	14.90	5.94
2	16.70	5.31
3	17.40	5.09
4	21.20	4.19
5	21.60	4.11
6	26.00	3.43

The subject matter of the invention is illustrated below by means of examples.

Examples

- 5 Example 1: Preparation of a β -hydroxyalkylamide from dimethyl 1,4-cyclohexyldicarboxylate and diethanolamine by the method of the invention

Substances employed	Product description, manufacturer
Diethanolamine (DEA)	Dow Chemical
Dimethyl 1,4-cyclohexyldicarboxylate (DMCD) trans content 15-35 mol%	Dimethyl ester of 1,4-cyclohexanedicarboxylic acid, EASTMAN
Sodium methyllate	30% strength in methanol

- 10 Three streams were operated:

Stream 1 consisted of DMCD

Stream 2 consisted of DEA

15

Stream 3 consisted of the catalyst, the methanolic sodium methyllate solution.

The streams were metered such that the molar ratio between dimethyl 1,4-cyclohexyldicarboxylate and diethanolamine was 1:1.95.

20

The total amount of catalyst (only sodium methyllate, calculated on solvent-free bases), based on the overall formula, was 0.50 to 3.0%.

- 25 Stream 1 was fed in at a rate of 10.0 kg/h into the first barrel of a twin-screw extruder (ZSK 30, 32 d) (stream temperature 80 to 130°C).

Stream 2 was fed in at a rate of 9.9 kg/h (stream temperature 65 to 145°C).

- 30 Stream 3 was introduced through nozzles from entry into the extruder in stream 2 (0.5 to 2.0 kg/h).

The extruder used consisted of 8 barrels, which were separately heatable and coolable. Barrels 1 – 5: 160°C, barrels 6 – 8: 120 – 160°C.

Barrels 3, 5, and 8 were provided with a reduced-pressure dome (100 to 600 mbar).

5

The extruder screws were fitted with conveying elements. Ahead of the reduced-pressure domes, kneading blocks were installed.

All temperatures represented setpoint temperatures. Regulation took place via electrical heating or water cooling. The extruder head was likewise heated electrically (100 – 160°C).

10

The screw speed was 300 rpm. The reaction product was conveyed out of the extruder via a gear pump. The overall throughput was 20 kg/h.

15

The end product was cooled via a pipe section and/or via an extruder, and was guided onto a cooling belt, and cooled further. The product was subsequently recrystallized from deionized water at 100°C and cooled to room temperature. The mother liquor was filtered off and the filtercake was subsequently washed three times in methanol at room temperature and then dried in a vacuum drying oven at 50°C and about 20 mbar. It was subsequently ground.

20

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Brief description of the drawings

Figures 1-4 show NMR spectra for trans-N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4 diamide.

Figure 5 is a DSC diagram of the β -hydroxyalkylamide of the formula XIIA described in example 3a.

Figure 6 is a DSC diagram of the β -hydroxyalkylamide of the formula XIIA described in example 3b.

Figure 7 is a DSC diagram of the β -hydroxyalkylamide of the formula XIIA described in example 4b.

10 Figure 8 is a DSC diagram of the β -hydroxyalkylamide described in example 4c.

Figure 9 is an XRPD (x-ray powder diffraction) analysis of the β -hydroxyalkylamide of the formula XIIA described in example 3a (matting material).

Figure 10 is an XRPD (x-ray powder diffraction) analysis of the β -hydroxyalkylamide described in example 4c (nonmatting material).

15 Figure 11 is an XRPD (x-ray powder diffraction) analysis of the β -hydroxyalkylamide of the formula XIIA described in example 4b (mattering material).

Figure 12 is an ortep plot (50%) with numbering scheme.

Figure 13 is a calculated powder diffractogram based on the single-crystal structural determination of N, N, N'N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide (vesta sample).

20

Table 1: End products and characterization

Product – Example		1
Preparation		Recrystallized
Trans-N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide ¹	% by mass	95.30
Cis-N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide ¹	% by mass	0.28
Σ N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide ¹	% by mass	95.58
DEA ¹	% by mass	0.18
OH number	mg KOH/g	616
Base number	mg KOH/g	3
Melting range	°C	194-201

¹Analytical values by GC

OH number: DIN 53240

Base number: DIN 53176

5 Melting range: DIN EN ISO 3146

The trans-N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide prepared (formula XII) was characterized by the NMR spectra of figures 1-4.

10 Example 2: Powder coating material and coating

The powder coating material with the inventive β-hydroxyalkylamide (matt curing agent) from example 1 was prepared in the melt by joint extrusion of all of the components as per table 2 at a temperature (barrel) of 90°C (approximately 130°C melt temperature). The composition of the raw materials is set out in table 2. The ratio of acid groups in the polyester to OH groups in the curing agent was 1:1.

The extrudate is subsequently cooled, ground and sieved to a particle size of < 100 μm. The powder coating material thus prepared was applied using an electrostatic powder spraying unit at 60 KV to degreased steel panels (deep-drawn steel from Krüppel 210 × 70 × 0.8 mm) and/or aluminum panels (Q-panel AL-36 5005

20

H 14/08 0.8 mm) and baked in a forced-air drying oven between 160 to 220°C. The cured coating films have a film thickness of about 65 µm. The example data are based on a baking time of 20 minutes at 200°C.

5 Substances used:

1) Crosslinker:

Inventive β-hydroxyalkylamide crosslinker according to example 1.

10 2) Amorphous polyester:

CRYLCOAT® 2617-3, AN number: 32.7 mg KOH/g, Tg: 61°C, (Cytec Inc., USA)

3) Other formulating ingredients:

15 Titanium dioxide KRONOS® 2160, (Kronos GmbH, D),
RESIFLOW® PV 88, (Worlée-Chemie GmbH, D),
Benzoin, (Merck-Schuchard OHG, D)

Table 2

Product		% by mass	Substances used
HAA crosslinker		3.00	β-Hydroxyalkylamide example 1
Amorphous polyester		60.70	CRYLCOAT® 2617-3
Pigment TiO ₂		35.00	KRONOS® 2160
Flow control agent		1.00	RESIFLOW® PV 88
Degassing agent		0.30	Benzoin
Total		100.00	

20

Properties of the coating:

Baking conditions: 20 min at 200°C

Gloss: 33 scale divisions at 60° angle

Gloss: 42 scale divisions at 85° angle

25 Erichsen cupping: > 8 mm

Ball impact (direct): > 80 in lb

Gloss: DIN 67530 / ISO 2813

Erichsen cupping: DIN ISO 1520

Ball impact: DIN EN ISO 6272

5 The DSC measurements

The DSC measurements were carried out in accordance with DIN EN ISO 11357-1 from March 2010.

A heat flow difference calorimeter from the manufacturer Mettler-Toledo model DSC 821, with the serial number 5116131417 was used. The samples are run once from -
10 30°C to 250°C at 10 K/min.

Comprehensive description of the measurement method:

1. type (heat flow difference calorimeter or power-compensated calorimeter),
model and manufacturer of the DSC instrument used;
- 15 2. material, nature and type, and, where necessary, mass of the crucible used;
3. nature, purity, and volume flow rate of the flushing gas used;
4. nature of calibration method and details of the calibrating substances used,
including source, mass, and other properties important for calibration;
5. details relating to sampling, sample preparation, and conditioning

20

1: Heat flow difference calorimeter
Manufacturer: Mettler-Toledo
Model: DSC 821
Serial number: 5116131417

25

2: Crucible material: ultrapure aluminum
Size: 40 µl, without pin,
Mettler order No.: ME-26763
Mass incl. lid: about 48 mg

30

3: Flushing gas: nitrogen
Purity: 5.0 (> 99.999% by volume)
Volume flow rate: 40 ml/min

35

4: Calibration method: single
Material 1: indium

Mettler calibrating set ME-51119991

Mass: about 6 mg per weighing

Calibration of temperature (onset) and heat flow

Material 2: deionized water

5 Taken from the in-house system

Mass: about 1 mg per weighing

Calibration of temperature (onset)

5: Sampling: from sample vials supplied

10 Sample mass: 8 to 10 mg

Sample preparation: pressed on the crucible base using a die

Crucible lid: perforated

Measurement program: -30 to 250°C 10 K/min 1×

15 Description of the XRPD measurement:

The powder sample is pressed into a powder holder and measured in the Philips x-ray diffractometer PW1800 using Cu K α radiation (1.541 Å) under the following conditions:

20

Excitation: 40 kV, 45 mA

Measuring range: $3^\circ \leq 2\theta \leq 40^\circ$

Step size: 0.1° (2Theta)

Time per step: 20 s

25 Rotation: 1/4 revolution/sec

Receiving slit: coarse

Divergence slit: automatic

30 **Examples 3-4**

Substances employed	Product description, manufacturer
Diethanolamine (DEA)	Dow Chemical
Dimethyl 1,4-cyclohexyldicarboxylate (DMCD) (distilled) trans content 15-35 mol%	Dimethyl ester of 1,4-cyclohexanedicarboxylic acid, EASTMAN

Sodium methylate	30% strength in methanol
------------------	--------------------------

Example 3a

A three-neck flask with reflux condenser and glass stirrer is charged with 92.24 g of dimethyl 1,4-cyclohexyldicarboxylate with 96.91 g of diethanolamine, 10.84 g of 30% strength sodium methylate in methanol, and 52 g of methanol. A homogeneous solution is formed.

The batch is boiled for six hours under reflux with stirring in an oil heating bath (bath temperature 80°C). Product begins to precipitate after about 0.5 hour.

10

The reaction batch is left to cool, and further product crystallizes. The precipitated product is subsequently separated from the methanol by filtration and then dried. The yield is more than 80% of theory. Table 3

15 This gives, then, an N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to formula XIA having two endothermic peaks (1st at about 160°C and 2nd at about 190°C) in the DSC as per figure 5 and the XRPD spectrum as per figure 9 and table 5. This product thus produced produces far-reaching matting in powder coating materials, with a gloss of less than 50 scale divisions at the 60° angle, table 3.

20

Example 3b

The product produced in 3a is dissolved in boiling water, then slowly cooled down again; after crystallization has taken place, the crystallized product is briefly washed with methanol. Table 3

25

This product shows the two endothermic peaks – see figure 6 – with a matting effect present in the resulting powder coatings, of 29 scale divisions at the 60 degree angle, table 3.

Table 3: End products from discontinuous preparation examples 3a-3b and characterization thereof by GC analysis ¹⁾

Example		3a	3b
Starting material		-	3a
Preparation		Batch preparation as described in example 3a	Boil 3a in deionized water - cool slowly - crystallize - wash with methanol - dry under reduced pressure
¹⁾ DEA	% by mass	1.22	< 0.1
¹⁾ Trans-N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide	% by mass	89.34	91.81
¹⁾ Cis-N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide	% by mass	0.74	0.00
Σ N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide	% by mass	90.08	91.81
Ratio of ¹⁾ trans-N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide	mol%	99.2	100.0
to ¹⁾ cis-N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide	mol%	0.8	0.0
OH number	mg KOH / g	629	-
Base number	-	22	-
DSC: 1 st endo. peak - Δ H	°C - J/g	159 - 54	164 - 63
DSC: 2 nd endo. peak - Δ H	°C - J/g	186 - 89	203 - 124

¹⁾ Analytical values by GC

5 GC after silylation with Silyl 991 (BSTFA-TMCS 99 : 1) from Macherey und Nagel order No. 701.490.150. Silylation: 1 ml of Silyl 991, 1 ml of pyridine, 35 mg of reaction product, 35 mg of C-18 hydrocarbon as internal standard, heat for 30 minutes at 80°C in a closed ampoule.

Continuation of table 3
Powder coating data

PC experiment number		3a	3b
HAA crosslinker	% by mass	3.00	3.00
CRYLCOAT® 2617-3	% by mass	60.70	60.70
KRONOS® 2160	% by mass	35.00	35.00
RESIFLOW® PV 88	% by mass	1.00	1.00
Benzoin	% by mass	0.30	0.30
Total	% by mass	100.00	100.00
Curing	min@°C	30 @ 200	30 @ 200
Film thickness	µm	64-70	70-73
Gloss at 60° x	Sc. div.	30	29

OH number: DIN 53240

5 Base number: DIN 53176

Example 4a and 4b**4a**

An N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide with the product data 4a
5 is prepared in the same way as described in example 1 in an extruder (Werner und
Pfleiderer ZSK 30, 32 d). Table 4

4b

This product, as prepared and described in example 4a, is recrystallized. For this
10 purpose, the product from example 4a is dissolved with boiling in deionized water
and then slowly cooled and crystallized, to convert it back into the solid form. It is
subsequently washed with methanol and dried in a vacuum drying oven at 50°C and
about 20 mbar. Table 4

15 This, then, gives an N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide having
two endothermic peaks (1st at about 160°C and 2nd at about 190°C) in the DSC. This
product with the two peaks in the DSC as per figure 7 and the XRPD spectrum as per
figure 11 produces far-reaching matting in powder coating materials, with a gloss of
30 scale divisions at the 60° angle. Table 4

20

Comparative example 4c

A noninventive N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide having the
DSC as per figure 8 was prepared.

This product exhibits only one endothermic peak in the DSC, at about 190°C, as per
25 figure 8, and an XRPD spectrum as per figure 10 and table 6. The powder coating
material produced from this product does not produce far-reaching matting, but
instead exhibits a gloss of 95 scale divisions at the 60 degree angle. Table 4

Example 4d

30 An N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide of the formula XIIA with
the product data 4d is prepared in the same way as described in example 1 in an
extruder (Werner und Pfleiderer ZSK 30, 32 d). Table 4

This product thus produced is run onto a cooling belt and collected. This material is
then subjected to thermal conditioning under reduced pressure in a drying oven at
35 80°C for 24 hours, and the product thus obtained is then comminuted.

This product produces far-reaching matting in powder coating materials, with a gloss
of 40 scale divisions at the 60° angle. Table 4

Table 4: End products from continuous preparation examples 4a-4b and characterization thereof by GC analysis ¹⁾

Product examples		4a	4b	4d
Starting material		-	SK 988	
Preparation		Extruder setting as described in example 1	Dissolve 4a in deionized water - cool slowly - crystallize - wash with methanol - dry under reduced pressure	Extruder setting as described in example 1 - thermal condition 24 h 80°C reduced pressure
¹⁾ DEA fraction	% by mass	2.17	0.11	1.2
¹⁾ Trans-N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide	% by mass	84.25	93.72	91.3
¹⁾ Cis-N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide	% by mass	1.60	0.11	0.66
Σ N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide	% by mass	85.85	93.83	91.96
Ratio of ¹⁾ trans-N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide to ¹⁾ cis-N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide	mol%	98.1	99.9	99.3
OH number	mg KOH / g	641	625	
Base number	-	24	1.1	
DSC: 1 st endo. peak - Δ H	°C - J/g		162 - 61	158-50
DSC: 2 nd endo. peak - Δ H	°C - J/g		200 - 128	188-115

¹⁾Analytical values by GC. GC after silylation with Silyl 991 (BSTFA-TMCS 99 : 1) from Macherey und Nagel order No. 701.490.150. Silylation: 1 ml of Silyl 991, 1 ml of pyridine, 35 mg of reaction product, 35 mg of C-18 hydrocarbon as internal standard, heat for 30 minutes at 80°C in a closed ampoule.

Continuation of table 4
Powder coating data

PC experiment number			4b	4d
HAA crosslinker	% by mass		3.00	3.00
CRYLCOAT® 2617-3	% by mass		60.70	60.70
KRONOS® 2160	% by mass		35.00	35.00
RESIFLOW® PV 88	% by mass		1.00	1.00
Benzoin	% by mass		0.30	0.30
Total	% by mass		100.00	100.00
Curing	min@°C		30 @ 200	30 @ 200
Film thickness	µm		52-55	58-68
Gloss at 60° x	Sc. div.		29-30	40

5 OH number: DIN 53240
Base number: DIN 53176

Table 4 Continued: End products from preparation of comparative example 4c and characterization by GC analysis ¹⁾ and powder coating material

Comparative example		4c
Starting material		
Preparation		- allow to cool at RT
¹⁾ DEA	% by mass	2.87
¹⁾ Trans-N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide	% by mass	64.11
¹⁾ Cis-N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide	% by mass	15.84
Σ N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide	% by mass	79.95
Ratio of ¹⁾ trans-N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide	mol%	80.19
to ¹⁾ cis-N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide	mol%	19.81
OH number	mg KOH/ g sample	-
Base number	-	-
DSC: 1 st endo. peak - Δ H	°C - J/g	
DSC: 2 nd endo. peak - Δ H	°C - J/g	171 - 87

5

Powder coating data

PC experiment number		4c
HAA crosslinker	% by mass	3.00
CRYLCOAT [®] 2617-3	% by mass	60.70
KRONOS [®] 2160	% by mass	35.00
RESIFLOW [®] PV 88	% by mass	1.00
Benzoin	% by mass	0.30
Total	% by mass	100.00
Curing	min@°C	30 @ 200
Film thickness	μm	65-78
Gloss at 60° ∅	Sc. div.	95

Example 5

A β -hydroxyalkylamide of the formula XIIA was prepared as in example 3a. A single crystal was grown from this product. The inventive of the formula XIIA was investigated by x-ray structural analysis of a single crystal. Comprehensive information on the measurement is compiled in annex 1.

ANNEX 1

Single-crystal x-ray structural analysis

Analytical method: Single Crystal X-ray Structure Analysis "2012-0573602-06D"

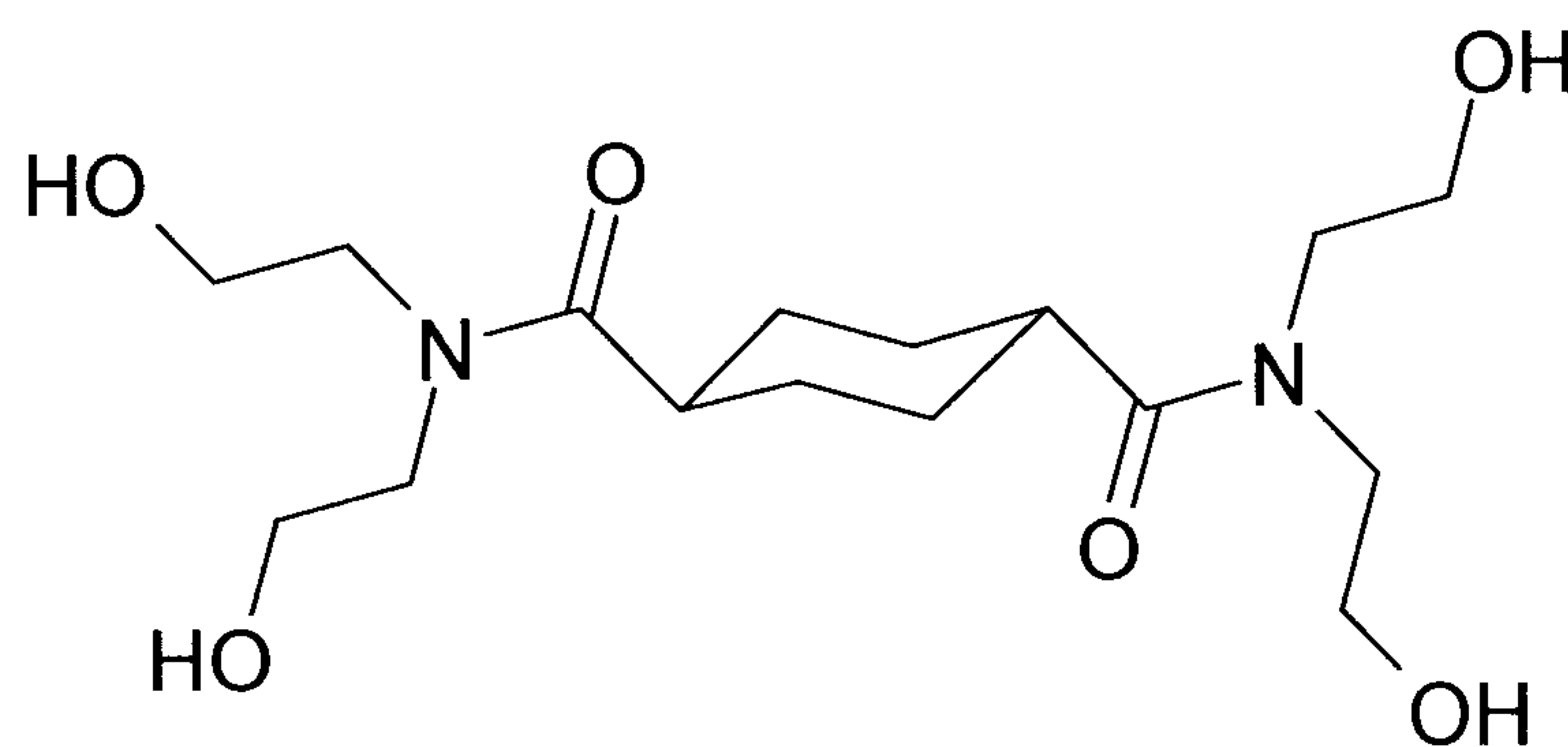
Report: WHC 11/11 EKS

Sample received: 2011-02-22

Report date: 2011-02-25

Objective: Determination of single-crystal structure.

Compound: N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide formula XIIA



Crystallization: by the chemist.

Crystal dimensions: colorless block, 0.50 × 0.40 × 0.40 mm³

Code: vesta

Comments: The asymmetric unit contains half a molecule.

Figure 12: Ortep Plot (50%) with numbering scheme.

Experimental details

The single-crystal structure determination was carried out using an instrument from Oxford Diffraction, which was fitted with a CCD detector (Ruby model), a conventional x-ray tube with $\text{Cu}_{\text{K}\alpha}$ radiation, osmic mirror as monochromator, and a low-temperature unit of the Cryojet type ($T = 100 \text{ K}$). The data collection was carried out in phi and omega scans. Data collection and reduction took place with CrysAlis (Oxford Diffraction 2007).

Structural elucidation and refinement took place with SHELXTL (V. 6.10, Sheldrick, University of Göttingen, 2000). All nonhydrogen atoms were refined anisotropically. The hydrogen atoms were refined as riding groups.

Tables

Table a: Crystal data and data relating to structural refinement for vesta.

Identification code	vesta
Empirical formula	C ₁₆ H ₃₀ N ₂ O ₆
Formula weight	346.42
Temperature	100 K
Wave length	1.54178 Å
Crystal system	Orthorhombic
Space group	Pbca
Unit cell	$a = 10.06350(10) \text{ Å}$ $\alpha = 90^\circ$ $b = 11.85290(10) \text{ Å}$ $\beta = 90^\circ$ $c = 14.6275(2) \text{ Å}$ $\gamma = 90^\circ$
Volume	1744.79(3) Å ³
Z	4
Density (calculated)	1.319 Mg/m ³
Absorption coefficient	0.832 mm ⁻¹
F(000)	752
Crystal dimensions	0.50 x 0.40 x 0.40 mm ³
Theta range for data collection	6.05 to 65.68°.
Index range	-11 ≤ h ≤ 10, -12 ≤ k ≤ 14, -14 ≤ l ≤ 17
Number of reflections collected	9191
Symmetry-independent reflections	1482 [R(int) = 0.0345]
Completeness to theta = 65.68°	98.5 %
Absorption correction	Crysalis
Refinement	Full-matrix least-squares on F ²
Data/restraints/parameters	1482 / 0 / 111
Goodness-of-fit on F ²	1.065
Final R values [I > 2 sigma(I)]	R1 = 0.0316, wR2 = 0.0792
R values (all data)	R1 = 0.0358, wR2 = 0.0817
Largest difference peaks	0.199 and -0.189 e.Å ⁻³

Table b: Bond lengths [Å] and angles [°] for vesta.

O(1)-C(4)	1.2478(15)	N(1)-C(7)	1.4727(15)	C(3)-C(1)#1	1.5291(17)
O(2)-C(6)	1.4221(15)	C(1)-C(3)#1	1.5291(17)	C(5)-C(6)	1.5182(16)
O(3)-C(8)	1.4205(16)	C(1)-C(2)	1.5398(16)	C(7)-C(8)	1.5159(17)
N(1)-C(4)	1.3479(16)	C(2)-C(4)	1.5189(17)		
N(1)-C(5)	1.4741(15)	C(2)-C(3)	1.5405(16)		

C(4)-N(1)-C(5)	124.59(10)	O(1)-C(4)-N(1)	119.97(11)
C(4)-N(1)-C(7)	117.87(10)	O(1)-C(4)-C(2)	120.15(10)
C(5)-N(1)-C(7)	117.54(9)	N(1)-C(4)-C(2)	119.84(10)
C(3)#1-C(1)-C(2)	110.62(10)	N(1)-C(5)-C(6)	113.66(9)
C(4)-C(2)-C(1)	111.04(10)	O(2)-C(6)-C(5)	110.97(10)
C(4)-C(2)-C(3)	108.67(10)	N(1)-C(7)-C(8)	113.52(10)
C(1)-C(2)-C(3)	110.09(10)	O(3)-C(8)-C(7)	113.31(10)
C(1)#1-C(3)-C(2)	111.18(10)		

Symmetry operations for generating equivalent atoms:

#1 -x+1,-y+1,-z

Table c: Torsional angles [°] for vesta.

C(3)#1-C(1)-C(2)-C(4)	177.11(9)	C(7)-N(1)-C(5)-C(6)	-99.58(12)
C(3)#1-C(1)-C(2)-C(3)	56.72(14)	N(1)-C(5)-C(6)-O(2)	61.92(13)
C(4)-C(2)-C(3)-C(1)#1	-178.85(9)	C(4)-N(1)-C(7)-C(8)	86.25(13)
C(1)-C(2)-C(3)-C(1)#1	-57.04(14)	C(5)-N(1)-C(7)-C(8)	-93.60(12)
C(5)-N(1)-C(4)-O(1)	176.19(10)	N(1)-C(7)-C(8)-O(3)	73.97(13)
C(7)-N(1)-C(4)-O(1)	-3.65(16)		
C(5)-N(1)-C(4)-C(2)	-6.21(16)		
C(7)-N(1)-C(4)-C(2)	173.95(10)		
C(1)-C(2)-C(4)-O(1)	-54.62(14)		
C(3)-C(2)-C(4)-O(1)	66.61(14)		
C(1)-C(2)-C(4)-N(1)	127.78(11)		
C(3)-C(2)-C(4)-N(1)	-110.98(12)		
C(4)-N(1)-C(5)-C(6)	80.57(13)		

Symmetry operations for generating equivalent atoms:

#1 $-x+1, -y+1, -z$

Figure 13:

Calculated powder diffractogram based on the single-crystal structure determination of N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide (vesta sample)

Peak #	Degrees 2theta ± 0.2 degree 2theta	d (Å)
1	14.90	5.94
2	16.70	5.31
3	17.40	5.09
4	21.20	4.19
5	21.60	4.11
6	26.00	3.43

Table 5: Characteristic XRPD peaks (in degrees 2theta) for the β -hydroxyalkylamide of the formula XIIA described in example 3a (matting material)

Peak #	Degrees 2theta ± 0.2 Grad 2theta	d (Å)
1	8.90	9.93
2	13.90	6.37
3	17.10	5.18
4	17.60	5.04
5	18.50	4.79
6	19.00	4.67
7	22.20	4.00
8	23.40	3.80
9	24.00	3.71
10	24.60	3.62
11	25.40	3.50
12	28.70	3.11
13	32.00	2.80
14	37.30	2.41

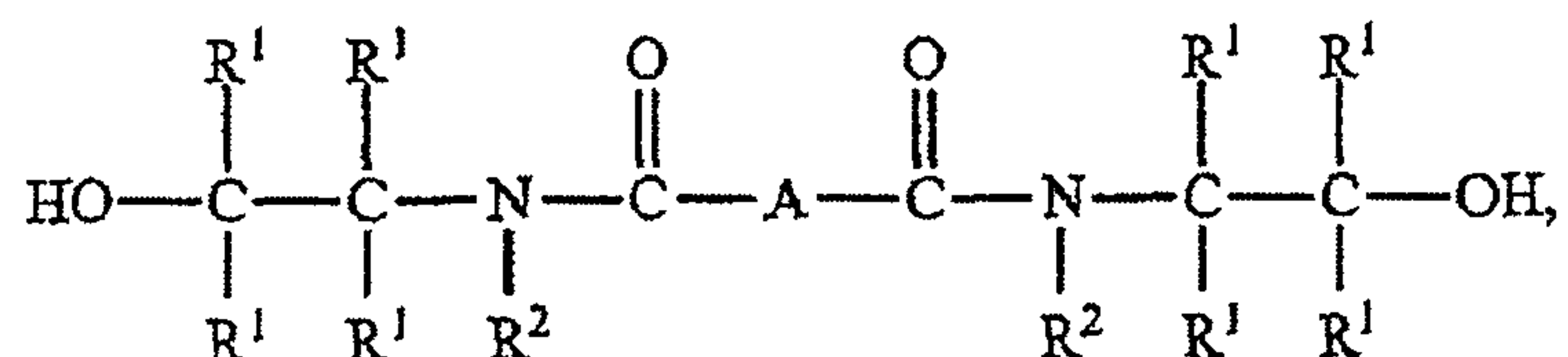
Table 6: Characteristic XRPD peaks (in degrees 2theta) for the β -hydroxyalkylamide described in example 4c (nonmatting material)

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CLAIMS:

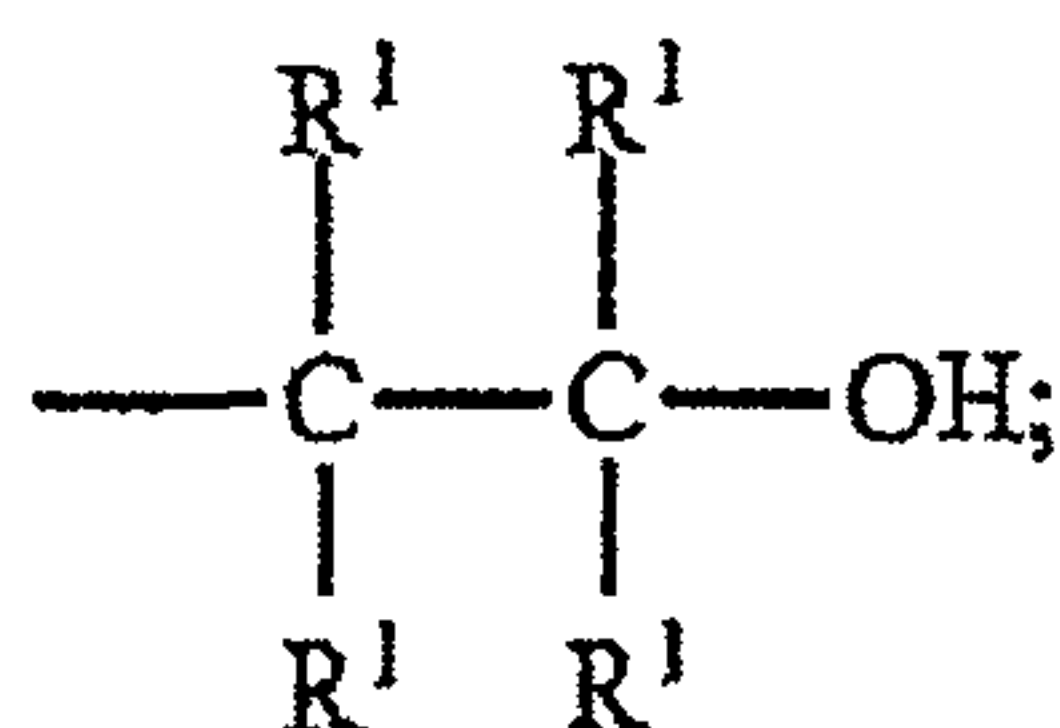
1. A β -hydroxyalkylamide, comprising two or three or four β -hydroxyalkylamide groups per molecule of the formula I:



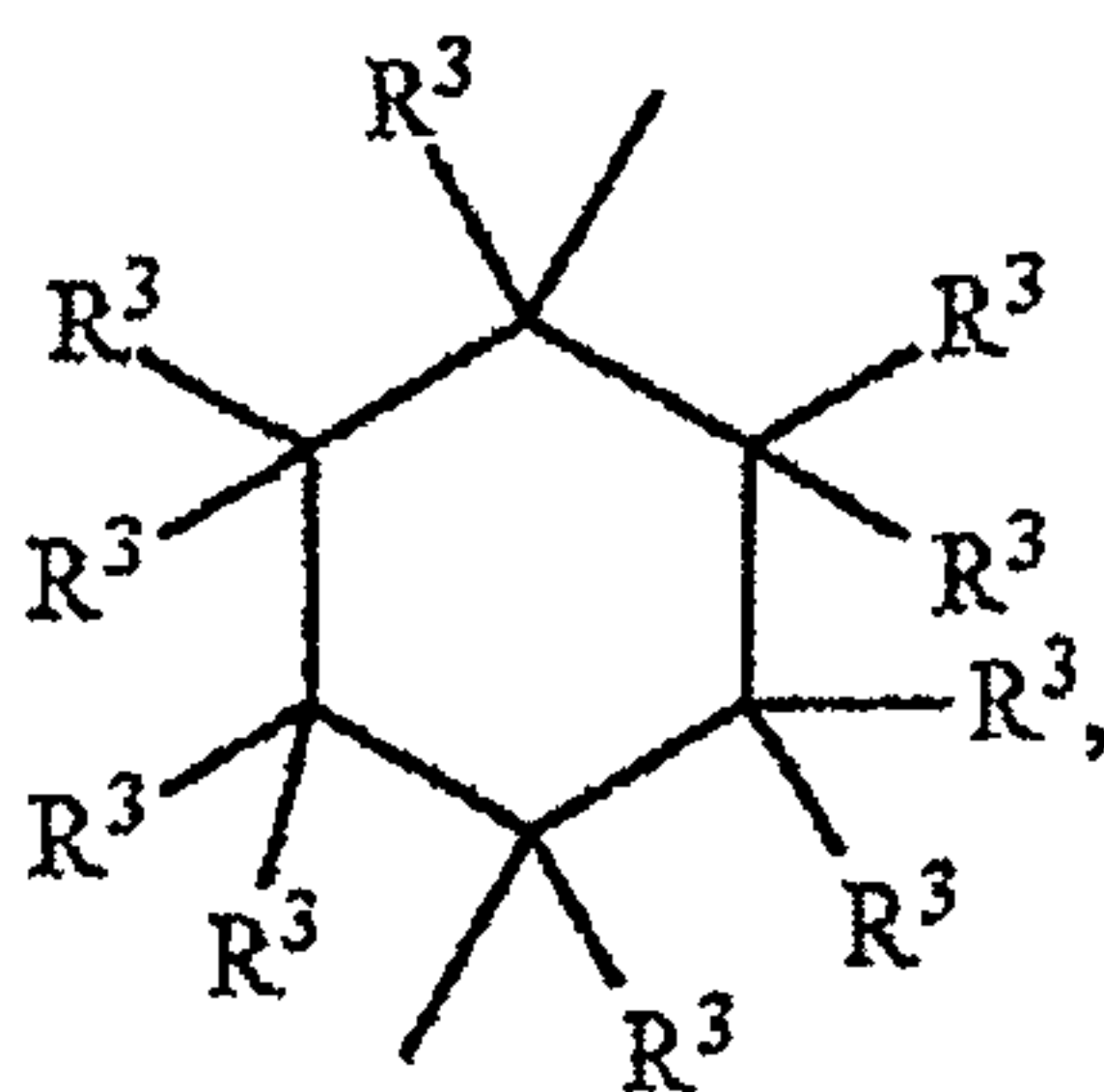
5 wherein:

R^1 and R^2 independently of one another are identical or different radicals selected from the group consisting of an alkyl radical, a cycloalkyl radical, an aryl radical, an aralkyl radical and an alkenyl radical having 1-24 carbon atoms, such that the radicals optionally comprise a heteroatom, R^1 is optionally a hydrogen, and

10 R^2 is optionally represented by the formula:



A is represented by A^1 :



15 wherein radicals R^3 independently of one another are identical or different radicals selected from the group consisting of hydrogen, an alkyl radical, a cycloalkyl radical, an aryl radical, an aralkyl radical, and an alkenyl radical having

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1-24 carbon atoms, such that the radicals optionally comprise a heteroatom, and two or more substituents R^3 are optionally linked with one another to form rings;

the β -hydroxyalkylamide is present in solid form below 150°C ;

wherein the β -hydroxyalkylamide has a trans content of greater than or
 5 equal to 70 mol % based on the total amount of all of the isomers of the
 β -hydroxyalkylamide;

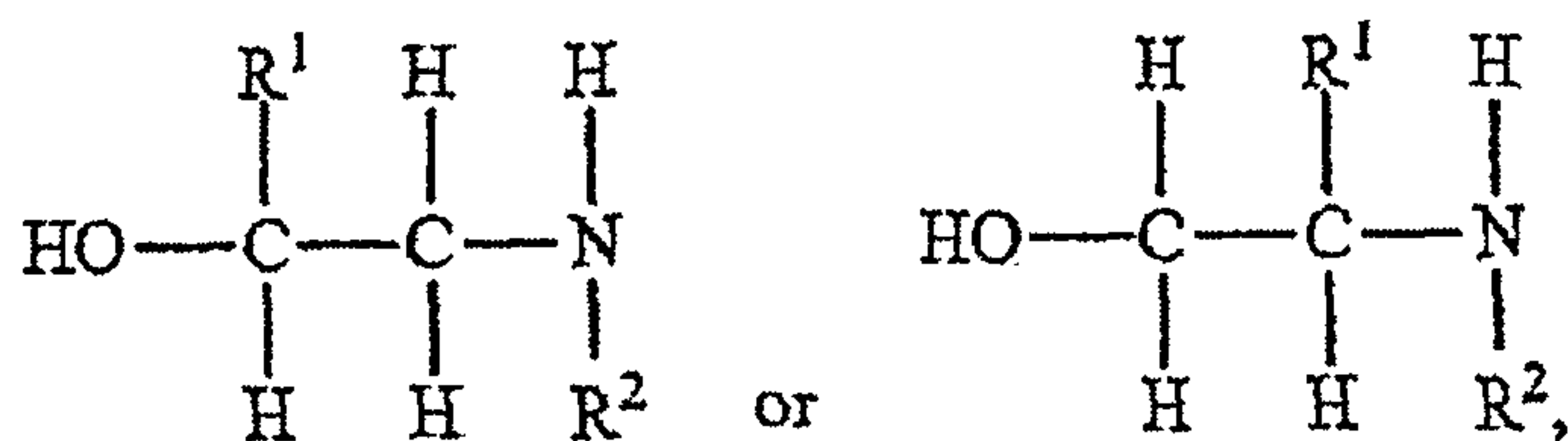
wherein the β -hydroxyalkylamide has two endothermic peaks according
 to DSC analysis (differential scanning calorimetry), where peak 1 is situated in the
 region of $140\text{--}170^\circ\text{C}$, with a maximum of $155\text{--}165^\circ\text{C}$, and peak 2 is situated in the
 10 region of $170\text{--}210^\circ\text{C}$, with a maximum of $175\text{--}207^\circ\text{C}$; and

wherein a ratio of the enthalpies of endothermic peak 1 to endothermic
 peak 2 is 1:1 to 1:5.

2. The β -hydroxyalkylamide of claim 1, wherein the β -hydroxyalkylamide
 derives from at least one β -hydroxyalkylamine comprising an alkyl group having at
 15 least 2 to 10 carbon atoms in the hydrocarbon framework, such that the alkyl groups
 are optionally linear, branched or cyclic, and the alkyl groups are optionally
 substituted with at least one heteroatom.

3. The β -hydroxyalkylamide of claim 1, comprising at least one
 β -hydroxyalkylamide formed from an N-alkyl-1,2-alkanolamine, an N,N,
 20 -bis-2-hydroxyalkylamine, or both, and an ester of a cyclohexanedicarboxylic acid.

4. The β -hydroxyalkylamide of claim 1 comprising a β -hydroxyalkylamide
 derived from at least one β -hydroxyalkylamine of formula II and/or III, formulae II:



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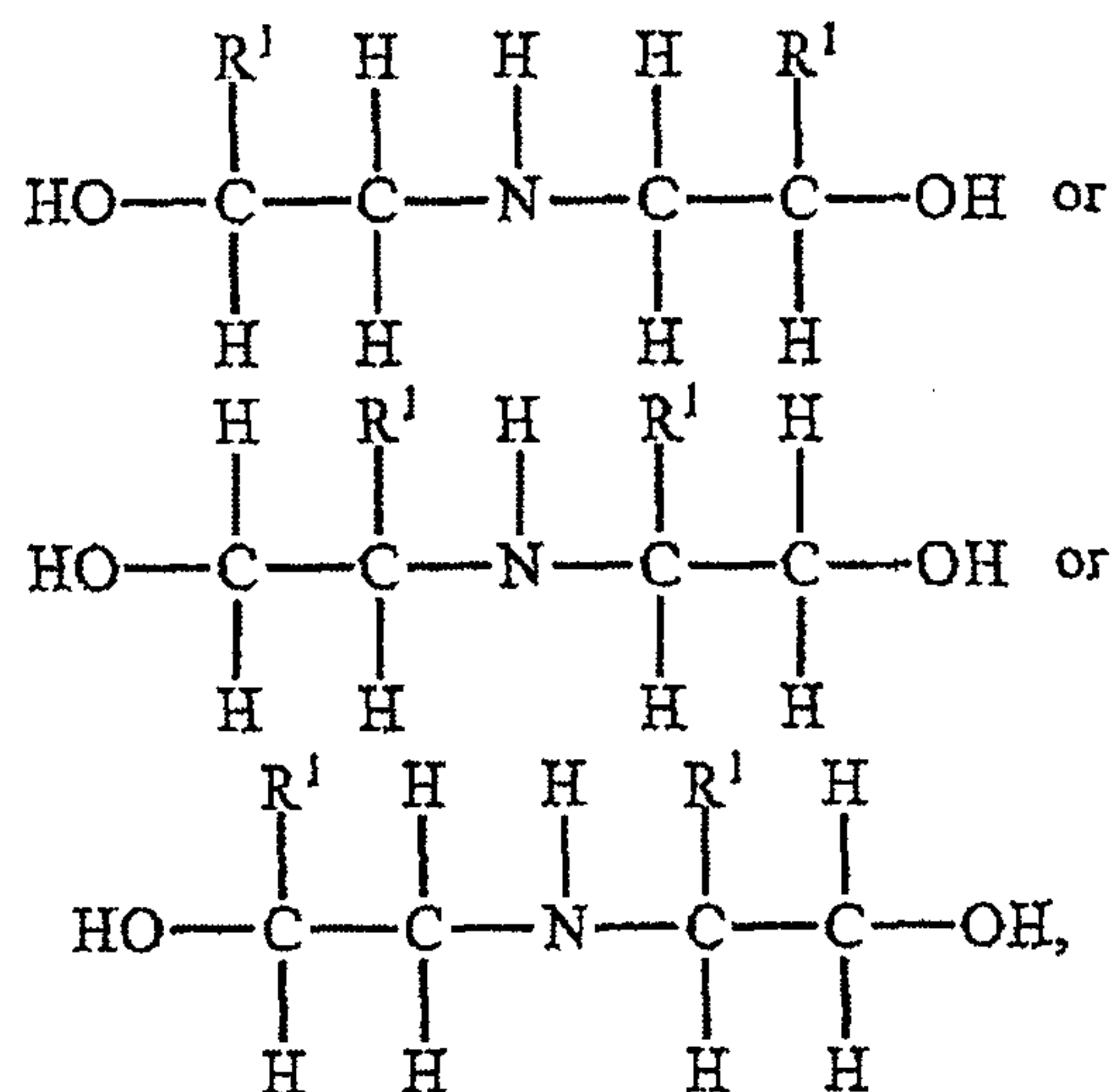
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wherein

R^1 is hydrogen, methyl, ethyl, or propyl; and

R^2 is methyl;

formulae III:



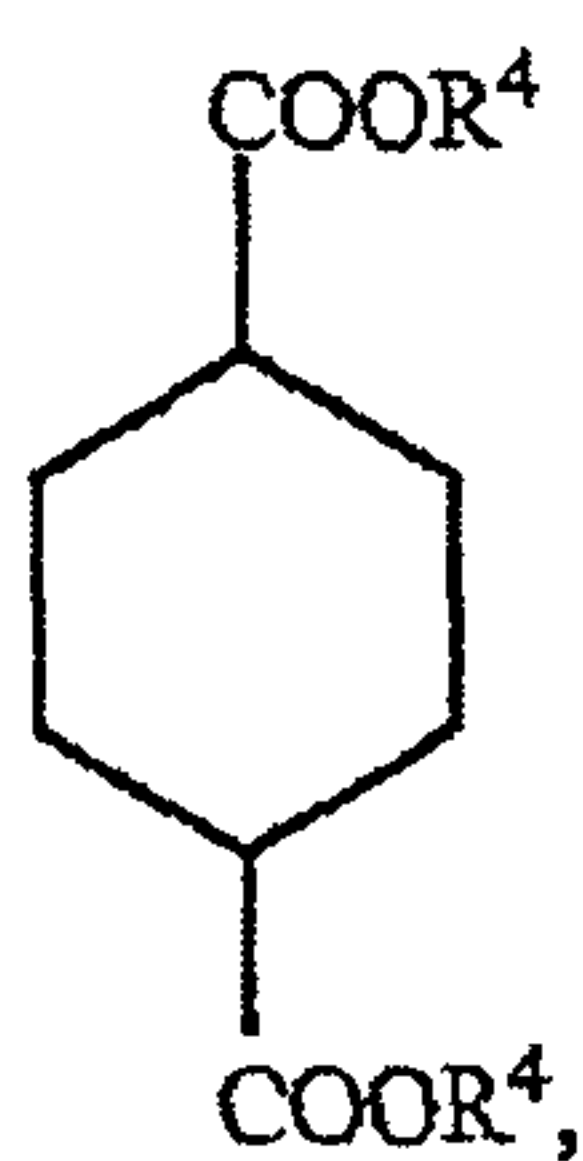
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wherein radicals R^1 simultaneously or independently of one another are hydrogen, methyl, ethyl, or propyl.

5. The β -hydroxyalkylamide of claim 1, comprising a β -hydroxyalkylamide derived from the following compounds: diethanolamine (DEA),
- 10 di-isopropanolamine (DIPA), di-sec-butanolamine, N-methylethanolamine, N-methylisopropanolamine.
6. The β -hydroxyalkylamide of claim 1, comprising a β -hydroxyalkylamide derived from at least one compound comprising the substituent A, said compound being a 1,4-substituted cyclohexanedicarboxylic acid derivative.
- 15 7. The β -hydroxyalkylamide of claim 1, comprising a β -hydroxyalkylamide derived from at least one compound of formula IV:

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wherein radicals R^4 simultaneously or independently of one another are methyl, ethyl, propyl, or butyl.

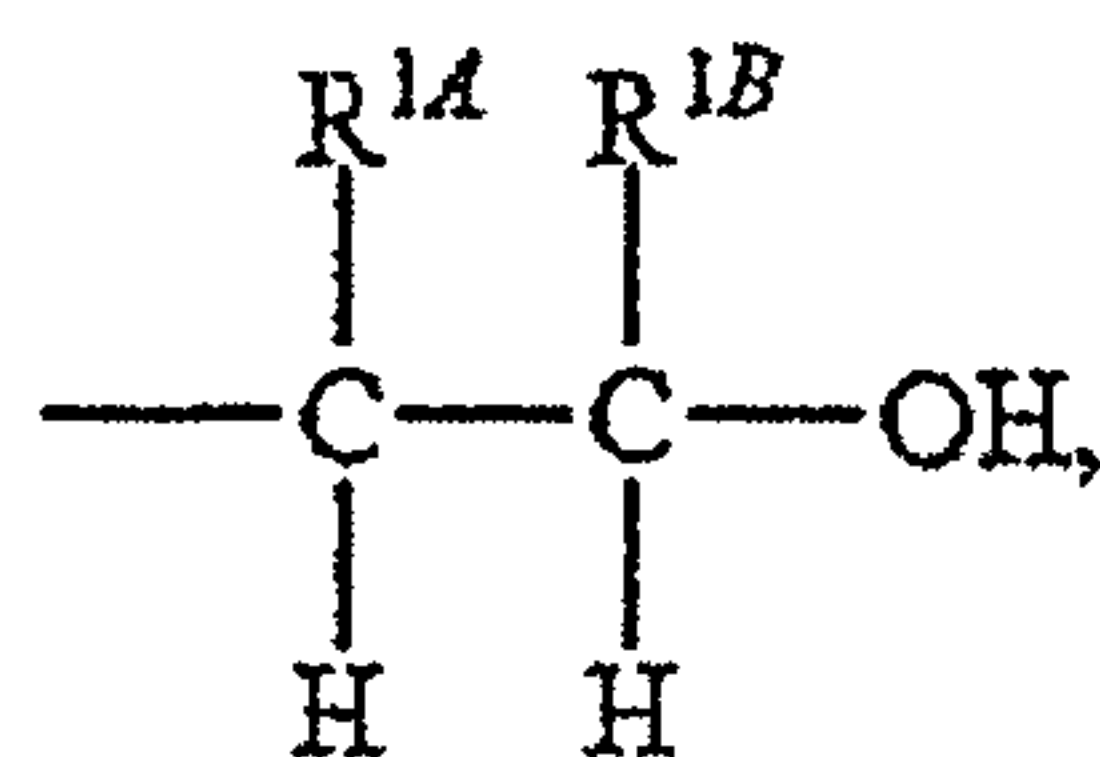
8. The β -hydroxyalkylamide of claim 1, comprising a β -hydroxyalkylamide
5 derived from at least one 1,4-substituted cyclohexanedicarboxylic ester.

9. The β -hydroxyalkylamide of claim 1, comprising at least one
 β -hydroxyalkylamide of the following formulae:

R^2 is methyl, or:

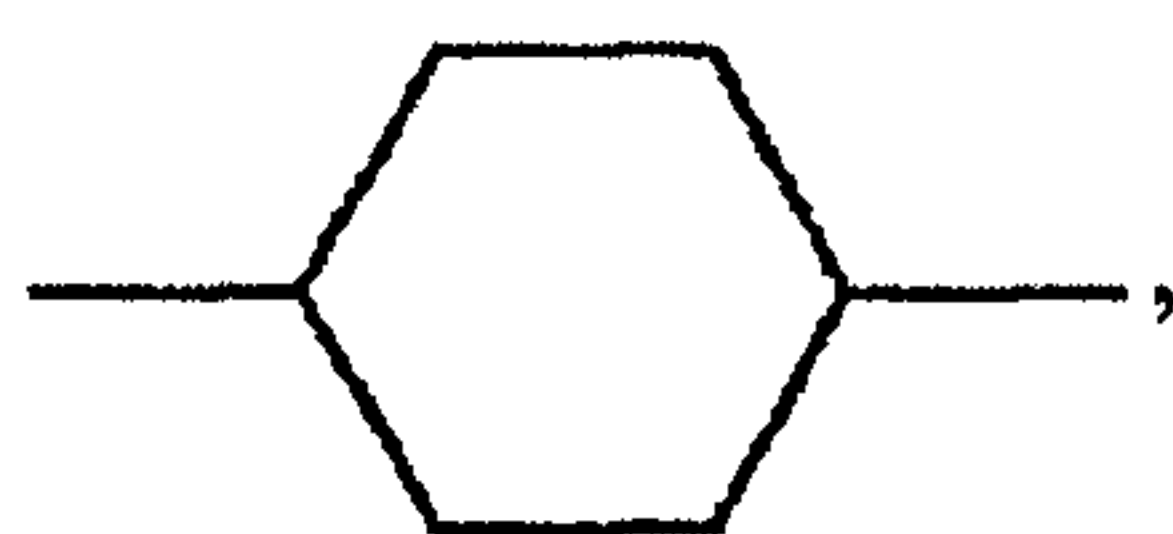
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such that R^{1A} is hydrogen and R^{1B} is methyl, ethyl, or propyl, or R^{1A} is methyl, ethyl, or propyl and R^{1B} is hydrogen;

A is a 1,4-disubstituted cyclohexane ring of the formula:



5

wherein a trans content of A is ≥ 70 mol %; and

the β -hydroxyalkylamide is in solid form below 150°C ;

wherein the β -hydroxyalkylamide has two endothermic peaks according to DSC analysis (differential scanning calorimetry), where peak 1 is situated in the region of 140 - 170°C , with a maximum of 155 - 165°C , and peak 2 is situated in the region of 170 - 210°C , with a maximum of 175 - 207°C ; and

wherein a ratio of the enthalpies of endothermic peak 1 to endothermic peak 2 is 1:1 to 1:5.

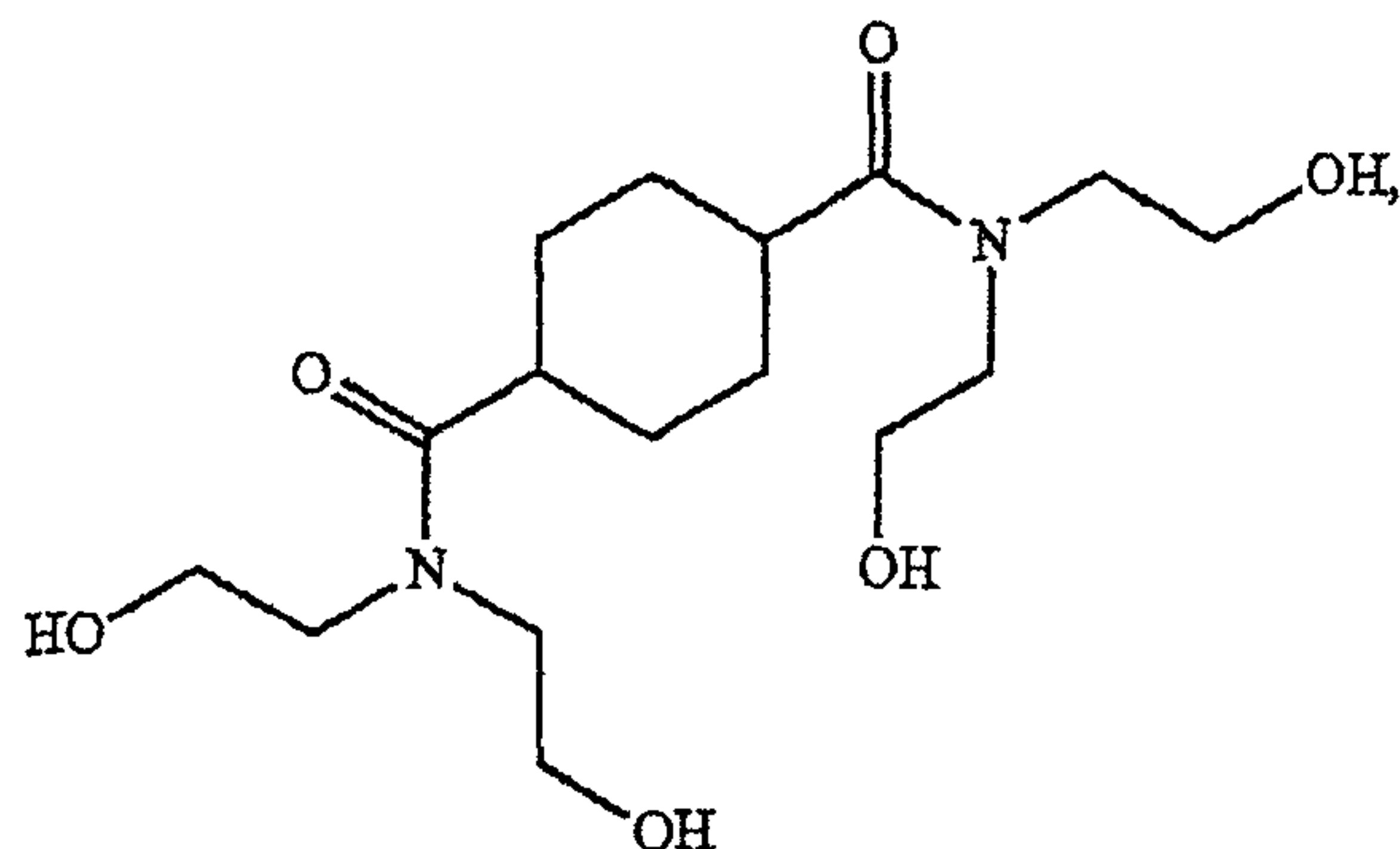
10. The β -hydroxyalkylamide of claim 1, formed from at least one dialkyl 1,4-cyclohexyldicarboxylate having a trans content, based on the position of the carboxyl groups on the cyclohexyl ring, of greater than or equal to 70 mol %.

11. The β -hydroxyalkylamide of claim 1, formed from a dimethyl 1,4-cyclohexyldicarboxylate and diethanolamine, having four β -hydroxyalkylamide groups per molecule, of the formula XII:

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formula XII

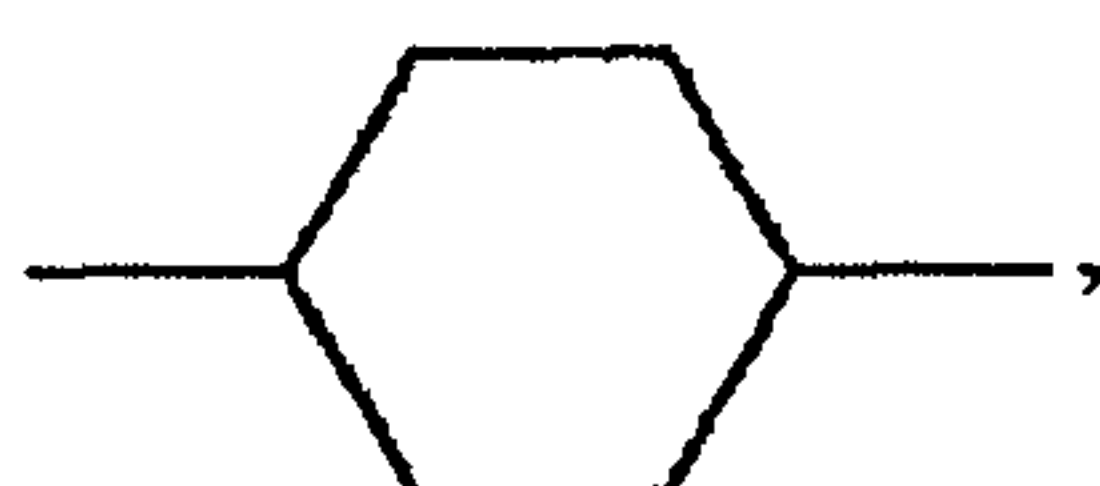


wherein a trans content on the cyclohexyl ring is greater than or equal to 70 mol %.

12. The β -hydroxyalkylamides of claim 1, comprising a β -hydroxyalkylamide
5 present in solid form below 150° C.

13. A method for the solvent-free, continuous preparation of the
 β -hydroxyalkylamide of claim 1, the method comprising reacting reactants
continuously in an extruder, intensive compounder, intensive mixer, or a static mixer
with intensive mixing to form a β -hydroxyalkylamide, wherein

10 A is a 1,4-disubstituted cyclohexane ring of the formula:



such that a trans content of A is ≥ 70 mol %.

14. The method of claim 13, wherein the reactants comprise a dialkyl 1,
4-cyclohexyldicarboxylate having a trans content on the cyclohexyl ring of greater
15 than or equal to 70 mol % and present in solid form below 150° C.

15. The method of claim 13, wherein the reactants comprise a dialkyl 1,
4-cyclohexyldicarboxylate.

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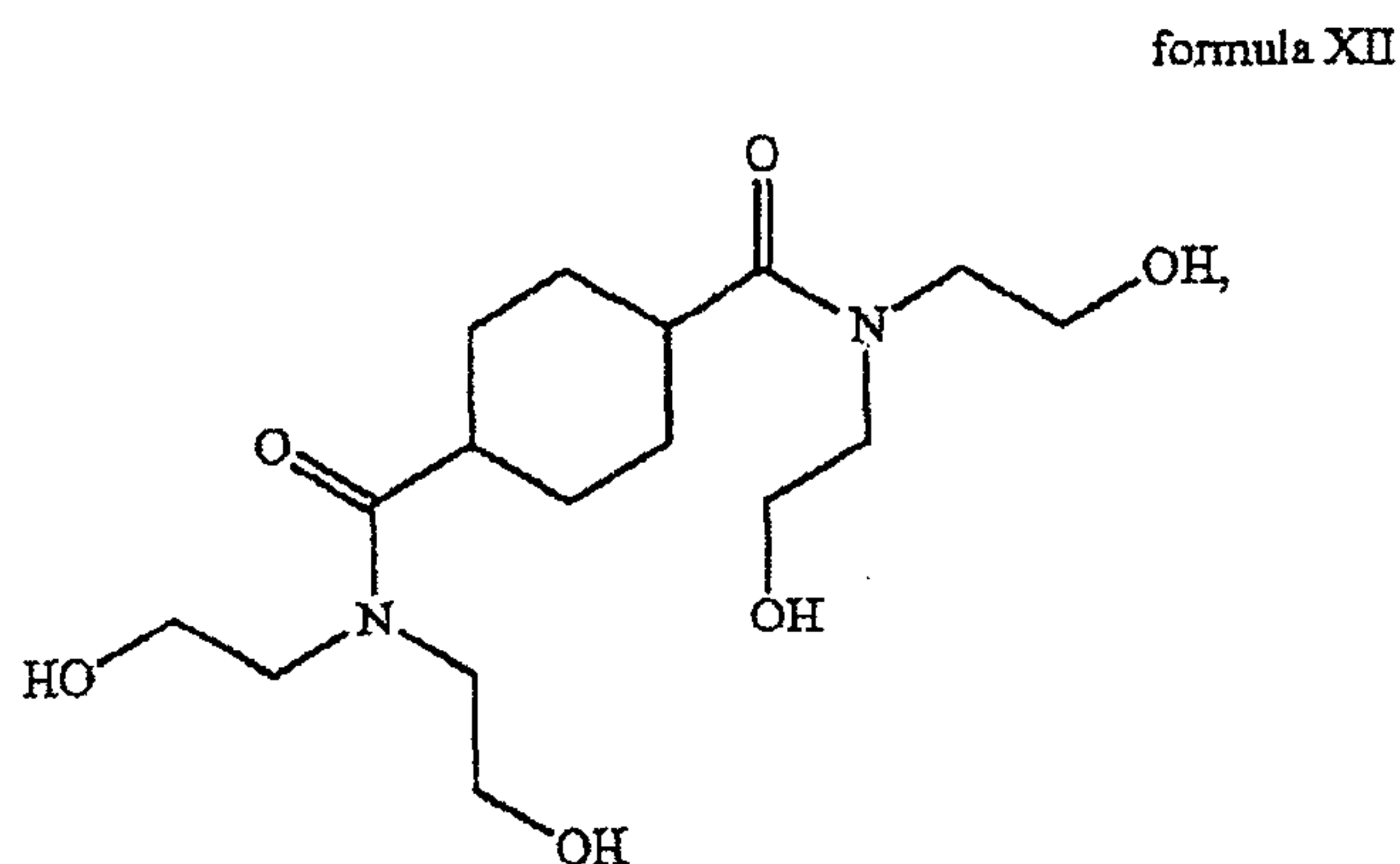
16. The method of claim 13, comprising reacting the reactants with intense mixing and a brief reaction with a supply of heat at temperatures $>50^{\circ}\text{C}$ followed by isolation of the β -hydroxyalkylamide by cooling.

17. The method of claim 13, wherein a residence time of the reactants
5 is 3 seconds to 15 minutes.

18. The method of claim 13, wherein the reaction occurs in a single-, twin- or multiscrew extruder, annular extruder or planetary roller extruder.

19. The method of claim 13, wherein a temperature in the extruder, intensive compounder, intensive mixer or static mixer is 50 to 325°C .

10 20. The method of claim 13, wherein the reactants comprise dimethyl 1, 4-cyclohexyldicarboxylate and diethanolamine, and the β -hydroxyalkylamide comprises four β -hydroxyalkylamide groups per molecule, and is represented by formula XII:



15 such that a trans content on the cyclohexyl ring of greater than or equal to 70 mol %.

21. A method for producing a coating having a matt surface, the method comprising apply the β -hydroxyalkylamide of claim 1 to a substrate.

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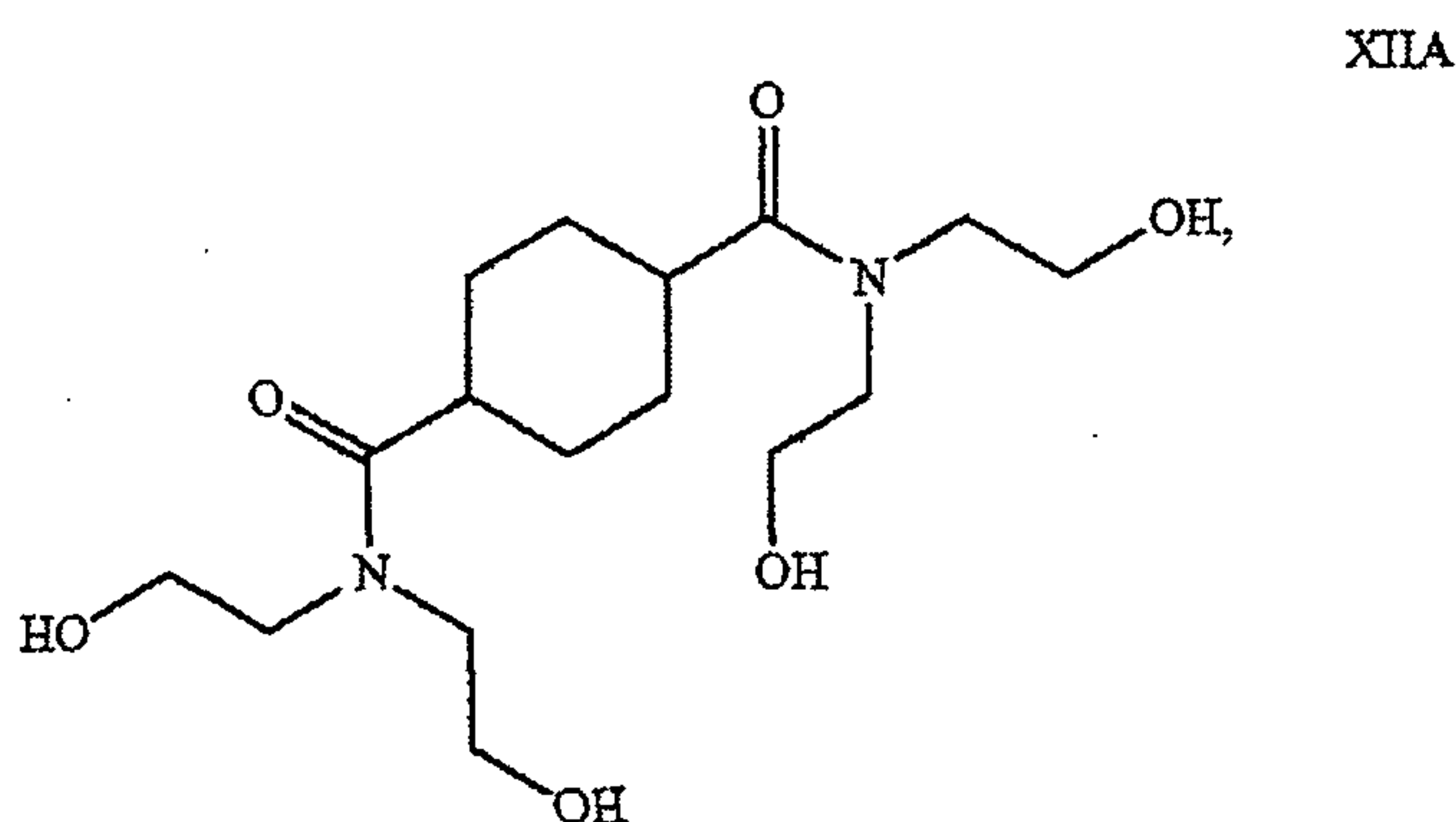
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22. A crosslinker for a polymer comprising carboxyl groups, the crosslinker comprising the β -hydroxyalkylamide of claim 1.

23. A powder coating material, comprising the crosslinker of claim 22.

24. The method of claim 21, wherein the matt coating has <50 gloss units,
5 measured as reflectometer values according to DIN 67530/ISO 2813 at an incident angle of 60°.

25. The β -hydroxyalkylamide of claim 11, comprising an N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide of formula XIIA:



10 having the following parameters:

a trans content on the cyclohexyl ring of greater than or equal to 70 mol %, based on the total amount of all of the isomers of N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide;

15 two endothermic peaks according to DSC analysis (differential scanning calorimetry), where peak 1 is situated in the region of 140-170° C, with a maximum of 155-165°C, and peak 2 is situated in the region of 170-210° C, with a maximum of 175-207° C;

a ratio of the enthalpies of endothermic peak 1 to endothermic peak 2 is 1:1 to 1:5; and

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a XRPD spectrum of the powder sample in an x-ray diffractometer, measured with Cu K α radiation (1.541 Å) has the following peaks:

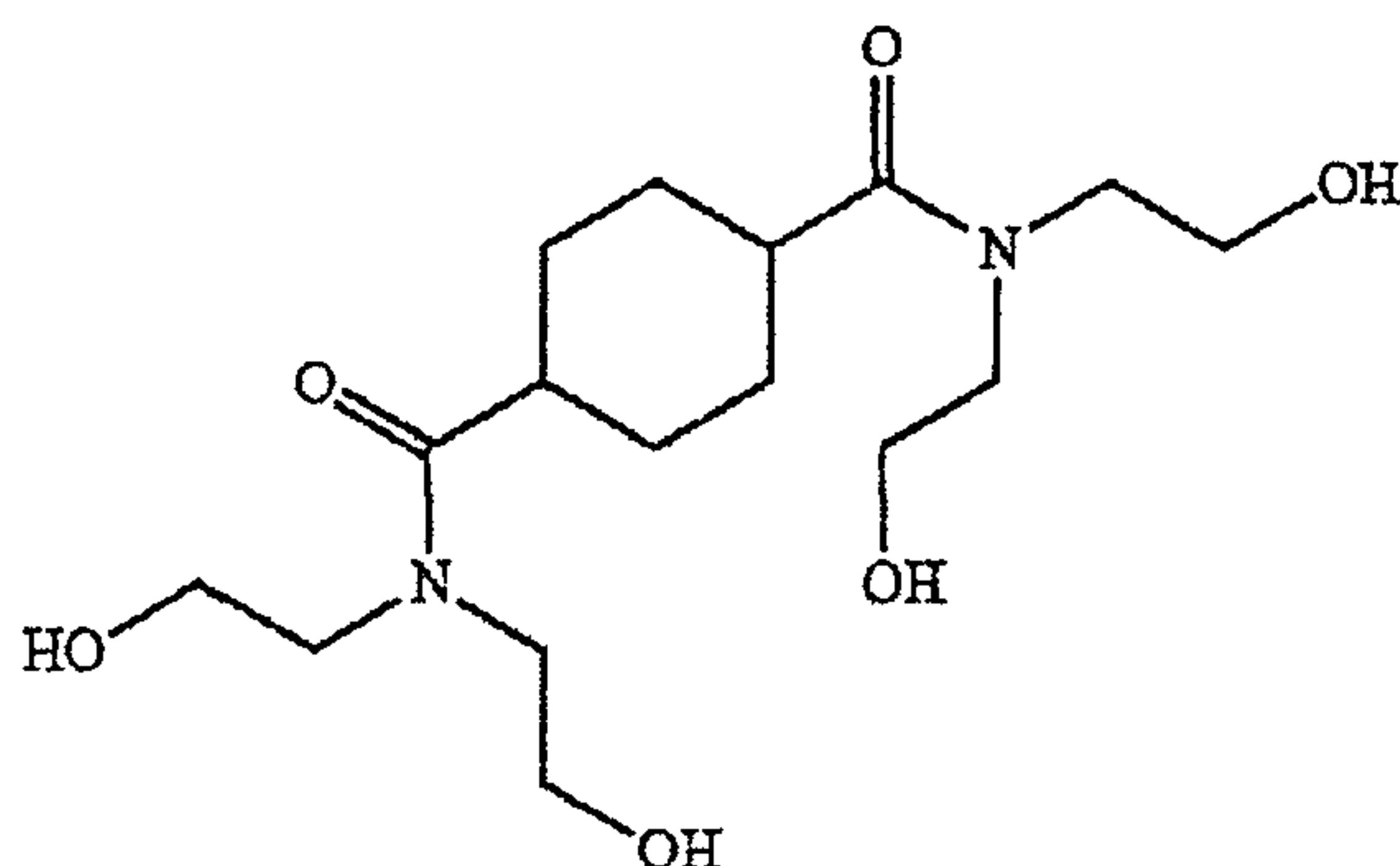
Peak #	Degrees 2theta \pm 0.2 degree 2theta	d (Å)
1	14.90	5.94
2	16.70	5.31
3	17.40	5.09
4	21.20	4.19
5	21.60	4.11
6	26.00	3.43

26. The β -hydroxyalkylamide of claim 25, wherein the N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide has a trans content on the cyclohexyl ring of greater than or equal to 92 mol %, based on a total amount of all of the isomers of N,N,N',N'-tetrakis(2-hydroxyethyl)-cyclohexyl-1,4-diamide.
27. The β -hydroxyalkylamide of claim 25, wherein the β -hydroxyalkylamide of the formula XIIA is present in solid form below 175° C.
28. The β -hydroxyalkylamide of claim 25, wherein a concentration of all of the isomers of N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide in the end product after its preparation is 75% by mass.
29. The β -hydroxyalkylamide of claim 25, wherein a ratio of the enthalpies of endothermic peak 1 (~160° C) to endothermic peak 2 (~190° C) is 1:1 to 1:3.
30. The β -hydroxyalkylamide of claim 25, wherein the N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide of formula XIIA:

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XIIA



has the following parameters:

a trans content on the cyclohexyl ring of greater than or equal to 70 mol %, based on the total amount of all of the isomers of N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide;

two endothermic peaks according to DSC analysis (differential scanning calorimetry), where peak 1 is situated in the region of 140-170° C, with a maximum of 155-165° C, and peak 2 is situated in the region of 170-210° C, with a maximum of 175-207° C;

a ratio of the enthalpies of endothermic peak 1 to endothermic peak 2 is 1:1 to 1:5;

a XRPD spectrum of the powder sample in an x-ray diffractometer, measured with Cu K α radiation (1.541 Å) has the following peaks:

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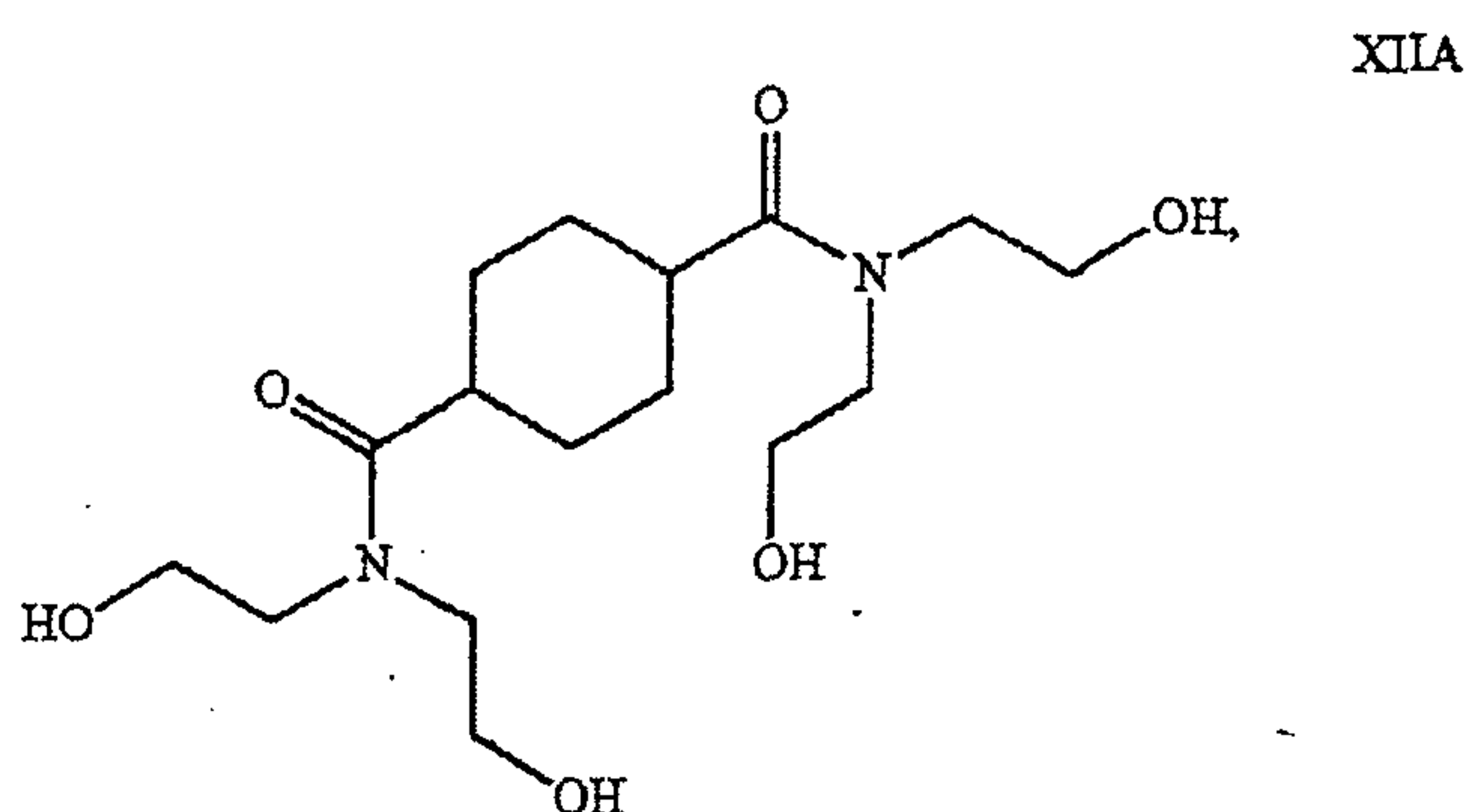
5

Peak #	Degrees 2theta ± 0.2 degree 2theta	d (Å)
1	14.90	5.94
2	16.70	5.31
3	17.40	5.09
4	21.20	4.19
5	21.60	4.11
6	26.00	3.43; and

10 according to x-ray structural analysis of a single crystal, has the following crystal parameters:

Crystal system:	Orthorhombic;	
Space group:	Pbca;	
Unit cell dimensions:	a = 10.06350(10) Å	$\alpha = 90^\circ$,
	b = 11.85290(10) Å	$\beta = 90^\circ$,
	c = 14.6275(2) Å	$\gamma = 90^\circ$,
Volume:	1744.79(3) Å ³ .	

31. A method for the discontinuous preparation of an N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to formula XIA:



15 the method comprising reacting dimethyl 1,4-cyclohexyldicarboxylate and diethanolamine in a solvent in an extruder, intensive compounder, intensive mixer or static mixer, wherein the N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide has the following parameters:

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a trans content on the cyclohexyl ring of greater than or equal to 70 mol %, based on the total amount of all of the isomers of N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide;

two endothermic peaks according to DSC analysis (differential scanning calorimetry), where peak 1 is situated in the region of 140-170° C, with a maximum of 155-165° C, and peak 2 is situated in the region of 170-210° C, with a maximum of 175-207° C;

a ratio of the enthalpies of endothermic peak 1 to endothermic peak 2 is 1:1 to 1:5; and

an XRPD spectrum of the powder sample in an x-ray diffractometer, measured with Cu K α radiation (1.541 Å) has the following peaks:

Peak #	Degrees 2theta \pm 0.2 degree 2theta	d (Å)
1	14.90	5.94
2	16.70	5.31
3	17.40	5.09
4	21.20	4.19
5	21.60	4.11
6	26.00	3.43

32. The method of claim 31, wherein the reacting occurs at temperatures from 20 to 120° C.

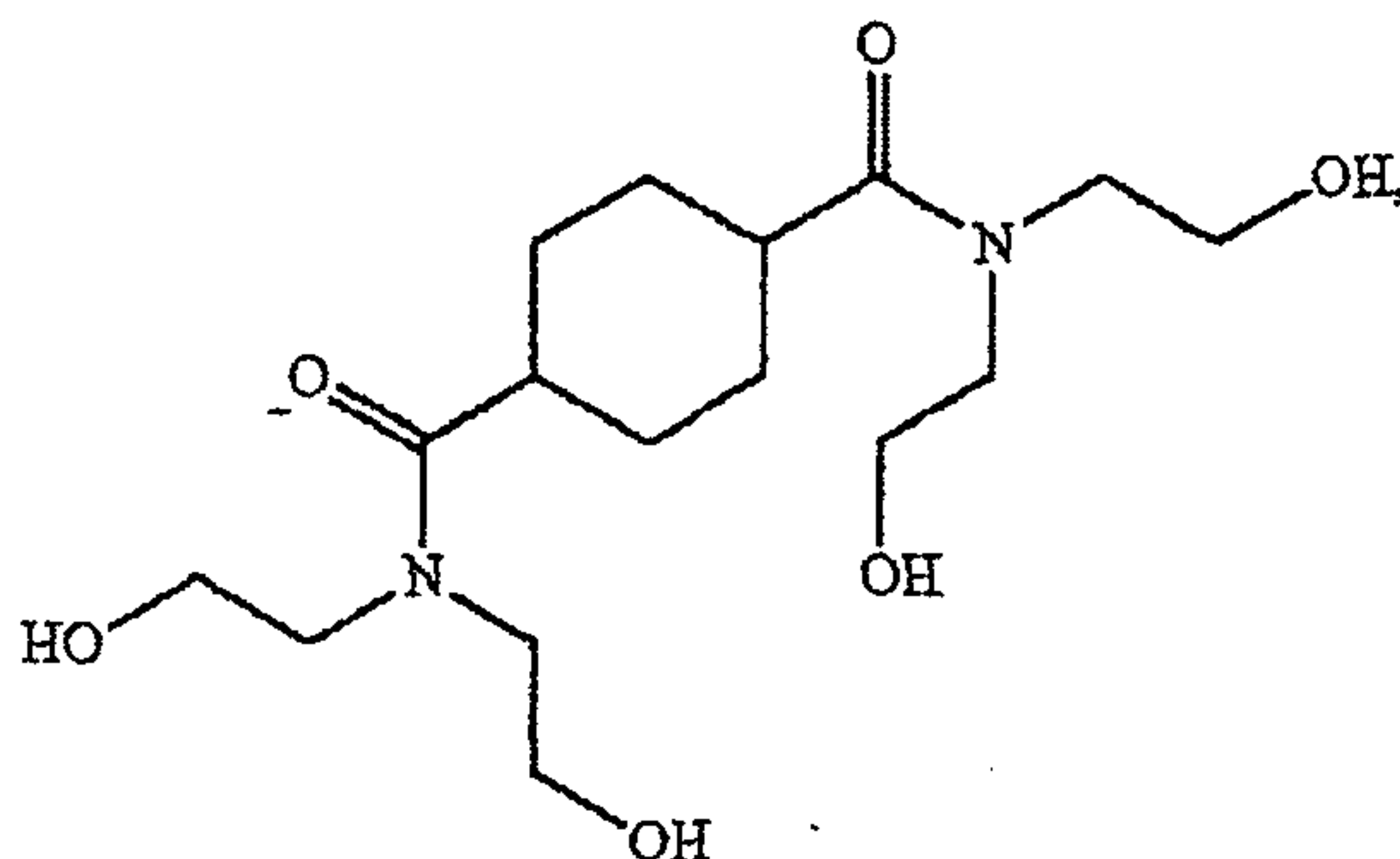
33. The method of claim 31, wherein an amount of solvent added is greater than 10% by weight, based on an total amount of all of the reactants.

34. A method for the discontinuous preparation of an N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide according to formula XIA:

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XIIA



the method comprising reacting dimethyl 1,4-cyclohexyldicarboxylate and diethanolamine in closed apparatus under pressure at temperatures from 60 to 140° C without addition of solvents, wherein the N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide has the following parameters:

a trans content on the cyclohexyl ring of greater than or equal to 70 mol %, based on the total amount of all of the isomers of N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide that are present;

two endothermic peaks according to DSC analysis (differential scanning calorimetry), where peak 1 is situated in the region of 140-170° C, with a maximum of 155-165° C, and peak 2 is situated in the region of 170-210° C, with a maximum of 175-207° C;

a ratio of the enthalpies of endothermic peak 1 to endothermic peak 2 is 1:1 to 1:5; and

an XRPD spectrum of the powder sample in an x-ray diffractometer, measured with Cu K α radiation (1.541 Å) has the following peaks:

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Peak #	Degrees 2theta \pm 0.2 degree 2theta	d (Å)
1	14.90	5.94
2	16.70	5.31
3	17.40	5.09
4	21.20	4.19
5	21.60	4.11
6	26.00	3.43

35. The method of claim 34, further comprising recrystallizing the N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide, wherein the reacting occurs at temperatures from 20 to 140° C

5 36. The method of claim 34, wherein:

the method further comprises:

recrystallizing the N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide; or

thermally conditioning the N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-
10 1,4-diamide at temperatures of 50-100° C

37. The method of claim 34

wherein an x-ray structural analysis of a single crystal of the N,N,N',N'-tetrakis-(2-hydroxyethyl)cyclohexyl-1,4-diamide, has the following parameters:

Crystal system:	Orthorhombic	
Space group:	Pbca	
Unit cell dimensions:	a = 10.06350(10) Å	$\alpha = 90^\circ$
	b = 11.85290(10) Å	$\beta = 90^\circ$
	c = 14.6275(2) Å	$\gamma = 90^\circ$
Volume:	1744.79(3) Å ³	

15 38. A process for producing a coating, the process comprising applying the β -hydroxyalkylamide of claim 25 to a substrate.

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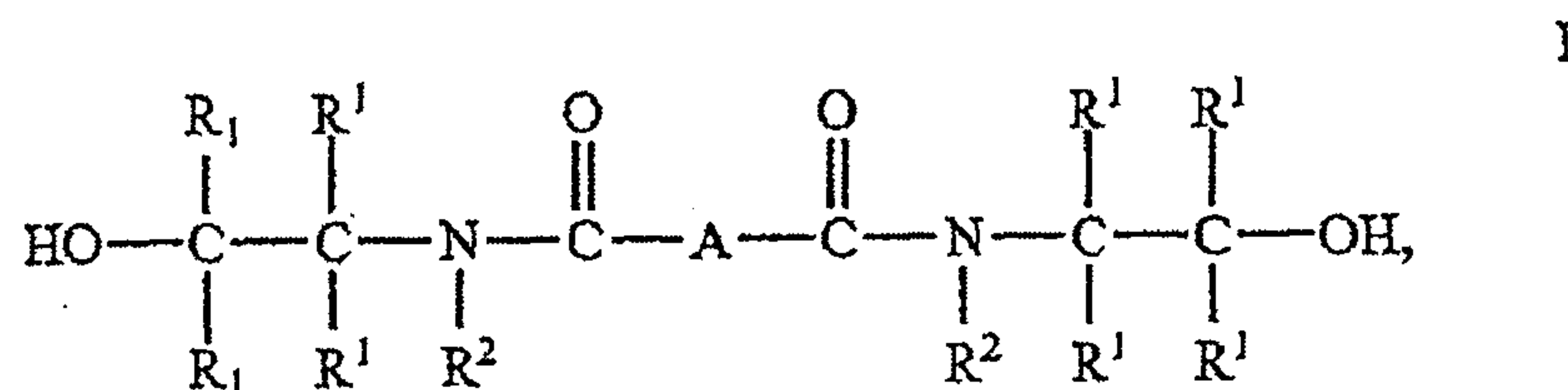
39. The process of claim 38, wherein an x-ray structural analysis of a single crystal of the N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide, has the following parameters:

Crystal system:	Orthorhombic	
Space group:	Pbca	
Unit cell dimensions:	a = 10.06350(10) Å	$\alpha = 90^\circ$.
	b = 11.85290(10) Å	$\beta = 90^\circ$.
	c = 14.6275(2) Å	$\gamma = 90^\circ$.
Volume:	1744.79(3) Å ³ .	

5 40. A powder coating material, comprising the β -hydroxyalkylamide of claim 25.

41. The method of claim 38 which produces a coating having a matt surface with <50 gloss units, measured as reflectometer values according to DIN 67530/ISO 2813 at an incident angle of 60°.

10 42. A β -hydroxyalkylamide, comprising two or three or four β -hydroxyalkylamide groups per molecule of the formula I:

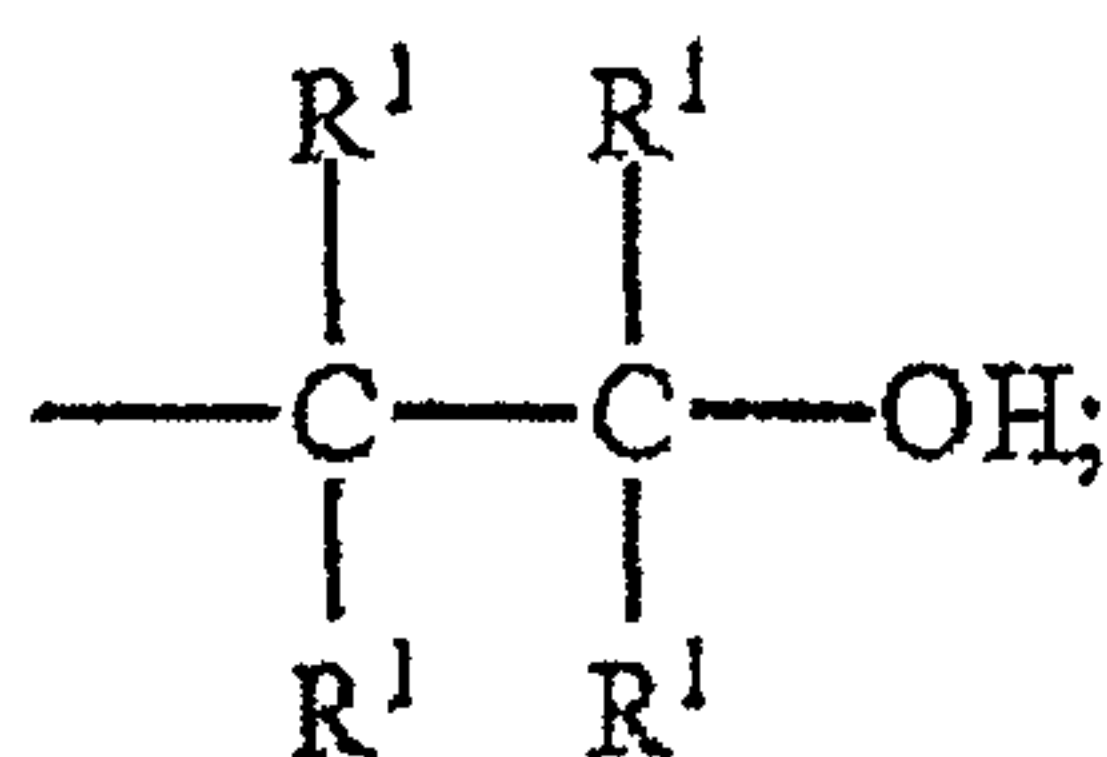


wherein:

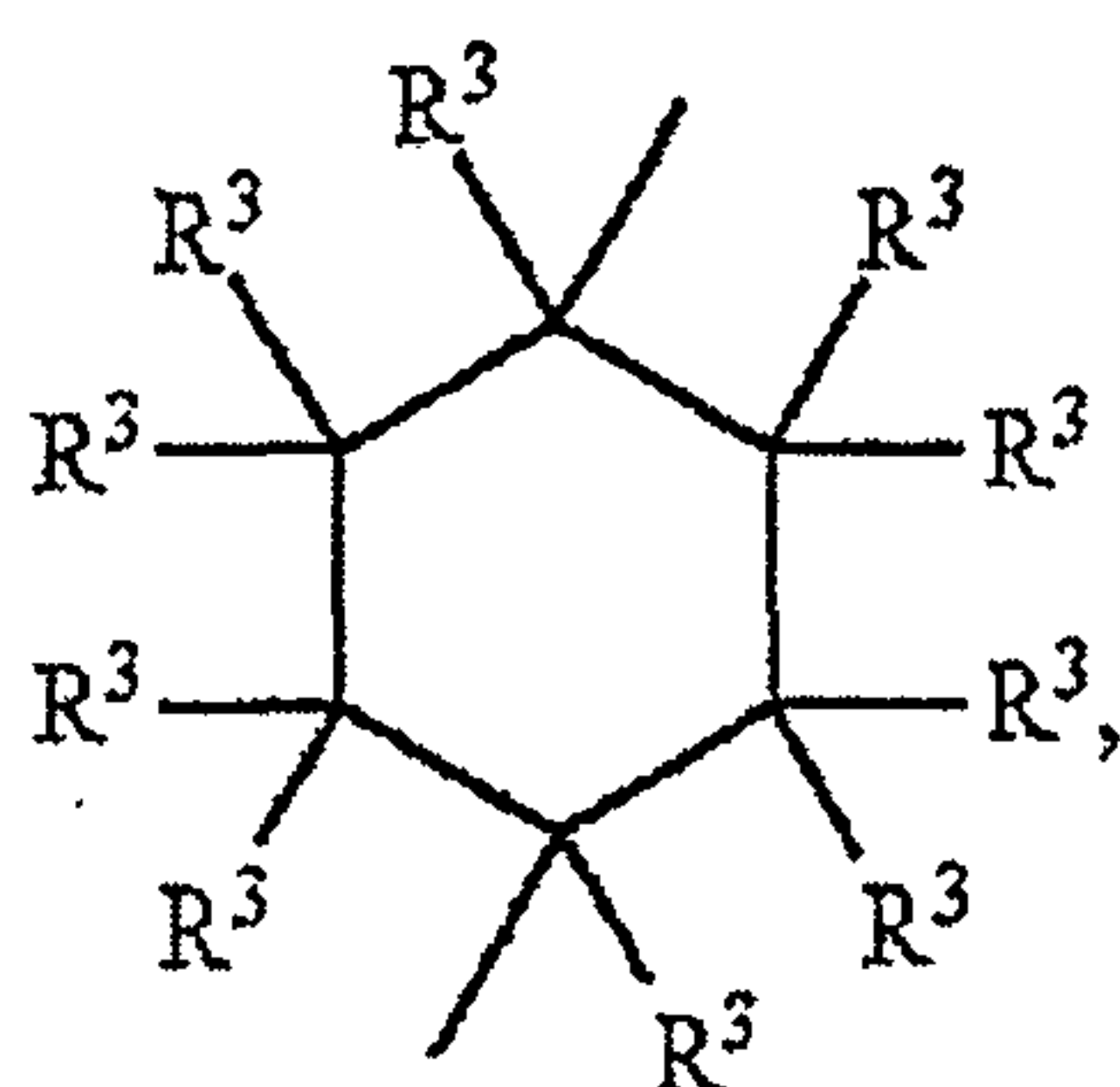
15 R^1 and R^2 independently of one another are identical or different radicals selected from the group consisting of an alkyl radical, a cycloalkyl radical, an aryl radical, an aralkyl radical and an alkenyl radical having 1-24 carbon atoms, such that the radicals optionally comprise a heteroatom, R^1 is optionally a hydrogen, and R^2 is optionally represented by the formula:

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A is represented by A¹:

A¹

- wherein radicals R³ independently of one another are identical or
 5 different radicals selected from the group consisting of hydrogen, an alkyl radical, a cycloalkyl radical, an aryl radical, an aralkyl radical, and an alkenyl radical having 1-24 carbon atoms, such that the radicals optionally comprise a heteroatom and two or more substituents R³ are optionally linked with one another to form rings;

wherein the β-hydroxyalkylamide is present in solid form below 150° C;

- 10 wherein the β-hydroxyalkylamide has a trans content of greater than or equal to 70 mol % based on the total amount of all of the isomers of the β-hydroxyalkylamide;

- two endothermic peaks according to DSC analysis (differential scanning calorimetry), where peak 1 is situated in the region of 140-170° C, with a maximum of
 15 155-165° C, and peak 2 is situated in the region of 170-210° C, with a maximum of 175-207° C;

a ratio of the enthalpies of endothermic peak 1 to endothermic peak 2 is 1:1 to 1:5; and

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a XRPD spectrum of the powder sample in an x-ray diffractometer, measured with Cu K α radiation (1.541 Å) has the following peaks:

Peak #	Degrees 2theta \pm 0.2 degree 2theta	d (Å)
1	14.90	5.94
2	16.70	5.31
3	17.40	5.09
4	21.20	4.19
5	21.60	4.11
6	26.00	3.43.

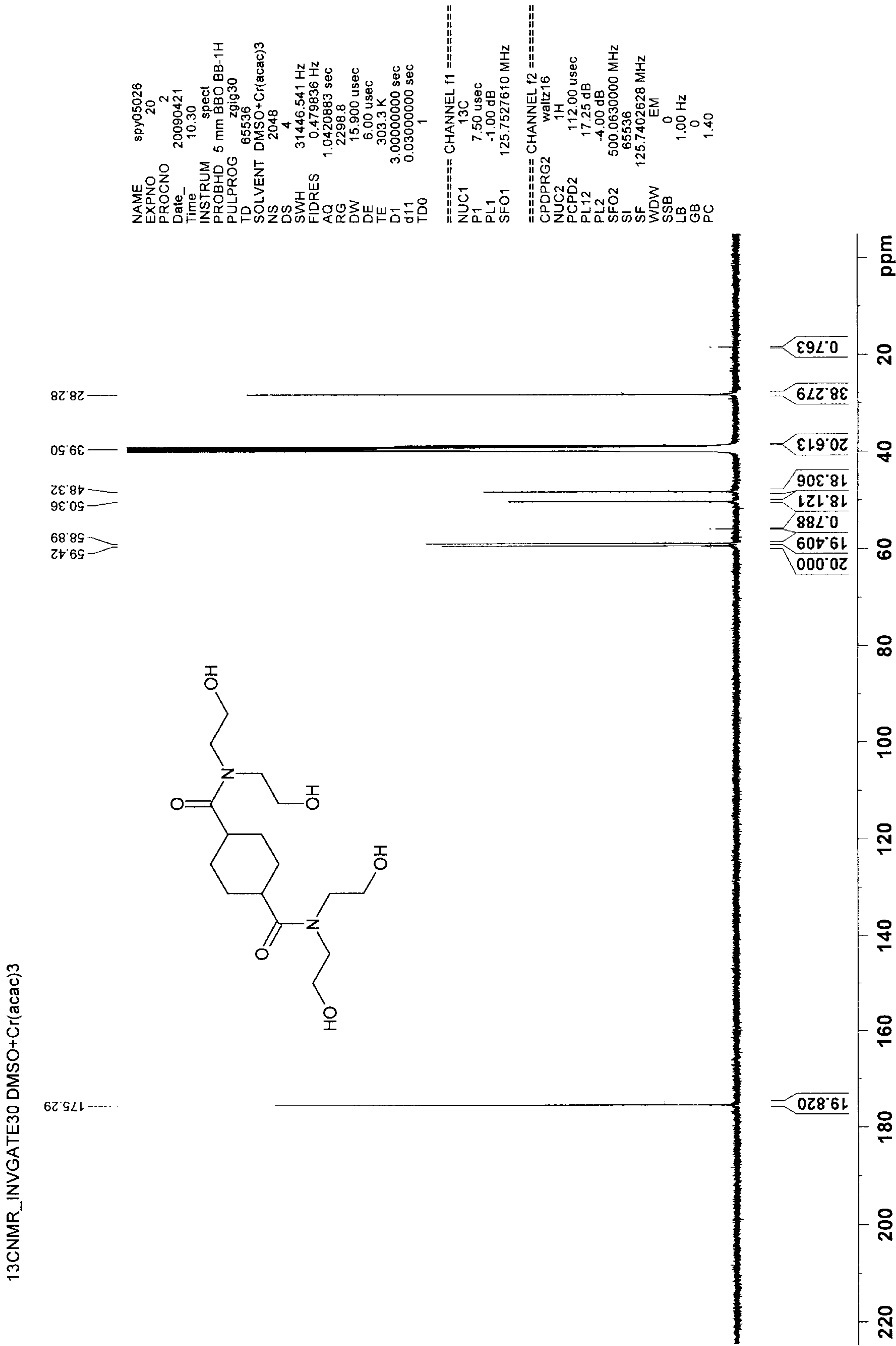
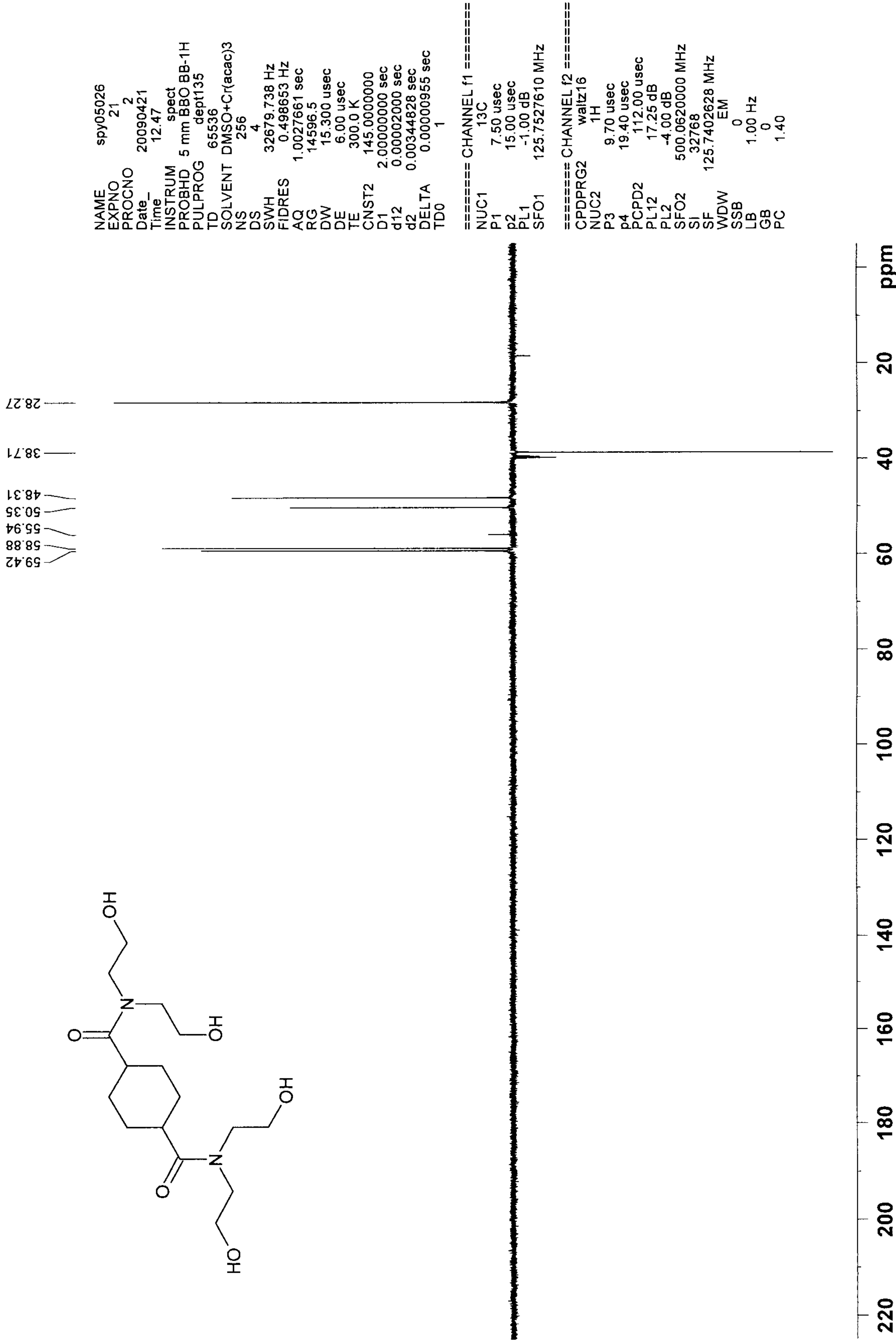
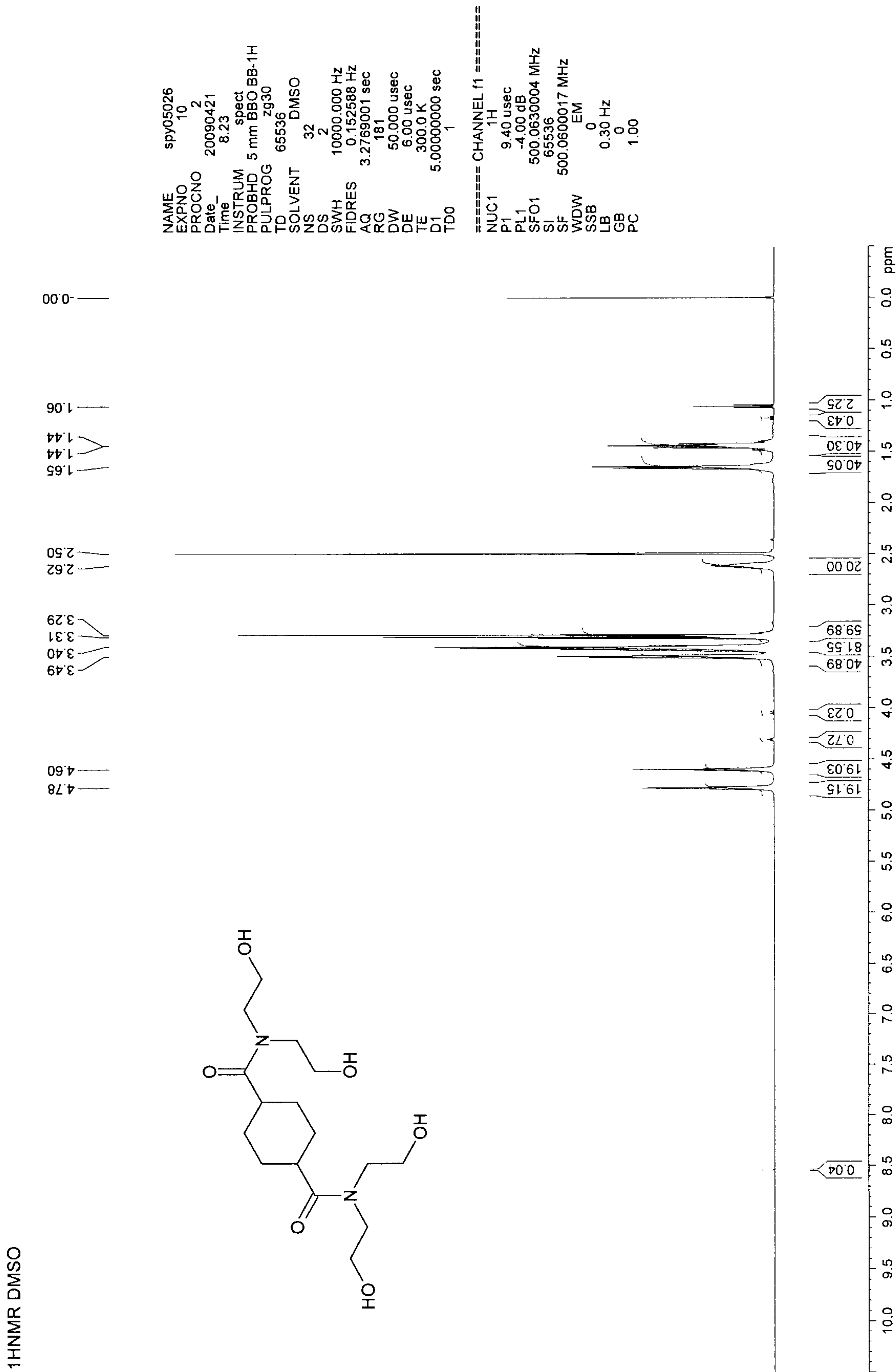


Figure 1

13CNMR_DEPT135 DMSO+Cr(acac)3

Figure 2





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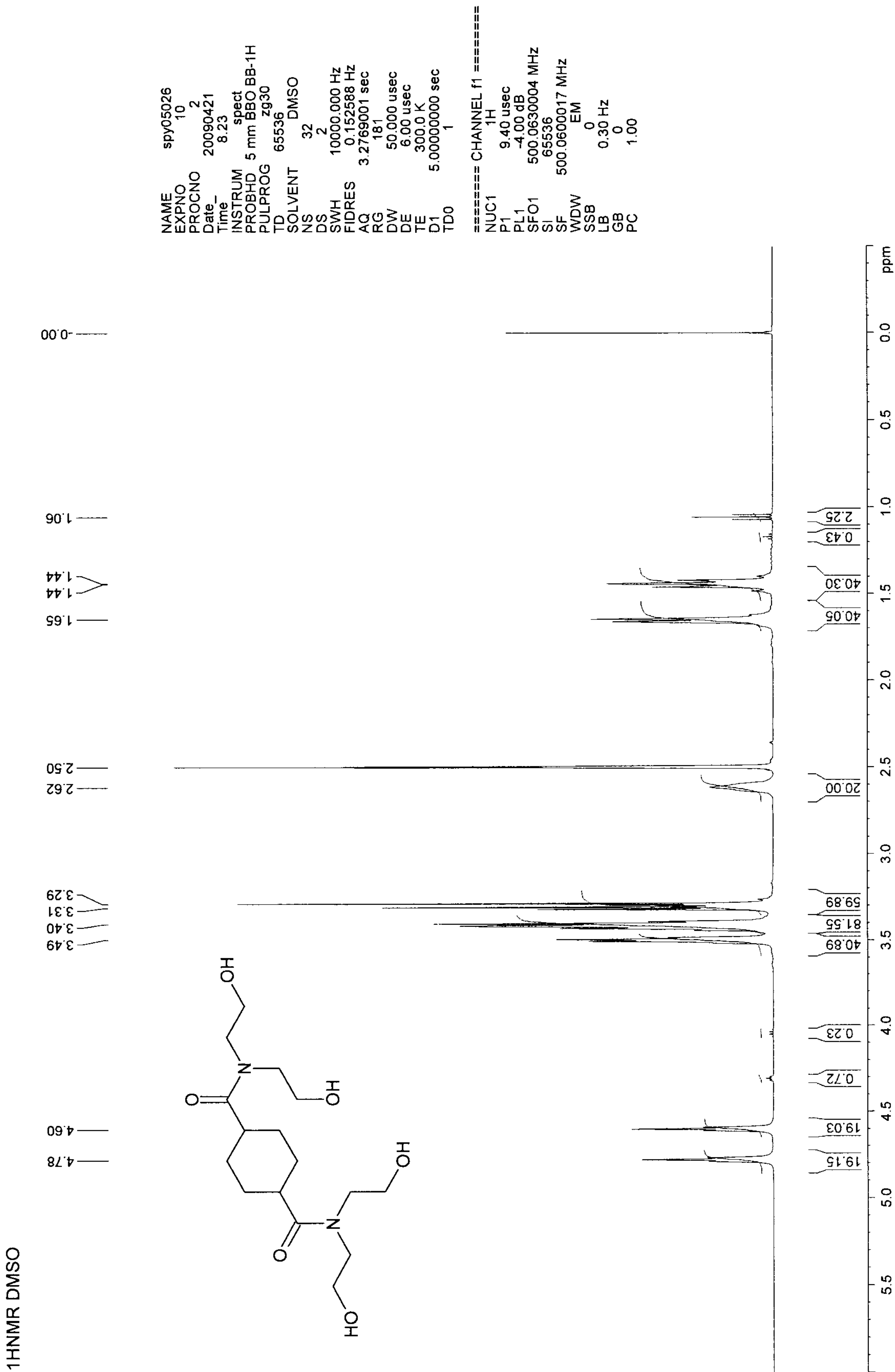


Figure 4

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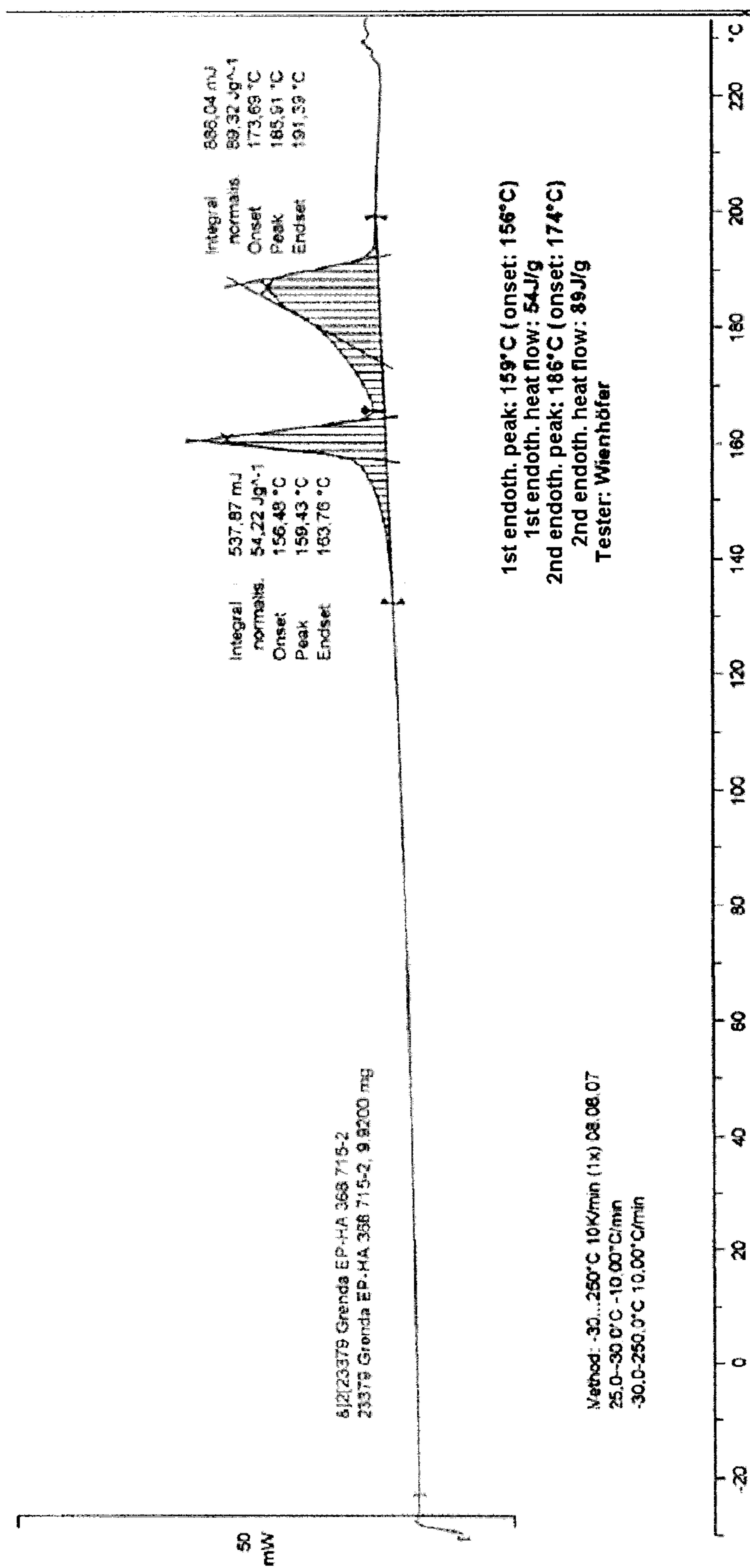


Figure 5: DSC diagram of the β -hydroxyalkylamide of the formula XIIA described in example 3a

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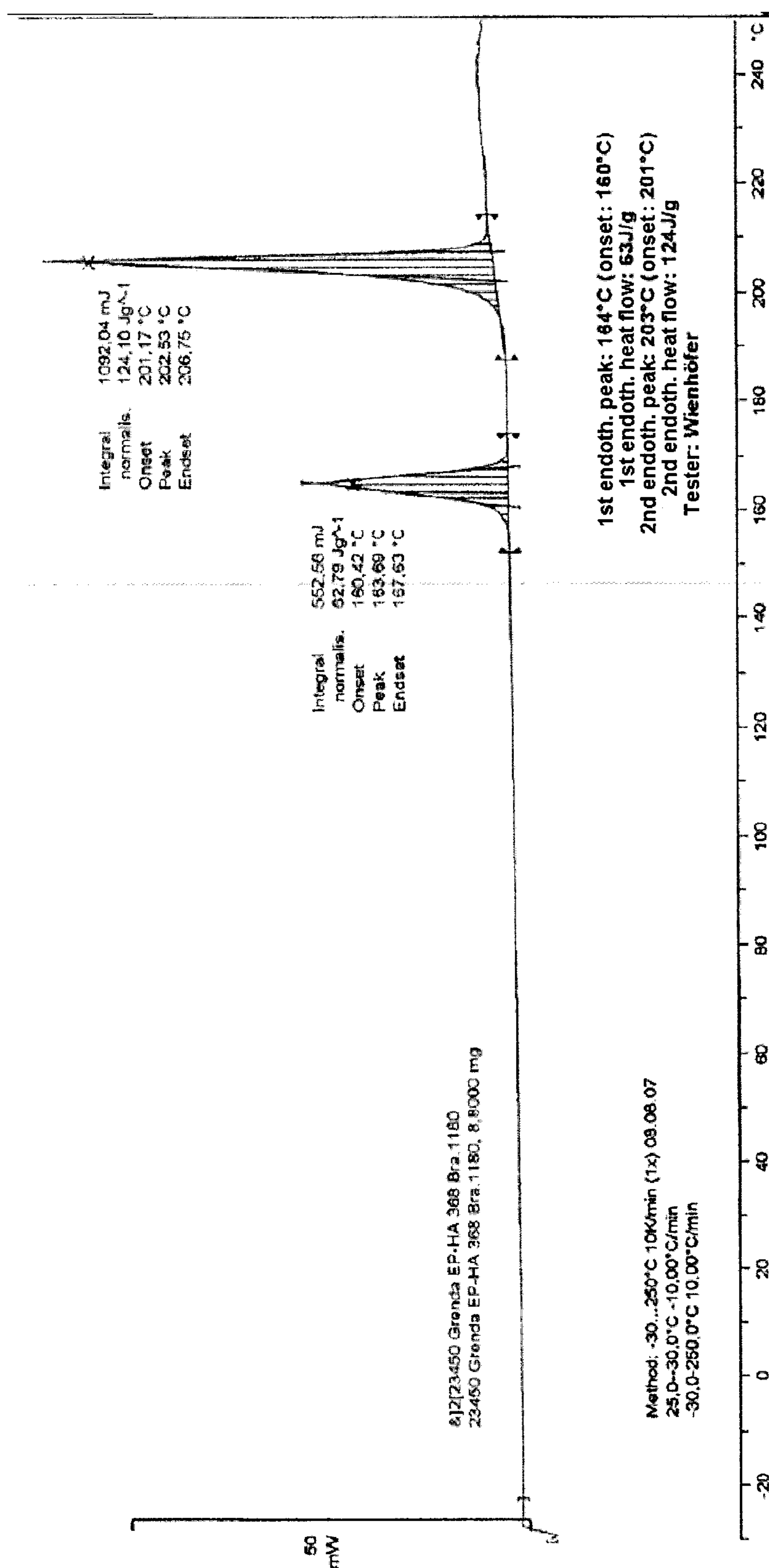


Figure 6: DSC diagram of the β -hydroxyalkylamide of the formula XIIA described in example 3b

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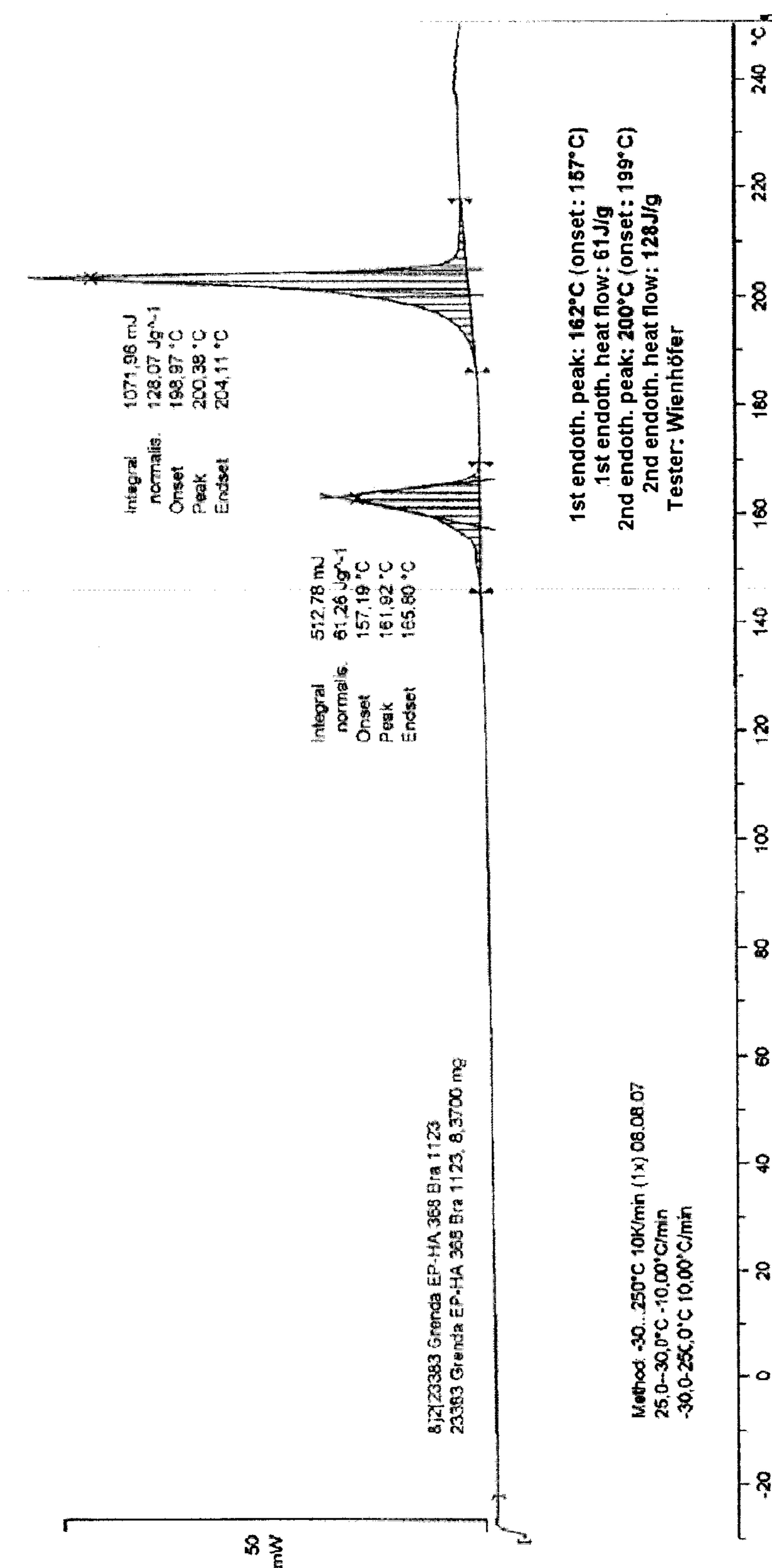


Figure 7: DSC diagram of the β -hydroxyalkylamide of the formula XIIA described in example 4b

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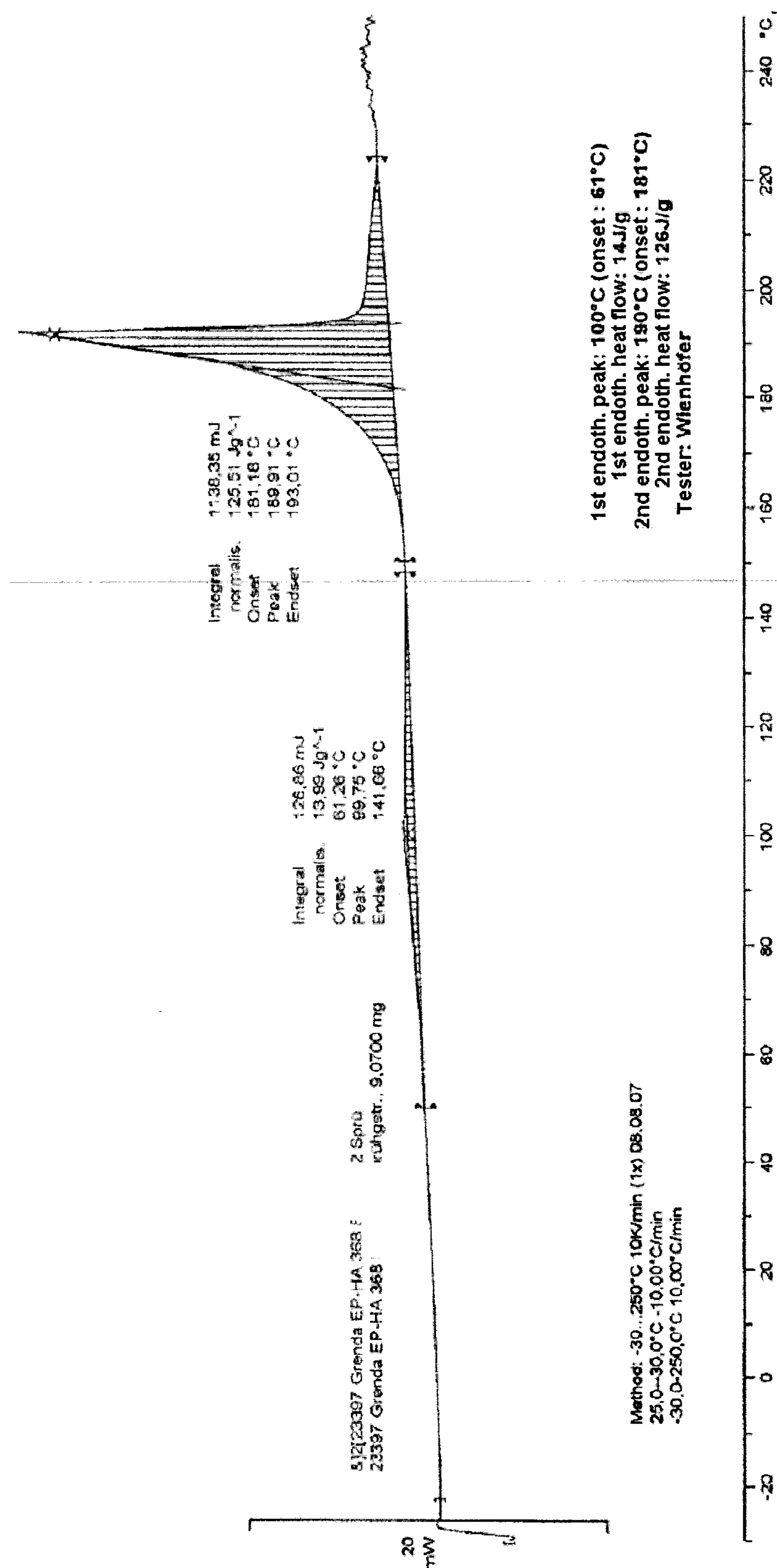


Figure 8: DSC diagram of the β -hydroxyalkylamide described in example 4c

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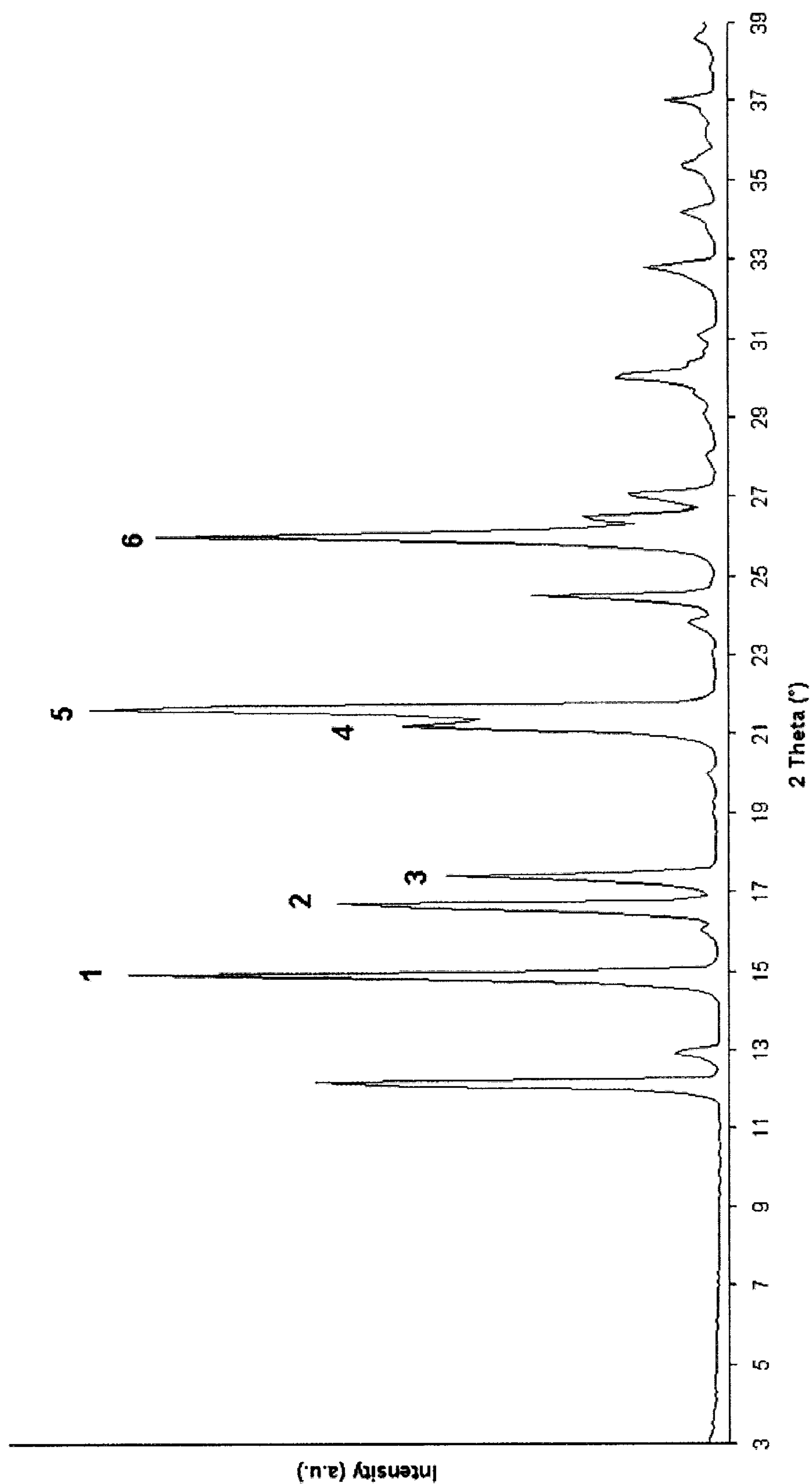


Figure 9: XRPD (x-ray powder diffraction) analysis of the β -hydroxyalkylamide of the formula XIIA described in example 3a (matting material)

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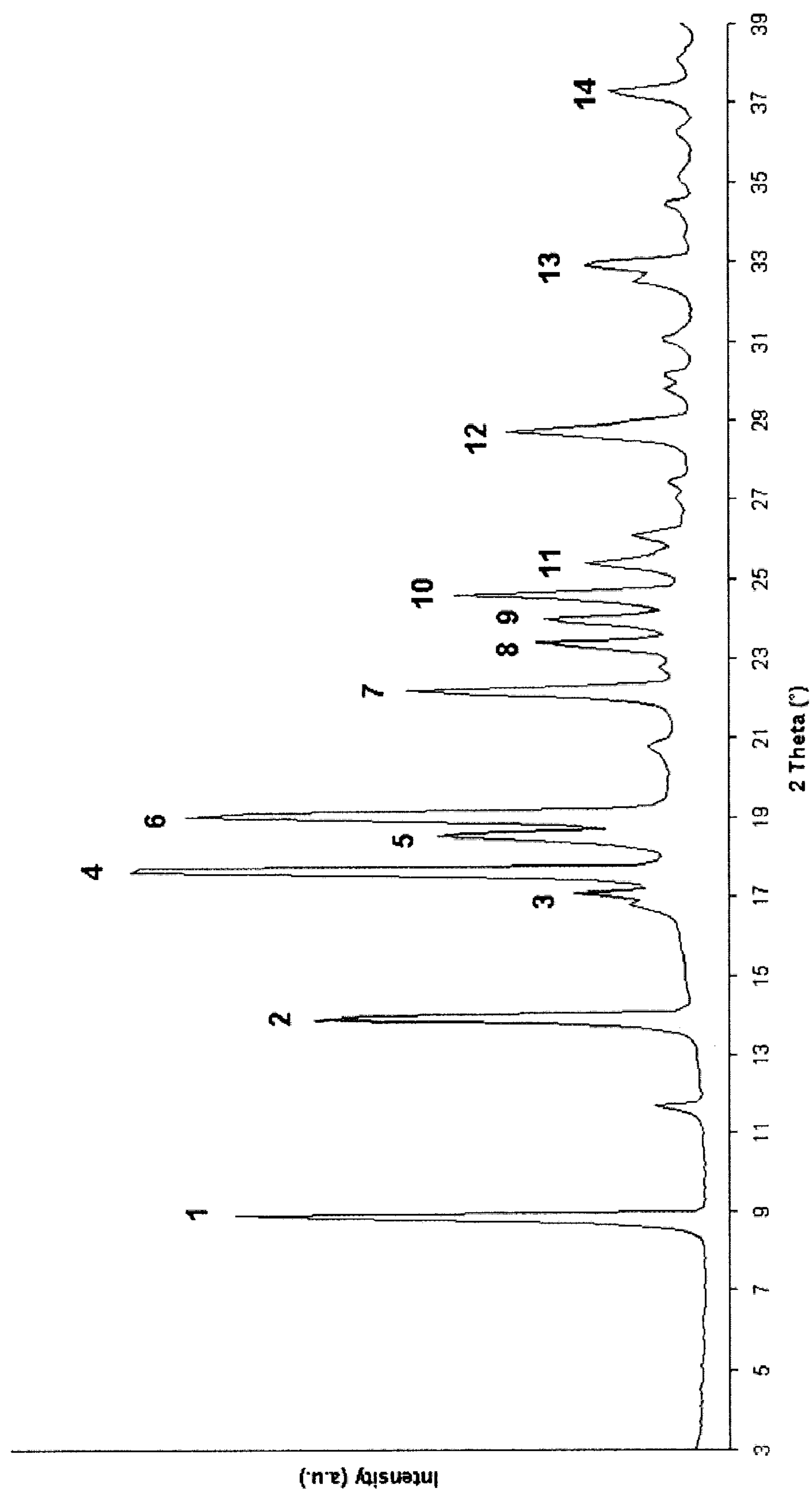


Figure 10: XRPD (x-ray powder diffraction) analysis of the β -hydroxyalkylamide described in example 4c (nonmatting material)

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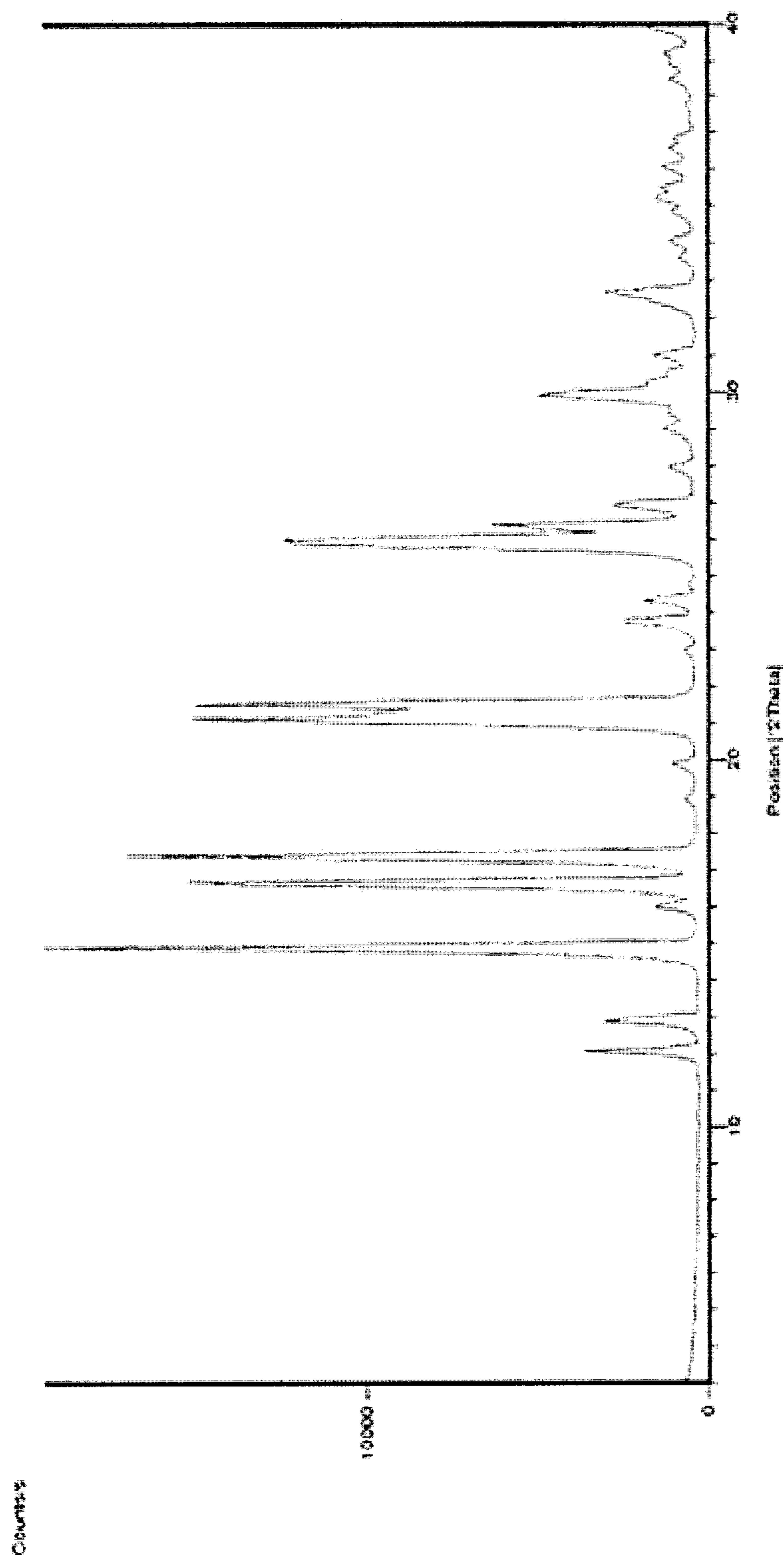
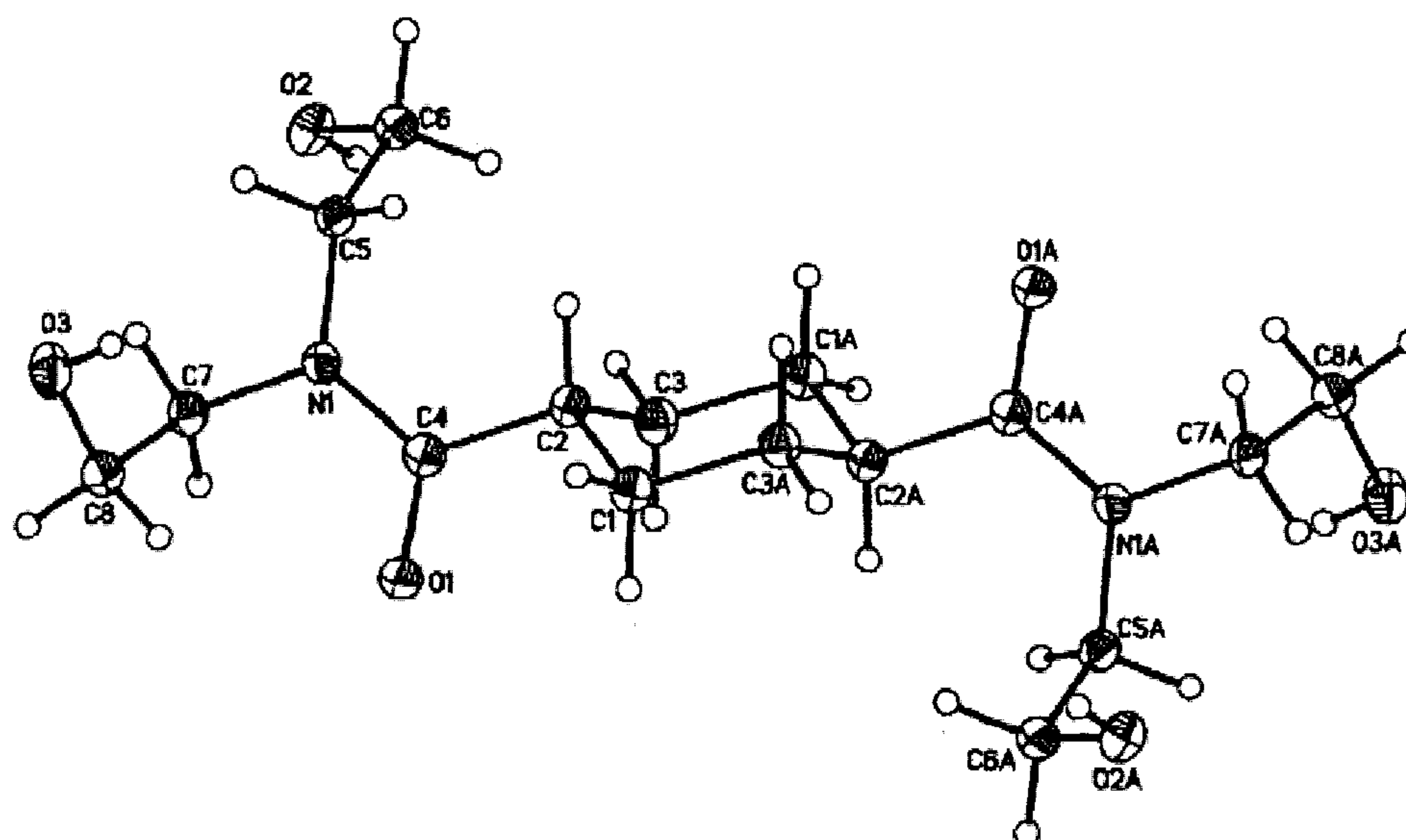


Figure 11: XRPD (x-ray powder diffraction) analysis of the β -hydroxyalkylamide of the formula XIIA described in example 4b (matting material)

Figure 12: Ortep plot (50%) with numbering scheme



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Figure 13:

Calculated powder diffractogram based on the single-crystal structural determination of N,N,N',N'-tetrakis(2-hydroxyethyl)cyclohexyl-1,4-diamide (vesta sample)

