⑲ Europäisches Patentamt
European Patent Office
Office européen des brevets

⑪ Publication number: **0 000 769**
B1

⑫ **EUROPEAN PATENT SPECIFICATION**

㊺ Date of publication of patent specification: **05.08.81**

(51) Int. Cl.^{3}: **C 08 G 73/06, C 08 K 5/34**

㉑ Application number: **78100573.1**

㉒ Date of filing: **02.08.78**

(54) Polymers containing polyalkylpiperidines and use thereof as stabilizers.

㉚ Priority: **08.08.77 JP 94808/77**

㊸ Date of publication of application:
21.02.79 Bulletin 79/4

㊺ Publication of the grant of the European patent:
05.08.81 Bulletin 81/31

(84) Designated Contracting States:
DE FR GB

(56) References cited:
FR - A - 2 351 144

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EP 0 000 769 B1

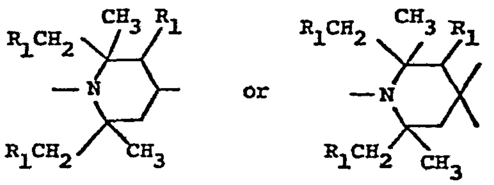
Courier Press, Leamington Spa, England.

Polymers containing polyalkylpiperidines and use thereof as stabilizers

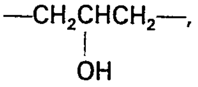
The present invention relates to novel polymers containing sterically hindered polyalkylpiperidines and which are useful as stabilizers for synthetic polymers, and synthetic polymer compositions stabilized against light- and heat-deterioration containing said novel polymers.

It is known that polyalkylpiperidine derivatives in which the 1- and/or 4-positions are substituted are effective polymer stabilizers against light- or heat-induced degradation thereof. For example, 4-spirohydantoins of 2,2,6,6-tetramethylpiperidine or 2,6-diethyl-2,3,6-trimethylpiperidine are disclosed in U.S. Patents nos. 3,542,729, 3,705,126, 3,941,744, 4,005,094 and 3,898,303 and German Offenlegungsschrift no. 2,623,464; amino derivatives are disclosed in U.S. Patents nos. 3,684,765 and 3,904,581 and German Offenlegungsschrift no. 2,621,870; esters, ethers and carbamates of 4-piperidinols are disclosed in U.S. Patents nos. 3,640,928, 3,840,494, 4,021,432, 3,940,363, 3,993,655 and 4,075,165 and German Offenlegungsschrift 2,647,452; ketals of 4-oxo compounds are disclosed in U.S. Patents nos. 3,899,464 and 3,940,363 and German Offenlegungsschrift no. 2,621,855; and pinacol type derivatives are disclosed in U.S. Patents nos. 4,061,616 and 4,055,536 as well as Japanese Provisional Patent Publication no. 51—145548, respectively. Furthermore, polymers containing 4-amino-polyalkylpiperidines as the side chain are proposed in German Offenlegungsschriften nos. 2,611,208, 2,636,144 and 2,636,130.

The invention provides polymers in which groups containing polyalkylpiperidines represented by the general formulae:



wherein R_1 represents a hydrogen atom or a methyl group, are linked in the main chain *via* bridging members containing groups having the formula

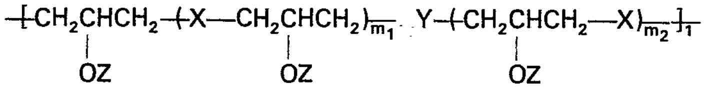


$$-\mathrm{CH_2\underset{OH}{CH}CH_2}-,$$

or derivatives thereof.

The new polymers show improved stabilizing effect in various polymeric materials against light- and heat-degradation thereof, with less sublimation and exudation therefrom.

The polymers of the present invention containing polyalkylpiperidines are represented by the following formula (I):

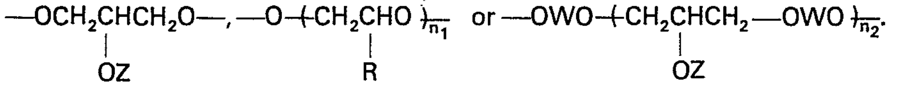


$$-\!\left[\mathrm{CH_2\underset{OZ}{CH}CH_2}\left(X-\mathrm{CH_2\underset{OZ}{CH}CH_2}\right)_{m_1}Y\left(\mathrm{CH_2\underset{OZ}{CH}CH_2}-X\right)_{m_2}\right]_{l}\!- \qquad (I)$$

In the above formula, l represents an integer of from 2 to 50, preferably 2—10, and most preferably 2—6.

Both m_1 and m_2 represent 0, or one of them represents 1 and the other represents 0.

X represents a group of formula



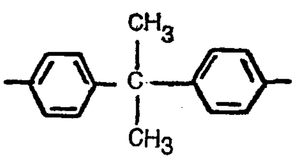
$$-\mathrm{OCH_2\underset{OZ}{CH}CH_2O}-,\ -\mathrm{O}\left(\mathrm{CH_2\underset{R}{CH}O}\right)_{n_1}\ \text{or}\ -\mathrm{OWO}\left(\mathrm{CH_2\underset{OZ}{CH}CH_2}-\mathrm{OWO}\right)_{n_2}.$$

R′ represents a hydrogen atom or a methyl group, preferably a hydrogen atom.

n_1 represents an integer of from 1 to 10, preferably 1.

n_2 represents 0 or an integer of from 1 to 10, preferably 0.

W represents a group of formula



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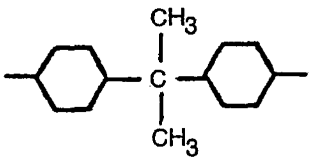
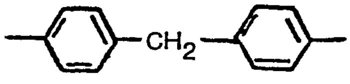


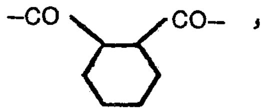


or $-\mathrm{CO}\left(\mathrm{CH_2}\right)_{n_3}\mathrm{CO}-$.

n_3 represents an integer of from 1 to 10, preferably 4 to 10, most preferably 4. As such groups may be mentioned, for example, the malonyl, succinyl, adipoyl, suberoyl, sebacoyl or decane-1,10-dicarbonyl group.

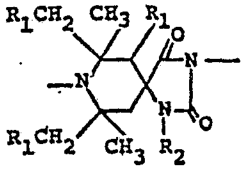
W further represents a group of formula

e.g. isophthaloyl or terephthaloyl, preferably phthaloyl, or a group of formula



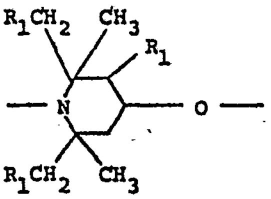
e.g. cyclohexane-1,2-dicarbonyl.

Y represents, when both m_1 and m_2 are 0, one of the following formulae (II) to (IV):



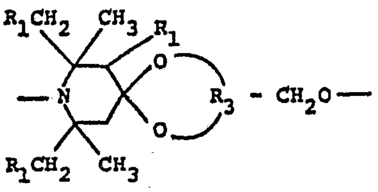
(II)

in which R_1 represents a hydrogen atom or a methyl group, preferably a hydrogen atom, and R_2 represents a hydrogen atom or an alkyl group having from 1 to 18 carbon atoms, e.g. methyl, ethyl, n-propyl, n-butyl, octyl, dodecyl or octadecyl, particularly methyl. Preferably, R_2 is a hydrogen atom.



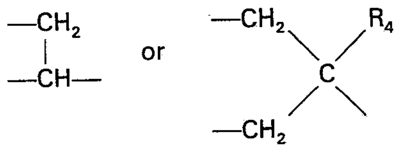
(III)

in which R_1 has the meaning given above.



(IV)

in which R_1 has the meaning defined above, R_3 represents a group of formula

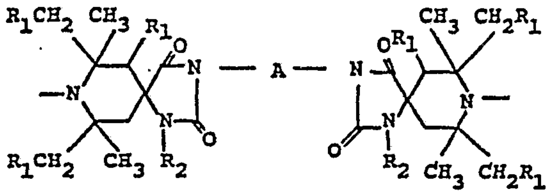


$$\begin{matrix}-CH_2\\ |\\ -CH-\end{matrix} \quad \text{or} \quad \begin{matrix}-CH_2 & & R_4\\ & C & \\ -CH_2 & & \end{matrix}$$

, and

R_4 represents the methyl or ethyl group.

Y represents, when one of m_1 and m_2 is 1 and the other is 0, a group of the following formulae (V) to (XI):

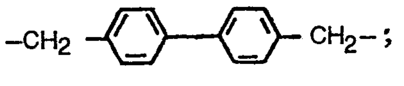


(V)

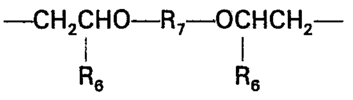
in which:

R_1 and R_2 have the meanings defined above.

A represents an alkylene group having from 2 to 12, preferably 2 to 6, and most preferably 6 carbon atoms, e.g. ethylene, tetramethylene, hexamethylene, octamethylene, decamethylene or dodecamethylene; a xylylene group, e.g. o-, m- or p-xylylene, preferably p-xylylene; a group of formula

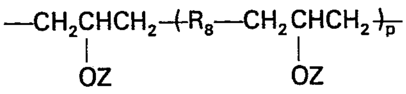


a group of formula $—CH_2COO—R_5—OCOCH_2—$ in which R_5 is an alkylene group having from 2 to 8 carbon atoms, e.g. ethylene, tetramethylene, hexamethylene or 2-ethyl-1,6-hexylene, preferably ethylene; a group of formula



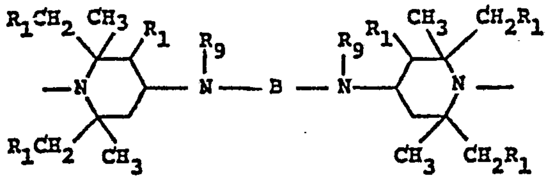
$$—CH_2\underset{R_6}{C}HO—R_7—O\underset{R_6}{C}HCH_2—$$

in which R_6 represents a hydrogen atom, a methyl group or a phenyl group, preferably a hydrogen atom, and R_7 represents an aliphatic, aromatic or alicyclic diacyl group having up to 12 carbon atoms, for example, an alkanedicarbonyl group having from 4 to 12 carbon atoms and which is optionally interrupted by sulfur, such as malonyl, succinyl, adipoyl, sebacoyl, decane-1,10-dicarbonyl or 3,3'-thiodipropionyl, phthaloyl, isophthaloyl, terephthaloyl or cyclohexane-1,4-dicarbonyl; or a group of formula



$$—CH_2\underset{OZ}{C}HCH_2—(R_8—CH_2\underset{OZ}{C}HCH_2)_p—$$

in which p is 0 or 1, preferably 1, R_8 has the same meaning as the above-defined X, and Z has the meaning defined below.

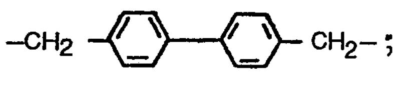


(VI)

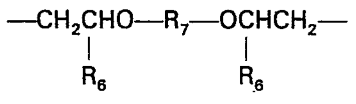
in which:

R_1 has the meaning defined above. R_9 represents an alkyl group having from 1 to 18, preferably 1 to 18 carbon atoms, e.g. methyl, ethyl, n-propyl, n-butyl, n-octyl 2-ethylhexyl, undecyl or octadecyl; a phenyl group; a benzyl group; a cyclohexyl group; or an aliphatic, aromatic, araliphatic or alicyclic acyl group having up to 18 carbon atoms, for example, an alkanoyl group having from 2 to 18 carbon atoms such as acetyl, propionyl, hexanoyl, octanoyl, 2-ethylhexanoyl, lauroyl or stearoyl, benzoyl, toluoyl or 3-(2,4-di-tert-butyl-4-hydroxyphenyl)propionyl. R_9 most preferably is an alkyl group having from 1 to 4 carbon atoms or the acetyl group.

B represents an alkylene group having from 2 to 10, preferably 2 to 6 carbon atoms, e.g. ethylene, tetramethylene, hexamethylene, octamethylene or decamethylene; a xylylene group, e.g. o-, m- or p-xylylene, preferably p-xyiylene; a group of formula

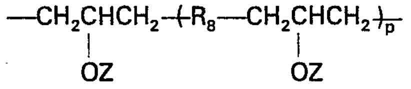


a group of formula $—CH_2COO—R_5—OCOCH_2—$ in which R_5 has the meaning defined above; a group of formula



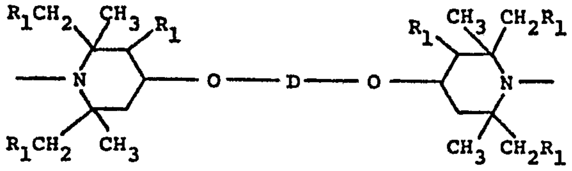
$$—CH_2\underset{R_6}{C}HO—R_7—O\underset{R_6}{C}HCH_2—$$

in which R_6 and R_7 have the meanings defined above; or a group of formula



$$—CH_2\underset{OZ}{C}HCH_2—(R_8—CH_2\underset{OZ}{C}HCH_2)_p—$$

in which R_8 and p have the meanings given above and Z has the meaning defined below.

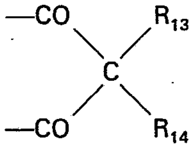


(VII)

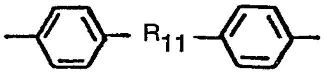
in which:

R_1 has the meaning defined above.

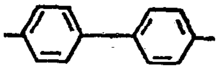
D represents an aliphatic, aromatic, araliphatic or alicyclic diacyl group having up to 36 carbon atoms, for example diacyl groups as described under R_7, an alkanedicarbonyl group having from 4 to 12 carbon atoms and which is optionally interrupted by sulfur, phthaloyl, isophthaloyl, terephthaloyl or cyclohexane-1,4-dicarbonyl, or a diacyl group derived from a dimeric acid, or a group of formula

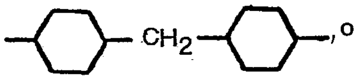
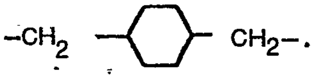


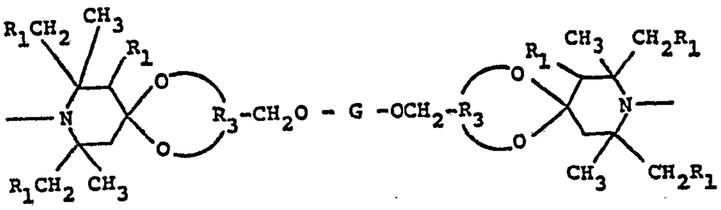
in which R_{13} represents an alkyl group having from 1 to 4 carbon atoms, a benzyl group or a 3,5-di-tert-butyl-4-hydroxybenzyl group and R_{14} has the same meaning as R_{13} or represents a hydrogen atom; or a group of formula —CONH—R_{10}—NHCO— in which R_{10} represents an alkylene group having from 2 to 10, preferably 6 carbon atoms, e.g. ethylene, tetramethylene, hexamethylene, octamethylene or decamethylene, a phenylene group optionally substituted with methyl, e.g. o-, m- or p-phenylene, particularly 2,4-tolylene, a naphthylene group, e.g. 1,5-naphthylene, a xylylene group, e.g. p-xylylene, a cyclohexylene group optionally substituted with methyl, e.g. 1,4-cyclohexylene or methyl-2,4-cyclohexylene, a group of formula



in which R_{11} represents an oxygen atom or a methylene group, a group of formula

,

a group of formula , or a group of formula .

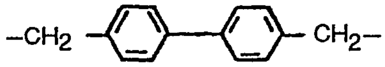


(VIII)

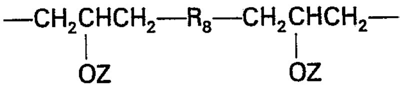
in which:

R_1 and R_3 have the meanings given above.

G has the same meaning as the above-defined D, or it represents an alkylene group having from 3 to 10, preferably 4 to 6 carbon atoms, e.g. trimethylene, tetramethylene, hexamethylene, octamethylene or decamethylene, a xylylene group, e.g. o-, m- or p-xylylene, preferably p-xylylene; a group of formula

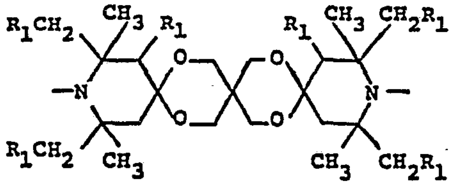


or a group of formula



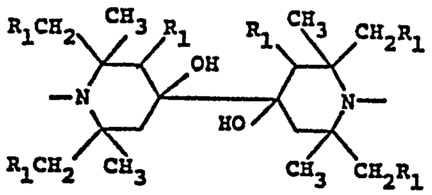
$$—\underset{\mathrm{OZ}}{\mathrm{CH_2\underset{|}{C}HCH_2}}—R_8—\underset{\mathrm{OZ}}{\mathrm{CH_2\underset{|}{C}HCH_2}}—$$

in which R_8 has the meaning defined above and Z has the meaning defined below.



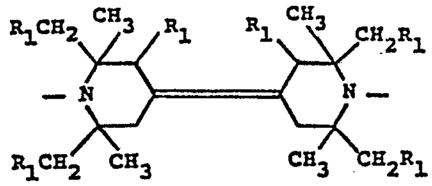
(IX)

in which R_1 has the meaning defined above.



(X)

in which R_1 has the meaning defined above.



(XI)

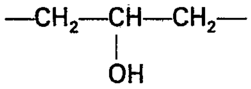
in which R_1 has the meaning defined above.

All of Z in the molecule represent a hydrogen atom; an alkyl group having from 1 to 18 carbon atoms, e.g. methyl, ethyl, n-propyl, n-butyl, octyl, dodecyl or octadecyl, particularly methyl; an aliphatic, aromatic, araliphatic or alicyclic acyl group having up to 18 carbon atoms, for example as described under R_9, an alkanoyl group having from 2 to 18, preferably 2 to 4 carbon atoms, benzoyl, toluoyl or 3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionyl; a group of formula $—CONHR_{12}$ in which R_{12} is an alkyl group having from 1 to 18, preferably 1 to 4 carbon atoms, a phenyl group or a cyclohexyl group.

Alternatively, a part of Z in the molecule may represent hydrogen atoms while the remaining Z represent above-defined groups other than hydrogen.

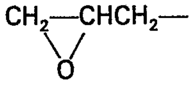
Preferably, all of Z in the molecule represent hydrogen atoms, acetyl or benzoyl groups, or a part of Z in the molecule are hydrogen atoms and the remaining Z are acetyl or benzoyl groups.

In formula (I), the terminal groups



$$—CH_2—\underset{\displaystyle OH}{\underset{|}{CH}}—CH_2—$$

may be present as groups of formula



which impart a similar stabilizing effect and are included within the scope of the polymers of the invention.

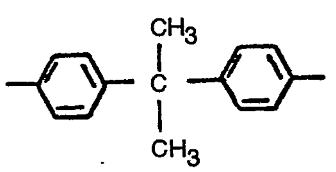
Among the polymers of formula (I), preferred groups are as follows:

1) in formula (I), when both m_1 and m_2 represent 0;

1-a) polymers in which Y is a group of formula (II) and R_1 and R_2 are hydrogen atoms,

1-b) polymers in which Y is a group of formula (III) or (IV) and R_1 is a hydrogen atom;

2) in formula (I), when one of m_1 and m_2 represents 1 and the other represents 0, X represents a group of formula $—OCH_2CH_2O—$ or —OWO— in which W represents a group of formula



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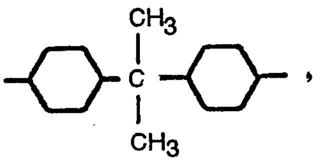

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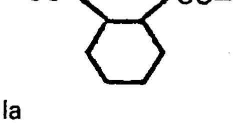
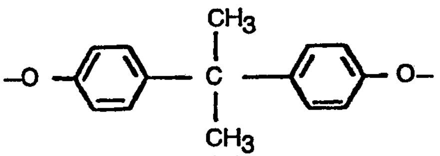
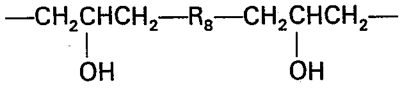
$—CO(CH_2)_{n_3}CO—$ (n_3 represents an integer of from 4 to 10, preferably 4), or a group of formula



most preferably X is a group of formula



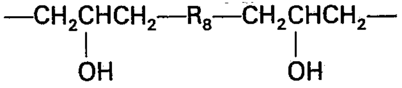
2-a) polymers in which Y is a group of formula (V), R_1 and R_2 are hydrogen atoms and A is an alkylene group having from 2 to 6, particularly 6 carbon atoms, or a group of formula



$$-\mathrm{CH_2}\underset{\mathrm{OH}}{\mathrm{CH}}\mathrm{CH_2}-\mathrm{R_8}-\mathrm{CH_2}\underset{\mathrm{OH}}{\mathrm{CH}}\mathrm{CH_2}-$$

wherein R_8 has the same meaning as the above-defined X,

2-b) polymers in which Y is a group of formula (VI), R_1 is a hydrogen atom, and R_9 is an alkanoyl group having from 2 to 4 carbon atoms or an alkyl group having from 1 to 8 carbon atoms and B is an alkylene group having from 2 to 6 carbon atoms, or R_9 is an alkyl group having from 1 to 8 carbon atoms and B is a group of formula

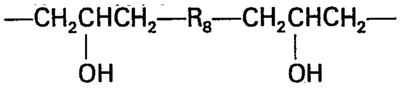


$$-\mathrm{CH_2}\underset{\mathrm{OH}}{\mathrm{CH}}\mathrm{CH_2}-\mathrm{R_8}-\mathrm{CH_2}\underset{\mathrm{OH}}{\mathrm{CH}}\mathrm{CH_2}-$$

wherein R_8 has the same meaning as X;

2-c) polymers in which Y is a group of formula (VII), R_1 is a hydrogen atom and D is an alkanedicarbonyl group having from 4 to 12 carbon atoms, particularly 10 carbon atoms, or a benzene dicarbonyl group;

2-d) polymers in which Y is a group of formula (VIII), R_1 is a hydrogen atom and G is an alkanedicarbonyl group having from 4 to 12 carbon atoms or a group of formula

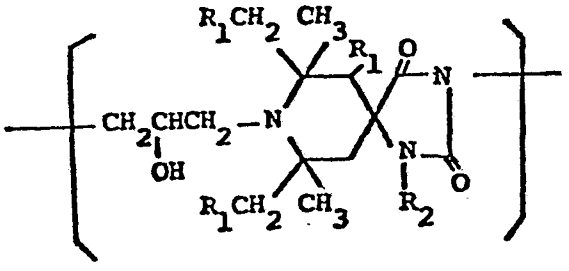


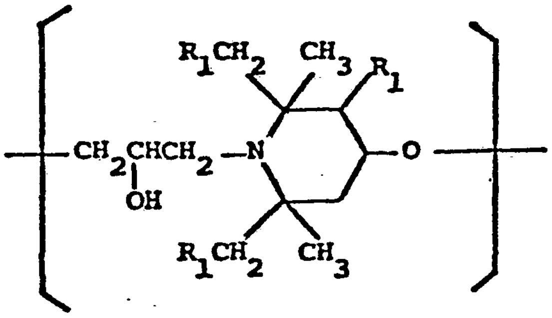
$$-\mathrm{CH_2}\underset{\mathrm{OH}}{\mathrm{CH}}\mathrm{CH_2}-\mathrm{R_8}-\mathrm{CH_2}\underset{\mathrm{OH}}{\mathrm{CH}}\mathrm{CH_2}-$$

wherein R_8 has the same meaning as the above-defined X,

2-e) polymers in which Y is a group of formula (IX), (X) or (XI) and R_1 is a hydrogen atom.

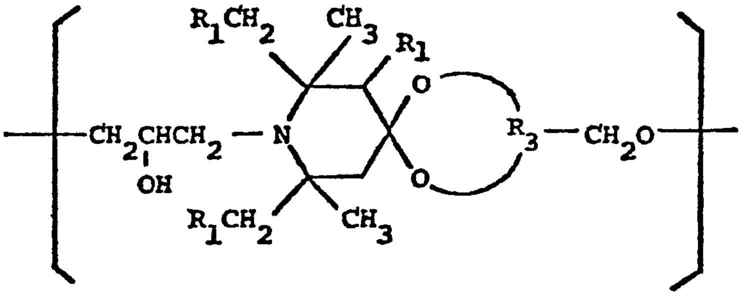
The following is a non-limiting list of recurring units of the polymers of the invention:

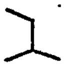
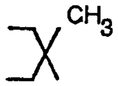
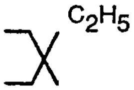


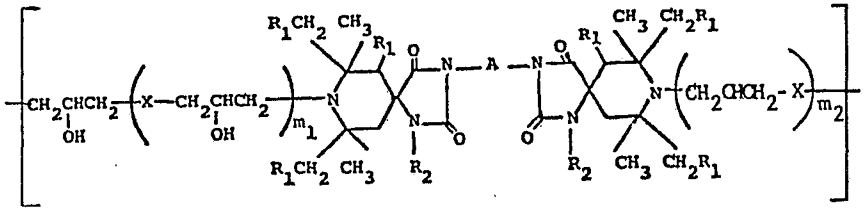


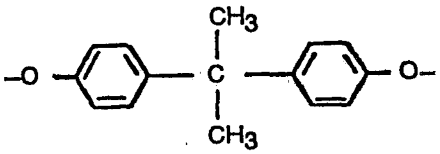
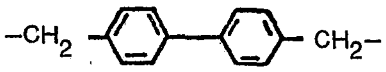
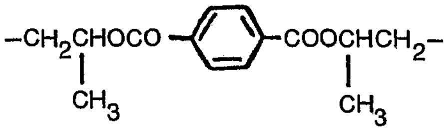
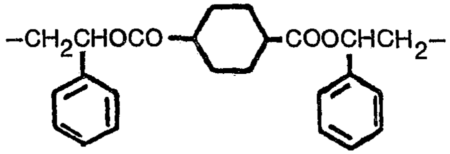
No.	R1	R2
II–1	H	H
II–2	CH_3	H

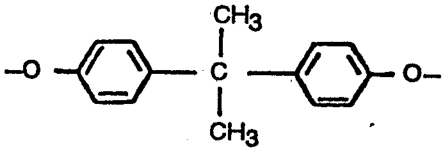
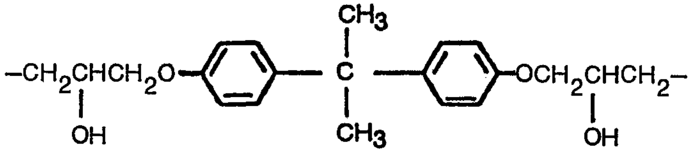
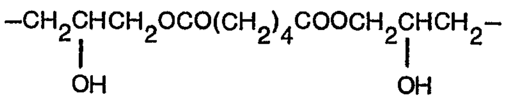
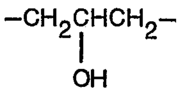
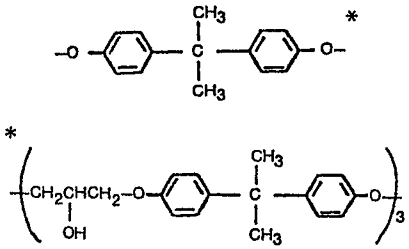
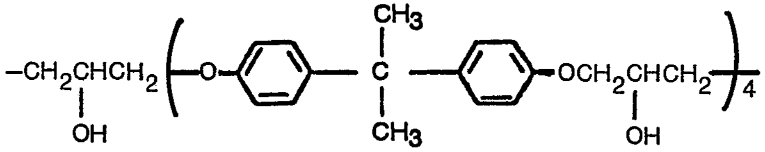
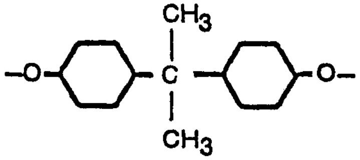
No.	R1
III–1	H

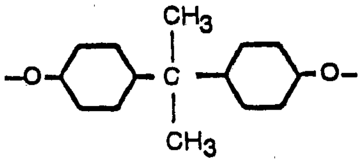
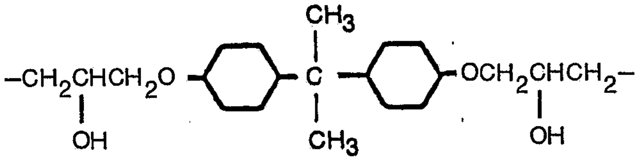
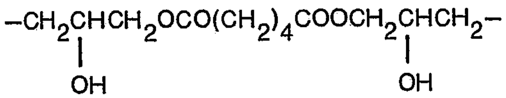
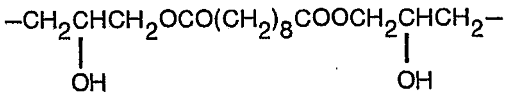
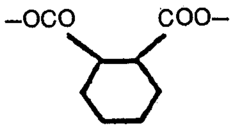
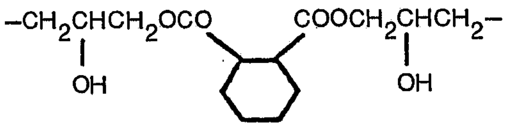
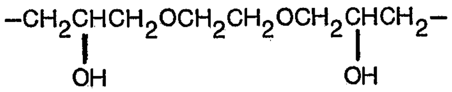
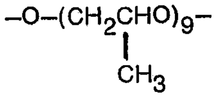
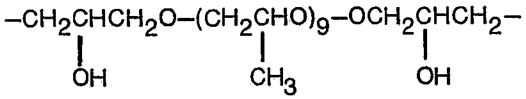


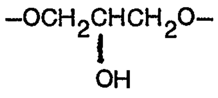
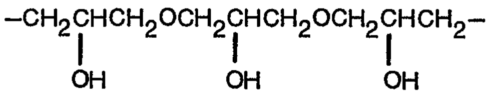
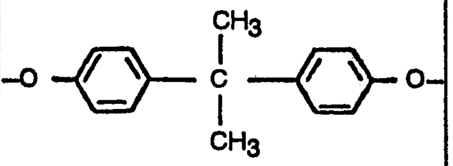
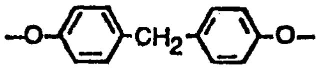
No.	R_1	R_3
IV–1	H	
IV–2	H	 CH_3
IV–3	H	 C_2H_5
IV–4	CH_3	

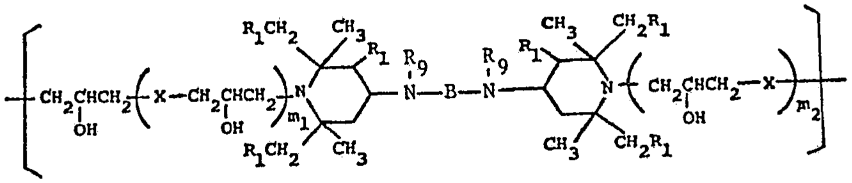


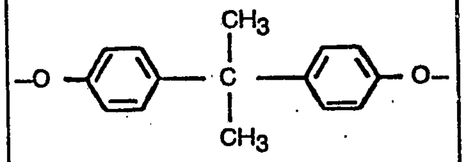
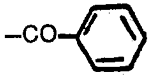
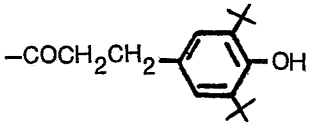
No.	X	R_1	R_2	A
V–1		H	H	$-(CH_2)_4-$
V–2	,,	H	H	$-(CH_2)_6-$
V–3	,,	H	H	
V–4	,,	H	H	
V–5	,,	H	H	$-CH_2COO(CH_2)_2OCOCH_2-$
V–6	,,	H	H	$-CH_2CH_2OCO(CH_2)_4COOCH_2CH_2-$
V–7	,,	H	H	
V–8	,,	H	H	

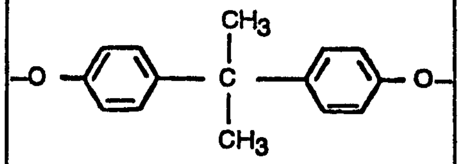
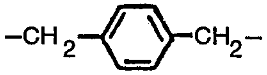
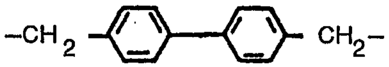
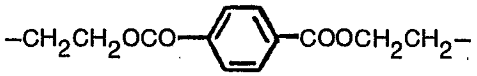
No.	X	R_1	R_2	A
V–9		H	H	
V–10	,,	H	H	 $-CH_2CH(OH)CH_2OCO(CH_2)_4COOCH_2CH(OH)CH_2-$
V–11	,,	H	H	 $-CH_2CH(OH)CH_2-$
V–12		H	H	
V–13		H	H	$-(CH_2)_6-$

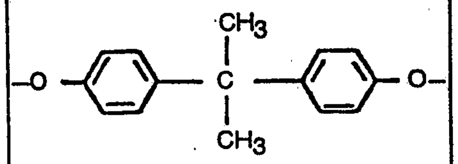
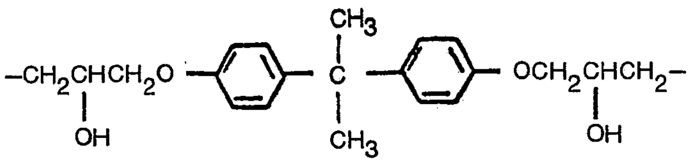
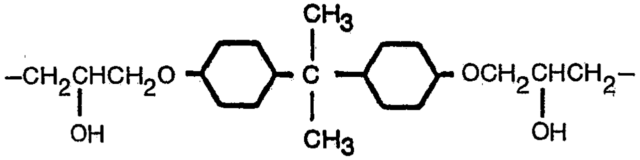
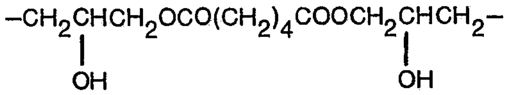
No.	X	R_1	R_2	A
V–14		H	H	
V–15	$-OCO(CH_2)_4COO-$	H	H	$-(CH_2)_4-$
V–16	,,	H	H	 $-CH_2\underset{\mathrm{OH}}{\underset{\vert}{C}}HCH_2OCO(CH_2)_4COOCH_2\underset{\mathrm{OH}}{\underset{\vert}{C}}HCH_2-$
V–17	$-OCO(CH_2)_8COO-$	H	H	 $-CH_2\underset{\mathrm{OH}}{\underset{\vert}{C}}HCH_2OCO(CH_2)_8COOCH_2\underset{\mathrm{OH}}{\underset{\vert}{C}}HCH_2-$
V–18		H	H	
V–19		H	H	
V–20	$-OCH_2CH_2O-$	H	H	 $-CH_2\underset{\mathrm{OH}}{\underset{\vert}{C}}HCH_2OCH_2CH_2OCH_2\underset{\mathrm{OH}}{\underset{\vert}{C}}HCH_2-$
V–21	 $-O-(CH_2\underset{\mathrm{CH_3}}{\underset{\vert}{C}}HO)_9-$	H	H	 $-CH_2\underset{\mathrm{OH}}{\underset{\vert}{C}}HCH_2O-(CH_2\underset{\mathrm{CH_3}}{\underset{\vert}{C}}HO)_9-OCH_2\underset{\mathrm{OH}}{\underset{\vert}{C}}HCH_2-$

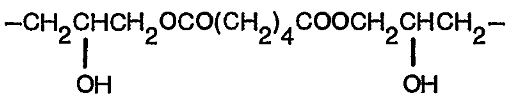
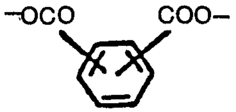
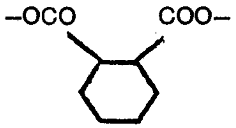
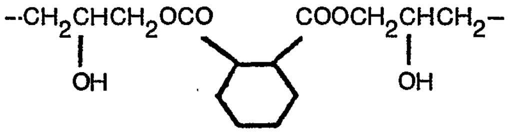
No.	X	R_1	R_2	A
V–22	 $-OCH_2\underset{OH}{\underset{\mid}{C}}HCH_2O-$	H	H	 $-CH_2\underset{OH}{\underset{\mid}{C}}HCH_2OCH_2\underset{OH}{\underset{\mid}{C}}HCH_2OCH_2\underset{OH}{\underset{\mid}{C}}HCH_2-$
V–23		H	CH_3	$-(CH_2)_4-$
V–24	,,	CH_3	H	,,
V–25		H	H	,,

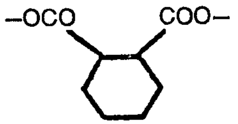
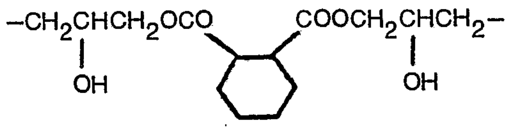
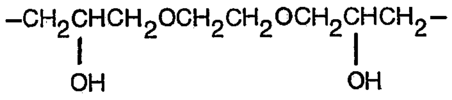
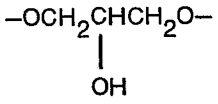
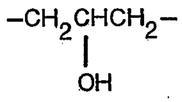
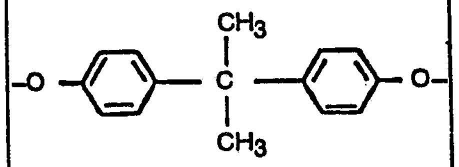
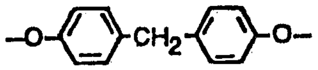
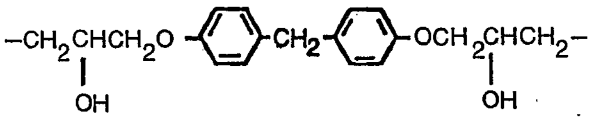


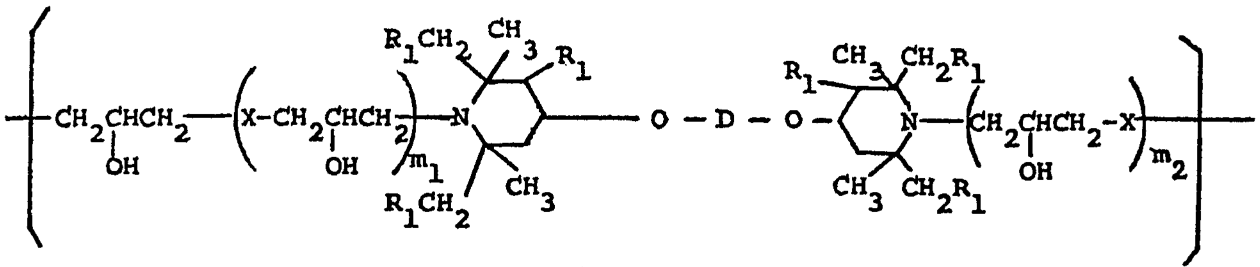
No.	X	R_1	R_9	B
VI–1		H	$COCH_3$	$-(CH_2)_2-$
VI–2	,,	H	$COCH_3$	$-(CH_2)_3-$
VI–3	,,	H	$COCH_3$	$-(CH_2)_4-$
VI–4	,,	H	$COCH_3$	$-(CH_2)_6-$
VI–5	,,	H	$-COC_2H_5$	$-(CH_2)_6-$
VI–6	,,	H	$-COC_{11}H_{23}$	,,
VI–7	,,	H	$-COC_{17}H_{35}$	,,
VI–8	,,	H		,,
VI–9	,,	H		,,

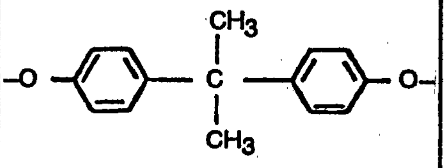
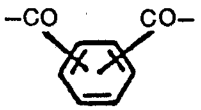
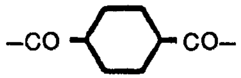
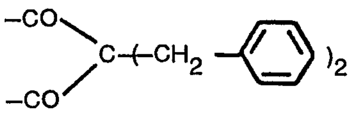
No.	X	R_1	R_9	B
VI–10		H	$-CH_3$	$-(CH_2)_6-$
VI–11	,,	H	$-C_4H_9$	,,
VI–12	,,	H	$-C_8H_{17}$	,,
VI–13	,,	H	$-COCH_3$	$-(CH_2)_{10}-$
VI–14	,,	H	$-C_4H_9$	
VI–15	,,	H	$-C_4H_9$	
VI–16	,,	H	$-C_4H_9$	$-CH_2COO(CH_2)_2OCOCH_2-$
VI–17	,,	H	$-C_4H_9$	$-CH_2CH_2OCO(CH_2)_2COOCH_2CH_2-$
VI–18	,,	H	$-C_4H_9$	$-CH_2CH_2OCO(CH_2)_8COOCH_2CH_2-$
VI–19	,,	H	$-C_4H_9$	

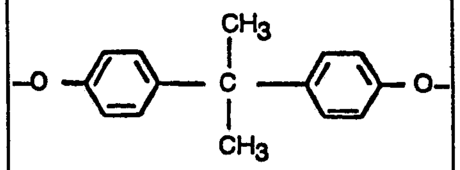
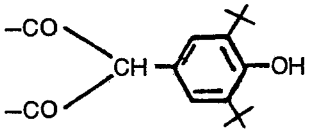
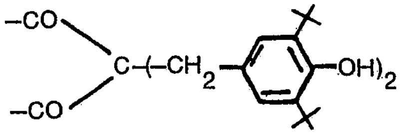
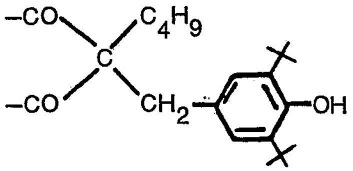
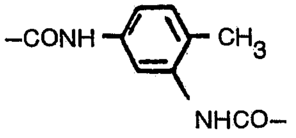
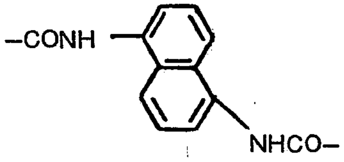
No.	X	R_1	R_9	B
VI–20		H	$-CH_3$	
VI–21	,,	H	$-C_2H_5$	,,
VI–22	,,	H	$-C_4H_9$	,,
VI–23	,,	H	$-C_8H_{17}$	,,
VI–24	,,	H	$-C_{12}H_{25}$	,,
VI–25	,,	H	$-C_{18}H_{37}$	,,
VI–26	,,	H	$-COCH_3$	$-(CH_2)_6-$
VI–27	,,	H	$-C_4H_9$	
VI–28	$-OCO(CH_2)_4COO-$	H	$-COCH_3$	$-(CH_2)_2-$
VI–29	,,	H	$-COCH_3$	$-(CH_2)_6-$
VI–30	,,	H	$-C_2H_5$	 $-CH_2CH(OH)CH_2OCO(CH_2)_4COOCH_2CH(OH)CH_2-$

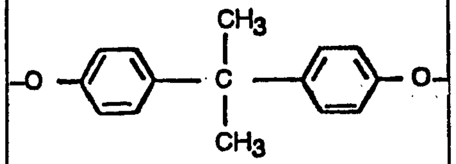
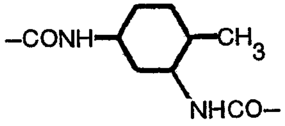
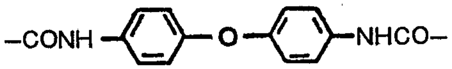
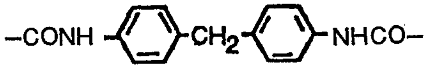
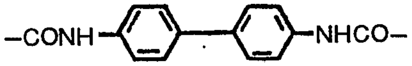
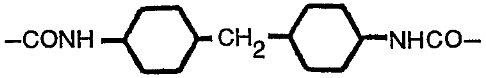
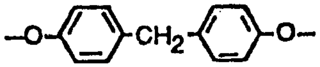
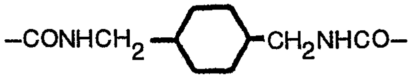
No.	X	R_1	R_9	B
VI–31	$-OCO(CH_2)_4COO-$	H	$-C_4H_9$	 $-CH_2\underset{OH}{\underset{\vert}{C}}HCH_2OCO(CH_2)_4COOCH_2\underset{OH}{\underset{\vert}{C}}HCH_2-$
VI–32	,,	H	$-C_8H_{17}$	,,
VI–33		H	$-COCH_3$	$-(CH_2)_6-$
VI–34	,,	H	$-C_4H_9$	
VI–35		H	$-COCH_3$	$-(CH_2)_2-$
VI–36	,,	H	$-COCH_3$	$-(CH_2)_6-$
VI–37	,,	H	$-C_4H_9$	,,
VI–38	,,	H	$-C_2H_5$	
VI–39	,,	H	$-C_4H_9$	,,

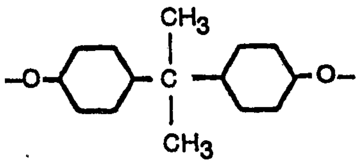
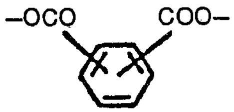
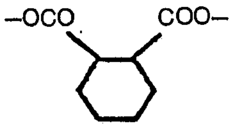
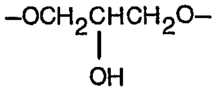
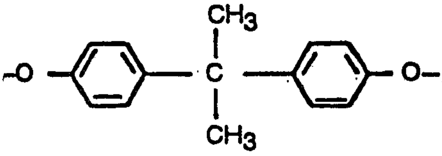
No.	X	R_1	R_9	B
VI–40		H	$-C_8H_{17}$	
VI–41	$-OCH_2CH_2O-$	H	$-COCH_3$	$-(CH_2)_6-$
VI–42	,,	H	$-C_4H_9$	 $-CH_2\underset{\mid \atop OH}{C}HCH_2OCH_2CH_2OCH_2\underset{\mid \atop OH}{C}HCH_2-$
VI–43	 $-OCH_2\underset{\mid \atop OH}{C}HCH_2O-$	H	$-C_4H_9$	 $-CH_2\underset{\mid \atop OH}{C}HCH_2-$
VI–44		CH_3	$-COCH_3$	$-(CH_2)_6-$
VI–45		H	$-COCH_3$	,,
VI–46	,,	H	$-C_2H_5$	

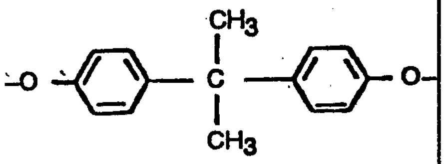
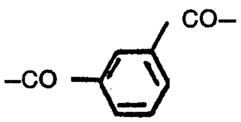
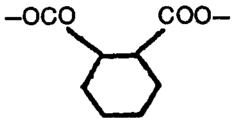
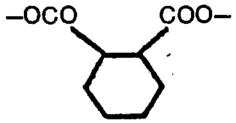


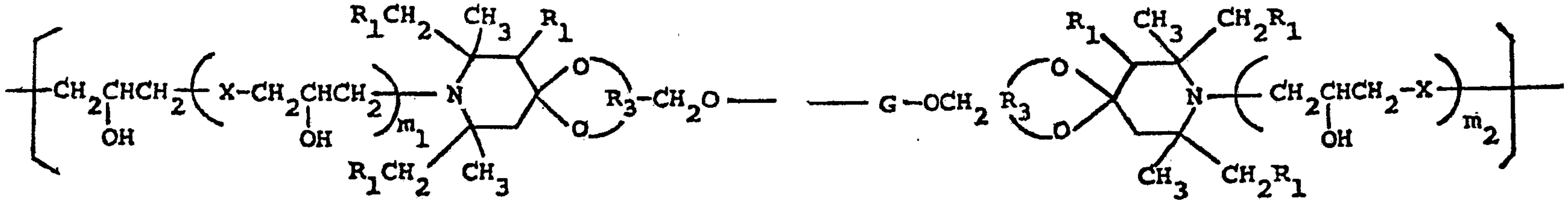
No.	X	R_1	D
VII–1		H	$-CO(CH_2)_2CO-$
VII–2	,,	H	$-CO(CH_2)_4CO-$
VII–3	,,	H	$-CO(CH_2)_8CO-$
VII–4	,,	H	$-CO(CH_2)_{10}CO-$
VII–5	,,	H	$-CO(CH_2)_2-S-(CH_2)_2CO-$
VII–6	,,	H	
VII–7	,,	H	
VII–8	,,	H	

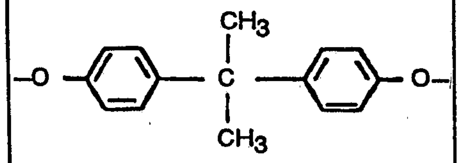
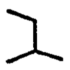
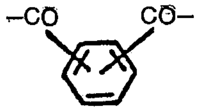
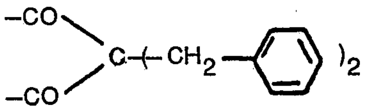
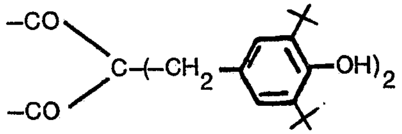
No.	X	R_1	D
VII–9		H	
VII–10	,,	H	
VII–11	,,	H	
VII–12	,,	H	$-COC_{34}H_{62}CO-$
VII–13	,,	H	$-CONH-(CH_2)_6-NHCO-$
VII–14	,,	H	
VII–15	,,	H	

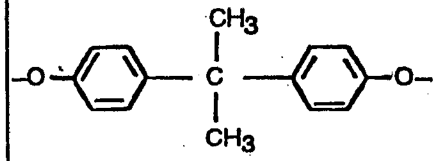
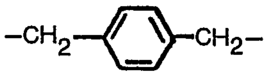
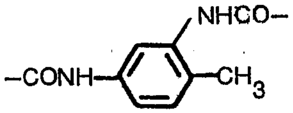
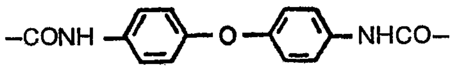
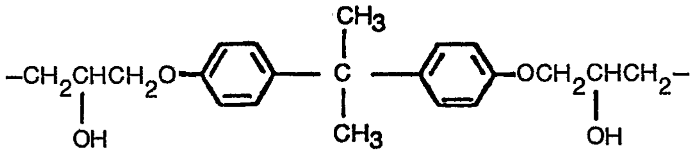
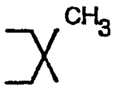
No.	X	R_1	D
VII–16		H	
VII–17	,,	H	
VII–18	,,	H	
VII–19	,,	H	
VII–20	,,	H	
VII–21	,,	H	
VII–22		H	

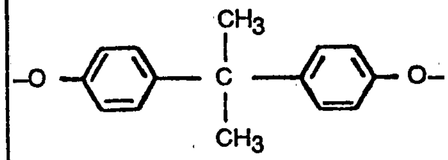
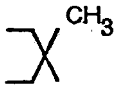
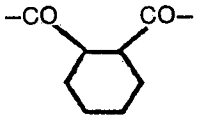
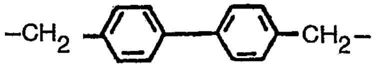
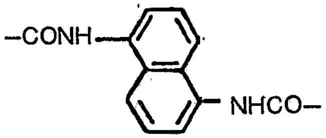
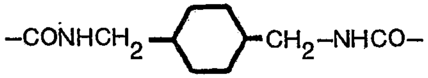
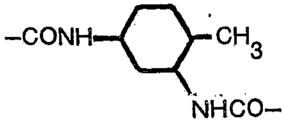
No.	X	R_1	D
VII–23		H	$-CO(CH_2)_8CO-$
VII–24	$-OCO(CH_2)_4COO-$	H	$-CO(CH_2)_4CO-$
VII–25	,,	H	$-CO(CH_2)_8CO-$
VII–26	$-OCO(CH_2)_8CO-$	H	,,
VII–27	$-OCO(CH_2)_8COO-$	H	,,
VII–28		H	,,
VII–29		H	,,
VII–30	$-OCH_2CH_2O-$	H	,,
VII–31	 $-OCH_2\underset{\displaystyle OH}{\underset{\vert}{C}}HCH_2O-$	H	,,
VII–32		CH_3	,,

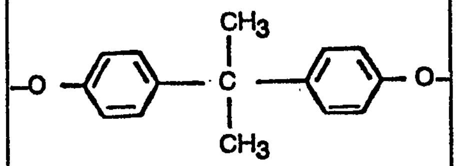
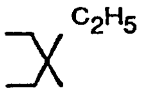
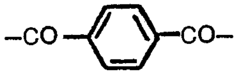
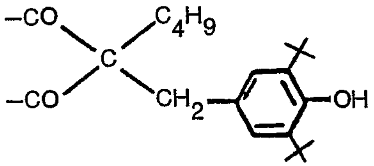
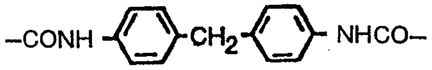
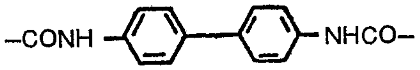
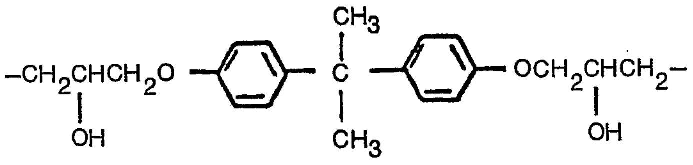
No.	X	R_1	D
VII–33		H	
VII–34		H	,,
VII–35	$-OCO(CH_2)_4COO-$	H	,,
VII–36	$-OCH_2CH_2O-$	H	,,
VII–37		H	$-CO(CH_2)_4CO-$

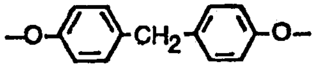
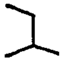
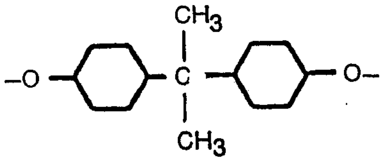
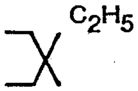
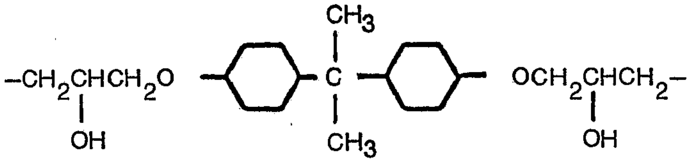
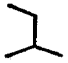
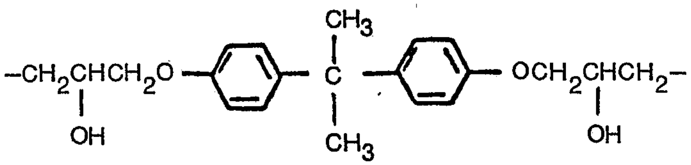
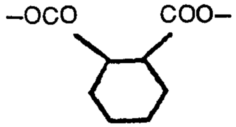
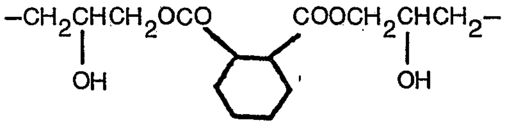


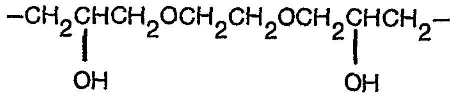
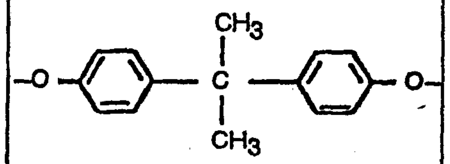
No.	X	R_1	R_3	G
VIII–1		H		$-CO(CH_2)_4CO-$
VIII–2	,,	H	,,	$-CO(CH_2)_8CO-$
VIII–3	,,	H	,,	
VIII–4	,,	H	,,	
VIII–5	,,	H	,,	
VIII–6	,,	H	,,	$-(CH_2)_4-$

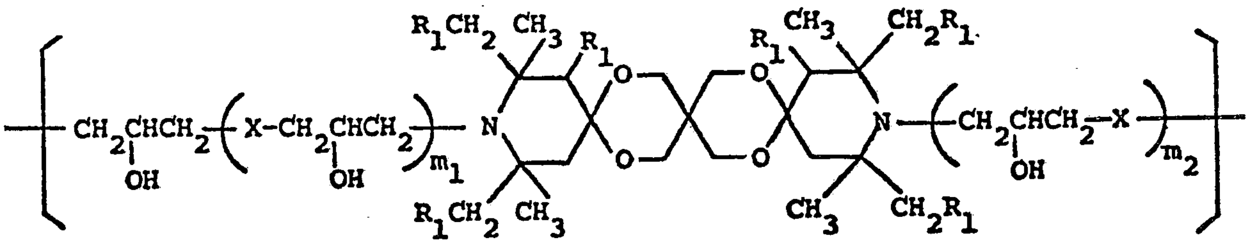
No.	X	R_1	R_3	G
VIII–7		H		
VIII–8	,,	H	,,	$-CONH(CH_2)_6NHCO-$
VIII–9	,,	H	,,	
VIII–10	,,	H	,,	
VIII–11	,,	H	,,	
VIII–12	,,	H	,,	
VIII–13	,,	H		$-CO(CH_2)_4CO-$

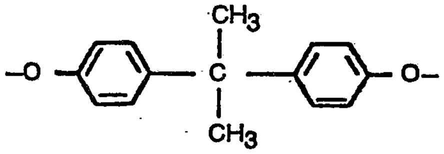
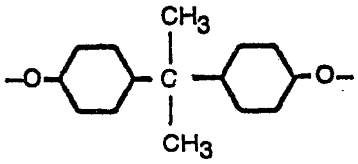
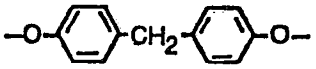
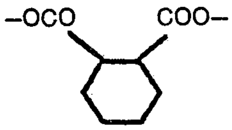
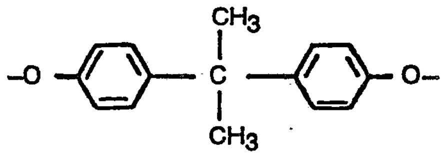
No.	X	R_1	R_3	G
VIII–14		H		$-CO(CH_2)_2-S-(CH_2)_2CO-$
VIII–15	,,	H	,,	
VIII–16	,,	H	,,	$-CO-C_{34}H_{62}-CO-$
VIII–17	,,	H	,,	
VIII–18	,,	H	,,	
VIII–19	,,	H	,,	
VIII–20	,,	H	,,	

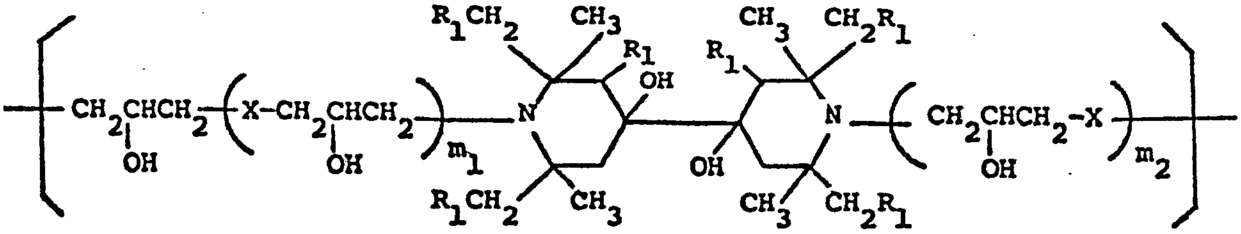
No.	X	R_1	R_3	G
VIII–21		H		$-CO(CH_2)_4CO-$
VIII–22	,,	H	,,	$-CO(CH_2)_{10}CO-$
VIII–23	,,	H	,,	
VIII–24	,,	H	,,	
VIII–25	,,	H	,,	$-(CH_2)_6-$
VIII–26	,,	H	,,	
VIII–27	,,	H	,,	
VIII–28	,,	H	,,	

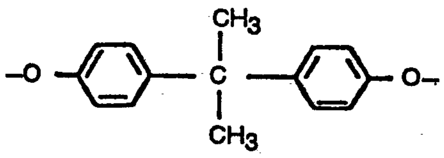
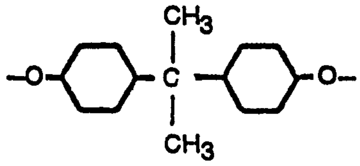
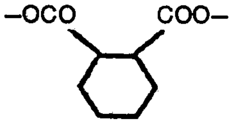
No.	X	R_1	R_3	G
VIII–29		H		$-CO(CH_2)_4CO-$
VIII–30		H		$-CO(CH_2)_4CO-$
VIII–31	,,	H		
VIII–32	$-OCO(CH_2)_4COO-$	H		$-CO(CH_2)_4CO-$
VIII–33	,,	H	,,	
VIII–34		H	,,	$-CO(CH_2)_4CO-$
VIII–35	,,	H	,,	

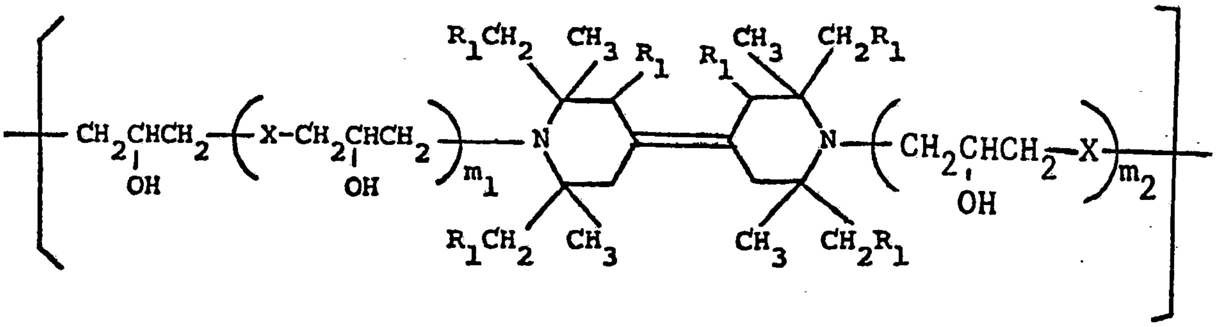
No.	X	R_1	R_3	G
VIII–36	$-OCH_2CH_2O-$	H		 $-CH_2\underset{OH}{\underset{\vert}{C}}HCH_2OCH_2CH_2OCH_2\underset{OH}{\underset{\vert}{C}}HCH_2-$
VIII–37		CH_3	,,	$-CO(CH_2)_4CO-$

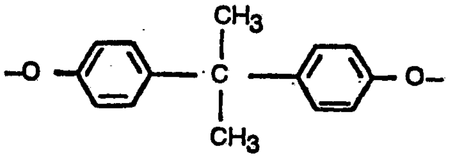
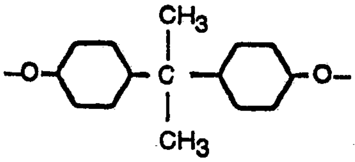
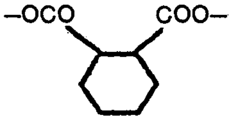


No.	X	R_1
IX–1		H
IX–2		H
IX–3		H
IX–4	$-OCO(CH_2)_8COO-$	H
IX–5		H
IX–6	$-OCH_2CH_2O-$	H
IX–7		CH_3



No.	X	R_1
X–1		H
X–2		H
X–3	$-OCO(CH_2)_4COO-$	H
X–4		H
X–5	$-OCH_2CH_2O-$	H



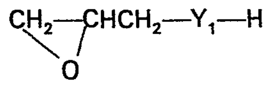
No.	X	R_1
XI–1		H
XI–2		H
XI–3	$-OCO(CH_2)_4COO-$	H
XI–4		H
XI–5	$-OCH_2CH_2O-$	H

The polymers of the invention having formula (I) may be either homopolymers or copolymers. In other words, each of W, X and Y may be all the same, partly different, or all different in their structures. In some instances, copolymers give better physical and/or stabilizing properties than homopolymers.

Polymers having the above-mentioned recurring units wherein a part or all of the hydroxy groups of the polymer are etherified, esterified or converted into carbamate groups are also representative polymers of the invention. Preferred such polymers with Z ≠ hydrogen are, e.g. methyl ethers, ethyl ethers, acetates, propionates, benzoates, N-methyl carbamates, N-ethyl carbamates, N-phenyl carbamates and N-cyclohexyl carbamates.

The polymers of formula (I) according to the invention may be prepared by any one of the following methods which are performed under conditions known *per se*.

1) Polymers in which Z is a hydrogen atom and both m_1 and m_2 are O may be prepared by polymerizing an epoxy compound having the formula



$$\underset{\diagdown O \diagup}{CH_2—CH}CH_2—Y_1—H$$

(XII)

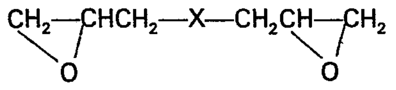
in which Y_1 is a group of formulae (II) to (IV), with the proviso that the 2,3-epoxypropyl group is linked to the nitrogen atom at 3-position, the oxygen atom at 4-position or the oxygen atom of —CH_2O— in formulae (II), (III) and (IV), respectively.

The reaction is performed preferably by heating the starting compound (XII) at 60 to 230°C in the presence or absence of an inert solvent. Suitable solvents are, e.g., ethers such as dioxane or diethyleneglycol dimethyl ether, dialkylamides such as dimethylformamide or dimethylacetamide; chlorinated or non-chlorinated aromatic hydrocarbons such as benzene, toluene, xylene, chlorobenzene or p-dichlorobenzene; and alcohols or mixtures thereof with water such as methanol, ethanol, n- or tert-butanol, n-octanol, aqueous methanol or aqueous ethanol.

In performing the reaction, there may be employed a catalyst, if necessary. Suitable catalysts are for instance those used for curing epoxy resins, e.g. tertiary amines such as triethylamine, triethanolamine or 1,5-diazabicyclo-[5.4.0]undecene-5 (DBU) or Lewis acids such as boron trifluoride or zinc borofluoride.

The starting compounds (XII) may be prepared by reacting a compound of formula H—Y_1—H with one mole equivalent of an epihalohydrin.

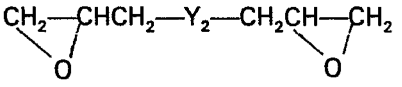
2) Polymers of formula (I) in which Z is a hydrogen atom and one of m_1 and m_2 is 1 and the other is 0 may be prepared by polymerizing an epoxy compound of formula



$$\underset{\diagdown O \diagup}{CH_2—CH}CH_2—X—CH_2\underset{\diagdown O \diagup}{CH—CH_2}$$

(XIII)

with a piperidine compound of formula H—Y_2—H, or by polymerizing an epoxy compound containing a piperidine of formula



$$\underset{\diagdown O \diagup}{CH_2—CH}CH_2—Y_2—CH_2\underset{\diagdown O \diagup}{CH—CH_2}$$

(XIV)

with a compound H—X—H in which Y_2 is an above-defined group of formula (V) to (XI) and X has the meaning defined above.

The reaction is performed under similar conditions as in the aforementined process variant 1).

The starting compounds (XIV) may be prepared by reacting a compound of formula H—Y_2—H with two mole equivalents of an epihalohydrin.

3) Compounds of formula (I) in which a part or all of Z are other than hydrogen may be prepared by introducing a substituent into a compound in which Z is a hydrogen atom and obtained in accordance with the above-mentioned process variants 1) or 2), by the methods described below:

3-a) Compounds in which Z is an alkyl group may be prepared by reacting the corresponding compound in which Z is a hydrogen atom with a strong basic alkali metal compound such as sodium hydride or potassium tert-butyrate and then with the desired alkyl halide.

3-b) Compounds in which Z is an acyl group may be prepared by reacting the corresponding compound in which Z is a hydrogen atom with a reactive derivative of the corresponding carboxylic acid. As reactive derivatives acid halides, acid lower alkyl esters or acid anhydrides, preferably acid anhydrides, may be employed.

3-c) Compounds in which Z is an N-substituted carbamoyl group may be prepared by reacting the corresponding compound in which Z is a hydrogen atom with the corresponding isocyanate.

The polymers of formula (I) according to the invention can effectively stabilize a wide variety of synthetic polymers, such as:

olefin and diene polymers, including homopolymers of olefins and dienes (e.g. low-density, highdensity and cross-linked polyethylenes, polypropylene, polyisobutylene, polymethylbutene-1, polymethylpentene-1, polyisoprene and polybutadiene), mixtures of such homopolymers (e.g. mixtures of polypropylene and polyethylene, polypropylene and polybutene-1, or polypropylene and polyisobutylene), and copolymers of olefins and dienes (e.g. ethylene/propylene copolymers, propylene/butene-1 copolymers, propylene/isobutylene copolymers, ethylene/butene-1 copolymers, and terpolymers of ethylene and propylene with dienes such as hexadiene, dicyclopentadiene or ethylidene norbornene);

styrene polymers, including polystyrene, copolymers of styrene and of α-methylstyrene (e.g. styrene/butadiene copolymers, styrene-acrylonitrile copolymers, styrene/acrylonitrile/methylmethacrylate copolymers, styrene/acrylonitrile/acrylic ester copolymers, styrene/acrylonitrile copolymers modified with acrylic ester polymers to provide impact strength, and styrene polymers modified with ethylene/propylene/diene elastomers to provide impact strength), and graft copolymers of styrene (e.g. polymers in which styrene is grafted onto polybutadiene, and polymers in which styrene and acrylonitrile are grafted onto polybutadiene as well as mixtures thereof with the aforementioned styrene copolymers, commonly known as acrylonitrile/butadiene/styrene or ABS plastics);

halogenated vinyl and vinylidene polymers, including polyvinyl chloride, polyvinylidene chloride, polyvinyl fluoride, polychloroprene, chlorinated rubbers, vinyl chloride/vinylidene chloride copolymers, vinyl chloride/vinyl acetate copolymers, and vinylidene chloride/vinyl acetate copolymers;

polymers derived from α,β-unsaturated acids, and derivatives thereof, including polyacrylates and polymethacrylates, polyacrylic amides and polyacrylonitrile;

polymers derived from unsaturated alcohols and amines, and from the acyl derivatives thereof or acetals, including polyvinyl alcohol, polyvinyl acetate, polyvinyl stearate, polyvinyl benzoate, polyvinyl maleate, polyvinyl butyral, polyallyl phthalate, and polyallyl melamine, and copolymers thereof with other ethylenically unsaturated monomers (e.g. ethylene/vinyl acetate copolymers);

epoxy polymers, including homopolymers and copolymers derived from epoxides (e.g. polyethylene oxide), and polymers derived from bis-glycidyl ethers;

polyacetals, polyalkylene oxides and polyphenylene oxides, including polyoxymethylene, oxymethylene/ethylene oxide copolymers, polyoxyethylene, polypropylene oxide, polyisobutylene oxide and polyphenylene oxides;

polyurethane and polyureas;

polycarbonates;

polysulphones;

polyamides and copolyamides derived from diamines and dicarboxylic acids and/or from amino-carboxylic acids or the corresponding lactams, including nylon-6, nylon-6,6, nylon-6,10, nylon-11 and nylon-12;

polyesters derived from dicarboxylic acids and dialcohols and/or from hydroxycarboxylic acids and the corresponding lactones, e.g. polyethylene glycol terephthalate and poly-1,4-dimethylol-cyclohexane terephthalate;

crosslinked polymers derived from aldehydes together with phenols, ureas or melamines, e.g. phenol/formaldehyde, urea/formaldehyde and melamine/formaldehyde resins;

alkyd resins, e.g. glycerol/phthalic acid resins and mixtures thereof with melamine/formaldehyde resins;

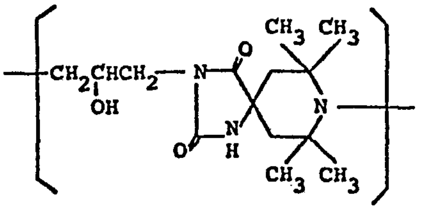
unsaturated polyester resins derived from copolyesters of saturated and unsaturated dicarboxylic acids with polyhydric alcohols as well as from vinyl compounds as cross-linking agents, and also halogenated flame-resistant modifications thereof.

The amount of stabilizers of the invention needed for effective stabilization of organic polymers will depend on a variety of factors, such as the type and properties of the polymer concerned, its intended use, and the presence of other stabilizers. It is generally satisfactory to use from 0.01 to 5.0% by weight of the stabilizers of the invention, based on the weight of the polymer, but the most effective range will vary with the type of the polymer; viz. 0.01% to 2.0%, preferably 0.02% to 1.0%, by weight for olefin, diene and styrene polymers; 0.01% to 1.0%, preferably 0.02% to 0.5%, by weight for vinyl and vinylidene polymers; and 0.01% to 5.0%, preferably 0.02% to 2.0%, by weight for polyurethanes and polyamides. If desired, two or more of the polymeric stabilizers of the invention may be used together.

The polymeric stabilizers of the invention may readily be incorporated into synthetic polymers to be stabilized by conventional techniques at any convenient stage prior to the manufacture of shaped articles therefrom. For example, the polymeric stabilizer may be mixed with the polymer to be stabilized in dry powder form, or a suspension or emulsion of the stabilizer may be mixed with a solution, suspension or emulsion of the polymer to be stabilized.

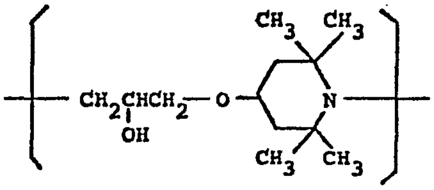
The stabilized polymeric composition of the invention may optionally also contain one or more of various additives conventionally used in polymer technology, such as the additives listed in British Patent Specification no. 1,401,924, at pages 11—13. In the following Examples the mean molecular weight was measured by the vapour pressure depression method.

Example 1



1.4 g of 3-(2,3-epoxypropyl)-7,7,9,9-tetramethyl-1,3,8-triazaspiro[4.5]decane-2,4-dione were heated at 180—190°C for 20 minutes in the absence of a solvent. Ethyl acetate was added to the reaction mixture, and insoluble residues were filtered off. A precipitate formed by adding n-hexane to the ethyl acetate solution, and said precipitate was collected by filtration and dried under reduced pressure, giving a white powder (Compound no. 1) softening at 205—208°C and having a mean molecular weight of 1,350.

Example 2

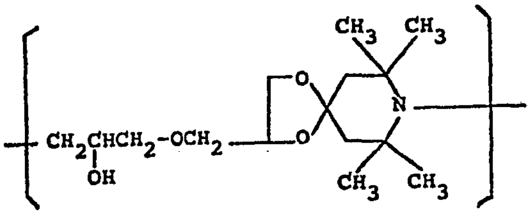


3.0 g of 4-(2,3-epoxypropoxy)-2,2,6,6-tetramethylpiperidine were heated at 210—220°C for 5 hours under an argon stream. Upon cooling, the reaction product solidified, yielding a vitreous solid which was triturated giving a pale yellow powder (Compound no. 2) softening at 140—145°C and having a mean molecular weight of 3,800.

Example 3

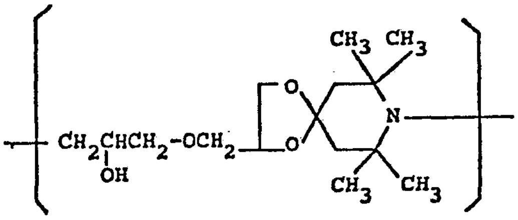
Acetate of compound no. 2. A mixture of 1.0 g of the pale yellow powder obtained according to Example 2 and 10 g of acetic anhydride was heated at 100—110°C for 5 hours under a nitrogen stream. The reaction mixture was poured into ice-water, neutralized with aqueous ammonia and extracted with ethyl acetate. The organic phase was washed with water and dried over anhydrous magnesium sulfate. The ethyl acetate was removed by evaporation under reduced pressure, yielding the desired compound (Compound no. 3) softening at 103—106°C. Examination of the compound by infrared and NMR spectrometry revealed that all of the hydroxy groups of the compound were acetylated.

Example 4



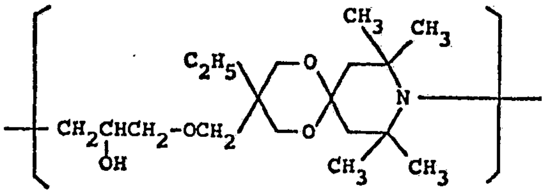
3.0 g of 2-(2,3-epoxypropoxymethyl)-7,7,9,9-tetramethyl-1,4-dioxa-8-azaspiro[4.5]decane were heated in a similar manner as in Example 2. Further treatment of the reaction product as described in said Example 2 yielded the desired compound (Compound no. 4) as a pale yellow powder softening at 128—135°C and having a mean molecular weight of 14,000. By heating the starting material to 210—220°C for 2 hours under an argon stream and treating the reaction product as described in Example 2, a pale yellow powder (Compound no. 5) was obtained, softening at 80—85°C and having a mean molecular weight of 2,200.

Example 5



To 6.0 g of 2 - hydroxymethyl - 8 - (2,3 - epoxypropyl) - 7,7,9,9 - tetramethyl - 1,4 - dioxa - 8 - azaspiro[4.5]decane were added 20 ml of 10% aqueous potassium hydroxide solution and the mixture was refluxed for 5 hours while stirring. The reaction mixture was then extracted with benzene and the benzene solution was washed with water and dried over potassium carbonate. The residue obtained by removing the solvent was purified by column chromatography (silica gel; ethyl acetate: triethylamine = 19:1), giving the desired compound (Compound no. 6) as a pale yellow viscous oily product having a mean molecular weight of 600.

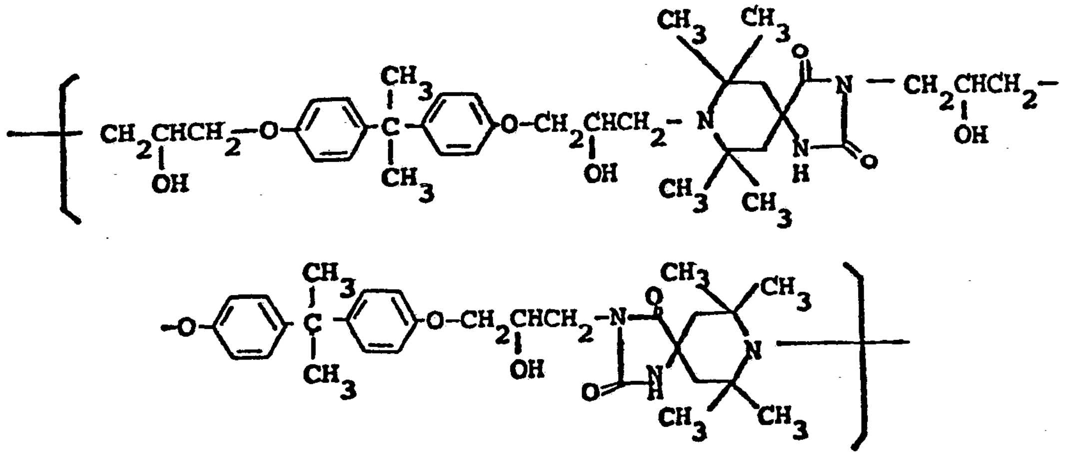
Example 6



5.0 g of 3 - ethyl - 3 - (2,3 - epoxypropoxymethyl) - 8,8,10,10 - tetramethyl - 1,5 - dioxa - 9 - azaspiro[5.5]undecane were heated to 220—230°C for 10 hours under an argon stream. The reaction mixture was then treated in a manner similar to that described in Example 2, yielding the desired compound (Compound no. 7) as a pale yellow powder softening at 130—140°C and having a mean molecular weight of 14,000.

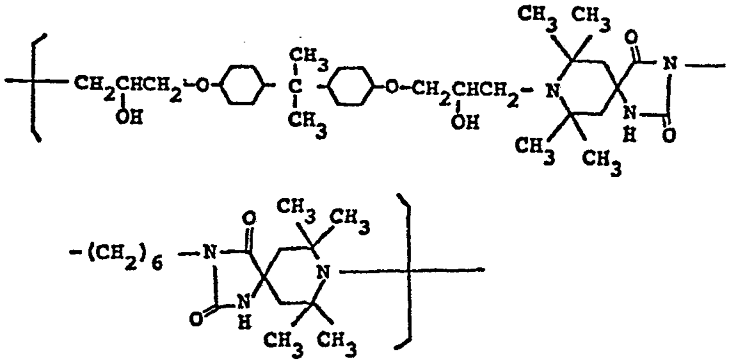
By heating the above-mentioned starting material to 210—220°C for 2 hours under an argon stream and treating the reaction product as described in Example 2, the desired compound (Compound no. 8) was obtained in the form of a white powder softening at 55—60°C and having a mean molecular weight of 1,200.

Example 7



7.9 g of 2,2-bis(p-[2-hydroxy-3-(7,7,9,9-tetramethyl-2,4-dioxo-1,3,8-triazaspiro[4.5]dec-3-yl)propoxy]phenyl)propane and 3.4 g of 2,2-bis[p-(2,3-epoxypropoxy)phenyl]propane were heated to 180—190°C for 20 minutes in the absence of a solvent. The reaction mixture was washed with ethyl acetate and dried under reduced pressure, giving the desired compound (Compound no. 9) as a white powder softening at 175—180°C and having a mean molecular weight of 3,200.

Example 8

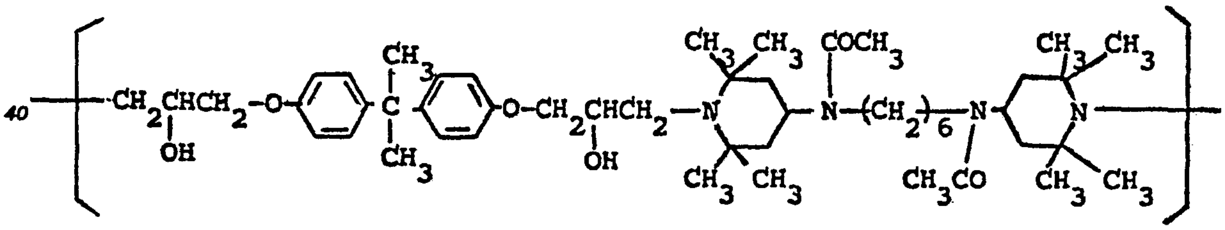


5.0 g of 3,3′-hexamethylene-bis-(7,7,9,9-tetramethyl-1,3,8-triazaspiro[4.5]decane-2,4-dione) and 3.3 g of 2,2-bis[4-(2,3-epoxypropoxy)cyclohexyl]propane were heated at 230°C for 3 hours under an argon stream. The reaction mixture was dissolved in chloroform and the solution was poured into n-hexane, whereby a precipitate was formed. The precipitate was collected by filtration and dried under reduced pressure, giving the desired compound (Compound no. 10) in the form of a white powder softening at 139—141°C and having a mean molecular weight of 1,900.

Example 9

Benzoate of Compound no. 10. 2.0 g of the white powder obtained according to Example 8 and 8.0 g of benzoic anhydride were heated to 150°C for 15 hours under argon. The reaction mixture was dissolved in ethyl acetate, washed with 5% aqueous potassium carbonate solution and water, and dried over sodium sulfate. The residue obtained by removing the solvent was purified by column chromatography. At first, elution with ethyl acetate gave a white powder (Compound no. 11) softening at 115—120°C. The NMR spectrum of the compound showed that the benzoylation rate was 95%. A second elution with ethyl acetate:ethanol:triethylamine = 20:1:1, gave a greyish white powder (Compound no. 12) softening at 115—118°C. The NMR spectrum of the compound showed that the benzoylation rate was 45%.

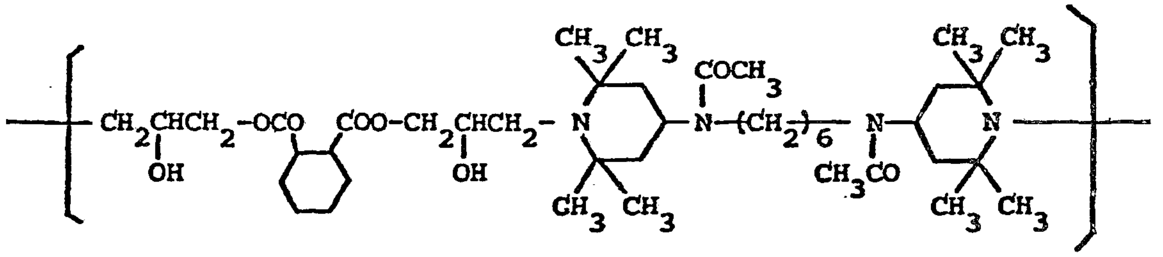
Example 10



4.8 g of N,N′-hexamethylene bis[N-(2,2,6,6-tetramethyl-4-piperidyl)acetamide and 3.4 g of 2,2-bis[p-(2,3-epoxypropoxy)phenyl]propane in 25 ml of n-octanol were refluxed at 180—200°C for 8 hours. n-Octanol was removed under reduced pressure and the residue was purified by column chromatography (silica gel; ethyl acetate: triethylamine: methanol = 8:1:1) giving a white powder (Compound no. 13) softening at 108—115°C and having a mean molecular weight of 2,600 from the first eluate, and a white powder (Compound no. 14) softening at 90—110°C and having a mean molecular weight of 1,650 from the second eluate.

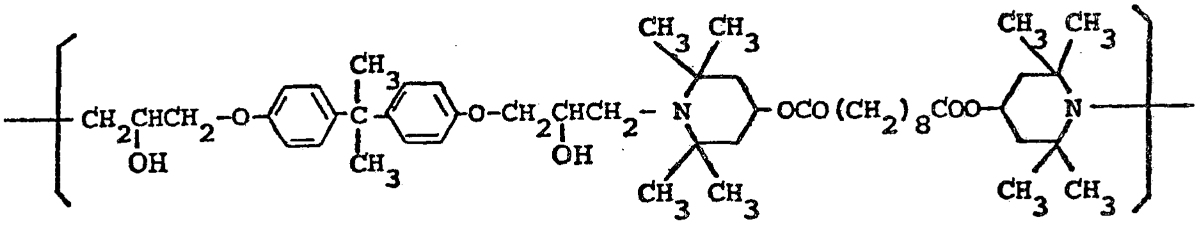
The above-mentioned starting material was heated to 200—220°C for 3 hours in the absence of a solvent and then treated and purified in a similar manner as described above, giving a white powder (Compound no. 15) softening at 150—160°C and having a mean molecular weight of 6,000 from the first eluate, and a white powder (Compound no. 16) softening at 120—130°C and having a mean molecular weight of 2,650 from the second eluate.

Example 11



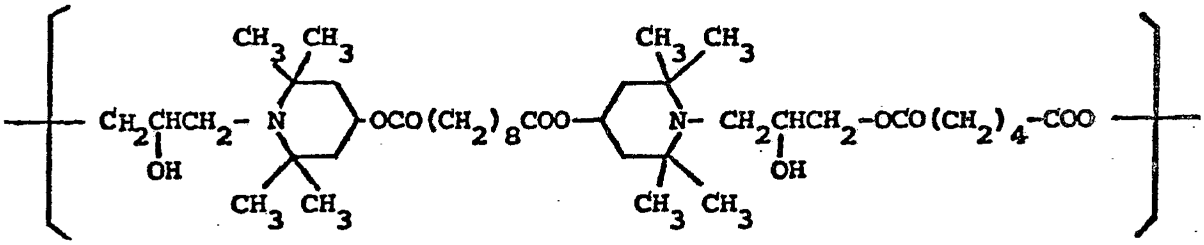
4.8 g of N,N'-hexamethylenebis[N-(2,2,6,6-tetramethyl-4-piperidyl)acetamide] and 2.8 g of bis(2,3-epoxypropyl)-1,2-cyclohexanedicarboxylate were heated to 200°C for 5 hours under a nitrogen stream. The reaction mixture was purified by column chromatography (silica gel; ethyl acetate:triethylamine:methanol = 8:1:1) and the crude product thus obtained was dissolved in benzene. The benzene solution was poured into n-hexane and the precipitate formed was collected by filtration, giving a white powder (Compound no. 17) softening at 115—125°C and having a mean molecular weight of 2,700.

Example 12



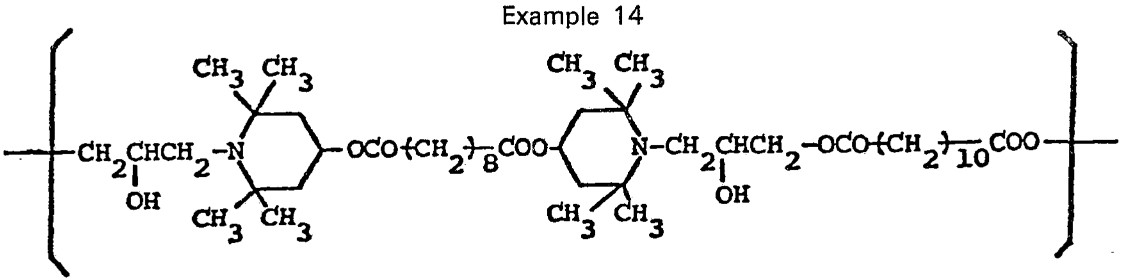
3.0 g of bis(2,2,6,6-tetramethyl-4-piperidyl)sebacate and 2.1 g of 2,2-bis[p-(2,3-epoxypropoxy)phenyl]propane were heated to 190—200°C for 4 hours under argon. The reaction mixture was purified by column chromatography (silica gel; ethyl acetate: triethylamine = 20:1), giving a pale yellow solid product (Compound no. 18) softening at 85—90°C and having a mean molecular weight of 2,500 from the first eluate, and a pale yellow solid product (Compound no. 19) softening at 74—77°C and having a mean molecular weight of 1,900 from the second eluate.

Example 13



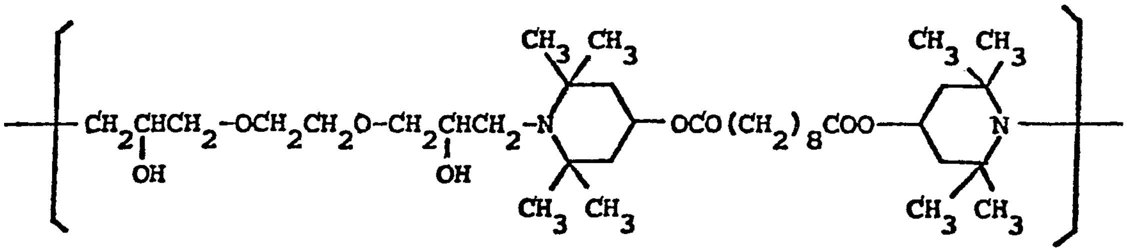
2.2 g of bis[1-(2,3-epoxypropyl)-2,2,6,6-tetramethyl-4-piperidyl]sebacate and 0.55 g of adipic acid were refluxed in 5 mol of n-butanol for 24 hours. The n-butanol was removed from the reaction mixture and the residue was washed with n-hexane and then treated with ethyl acetate. The ethyl acetate soluble fraction was purified by column chromatography (silica gel: ethyl acetate: ethanol = 4:1), giving the desired compound (Compound no. 20) as a pale yellow oil having a mean molecular weight of 1,600.

Example 14



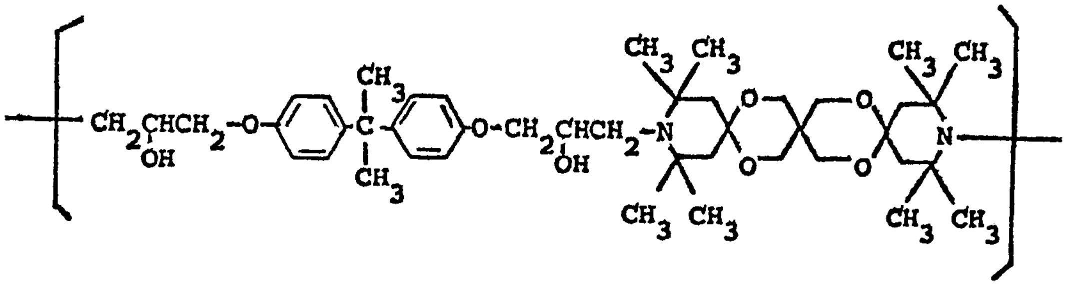
2.7 g of bis[1-(2,3-epoxypropyl)-2,2,6,6-tetramethyl-4-piperidyl]sebacate and 0.95 g of 1,10-decanedicarboxylic acid were refluxed in 6 ml of n-amyl alcohol for 17 hours. The reaction mixture was then treated in a similar manner as described in Example 12, yielding the desired compound (Compound no. 21) as a pale yellow oil having a mean molecular weight of 1,820.

Example 15



3.0 g of bis(2,2,6,6-tetramethyl-4-piperidyl)sebacate and 1.1 g of ethyleneglycol diglycidyl ether were heated at 200—200°C for 8 hours under an argon stream. The reaction mixture was purified by column chromatography (silica gel; ethyl acetate: ethanol:triethylamine = 20:1:1), giving a pale yellow viscous oil (Compound no. 22) having a mean molecular weight of 3,000 from the first eluate, and a pale yellow viscous oil (Compound no. 23) having a mean molecular weight of 2,300 from the second eluate.

Example 16

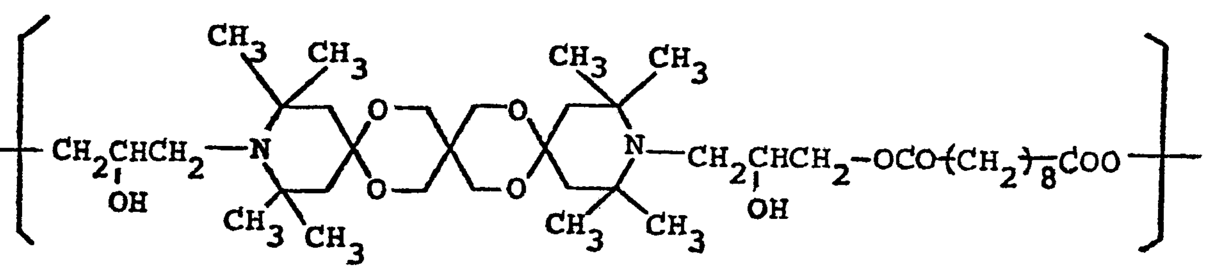


4.4 g of 2,2,4,4,14,14,16,16 - octamethyl - 7,11,18,21 - tetraoxa - 3,15 - diazatrispiro - [5.2.2.5.2.2]heneicosane and 3.4 g of 2,2-bis[p-(2,3-epoxypropoxy)phenyl]propane were heated to 180°C for 8 hours under a nitrogen stream. The reaction mixture was purified by column chromatography (silica gel; ethyl acetate: triethylamine = 9:1), and the product was dissolved in benzene and precipitated with n-hexane, yielding the desired compound (Compound no. 24) as a white powder softening at 180—200°C and having a mean molecular weight of 3,450.

Example 17

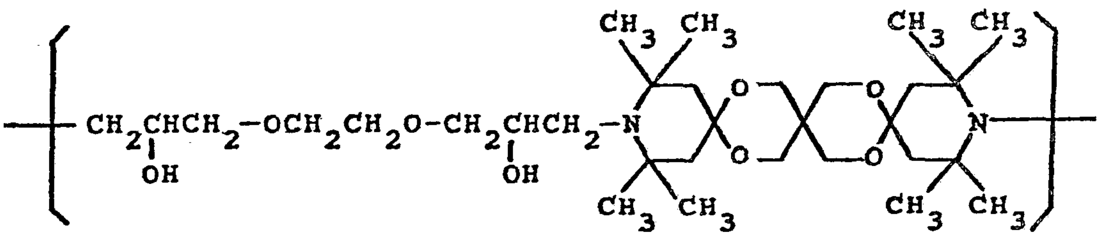
Acetate of Compound no. 24. 2 g of the white powder obtained according to Example 16 and 10 g of acetic anhydride were refluxed in 20 ml of benzene for 5 hours. The reaction mixture was washed with 10% aqueous potassium carbonate solution, and the benzene layer was dried over potassium carbonate. The residue obtained by removing the solvent was purified by column chromatography (silica gel; ethyl acetate) and the obtained crude product was dissolved in benzene. By pouring the benzene solution into n-hexane, a precipitate was formed, which precipitate was collected by filtration, yielding a white powder (Compound no. 25) softening at 155—175°C. The IR and NMR spectra showed that the hydroxy groups of the polymer had been completely acetylated.

Example 18



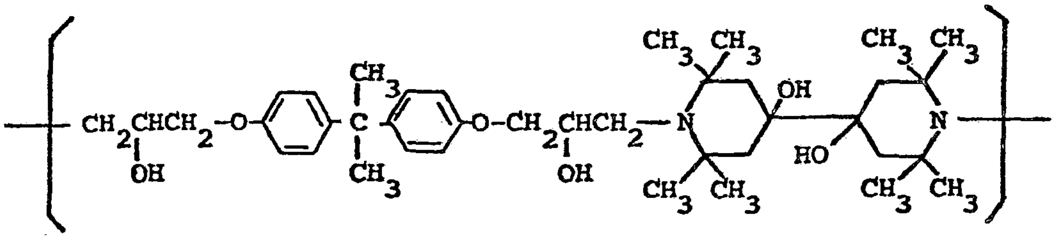
2.1 g of 3,15-bis(2,3-epoxypropyl)-2,2,4,4,14,14,16,16-octamethyl-7,11,18,21-tetraoxa-3,15-diaza-trispiro[5.2.2.5.2.2]heneicosane and 0.8 g of sebacic acid were refluxed in a mixture of 10 ml of tert-butanol and 10 ml of toluene for 14 hours. After removal of the solvent under reduced pressure, the residue was purified by column chromatography (silica gel; ethyl acetate: methanol = 1:1), giving the desired compound (Compound no. 26) as a white powder softening at 149—158°C and having a mean molecular weight of 5,100.

Example 19



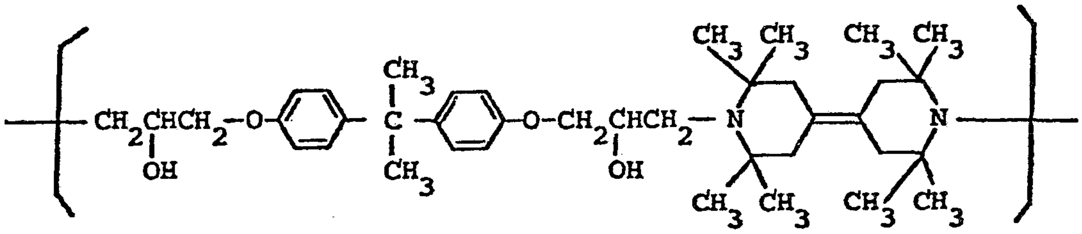
4.1 g of 2,2,4,4,14,14,16,16 - octamethyl - 7,11,18,21 - tetraoxa - 3,15 - diazatrispiro - [5.2.2.5.2.2.]heneicosane and 1.74 g of ethyleneglycol diglycidyl ether were heated to 200°C in 4 ml of octanol for 20 hours while stirring. After removal of the octanol under reduced pressure, the residue was purified by column chromatography (silica gel; ethyl acetate:methanol:triethylamine = 2:2:1), giving the desired compound (Compound no. 27) as a pale yellow softening at 140—154°C and having a mean molecular weight of 2,770.

Example 20



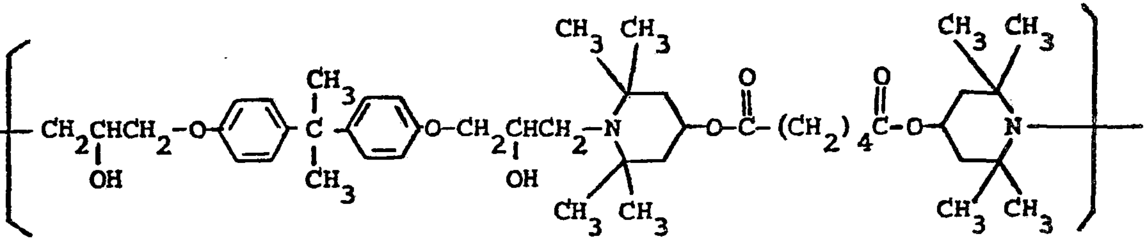
6.4 g of 4,4′-bis(2,2,6,6-tetramethyl-4-piperidinol) and 6.8 g of 2,2-bis[p-(2,3-epoxypropoxy)phenyl]propane were heated in 80 ml of octanol to 150—160°C for 8 hours while stirring. After removal of the octanol under reduced pressure, the remaining vitreous mass was triturated, giving the desired compound (Compound no. 28) as a white powder softening at 106—109°C and having a mean molecular weight of 1,850.

Example 21



1.5 g of 2,2,2′,2′,6,6,6′,6′-octamethyl-4,4′-bipiperidylydene and 1.8 g of 2,2-bis[p-(2,3-epoxypropoxy)phenyl]propane were heated to 180—220°C for 2 hours under an argon stream. The reaction mixture was dissolved in chloroform, methanol was added thereto whereupon a precipitate formed. The precipitate was collected by filtration, washed with methanol and dried under reduced pressure, giving the desired compound (Compound no. 29) as a white solid softening at 170—180°C and having a mean molecular weight of 5,300.

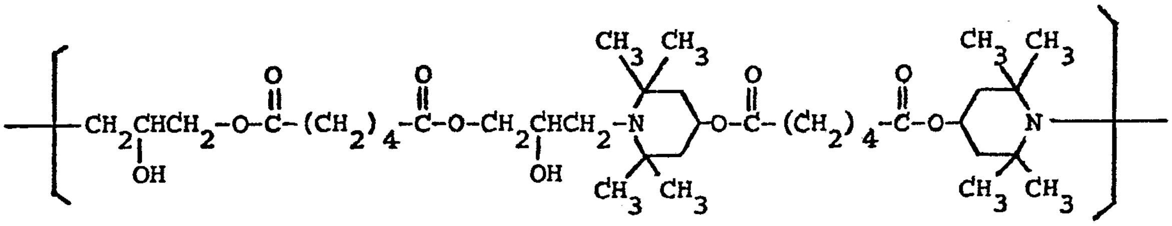
Example 22



A mixture of 5.24 g of bis(2,2,6,6-tetramethyl-4-piperidyl)adipate and 4.2 g of 2,2-bis[p-(2,3-

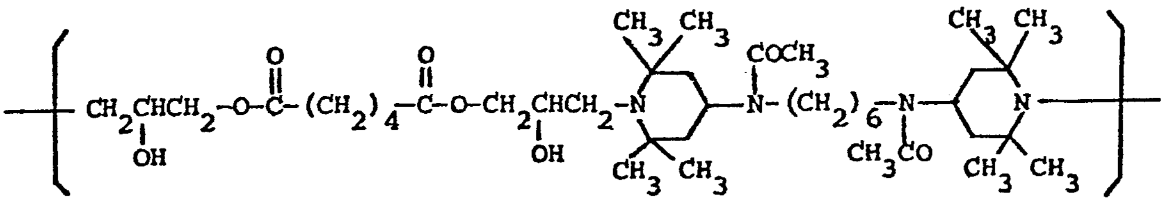
epoxypropoxy)phenyl]propane was heated to 160—170°C for 6 hours under a nitrogen stream. The reaction mixture was purified by column chromatography through silica gel eluted with ethyl acetate, giving the desired compound (Compound no. 30) in the form of a white powder softening at 95—115°C and having a mean molecular weight of 3,700.

Example 23



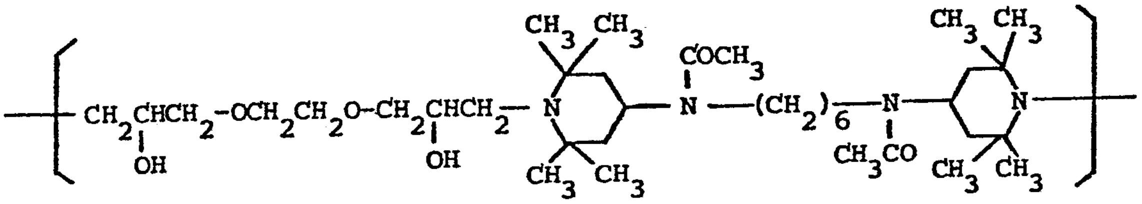
A mixture of 8.5 g of bis (2,2,6,6-tetramethyl-4-piperidyl)adipate and 5.2 g of bis(2,3-epoxypropyl) 1,4-butanedicarboxylate was heated to 160—170°C for 4 hours under a nitrogen stream. The reaction mixture was dissolved in ethyl acetate and the solution was poured into n-hexane yielding an oily substance which was insoluble in n-hexane. The oily substance was dried under reduced pressure, giving the desired compound (Compound no. 31) in the form of a colourless solid softening at 97—115°C and having a mean molecular weight of 3,700.

Example 24



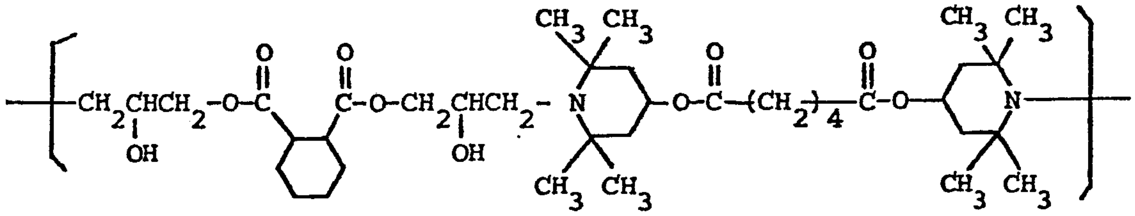
A mixture of 4.8 g of N,N'-hexamethylenebis[N-(2,2,6,6-tetramethyl-4-piperidyl)acetamide] and 2.6 g of bis(2,3-epoxypropyl) 1,4-butanedicarboxylate was heated to 200°C for 3 hours under a nitrogen stream. The reaction mixture was washed with hot n-hexane and dried under reduced pressure, yielding the desired compound (Compound no. 32) in the form of a pale yellow powder softening at 50—60°C and having a mean molecular weight of 2,020.

Example 25



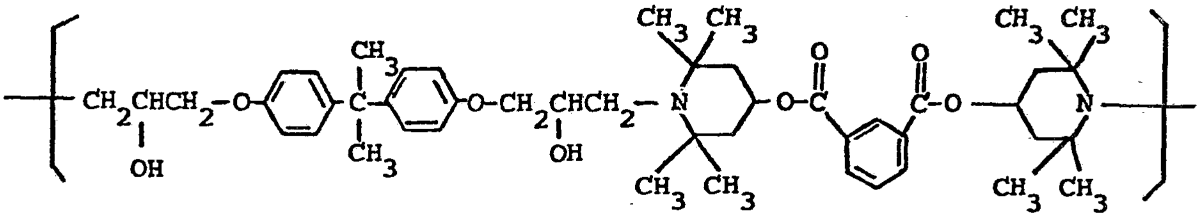
A mixture of 4.8 g of N,N'-hexamethylenebis[N-(2,2,6,6-tetramethyl-4-piperidyl)acetamide] and 1.74 g of ethylene glycol diglycidyl ether was heated to 250°C for 1 hour under a nitrogen stream. The reaction mixture was dissolved in benzene and the solution was poured into n-hexane to give a precipitate. The precipitate was collected by filtration and dried under reduced pressure, affording the desired compound (Compound no. 33) in the form of a pale yellow powder softening at 85—95°C and having a mean molecular weight of 2,360.

Example 26



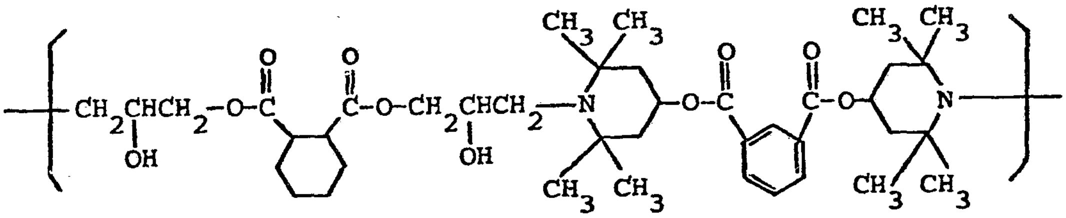
A mixture of 4.25 g of bis(2,2,6,6-tetramethyl-4-piperidyl)adipate and 2.84 g of bis(2,3-epoxypropyl) 1,2-cyclohexane-dicarboxylate was heated to 170—180°C for 16 hours under an argon stream. The reaction mixture was dissolved in benzene and the solution was poured into n-hexane to give a precipitate. The precipitate was collected by filtration and dried under reduced pressure, giving the desired compound (Compound no. 34) in the form of a colourless solid softening at 89—95°C and having a mean molecular weight of 4,530.

Example 27



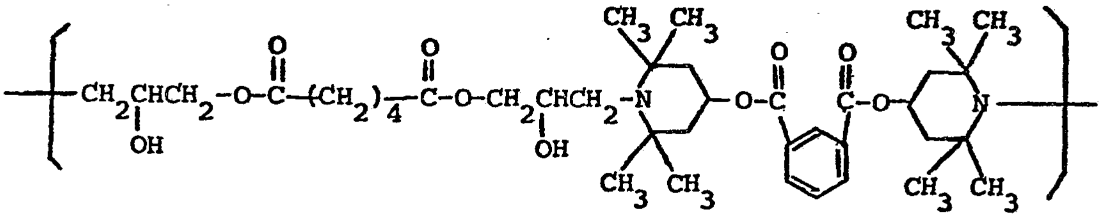
A mixture of 3.0 g of bis(2,2,6,6-tetramethyl-4-piperidyl)isophthalate and 2.3 g of bis[p-(2,3-epoxypropoxy)phenyl]propane was heated to 180°C for 7 hours under a nitrogen stream. The reaction mixture was dissolved in benzene and the solution was poured into n-hexane to give a precipitate. The precipitate was collected by filtration and dried under reduced pressure to give the desired compound (Compound no. 35) in the form of a white powder softening at 155—165°C and having a mean molecular weight of 4,270.

Example 28



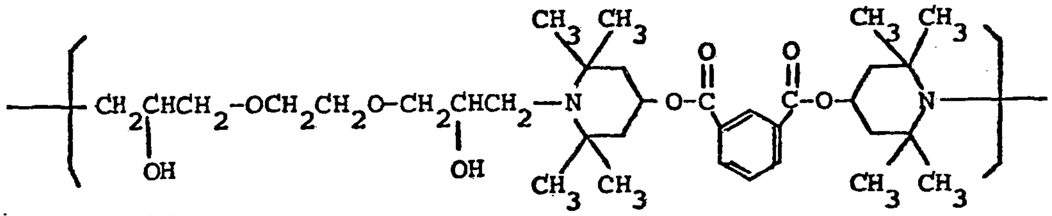
A mixture of 3.5 g of bis(2,2,6,6-tetramethyl-4-piperidyl)isophthalate and 2.2 g of bis(2,3-epoxypropyl) 1,2-cyclohexanedicarboxylate was heated to 180°C for 7 hours under a nitrogen stream. The reaction mixture was dissolved in benzene and the solution was poured into n-hexane to give a precipitate. The precipitate was collected by filtration and dried under reduced pressure, yielding the desired compound (Compound no. 36) in the form of a white powder softening at 94—104°C and having a mean molecular weight of 1,660.

Example 29



A mixture of 3.5 g of bis(2,2,6,6-tetramethyl-4-piperidyl)isophthalate and 2.0 g of bis(2,3-epoxypropoxy)adipate was heated to 180°C for 7 hours under a nitrogen stream. The reaction mixture was dissolved in benzene and the solution was filtered and poured into n-hexane to give a precipitate. The precipitate was collected by filtration and dried under reduced pressure to give the desired compound (Compound no. 37) in the form of a pale yellow powder softening at 90—100°C and having a mean molecular weight of 3,000.

Example 30



A mixture of 3.5 g of bis (2,2,6,6-tetramethyl-4-piperidyl)isophthalate and 1.4 g of ethylene glycol diglycidyl ether was heated to 180°C for 7 hours under a nitrogen stream. The reaction mixture was dissolved in benzene and the solution was poured into n-hexane to give a precipitate. The precipitate was collected by filtration and dried under reduced pressure, yielding the desired compound (Compound no. 38) in the form of a pale yellow powder softening at 98—105°C and having a mean molecular weight of 3,200.

Example 31

Stabilisation of polypropylene. Mixtures were prepared from 100 parts of unstabilized polypropylene powder (melt flow index = 18), 0.2 parts of stearyl 3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate (a conventional antioxidant) and 0.25 parts of each of the stabilizers, listed in the following Table 1. [All parts are parts by weight]. The mixtures were blended and homogenized with a Brabender Plastograph at 200°C for 10 minutes and the obtained mass was pressed into a sheet of 2—3 mm thickness with a laboratory press. The sheet was then pressed in a hydraulic press at 260°C for 6 minutes (12 tons) and put immediately into cold water to form a film of 0.5 mm thickness, from which a film of 0.1 mm thickness was obtained by the same procedure.

The film was cut into test specimens of 50 x 120 mm. The test specimens were exposed to light in a Sunshine Weather Meter at a black panel temperature of 63±3°C and examined periodically to determine the percentage of elongation at break. From the test results the ratio of the time required for the test specimens containing stabilizer to reach 50% elongation at break was determined, to the time required for an unstabilized test specimen to reach 50% elongation at break. The results are summarized in Table 1.

TABLE 1

Compound No.	Ratio	Compound No.	Ratio
1	7.8	21	6.2
2	7.2	22	7.8
3	7.4	27	6.4
5	5.4	28	4.3
8	6.1	29	4.4
10	4.8	30	5.0
14	7.1	31	7.1
17	6.8	32	5.3
19	6.0	33	5.9
20	7.7	34	>6.7

Example 32

Stabilization of polystyrene.

Mixtures were prepared from 100 parts of polystyrene ("Styron 666", "Styron" is a Registered Trade Mark, manufactured and sold by Asahi Dow Co. Ltd.) and 0.25 parts of each of the stabilizers listed in the following Table 2. The resultant mixtures were blended and homogenized by means of a Brabender Plastograph at 200°C for 5 minutes, then compression-molded at 180°C for 2 minutes, forming sheets of 1.5 mm thickness. Each test sheet was exposed to light in a Xenon Weather-O-Meter (6.5 kw; ASTM G 26—77) for 1500 hours, and the yellowness index of the sheet before and after irradiation was determined in accordance with ASTM D 1925. The results are shown in Table 2.

TABLE 2

Compound No.	Yellowness Index	
	before irradiation	after irradiation
2	1.9	7.1
8	1.8	8.7
10	2.0	8.6
17	2.2	10.2
19	1.8	10.8
20	1.7	8.3
22	1.9	9.5
28	2.1	10.4
without stabilizer	1.8	35.2

Example 33

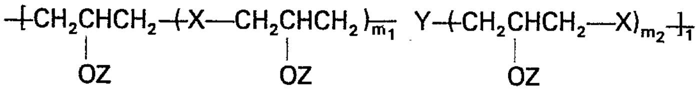
Stabilization of polyurethane. Mixtures were prepared from 10 g of polyurethane ("Paraprene 22S", trade name, manufactured and sold by Nippon Polyurethane Kogyo KK), and 0.05 g of each of the stabilizers given in the following Table 3. Each mixture was dissolved in 30 ml of dimethylformamide and the resulting solution was drawn on a glass plate to form a sheet of 0.4 mm thickness. The sheets were maintained at 60°C for 20 minutes and dried at 120°C for 15 minutes to cast films of 0.1 mm thickness. The films thus obtained were exposed to ultraviolet radiation in a Sunshine Carbon Arc Lamp Weather Meter (type "WEL-SUN-HC", trade name of Suga Test Instruments Co. Ltd.) for 300 hours and the yellowness index before and after irradiation was determined in accordance with ASTM D1925. The results are shown in Table 3.

TABLE 3

Compound No.	Yellowness Index	
	before irradiation	after irradiation
2	1.5	23.7
8	1.9	25.5
10	1.7	22.8
17	2.0	26.5
19	1.6	23.1
20	1.8	21.9
22	1.6	22.8
without stabilizer	1.5	47.6

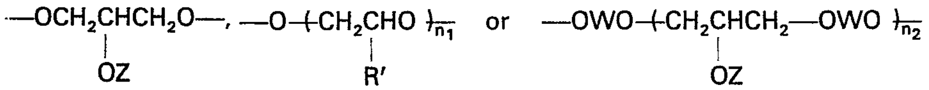
Claims

1. A polymer represented by the formula (I):



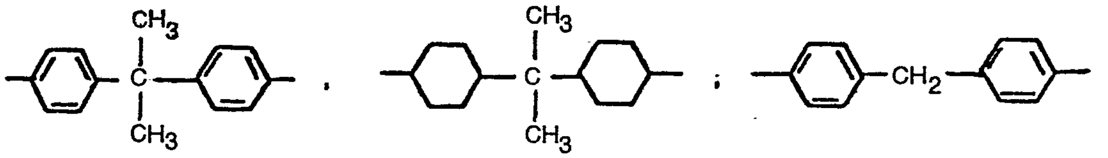
$$-\!\!\left[CH_2\underset{OZ}{C}HCH_2\left(X-CH_2\underset{OZ}{C}HCH_2\right)_{m_1}Y\left(CH_2\underset{OZ}{C}HCH_2-X\right)_{m_2}\right]_{l} \qquad (I)$$

wherein
l is an integer of from 2 to 50,
m_1 and m_2 are 0, or one of them is 1 and the other is 0,
X represents a group of formula

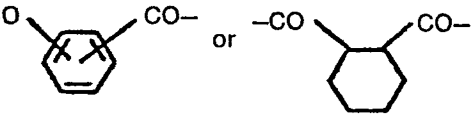


$$-OCH_2\underset{OZ}{C}HCH_2O-,\ -O\left(CH_2\underset{R'}{C}HO\right)_{n_1}\ \text{or}\ -OWO\left(CH_2\underset{OZ}{C}HCH_2-OWO\right)_{n_2}$$

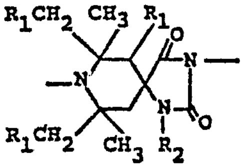
in which
R′ is a hydrogen atom or a methyl group,
n_1 is an integer of from 1 to 10,
n_2 is 0 or an integer of from 1 to 10,
W represents a group of formula



$-CO\left(CH_2\right)_{n_3}CO-$

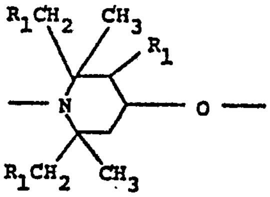


in which
n_3 is an integer of from 1 to 10, and
Z has the meaning defined below,
Y represents, when m_1 and m_2 are 0, a group of the following formulae II to IV:



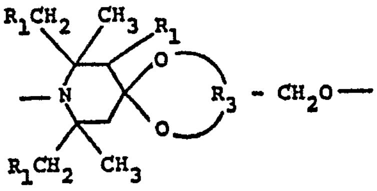
(II)

in which
R_1 represents a hydrogen atom or a methyl group,
R_2 represents a hydrogen atom or an alkyl group having from 1 to 18 carbon atoms,



(III)

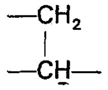
in which R_1 has the meaning defined above,



(IV)

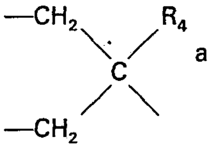
in which R_1 has the meaning defined above,

R_3 represents a group of formula



$$\underset{-\underset{}{CH}-}{-CH_2}$$

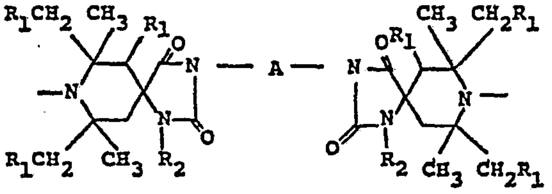
or



and

R_4 represents the methyl or ethyl group,

Y represents, when one of m_1 and m_2 is 1 and the other is 0, a group of the following formulae V to XI:

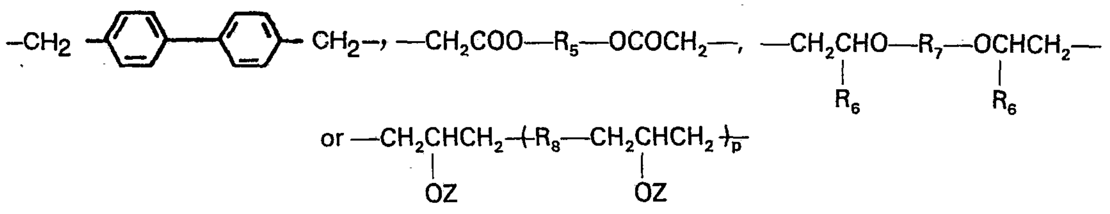


(V)

in which:

R_1 and R_2 have the meanings defined above,

A represents an alkylene group having from 2 to 12 carbon atoms, a xylylene group, a group of formula



$-CH_2COO-R_5-OCOCH_2-$, $-CH_2\underset{R_6}{CH}O-R_7-O\underset{R_6}{CH}CH_2-$

or $-CH_2\underset{OZ}{CH}CH_2\left(R_8-CH_2\underset{OZ}{CH}CH_2\right)_p$

in which

R_5 represents an alkylene group having from 2 to 8 carbon atoms,

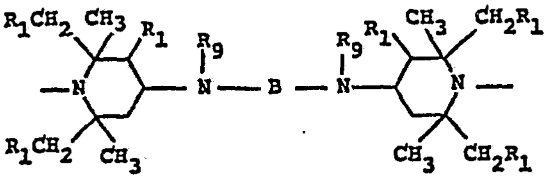
R_6 represents a hydrogen atom, the methyl or phenyl group,

R_7 represents an aliphatic, aromatic or alicyclic diacyl group having up to 12 carbon atoms,

R_8 has the same meaning as the above-defined X,

p is 0 or 1, and

Z has the meaning defined below,



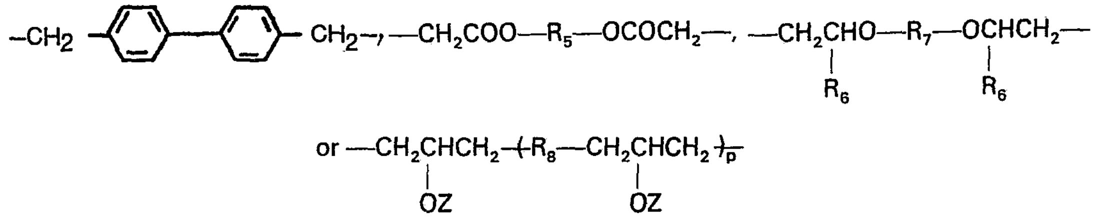
(VI)

in which:

R_1 has the meaning defined above,

R_9 represents an alkyl group having from 1 to 18 carbon atoms, a phenyl group, a benzyl group, a cyclohexyl group, or an aliphatic, aromatic, araliphatic or alicyclic acyl group having up to 18 carbon atoms,

B represents an alkylene group having from 2 to 10 carbon atoms, a xylylene group, a group of formula



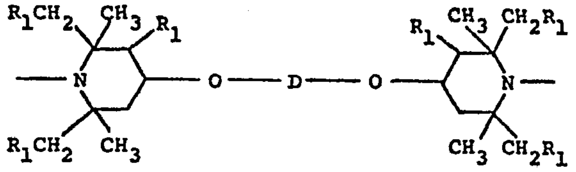
$-CH_2COO-R_5-OCOCH_2-$, $-CH_2\underset{R_6}{CH}O-R_7-O\underset{R_6}{CH}CH_2-$

or $-CH_2\underset{OZ}{CH}CH_2\left(R_8-CH_2\underset{OZ}{CH}CH_2\right)_p$

in which

R_5, R_6, R_7, R_8 and p have the meanings defined above, and

Z has the meaning defined below,



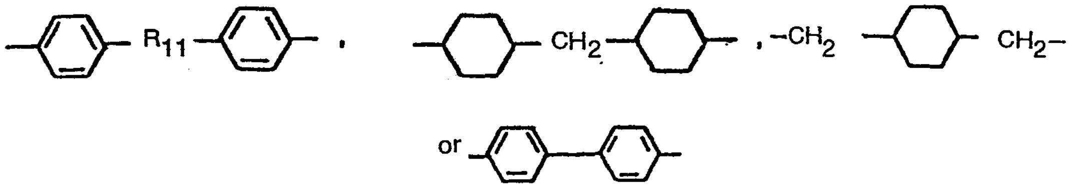
(VII)

in which:

R_1 has the meaning defined above,

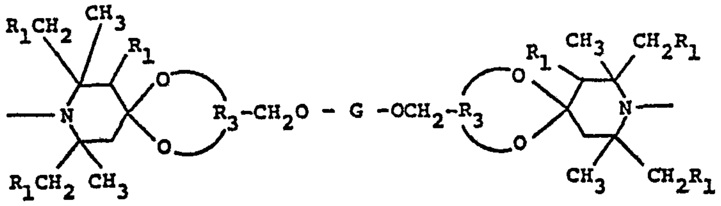
D represents an aliphatic, aromatic, araliphatic or alicyclic diacyl group having up to 36 carbon atoms or a group of formula —CONH—R_{10}—NHCO— in which

R_{10} represents an alkylene group having from 2 to 10 carbon atoms, a phenylene group optionally substituted with methyl, a naphthylene group, a xylylene group, a cyclohexylene group optionally substituted with methyl, a group of formula



in which

R_{11} represents an oxygen atom or the methylene group,

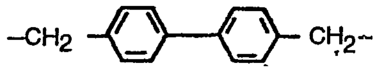


(VIII)

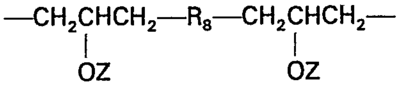
in which:

R_1 and R_3 have the meanings defined above,

G has the same meaning as the above-defined D, or represents an alkylene group having from 3 to 10 carbon atoms, a xylylene group, a group of formula



or a group of formula

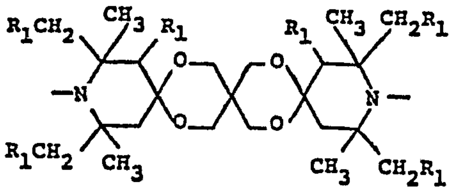


$$—CH_2\underset{\displaystyle OZ}{CH}CH_2—R_8—CH_2\underset{\displaystyle OZ}{CH}CH_2—$$

in which

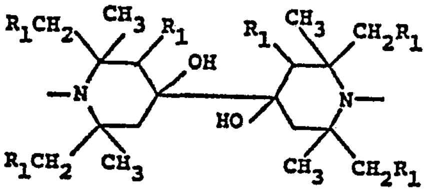
R_8 has the meaning defined above, and

Z has the meaning defined below,



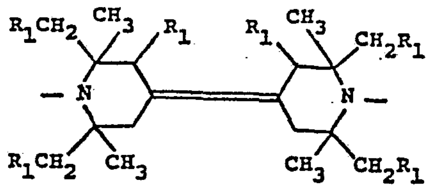
(IX)

in which R_1 has the meaning defined above,



(X)

in which R_1 has the meaning defined above,



(XI)

in which R_1 has the meaning defined above, and all of Z in the molecule represent a hydrogen atom, an alkyl group having from 1 to 18 carbon atoms, an aliphatic, aromatic, araliphatic or alicyclic acyl group having up to 18 carbon atoms or a group of formula —CONHR$_{12}$ in which R_{12} represents an alkyl group having from 1 to 18 carbon atoms, a phenyl or cyclohexyl group,
or a part of Z represents a hydrogen atom and the remaining Z represent above-defined groups other than hydrogen.

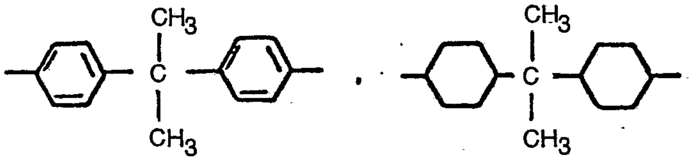
2. A polymer as claimed in claim 1, wherein Y is a group of formula (II) to (XI) and R_1 is a hydrogen atom.

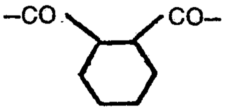
3. A polymer as claimed in claim 2, wherein both m_1 and m_2 are 0, Y is a group of formula (II) in which R_1 and R_2 are hydrogen atoms.

4. A polymer as claimed in claim 2, wherein both m_1 and m_2 are 0, Y is a group of formula (III) in which R_1 is a hydrogen atom.

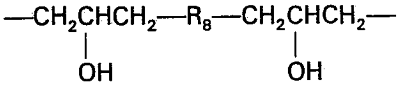
5. A polymer as claimed in claim 2, wherein both m_1 and m_2 are 0, Y is a group of formula (IV) in which R_1 is a hydrogen atom.

6. A polymer as claimed in claim 2, wherein one of m_1 and m_2 is 1 and the other is 0, X is a group of formula $—OCH_2CH_2O—$ or —OWO— in which
W is a group of formula



$—CO—(CH_2)_{n_3}CO—$, n_3 being an integer of from 4 to 10, or 

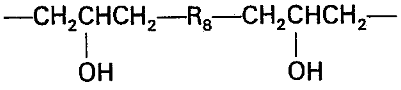
7. A polymer as claimed in claim 6, wherein Y is a group of formula (V) in which R_1 and R_2 are hydrogen atoms and A is an alkylene group having from 2 to 6 carbon atoms or a group of formula



$$—CH_2\underset{\displaystyle OH}{\underset{|}{C}}HCH_2—R_8—CH_2\underset{\displaystyle OH}{\underset{|}{C}}HCH_2—$$

wherein R_8 has the same meaning as X.

8. A polymer as claimed in claim 6, wherein Y is a group of formula (VI) in which R_1 is a hydrogen atom, and R_9 is an alkanoyl group having from 2 to 4 carbon atoms or an alkyl group having from 1 to 8 carbon atoms, and B is an alkylene group having from 2 to 6 carbon atoms, or R_9 is an alkyl group having from 1 to 8 carbon atoms, and B is a group of formula



$$—CH_2\underset{\displaystyle OH}{\underset{|}{C}}HCH_2—R_8—CH_2\underset{\displaystyle OH}{\underset{|}{C}}HCH_2—$$

wherein R_8 has the same meaning as X.

9. A polymer as claimed in claim 6, wherein Y is a group of formula (VII) in which R_1 is a hydrogen atom and D is an alkanedicarbonyl group having from 4 to 12 carbon atoms or a benzene dicarbonyl group.

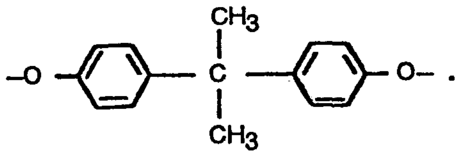
10. A polymer as claimed in claim 6, wherein Y is a group of formula (VIII) in which R_1 is a hydrogen atom and G is an alkanedicarbonyl group having from 4 to 12 carbon atoms or a group of formula

$$—CH_2\underset{\displaystyle OH}{\underset{|}{C}}HCH_2—R_8—CH_2\underset{\displaystyle OH}{\underset{|}{C}}HCH_2—$$

wherein R_8 has the same meaning as X.

11. A polymer as claimed in claim 6, wherein Y is a group of formula (IX), (X) or (XI) in which R_1 is a hydrogen atom.

12. A polymer as claimed in any one of claims 7 to 11, wherein X is a group of formula



13. A polymer as claimed in claim 1 wherein all of Z in the molecule are hydrogen atoms.

14. A polymer as claimed in claim 1, wherein all of Z in the molecule are acetyl or benzoyl groups, or a part of Z are hydrogen atoms and the remaining Z are acetyl or benzoyl groups.

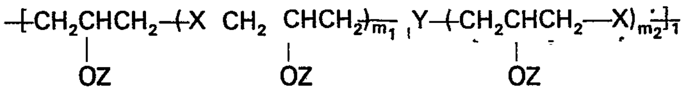
15. A polymer as claimed in claim 1, wherein *l* is an integer of from 2 to 10.

16. A synthetic polymer composition stabilized against light- and heat-degradation thereof, which is characterized in that there is incorporated into a synthetic polymer to be stabilized from 0.01 to 5.0% by weight, based on the weight of said synthetic polymer, of at least one polymer represented by the formula (I) as defined in claim 1.

17. A synthetic polymer composition as claimed in claim 16, wherein the synthetic polymer is an olefin or diene polymer, a styrene polymer or a polyurethane.

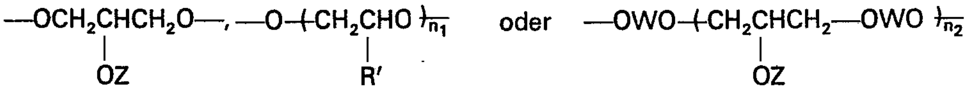
Patentansprüche

1. Polymere der Formel (I):



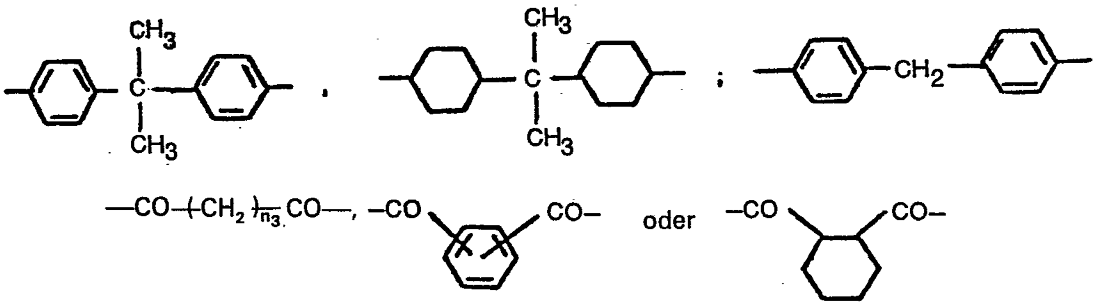
$$-\!\!\left[CH_2\underset{OZ}{CH}CH_2-\!\!\left(X\ CH_2\ \underset{OZ}{CH}CH_2\right)_{m_1} Y-\!\!\left(CH_2\underset{OZ}{CH}CH_2-X\right)_{m_2}\right]_l \qquad (I)$$

worin *l* eine ganze Zahl von 2 bis 50 ist, m_1 und m_2 0 sind bzw. eines davon 1 und das andere 0 ist, X für eine Gruppe der Formel



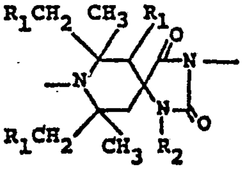
$$-OCH_2\underset{OZ}{CH}CH_2O-,\ -O-\!\!\left(CH_2\underset{R'}{CH}O\right)_{n_1} \quad \text{oder} \quad -OWO-\!\!\left(CH_2\underset{OZ}{CH}CH_2-OWO\right)_{n_2}$$

in denen R' ein Wasserstoffatom oder eine Methylgruppe bedeutet, n_1 eine ganze Zahl von 1 bis 10 und n_2 0 oder eine ganze Zahl von 1 bis 10 sind und W eine Gruppe der Formel



$-CO-(CH_2)_{n_3}CO-$, $-CO-C_6H_4-CO-$ oder $-CO-C_6H_{10}-CO-$

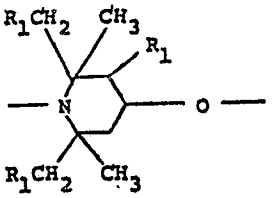
darstellt, wobei n_3 eine ganze Zahl von 1 bis 10 ist, steht und Z die unten angegebene Bedeutung hat, und Y, falls m_1 und m_2 0 sind, eine Gruppe der folgenden Formeln II bis IV:



(II)

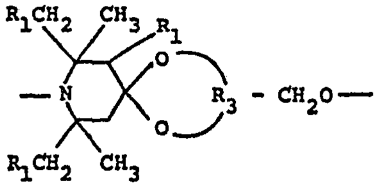
worin R_1 für ein Wasserstoffatom oder eine Methylgruppe und R_2 für ein Wasserstoffatom oder eine

Alkylgruppe mit 1 bis 18 Kohlenstoffatomen stehen,



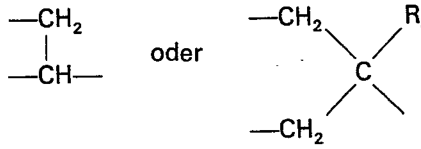
(III)

worin R_1 die oben angegebene Bedeutung hat,

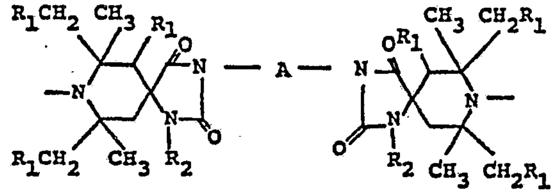


(IV)

worin R_1 die oben angegebene Bedeutung hat und R_3 für eine Gruppe der Formel

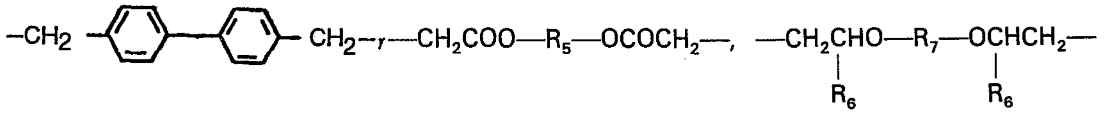


und R_4 für die Methyl- oder Äthylgruppe stehen, darstellt oder Y, falls eines von m_1 und m_2 1 und das andere 0 ist, eine Gruppe der folgenden Formeln V bis XI:

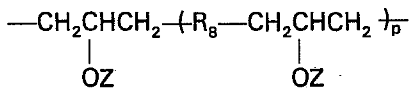


(V)

worin R_1 und R_2 die oben angegebenen Bedeutungen haben und A eine Alkylengruppe mit 2 bis 12 Kohlenstoffatomen, eine Xylylengruppe oder eine Gruppe der Formel

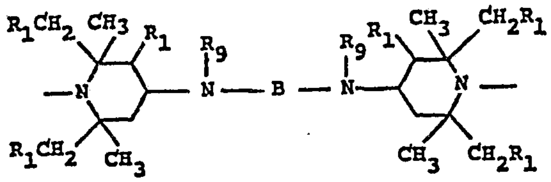


$$—CH_2CH_2COO—R_5—OCOCH_2—,\quad —\underset{R_6}{CH_2CHO}—R_7—\underset{R_6}{OCHCH_2}—$$



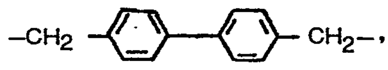
$$\text{oder}\quad —CH_2\underset{OZ}{CH}CH_2—(R_8—CH_2\underset{OZ}{CH}CH_2)_p—$$

bedeutet, wobei R_5 für eine Alkylengruppe mit 2 bis 8 Kohlenstoffatomen, R_6 für ein Wasserstoffatom oder die Methyl- oder Phenylgruppe und R_7 für eine aliphatische, aromatische oder alicyclische Diacylgruppe mit bis zu 12 Kohlenstoffatomen stehen, R_8 dieselbe Bedeutung wie das oben definierte X hat, p 0 oder 1 ist und Z die unten angegebene Bedeutung hat,

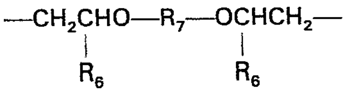


(VI)

worin R_1 die oben angegebene Bedeutung hat, R_9 für eine Alkylgruppe mit 1 bis 18 Kohlenstoffatomen, eine Phenylgruppe, eine Benzylgruppe, eine Cyclohexylgruppe oder eine aliphatische, aromatische, araliphatische oder alicyclische Acylgruppe mit bis zu 18 Kohlenstoffatom steht und B eine Alkylengruppe mit 2 bis 10 Kohlenstoffatomen, eine Xylylengruppe oder eine Gruppe der Formel

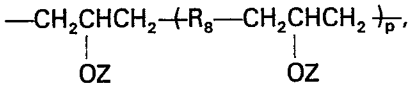


$-CH_2COO-R_5-OCOCH_2-$,



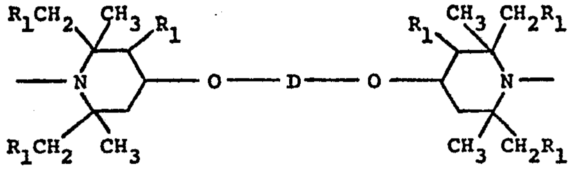
$-CH_2\underset{\substack{|\\R_6}}{C}HO-R_7-O\underset{\substack{|\\R_6}}{C}HCH_2-$

oder



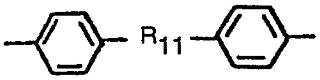
$-CH_2\underset{\substack{|\\OZ}}{C}HCH_2-(R_8-CH_2\underset{\substack{|\\OZ}}{C}HCH_2)_p-$,

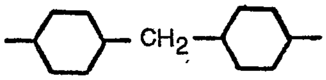
bedeutet, wobei R_5, R_6, R_7, R_8 und p die oben angegebenen Bedeutungen und Z die unten angegebene Bedeutung hat,

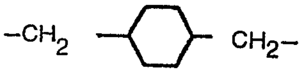


(VII)

worin R_1 die oben angegebene Bedeutung hat und D eine aliphatische, aromatische, araliphatische oder alicyclische Diacylgruppe mit bis zu 36 Kohlenstoffatomen oder eine Gruppe der Formel $-CONH-R_{10}-NHCO-$ bedeutet, in der R_{10} für eine Alkylengruppe mit 2 bis 10 Kohlenstoffatomen, eine gegebenenfalls methylsubstituierte Phenylengruppe, eine Naphthylengruppe, eine Xylylengruppe, eine gegebenenfalls methylsubstituierte Cyclohexylengruppe oder eine Gruppe der Formel







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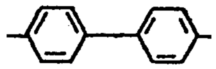
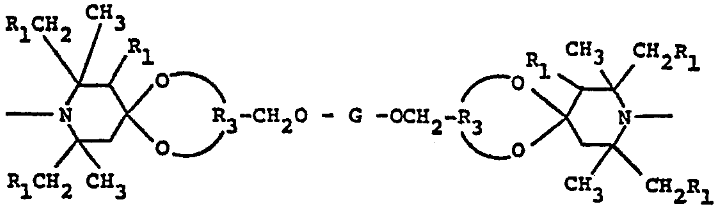
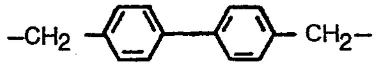


darstellt, wobei R_{11} für eine Sauerstoffatom oder die Methylengruppe steht,

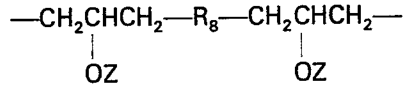


(VIII)

worin R_1 und R_3 die oben angegebenen Bedeutungen haben und G dieselbe Bedeutung wie das oben definierte D hat oder eine Alkylengruppe mit 3 bis 10 Kohlenstoffatomen, eine Xylylengruppe, eine Gruppe der Formel

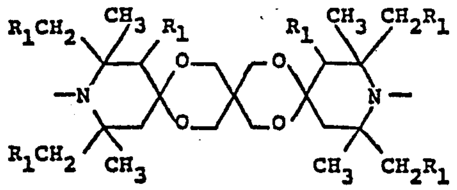


oder eine Gruppe der Formel



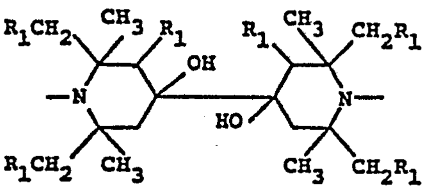
$-CH_2\underset{\substack{|\\OZ}}{C}HCH_2-R_8-CH_2\underset{\substack{|\\OZ}}{C}HCH_2-$

darstellt, wobei R_8 die oben angegebene Bedeutung und Z die unten angegebene Bedeutung haben,



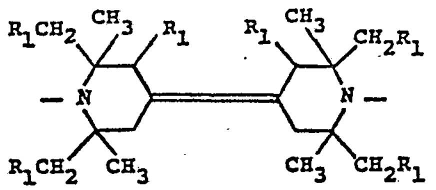
(IX)

worin R_1 die oben angegebene Bedeutung hat,



(X)

worin R_1 die oben angegebene Bedeutung hat,



(XI)

worin R_1 die oben angegebene Bedeutung hat, darstellt und sämtliche Z im Molekül für ein Wasserstoffatom, eine Alkylgruppe mit 1 bis 18 Kohlenstoffatomen, eine aliphatische, aromatische, araliphatische oder alicyclische Acylgruppe mit bis zu 18 Kohlenstoffatomen oder eine Gruppe der Formel $-CONHR_{12}$ stehen, worin R_{12} eine Alkylgruppe mit 1 bis 18 Kohlenstoffatomen oder eine Phenyl- oder Cyclohexylgruppe bedeutet, bzw. ein Teil der Z für ein Wasserstoffatom steht und die übrigen Z wie oben definierte, von Wasserstoff verschiedene Gruppen bedeuten.

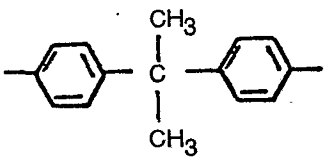
2. Polymere nach Anspruch 1, dadurch gekennzeichnet, dass Y eine Gruppe der Formel (II) bis (XI) darstellt und R_1 für ein Wasserstoffatom steht.

3. Polymere nach Anspruch 2, dadurch gekennzeichnet, dass sowohl m_1 als auch m_2 0 sind und Y eine Gruppe der Formel (II) darstellt, worin R_1 und R_2 für Wasserstoffatome stehen.

4. Polymere nach Anspruch 2, dadurch gekennzeichnet, dass sowohl m_1 als auch m_2 0 sind und Y eine Gruppe der Formel (III) darstellt, worin R_1 für ein Wasserstoffatom steht.

5. Polymere nach Anspruch 2, dadurch gekennzeichnet, dass sowohl m_1 als auch m_2 0 sind und Y eine Gruppe der Formel (IV) darstellt, worin R_1 für ein Wasserstoffatom steht.

6. Polymere nach Anspruch 2, dadurch gekennzeichnet, dass eines von m_1 und m_2 1 und das andere 0 ist und X eine Gruppe der Formel $-OCH_2CH_2O-$ oder $-OWO-$ darstellt, worin W eine Gruppe der Formel



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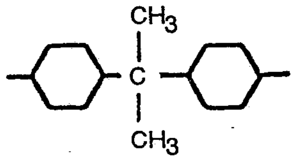
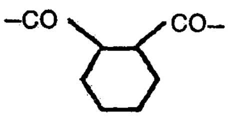


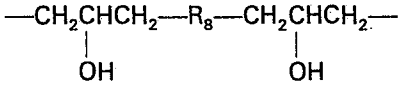
, $-CO-(CH_2)_{n_3}-CO-$,

wobei n_3 eine ganze Zahl von 4 bis 10 ist, oder



bedeutet.

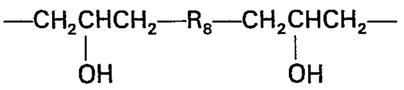
7. Polymere nach Anspruch 6, dadurch gekennzeichnet, dass Y eine Gruppe der Formel (V) darstellt, in der R_1 und R_2 für Wasserstoffatome und A für eine Alkylengruppe mit 2 bis 6 Kohlenstoffatomen oder eine Gruppe der Formel



$$-CH_2\underset{OH}{CH}CH_2-R_8-CH_2\underset{OH}{CH}CH_2-$$

worin R_8 dieselbe Bedeutung wie X hat, stehen.

8. Polymere nach Anspruch 6, dadurch gekennzeichnet, dass Y eine Gruppe der Formel (VI) darstellt, in der R_1 für ein Wasserstoffatom, R_9 für eine Alkanoylgruppe mit 2 bis 4 Kohlenstoffatomen oder eine Alkylgruppe mit 1 bis 8 Kohlenstoffatomen und B für eine Alkylengruppe mit 2 bis 6 Kohlenstoffatomen stehen oder R_9 für eine Alkylgruppe mit 1 bis 8 Kohlenstoffatomen und B für eine Gruppe der Formel

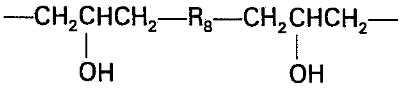


$$-CH_2\underset{OH}{CH}CH_2-R_8-CH_2\underset{OH}{CH}CH_2-$$

worin R_8 dieselbe Bedeutung wie X hat, stehen.

9. Polymere nach Anspruch 6, dadurch gekennzeichnet, dass Y eine Gruppe der Formel (VII) darstellt, in der R_1 für ein Wasserstoffatom und D für eine Alkandicarbonylgruppe mit 4 bis 12 Kohlenstoffatomen oder eine Benzoldicarbonylgruppe stehen.

10. Polymere nach Anspruch 6, dadurch gekennzeichnet, dass Y eine Gruppe der Formel (VIII) darstellt, in der R_1 für ein Wasserstoffatom und G für eine Alkandicarbonylgruppe mit 4 bis 12 Kohlenstoffatomen oder eine Gruppe der Formel

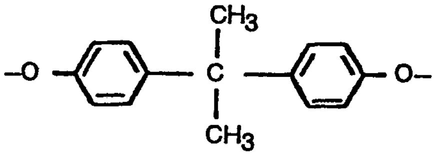


$$-CH_2\underset{OH}{CH}CH_2-R_8-CH_2\underset{OH}{CH}CH_2-$$

worin R_8 dieselbe Bedeutung wie X hat, stehen.

11. Polymere nach Anspruch 6, dadurch gekennzeichnet, dass Y eine Gruppe der Formel (IX), (X) oder (XI) darstellt, in der R_1 für ein Wasserstoffatom steht.

12. Polymere nach einem der Ansprüche 7 bis 11, dadurch gekennzeichnet, das X eine Gruppe der Formel



darstellt.

13. Polymere nach Anspruch 1, dadurch gekennzeichnet, dass sämtliche Z im Molekül Wasserstoffatome bedeuten.

14. Polymere nach Anspruch 1, dadurch gekennzeichnet, dass sämtliche Z im Molekül Acetyl- oder Benzoylgruppen bedeuten oder ein Teil der Z Wasserstoffatome und die übrigen Z Acetyl- oder Benzoylgruppen sind.

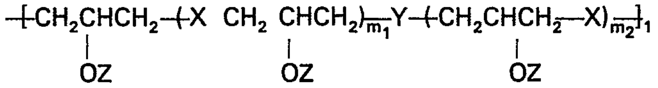
15. Polymere nach Anspruch 1, dadurch gekennzeichnet, dass *1* eine ganze Zahl von 2 bis 10 ist.

16. Gegen Licht- und Hitzeabbau stabilisierte synthetische Polymermassen, dadurch gekennzeichnet, dass dabei in ein zu stabilisierendes synthetisches Polymer 0,01 bis 5,0 Gew.-% bezogen auf das Gewicht dieses synthetischen Polymeren, mindestens eines durch die wie in Anspruch 1 definierte Formel (I) dargestellten Polymeren eingearbeitet sind.

17. Synthetische Polymermassen nach Anspruch 16, dadurch gekennzeichnet, dass als synthetisches Polymer ein Olefin- oder Dienpolymer, ein Styrolpolymer oder ein Polyurethan vorliegt.

Revendications

1. Les polymères de formule générale I ci-dessous:



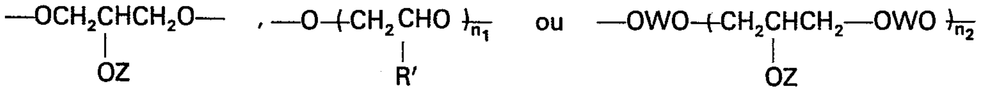
$$-\!\left[CH_2\underset{OZ}{CH}CH_2-(X\ CH_2\ \underset{OZ}{CH}CH_2)_{m_1}Y-(CH_2\underset{OZ}{CH}CH_2-X)_{m_2}\right]_1$$

dans laquelle

1 est un entier de 2 à 50,

m_1 et m_2 sont tous les deux nuls, ou bien l'un d'eux est le chiffre 1 et l'autre nul,

X représente un groupe



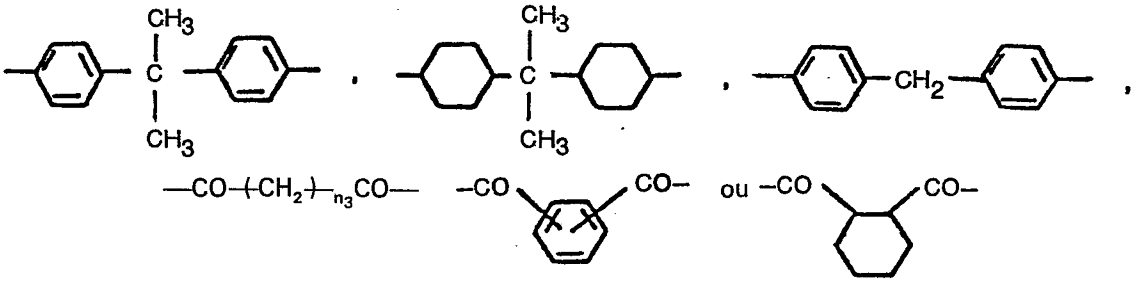
$$-OCH_2\underset{OZ}{CH}CH_2O- \quad , -O-(CH_2\underset{R'}{CH}O)_{n_1} \quad \text{ou} \quad -OWO-(CH_2\underset{OZ}{CH}CH_2-OWO)_{n_2}$$

R' étant l'hydrogène ou un méthyle,

n_1 un entier de 1 à 10,

n_2 un entier de 0 à 10,

W représentant un groupe

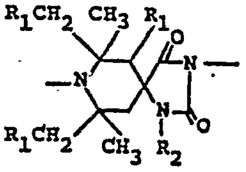


$—CO—(CH_2)_{n_3}CO—$

n_3 étant un entier de 1 à 10, et

Z ayant la signification donnée plus loin,

Y représente, si m_1 et m_2 sont tous les deux nuls, un groupe répondant à l'une des formules II à IV ci-après:

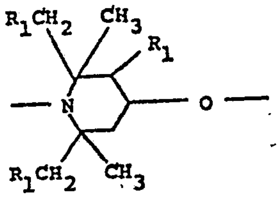


(II)

dans laquelle

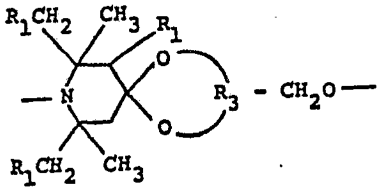
R_1 représente un atome d'hydrogène ou un méthyle, et

R_2 un atome d'hydrogène ou un alkyle en C_1 à C_{18},



(III)

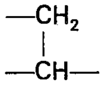
R_1 ayant la signification ci-dessus



(IV)

R_1 ayant la signification ci-dessus,

R_3 représentant un groupe



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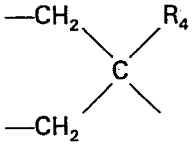
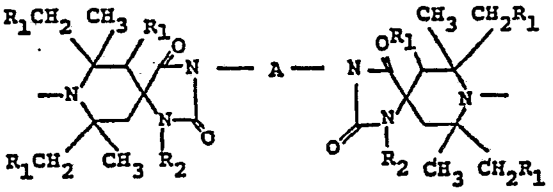


et

R_4 un méthyle ou un éthyle, tandis que

Y représente, si parmi m_1 et m_2, l'un est le nombre 1 et l'autre est nul, l'un des groupes de formules V à XI ci-après:

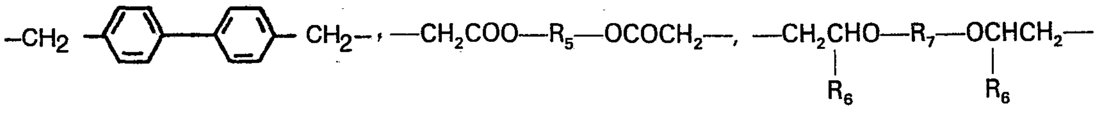


(V)

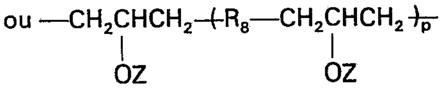
dans laquelle

R_1 et R_2 ont les significations données plus haut, et

A représente un alkylène en C_2 à C_{12}, un groupe xylylène ou un groupe



$$-CH_2-C_6H_4-C_6H_4-CH_2-,\ -CH_2COO-R_5-OCOCH_2-,\ -CH_2\underset{R_6}{CH}O-R_7-O\underset{R_6}{CH}CH_2-$$



$$\text{ou } -CH_2\underset{OZ}{CH}CH_2\!-\!(R_8-CH_2\underset{OZ}{CH}CH_2)_p-$$

R_5 étant un alkylène en C_2 à C_8,

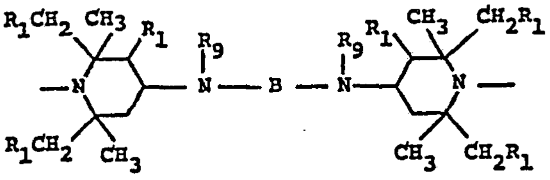
R_6 l'hydrogène, un méthyle ou un phényle,

R_7 un radical diacyle aliphatique, aromatique ou alicyclique jusqu'en C_{12},

R_8 ayant la signification donnée ci-dessus pour X,

p étant le nombre 0 ou 1, et

Z ayant la signification donnée plus loin,



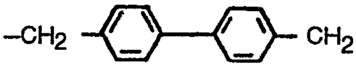
(VI)

dans laquelle

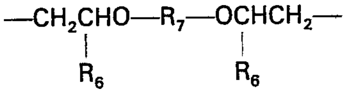
R_1 a la signification ci-dessus,

R_9 représente un alkyle en C_1 à C_{18} ou bien un phényle, un benzyle, un cyclohexyle ou un radical acyle aliphatique, aromatique, araliphatique ou alicyclique jusqu'en $C_1$8, et

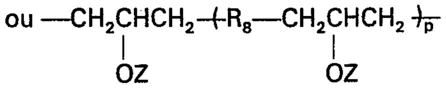
B représente un alkylène en C_2 à C_{10}, un xylylène ou un groupe



—CH_2COO—R_5—$OCOCH_2$—,



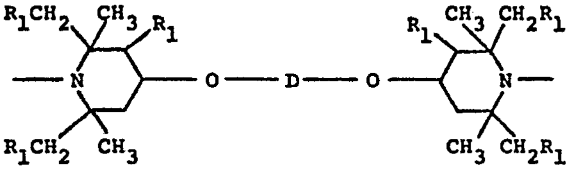
$$-CH_2\underset{\displaystyle R_6}{C}HO-R_7-O\underset{\displaystyle R_6}{C}HCH_2-$$



$$\text{ou }-CH_2\underset{\displaystyle OZ}{C}HCH_2\left(R_8-CH_2\underset{\displaystyle OZ}{C}HCH_2\right)_p$$

R_5, R_6, R_7, R_8 et p ayant les significations ci-dessus et

Z la signification qui sera donnée plus loin,



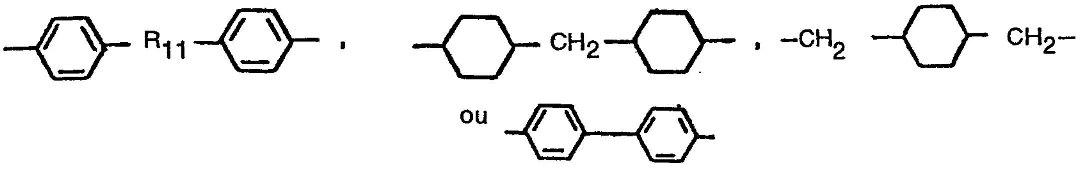
(VII)

dans laquelle

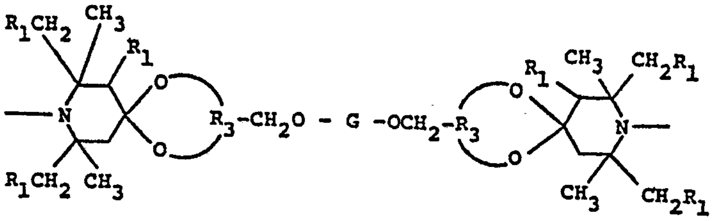
R_1 a la signification ci-dessus, et

D représente un radical diacyle aliphatique, aromatique, araliphatique ou alicyclique jusqu'en C_{36} ou bien un groupe —CONH—R_{10}—NHCO—,

R_{10} étant un alkylène en C_2 à C_{10}, un phénylène éventuellement substitué par un méthyle, un groupe naphtylène ou xylylène, un groupe cyclohexylène éventuellement substitué par un méthyle ou un groupe



R_{11} représentant un atome d'oxygène ou le groupe méthylène,

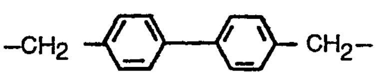


(VIII)

dans laquelle

R_1 et R_3 ont les significations ci-dessus, et

G a la signification donnée plus haut pour D ou bien représente un alkylène en C_3 à C_{10}, un xylylène ou un groupe



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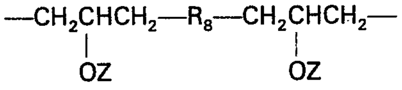
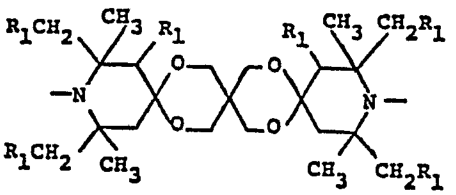


$$-CH_2\underset{\displaystyle OZ}{C}HCH_2-R_8-CH_2\underset{\displaystyle OZ}{C}HCH_2-$$

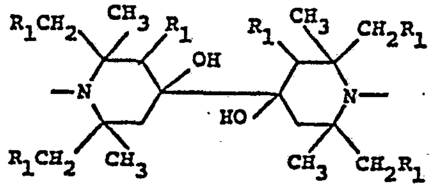
R_8 ayant la signification donnée plus haut et

Z la signification donnée plus loin,



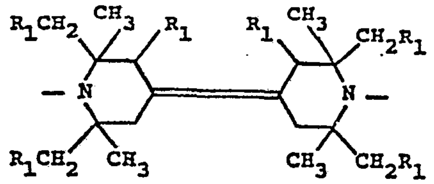
(IX)

R_1 ayant la signification ci-dessus,



(X)

R_1 ayant la signification ci-dessus,



(XI)

R_1 ayant la signification ci-dessus, et tous les radicaux Z de la molécule sont chacun un atome d'hydrogène, un alkyle en C_1 à C_{18}, un radical acyle aliphatique, aromatique, araliphatique ou alicyclique jusqu'en C_{18}, ou encore un groupe $-CONHR_{12}$, R_{12} représentant un alkyle en C_1 à C_{18}, un phényle ou un cyclohexyle, ou bien

certains seulement des radicaux Z sont des atomes d'hydrogène et les autres des groupes tels que définis plus haut, à l'exception de l'hydrogène.

2. Polymère selon la revendication 1, dans lequel Y est un des groupes de formules II à XI et R_1 un atome d'hydrogène.

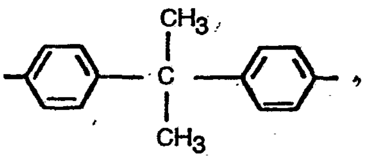
3. Polymère selon la revendication 2, dans lequel m_1 et m_2 sont tous les deux nuls et Y est un groupe de formule II, R_1 et R_2 étant des atomes d'hydrogène.

4. Polymère selon la revendication 2, dans lequel m_1 et m_2 sont nuls tous les deux, et Y est un groupe de formule III, R_1 étant un atome d'hydrogène.

5. Polymère selon la revendication 2, dans lequel m_1 et m_2 sont nuls tous les deux et Y est un groupe de formule IV, R_1 étant un atome d'hydrogène.

6. Polymère selon la revendication 2, dans lequel l'un des nombres m_1 et m_2 est égal à un et l'autre est nul, et X est un groupe $-OCH_2CH_2O-$ ou $-OWO-$,

W étant un groupe



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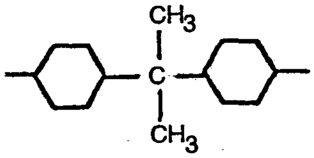
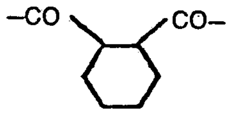
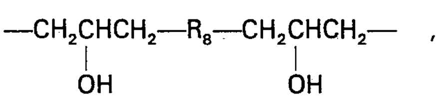


ou $-CO\!\left(CH_2\right)_{n_3}CO-$, n_3 étant un entier de 4 à 10, ou le groupe



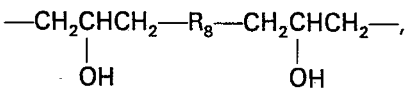
7. Polymère selon la revendication 6, dans lequel Y est un groupe de formule V, R_1 et R_2 étant des atomes d'hydrogène et A un alkylène en C_2 à C_6 ou un groupe



$$-CH_2\underset{\substack{|\\ OH}}{C}HCH_2-R_8-CH_2\underset{\substack{|\\ OH}}{C}HCH_2- \quad ,$$

R_8 ayant la même signification que X.

8. Polymère selon la revendication 6, dans lequel Y est un groupe de formule VI, R_1 étant un atome d'hydrogène et R_9 un alcanoyle en C_2 à C_4 ou un alkyle en C_1 à C_8, et B un alkylène en C_2 à C_6, ou bien R_9 est un alkyle en C_1 à C_8 et B un groupe

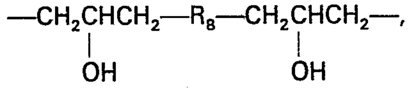


$$-CH_2\underset{\substack{|\\ OH}}{C}HCH_2-R_8-CH_2\underset{\substack{|\\ OH}}{C}HCH_2-,$$

R_8 ayant la même signification que X.

9. Polymère selon la revendication 6, dans lequel Y est un groupe de formule VII, R_1 étant un atome d'hydrogène et D un radical alcane-dicarbonyle en C_4 à C_{12} ou un radical benzène-dicarbonyle.

10. Polymère selon la revendication 6, dans lequel Y est un groupe de formule VIII, R_1 étant un atome d'hydrogène et G un radical alcane-dicarbonyle en C_4 à C_{12} ou un groupe

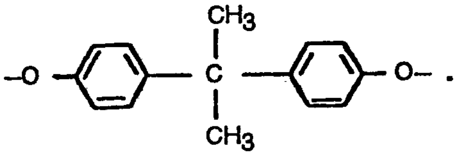


$$—CH_2\underset{OH}{\underset{|}{C}}HCH_2—R_8—CH_2\underset{OH}{\underset{|}{C}}HCH_2—,$$

R_8 ayant la même signification que X.

11. Polymère selon la revendication 6, dans Y est un groupe de formule IX, X ou XI, R_1 étant un atome d'hydrogène.

12. Polymère selon l'une quelconque des revendications 7 à 11, dans lequel X est un groupe de formule



13. Polymère selon l'une quelconpue des revendications précédentes, dans lequel tous les radicaux Z de la molécule sont des atomes d'hydrogène.

14. Polymère selon l'une quelconque des revendications précédentes, dans lequel tous les radicaux Z sont des groupes acétyle ou benzoyle, ou bien certains des radicaux Z sont des atomes d'hydrogène et les autres des groupes acétyle ou benzoyle.

15. Polymère selon l'une quelconque des revendications précédentes, dans lequel l est un entier de 2 à 10.

16. Composition de matière polymère synthétique protégée contre les dégradations sous l'action de la lumière et de la chaleur, composition caractérisée en ce qu'elle comprend 0,01 à 5%, du poids de la matière synthétique, d'un ou de plusieurs polymères de formule générale I selon la revendication 1.

17. Composition selon la revendication 16, dans laquelle la matière polymère synthétique est une polyoléfine, un polymère de diènes ou du styrène ou un polyuréthanne.