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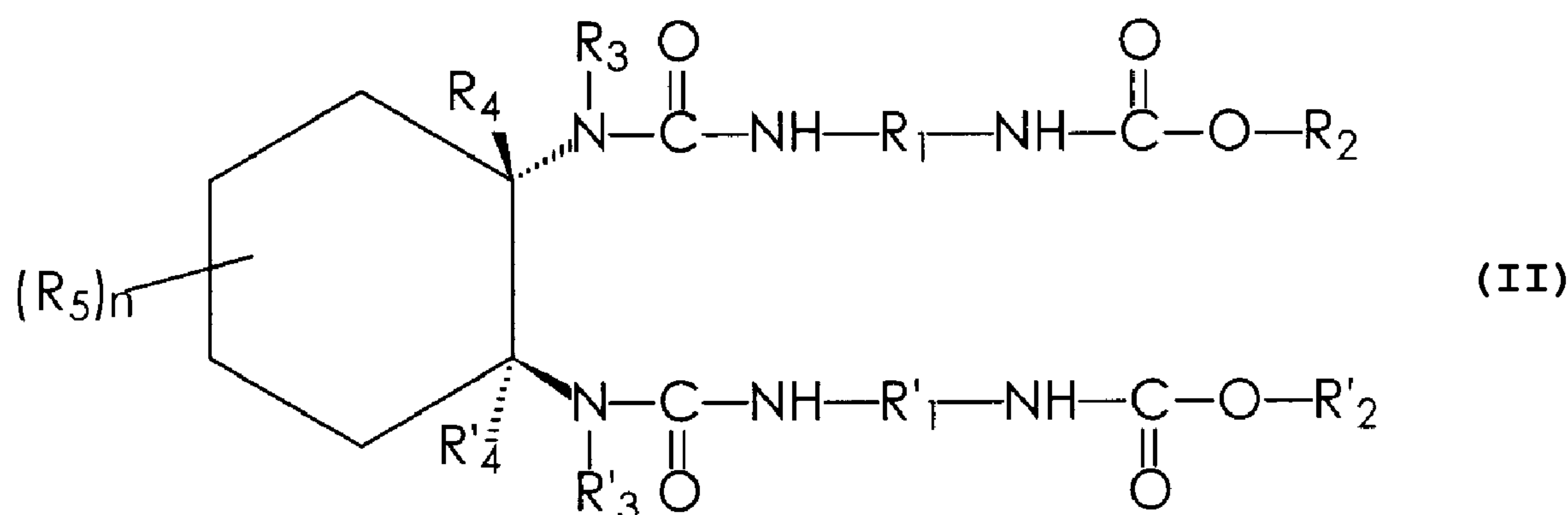
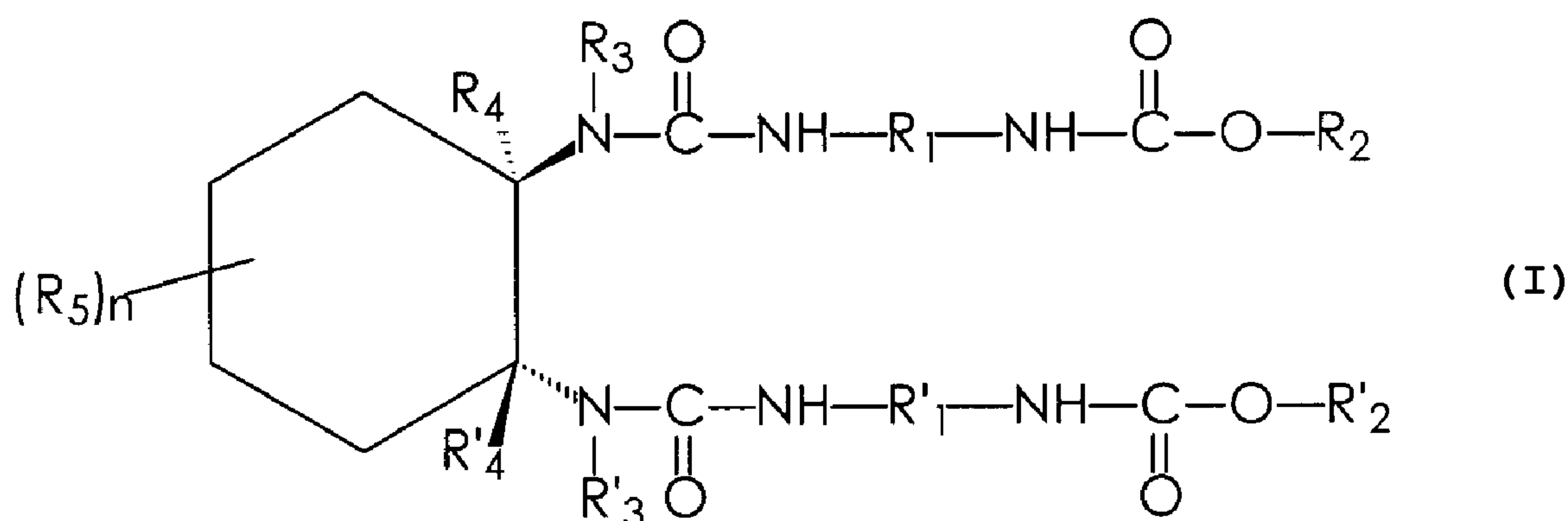
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(54) Titre : ENCRE A CHANGEMENT DE PHASE CONTENANT DES COMPOSES DE TRANS-1,2-BIS[UREE-URETHANE]CYCLOHEXANE

(54) Title: PHASE CHANGE INKS CONTAINING TRANS-1,2-CYCLOHEXANE BIS[UREA-URETHANE] COMPOUNDS



(57) Abrégé/Abstract:

Disclosed are phase change inks comprising a phase change ink carrier and a trans-1,2-cyclohexane bis[urea-urethane] compound of the formula (see formula I) (see formula II) or mixtures thereof, wherein R_1 and R'_1 each, independently of the other,

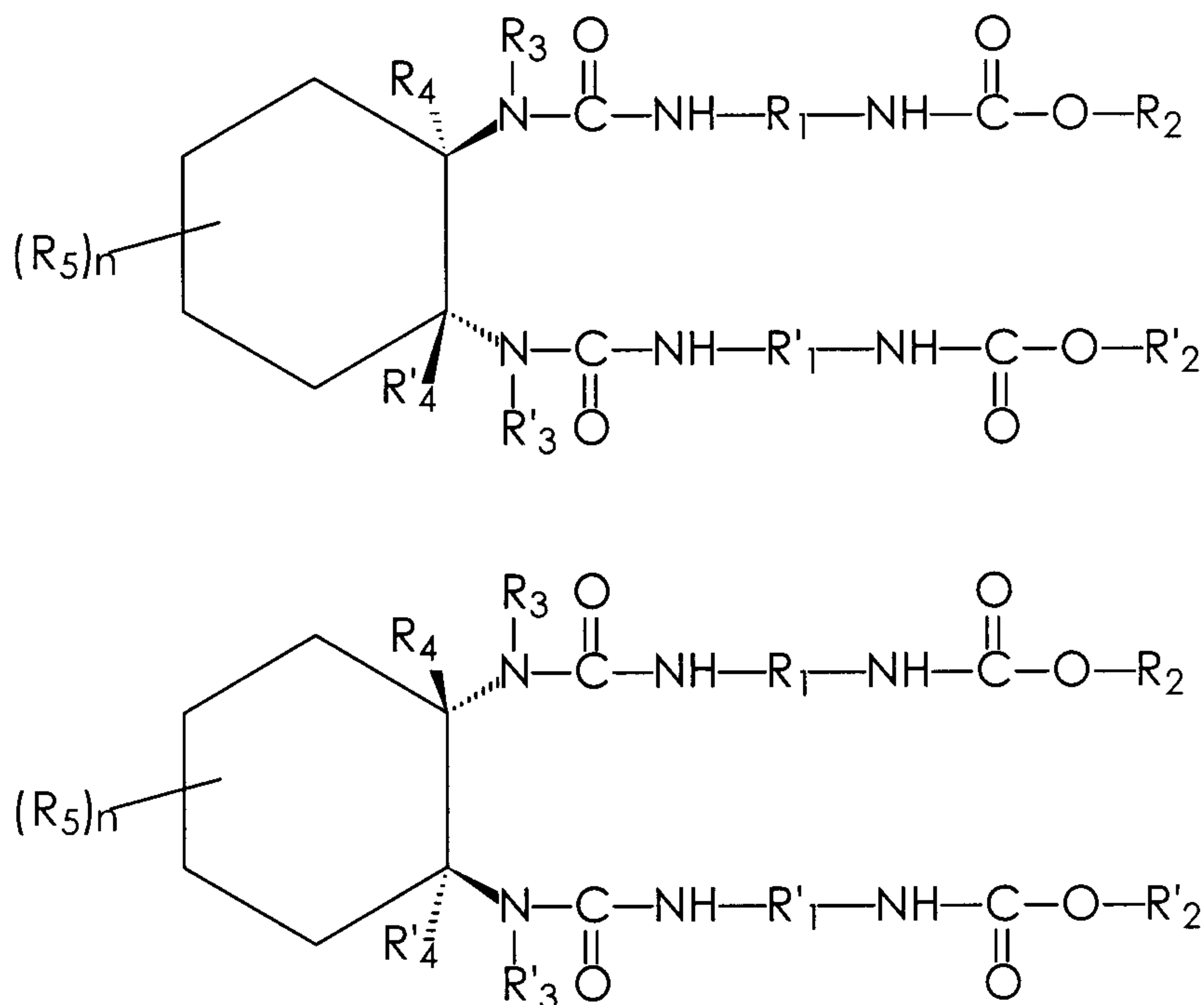


(57) Abrégé(suite)/Abstract(continued):

is an alkylene group, an arylene group, an arylalkylene group, or an alkylarylene group, R_2 and R'_2 each, independently of the other, is an alkyl group, an aryl group, an arylalkyl group, or an alkylaryl group, R_3 and R'_3 each, independently of the other, is a hydrogen atom or an alkyl group, R_4 and R'_4 each, independently of the other, is a hydrogen atom, a fluorine atom, an alkyl group, or a phenyl group, n is an integer of 0, 1, 2, 3, or 4, and R_5 is an alkyl group, an aryl group, an arylalkyl group, an alkylaryl group, or a substituent other than an alkyl, aryl, arylalkyl, or alkylaryl group.

ABSTRACT OF THE DISCLOSURE

Disclosed are phase change inks comprising a phase change ink carrier and a *trans*-1,2-cyclohexane bis[urea-urethane] compound of the formula

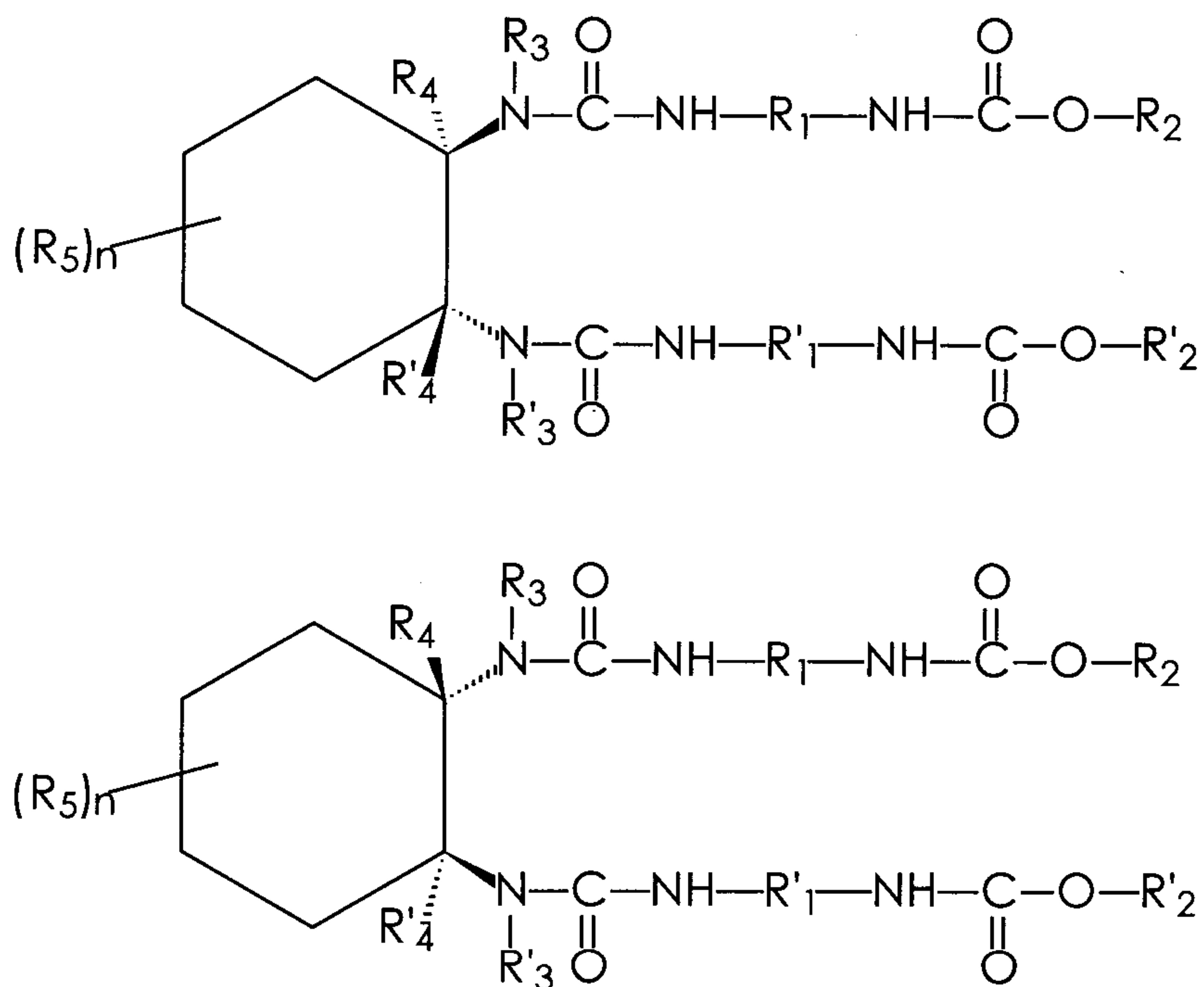


or mixtures thereof, wherein R_1 and R'_1 each, independently of the other, is an alkylene group, an arylene group, an arylalkylene group, or an alkylarylene group, R_2 and R'_2 each, independently of the other, is an alkyl group, an aryl group, an arylalkyl group, or an alkylaryl group, R_3 and R'_3 each, independently of the other, is a hydrogen atom or an alkyl group, R_4 and R'_4 each, independently of the other, is a hydrogen atom, a fluorine atom, an alkyl group, or a phenyl group, n is an integer of 0, 1, 2, 3, or 4, and R_5 is an alkyl group, an aryl group, an arylalkyl group, an alkylaryl group, or a substituent other than an alkyl, aryl, arylalkyl, or alkylaryl group.

PHASE CHANGE INKS CONTAINING
TRANS-1,2-CYCLOHEXANE BIS[UREA-URETHANE] COMPOUNDS

BACKGROUND

[0001] Disclosed herein are hot melt or phase change inks containing *trans*-1,2-cyclohexane bis[urea-urethane] compounds. One embodiment is directed to a phase change ink composition comprising a phase change ink carrier and a *trans*-1,2-cyclohexane bis[urea-urethane] compound of the formula



or mixtures thereof, wherein R_1 and R'_1 each, independently of the other, is an alkylene group, an arylene group, an arylalkylene group, or an alkylarylene group, R_2 and R'_2 each, independently of the other, is an alkyl group, an aryl group, an arylalkyl group, or an alkylaryl group, R_3 and R'_3 each, independently of the other, is a hydrogen atom or an alkyl group, R_4 and R'_4 each, independently of the other, is a hydrogen atom, a fluorine atom, an alkyl group, or a phenyl group, n is an integer

of 0, 1, 2, 3, or 4, and R_5 is an alkyl group, an aryl group, an arylalkyl group, an alkylaryl group, or a substituent other than an alkyl, aryl, arylalkyl, or alkylaryl group.

[0002] In general, phase change inks (sometimes referred to as "hot melt inks") are in the solid phase at ambient temperature, but exist in the liquid phase at the elevated operating temperature of an ink jet printing device. At the jet operating temperature, droplets of liquid ink are ejected from the printing device and, when the ink droplets contact the surface of the recording substrate, either directly or via an intermediate heated transfer belt or drum, they quickly solidify to form a predetermined pattern of solidified ink drops. Phase change inks have also been used in other printing technologies, such as gravure printing, as disclosed in, for example, U.S. Patent 5,496,879 and German Patent Publications DE 4205636AL and DE 4205713AL.

[0003] Phase change inks for color printing typically comprise a phase change ink carrier composition which is combined with a phase change ink compatible colorant. In a specific embodiment, a series of colored phase change inks can be formed by combining ink carrier compositions with compatible subtractive primary colorants. The subtractive primary colored phase change inks can comprise four component dyes, namely, cyan, magenta, yellow and black, although the inks are not limited to these four colors. These subtractive primary colored inks can be formed by using a single dye or a mixture of dyes. For example, magenta can be obtained by using a mixture of Solvent Red Dyes or a composite black can be obtained by mixing several dyes. U.S. Patent 4,889,560, U.S. Patent 4,889,761, and U.S. Patent 5,372,852, teach that the subtractive primary colorants employed can comprise dyes from the classes of Color Index (C.I.) Solvent Dyes, Disperse Dyes, modified Acid and Direct Dyes, and Basic Dyes. The

colorants can also include pigments, as disclosed in, for example, U.S. Patent 5,221,335. U.S. Patent 5,621,022, discloses the use of a specific class of polymeric dyes in phase change ink compositions.

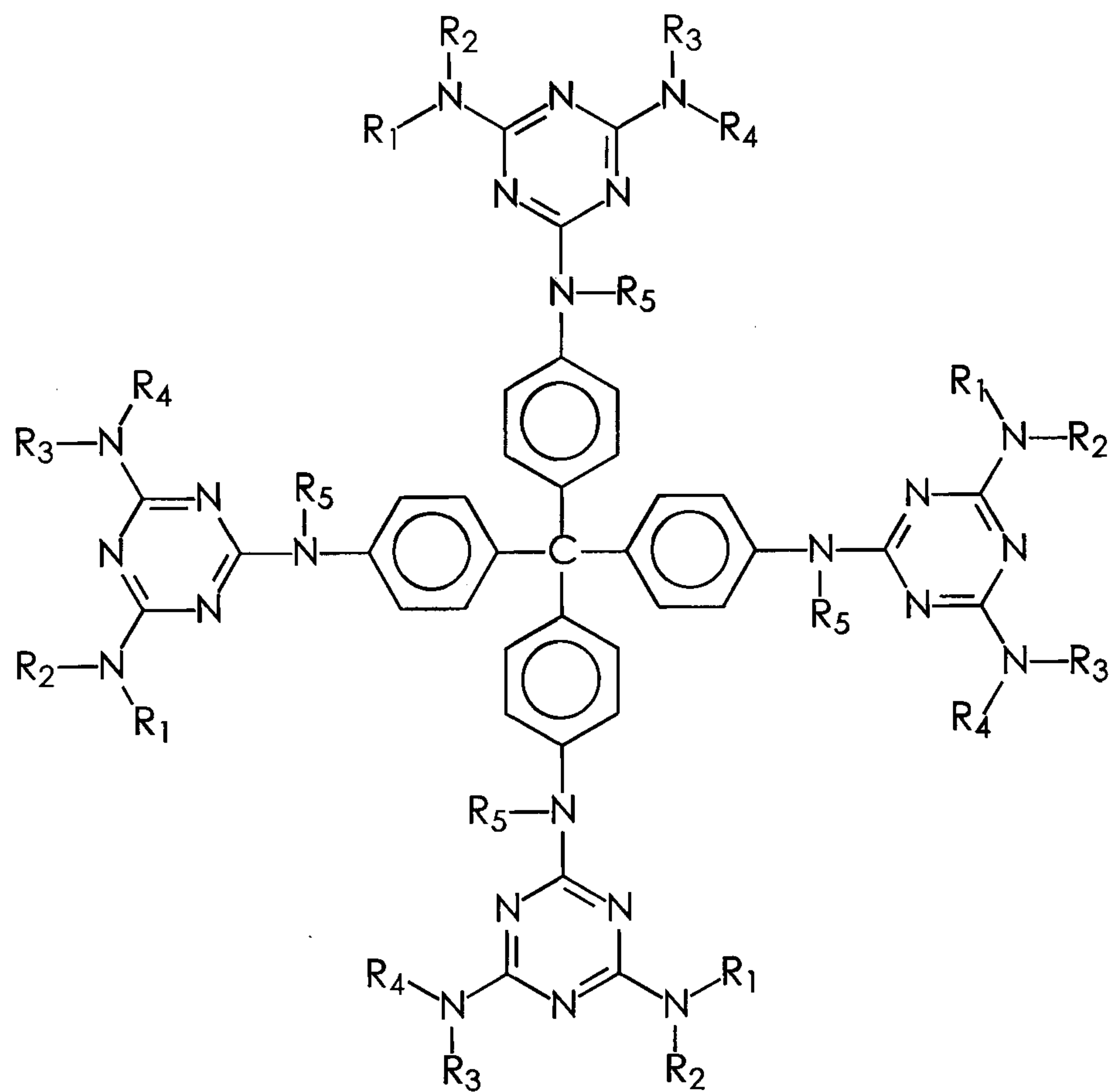
[0004] Phase change inks have also been used for applications such as postal marking, industrial marking, and labelling.

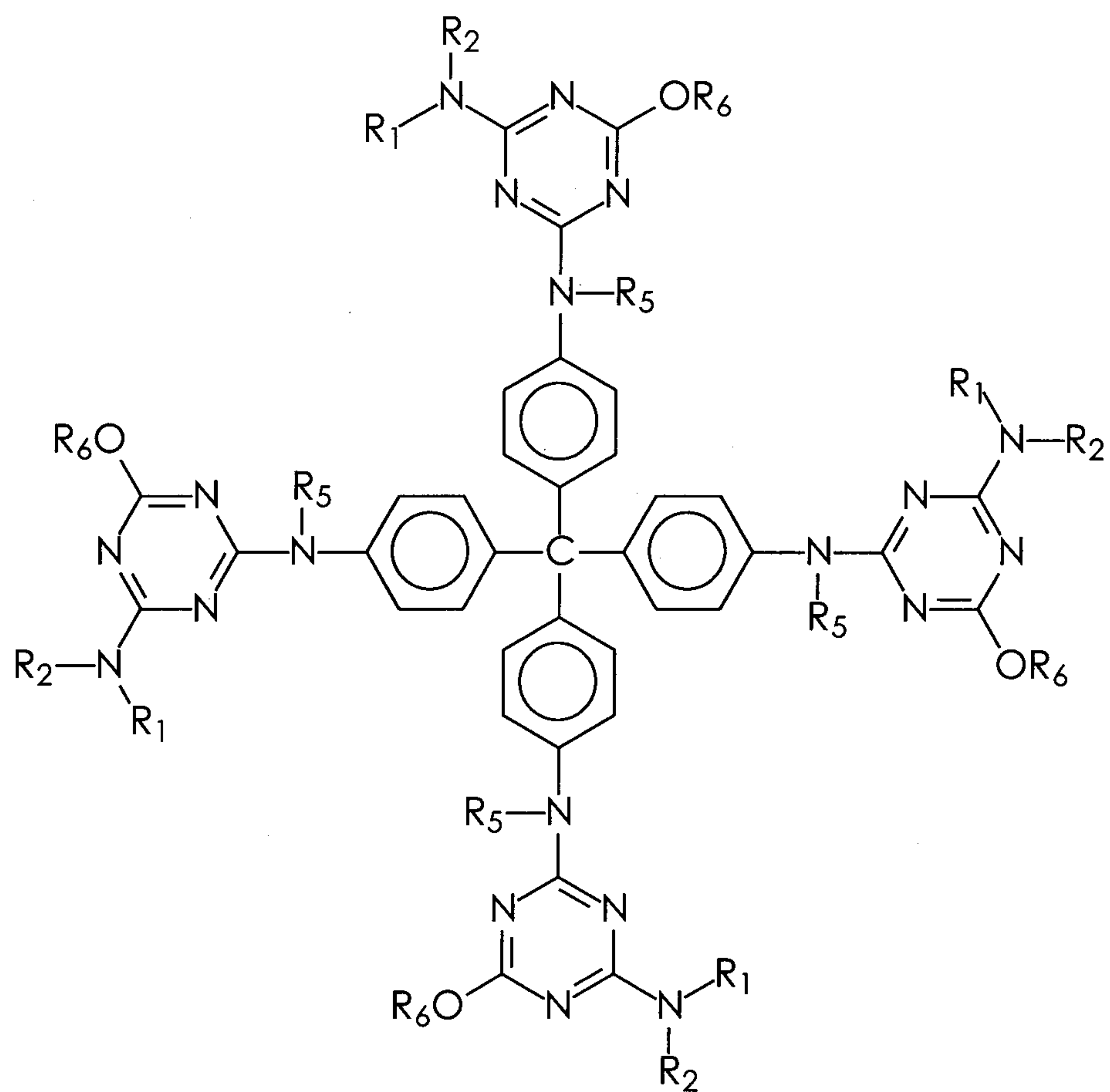
[0005] Phase change inks are desirable for ink jet printers because they remain in a solid phase at room temperature during shipping, long term storage, and the like. In addition, the problems associated with nozzle clogging as a result of ink evaporation with liquid ink jet inks are largely eliminated, thereby improving the reliability of the ink jet printing. Further, in phase change ink jet printers wherein the ink droplets are applied directly onto the final recording substrate (for example, paper, transparency material, and the like), the droplets solidify immediately upon contact with the substrate, so that migration of ink along the printing medium is prevented and dot quality is improved.

[0006] Compositions suitable for use as phase change ink carrier compositions are known. Some representative examples of references disclosing such materials include U.S. Patent 3,653,932, U.S. Patent 4,390,369, U.S. Patent 4,484,948, U.S. Patent 4,684,956, U.S. Patent 4,851,045, U.S. Patent 4,889,560, U.S. Patent 5,006,170, U.S. Patent 5,151,120, U.S. Patent 5,372,852, U.S. Patent 5,496,879, European Patent Publication 0187352, European Patent Publication 0206286, German Patent Publication DE 4205636AL, German Patent Publication DE 4205713AL, and PCT Patent Application WO 94/04619. Suitable carrier materials can include paraffins, microcrystalline waxes, polyethylene waxes, ester waxes, fatty acids and other waxy materials, fatty amide containing materials, sulfonamide materials, resinous materials made from different natural sources (tall oil rosins and rosin esters, for example), and many synthetic resins, oligomers, polymers,

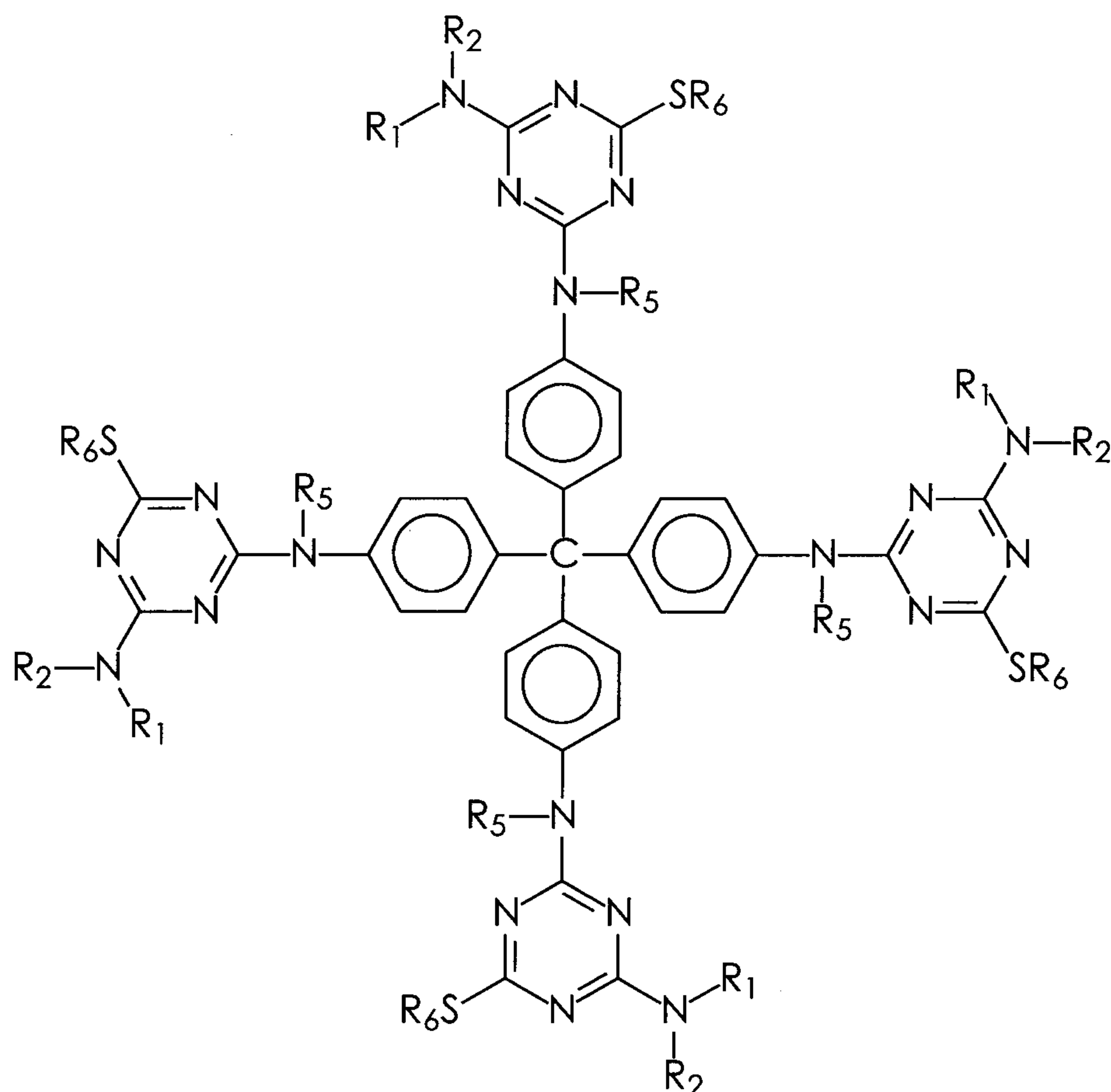
and copolymers.

[0007] U.S. Patent 6,761,758 (Boils-Boissier et al.), discloses compounds of the formulae





and



wherein, provided that at least one of R₁, R₂, R₃, R₄, R₅, and R₆ is a hydrogen atom, and provided that at least one of R₁, R₂, R₃, R₄, R₅, and R₆ is not a hydrogen atom, R₁, R₂, R₃, R₄, R₅, and R₆ each, independently of the others, is (i) a hydrogen atom, (ii) an alkyl group, (iii) an aryl group, (iv) an arylalkyl group, or (v) an alkylaryl group. Also disclosed are phase change ink compositions comprising a colorant and a phase change ink carrier comprising a material of this formula.

[0008] U.S. Patent 6,471,758 and European Patent Publication EP 1 067 157 (Kelderman et al.), disclose an ink composition for a meltable ink usable in a printing device in which ink drops are ejected from ink ducts, which comprises agents which reversibly cross-link the ink, the said agents containing a gelling agent. When an ink drop which has been transferred to a substrate passes over into a gel during

the cooling process, the consequence is that the viscosity of the melted ink drop increases greatly so that the drops become relatively immobile. In this way the ink drops are prevented from uncontrollably flowing into the paper. As a result, inks of this kind are suitable for use on both porous and smooth substrates. In addition, these inks have been found suitable for use in a printing device in which printed substrates are subjected to thermal after-treatment.

[0009] "Cyclic Bis-Urea Compounds as Gelators for Organic Solvents," J. van Esch et al., *Chem. Eur. J.* **1999**, 5, No. 3, pp. 937-950, discloses the study of the gelation properties of bis-urea compounds derived from optically pure trans-1,2-diaminocyclohexane and 1,2-diaminobenzene, with pendant aliphatic, aromatic, or ester groups, as well as the structure of the resulting gels.

[0010] "The Design of Organic Gelators Based on a Family of Bis-Ureas," R. E. Meléndez et al., *Mat. Res. Soc. Symp. Proc.* **2000**, 604, pp. 335-340, discloses a study of the organogelation properties of a family of bis-ureas.

[0011] "Formation of Organogels by Intermolecular Hydrogen Bonding Between Ureylene Segment," K. Hanabusa et al., *Chem. Lett.* **1996** pp. 885-886, discloses low molecular weight compounds having ureylene segment causing physical gelation in organic solvents. The main driving force for gelation was intermolecular hydrogen bonding between ureylene units.

[0012] "Low Molecular Weight Gelators for Organic Solvents," J. van Esch et al., in *Supramolecular Science: Where Is It and Where It Is Going*, R. Ungaro and E. Dalcanale, Eds., **1999**, Netherlands: Kluwer Academic Publishers, pp. 233-259, discloses the gelation of solvents by organogelators.

[0013] "Organogels and Low Molecular Mass Organic Gelators," D.

J. Abdallah and R. G. Weiss, *Adv. Mater.* **2000**, 12, No. 17, September 1, pp. 1237-1247, discloses the stepwise simplification of low molecular-mass organic gelator structures and the development of methods to determine their packing in organogels at the micrometer-to-angstrom distance regimes, as well as an overview of current and potential applications for these materials.

[0014] "Remarkable Stabilization of Self-Assembled Organogels by Polymerization," M. de Loos et al., *J. Am. Chem. Soc.* **1997**, 119, 12675-12676, discloses studies of polymerizable bis(amido)cyclohexane and bis(ureido)cyclohexane derivatives, investigating their gelating capacity for organic solvents.

[0015] "Low-molecular weight organogelators," P. Terech, in *Specialist Surfactants*, I.D. Robb, Ed., **1997**, London: Chapman & Hall, pp. 208-68, discloses a special class of surfactants which have the ability to form viscoelastic fluids or solid-like materials in organic solvents at concentrations lower than about 2 percent.

[0016] "New Functional Materials Based on Self-Assembling Organogels: From Serendipity Towards Design," J. H. van Esch and B. L. Feringa, *Angew. Chem. Int. Ed.* **2000**, 39, No. 13, pp. 2263-2266, discloses a review of developments in the field of organogels.

[0017] "Synthesis and Self-Assembling Properties of Polymerizable Organogelators," G. Wang and A. D. Hamilton, *Chem. Eur. J.* **2002**, 8, No. 8, pp. 1954-1961, discloses the development of a family of polymerizable urea derivatives that are gelators for organic solvents.

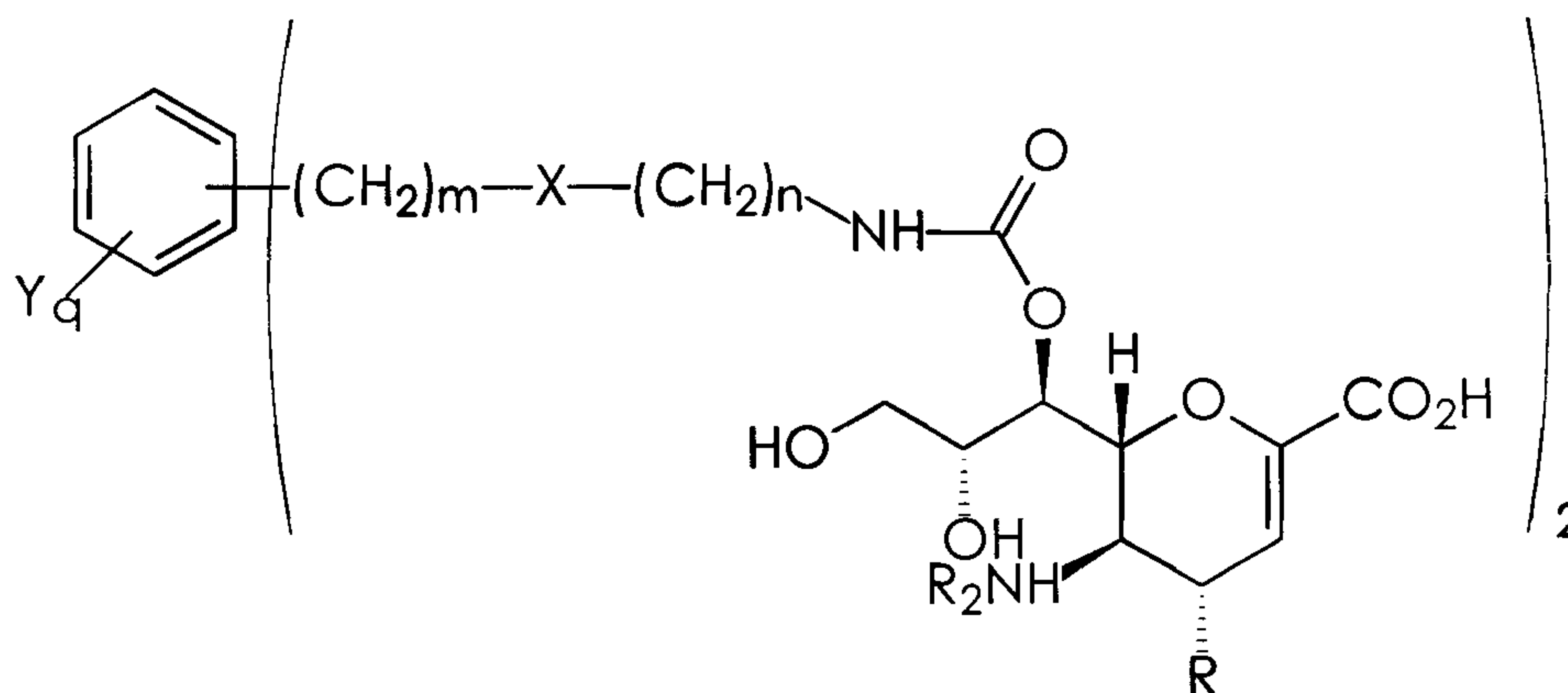
[0018] "Low Molecular Mass Gelators of Organic Liquids and the Properties of their Gels," P. Terech and R.G. Weiss, *Chem. Rev.* **1997**, 97, pp. 3133-3159, discloses a review of the properties of thermally-reversible viscoelastic liquidlike or solidlike organogels comprising an organic liquid and low concentrations of relatively low molecular mass

gelator molecules.

[0019] "Towards a Phenomenological Definition of the Term 'Gel'," K. Amdal et al., *Polymer Gels and Networks*, **1993**, 1, pp. 5-17, discusses existing definitions of the term "gel" and proposes specific uses of the term.

[0020] PCT Patent Publication WO 03/084508 and European Patent Publication EP 1 350 507 (Friesen et al.), disclose delivery vehicles for delivering a substance of interest to a predetermined site, said vehicle comprising said substance and a means for inducing availability of at least one compartment of said vehicle toward the exterior, thereby allowing access of said substance to the exterior of said vehicle at said predetermined site. The invention is further concerned with uses of said vehicle and methods for preparing it.

[0021] PCT Patent Publication WO 03/040135 (Dowle et al.), discloses compounds of the formula



in which R is an amino or guanidino group, R₂ is acetyl or trifluoroacetyl, X is CONH, SO₂NH, NHCO, or NHCONH, m is either 0 or 1, n is an integer from 2 to 6, q is an integer from 0 to 3, and Y is hydrogen or an aromatic substituent, or a pharmaceutically acceptable derivative thereof. Also disclosed are methods for their preparation, pharmaceutical formulations containing them, and their use in the prevention or

treatment of a viral infection.

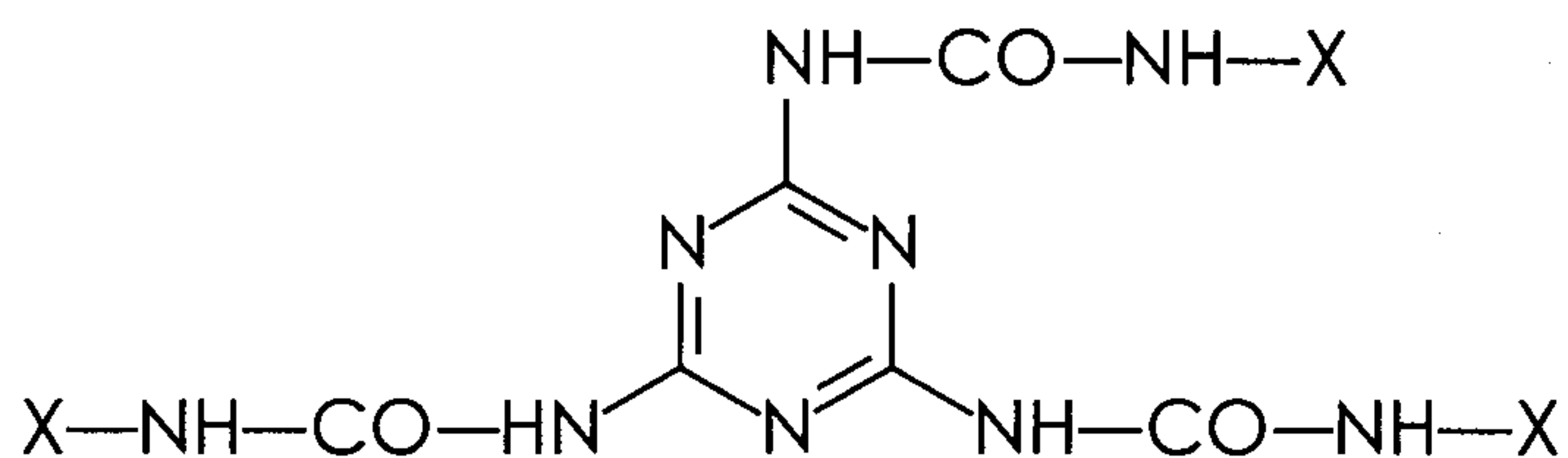
[0022] PCT Patent Publication WO 00/55149 and U.S. Patent 6,548,476 (Wu et al.), disclose dimeric compounds, methods for their preparation, pharmaceutical formulations thereof, and their use as antiviral agents. The compounds are particularly useful against influenza virus. In particular the references disclose a dimeric compound which comprises two neuraminidase binding groups attached to a spacer or linking group. Preferably the dimeric molecule comprises two neuraminidase-binding neuraminic acid (sialic acid) or cyclopentyl or cyclohexenyl carboxylic acid derivatives covalently attached to a common spacer group. Pharmaceutical compositions and methods of treatment, prophylaxis and diagnosis are disclosed and claimed.

[0023] U.S. Patent Publication 20010044553 (Kabashima et al.), discloses a urea-urethane compound having one or more urea groups and one or more urethane groups in the molecular structure, the number of said urea groups (A) and the number of said urethane groups (B) satisfying the following numerical formula: $10 \geq (A+B) \geq 3$ wherein each of A and B is an integer of 1 or more.

[0024] European Patent Publication EP 1 048 681 and U.S. Patent 6,420,466 (Haubennestel et al.), disclose a process for preparing a solution that is active as a thixotropic agent and contains urea urethanes, in which monohydroxyl compounds are reacted with an excess of toluene diisocyanate, the unreacted portion of the toluene diisocyanate is removed from the reaction mixture, and the monosiocyanate adduct obtained is further reacted with diarines in the presence of a lithium salt to form urea urethanes. The invention also relates to the use of the solution for imparting thixotropic properties to coating compounds.

[0025] Japanese Patent Publication JP 10310633, discloses a cationic curing catalyst composition improved in stability during storage at room temperature or above and suppressed in increase in viscosity, using at least one stabilizer selected from the compounds containing a urethane bond, an amide bond, a urea bond and a carbodiimide group in the molecule and a dialkylaminopyridine compound or a proton acid compound.

[0026] European Patent Publication EP 0 056 153 and U.S. Patent 4,384,102 (Rasshofer et al.), disclose compounds having both s-triazine units and epoxide groups present that are prepared by reacting an epoxide containing an isocyanate-reactive group with a triisocyanate corresponding to the formula



in which X is as defined therein. These reactants are used in quantities such that the equivalent ratio of isocyanate groups to isocyanate-reactive groups is maintained at less than or equal to 1 to 1. The compounds thus produced are particularly useful as reactive cross-linkers in the production of polyurethanes and polyepoxides.

[0027] European Patent Publication EP 0 160 402 and U.S. Patent 4,566,981 (Howells), disclose cationic and non-ionic fluorochemicals, mixtures of cationic and non-ionic fluorochemicals, blends of the mixtures with fluorochemical poly(oxyalkylenes), and compositions of the fluorochemicals with hydrocarbon nonionic surfactants. These fluorochemicals and compositions, in dispersions, emulsions and microemulsions, may be applied to porous fibrous substrates to give oil

and water repellancy and soil resistance.

[0028] Japanese Patent Publication JP 59030919, discloses a method to prevent the bad influence of a treatment on spinning properties and drawing properties of synthetic yarn, by providing undrawn yarn of melt spinning with a spinning oil, applying a specific treatment to it, drawing and heat-treating it. The undrawn yarn which is prepared by melt spinning and cooled is provided with a spinning oil by the oil applicator, coated with a treatment by the treatment applicator, sent through the taking up roller and the drawing rollers, and wound around the winder. The treatment is a compound shown by the formula $[R_f-A-B_1-CONH-X-NHCO-B_2-]_nY$ [R_f is 4-16C perfluoroalkyl; A is $-(CH_2)_{x1}-$, $CON(R_1)-(CH_2)_{x2}-$, or $SO_2N(R_1)-(CH_2)_{x2}-$; $x1$ is 1-20 integer; $x2$ is 1-12 integer; R_1 is H, or 1-6C alkyl; B_1 and B_2 are $-O-$, $-S-$, or $-N(R_2)-$; R_2 is H, or 1-4C alkyl; X is bifunctional organic group; Y is polyfunctional organic group; n is 2-10 integer] and its pickup is 0.03-2.0 wt%.

[0029] Compounds that enable gelation are also disclosed in, for example: "Reversible Polymers Formed from Self-Complementary Monomers Using Quadruple Hydrogen Bonding," R. P. Sijbesma et al., *Science*, Vol. 278, p. 1601 (1997); "Supramolecular Polymers," R. Dagani, *Chemical and Engineering News*, p. 4 (December 1997); "Supramolecular Polymers from Linear Telechelic Siloxanes with Quadruple-Hydrogen-Bonded Units," J.H.K. Hirschberg et al., *Macromolecules*, Vol. 32, p. 2696 (1999); "Design and Synthesis of 'Smart' Supramolecular Liquid Crystalline Polymers via Hydrogen-Bond Associations," A.C. Griffin et al., *PMSE Proceedings*, Vol. 72, p. 172 (1995); "The Design of Organic Gelators: Solution and Solid State Properties of a Family of Bis-Ureas," Andrew J. Carr et al., *Tetrahedron Letters*, Vol. 39, p. 7447 (1998); "Hydrogen-Bonded Supramolecular Polymer Networks," Ronald F.M. Lange et al., *Journal of Polymer*

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Organic Liquids)," D. Abdallah et al., *Langmuir*, 16, 352-355 (2000); "Low Molecular Mass Gelators of Organic Liquids and the Properties of their Gels," P. Terech et al., *Chem. Rev.*, 97, 3133-3159 (1997); "Organogels and Low Molecular Mass Organic Gelators," D. Abdallah et al., *Adv. Mater.*, 12, No. 17, 1237 (2000); "Making it All Stick Together: the Gelation of Organic Liquids by Small Organic Molecules," F. Schoonbeek, Doctoral Thesis, U. of Groningen, Netherlands, April 2001; Twieg et al., *Macromolecules*, Vol. 18, p. 1361 (1985); "Synthesis and Reactions of Polyhydric Alcohols I. Synthesis and Reactions of p-Toluenesulfonates of Polyhydric Alcohols," *Zhurnal Obshchei Khimii*, Vol. 35, No. 5, p. 804-807 (1965); "The Chemotherapy of Schistosomiasis. Part I. Derivatives and Analogs of $\alpha\omega$ -Di-(p-aminophenoxy)alkanes," J. Ashley et al., *J. Chem. Soc.* 1958, 3293; "Remarkably Simple Small Organogelators: Di-n-alkoxy-benzene Derivatives," G. Clavier et al., *Tetrahedron Letters*, 40, 9021-9024 (1999); "Rational Design of Low Molecular Mass Organogelators: Toward a Library of Functional N-Acyl-1- ω -Amino Acid Derivatives," G. Mieden-Gundert et al., *Angew. Chem. Int. Ed.*, 40, No. 17, 3164-3166 (2001); U.S. Patent 2,703,808; "Rational Design of New Acid-Sensitive Organogelators," J-L. Pozzo et al., *J. Mater. Chem.*, Vol. 8, pp. 2575-2577 (1998); J. T. Thurston et al., *J. Am. Chem. Soc.*, Vol. 73, pp. 2981-3008 (1951); *J. Am. Chem. Soc.*, Vol. 96, pp. 1082-1087 (1974); J-L. Pozzo et al., *Tetrahedron*, Vol. 53, No. 18, pp. 6377-6390 (1997); J-L. Pozzo et al., *Mol. Cryst. Liq. Cryst.*, Vol. 344, pp. 101-106 (2000); Y.C. Lin, R.G. Weiss, *Macromolecules*, Vol. 20, p. 414 (1987); U.S. Patent 4,790,961; Murata et al, *J. Am. Chem. Soc.*, Vol. 116, No 15, pp. 6664-6676 (1994); A. Ikeda et al., *Rep. Asahi Glass Found. Ind. Technol.*, Vol. 61, p. 115, (1992); Rabolt et al., *Macromolecules*, Vol. 17, p. 2786 (1984); D.J. Abdallah et al., *Chem. Mater.*, Vol. 11, p. 2907 (1999); Ralston et al., *J. Org. Chem.*, Vol. 9, p. 259 (1944); L. Lu et al.,

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[0030] The *trans*-1,2-cyclohexane bis-urea organogelator compounds exhibit some disadvantages for performing in a phase-change solid ink vehicle, such as high melting point and high degree of crystallinity. In addition, these compounds are commonly prepared by the reaction of *trans*-1,2-diaminocyclohexane with two molar equivalents of a monofunctional isocyanate, and their large-scale commercial preparation is often limited to the use of available monofunctional isocyanate raw materials that are regulated for health and safety reasons.

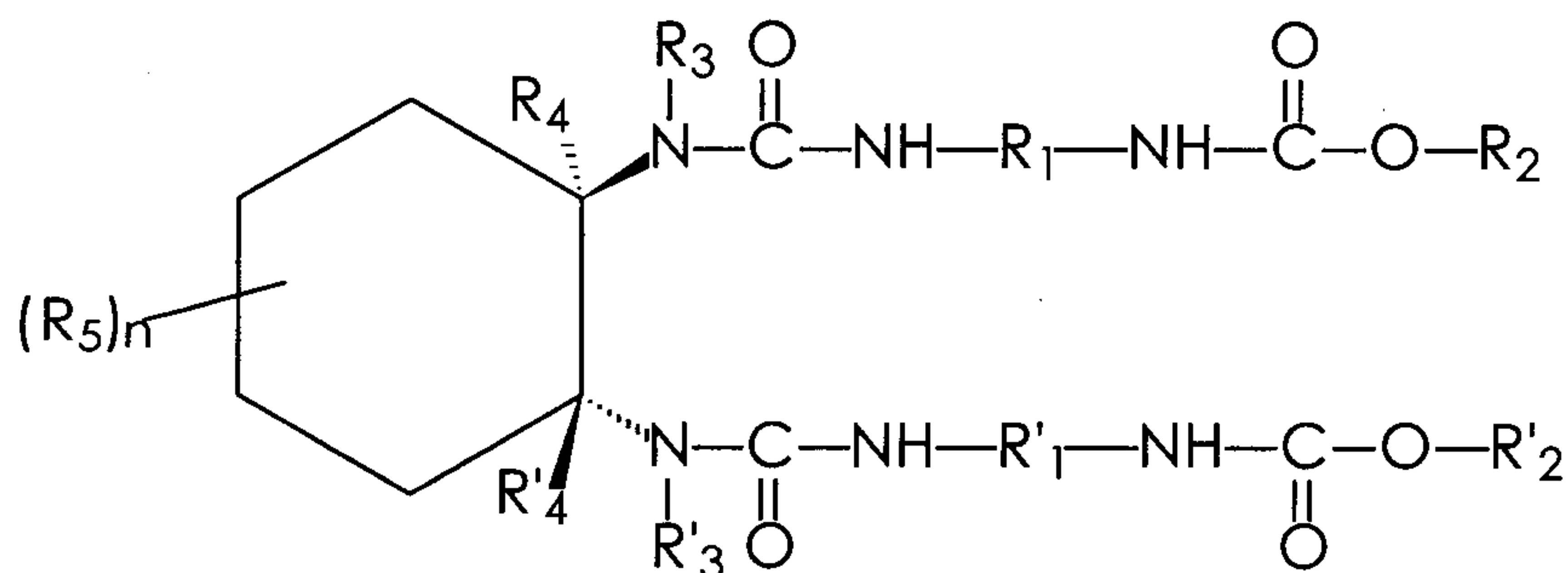
[0031] Many currently used phase change inks require high jetting temperatures of about 140°C or greater and also require relatively long warm-up times for the printer. In addition, many currently used phase change inks generate images with relatively poor scratch resistance and relatively poor image permanence.

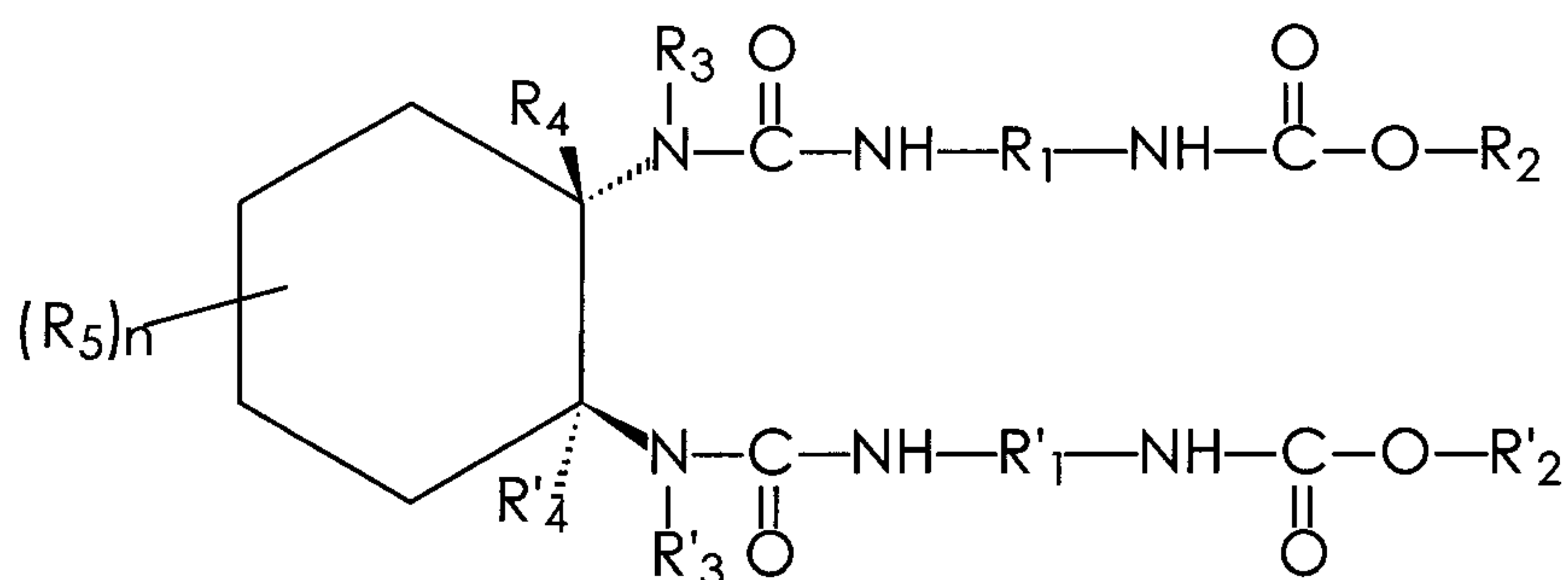
[0032] While known compositions and processes are suitable for their intended purposes, a need remains for improved phase change ink compositions. In addition, a need remains for phase change inks that can be jetted at reduced temperatures of about 110°C or lower, thereby enabling cost and energy savings. Further, a need remains for phase change inks that enable printing with reduced printer warm-up times. Additionally, a need remains for phase change inks that generate images with improved scratch resistance. There is also a need for phase change inks that generate images with improved image permanence. In addition, there is a need for phase change inks that generate images with improved image quality. Further, there is a need for phase change inks that exhibit the aforementioned advantages when used in a printing process wherein the ink is first jetted onto an intermediate transfer member and subsequently transferred from the intermediate transfer member to a final print substrate such as plain or coated paper or a transparency. Additionally, there is a need for phase change inks that exhibit the aforementioned advantages when used in a printing process wherein the ink is jetted directly onto a final print substrate such as plain or coated paper or a transparency. A need also remains for phase change inks that exhibit the aforementioned advantages when used in printing processes at relatively high speeds. In addition, a need remains for phase change inks having desirably low melting points that also contain gelator compounds which enable additional advantages in the phase change inks. Further, a need remains for gelator compounds for use in phase change inks and other applications that have a desirably low degree of crystallinity. Additionally, a need remains for gelator compounds that are soluble in phase change ink carriers. There is also a need for phase change inks that exhibit an intermediate gel

phase between the solid phase and the liquid phase. In addition, there is a need for phase change inks exhibiting an intermediate gel phase wherein the gel phase transition is desirably narrow. Further, there is a need for gelator compounds that enable desirably narrow gel phase transitions. Additionally, there is a need for phase change inks exhibiting an intermediate gel phase wherein the gel phase transition entails a tan-delta of less than about 10. A need also remains for gelator compounds that enable gel phase transitions entailing a tan-delta of less than about 10. In addition, a need remains for gelator compounds that are less highly crystalline and do not pack as tightly within a molecular network as do more crystalline materials, thereby enabling them to be soluble within molten phase change inks.

SUMMARY

[0033] Disclosed herein are phase change ink compositions comprising a phase change ink carrier and a *trans*-1,2-cyclohexane bis[urea-urethane] compound of the formula

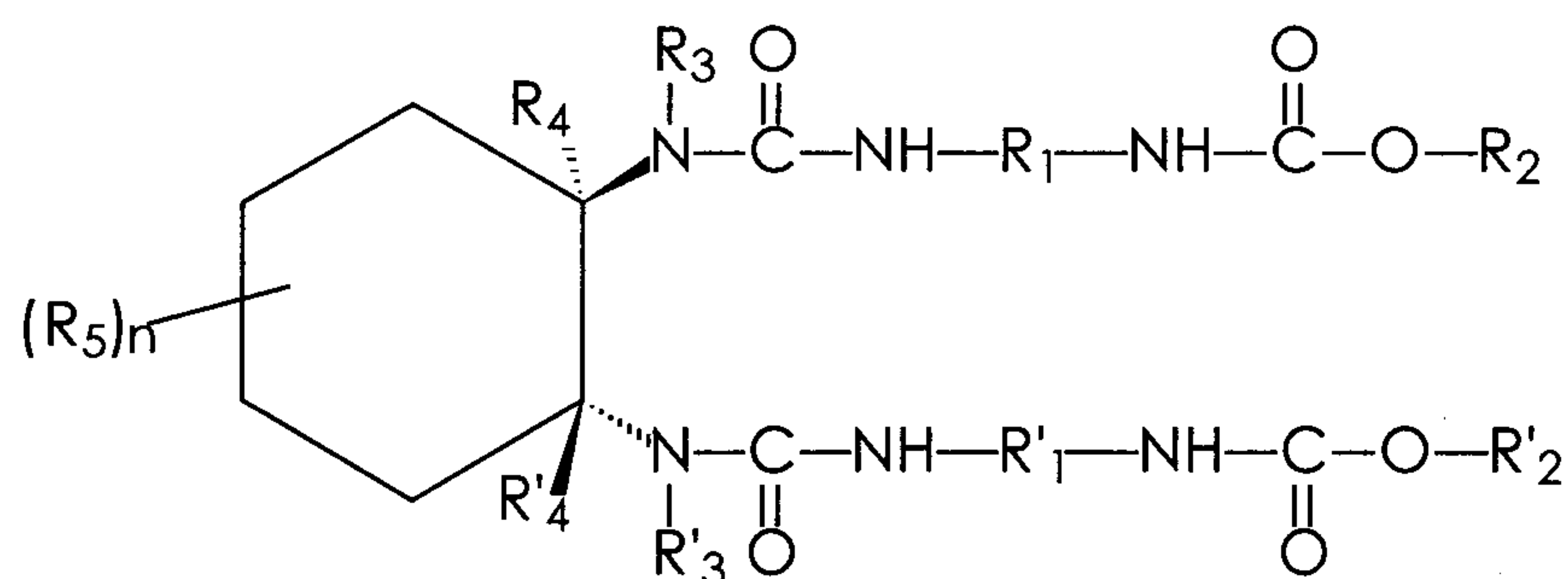




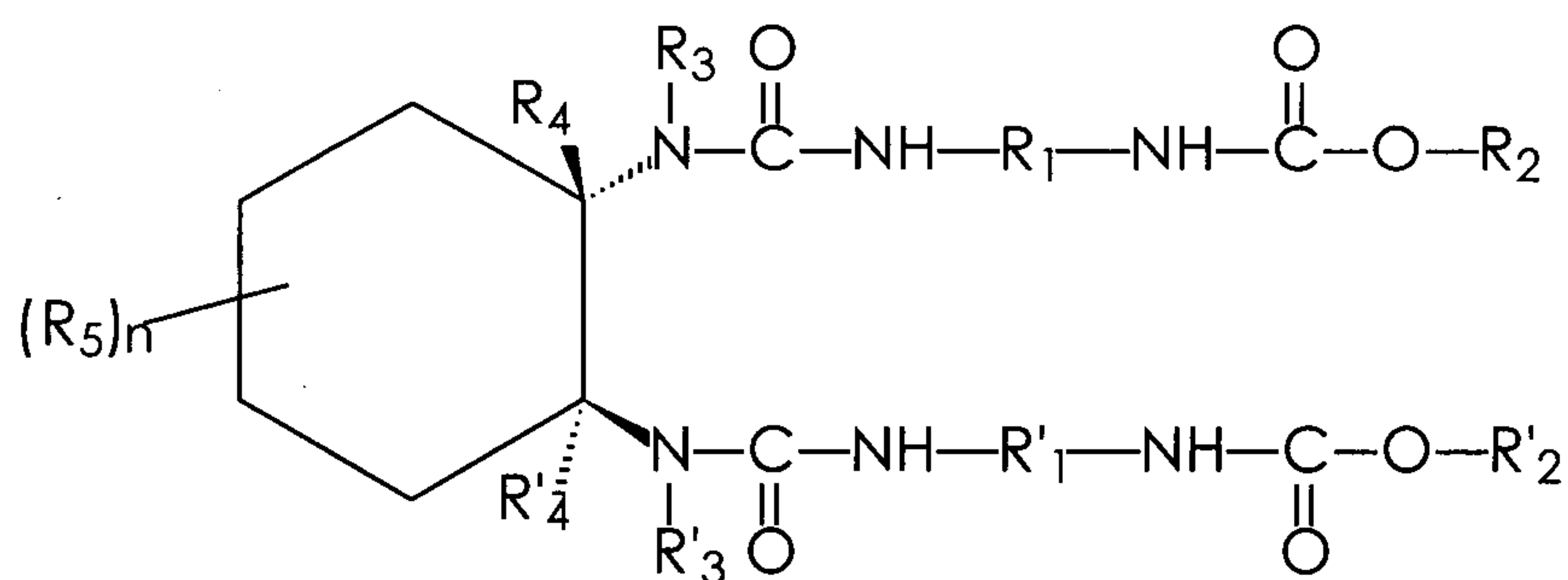
or mixtures thereof, wherein R_1 and R'_1 each, independently of the other, is an alkylene group, an arylene group, an arylalkylene group, or an alkylarylene group, R_2 and R'_2 each, independently of the other, is an alkyl group, an aryl group, an arylalkyl group, or an alkylaryl group, R_3 and R'_3 each, independently of the other, is a hydrogen atom or an alkyl group, R_4 and R'_4 each, independently of the other, is a hydrogen atom, a fluorine atom, an alkyl group, or a phenyl group, n is an integer of 0, 1, 2, 3, or 4, and R_5 is an alkyl group, an aryl group, an arylalkyl group, an alkylaryl group, or a substituent other than an alkyl, aryl, arylalkyl, or alkylaryl group.

DETAILED DESCRIPTION

[0034] The *trans*-1,2-cyclohexane bis[urea-urethane] compounds are of the formulae



and

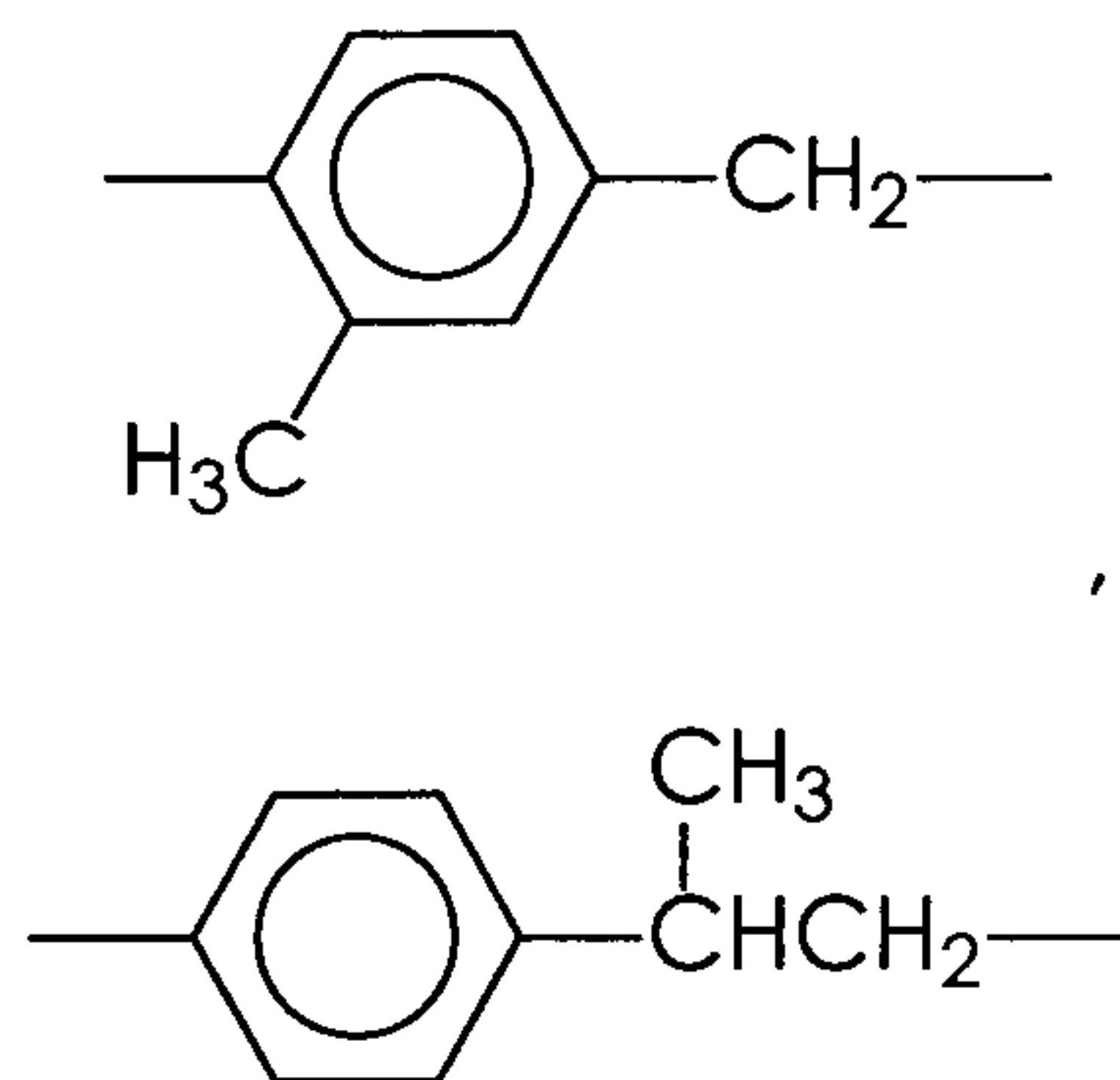


wherein R₁ and R'₁ each, independently of the other, is (i) an alkylene group (including linear, branched, cyclic, substituted, and unsubstituted alkylene groups, and wherein hetero atoms, such as oxygen, nitrogen, sulfur, silicon, phosphorus, boron, and the like either may or may not be present in the alkylene group), in one embodiment with at least about 2 carbon atoms, in another embodiment with at least about 4 carbon atoms, and in yet another embodiment with at least about 6 carbon atoms, and in one embodiment with no more than about 100 carbon atoms, in another embodiment with no more than about 60 carbon atoms, and in yet another embodiment with no more than about 20 carbon atoms, although the number of carbon atoms can be outside of these ranges, including (but not limited to) (1) linear saturated unsubstituted aliphatic groups containing no hetero atoms, (2) branched saturated unsubstituted aliphatic groups containing no hetero atoms, (3) cyclic saturated unsubstituted aliphatic groups containing no hetero atoms, (4) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being saturated, unsubstituted, and containing no hetero atoms, (5) linear ethylenically unsaturated unsubstituted aliphatic groups containing no hetero atoms, (6) branched ethylenically unsaturated unsubstituted aliphatic groups containing no hetero atoms, (7) cyclic ethylenically unsaturated unsubstituted aliphatic groups containing no hetero atoms, (8) aliphatic

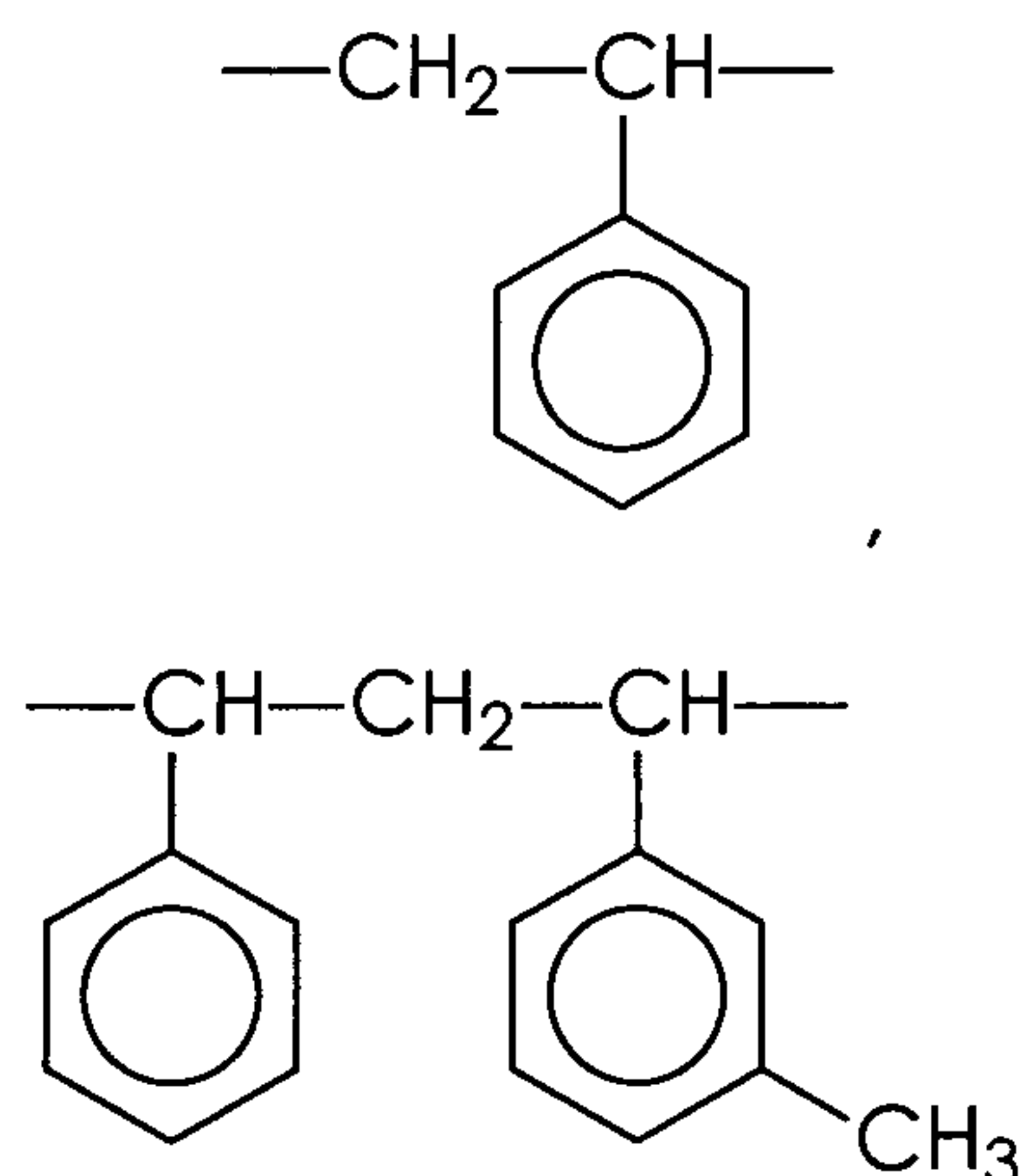
groups containing both cyclic and acyclic portions, said aliphatic groups being ethylenically unsaturated, unsubstituted, and containing no hetero atoms, (9) linear saturated substituted aliphatic groups containing no hetero atoms, (10) branched saturated substituted aliphatic groups containing no hetero atoms, (11) cyclic saturated substituted aliphatic groups containing no hetero atoms, (12) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being saturated, substituted, and containing no hetero atoms, (13) linear ethylenically unsaturated substituted aliphatic groups containing no hetero atoms, (14) branched ethylenically unsaturated substituted aliphatic groups containing no hetero atoms, (15) cyclic ethylenically unsaturated substituted aliphatic groups containing no hetero atoms, (16) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being ethylenically unsaturated, substituted, and contain no hetero atoms, (17) linear saturated unsubstituted aliphatic groups containing hetero atoms, (18) branched saturated unsubstituted aliphatic groups containing hetero atoms, (19) cyclic saturated unsubstituted aliphatic groups containing hetero atoms, (20) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being saturated, unsubstituted, and containing hetero atoms, (21) linear ethylenically unsaturated unsubstituted aliphatic groups containing hetero atoms, (22) branched ethylenically unsaturated unsubstituted aliphatic groups containing hetero atoms, (23) cyclic ethylenically unsaturated unsubstituted aliphatic groups containing hetero atoms, (24) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being ethylenically unsaturated, unsubstituted, and containing hetero atoms, (25) linear saturated substituted aliphatic groups containing hetero atoms, (26) branched saturated substituted aliphatic groups

containing hetero atoms, (27) cyclic saturated substituted aliphatic groups containing hetero atoms, (28) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being saturated, substituted, and containing hetero atoms, (29) linear ethylenically unsaturated substituted aliphatic groups containing hetero atoms, (30) branched ethylenically unsaturated substituted aliphatic groups containing hetero atoms, (31) cyclic ethylenically unsaturated substituted aliphatic groups containing hetero atoms, and (32) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being ethylenically unsaturated, substituted, and containing hetero atoms, (ii) an arylene group (including substituted and unsubstituted arylene groups, and wherein hetero atoms, such as oxygen, nitrogen, sulfur, silicon, phosphorus, boron, and the like either may or may not be present in the arylene group), in one embodiment with at least about 5 carbon atoms, and in another embodiment with at least about 6 carbon atoms, and in one embodiment with no more than about 18 carbon atoms, in another embodiment with no more than about 12 carbon atoms, and in yet another embodiment with no more than about 6 carbon atoms, although the number of carbon atoms can be outside of these ranges, (iii) an arylalkylene group (including substituted and unsubstituted arylalkylene groups, and wherein hetero atoms, such as oxygen, nitrogen, sulfur, silicon, phosphorus, boron, and the like either may or may not be present in either the aryl or the alkyl portion of the arylalkylene group), in one embodiment with at least about 6 carbon atoms, and in another embodiment with at least about 7 carbon atoms, and in one embodiment with no more than about 100 carbon atoms, in another embodiment with no more than about 60 carbon atoms, and in yet another embodiment with no more than about 20 carbon atoms,

although the number of carbon atoms can be outside of these ranges, such as benzylene or the like, including (a) arylalkylene groups wherein both the aryl and the alkyl portions form the linkage between the two -NH- groups, such as

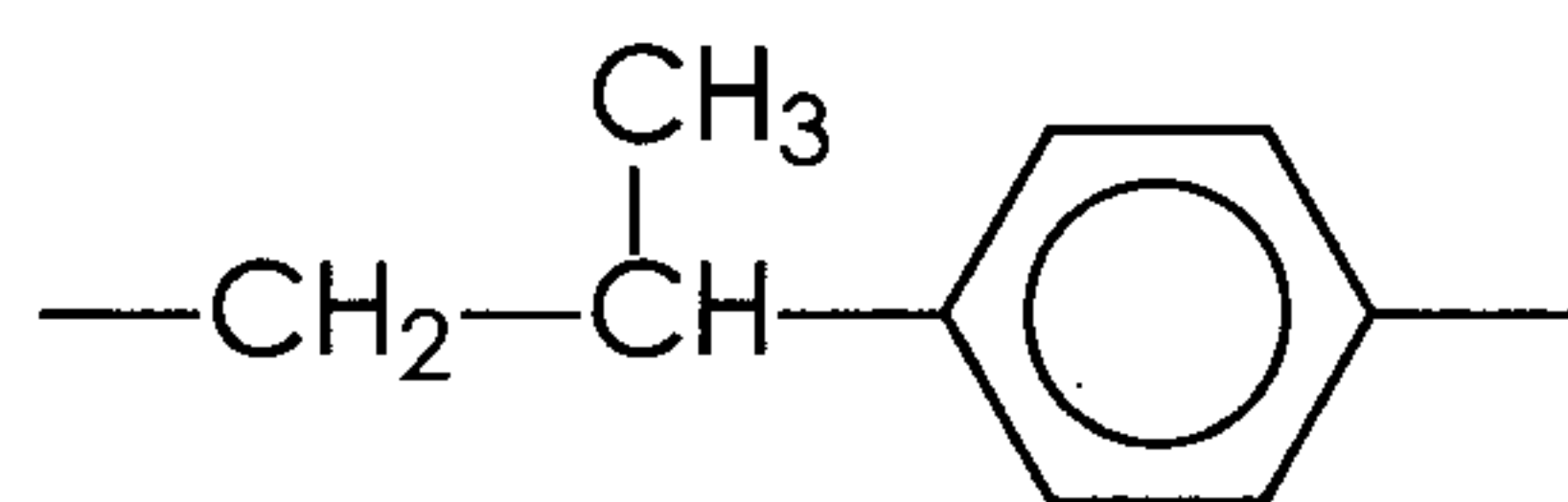
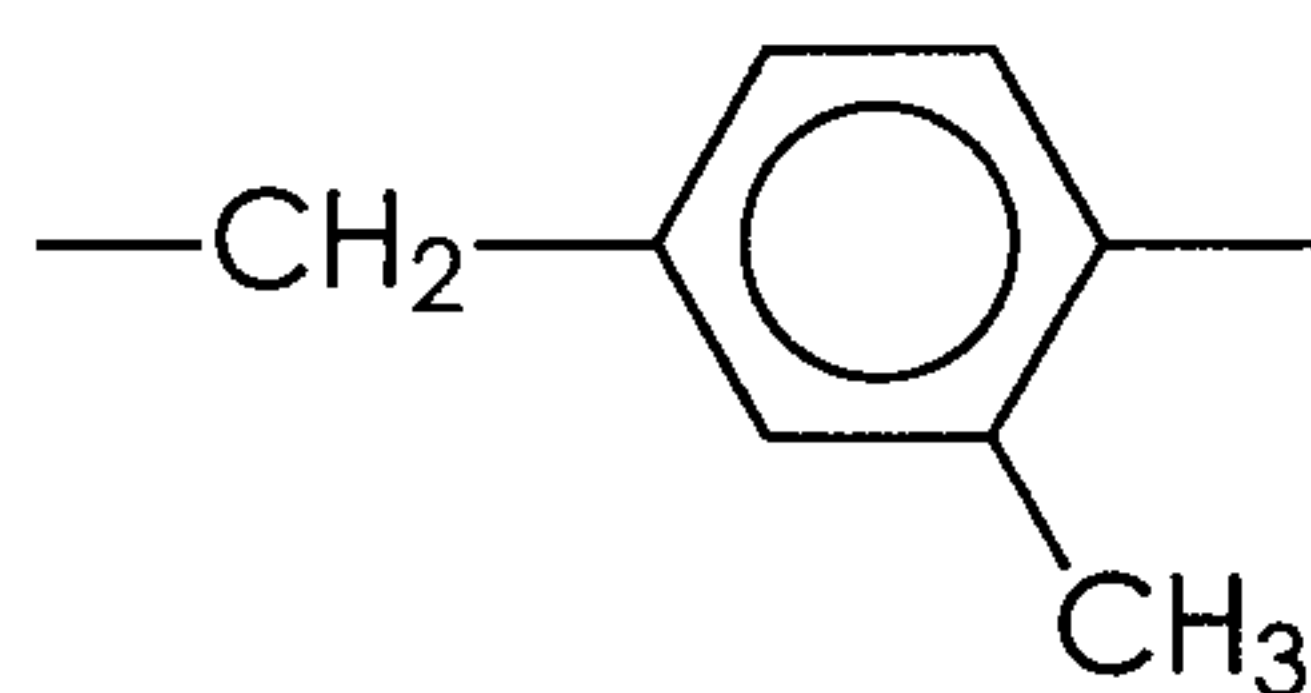


and the like, and (b) arylalkylene groups wherein only the alkyl portion forms the linkage between the two -NH- groups, such as

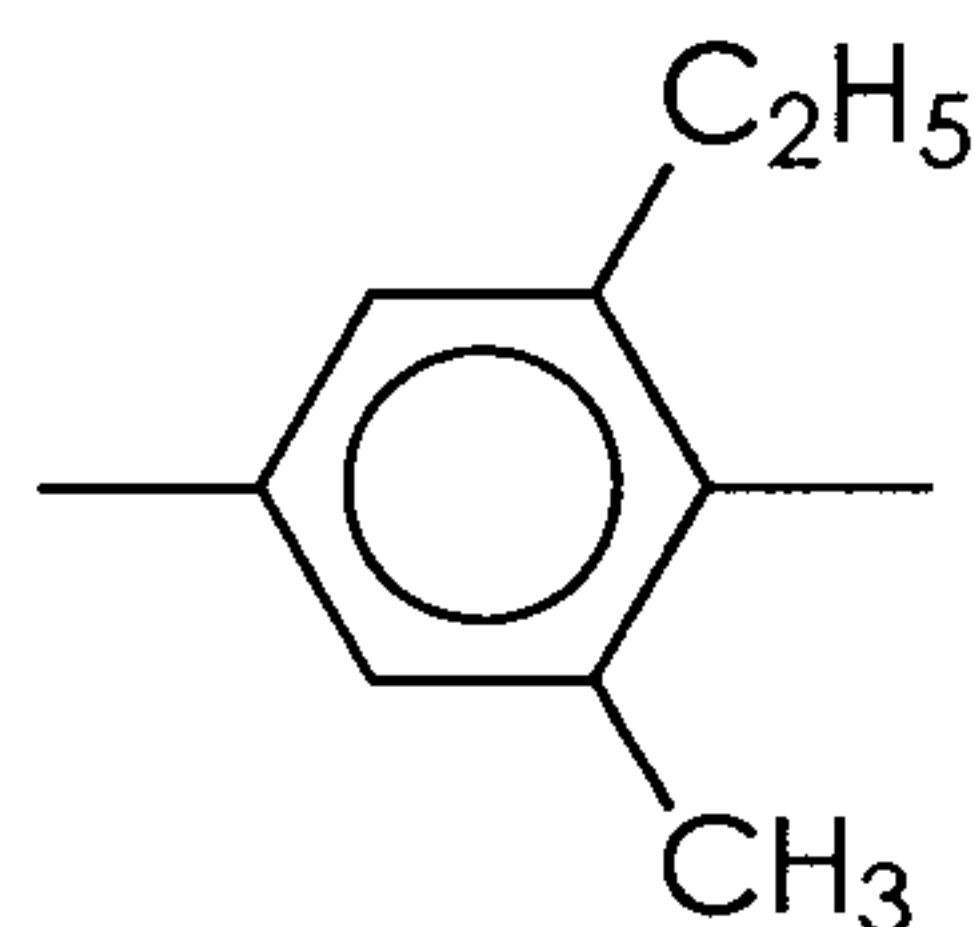
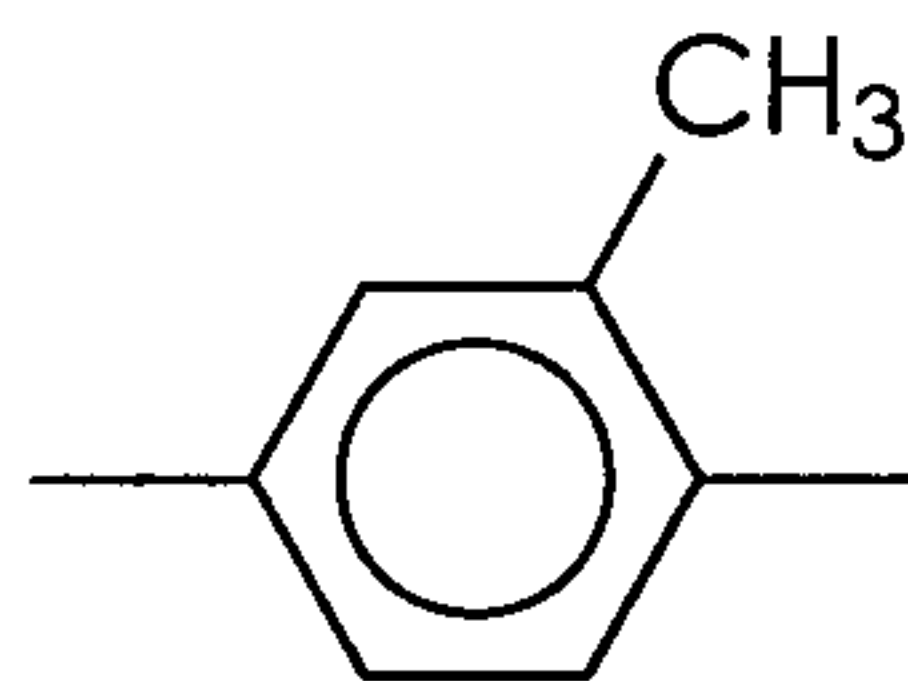


and the like, or (iv) an alkylarylene group (including substituted and unsubstituted alkylarylene groups, and wherein hetero atoms, such as oxygen, nitrogen, sulfur, silicon, phosphorus, boron, and the like either may or may not be present in either the aryl or the alkyl portion of the alkylarylene group), in one embodiment with at least about 6 carbon atoms, and in another embodiment with at least about 7 carbon atoms, and in one embodiment with no more than about 100 carbon atoms, in another embodiment with no more than about 60 carbon

atoms, and in yet another embodiment with no more than about 20 carbon atoms, although the number of carbon atoms can be outside of these ranges, such as tolylene or the like, including (a) alkylarylene groups wherein both the alkyl and the aryl portions form the linkage between the two -NH- groups, such as



and the like, and (b) alkylarylene groups wherein only the aryl portion forms the linkage between the two -NH- groups, such as



and the like, R_2 and R'_2 each, independently of the other, is (i) an alkyl group (including linear, branched, cyclic, substituted, and unsubstituted alkyl groups, and wherein hetero atoms, such as oxygen, nitrogen, sulfur, silicon, phosphorus, boron, and the like either may or may not be present in the alkyl group), in one embodiment with at least 1 carbon atom, in another embodiment with at least about 4 carbon atoms, and in yet another embodiment with at least about 10 carbon

atoms, and in one embodiment with no more than about 100 carbon atoms, in another embodiment with no more than about 60 carbon atoms, and in yet another embodiment with no more than about 20 carbon atoms, although the number of carbon atoms can be outside of these ranges, including (but not limited to) (1) linear saturated unsubstituted aliphatic groups containing no hetero atoms, (2) branched saturated unsubstituted aliphatic groups containing no hetero atoms, (3) cyclic saturated unsubstituted aliphatic groups containing no hetero atoms, (4) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being saturated, unsubstituted, and containing no hetero atoms, (5) linear ethylenically unsaturated unsubstituted aliphatic groups containing no hetero atoms, (6) branched ethylenically unsaturated unsubstituted aliphatic groups containing no hetero atoms, (7) cyclic ethylenically unsaturated unsubstituted aliphatic groups containing no hetero atoms, (8) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being ethylenically unsaturated, unsubstituted, and containing no hetero atoms, (9) linear saturated substituted aliphatic groups containing no hetero atoms, (10) branched saturated substituted aliphatic groups containing no hetero atoms, (11) cyclic saturated substituted aliphatic groups containing no hetero atoms, (12) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being saturated, substituted, and containing no hetero atoms, (13) linear ethylenically unsaturated substituted aliphatic groups containing no hetero atoms, (14) branched ethylenically unsaturated substituted aliphatic groups containing no hetero atoms, (15) cyclic ethylenically unsaturated substituted aliphatic groups containing no hetero atoms, (16) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being ethylenically unsaturated,

substituted, and contain no hetero atoms, (17) linear saturated unsubstituted aliphatic groups containing hetero atoms, (18) branched saturated unsubstituted aliphatic groups containing hetero atoms, (19) cyclic saturated unsubstituted aliphatic groups containing hetero atoms, (20) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being saturated, unsubstituted, and containing hetero atoms, (21) linear ethylenically unsaturated unsubstituted aliphatic groups containing hetero atoms, (22) branched ethylenically unsaturated unsubstituted aliphatic groups containing hetero atoms, (23) cyclic ethylenically unsaturated unsubstituted aliphatic groups containing hetero atoms, (24) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being ethylenically unsaturated, unsubstituted, and containing hetero atoms, (25) linear saturated substituted aliphatic groups containing hetero atoms, (26) branched saturated substituted aliphatic groups containing hetero atoms, (27) cyclic saturated substituted aliphatic groups containing hetero atoms, (28) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being saturated, substituted, and containing hetero atoms, (29) linear ethylenically unsaturated substituted aliphatic groups containing hetero atoms, (30) branched ethylenically unsaturated substituted aliphatic groups containing hetero atoms, (31) cyclic ethylenically unsaturated substituted aliphatic groups containing hetero atoms, and (32) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being ethylenically unsaturated, substituted, and containing hetero atoms, (ii) an aryl group (including substituted and unsubstituted aryl groups, and wherein hetero atoms, such as oxygen, nitrogen, sulfur, silicon, phosphorus, boron, and the like either may or may not be present in the aryl group), in one embodiment with at least about 5

carbon atoms, and in another embodiment with at least about 6 carbon atoms, and in one embodiment with no more than about 18 carbon atoms, in another embodiment with no more than about 12 carbon atoms, and in yet another embodiment with no more than about 6 carbon atoms, although the number of carbon atoms can be outside of these ranges, (iii) an arylalkyl group (including substituted and unsubstituted arylalkyl groups, and wherein hetero atoms, such as oxygen, nitrogen, sulfur, silicon, phosphorus, boron, and the like either may or may not be present in either the aryl or the alkyl portion of the arylalkyl group), in one embodiment with at least about 6 carbon atoms, and in another embodiment with at least about 7 carbon atoms, and in one embodiment with no more than about 100 carbon atoms, in another embodiment with no more than about 60 carbon atoms, and in yet another embodiment with no more than about 20 carbon atoms, although the number of carbon atoms can be outside of these ranges, such as benzyl or the like, or (iv) an alkylaryl group (including substituted and unsubstituted alkylaryl groups, and wherein hetero atoms, such as oxygen, nitrogen, sulfur, silicon, phosphorus, boron, and the like either may or may not be present in either the aryl or the alkyl portion of the alkylaryl group), in one embodiment with at least about 6 carbon atoms, and in another embodiment with at least about 7 carbon atoms, and in one embodiment with no more than about 100 carbon atoms, in another embodiment with no more than about 60 carbon atoms, and in yet another embodiment with no more than about 20 carbon atoms, although the number of carbon atoms can be outside of these ranges, such as tolyl or the like, R_3 and R'_3 each, independently of the other, is a hydrogen atom or an alkyl group (including linear, branched, substituted, and unsubstituted alkyl groups), in one embodiment with at least 1 carbon atom, and in one

embodiment with no more than about 3 carbon atoms, although the number of carbon atoms can be outside of these ranges, R_4 and R'_4 each, independently of the other, is a hydrogen atom, a fluorine atom, an alkyl group (including linear, branched, substituted, and unsubstituted alkyl groups, and wherein hetero atoms, such as oxygen, nitrogen, sulfur, silicon, phosphorus, boron, and the like either may or may not be present in the alkyl group), in one embodiment with at least 1 carbon atom, and in one embodiment with no more than about 6 carbon atoms, in another embodiment with no more than about 3 carbon atoms, and in yet another embodiment with no more than about 2 carbon atoms, although the number of carbon atoms can be outside of these ranges, including (but not limited to) (1) linear saturated unsubstituted aliphatic groups containing no hetero atoms, (2) branched saturated unsubstituted aliphatic groups containing no hetero atoms, (3) cyclic saturated unsubstituted aliphatic groups containing no hetero atoms, (4) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being saturated, unsubstituted, and containing no hetero atoms, (5) linear ethylenically unsaturated unsubstituted aliphatic groups containing no hetero atoms, (6) branched ethylenically unsaturated unsubstituted aliphatic groups containing no hetero atoms, (7) cyclic ethylenically unsaturated unsubstituted aliphatic groups containing no hetero atoms, (8) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being ethylenically unsaturated, unsubstituted, and containing no hetero atoms, (9) linear saturated substituted aliphatic groups containing no hetero atoms, (10) branched saturated substituted aliphatic groups containing no hetero atoms, (11) cyclic saturated substituted aliphatic groups containing no hetero atoms, (12) aliphatic groups containing both cyclic and acyclic portions, said aliphatic

groups being saturated, substituted, and containing no hetero atoms, (13) linear ethylenically unsaturated substituted aliphatic groups containing no hetero atoms, (14) branched ethylenically unsaturated substituted aliphatic groups containing no hetero atoms, (15) cyclic ethylenically unsaturated substituted aliphatic groups containing no hetero atoms, (16) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being ethylenically unsaturated, substituted, and contain no hetero atoms, (17) linear saturated unsubstituted aliphatic groups containing hetero atoms, (18) branched saturated unsubstituted aliphatic groups containing hetero atoms, (19) cyclic saturated unsubstituted aliphatic groups containing hetero atoms, (20) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being saturated, unsubstituted, and containing hetero atoms, (21) linear ethylenically unsaturated unsubstituted aliphatic groups containing hetero atoms, (22) branched ethylenically unsaturated unsubstituted aliphatic groups containing hetero atoms, (23) cyclic ethylenically unsaturated unsubstituted aliphatic groups containing hetero atoms, (24) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being ethylenically unsaturated, unsubstituted, and containing hetero atoms, (25) linear saturated substituted aliphatic groups containing hetero atoms, (26) branched saturated substituted aliphatic groups containing hetero atoms, (27) cyclic saturated substituted aliphatic groups containing hetero atoms, (28) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being saturated, substituted, and containing hetero atoms, (29) linear ethylenically unsaturated substituted aliphatic groups containing hetero atoms, (30) branched ethylenically unsaturated substituted aliphatic groups containing hetero atoms, (31) cyclic ethylenically unsaturated

substituted aliphatic groups containing hetero atoms, and (32) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being ethylenically unsaturated, substituted, and containing hetero atoms, or a phenyl group, n is an integer of 0, 1, 2, 3, or 4, and each R_5 , independently of the others, is (i) an alkyl group (including linear, branched, cyclic, substituted, and unsubstituted alkyl groups, and wherein hetero atoms, such as oxygen, nitrogen, sulfur, silicon, phosphorus, boron, and the like either may or may not be present in the alkyl group), in one embodiment with at least 1 carbon atom, and in one embodiment with no more than about 100 carbon atoms, in another embodiment with no more than about 60 carbon atoms, and in yet another embodiment with no more than about 20 carbon atoms, although the number of carbon atoms can be outside of these ranges, including (but not limited to) (1) linear saturated unsubstituted aliphatic groups containing no hetero atoms, (2) branched saturated unsubstituted aliphatic groups containing no hetero atoms, (3) cyclic saturated unsubstituted aliphatic groups containing no hetero atoms, (4) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being saturated, unsubstituted, and containing no hetero atoms, (5) linear ethylenically unsaturated unsubstituted aliphatic groups containing no hetero atoms, (6) branched ethylenically unsaturated unsubstituted aliphatic groups containing no hetero atoms, (7) cyclic ethylenically unsaturated unsubstituted aliphatic groups containing no hetero atoms, (8) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being ethylenically unsaturated, unsubstituted, and containing no hetero atoms, (9) linear saturated substituted aliphatic groups containing no hetero atoms, (10) branched saturated substituted aliphatic groups containing no hetero atoms, (11) cyclic saturated

substituted aliphatic groups containing no hetero atoms, (12) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being saturated, substituted, and containing no hetero atoms, (13) linear ethylenically unsaturated substituted aliphatic groups containing no hetero atoms, (14) branched ethylenically unsaturated substituted aliphatic groups containing no hetero atoms, (15) cyclic ethylenically unsaturated substituted aliphatic groups containing no hetero atoms, (16) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being ethylenically unsaturated, substituted, and contain no hetero atoms, (17) linear saturated unsubstituted aliphatic groups containing hetero atoms, (18) branched saturated unsubstituted aliphatic groups containing hetero atoms, (19) cyclic saturated unsubstituted aliphatic groups containing hetero atoms, (20) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being saturated, unsubstituted, and containing hetero atoms, (21) linear ethylenically unsaturated unsubstituted aliphatic groups containing hetero atoms, (22) branched ethylenically unsaturated unsubstituted aliphatic groups containing hetero atoms, (23) cyclic ethylenically unsaturated unsubstituted aliphatic groups containing hetero atoms, (24) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being ethylenically unsaturated, unsubstituted, and containing hetero atoms, (25) linear saturated substituted aliphatic groups containing hetero atoms, (26) branched saturated substituted aliphatic groups containing hetero atoms, (27) cyclic saturated substituted aliphatic groups containing hetero atoms, (28) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being saturated, substituted, and containing hetero atoms, (29) linear ethylenically unsaturated substituted aliphatic groups containing hetero atoms, (30)

branched ethylenically unsaturated substituted aliphatic groups containing hetero atoms, (31) cyclic ethylenically unsaturated substituted aliphatic groups containing hetero atoms, and (32) aliphatic groups containing both cyclic and acyclic portions, said aliphatic groups being ethylenically unsaturated, substituted, and containing hetero atoms, (ii) an aryl group (including substituted and unsubstituted aryl groups, and wherein hetero atoms, such as oxygen, nitrogen, sulfur, silicon, phosphorus, boron, and the like either may or may not be present in the aryl group), in one embodiment with at least about 5 carbon atoms, and in another embodiment with at least about 6 carbon atoms, and in one embodiment with no more than about 18 carbon atoms, in another embodiment with no more than about 12 carbon atoms, and in yet another embodiment with no more than about 6 carbon atoms, although the number of carbon atoms can be outside of these ranges, (iii) an arylalkyl group (including substituted and unsubstituted arylalkyl groups, and wherein hetero atoms, such as oxygen, nitrogen, sulfur, silicon, phosphorus, boron, and the like either may or may not be present in either the aryl or the alkyl portion of the arylalkyl group), in one embodiment with at least about 6 carbon atoms, and in another embodiment with at least about 7 carbon atoms, and in one embodiment with no more than about 100 carbon atoms, in another embodiment with no more than about 60 carbon atoms, and in yet another embodiment with no more than about 20 carbon atoms, although the number of carbon atoms can be outside of these ranges, such as benzyl or the like, (iv) an alkylaryl group (including substituted and unsubstituted alkylaryl groups, and wherein hetero atoms, such as oxygen, nitrogen, sulfur, silicon, phosphorus, boron, and the like either may or may not be present in either the aryl or the alkyl portion of the alkylaryl group), in one embodiment with at

least about 6 carbon atoms, and in another embodiment with at least about 7 carbon atoms, and in one embodiment with no more than about 100 carbon atoms, in another embodiment with no more than about 60 carbon atoms, and in yet another embodiment with no more than about 20 carbon atoms, although the number of carbon atoms can be outside of these ranges, such as tolyl or the like, or (v) a substituent other than an alkyl, aryl, arylalkyl, or alkylaryl group, wherein the substituents on the substituted alkyl, alkylene, aryl, arylene, arylalkyl, arylalkylene, alkylaryl, and alkylarylene groups for R_1 , R'_1 , R_2 , R'_2 , R_3 , R'_3 , R_4 , R'_4 , and R_5 and the substituents other than alkyl, aryl, arylalkyl, or alkylaryl groups can be (but are not limited to) halogen atoms, including fluorine, chlorine, bromine, and iodine atoms, imine groups, ammonium groups, cyano groups, pyridinium groups, ether groups, aldehyde groups, ketone groups, ester groups, carbonyl groups, thiocarbonyl groups, sulfide groups, sulfoxide groups, phosphine groups, nitrile groups, mercapto groups, nitro groups, nitroso groups, sulfone groups, acyl groups, urethane groups, urea groups, mixtures thereof, and the like, wherein two or more substituents can be joined together to form a ring.

[0035] Since hetero atoms can be included in the R_1 and R'_1 groups, R_1 and R'_1 also include alkyleneoxy, aryleneoxy, arylalkyleneoxy, alkylaryleneoxy, polyalkyleneoxy, alkoxyalkylene, alkoxyarylene, pyrrolidine, imidazole, pyrimidinone, oxazoline, thiazoline, and like groups, provided that no oxygen atom is directly bonded to one of the nitrogen atoms. In addition, since hetero atoms can be included in the R_1 and R'_1 groups, R_1 and R'_1 also include heterocyclic groups.

[0036] Since hetero atoms can be included in the R_2 and R'_2 groups, R_2 and R'_2 also include alkoxy, aryloxy, arylalkoxy, alkylaryloxy, polyalkyleneoxy, alkoxyalkyl, alkoxyaryl, pyrrolidine, imidazole,

pyrimidinone, oxazoline, thiazoline, and like groups, provided that no oxygen atom is directly bonded to one of the nitrogen atoms. In addition, since hetero atoms can be included in the R_2 and R'_2 groups, R_2 and R'_2 also include heterocyclic groups.

[0037] Since hetero atoms can be included in the R_5 groups, these groups also includes alkoxy, aryloxy, arylalkoxy, alkylaryloxy, polyalkyleneoxy, alkoxyalkyl, alkoxyaryl, pyrrolidine, imidazole, pyrimidinone, oxazoline, thiazoline, and like groups. In addition, since hetero atoms can be included in the R_5 groups, these groups also include heterocyclic groups.

[0038] In one specific instance, at least one of R_1 and R'_1 have in one embodiment at least about 2 carbon atoms, in another embodiment at least about 4 carbon atoms, and in yet another embodiment at least about 6 carbon atoms, although the number of carbon atoms can be outside of these ranges. In another specific instance, R_1 and R'_1 each have in one embodiment at least about 2 carbon atoms, in another embodiment at least about 4 carbon atoms, and in yet another embodiment at least about 6 carbon atoms, although the number of carbon atoms can be outside of these ranges.

[0039] In one specific instance, R_1 and R'_1 each have in one embodiment no more than about 50 carbon atoms, in another embodiment no more than about 36 carbon atoms, and in yet another embodiment no more than about 12 carbon atoms, although the number of carbon atoms can be outside of these ranges.

[0040] In one specific instance, at least one of R_1 and R'_1 have in one embodiment at least about 6 carbon atoms, in another embodiment at least about 8 carbon atoms, and in yet another embodiment at least about 12 carbon atoms, although the number of carbon atoms can be outside of these ranges. In another specific

instance, R_2 and R'_2 each have in one embodiment at least about 6 carbon atoms, in another embodiment at least about 8 carbon atoms, and in yet another embodiment at least about 12 carbon atoms, although the number of carbon atoms can be outside of these ranges.

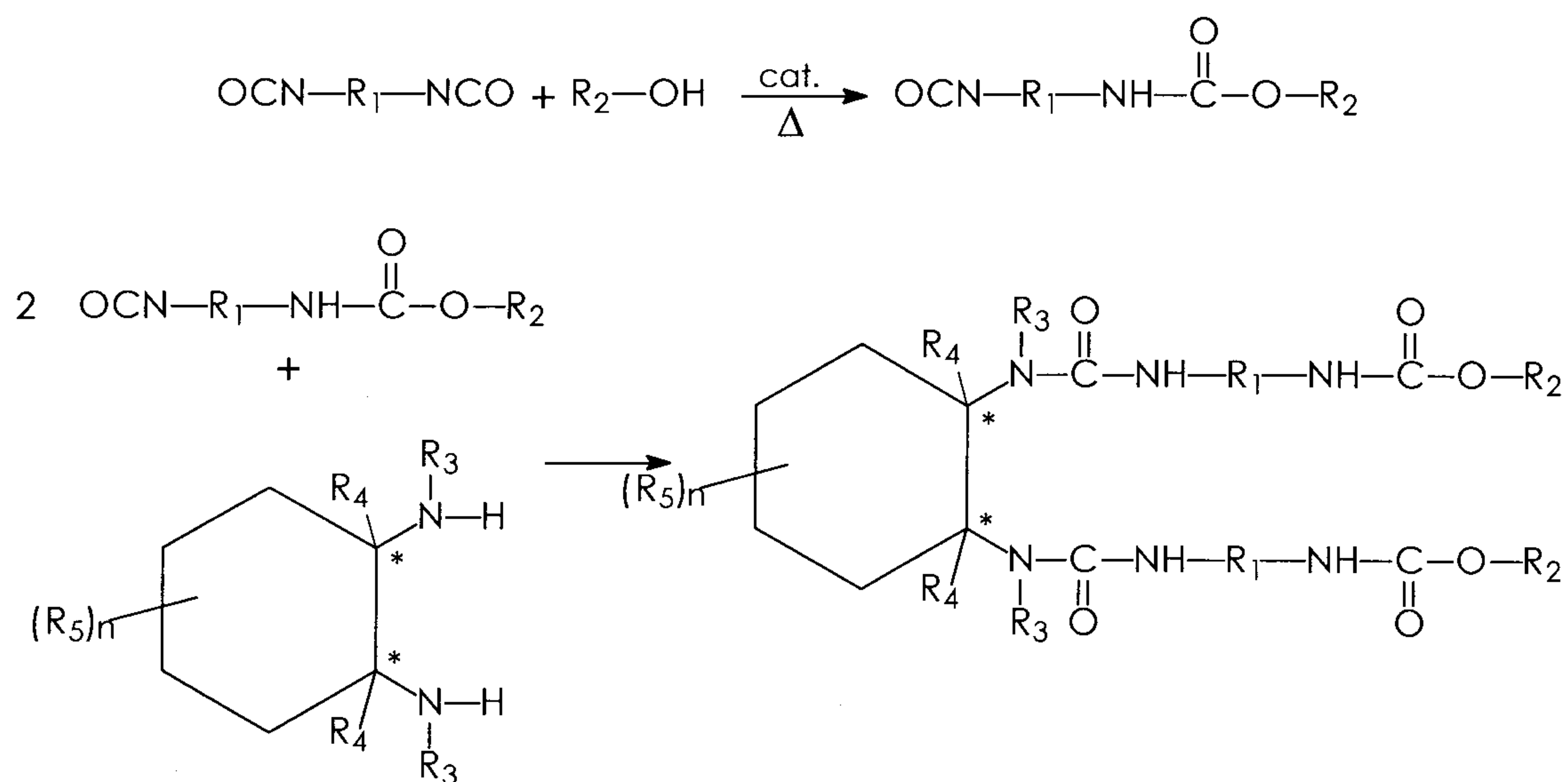
[0041] In one specific instance, R_2 and R'_2 each have in one embodiment no more than about 50 carbon atoms, in another embodiment no more than about 30 carbon atoms, and in yet another embodiment no more than about 18 carbon atoms, although the number of carbon atoms can be outside of these ranges.

[0042] In one specific instance, R_1 , R'_1 , R_2 , and R'_2 have no ethylenic unsaturations. In another specific instance, R_1 , R'_1 , R_2 , R'_2 , R_3 , R'_3 , R_4 , R'_4 , and R_5 have no ethylenic unsaturations.

[0043] In one specific embodiment, R_1 and R'_1 are the same. In another specific embodiment, R_1 and R'_1 are the same and R_2 and R'_2 are the same. In yet another specific embodiment, R_1 and R'_1 are the same, R_2 and R'_2 are the same, R_3 and R'_3 are the same, and R_4 and R'_4 are the same. In still another specific embodiment, R_1 and R'_1 are the same, R_2 and R'_2 are the same, R_3 and R'_3 are the same, R_4 and R'_4 are the same, and n is 0. In another specific embodiment, R_1 and R'_1 are the same, R_2 and R'_2 are the same, R_3 and R'_3 are both hydrogen, R_4 and R'_4 are both hydrogen, and n is 0. In yet another specific embodiment, R_1 and R'_1 are the same, R_2 and R'_2 are the same, R_3 and R'_3 are both hydrogen, R_4 and R'_4 are both fluorine, and n is 0.

[0044] The *trans*-1,2-cyclohexane bis[urea-urethane] compounds can be prepared by any desired or effective method. For example, a monoalcohol of the formula $R_2\text{-OH}$ can be reacted with a diisocyanate of the formula $\text{OCN-}R_1\text{-NCO}$ in approximately equimolar amounts at elevated temperatures, optionally in the presence of a catalyst, and optionally in the presence of a solvent. Thereafter, the resulting

product can be cooled to about room temperature and reacted with about 2 moles of product per every 1 mole of 1,2-diaminocyclohexane substituted as desired, optionally in the presence of a solvent, at room temperature. The reaction proceeds as follows (shown below without representing the stereochemistry; the asterisks indicate the chiral centers):



[0045] The monoalcohol and the diisocyanate are present in any desired or effective relative amounts, in one embodiment at least about 0.4 mole of monoalcohol per every one mole of diisocyanate, in another embodiment at least about 0.6 mole of monoalcohol per every one mole of diisocyanate, and in yet another embodiment at least about 0.8 mole of monoalcohol per every one mole of diisocyanate, and in one embodiment no more than about 1.4 moles of monoalcohol per every one mole of diisocyanate, in another embodiment no more than about 1.2 moles of monoalcohol per every one mole of diisocyanate, and in yet another embodiment no more

than about 1 mole of monoalcohol per every one mole of diisocyanate, although the relative amounts can be outside of these ranges.

[0046] Examples of suitable catalysts include (but are not limited to) Lewis acid catalysts such as dibutyl tin dilaurate, bismuth tris-neodecanoate, cobalt benzoate, lithium acetate, stannous octoate, triethylamine, ferric chloride, aluminum trichloride, boron trichloride, boron trifluoride, titanium tetrachloride, tin tetrachloride, and the like. The catalyst, when present, is present in any desired or effective amount, in one embodiment at least about 0.2 mole percent, in another embodiment at least about 0.5 mole percent, and in yet another embodiment at least about 1 mole percent, and in one embodiment no more than about 10 mole percent, in another embodiment no more than about 7.5 mole percent, and in yet another embodiment no more than about 5 mole percent, based on the amount of diisocyanate, although the amount can be outside of these ranges.

[0047] Examples of suitable solvents for the first part of the reaction include (but are not limited to) toluene, hexane, heptane, methylene chloride, tetrahydrofuran, diethyl ether, ethyl acetate, methyl ethyl ketone, and the like, as well as mixtures thereof. When present, the solvent is present in any desired amount, in one embodiment at least about 10 milliliters per millimole of diisocyanate, in another embodiment at least about 20 milliliters per millimole of diisocyanate, in another embodiment at least about 30 milliliters per millimole of diisocyanate, and in one embodiment no more than about 100 milliliters per millimole of diisocyanate, in another embodiment no more than about 80 milliliters per millimole of diisocyanate, and in yet another embodiment no more than about 50 milliliters per millimole of

diisocyanate, although the amount can be outside of these ranges.

[0048] The diisocyanate and the monoalcohol are heated to any desired or effective temperature, in one embodiment at least about 25°C, in another embodiment at least about 40°C, and in yet another embodiment at least about 50°C, and in one embodiment no more than about 125°C, in another embodiment no more than about 100°C, and in yet another embodiment no more than about 75°C, although the amounts can be outside of these ranges.

[0049] The diisocyanate and the monoalcohol are heated for any desired or effective period of time, in one embodiment at least about 5 minutes, in another embodiment at least about 10 minutes, and in yet another embodiment at least about 15 minutes, and in one embodiment no more than about 80 minutes, in another embodiment no more than about 40 minutes, and in yet another embodiment no more than about 30 minutes, although the time can be outside of these ranges.

[0050] Subsequent to the reaction between the diisocyanate and the monoalcohol, the first reaction product need not be recovered; the reaction mixture can be cooled to room temperature and the appropriately substituted 1,2-diaminocyclohexane can be added to the reaction mixture, along with additional solvent if desired, to complete the reaction.

[0051] The first reaction product and the 1,2-diaminocyclohexane are present in any desired or effective relative amounts, in one embodiment at least about 1.75 moles of first reaction product per every one mole of 1,2-diaminocyclohexane, in another embodiment at least about 1.9 moles of first reaction product per every one mole of 1,2-diaminocyclohexane, and in yet another embodiment at least about 2 moles of first reaction product per every one mole of 1,2-

diaminocyclohexane, and in one embodiment no more than about 2.3 moles of first reaction product per every one mole of 1,2-diaminocyclohexane, in another embodiment no more than about 2.1 moles of first reaction product per every one mole of 1,2-diaminocyclohexane, and in yet another embodiment no more than about 2 moles of first reaction product per every one mole of 1,2-diaminocyclohexane, although the relative amounts can be outside of these ranges.

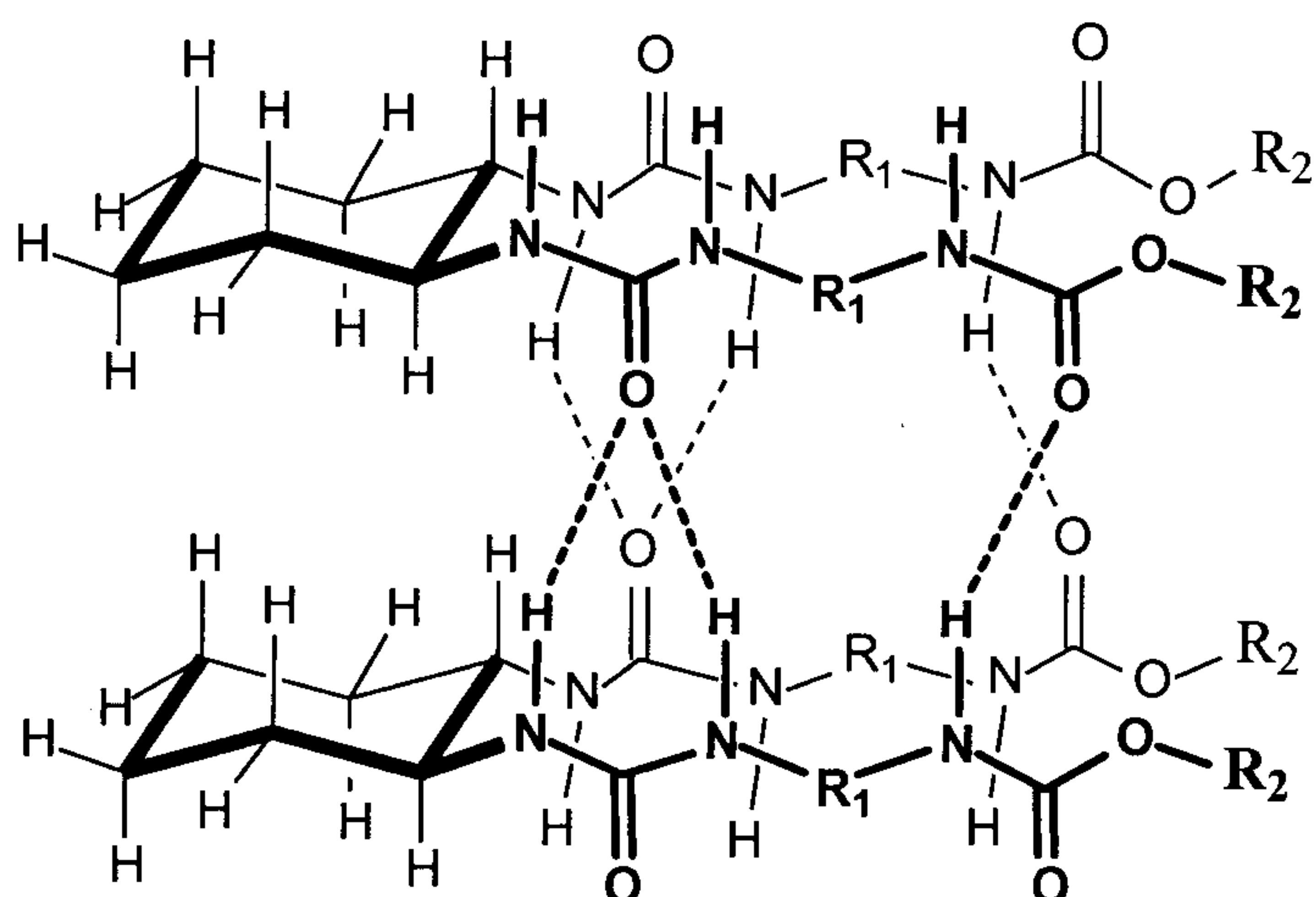
[0052] The first reaction product and the 1,2-diaminocyclohexane are allowed to react at any desired or effective temperature, in one embodiment at least about 10°C, in another embodiment at least about 20°C, and in yet another embodiment at least about 30°C, and in one embodiment no more than about 75°C, in another embodiment no more than about 50°C, and in yet another embodiment no more than about 40°C, although the temperature can be outside of these ranges.

[0053] The first reaction product and the 1,2-diaminocyclohexane are allowed to react for any desired or effective period of time, in one embodiment at least about 5 minutes, in another embodiment at least about 10 minutes, and in yet another embodiment at least about 20 minutes, and in one embodiment no more than about 3 hours, in another embodiment no more than about 1.5 hours, and in yet another embodiment no more than about 1 hour, although the time can be outside of these ranges.

[0054] Thereafter, the product can be precipitated by addition of a small amount of a non-solvent, such as hexane or methylene chloride, followed by good stirring. The product can then be recovered by filtration.

[0055] While not being limited to any particular theory, it is

believed that the *trans*-1,2-cyclohexane bis[urea-urethane] compounds disclosed herein form reversible hydrogen bonds, resulting in the formation of oligomers and oligomer networks held together by non-covalent hydrogen bonds instead of covalent bonds. One example of such bond formation is illustrated as follows:



While not being limited to any particular theory, it is believed that in the inks containing these *trans*-1,2-cyclohexane bis[urea-urethane] compounds, at least some and perhaps all of these hydrogen bonds can be broken at the temperatures at which hot melt ink jet printing occurs (typically, although not necessarily, over 100°C). When the ink is printed onto an intermediate transfer member or a final recording substrate, the ink cools as it is printed, which results in reformation of any hydrogen bonds broken by heating. The polymer-like materials thus formed behave like conventional covalently-bonded polymers to enhance image permanence. The image robustness can be increased by adding a *trans*-1,2-cyclohexane bis[urea-urethane] gelator compound to the ink. The gelator molecules can self-assemble into 3-dimensional fibrous networks by intermolecular hydrogen bonding and van der Waals interactions. The molten ink is expected to get trapped into these gel networks and form a semi-solid or a gel. In addition, the

gelled inks exhibit visco-elastic rheological characteristics that are different from those of conventional hot melt or phase change inks in that they show an elastic behavior in a region where the ink is supposed to be in the liquid state. This behavior is evidenced by the crossover of G' (storage modulus) and G'' (loss modulus), with G' being higher than G'' , indicating that the material is elastic. The elasticity of the material can also be expressed using tan-delta, which is defined as the ratio of G'' to G' , or G''/G' . A material which has a tan-delta of less than one is elastic, whereas a non-elastic material will not have a tan-delta of less than one above its melting point. The *trans*-1,2-cyclohexane bis[urea-urethane] gelator compounds, when present in phase change inks, can enable an intermediate gel phase wherein the gel phase transition entails a tan-delta of in one embodiment less than about 10, in another embodiment less than about 5, and in yet another embodiment less than about 1, although the tan-delta can be outside of these ranges. This elasticity can further enhance the robustness of images generated with the inks containing the *trans*-1,2-cyclohexane bis[urea-urethane] compounds. The *trans*-1,2-cyclohexane bis[urea-urethane] gelator compounds can also enable desirably narrow gel phase transitions in the inks, in one embodiment gel phase transitions 0.1 to 40°C wide, in another embodiment gel phase transitions 0.1 to 20°C wide, and in yet another embodiment gel phase transitions 0.1 to 15°C wide, although the gel phase transitions can be outside of these ranges.

[0056] Phase change inks as disclosed herein in one specific embodiment exhibit a gel phase or state from about 1°C to about 40°C above the ink melting point, in another specific embodiment exhibit a gel phase or state from about 1°C to about 20°C above the ink melting point, and in yet another specific embodiment exhibit a gel phase or state from about 2°C to about 15°C above the ink melting point,

although the gel phase or state can be exhibited outside of these ranges.

[0057] The formation of hydrogen-bonded oligomers or polymers from specific ink carrier materials can be determined by any desired method. For example, a dramatic onset of resinous and viscoelastic characteristics on cooling is indicative of the formation of hydrogen-bonded oligomers or polymers from the ink carrier material or combination of materials. The formation of hydrogen bonds and hydrogen-bonded oligomers or polymers can also be detected by IR spectroscopy. NMR spectroscopy may also help to detect the presence of hydrogen-bonded oligomers or polymers. In situations wherein the ink carrier material is crystalline, X-ray crystallography can be used to define the oligomeric or polymeric structure.

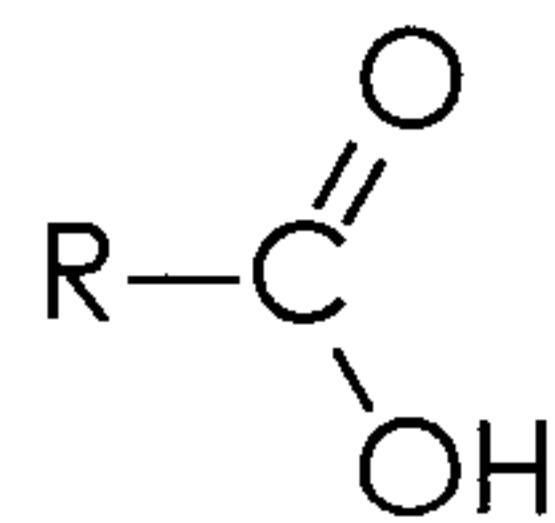
[0058] Further information on gels is disclosed in, for example, *Gels Handbook*, Vol. 1-4, Editors-in-Chief, Y. Osada and K. Kajiwara (translated by H. Ishida), **2001**, Academic Press.

[0059] Phase change inks as disclosed herein contain a phase change carrier system or composition. The phase change carrier composition is typically designed for use in either a direct printing mode or an indirect or offset printing transfer system.

[0060] In the direct printing mode, the phase change carrier composition in one embodiment contains one or more materials that enable the phase change ink (1) to be applied in a thin film of uniform thickness on the final recording substrate (such as paper, transparency material, and the like) when cooled to ambient temperature after printing directly to the recording substrate, (2) to be ductile while retaining sufficient flexibility so that the applied image on the substrate will not fracture upon bending, and (3) to possess a high degree of lightness, chroma, transparency, and thermal stability.

[0061] In an offset printing transfer or indirect printing mode, the phase change carrier composition in one embodiment exhibits not only the characteristics desirable for direct printing mode inks, but also certain fluidic and mechanical properties desirable for use in such a system, as described in, for example, U.S. Patent 5,389,958.

[0062] Any desired or effective carrier composition can be used. Examples of suitable ink carrier materials include fatty amides, such as monoamides, tetra-amides, mixtures thereof, and the like. Specific examples of suitable fatty amide ink carrier materials include stearyl stearamide, a dimer acid based tetra-amide that is the reaction product of dimer acid, ethylene diamine, and stearic acid, a dimer acid based tetra-amide that is the reaction product of dimer acid, ethylene diamine, and a carboxylic acid having at least about 36 carbon atoms, and the like, as well as mixtures thereof. When the fatty amide ink carrier is a dimer acid based tetra-amide that is the reaction product of dimer acid, ethylene diamine, and a carboxylic acid having at least about 36 carbon atoms, the carboxylic acid is of the general formula



wherein R is an alkyl group, including linear, branched, saturated, unsaturated, and cyclic alkyl groups, said alkyl group in one embodiment having at least about 36 carbon atoms, in another embodiment having at least about 40 carbon atoms, said alkyl group in one embodiment having no more than about 200 carbon atoms, in another embodiment having no more than about 150 carbon atoms, and in yet another embodiment having no more than about 100

carbon atoms, although the number of carbon atoms can be outside of these ranges. Carboxylic acids of this formula are commercially available from, for example, Baker Petrolite, Tulsa, OK, and can also be prepared as described in Example 1 of U.S. Patent 6,174,937. Further information on fatty amide carrier materials is disclosed in, for example, U.S. Patent 4,889,560, U.S. Patent 4,889,761, U.S. Patent 5,194,638, U.S. Patent 4,830,671, U.S. Patent 6,174,937, U.S. Patent 5,372,852, U.S. Patent 5,597,856, U.S. Patent 6,174,937, and British Patent GB 2 238 792.

[0063] Also suitable as phase change ink carrier materials are isocyanate-derived resins and waxes, such as urethane isocyanate-derived materials, urea isocyanate-derived materials, urethane/urea isocyanate-derived materials, mixtures thereof, and the like. Further information on isocyanate-derived carrier materials is disclosed in, for example, U.S. Patent 5,750,604, U.S. Patent 5,780,528, U.S. Patent 5,782,966, U.S. Patent 5,783,658, U.S. Patent 5,827,918, U.S. Patent 5,830,942, U.S. Patent 5,919,839, U.S. Patent 6,255,432, U.S. Patent 6,309,453, British Patent GB 2 294 939, British Patent GB 2 305 928, British Patent GB 2 305 670, British Patent GB 2 290 793, PCT Publication WO 94/14902, PCT Publication WO 97/12003, PCT Publication WO 97/13816, PCT Publication WO 96/14364, PCT Publication WO 97/33943, and PCT Publication WO 95/04760.

[0064] Mixtures of fatty amide materials and isocyanate-derived materials can also be employed as the ink carrier composition.

[0065] Additional suitable phase change ink carrier materials include paraffins, microcrystalline waxes, polyethylene waxes, ester waxes, amide waxes, fatty acids, fatty alcohols, such as the UNILIN® products available from Baker Petrolite, Tulsa, OK, fatty amides and other waxy materials, sulfonamide materials, resinous materials made from different natural sources (such as, for example, tall oil rosins and

rosin esters), and many synthetic resins, oligomers, polymers and copolymers, such as ethylene/vinyl acetate copolymers, ethylene/acrylic acid copolymers, ethylene/vinyl acetate/acrylic acid copolymers, copolymers of acrylic acid with polyamides, and the like, ionomers, and the like, as well as mixtures thereof. One or more of these materials can also be employed in a mixture with a fatty amide material and/or an isocyanate-derived material.

[0066] The ink carrier can also optionally contain an antioxidant. The optional antioxidants protect the images from oxidation and also protect the ink components from oxidation during the heating portion of the ink preparation process. Specific examples of suitable antioxidants include NAUGUARD® 445, NAUGUARD® 524, NAUGUARD® 76, and NAUGUARD® 512 (commercially available from Uniroyal Chemical Company, Oxford, CT), IRGANOX® 1010 (commercially available from Ciba Geigy), and the like. When present, the optional antioxidant is present in the ink in any desired or effective amount, in one embodiment of at least about 0.01 percent by weight of the ink carrier, in another embodiment of at least about 0.1 percent by weight of the ink carrier, and in yet another embodiment of at least about 1 percent by weight of the ink carrier, and in one embodiment of no more than about 20 percent by weight of the ink carrier, in another embodiment of no more than about 5 percent by weight of the ink carrier, and in yet another embodiment of no more than about 3 percent by weight of the ink carrier, although the amount can be outside of these ranges.

[0067] The ink carrier can also optionally contain a viscosity modifier. Examples of suitable viscosity modifiers include aliphatic ketones, such as stearone, and the like. When present, the optional viscosity modifier is present in the ink in any desired or effective amount,

in one embodiment of at least about 0.1 percent by weight of the ink carrier, in another embodiment of at least about 1 percent by weight of the ink carrier, and in yet another embodiment of at least about 10 percent by weight of the ink carrier, and in one embodiment of no more than about 99 percent by weight of the ink carrier, in another embodiment of no more than about 30 percent by weight of the ink carrier, and in yet another embodiment of no more than about 15 percent by weight of the ink carrier, although the amount can be outside of these ranges.

[0068] Other optional additives to the ink carrier include clarifiers, such as UNION CAMP® X37-523-235 (commercially available from Union Camp), in an amount in one embodiment of at least about 0.01 percent by weight of the ink carrier, in another embodiment of at least about 0.1 percent by weight of the ink carrier, and in yet another embodiment of at least about 5 percent by weight of the ink carrier, and in one embodiment of no more than about 98 percent by weight of the ink carrier, in another embodiment of no more than about 50 percent by weight of the ink carrier, and in yet another embodiment of no more than about 10 percent by weight of the ink carrier, although the amount can be outside of these ranges, tackifiers, such as FORAL® 85, a glycerol ester of hydrogenated abietic (rosin) acid (commercially available from Hercules), FORAL® 105, a pentaerythritol ester of hydroabietic (rosin) acid (commercially available from Hercules), CELLOLYN® 21, a hydroabietic (rosin) alcohol ester of phthalic acid (commercially available from Hercules), ARAKAWA KE-311 and KE-100 Resins, triglycerides of hydrogenated abietic (rosin) acid (commercially available from Arakawa Chemical Industries, Ltd.), synthetic polyterpene resins such as NEVTAC® 2300, NEVTAC® 100, and NEVTAC® 80 (commercially available from Neville Chemical

Company), WINGTACK® 86, a modified synthetic polyterpene resin (commercially available from Goodyear), and the like, in an amount in one embodiment of at least about 0.1 percent by weight of the ink carrier, in another embodiment of at least about 5 percent by weight of the ink carrier, and in yet another embodiment of at least about 10 percent by weight of the ink carrier, and in one embodiment of no more than about 98 percent by weight of the ink carrier, in another embodiment of no more than about 75 percent by weight of the ink carrier, and in yet another embodiment of no more than about 50 percent by weight of the ink carrier, although the amount can be outside of these range, adhesives, such as VERSAMID® 757, 759, or 744 (commercially available from Henkel), in an amount in one embodiment of at least about 0.1 percent by weight of the ink carrier, in another embodiment of at least about 1 percent by weight of the ink carrier, and in yet another embodiment of at least about 5 percent by weight of the ink carrier, and in one embodiment of no more than about 98 percent by weight of the ink carrier, in another embodiment of no more than about 50 percent by weight of the ink carrier, and in yet another embodiment of no more than about 10 percent by weight of the ink carrier, although the amount can be outside of these ranges, plasticizers, such as UNIPLEX® 250 (commercially available from Uniplex), the phthalate ester plasticizers commercially available from Monsanto under the trade name SANTICIZER®, such as dioctyl phthalate, diundecyl phthalate, alkylbenzyl phthalate (SANTICIZER® 278), triphenyl phosphate (commercially available from Monsanto), KP-140®, a tributoxyethyl phosphate (commercially available from FMC Corporation), MORFLEX® 150, a dicyclohexyl phthalate (commercially available from Morflex Chemical Company Inc.), trioctyl trimellitate (commercially available from Eastman Kodak Co.), and the like, in an

amount in one embodiment of at least about 0.1 percent by weight of the ink carrier, in another embodiment of at least about 1 percent by weight of the ink carrier, and in yet another embodiment of at least about 2 percent by weight of the ink carrier, and in one embodiment of no more than about 50 percent by weight of the ink carrier, in another embodiment of no more than about 30 percent by weight of the ink carrier, and in yet another embodiment of no more than about 10 percent by weight of the ink carrier, although the amount can be outside of these ranges, and the like.

[0069] The phase change ink can contain the *trans*-1,2-cyclohexane bis[urea-urethane] compound as a stereoisomerically pure form of the 1*R*,2*R* form or the 1*S*,2*S* form, or as a mixture of the two stereoisomeric forms, including but not limited to racemic mixtures. The *trans*-1,2-cyclohexane bis[urea-urethane] compound is present in the phase change ink in any desired or effective amount, in one embodiment at least about 0.1 percent by weight of the phase change ink, in another embodiment at least about 1 percent by weight of the phase change ink, and in yet another embodiment at least about 5 percent by weight of the phase change ink, and in one embodiment no more than about 30 percent by weight of the phase change ink, in another embodiment no more than about 15 percent by weight of the phase change ink, and in yet another embodiment no more than about 10 percent by weight of the phase change ink, although the amount can be outside of these ranges.

[0070] The ink carrier is present in the phase change ink in any desired or effective amount, in one embodiment of at least about 25 percent by weight of the ink, in another embodiment of at least about 50 percent by weight of the ink, and in yet another embodiment of at least about 90 percent by weight of the ink, and in one embodiment of

no more than about 99 percent by weight of the ink, in another embodiment of no more than about 98 percent by weight of the ink, and in yet another embodiment of no more than about 95 percent by weight of the ink, although the amount can be outside of these ranges.

[0071] In one specific embodiment, the ink carrier has a melting point of higher than about 110°C, and in another embodiment of higher than about 100°C, although the melting point of the ink carrier can be outside of these ranges.

[0072] The phase change ink compositions also contain a colorant. Any desired or effective colorant can be employed, including dyes, pigments, mixtures thereof, and the like, provided that the colorant can be dissolved or dispersed in the ink vehicle. The phase change carrier compositions can be used in combination with conventional phase change ink colorant materials, such as Color Index (C.I.) Solvent Dyes, Disperse Dyes, modified Acid and Direct Dyes, Basic Dyes, Sulphur Dyes, Vat Dyes, and the like. Examples of suitable dyes include Neozapon Red 492 (BASF); Orasol Red G (Ciba-Geigy); Direct Brilliant Pink B (Crompton & Knowles); Aizen Spilon Red C-BH (Hodogaya Chemical); Kayanol Red 3BL (Nippon Kayaku); Levanol Brilliant Red 3BW (Mobay Chemical); Levaderm Lemon Yellow (Mobay Chemical); Spirit Fast Yellow 3G; Aizen Spilon Yellow C-GNH (Hodogaya Chemical); Sirius Supra Yellow GD 167; Cartasol Brilliant Yellow 4GF (Sandoz); Pergasol Yellow CGP (Ciba-Geigy); Orasol Black RLP (Ciba-Geigy); Savinyl Black RLS (Sandoz); Dermacarbon 2GT (Sandoz); Pyrazol Black BG (ICI); Morfast Black Conc. A (Morton-Thiokol); Diaazol Black RN Quad (ICI); Orasol Blue GN (Ciba-Geigy); Savinyl Blue GLS (Sandoz); Luxol Blue MBSN (Morton-Thiokol); Sevron Blue 5GMF (ICI); Basacid Blue 750 (BASF), Neozapon Black X51 [C.I. Solvent Black, C.I. 12195] (BASF), Sudan Blue 670 [C.I. 61554] (BASF), Sudan Yellow 146 [C.I. 12700] (BASF), Sudan Red

462 [C.I. 26050] (BASF), Intratherm Yellow 346 from Crompton and Knowles, C.I. Disperse Yellow 238, Neptune Red Base NB543 (BASF, C.I. Solvent Red 49), Neopen Blue FF-4012 from BASF, Lampronol Black BR from ICI (C.I. Solvent Black 35), Morton Morplas Magenta 36 (C.I. Solvent Red 172), metal phthalocyanine colorants such as those disclosed in U.S. Patent 6,221,137 and the like. Polymeric dyes can also be used, such as those disclosed in, for example, U.S. Patent 5,621,022 and U.S. Patent 5,231,135, and commercially available from, for example, Milliken & Company as Milliken Ink Yellow 12, Milliken Ink Blue 92, Milliken Ink Red 357, Milliken Ink Yellow 1800, Milliken Ink Black 8915-67, uncut Reactant Orange X-38, uncut Reactant Blue X-17, Solvent Yellow 162, Acid Red 52, Solvent Blue 44, and uncut Reactant Violet X-80.

[0073] Pigments are also suitable colorants for the phase change inks. Examples of suitable pigments include Violet Toner VT-8015 (Paul Uhlich); Paliogen Violet 5100 (BASF); Paliogen Violet 5890 (BASF); Permanent Violet VT 2645 (Paul Uhlich); Heliogen Green L8730 (BASF); Argyle Green XP-111-S (Paul Uhlich); Brilliant Green Toner GR 0991 (Paul Uhlich); Lithol Scarlet D3700 (BASF); Toluidine Red (Aldrich); Scarlet for Thermoplast NSD PS PA (Ugine Kuhlmann of Canada); E.D. Toluidine Red (Aldrich); Lithol Rubine Toner (Paul Uhlich); Lithol Scarlet 4440 (BASF); Bon Red C (Dominion Color Company); Royal Brilliant Red RD-8192 (Paul Uhlich); Oracet Pink RF (Ciba-Geigy); Paliogen Red 3871K (BASF); Paliogen Red 3340 (BASF); Lithol Fast Scarlet L4300 (BASF); Heliogen Blue L6900, L7020 (BASF); Heliogen Blue K6902, K6910 (BASF); Heliogen Blue D6840, D7080 (BASF); Sudan Blue OS (BASF); Neopen Blue FF4012 (BASF); PV Fast Blue B2G01 (American Hoechst); Irgalite Blue BCA (Ciba-Geigy); Paliogen Blue 6470 (BASF); Sudan III (Red Orange) (Matheson, Coleman Bell); Sudan II (Orange) (Matheson, Coleman Bell); Sudan Orange G

(Aldrich), Sudan Orange 220 (BASF); Paliogen Orange 3040 (BASF); Ortho Orange OR 2673 (Paul Uhlich); Paliogen Yellow 152, 1560 (BASF); Lithol Fast Yellow 0991K (BASF); Palitol Yellow 1840 (BASF); Novoperm Yellow FGL (Hoechst); Permanent Yellow YE 0305 (Paul Uhlich); Lumogen Yellow D0790 (BASF); Suco-Yellow L1250 (BASF); Suco-Yellow D1355 (BASF); Suco Fast Yellow D1355, D1351 (BASF); Hostaperm Pink E (American Hoechst); Fanal Pink D4830 (BASF); Cinquasia Magenta (Du Pont); Paliogen Black L0084 (BASF); Pigment Black K801 (BASF); and carbon blacks such as REGAL 330® (Cabot), Carbon Black 5250, Carbon Black 5750 (Columbia Chemical), and the like.

[0074] Also suitable are the colorants disclosed in U.S. Patent 6,472,523, U.S. Patent 6,726,755, U.S. Patent 6,476,219, U.S. Patent 6,576,747, U.S. Patent 6,713,614, U.S. Patent 6,663,703, U.S. Patent 6,755,902, U.S. Patent 6,590,082, U.S. Patent 6,696,552, U.S. Patent 6,576,748, U.S. Patent 6,646,111, U.S. Patent 6,673,139, Copending Application U.S. Serial No. 10/260,146, filed September 27, 2002, entitled "Colorant Compounds," and Copending Application U.S. Serial No. 10/260,379, filed September 27, 2002, entitled "Methods for Making Colorant Compounds."

[0075] Other ink colors besides the subtractive primary colors can be desirable for applications such as postal marking, industrial marking, and labelling using phase change printing, and the inks are applicable to these needs. Further, infrared (IR) or ultraviolet (UV) absorbing dyes can also be incorporated into the inks for use in applications such as "invisible" coding or marking of products. Examples of such infrared and ultraviolet absorbing dyes are disclosed in, for example, U.S. Patent 5,378,574, U.S. Patent 5,146,087, U.S. Patent 5,145,518, U.S. Patent 5,543,177, U.S. Patent 5,225,900, U.S. Patent 5,301,044, U.S. Patent 5,286,286, U.S. Patent 5,275,647, U.S. Patent 5,208,630, U.S. Patent

5,202,265, U.S. Patent 5,271,764, U.S. Patent 5,256,193, U.S. Patent 5,385,803, and U.S. Patent 5,554,480.

[0076] In a specific embodiment, the colorant is an isocyanate-derived colored resin as disclosed in, for example, U.S. Patent 5,780,528 and U.S. Patent 5,919,839. In this embodiment, the colorant is the reaction product of a hydroxyl-substituted or primary or secondary amino-substituted chromophore with an isocyanate. Examples of suitable isocyanates include monoisocyanates, diisocyanates, triisocyanates, copolymers of a diisocyanate, copolymers of a triisocyanate, polyisocyanates (having more than three isocyanate functional groups), and the like, as well as mixtures thereof. Specific examples of suitable isocyanates include those listed hereinabove as being suitable for reaction with the hydroxyl-substituted or amino-substituted antioxidant. Examples of suitable hydroxyl-substituted and primary or secondary amino-substituted chromophores include those disclosed in, for example, U.S. Patent 3,157,633, U.S. Patent 3,927,044, U.S. Patent 3,994,835, U.S. Patent 4,102,644, U.S. Patent 4,113,721, U.S. Patent 4,132,840, U.S. Patent 4,137,243, U.S. Patent 4,170,564, U.S. Patent 4,284,729, U.S. Patent 4,507,407, U.S. Patent 4,640,690, U.S. Patent 4,732,570, U.S. Patent 4,751,254, U.S. Patent 4,751,254, U.S. Patent 4,761,502, U.S. Patent 4,775,748, U.S. Patent 4,812,141, U.S. Patent 4,846,846, U.S. Patent 4,871,371, U.S. Patent 4,912,203, U.S. Patent 4,978,362, U.S. Patent 5,043,013, U.S. Patent 5,059,244, U.S. Patent 5,149,800, U.S. Patent 5,177,200, U.S. Patent 5,270,363, U.S. Patent 5,290,921, and U.S. Patent 5,731,398. Hydroxyl-containing and primary or secondary amino-containing colorants from the classes of Color Index (C.I.) Solvent Dyes, Disperse Dyes, modified Acid and Direct Dyes, Basic Dyes, Sulphur Dyes, Vat Dyes, and the like can also be used. These colorants can also affect the rheological properties of the inks

containing them.

[0077] The colorant is present in the phase change ink in any desired or effective amount to obtain the desired color or hue, typically at least about 0.1 percent by weight of the ink, preferably at least about 0.2 percent by weight of the ink, and more preferably at least about 0.5 percent by weight of the ink, and typically no more than about 50 percent by weight of the ink, preferably no more than about 20 percent by weight of the ink, and more preferably no more than about 10 percent by weight of the ink, although the amount can be outside of these ranges.

[0078] The ink compositions in one embodiment have melting points of no lower than about 40°C, in another embodiment of no lower than about 60°C, and in yet another embodiment of no lower than about 70°C, and have melting points in one embodiment of no higher than about 140°C, in another embodiment of no higher than about 120°C, and in yet another embodiment of no higher than about 100°C, although the melting point can be outside of these ranges.

[0079] The ink compositions generally have melt viscosities at the jetting temperature (in one embodiment no lower than about 75°C, in another embodiment no lower than about 100°C, and in yet another embodiment no lower than about 120°C, and in one embodiment no higher than about 180°C, and in another embodiment no higher than about 150°C, although the jetting temperature can be outside of these ranges) in one embodiment of no more than about 30 centipoise, in another embodiment of no more than about 20 centipoise, and in yet another embodiment of no more than about 15 centipoise, and in one embodiment of no less than about 2 centipoise, in another embodiment of no less than about 5 centipoise, and in yet another embodiment of no less than about 7 centipoise, although the melt

viscosity can be outside of these ranges.

[0080] The ink compositions can be prepared by any desired or suitable method. For example, the ink ingredients can be mixed together, followed by heating, to a temperature in one embodiment of at least about 100°C, and in one embodiment of no more than about 140°C, although the temperature can be outside of these ranges, and stirring or milling until a homogeneous ink composition is obtained, followed by cooling the ink to ambient temperature (typically from about 20 to about 25°C). The inks are solid at ambient temperature. In a specific embodiment, during the formation process, the inks in their molten state are poured into molds and then allowed to cool and solidify to form ink sticks.

[0081] The inks can be employed in apparatus for direct printing ink jet processes and in indirect (offset) printing ink jet applications. Another embodiment disclosed herein is directed to a process which comprises incorporating an ink as disclosed herein into an ink jet printing apparatus, melting the ink, and causing droplets of the melted ink to be ejected in an imagewise pattern onto a recording substrate. A direct printing process is also disclosed in, for example, U.S. Patent 5,195,430. Yet another embodiment disclosed herein is directed to a process which comprises incorporating an ink as disclosed herein into an ink jet printing apparatus, melting the ink, causing droplets of the melted ink to be ejected in an imagewise pattern onto an intermediate transfer member, and transferring the ink in the imagewise pattern from the intermediate transfer member to a final recording substrate. In a specific embodiment, the intermediate transfer member is heated to a temperature above that of the final recording sheet and below that of the melted ink in the printing apparatus. In one embodiment, the intermediate transfer member is heated to a temperature of from

about 4°C above to about 60°C below the ink melting temperature, and in another embodiment, the intermediate transfer member is heated to a temperature of from about 2°C above to about 50°C below the ink melting temperature, although the temperature of the intermediate transfer member can be outside of these ranges. An offset or indirect printing process is also disclosed in, for example, U.S. Patent 5,389,958. In one specific embodiment, the printing apparatus employs a piezoelectric printing process wherein droplets of the ink are caused to be ejected in imagewise pattern by oscillations of piezoelectric vibrating elements. Inks as disclosed herein can also be employed in other hot melt printing processes, such as hot melt acoustic ink jet printing, hot melt thermal ink jet printing, hot melt continuous stream or deflection ink jet printing, and the like. Phase change inks as disclosed herein can also be used in printing processes other than hot melt ink jet printing processes.

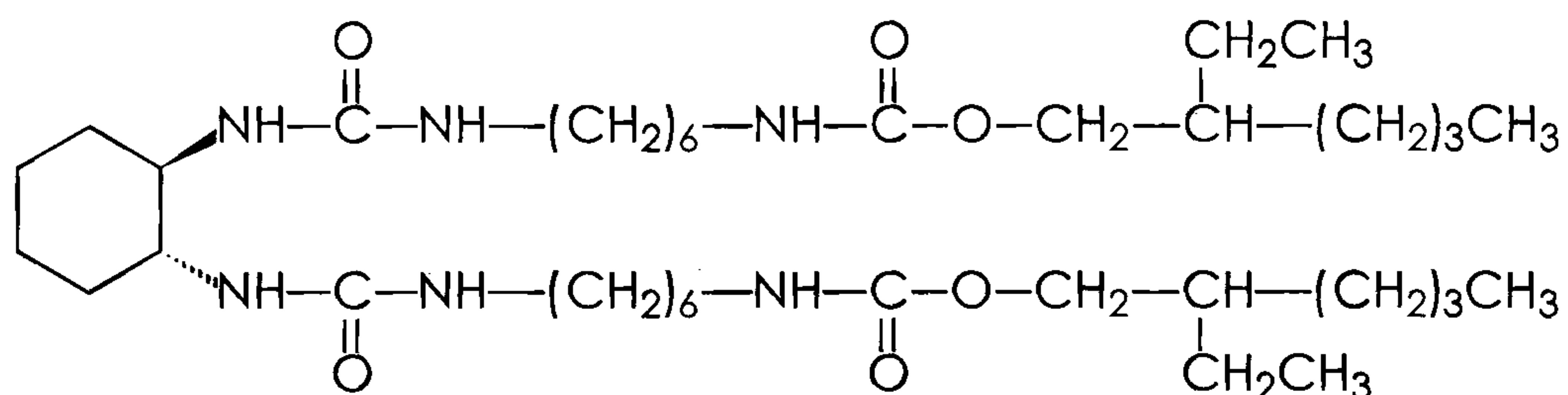
[0082] Any suitable substrate or recording sheet can be employed, including plain papers such as XEROX® 4024 papers, XEROX® Image Series papers, Courtland™ 4024 DP paper, ruled notebook paper, bond paper, silica coated papers such as Sharp Company silica coated paper, JuJo™ paper, HAMMERMILL LASERPRINT® paper, and the like, transparency materials, fabrics, textile products, plastics, polymeric films, inorganic substrates such as metals and wood, and the like.

[0083] Specific embodiments will now be described in detail. These examples are intended to be illustrative, and the claims are not limited to the materials, conditions, or process parameters set forth in these embodiments. All parts and percentages are by weight unless otherwise indicated.

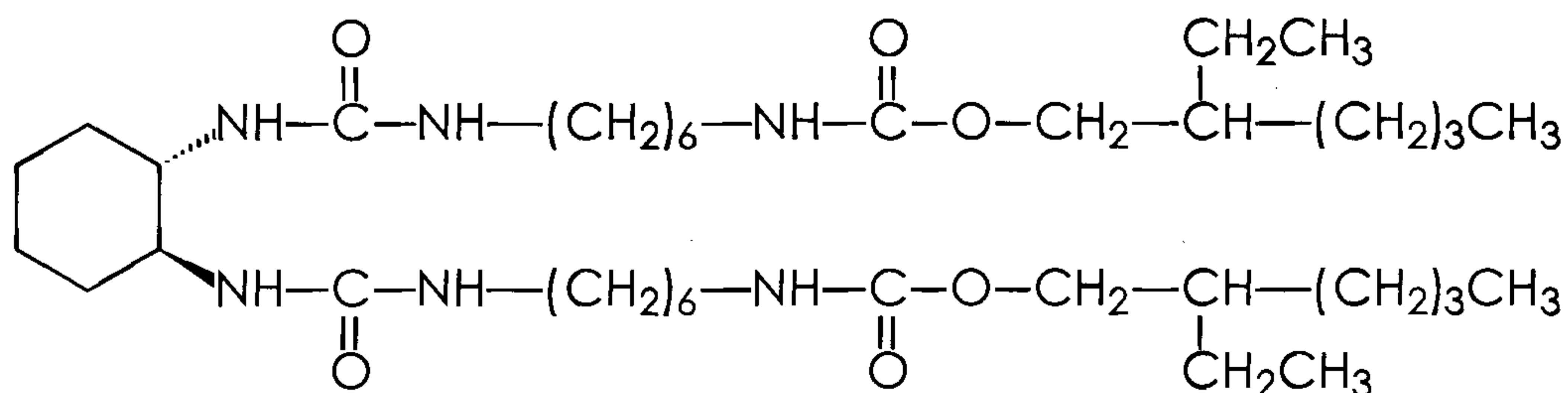
EXAMPLE I

[0084] Into a solution containing 1,6-diisocyanatohexane (4.04 grams, 24.0 mmol; obtained from Sigma-Aldrich Fine Chemicals, Milwaukee, WI) and anhydrous tetrahydrofuran (100 mL, Sigma-Aldrich Fine Chemicals, Milwaukee, WI) stirring at room temperature was added 2-ethylhexanol (3.13 grams, 24.0 mmol, obtained from Sigma-Aldrich Fine Chemicals) and dibutyltin dilaurate (0.38 grams, 0.6 mmol, obtained from Sigma-Aldrich Fine Chemicals) as the catalyst. The mixture was stirred and heated to an internal temperature of about 70°C. The progress of the reaction was monitored by ¹H-NMR spectroscopy for the consumption of 2-ethylhexanol starting material, indicated by the disappearance of the -CH₂OH multiplet, which appears at 3.5 ppm as a shoulder peak on the downfield end of the intermediate isocyanate product whose signal is located at 3.35-3.40 ppm. The mixture was cooled to about 5°C internal temperature; thereafter, to this mixture was added dropwise a solution of *trans*-1,2-diaminocyclohexane (1.37 grams, 12 mmol; obtained as a racemic mixture of (1*R*,2*R*) and (1*S*,2*S*) stereoisomers from Sigma-Aldrich Fine Chemicals) dissolved in anhydrous tetrahydrofuran (10 mL). The mixture was stirred for about 30 minutes while warming up to room temperature, and thickened to form a gelatinous slurry. FTIR spectroscopic analysis of a reaction sample showed very little unreacted isocyanate (peak at 2180 cm⁻¹, sample prepared as a KBr pellet). Residual isocyanate was quenched by addition of 5 mL of methanol. A crystalline product was isolated from the slurry by first adding methylene chloride (40 mL) followed with stirring for approximately 20 minutes to ensure full precipitation out of the gel

slurry. The solid was filtered by suction on a paper filter, rinsed with methylene chloride (about 10 mL), and then dried in air to give 7.36 grams of off-white solid (86% yield). The product was believed to be of the formulae



and



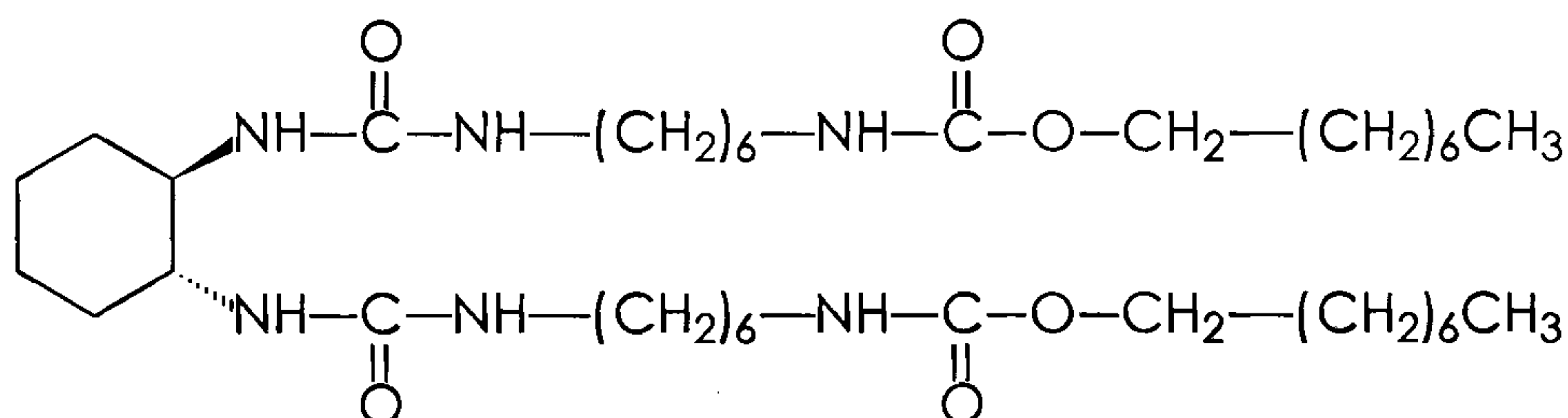
^1H -NMR spectroscopic analysis of the solid was performed in $\text{DMSO-}d_6$ (300 MHz) at high temperature (60°C) and indicated the above structure, with the following assigned peaks: 0.90 ppm (multiplet, 6H integration, $-\text{OCH}_2\text{CH}(\text{CH}_2\text{CH}_3)\text{CH}_2\text{CH}_2\text{CH}_2\text{CH}_3$); 1.0-1.95 ppm (broad multiplets, 20 H integration, 8 methylene protons from 2-ethylhexanol portion, 8 methylene protons from the 1,6-diisocyanatohexane portion, and 4 methylene protons from the cyclohexane ring portion); 2.95 ppm (narrow multiplet, 4H integration, $-\text{NH}(\text{C}=\text{O})\text{NHCH}_2(\text{CH}_2)_4\text{CH}_2\text{NH}(\text{C}=\text{O})\text{O}$); 3.20 ppm (broad singlet, 1 H integration, tertiary methine proton adjacent to urea group on cyclohexane ring); 3.90 ppm (doublet, 2H integration, $\text{OCH}_2\text{CH}(\text{CH}_2\text{CH}_3)\text{CH}_2\text{CH}_2\text{CH}_2\text{CH}_3$); 5.65 ppm and 5.75 ppm (each a broad singlet, 1H integration, urea NH protons); 6.75 ppm (broad singlet,

¹H integration, urethane NH proton). Elemental analysis calculated for C: 64.19%, H: 10.49%, N: 11.82%; found for C: 61.70%, H: 9.86%, N: 14.91%.

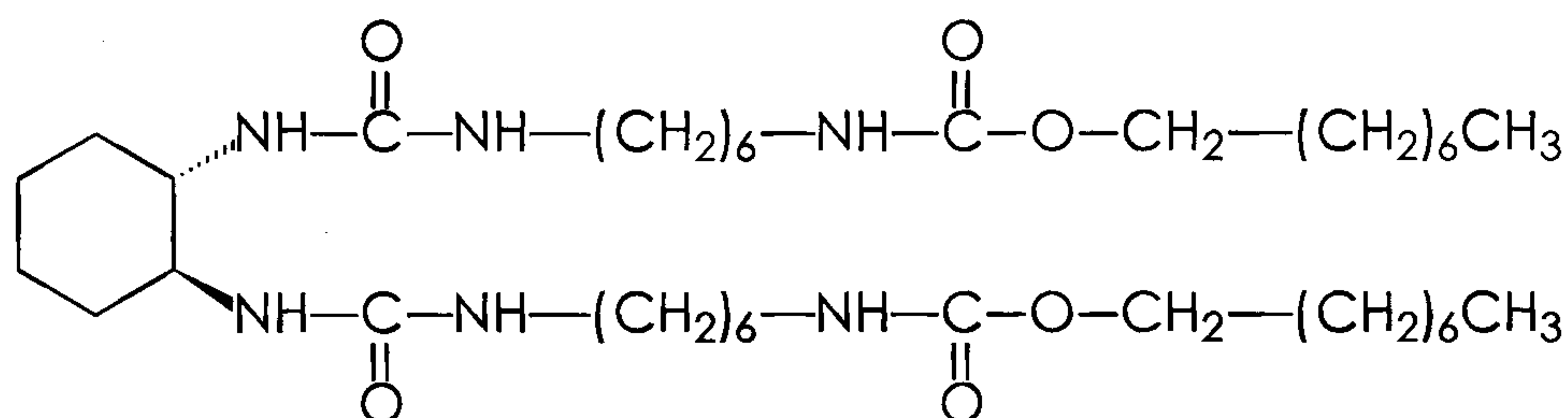
EXAMPLE II

[0085] Into a solution containing 1,6-diisocyanatohexane (4.04 grams, 24.0 mmol; obtained from Sigma-Aldrich Fine Chemicals) and anhydrous tetrahydrofuran (100 mL, obtained from Sigma-Aldrich Fine Chemicals) stirring at room temperature was added 1-octanol (3.13 grams, 24.0 mmol, obtained from Sigma-Aldrich Fine Chemicals) and dibutyltin dilaurate (0.15 grams, 0.24 mmol, obtained from Sigma-Aldrich Fine Chemicals) as the catalyst. The mixture was stirred and heated to an internal temperature of about 65°C. The progress of the reaction was monitored by ¹H-NMR spectroscopy for the consumption of 1-octanol starting material, indicated by the disappearance of the -CH₂OH multiplet, which appears at 3.6 ppm downfield of the intermediate isocyanate product whose signal is located at 3.35 ppm. The mixture was cooled to about 15°C internal temperature; thereafter, to this mixture was added dropwise a solution of *trans*-1,2-diaminocyclohexane (1.37 grams, 12 mmol; obtained as a racemic mixture of (1*R*,2*R*) and (1*S*,2*S*) stereoisomers from Sigma-Aldrich Fine Chemicals) dissolved in anhydrous tetrahydrofuran (10 mL). The mixture was stirred for about 60 minutes while warming up to room temperature, and thickened to form a gelatinous slurry. FTIR spectroscopic analysis of a reaction sample showed very little unreacted isocyanate (peak at 2180 cm⁻¹, sample prepared as a KBr pellet). Residual isocyanate was quenched by addition of 5 mL of methanol. A crystalline product was isolated from the slurry by first adding diethyl ether (20 mL) followed with stirring for approximately 30

minutes to ensure full precipitation out of the gel slurry. The solid was filtered by suction on a paper filter, rinsed with diethyl ether, and then dried in air to give 6.20 grams of off-white solid (77.5% yield). The product was believed to be of the formulae



and

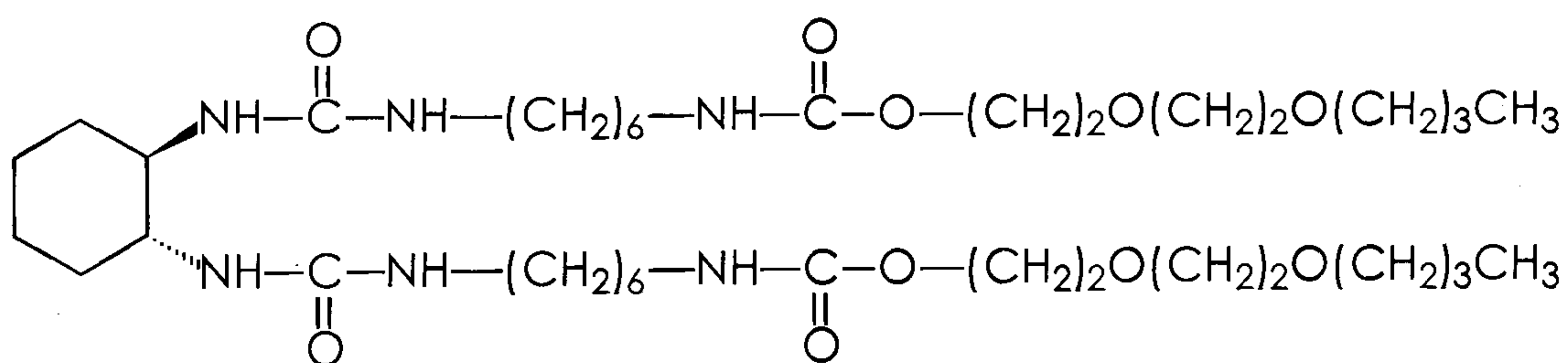


^1H -NMR spectroscopic analysis of the solid was performed in $\text{DMSO-}d_6$ (300 MHz) at high temperature (60°C) and indicated the above structure with the following assigned peaks: 0.90 ppm (multiplet, 3H integration, $-\text{OCH}_2(\text{CH}_2)_6\text{CH}_3$); 1.05-1.95 ppm (broad multiplets, 24H integration, 12 methylene protons from 2-ethylhexanol portion, 8 methylene protons from the 1,6-diisocyanatohexane portion, and 4 methylene protons from the cyclohexane ring portion); 2.95 ppm (narrow multiplet, 4H integration, $-\text{NH}(\text{C}=\text{O})\text{NHCH}_2(\text{CH}_2)_4\text{CH}_2\text{NH}(\text{C}=\text{O})\text{O}$); 3.35 ppm (doublet, 1H integration, tertiary methine proton adjacent to urea group on cyclohexane ring); 3.90 ppm (doublet of doublets, 2H integration, $\text{NH}(\text{C}=\text{O})\text{OCH}_2(\text{CH}_2)_6\text{CH}_3$); 5.70 ppm and 5.85 ppm (each a broad singlet, 1H integration, urea NH protons); 7.00 ppm (broad singlet, 1H

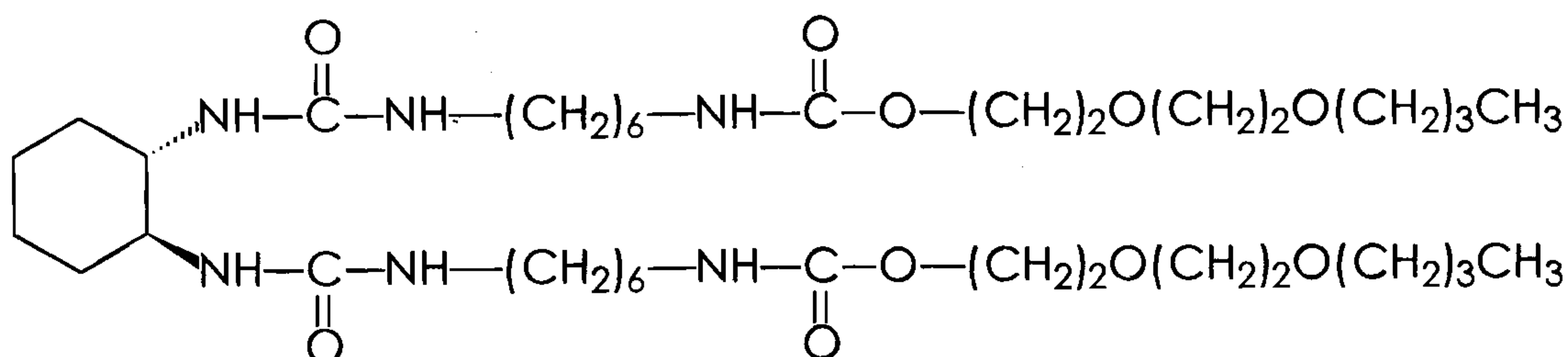
integration, urethane NH proton). Elemental analysis calculated for C: 64.19%, H: 10.49%, N: 11.82%; found for C: 64.46%, H: 10.63%, N: 10.69%.

EXAMPLE III

[0086] Into a solution containing 1,6-diisocyanatohexane (2.35 grams, 13.95 mmol; obtained from Sigma-Aldrich Fine Chemicals) and anhydrous hexane (100 mL, obtained from Sigma-Aldrich Fine Chemicals) stirring at room temperature was added diethyleneglycol butyl ether (2.27 grams, 14.0 mmol, obtained from Sigma-Aldrich Fine Chemicals), which was previously dried over calcium chloride granules, and dibutyltin dilaurate as catalyst (0.095 grams, 0.15 mmol, obtained from Sigma-Aldrich Fine Chemicals). The mixture was stirred and heated to an internal temperature of about 45°C. The progress of the reaction was monitored by ¹H-NMR spectroscopy for the consumption of the diethyleneglycol butyl ether starting material. The mixture was cooled to about 15°C internal temperature; thereafter, to this mixture was added dropwise a solution of *trans*-1,2-diaminocyclohexane (0.80 grams, 7.0 mmol; obtained as a racemic mixture of (1*R*,2*R*) and (1*S*,2*S*) stereoisomers from Sigma-Aldrich Fine Chemicals) dissolved in anhydrous hexane (20 mL). The mixture was stirred for about 30 minutes while warming up to room temperature, and FTIR spectroscopic analysis of a reaction sample indicated no unreacted isocyanate (peak at 2180 cm⁻¹, sample prepared as a KBr pellet). The crystalline product was isolated by vacuum filtration on filter paper, rinsed with hexane, and then dried in air to give 4.82 grams of a white powder (88.8% yield). The product was believed to be of the formulae



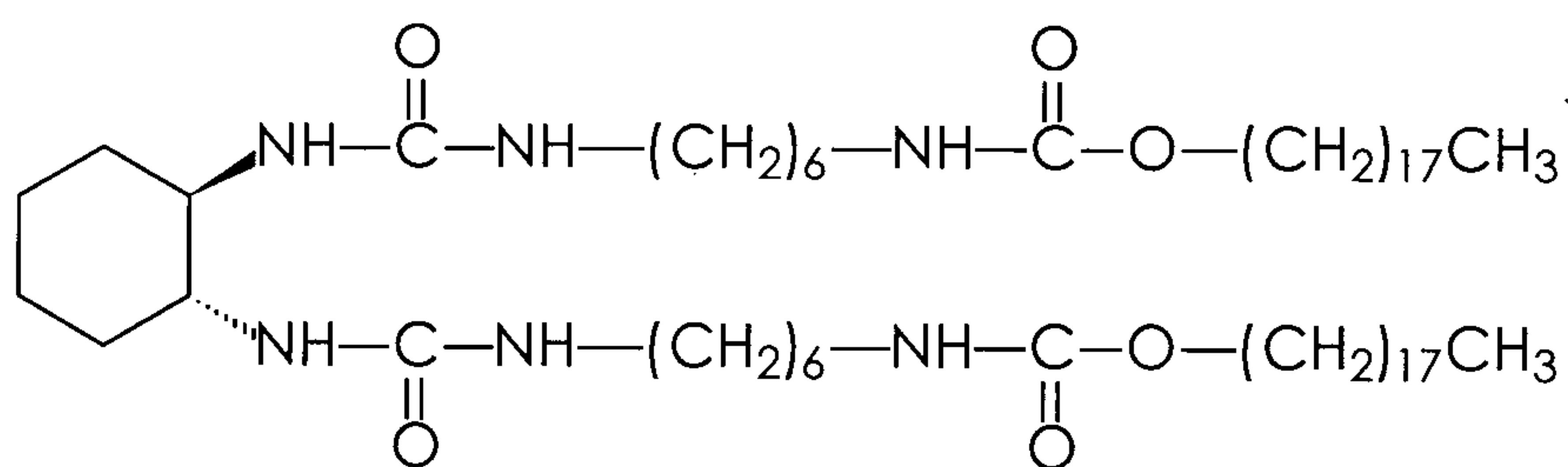
and



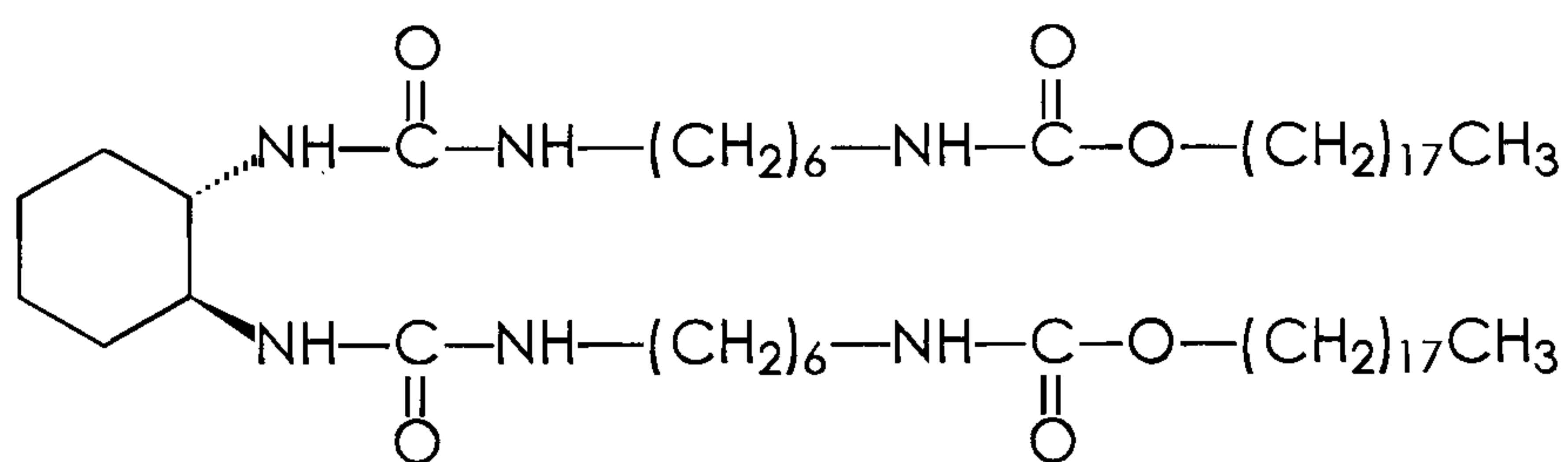
^1H -NMR spectroscopic analysis of the solid was performed in $\text{DMSO-}d_6$ (300 MHz) at 80°C and indicated the above structure with the following assigned peaks: 0.90 ppm (multiplet, 3H integration, $-\text{OCH}_2\text{CH}_2\text{OCH}_2\text{CH}_2\text{OCH}_2\text{CH}_2\text{CH}_2\text{CH}_3$); 1.05-1.95 ppm (broad multiplets, 16H integration, 4 methylene protons from butyl ether terminus, 8 methylene protons from the 1,6-diisocyanatohexane portion, and 4 methylene protons from the cyclohexane ring portion); 3.0 ppm (narrow multiplet, 5H integration, $-\text{NH}(\text{C}=\text{O})\text{NHCH}_2(\text{CH}_2)_4\text{CH}_2\text{NH}(\text{C}=\text{O})\text{O}$ and also tertiary methine proton adjacent to urea group on cyclohexane ring); 3.40-3.70 ppm (multiplets, 8H integration, $\text{NH}(\text{C}=\text{O})\text{OCH}_2\text{CH}_2\text{OCH}_2\text{CH}_2\text{OCH}_2\text{CH}_2\text{CH}_2\text{CH}_3$); 4.10 ppm (singlet, 2H integration, $\text{NH}(\text{C}=\text{O})\text{OCH}_2\text{CH}_2\text{OCH}_2\text{CH}_2\text{OCH}_2\text{CH}_2\text{CH}_2\text{CH}_3$); 5.60 ppm and 5.70 ppm (each a broad singlet, 1H integration, urea NH protons); 6.75 ppm (broad singlet, 1H integration, urethane NH proton). Elemental analysis calculated for C: 58.83%, H: 9.54%, N: 10.83%; found for C: 58.81%, H: 9.58%, N: 12.17%.

EXAMPLE IV

[0087] Into a solution containing 1,6-diisocyanatohexane (1.86 grams, 11.09 mmol; obtained from Aldrich Fine Chemicals) and hexane (250 milliliters) stirring at room temperature was added a solution of 1-octadecanol (3.0 grams, 11.09 mmol; obtained from Aldrich Fine Chemicals) in anhydrous tetrahydrofuran (50 milliliters, obtained from Aldrich Fine Chemicals) and dibutyl tin dilaurate (0.07 gram, 1 mol%; obtained from Aldrich Fine Chemicals) as catalyst. The resulting solution was heated to 60°C for 1 hour, during which a white precipitate was formed. The mixture was cooled to room temperature (20 to 25°C). A solution of *trans*-1,2-diaminocyclohexane (0.69 gram, 6.09 mmol; obtained from Aldrich Fine Chemicals) in hexane (50 milliliters) was then slowly added to the reaction mixture through an addition funnel. The mixture was stirred vigorously at room temperature for 2 hours, during which a more viscous white precipitate was formed. An IR spectrum indicated the presence of trace amounts of isocyanate. More *trans*-1,2-diaminocyclohexane was added (0.1 gram, 0.87 mmol) and stirred for an additional 30 minutes, during which all of the isocyanate was consumed as shown by IR. The product was isolated by vacuum filtration on a paper filter, rinsed with hexane, and dried under vacuum at 60°C for 2 hours to give 5 grams of an off-white powder (91 percent yield). The product was believed to be of the formulae



and

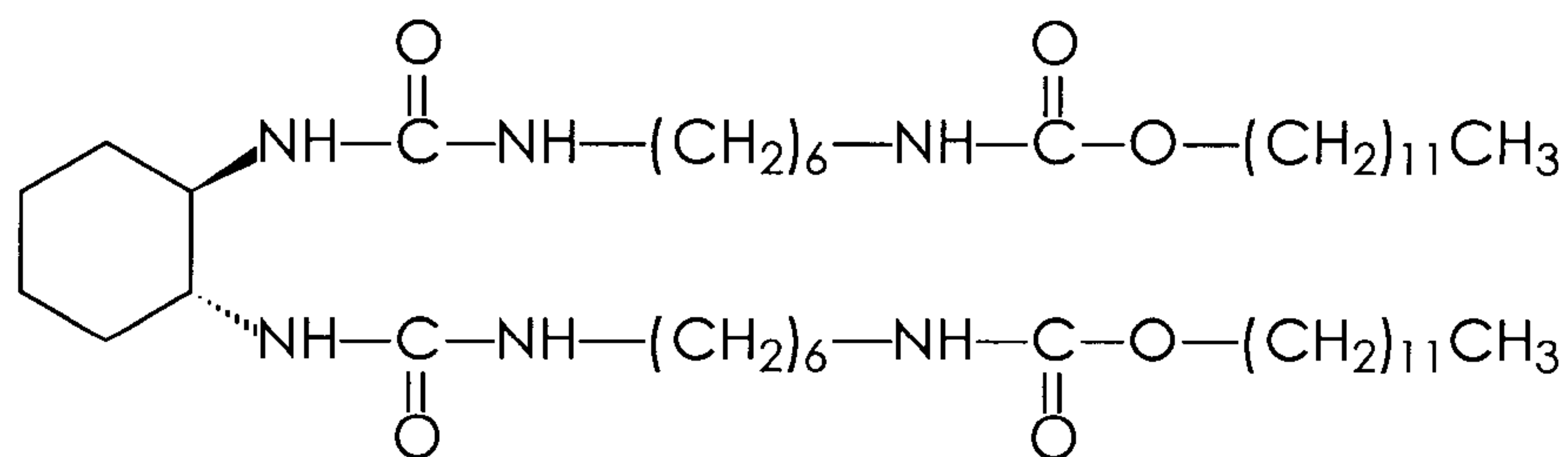


IR and ^1H NMR analysis of the product indicated that product was of high purity. IR (KBr) 3318, 2921, 2849, 1684, 1634, 1539, 1276 cm^{-1} ; ^1H NMR ($\text{DMSO}-d_6$, at 100°C); 0.89 ppm (triplet, 6H integration, $\text{CH}_3(\text{CH}_2)_{16}\text{CH}_2\text{CONH}-$), 1.01-1.82 ppm (multiplet, 86H, 6 methylene protons on cyclohexyl ring, $-\text{NHCONHCH}_2(\text{CH}_2)_4\text{CH}_2\text{NHCO}_2-$, $\text{CH}_3(\text{CH}_2)_{16}\text{CH}_2\text{CONH}-$), 1.83 ppm (broad doublet, 0.4 H, CH on the cyclohexyl rings adjacent to carbons bonded to the NH urea), 1.89 ppm (broad doublet, 1.6 H, tertiary methine proton adjacent to urea group on cyclohexane ring), 2.25 ppm (doublet of triplet, 0.2 H $-\text{NHCONHCH}_2(\text{CH}_2)_4\text{CH}_2\text{NHCO}_2-$), 2.8 ppm (doublet of doublets, 0.3 H, CH on cyclohexyl ring next to NH urea), 3.00 ppm (quartet, 7.8 H, $-\text{NHCONHCH}_2(\text{CH}_2)_4\text{CH}_2\text{NHCO}_2-$), 3.18 ppm (multiplet, 1.7 H, CH on cyclohexyl ring next to NH urea), 4.02 ppm (triplet, 4 H, $-\text{NHCO}_2\text{CH}_2(\text{CH}_2)_{16}\text{CH}_3$), 5.37 (broad triplet, 0.7 H, $-\text{NHCONH}-$), 5.71 ppm (broad doublet, 3.3 H, $-\text{NHCONH}-$), 6.48 ppm (broad singlet, 2 H, $-\text{NHCO}_2-$), melting point by DSC 119.5°C .

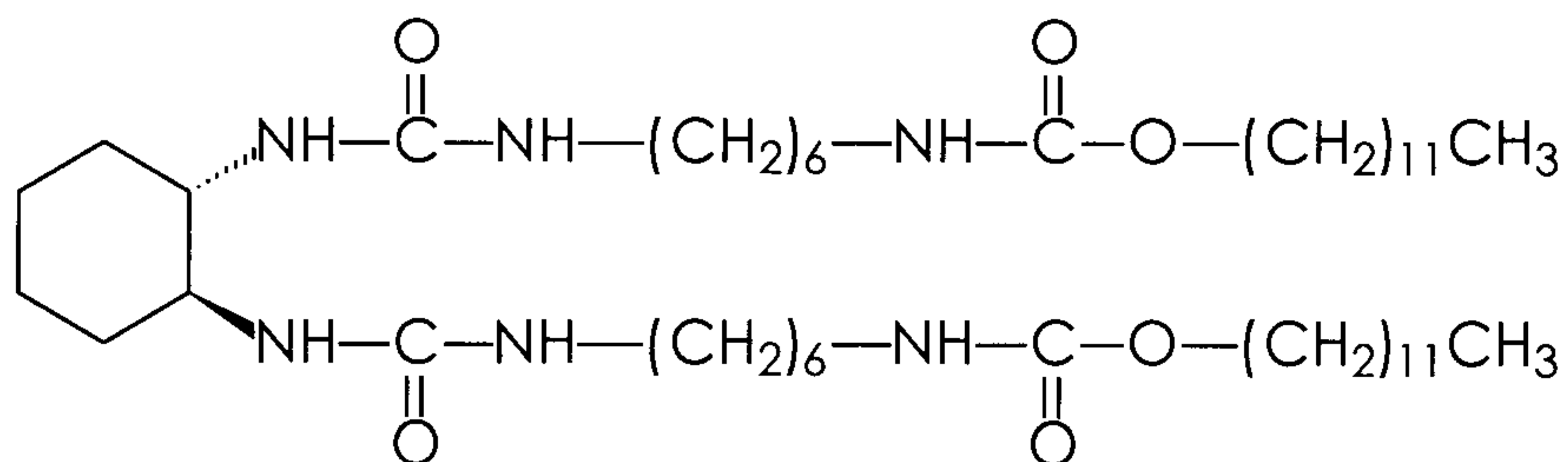
EXAMPLE V

[0088] Into a solution containing 1,6-diisocyanatohexane (2.07 grams, 12.34 mmol; obtained from Sigma-Aldrich Fine Chemicals) and hexane (250 milliliters) with stirring at room temperature was added a solution of 1-dodecanol (2.30 grams, 12.34 mmol; obtained from Sigma-Aldrich Fine Chemicals) in anhydrous tetrahydrofuran (50 milliliters,

obtained from Aldrich Fine Chemicals) and dibutyl tin dilaurate (0.08 gram, 1 mol%; obtained from Sigma-Aldrich Chemical Company) as catalyst. The resulting solution was heated to 45°C for 1 hour, during which a white precipitate was formed. The mixture was cooled to room temperature (20 to 25°C). A solution of *trans*-1,2-diaminocyclohexane (0.775 gram, 6.79 mmol; obtained as a racemic mixture of (1*R*,2*R*) and (1*S*,2*S*) stereoisomers from Sigma-Aldrich Fine Chemicals) in hexane (50 milliliters) was then slowly added to the reaction mixture through an addition funnel. The mixture was stirred at room temperature for 1 hour, during which a more viscous white precipitate was formed. An IR spectrum indicated the presence of trace amount of isocyanate. More *trans*-1,2-diaminocyclohexane was added (0.07 gram, 0.6 mmol) and stirred for an additional 30 minutes, during which all the isocyanate was consumed as shown by IR. The product was isolated by vacuum filtration on a paper filter, rinsed with hexane, and dried under vacuum at 60°C for 2 hours to give 4.6 grams of product as an off-white powder (90 percent yield). The product was believed to be of the formulae



and

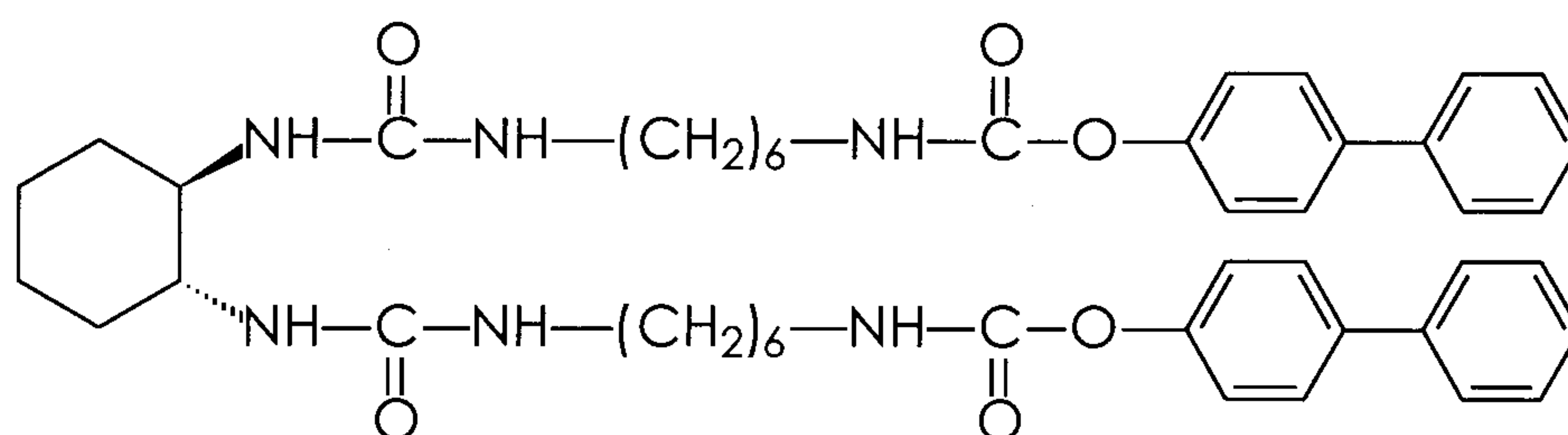


^1H NMR analysis of the product indicated that product was of high purity. IR (KBr) 3320, 2919, 2851, 1684, 1635, 1538, 1265 cm^{-1} ; ^1H NMR ($\text{DMSO}-d_6$, at 80°C); 0.89 ppm (triplet, 6 H, $\text{CH}_3(\text{CH}_2)_{16}\text{CH}_2\text{CONH-}$), 1.01-1.80 ppm (multiplet, 62 H, 6 methylene protons on cyclohexyl ring, $\text{-NHCONHCH}_2(\text{CH}_2)_4\text{CH}_2\text{NHCO}_2\text{-}$, $\text{CH}_3(\text{CH}_2)_{16}\text{CH}_2\text{CONH-}$), 1.87 ppm (broad doublet, 2 H, one of the CH_2 hydrogens on the cyclohexyl ring adjacent to carbons bonded to the NH urea), 2.98 ppm (quartet, 8 H, $\text{-NHCONHCH}_2(\text{CH}_2)_4\text{CH}_2\text{NHCO}_2\text{-}$), 3.24 ppm (multiplet, 2 H, CH on cyclohexyl ring next to NH urea), 4.93 ppm (triplet, 4 H, $\text{-NHCO}_2\text{CH}_2(\text{CH}_2)_{16}\text{CH}_3$), 5.56 ppm (broad singlet, 2 H, -NHCONH-), 5.60 ppm (broad multiplet, 2 H, -NHCONH-), 6.60 ppm (broad singlet, 2 H, $\text{-NHCO}_2\text{-}$), melting point by DSC 111.7°C .

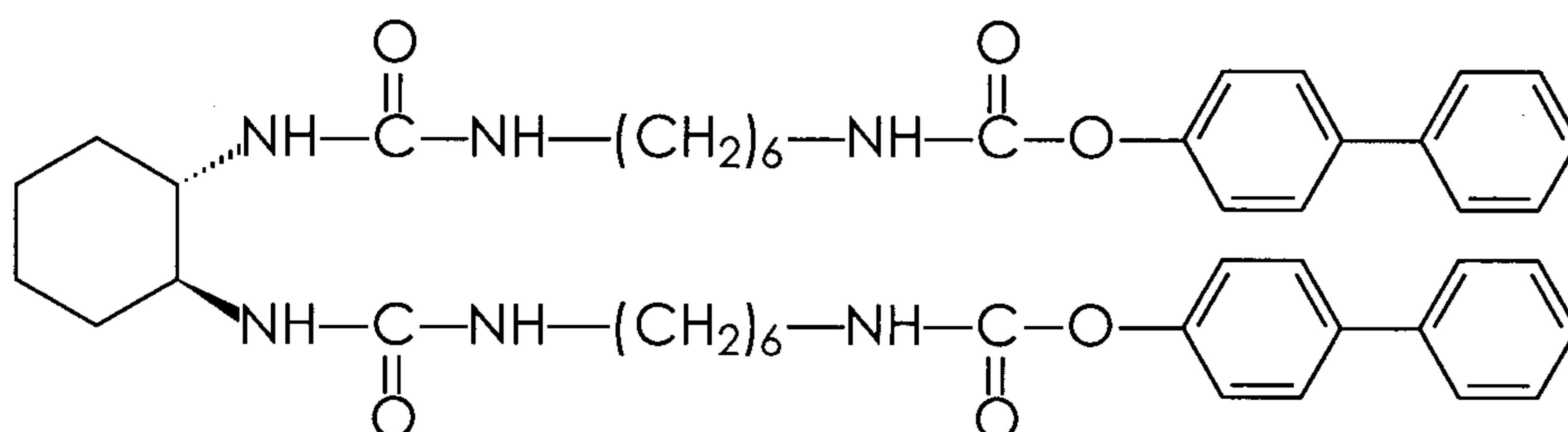
EXAMPLE VI

[0100] The process of Example I is repeated except that 4-phenylphenol is used instead of 2-ethylhexanol. A solution of 4-phenylphenol (4.08 grams, 24.0 mmol; available from Sigma-Aldrich Fine Chemicals, Milwaukee, WI) dissolved in anhydrous tetrahydrofuran (100 mL, available from Sigma-Aldrich Fine Chemicals) is added into a second solution containing 1,6-diisocyanatohexane (4.04 grams, 24.0 mmol; available from Sigma-Aldrich Fine Chemicals) dissolved in anhydrous tetrahydrofuran (100 mL) stirring at room temperature. Dibutyltin dilaurate (0.38 grams, 0.6 mmol, available from Sigma-Aldrich Fine Chemicals) is added as the catalyst, and the mixture is heated to an internal temperature of about 80°C for 30 to 60 minutes. The mixture is then cooled to about 20°C internal temperature, after which is then added dropwise to the mixture a solution of *trans*-1,2-diaminocyclohexane (1.37 grams, 12 mmol; available as a racemic

mixture of (1*R*,2*R*) and (1*S*,2*S*) stereoisomers from Sigma-Aldrich Fine Chemicals) dissolved in anhydrous tetrahydrofuran (10 mL). The mixture is stirred for about 60 minutes while warming up to room temperature. Residual isocyanate is quenched by addition of 5 mL of methanol. It is believed that a crystalline product can be precipitated from the mixture by the addition of hexane (40 mL) followed with stirring for approximately 30 minutes. The solid can be recovered by vacuum filtration, rinsed with hexane and diethyl ether (about 10 mL each), and then dried in air. It is believed that compounds of the formulae



and

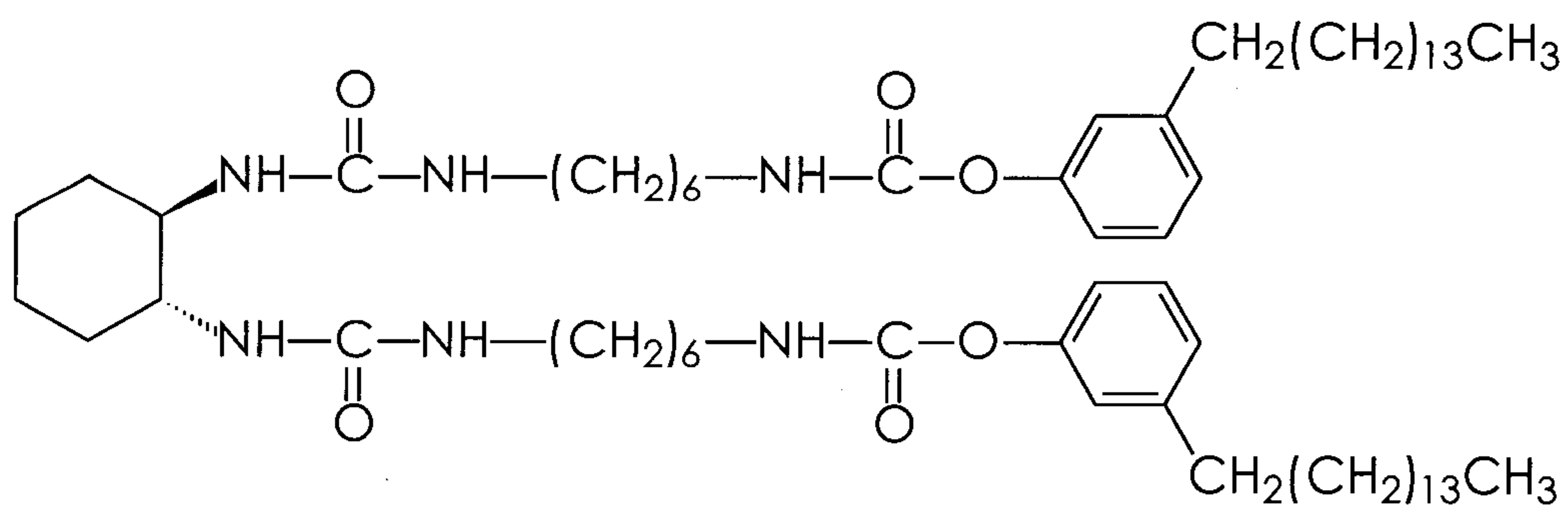


will be obtained.

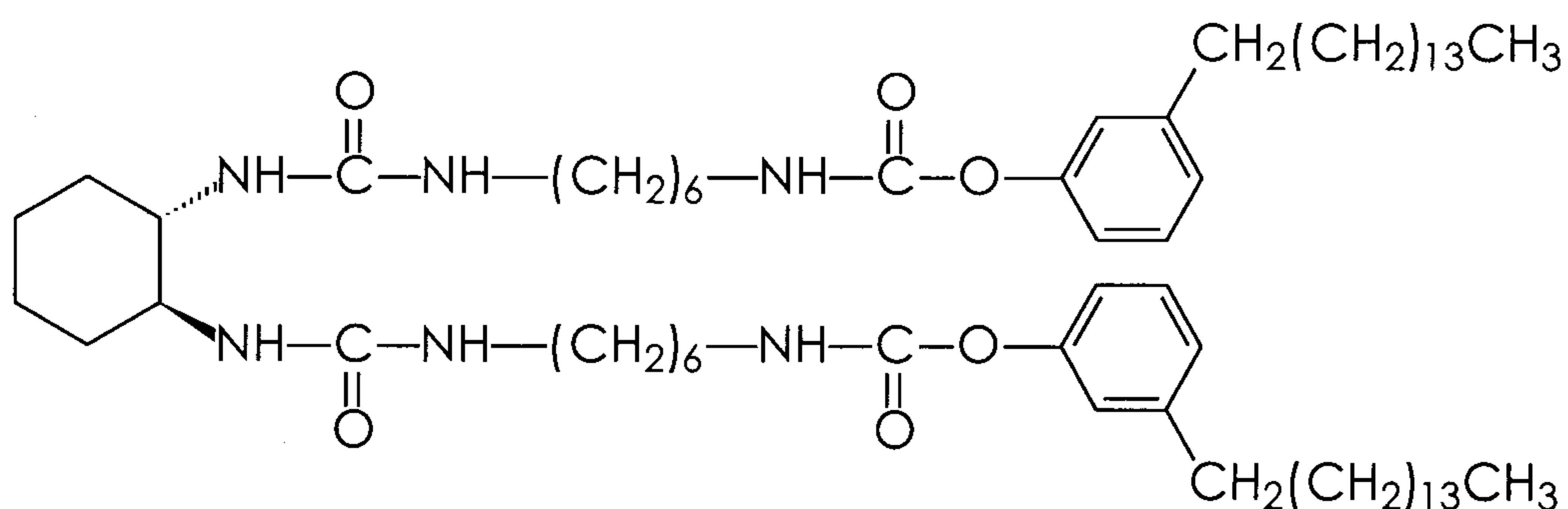
EXAMPLE VII

[0101] The process of Example VI is repeated except that 3-pentadecylphenol is used in place of 4-phenylphenol. A solution of 3-pentadecylphenol (7.31 grams, 24.0 mmol; available from Sigma-Aldrich Fine Chemicals) dissolved in anhydrous tetrahydrofuran (100 mL, available from Sigma-Aldrich Fine Chemicals) is added into a second

solution containing 1,6-diisocyanatohexane (4.04 grams, 24.0 mmol; available from Sigma-Aldrich Fine Chemicals) dissolved in anhydrous tetrahydrofuran (100 mL) stirring at room temperature. Dibutyltin dilaurate (0.38 grams, 0.6 mmol, available from Sigma-Aldrich Fine Chemicals) is added as the catalyst, and the mixture is heated to an internal temperature of about 80°C for 30 to 60 minutes. The mixture is then cooled to about 20°C internal temperature, after which is added dropwise to the mixture a solution of *trans*-1,2-diaminocyclohexane (1.37 grams, 12 mmol; available as a racemic mixture of (1*R*,2*R*) and (1*S*,2*S*) stereoisomers from Sigma-Aldrich Fine Chemicals) dissolved in anhydrous tetrahydrofuran (10 mL). The mixture is stirred for about 60 minutes while warming up to room temperature. Residual isocyanate is quenched by addition of 5 mL of methanol. It is believed that a crystalline product can be precipitated from the mixture by the addition of hexane (40 mL) followed with stirring for approximately 30 minutes. The solid can be recovered by vacuum filtration, rinsed with hexane and diethyl ether (about 10 mL each), and then dried in air. It is believed that compounds of the formulae



and

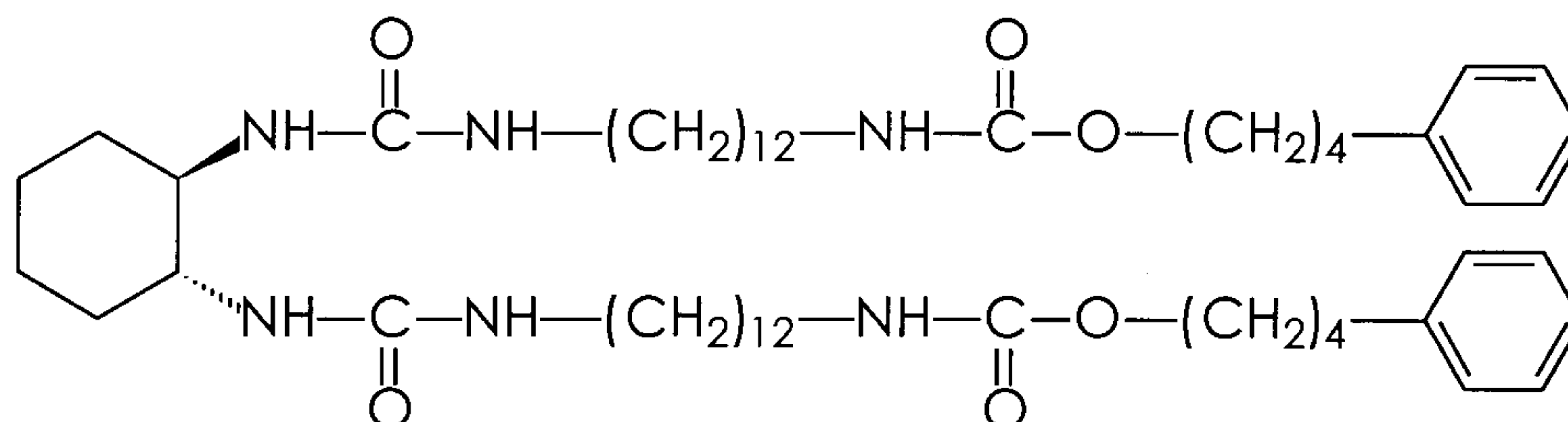


will be obtained.

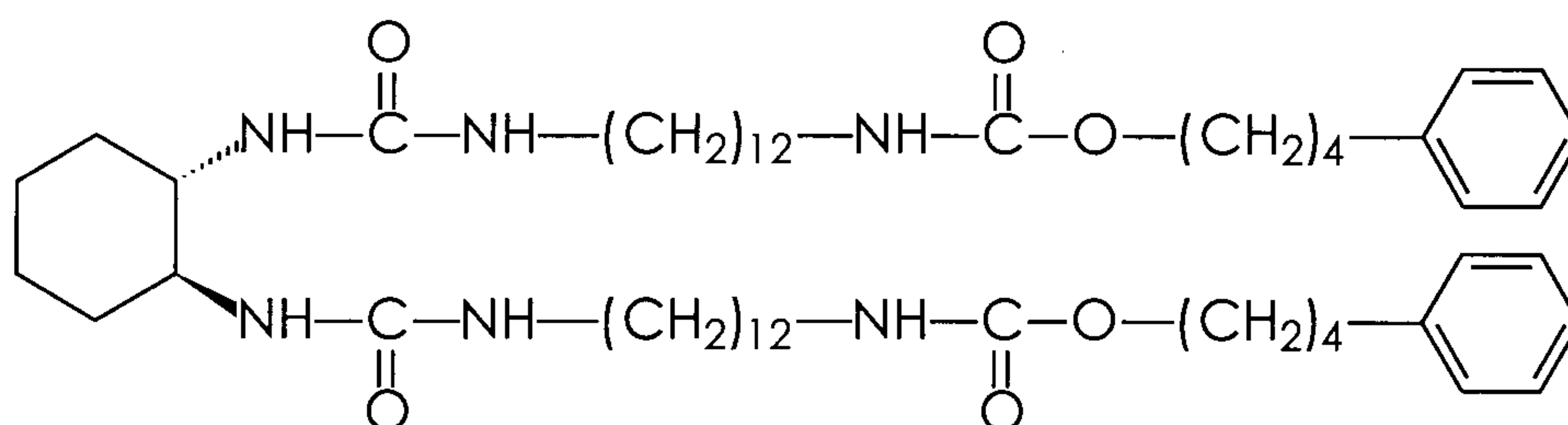
EXAMPLE VIII

[0102] The process of Example VI is repeated except that 4-phenyl-1-butanol is used in place of 4-phenylphenol. A solution of 4-phenyl-1-butanol (3.60 grams, 24.0 mmol; available from Sigma-Aldrich Fine Chemicals) dissolved in anhydrous tetrahydrofuran (100 mL, available from Sigma-Aldrich Fine Chemicals) is added into a second solution containing 1,12-diisocyanatododecane (6.06 grams, 24.0 mmol; available from Sigma-Aldrich Fine Chemicals) dissolved in anhydrous tetrahydrofuran (100 mL) stirring at room temperature. Dibutyltin dilaurate (0.38 grams, 0.6 mmol, available from Sigma-Aldrich Fine Chemicals) is added as the catalyst, and the mixture is heated to an internal temperature of about 80°C for 30 to 60 minutes. The mixture is then cooled to about 20°C internal temperature, after which is added dropwise to the mixture a solution of *trans*-1,2-diaminocyclohexane (1.37 grams, 12 mmol; available as a racemic mixture of (1*R*,2*R*) and (1*S*,2*S*) stereoisomers from Sigma-Aldrich Fine Chemicals) dissolved in anhydrous tetrahydrofuran (10 mL). The mixture is stirred for about 60 minutes while warming up to room temperature. Residual isocyanate is quenched by addition of 5 mL of methanol. It is believed that a crystalline product can be precipitated from the mixture by the

addition of hexane (40 mL) followed with stirring for approximately 30 minutes. The solid can be recovered by vacuum filtration, rinsed with hexane and diethyl ether (about 10 mL each), and then dried in air. It is believed that compounds of the formulae



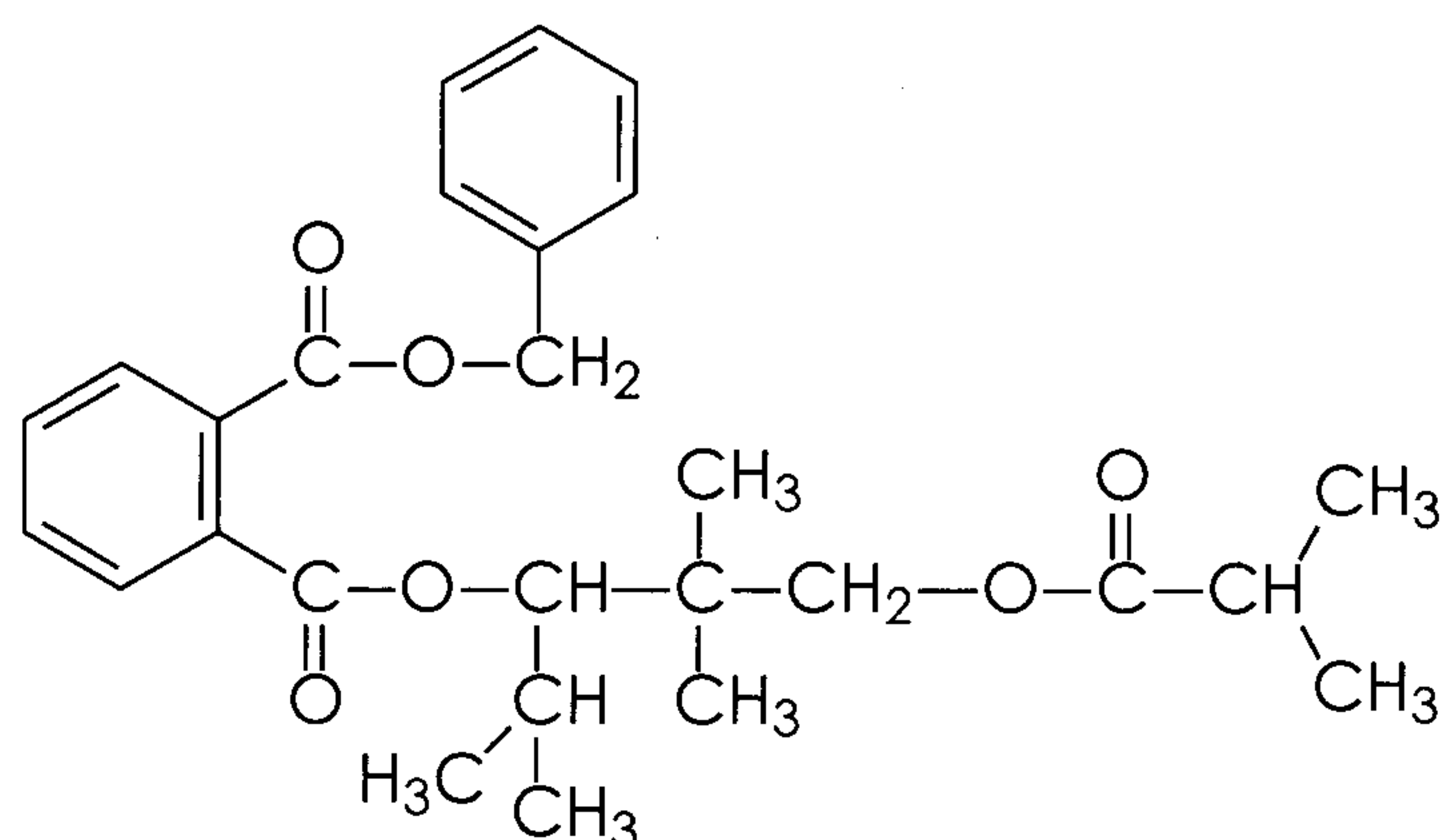
and



will be obtained.

INK EXAMPLE 1

[0103] A cyan ink composition was prepared in a beaker by adding (1) 21.6 grams (61.03 parts by weight) of polyethylene wax (PE 500, obtained from Baker Petrolite, Tulsa, OK, a polyethylene homopolymer with an average chain length of C-36), (2) 9.76 grams (27.41 parts by weight) of a linear primary long chain alcohol (UNILIN® 425, obtained from Baker Petrolite, Tulsa, OK, with an average chain length of C-30), (3) 1.27 grams (3.59 parts by weight) of a glycerol ester of hydrogenated (rosin) acid (KE-100, obtained from Arakawa Chemical Industries, Ltd, Osaka, Japan), (4) 0.91 gram (2.57 parts by weight) of an alkylbenzyl phthalate of the formula



(SANTICIZER® 278, obtained from Ferro Corporation, Bridgeport, NJ), (5) 0.03 gram (0.08 part by weight) of NAUGUARD® 445 antioxidant (obtained from Uniroyal Chemical Co., Middlebury, CT), and (6) 1.04 grams (2.83 parts by weight) of the *trans*-1,2-cyclohexane bis(urea-urethane) prepared in Example IV. The materials were melted together at a temperature of about 135°C in a reaction block (from H + P Labortechnik GmbH, München) controlled with a telemode 40CT, and stirred for 2 hours at 500 rpm. To this mixture was then added (7) 0.89 gram (2.49 parts by weight) of the cyan colorant disclosed in Example V of U.S. Patent 6,472,523. The ink was stirred for 2 additional hours and then cooled to room temperature. The cyan ink thus prepared exhibited a viscosity of 7.2 centipoise as measured by an RFS3 Rheometrics parallel-plate viscometer at 110°C.

INK EXAMPLE 2

[0104] A cyan ink was prepared as described in Ink Example 1 except that 3.5 parts by weight of the *trans*-1,2-cyclohexane bis(urea-urethane) was used. The cyan ink thus prepared exhibited a viscosity of 26 centipoise as measured by an RFS3 Rheometrics parallel-plate viscometer at 120°C.

COMPARATIVE EXAMPLE A

[0105] A cyan ink was prepared as described in Ink Example 1 except that no *trans*-1,2-cyclohexane bis(urea-urethane) was present. Relative amounts of the ingredients in this ink, expressed in parts by weight of the ink, is indicated in the table below:

Component	Ink 1	Ink 2	Ink A
POLYWAX 500	61.03	60.60	62.81
UNILIN® 425	27.41	27.23	28.21
KE-100	3.59	3.57	3.69
SANTICIZER® 278	2.57	2.55	2.65
urea-urethane	2.83	3.50	0
NAUGUARD® 445	0.08	0.08	0.09
cyan colorant	2.49	2.47	2.56
total	100	100	100

Rheology of the inks was measured using a controlled strain rheometer, RFS3 obtained from Rheometrics™ Scientific, in a conventional parallel plate configuration. The table below shows the tan-delta (ratio of loss modulus or viscous modulus, G'' , to storage modulus or elastic modulus, G') of the two inks in a region above their melting point (melting point of the inks is around 90°C as determined by the rheometer). Ink 1 and Ink 2 have a lower tan-delta in this region, indicating an increase in G' (elastic modulus), and a tan-delta of less than one at 95°C, indicating that G' is much higher than G'' , which suggests that the material is elastic in that region. Comparative Ink A, on the other hand, has a high tan-delta in the same region, suggesting that very low elasticity

compared to Inks 1 and 2. These data demonstrate that the *trans*-1,2-cyclohexane bis(urea-urethane) significantly affects the rheological properties of the solid inks containing them. The increase of elasticity of the ink above its melting point is expected to translate into a more robust image.

Temperature (°C)	Ink 1 tan-delta	Ink 2 tan-delta	Ink A tan-delta
105	1.84	1.41	14
100	1.28	1.00	17
95	0.76	0.91	15

INK EXAMPLE 3

[0106] An ink is prepared as described in Ink Example 1 except that the cyan dye is replaced with 3 parts by weight of the yellow colorant disclosed in Example I of U.S. Patent 6,713,614. A yellow phase change ink is thus prepared.

INK EXAMPLE 4

[0107] An ink is prepared as described in Ink Example 1 except that the *trans*-1,2-cyclohexane bis(urea-urethane) prepared in Example IV is replaced by 5 parts by weight of the *trans*-1,2-cyclohexane bis(urea-urethane) prepared in Example V.

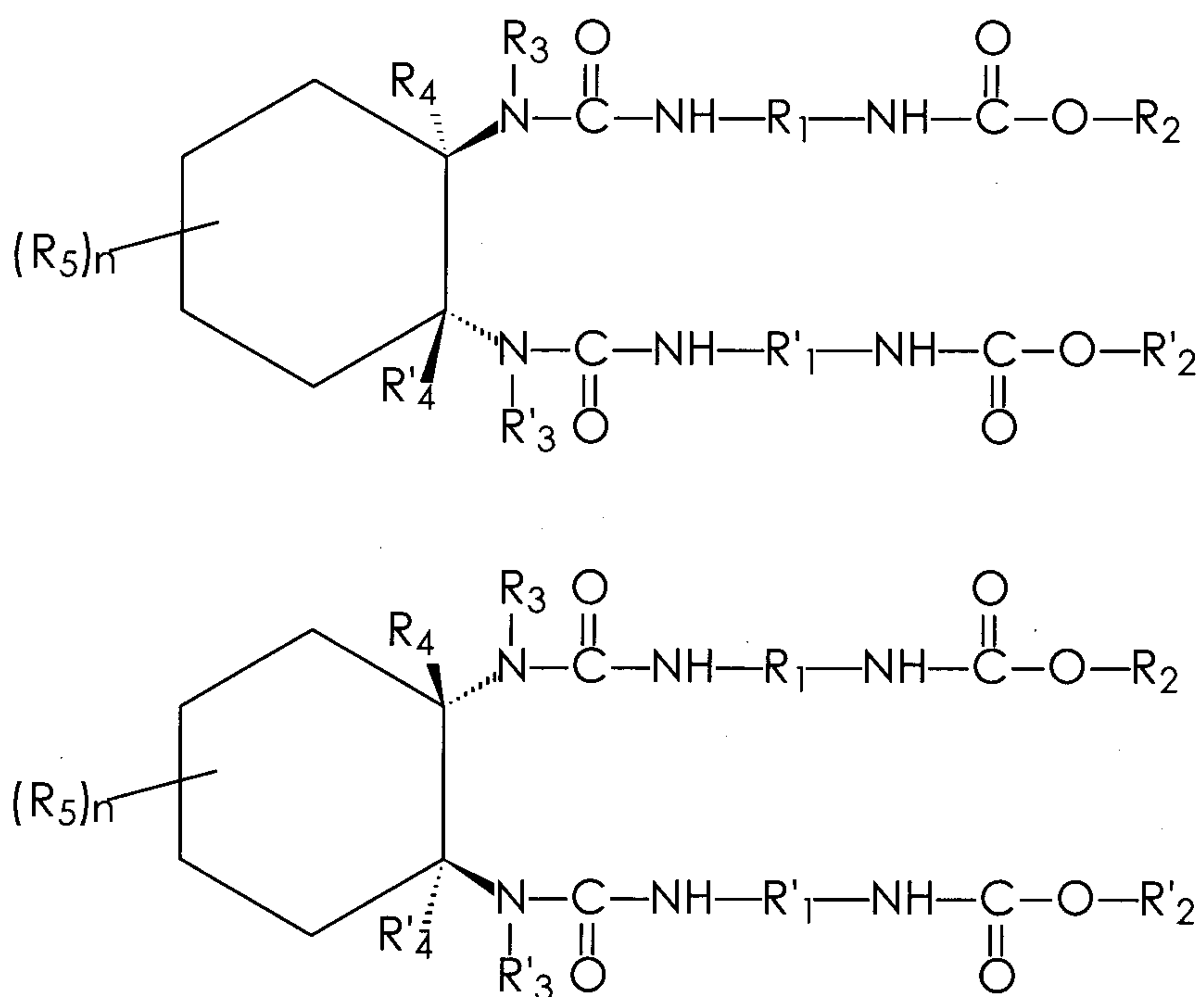
[0108] Other embodiments and modifications of the present invention may occur to those of ordinary skill in the art subsequent to a

review of the information presented herein; these embodiments and modifications, as well as equivalents thereof, are also included within the scope of this invention.

[0109] The recited order of processing elements or sequences, or the use of numbers, letters, or other designations therefor, is not intended to limit a claimed process to any order except as specified in the claim itself.

WHAT IS CLAIMED IS:

1. A phase change ink composition comprising a phase change ink carrier and a *trans*-1,2-cyclohexane bis[urea-urethane] compound of the formula



or mixtures thereof, wherein R₁ and R'₁ each, independently of the other, is an alkylene group, an arylene group, an arylalkylene group, or an alkylarylene group, R₂ and R'₂ each, independently of the other, is an alkyl group, an aryl group, an arylalkyl group, or an alkylaryl group, R₃ and R'₃ each, independently of the other, is a hydrogen atom or an alkyl group, R₄ and R'₄ each, independently of the other, is a hydrogen atom, a fluorine atom, an alkyl group, or a phenyl group, n is an integer of 0, 1, 2, 3, or 4, and R₅ is an alkyl group, an aryl group, an arylalkyl group, an alkylaryl group, or a substituent other than an alkyl, aryl, arylalkyl, or alkylaryl group.

2. An ink according to claim 1 wherein at least one of R_1 and R'_1 is an alkylene group.
3. An ink according to claim 2 wherein at least one of R_1 and R'_1 is a linear alkylene group.
4. An ink according to claim 2 wherein at least one of R_1 and R'_1 is a branched alkylene group.
5. An ink according to claim 2 wherein at least one of R_1 and R'_1 is a cyclic alkylene group.
6. An ink according to claim 2 wherein at least one of R_1 and R'_1 is a substituted alkylene group.
7. An ink according to claim 2 wherein at least one of R_1 and R'_1 is an unsubstituted alkylene group.
8. An ink according to claim 2 wherein at least one of R_1 and R'_1 is an alkylene group having hetero atoms therein.
9. An ink according to claim 2 wherein at least one of R_1 and R'_1 is an alkylene group having no hetero atoms therein.
10. An ink according to claim 2 wherein at least one of R_1 and R'_1 is an alkylene group having at least 2 carbon atoms.
11. An ink according to claim 2 wherein at least one of R_1 and R'_1 is an alkylene group having at least about 6 carbon atoms.

12. An ink according to claim 2 wherein at least one of R_1 and R'_1 is an alkylene group having no more than about 12 carbon atoms.

13. An ink according to claim 1 wherein at least one of R_1 and R'_1 is an arylene, arylalkylene, or alkylarylene group.

14. An ink according to claim 13 wherein at least one of R_1 and R'_1 is a substituted arylene, arylalkylene, or alkylarylene group.

15. An ink according to claim 13 wherein at least one of R_1 and R'_1 is an unsubstituted arylene, arylalkylene, or alkylarylene group.

16. An ink according to claim 13 wherein at least one of R_1 and R'_1 is an arylene, arylalkylene, or alkylarylene group having hetero atoms therein.

17. An ink according to claim 13 wherein at least one of R_1 and R'_1 is an arylene, arylalkylene, or alkylarylene group having no hetero atoms therein.

18. An ink according to claim 1 wherein R_1 and R'_1 are the same as each other.

19. An ink according to claim 1 wherein R_1 and R'_1 are different from each other.

20. An ink according to claim 1 wherein at least one of R_2 and R'_2 is an alkyl group.

21. An ink according to claim 20 wherein at least one of R_2 and R'_2 is a linear alkyl group.

22. An ink according to claim 20 wherein at least one of R_2 and R'_2 is a branched alkyl group.

23. An ink according to claim 20 wherein at least one of R_2 and R'_2 is a cyclic alkyl group.

24. An ink according to claim 20 wherein at least one of R_2 and R'_2 is a substituted alkyl group.

25. An ink according to claim 20 wherein at least one of R_2 and R'_2 is an unsubstituted alkyl group.

26. An ink according to claim 20 wherein at least one of R_2 and R'_2 is an alkyl group having hetero atoms therein.

27. An ink according to claim 20 wherein at least one of R_2 and R'_2 is an alkyl group having no hetero atoms therein.

28. An ink according to claim 20 wherein at least one of R_2 and R'_2 is an alkyl group having at least about 6 carbon atoms.

29. An ink according to claim 20 wherein at least one of R_2 and R'_2 is an alkyl group having at least about 12 carbon atoms.

30. An ink according to claim 20 wherein at least one of R_2 and R'_2 is an alkyl group having no more than about 18 carbon atoms.

31. An ink according to claim 1 wherein at least one of R_2 and R'_2 is an aryl, arylalkyl, or alkylaryl group.

32. An ink according to claim 31 wherein at least one of R_2 and R'_2 is a substituted aryl, arylalkyl, or alkylaryl group.

33. An ink according to claim 31 wherein at least one of R_2 and R'_2 is an unsubstituted aryl, arylalkyl, or alkylaryl group.

34. An ink according to claim 31 wherein at least one of R_2 and R'_2 is an aryl, arylalkyl, or alkylaryl group having hetero atoms therein.

35. An ink according to claim 31 wherein at least one of R_2 and R'_2 is an aryl, arylalkyl, or alkylaryl group having no hetero atoms therein.

36. An ink according to claim 1 wherein R_2 and R'_2 are the same as each other.

37. An ink according to claim 1 wherein R_2 and R'_2 are different from each other.

38. An ink according to claim 1 wherein R_1 and R'_1 are the same as each other and wherein R_2 and R'_2 are the same as each other.

39. An ink according to claim 1 wherein R_3 and R'_3 are each hydrogen atoms.

40. An ink according to claim 1 wherein at least one of R_3 and R'_3 is an alkyl group with from 1 to about 3 carbon atoms.

41. An ink according to claim 1 wherein R_3 and R'_3 are each hydrogen atoms, wherein R_1 and R'_1 are the same as each other, and wherein R_2 and R'_2 are the same as each other.

42. An ink according to claim 1 wherein R_4 and R'_4 are each hydrogen atoms.

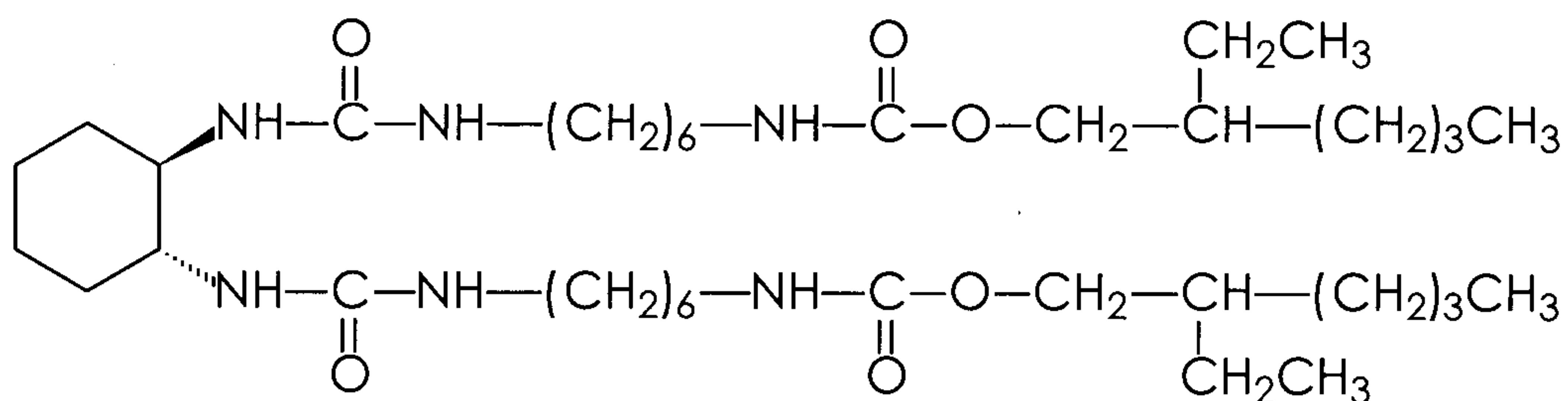
43. An ink according to claim 1 wherein R_4 and R'_4 are each fluorine atoms.

44. An ink according to claim 1 wherein at least one of R_4 and R'_4 is an alkyl group.

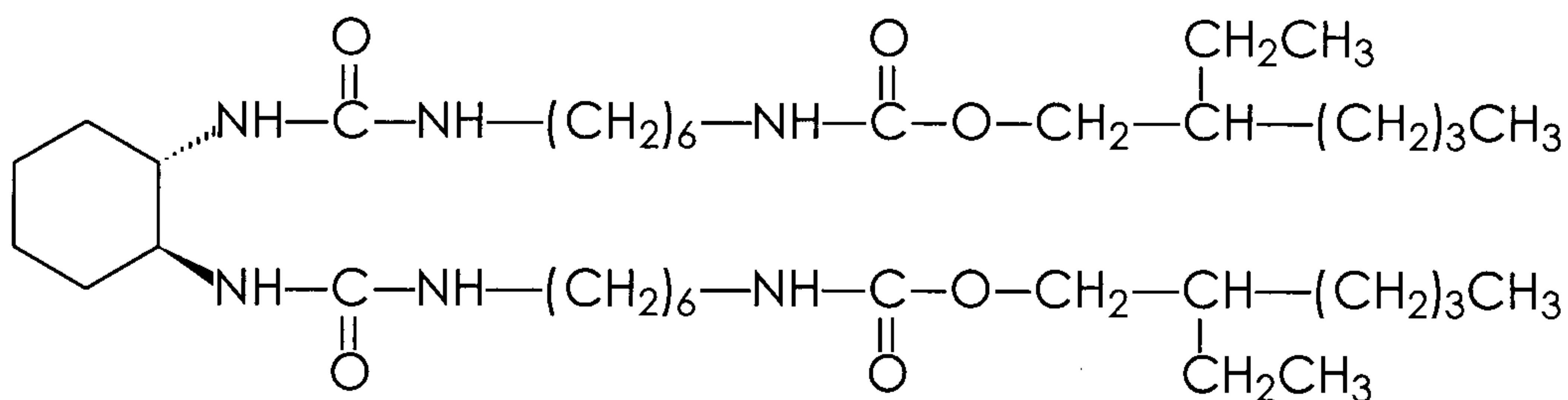
45. An ink according to claim 1 wherein R_1 and R'_1 are the same as each other, R_2 and R'_2 are the same as each other, R_3 and R'_3 are each hydrogen atoms, R_4 and R'_4 are the same as each other, and R_4 and R'_4 are hydrogen atoms or fluorine atoms.

46. An ink according to claim 1 wherein R_1 and R'_1 are the same as each other, R_2 and R'_2 are the same as each other, R_3 and R'_3 are each hydrogen atoms, R_4 and R'_4 are the same as each other, R_4 and R'_4 are hydrogen atoms or fluorine atoms, and n is 0.

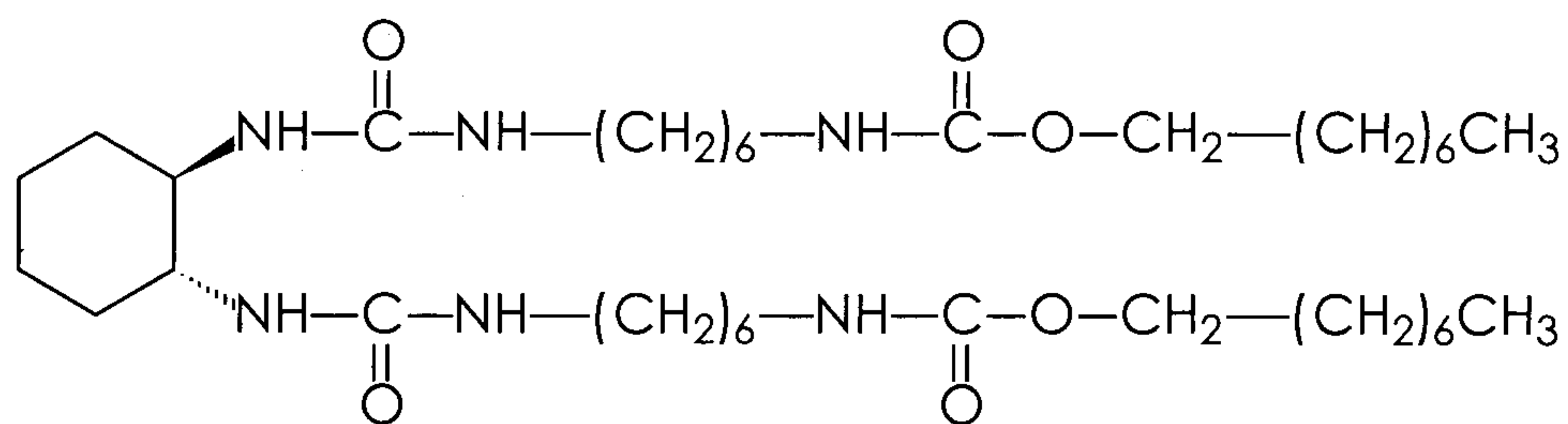
47. An ink according to claim 1 wherein the compounds are of the formulae



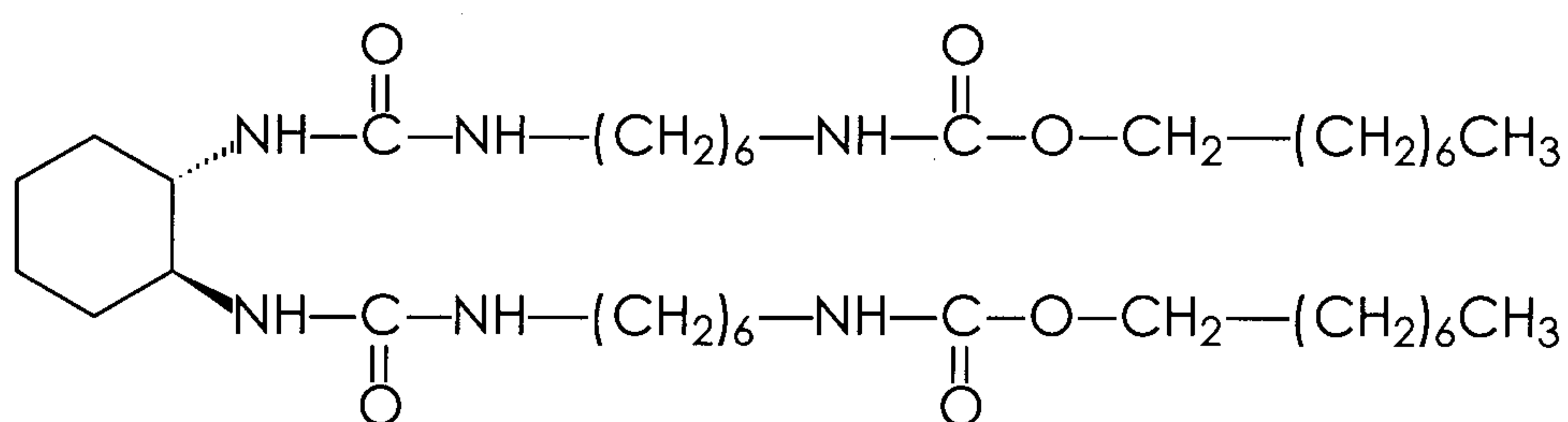
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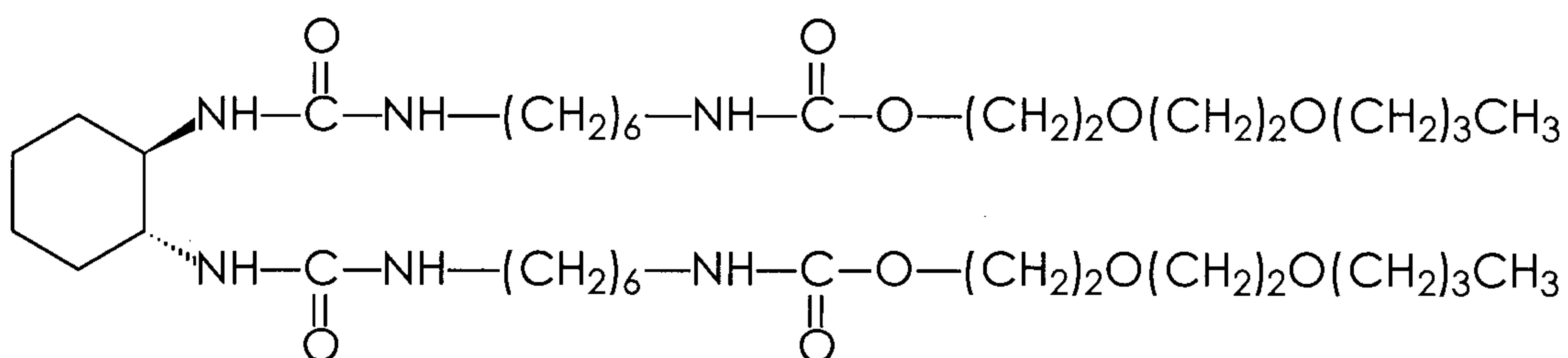
48. An ink according to claim 1 wherein the compounds are of the formulae



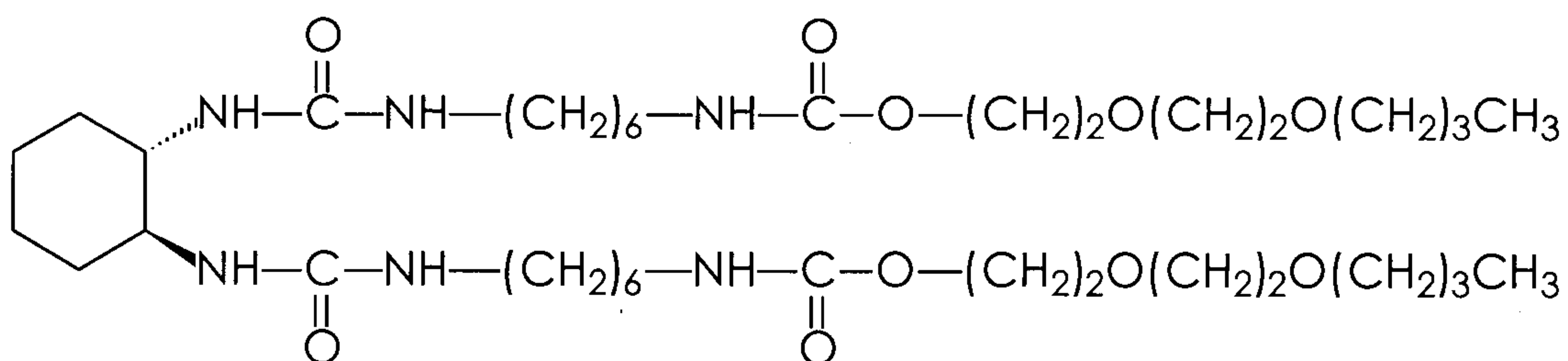
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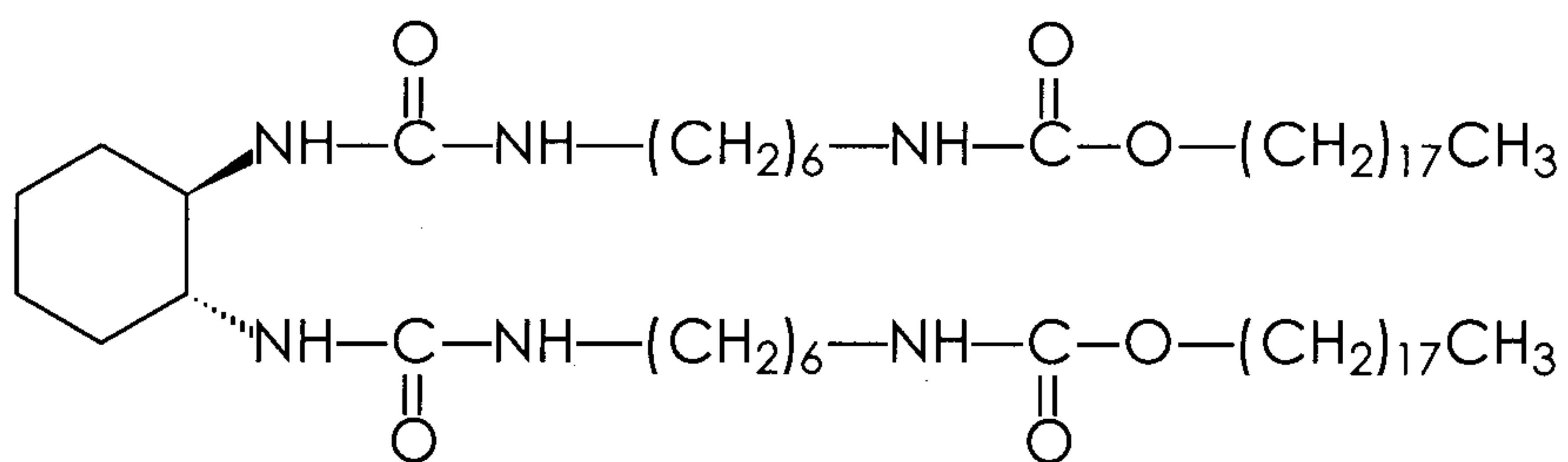
49. An ink according to claim 1 wherein the compounds are of the formulae



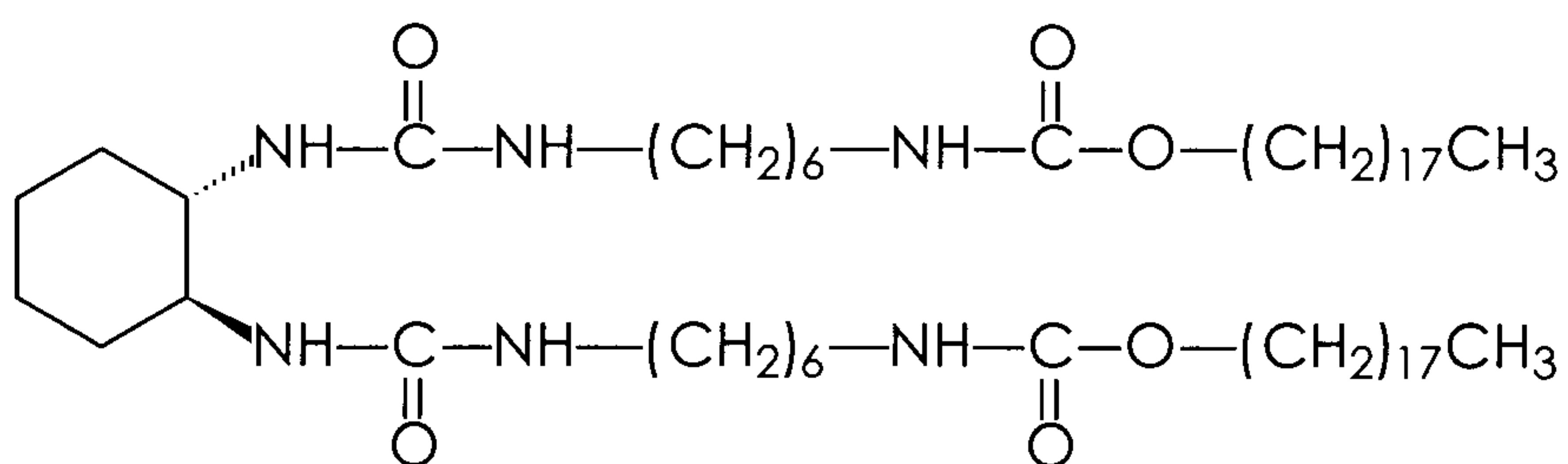
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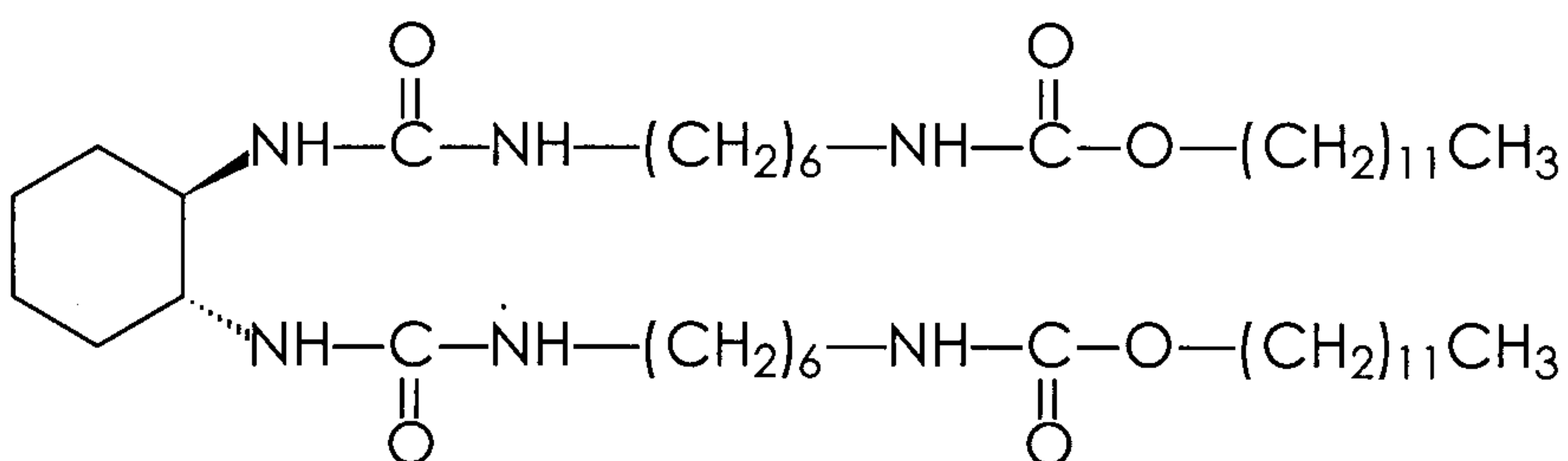
50. An ink according to claim 1 wherein the compounds are of the formulae



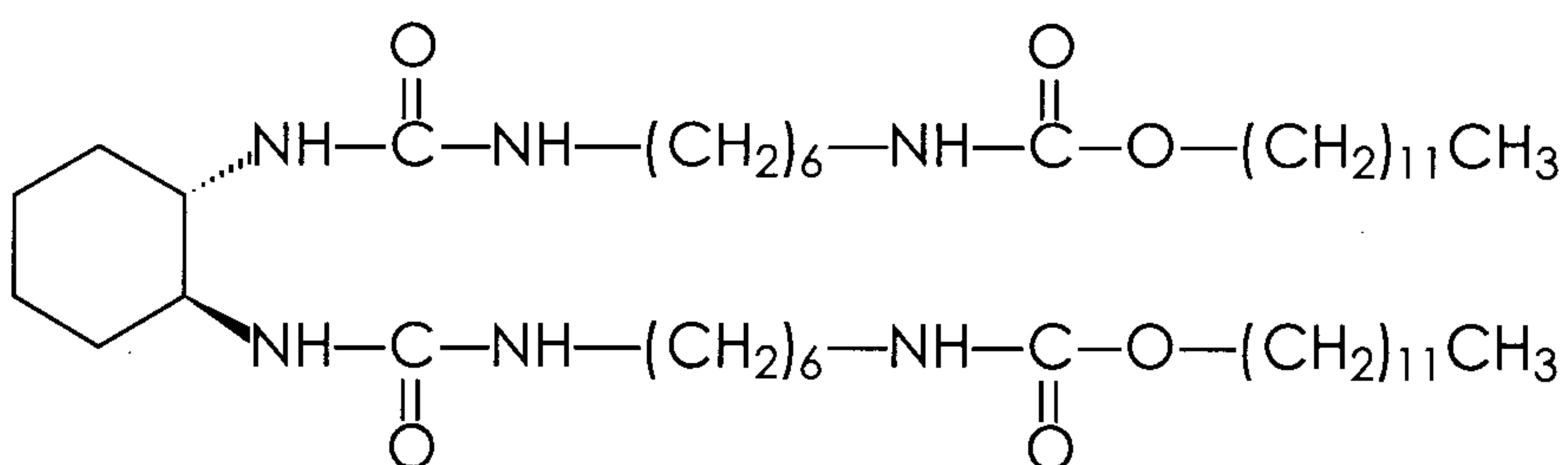
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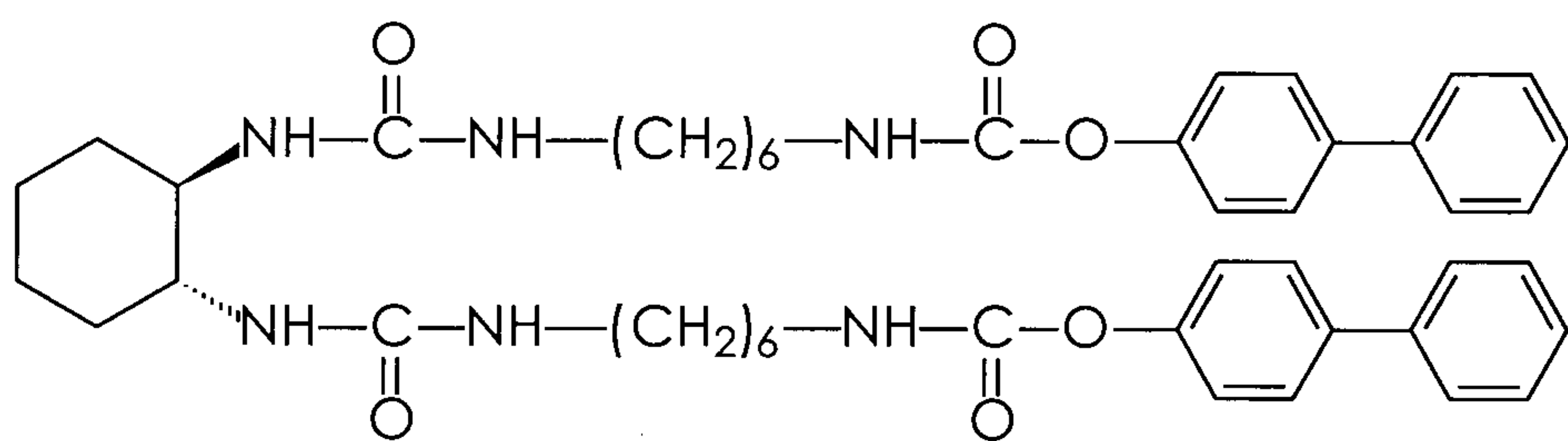
51. An ink according to claim 1 wherein the compounds are of the formulae



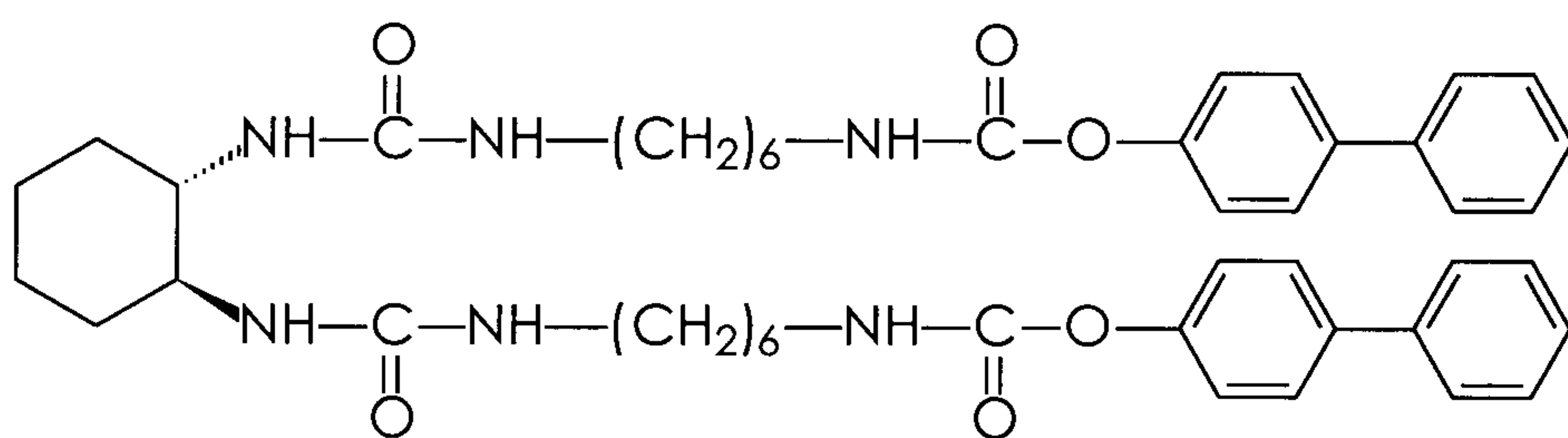
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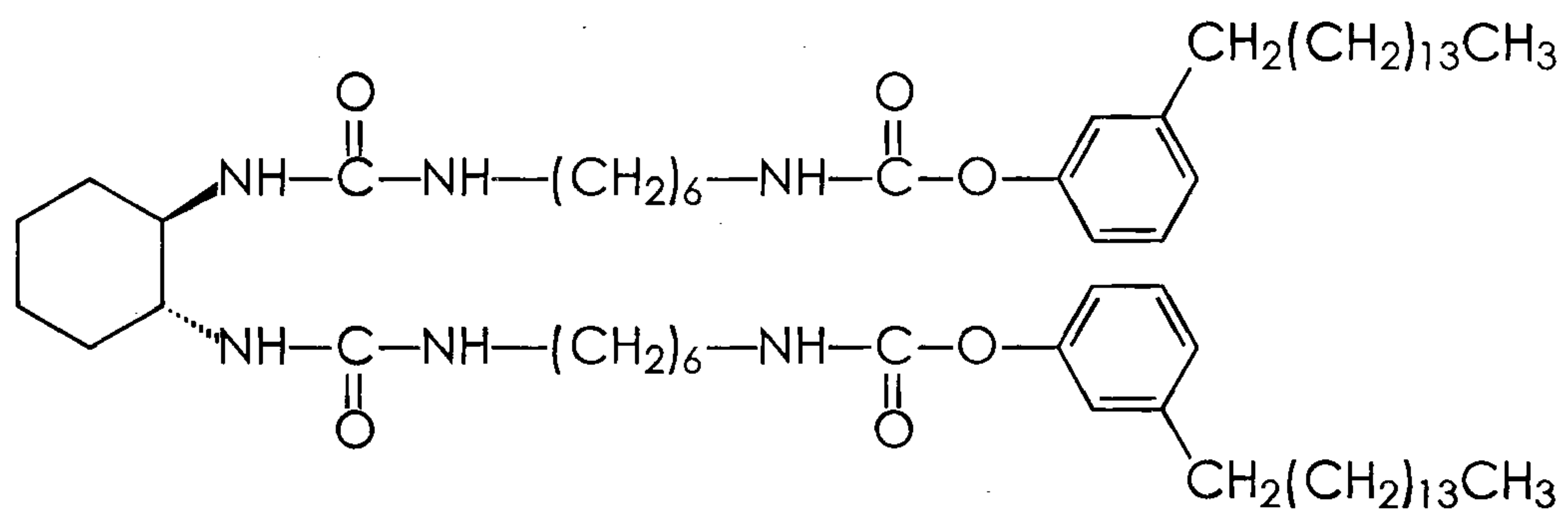
52. An ink according to claim 1 wherein the compounds are of the formulae



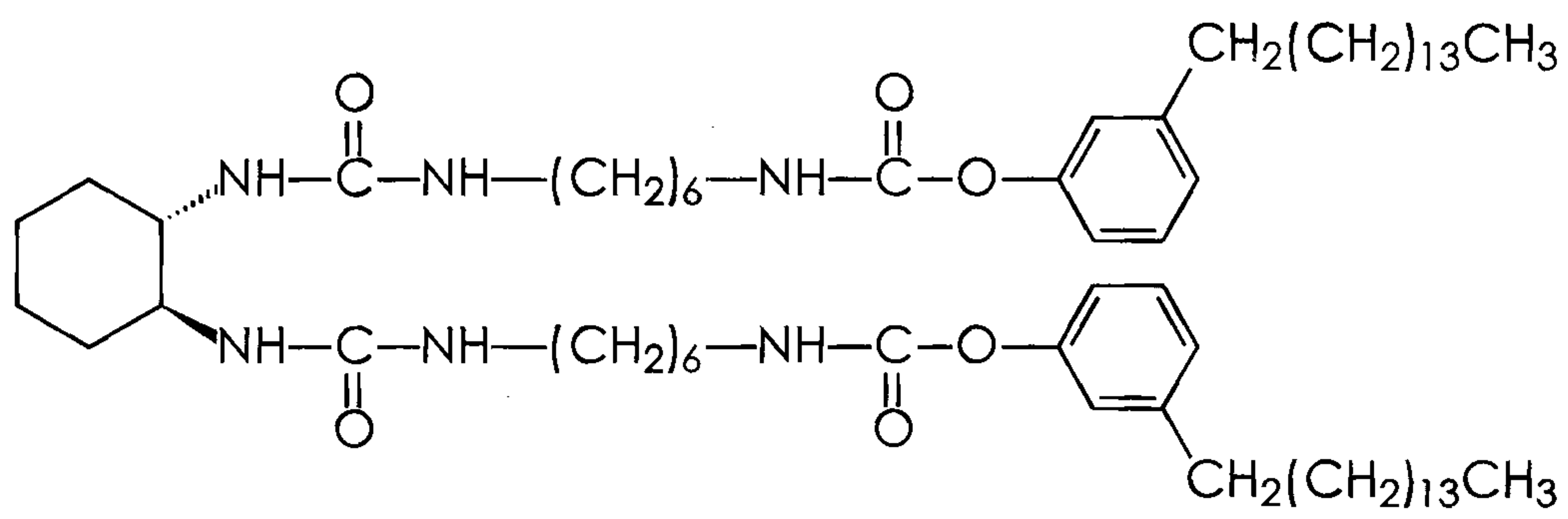
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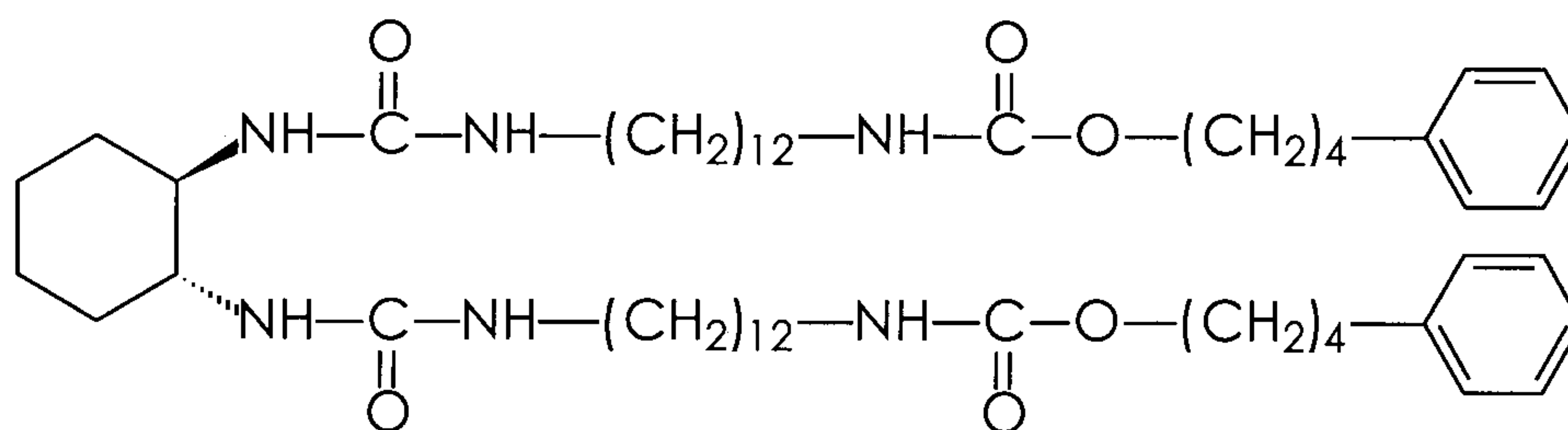
53. An ink according to claim 1 wherein the compounds are of the formulae



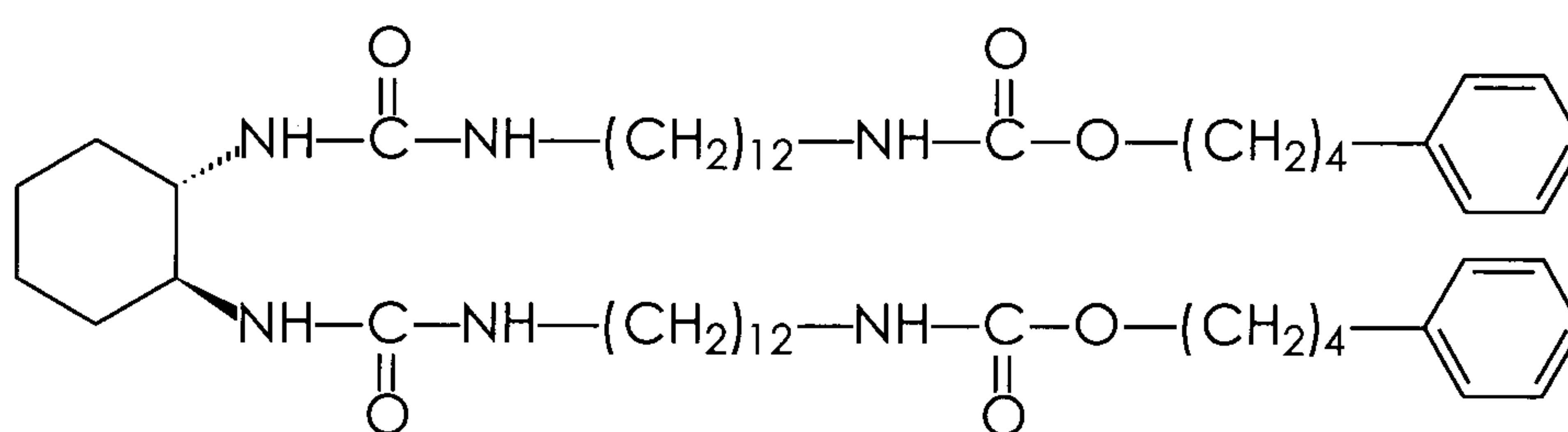
and



54. An ink according to claim 1 wherein the compounds are of the formulae



and



55. An ink according to claim 1 wherein the *trans*-1,2-cyclohexane bis[urea-urethane] compound is present in the ink in an amount of at least about 0.1 percent by weight of the ink.

56. An ink according to claim 1 wherein the *trans*-1,2-cyclohexane bis[urea-urethane] compound is present in the ink in an amount of at least about 1 percent by weight of the ink.

57. An ink according to claim 1 wherein the *trans*-1,2-cyclohexane bis[urea-urethane] compound is present in the ink in an amount of at least about 5 percent by weight of the ink.

58. An ink according to claim 1 wherein the *trans*-1,2-cyclohexane bis[urea-urethane] compound is present in the ink in an amount of no more than about 30 percent by weight of the ink.

59. An ink according to claim 1 wherein the *trans*-1,2-cyclohexane bis[urea-urethane] compound is present in the ink in an amount of no more than about 15 percent by weight of the ink.

60. An ink according to claim 1 wherein the *trans*-1,2-cyclohexane bis[urea-urethane] compound is present in the ink in an amount of no more than about 10 percent by weight of the ink.

61. An ink according to claim 1 wherein the ink contains a stereoisomerically pure *trans*-1,2-cyclohexane bis[urea-urethane] compound.

62. An ink according to claim 1 wherein the ink contains a mixture of both stereoisomeric forms of the *trans*-1,2-cyclohexane bis[urea-urethane] compound.

63. An ink according to claim 62 wherein the mixture is a racemic mixture.

64. An ink according to claim 1 having a tan-delta of less than about 10 in a region above the ink melting point.

65. An ink according to claim 1 having a tan-delta of less than about 5 in a region above the ink melting point.

66. An ink according to claim 1 having a tan-delta of less than about 1 in a region above the ink melting point.

67. An ink according to claim 1 exhibiting a gel phase at a temperature no more than about 40°C above the ink melting point.

68. An ink according to claim 1 exhibiting a gel phase at a temperature no more than about 20°C above the ink melting point.

69. An ink according to claim 1 further containing a material selected from the group consisting of monoamides, tetra-amides, and mixtures thereof.

70. An ink according to claim 1 further containing a material selected from the group consisting of polyethylene waxes.

71. An ink according to claim 1 further containing a material selected from the group consisting of fatty alcohols.

72. An ink according to claim 1 wherein the ink carrier has a melting point of higher than about 110°C.

73. An ink according to claim 1 wherein the ink carrier has a melting point of higher than about 100°C.

74. An ink according to claim 1 wherein the ink has a melting point of no lower than about 40°C.

75. An ink according to claim 1 wherein the ink has a melting point of no lower than about 70°C.

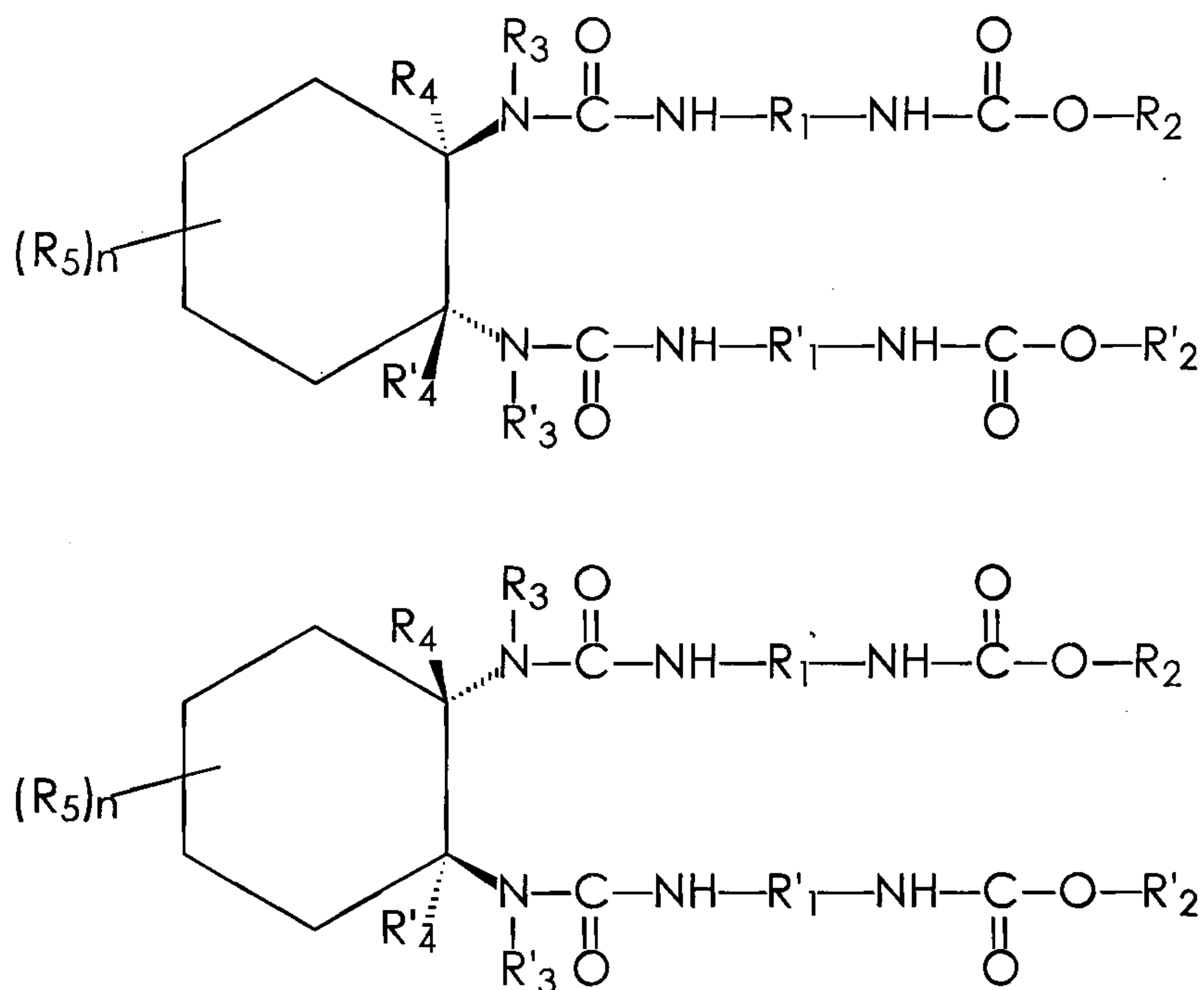
76. An ink according to claim 1 wherein the ink has a melting point of no higher than about 140°C.

77. An ink according to claim 1 wherein the ink has a melting point of no higher than about 100°C.

78. An ink according to claim 1 wherein the ink exhibits a melt viscosity at jetting temperatures of no more than about 30 centipoise.

79. An ink according to claim 1 wherein the ink exhibits a melt viscosity at jetting temperatures of no more than about 15 centipoise.

80. A process which comprises (1) incorporating into an ink jet printing apparatus a phase change ink composition comprising a phase change ink carrier and a *trans*-1,2-cyclohexane bis[urea-urethane] compound of the formula



or mixtures thereof, wherein R₁ and R'₁ each, independently of the other, is an alkylene group, an arylene group, an arylalkylene group, or an alkylarylene group, R₂ and R'₂ each, independently of the other, is an alkyl group, an aryl group, an arylalkyl group, or an alkylaryl group, R₃ and R'₃ each, independently of the other, is a hydrogen atom or an alkyl group, R₄ and R'₄ each, independently of the other, is a hydrogen atom, a fluorine atom, an alkyl group, or a phenyl group, n is an integer of 0, 1, 2, 3, or 4, and R₅ is an alkyl group, an aryl group, an arylalkyl group, an alkylaryl group, or a substituent other than an alkyl, aryl, arylalkyl, or alkylaryl group; (2) melting the ink; and (3) causing droplets of the melted ink to be ejected in an imagewise pattern onto a substrate.

81. A process according to claim 80 wherein the printing apparatus employs a piezoelectric printing process wherein droplets of the ink are caused to be ejected in imagewise pattern by oscillations of piezoelectric vibrating elements.

82. A process according to claim 80 wherein the substrate is a final recording sheet and droplets of the melted ink are ejected in an imagewise pattern directly onto the final recording sheet.

83. A process according to claim 80 wherein the substrate is an intermediate transfer member and droplets of the melted ink are ejected in an imagewise pattern onto the intermediate transfer member followed by transfer of the imagewise pattern from the intermediate transfer member to a final recording sheet.

84. A process according to claim 83 wherein the intermediate transfer member is heated to a temperature above that of the final recording sheet and below that of the melted ink in the printing apparatus.

85. A process according to claim 83 wherein the intermediate transfer member is heated to a temperature of from about 4°C above to about 60°C below the ink melting temperature.

86. A process according to claim 83 wherein the intermediate transfer member is heated to a temperature of from about 2°C above to about 50°C below the ink melting temperature.

