

A THERMOPLASTIC RESIN COMPOSITIONBACKGROUND OF THE INVENTIONField of The Invention

The present invention relates to a thermoplastic resin composition which comprises a polyphenylene ether and a non-crystalline thermoplastic resin, having improved properties in process stability and in heat stability on practically using.

Description of Related Art

A polyphenylene ether resin is a thermoplastic resin superior in various characteristics such as mechanical properties, heat resistance and dimensional stability. However, it is apt to undergo coloration and deteriorations such as cross-linking when molded at high temperatures. Especially, in the presence of oxygen, it undergoes serious deterioration due to the heat to cause gelation, increasing in molecular weight or sometimes produce black specks. Furthermore, polyphenylene ether resin readily undergoes discoloration with light.

In order to offset these defects of the resin, it has been proposed to add various additives. For example, phosphorus additives are proposed in Japanese Patent Kokoku No. 44-29751 and phenolic additives are proposed in Japanese Patent Kokoku No. 46-24782. Furthermore, many attempts such

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as use of various stabilizers containing steric hindrance type amines or containing steric hindrance type amines and phenolic additives have been made mainly for inhibition of the discoloration with light as disclosed in Japanese Patent Kokai Nos. 60-149646 and 60-203150; U.S. Patent Nos. 4232131, 4233410, 4233412, 4234699, 4260689, 4260691 and 4299926; EP 094048A2, EP 134523A1, EP 146879A2, EP 146878A2, EP 151961A1, EP 149454A2, EP 233153A2, EP 240723A2, EP 229363A2 and EP 313118A1. Many of these attempts may be effective for improving evaporation of stabilizers at the time of processing or actual use, and various compounds containing steric hindrance type amines alone or copolymers (oligomers) with other components have been proposed. However, none of these proposals have provided sufficient effects to inhibit deterioration at heating.

#### SUMMARY OF THE INVENTION

The present invention relates to a thermoplastic resin composition comprising a polyphenylene ether resin and a non-crystalline thermoplastic resin, which is inhibited from deterioration at the time of processing, especially with heat.

The present inventors have found that a conspicuous effect to inhibit the deterioration with heat can be obtained by adding a compound containing steric hindrance type amine having a specific structure to a thermoplastic resin composition comprising a polyphenylene ether resin and a non-

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crystalline thermoplastic resin.

The present invention relates to a thermoplastic resin composition which comprises :

(a) 1 to 99 % by weight of a polyphenylene ether resin,

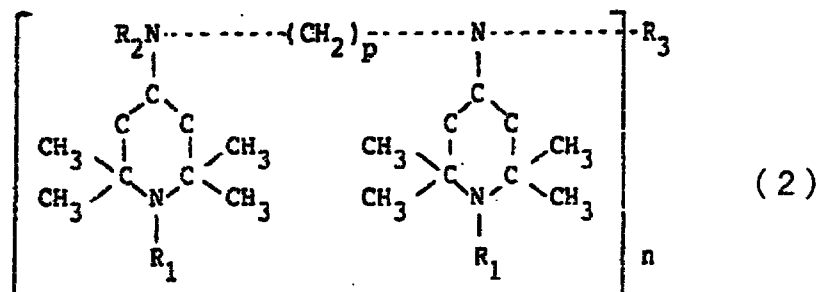
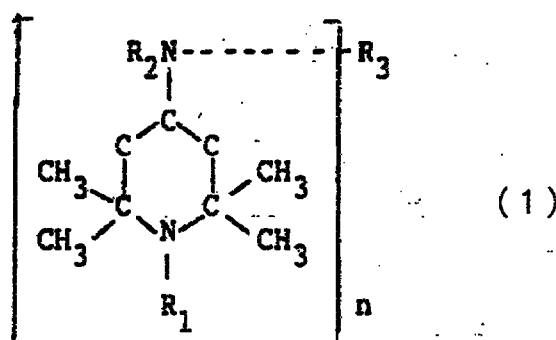
(b) 99 to 1 % by weight of at least one non-crystalline thermoplastic resin selected from an alkenyl aromatic resin, a non-crystalline aromatic polyester resin, a methacrylate resin, a polyetherimide resin and an aromatic polysulfone

resin, and per 100 parts by weight of the sum of (a) and (b),

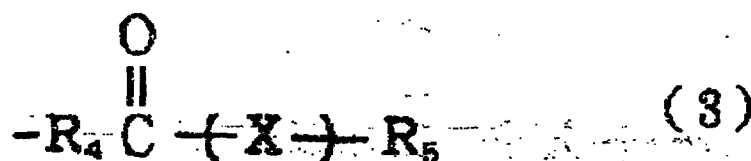
(c) 0.001 to 15 parts by weight of at least one 4-amino-

2,2,6,6-tetramethylpiperidyl compound represented by the

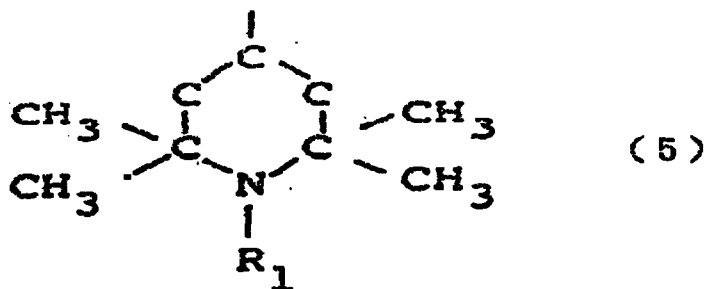
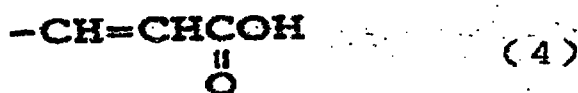
following formula (1) or (2) ;



; wherein  $R_1$  represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group, and  $R_2$  represents a hydrogen atom, an alkyl group or a group represented by the following formula (3),



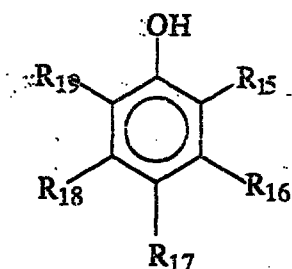
in which X represents a direct bond, -O- or -NH-,  $R_4$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and  $R_5$  represents a hydrogen atom, an alkyl group, an aryl group or a group represented by the following formula (4) or (5),



; and wherein  $n$  represents an integer of 1 to 4,  $p$  represents an integer of 2 to 6, and  $R_3$  represents an  $n$  valent organic group.

#### DETAILED DESCRIPTION OF THE PRESENT INVENTION

The polyphenylene ether resin (a) of the present invention is a polymer obtained by oxidation polymerization of at least one phenol compound represented by the following formula with oxygen or a gas containing oxygen using an oxidation coupling catalyst ;



wherein  $R_{15}$ ,  $R_{16}$ ,  $R_{17}$ ,  $R_{18}$  and  $R_{19}$  each represents a hydrogen atom, a halogen atom or a substituted or unsubstituted hydrocarbon group and at least one of them is a hydrogen atom.

Examples of  $R_{15}$ ,  $R_{16}$ ,  $R_{17}$ ,  $R_{18}$  and  $R_{19}$  are hydrogen, chlorine, bromine, fluorine, iodine, methyl, ethyl, *n*- or isopropyl, *pri*-, *sec*- or *t*-butyl, chloroethyl, hydroxyethyl, phenylethyl, benzyl, hydroxymethyl, carboxyethyl, methoxycarbonyl, cyanoethyl, phenyl, chlorophenyl, methylphenyl, dimethylphenyl, ethylphenyl and allyl.

Examples of the compounds represented by the above



formula are phenol, o-, m- or p-cresol, 2,6-, 2,5-, 2,4- or 3,5-dimethylphenol, 2-methyl-6-phenylphenol, 2,6-diphenylphenol, 2,6-diethylphenol, 2-methyl-6-ethylphenol, 2,3,5-, 2,3,6- or 2,4,6-trimethylphenol, 3-methyl-6-t-butylphenol, thymol and 2-methyl-6-allylphenol.

The phenol compounds of the above formula can also be copolymerized with other phenol compounds, for example, polyhydric aromatic compounds such as bisphenol A, tetrabromobisphenol A, resorcin, hydroquinone and novolak resins.

As preferable examples of the polyphenylene ether resin (a), mention may be made of homopolymers of 2,6-dimethylphenol, homopolymers of 2,6-diphenylphenol, copolymers of a larger amount of 2,6-dimethylphenol and a smaller amount of 3-methyl-6-t-butylphenol or 2,3,6-trimethylphenol.

Any oxidation coupling catalysts may be used for oxidizing polymerization of the phenol compound as far as they have polymerization ability.

The non-crystalline thermoplastic resin (b) of the present invention is at least one selected from an alkenyl aromatic resin, a non-crystalline aromatic polyester resin, a methacrylate resin, a polyether imide resin and an aromatic polysulfone resin.

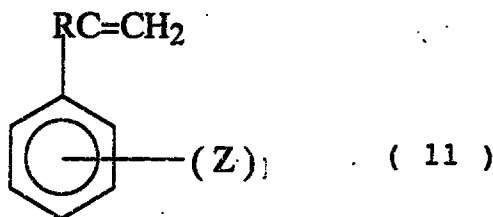
The alkenyl aromatic resin includes a homopolymer and a copolymer containing an aromatic vinyl compound represented by

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the following formula (11) ;



wherein R represents a hydrogen atom, an alkyl group having 1 to 6 carbon atoms or a halogen atom, Z represents a hydrogen atom, a vinyl group, a halogen atom, an amino group or an alkyl group having 1-6 carbon atoms, and l is 0 or an integer of 1 to 5.

The example of the alkenyl aromatic resin includes a polymer containing one kind or plural kinds of polymerization unit selected from styrene,  $\alpha$ -methylstyrene and p-methylstyrene, polystyrene (PS resin), polychlorostyrene, poly- $\alpha$ -methylstyrene, styrene-acrylonitrile copolymer (AS resin), styrene- $\alpha$ -methylstyrene copolymer, styrene-4-methylstyrene copolymer, styrene-maleic anhydride copolymer and styrene-methylmethacrylate copolymer (MS resin).

Besides, it includes a copolymer of a compound containing 2 or more vinyl groups such as divinyl benzene and ethyleneglycol dimethacrylate, with the above-mentioned aromatic vinyl compound.

Further, the alkenyl aromatic resin includes a rubber-modified polymer containing a dispersed rubber phase in the matrix of the alkenyl aromatic resin phase. The rubber used



here includes a homopolymer or copolymer containing a diene component such as polybutadiene rubber (BR), polyisoprene rubber (IR), polychlorobutadiene rubber, a butadiene-styrene copolymer rubber, an isoprene-styrene copolymer rubber, a butadiene-acrylonitrile copolymer rubber (NBR), an isobutylene-butadiene or -isoprene copolymer rubber, an ethylene-propylene copolymer rubber (EPM) and an ethylene-propylene-non-conjugated diene copolymer rubber (EPDM).

Various methods are known as a method for dispersing the rubber into the matrix of the alkenyl aromatic resin phase, and for example, a method by blending mechanically, a method by solution blending, and a method by dissolving a rubber into an alkenyl aromatic monomer and graft copolymerizing said monomer, are mentioned, and any one of them may be used.

Besides, a polymerization method may be any one of polymerization methods such as an emulsion polymerization process, a bulk polymerization process (Japanese Examined Patent Kokoku No.42-662 and USP 3435096), a solution polymerization process (USP 3538190 and USP 3538191) and a suspension polymerization process (Japanese Examined Patents Kokoku No.49-10831, No.57-40166 and No.62-10565). The ratio of the rubber to the alkenyl aromatic monomer in the rubber-modified alkenyl aromatic resin is preferably 0.5 to 80 %, and more preferably 0.5 to 50% by weight of the rubber as total amount of 100.



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Various methods are known as a method for dispersing the rubber into the matrix of the alkenyl aromatic resin phase, and for example, a method by blending mechanically, a method by solution-blending, and a method by dissolving a rubber into an alkenyl aromatic monomer and graft-copolymerizing said monomer, are mentioned, and any one of them may be used.

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As the examples of the rubber-modified alkenyl aromatic resin, a high impact polystyrene (HIPS) such as polystyrene modified with polybutadiene rubber, polystyrene modified with styrene-butadiene copolymer rubber and polystyrene modified with ethylene-propylene-non-conjugated diene copolymer rubber, an acrylonitrile-butadiene-styrene copolymer resin known as ABS resin and an acrylonitrile-(ethylene-propylene-non-conjugated diene copolymer rubber)-styrene resin known as AES resin are mentioned.

The non-crystalline aromatic polyester resin is polycarbonate resin and polyarylate resin.

The polycarbonate resin of the present invention is 4,4-dioxydialkanic polycarbonate including 4,4-dihydroxydiphenyl-2,2-propane (bisphenol A), and among them, 4,4-dihydroxydiphenyl-2,2-propane of polycarbonate having number average molecular weight of 15,000 to 80,000 is especially preferred. These polycarbonates are prepared with an arbitrary method. For example, a method of preparing by using 4,4-dihydroxydiphenyl-2,2-propane and phosgene in the presence of sodium hydroxide aqueous solution and a solvent, and a method of preparing by ester interchanging of 4,4-dihydroxydiphenyl-2,2-propane with carbonic diester in the presence of a catalyst are mentioned.

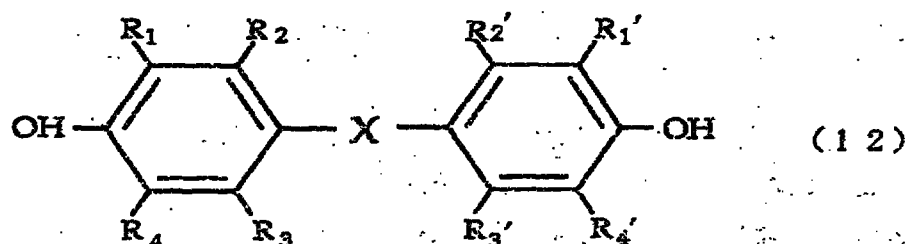
The polyarylate resin of the present invention is a polyester resin obtained from bisphenols and terephthalic acid

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and/or isophthalic acid. As the bisphenols, those having a structure represented by the following formula (12) are used ;



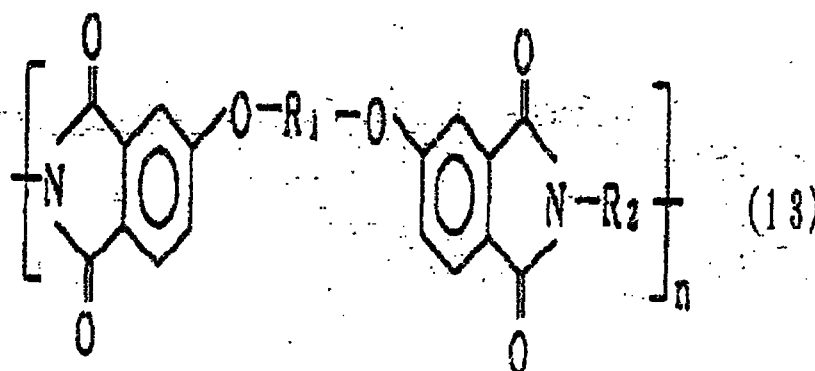
In the formula (12), X is a group selected from -O-, -S-, -CO- and a hydrocarbon group having 1 to 10 carbon atoms, and  $R_1$  to  $R_4$  and  $R'_1$  to  $R'_4$  represent a group selected from a hydrogen atom, a halogen atom and a hydrocarbon group. When X represents a hydrocarbon group having 1 to 10 carbon atoms, an alkylene group, a branched alkylene group and a halogen-substituted alkyl group are included. On the other hand, each terephthalic acid and isophthalic acid can be used alone or in combination. It is obtained by polymerizing the above-mentioned bisphenols with terephthalic acid and/or isophthalic acid, and as a polymerization method, either of usual interfacial polymerization process or solution polymerization process may be used. Besides, these polyarylate resins having a molecular weight of 5,000 to 70,000 are preferred.

The methacrylate resin of the present invention is a homopolymer of methyl methacrylate or a copolymer of methyl methacrylate with acrylate, methacrylate, acrylic acid, methacrylic acid, styrene, acrylonitrile, glutaric anhydride, glutarimide or a diene rubber.

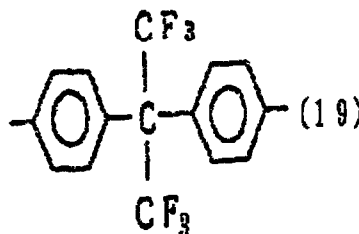
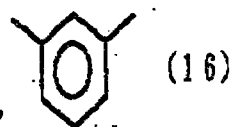
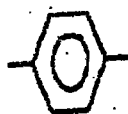
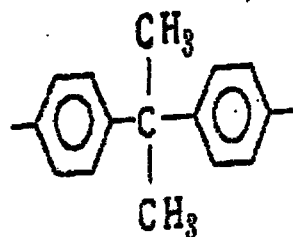


Acrylate includes methyl acrylate, ethyl acrylate and butyl acrylate, and methacrylate includes methyl methacrylate, ethyl methacrylate, butyl methacrylate and cyclohexyl methacrylate.

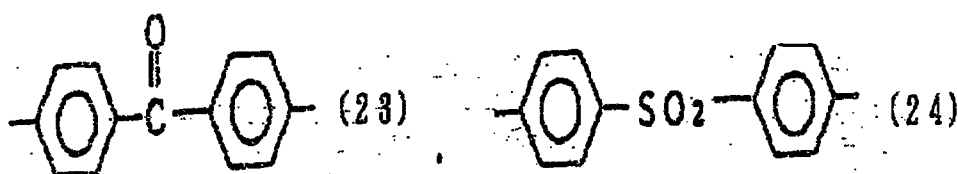
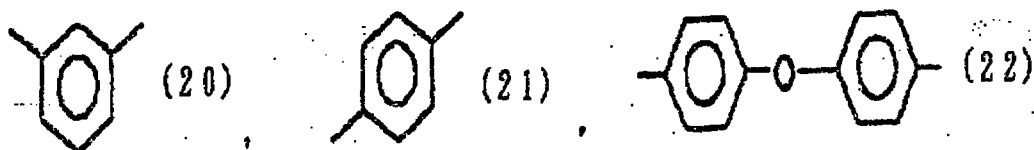
The polyetherimide resin of the present invention is a resin represented by the following formula (13),



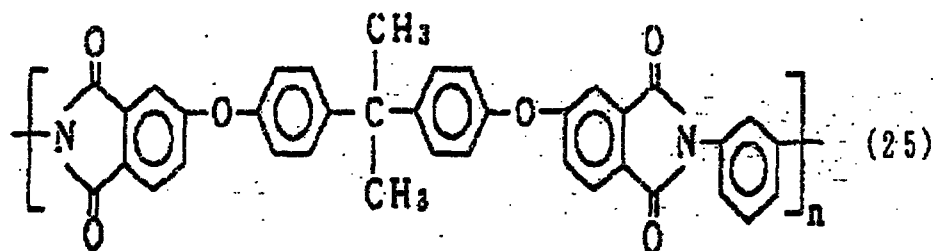
In the formula (13),  $n$  represents an integer of 1 to 1000, and  $R_1$  represents a group represented by the following formulas (14) to (19).



$R_2$  represents a group represented by the following formulas (20) to (24).



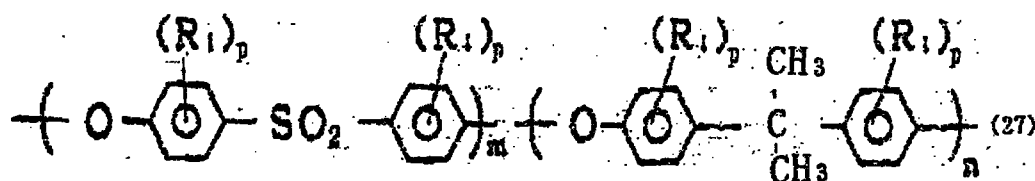
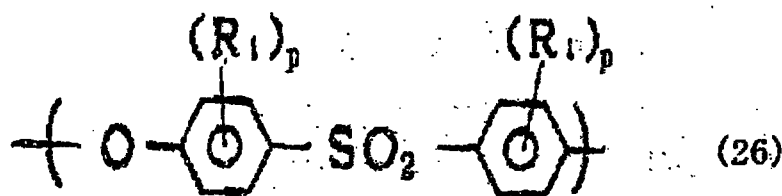
Preferable example from the view points of property and production cost includes the structure represented by the following formula (25),



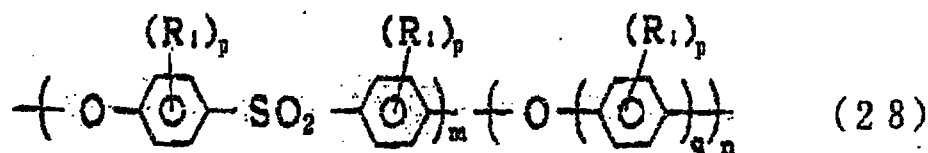
wherein  $n$  represents an integer of 1 to 1000.

The aromatic polysulfone resin used in the present invention is defined as the polyarylene compound wherein arylene units are located in disorder or order with ether bond and sulfone bond. Those having a repeating unit represented by the following formulas (26) to (28) are mentioned as typical examples, but it is not limited to these.





The compound of the formula (26) includes a homopolymer and the compound of the formula (27) includes a random copolymer, wherein  $\text{R}_1$  represents an alkyl group having 1 to 6 carbon atoms, an alkenyl group having 3 to 10 carbon atoms, a phenyl group or a halogen atom,  $p$  is an integer of 0 to 4, besides,  $m$  and  $n$  represent mean repeating unit numbers and are integers of 1 to 1000, each  $\text{R}_1$  on the same or different nucleus may be different mutually, and each  $p$  may be different mutually.



The compound of the formula (28) includes a random copolymer, wherein  $\text{R}_1$  represents an alkyl group having 1 to 6 carbon atoms, an alkenyl group having 3 to 10 carbon atoms, a

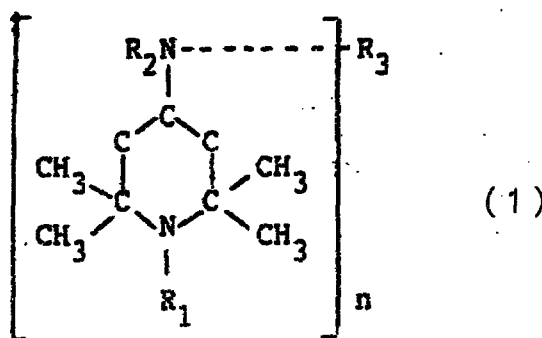


phenyl group or halogen atom,  $p$  is an integer of 0 to 4, besides,  $q$ ,  $m$  and  $n$  represent mean repeating unit numbers,  $q$  is an integer of 1 to 3,  $m$  and  $n$  are an integer of 0 to 100, each  $R_1$  on the same or different nucleus may be different mutually, and each  $p$  may be different mutually.

As the aromatic polysulfone resin used in the present invention,  $m/(m+n)$  in the structure unit of the formula (27) is preferably 0.8 or more. Besides, it is preferred that  $m/(m+n)$  in the structure unit of the formula (28) is 0.8 or more and  $q$  is 1.

Among these, the one having the repeating unit of the formula (26) or (27) is preferred.

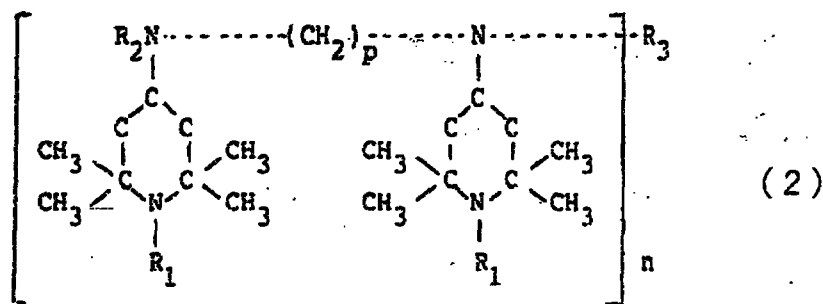
The 4-amino-2,2,6,6-tetramethylpiperidyl compound (c) having a steric hindrance type amino group as a basic skeleton is represented by the following formula (1) or (2):



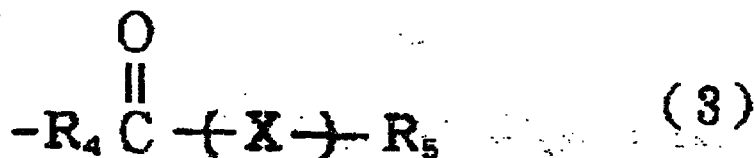
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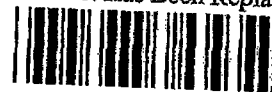


; wherein  $\text{R}_1$  represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group.  $\text{R}_2$  represents a hydrogen atom, an alkyl group or a group represented by the following formula (3),

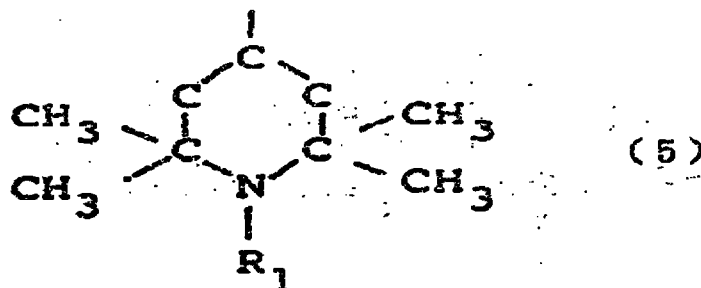
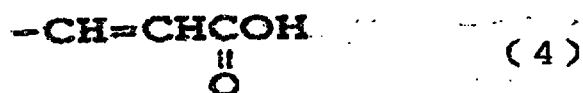


in which X represents a direct bond, -O- or -NH-,  $\text{R}_4$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and  $\text{R}_5$  represents a hydrogen atom, an alkyl group, an aryl group or a group represented by the following formula (4) or (5),

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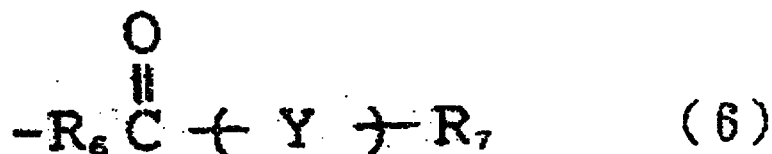
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; and wherein n represents an integer of 1 to 4, p represents an integer of 2 to 6, and R<sub>1</sub> represents an n valent organic group.

Preferable examples of the 4-amino-2,2,6,6-tetramethyl-piperidyl compound represented by the formula (1) or (2) are [I] to [VII] shown below.

[I] When n is 1 and R<sub>1</sub> is a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group, a benzyl group, an aryl group or a group represented by the following formula (6);



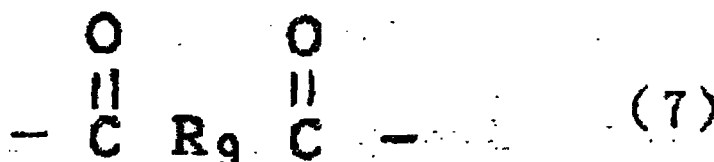
, wherein Y represents a direct bond, -O- or -NR<sub>8</sub>-, R<sub>6</sub> represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and R<sub>7</sub> and R<sub>8</sub>



each represents a hydrogen atom, a hydroxyl group, an alkyl group or an aryl group.

More preferably, when n is 1, R<sub>1</sub> and R<sub>2</sub> each represents a hydrogen atom or an alkyl group having 1-18 carbon atoms and R<sub>3</sub> represents a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group or the group represented by the formula (3) in which X represents a direct bond, -O- or -NR<sub>6</sub>-, R<sub>4</sub> represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group or arylene group, and R<sub>5</sub> and R<sub>6</sub> each represent a hydrogen atom, an alkyl group having 1-18 carbon atoms, an aryl group having 6-21 carbon atoms or a group represented by the formula (4) or (5).

[II] When n is 2, R<sub>3</sub> represents an alkylene group or a group represented by the following formula (7);



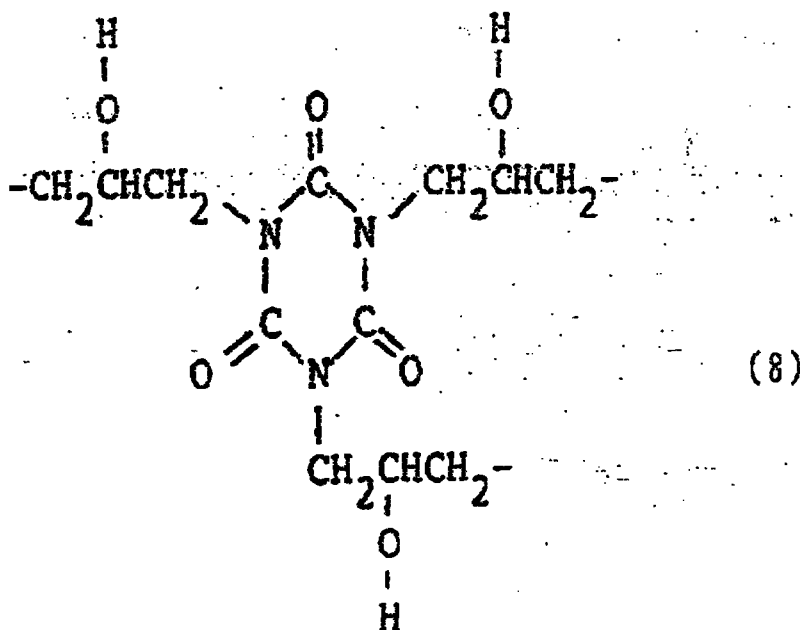
wherein R<sub>9</sub> represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group.

More preferably, when n is 2, R<sub>1</sub> and R<sub>2</sub> each represents a hydrogen atom or an alkyl group having 1-18 carbon atoms and R<sub>3</sub> represents an alkylene group or a group represented by the formula (7) in which R<sub>9</sub> represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group or an arylene group.



The compound in the formula (1), when n is 2,  $R_2$  represents a group represented by the formula (3) in which X represents a direct bond, -O- or -NH-,  $R_4$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group,  $R_5$  represents a hydrogen atom, an alkyl group, an aryl group or a group represented by the formula (4) or (5), and  $R_6$  represents an alkylene group, is also more preferable.

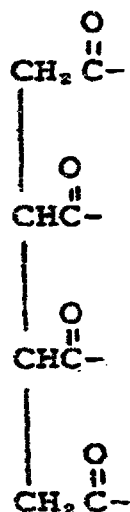
[III] When n is 3,  $R_3$  is a group represented by the following formula (8);



[IV] When n is 4,  $R_4$  is a group represented by the following formula (9) or (10);

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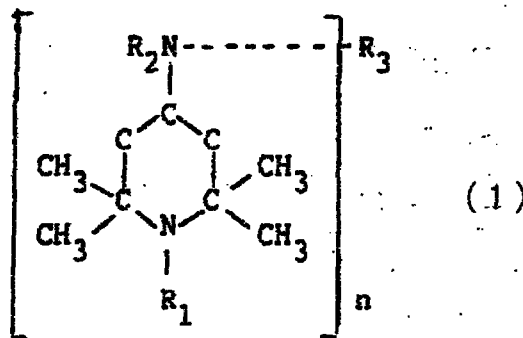




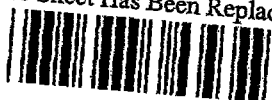
(9)



[V] Further, the compound represented by the following formula (1) is also preferable.

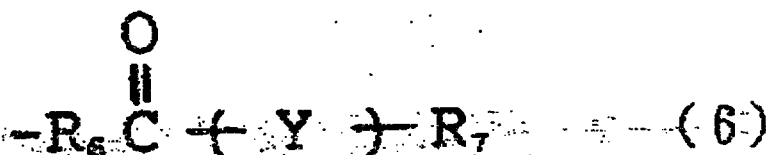


In the formula (1), R<sub>1</sub> represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group having 1-8 carbon atoms, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group, and n is an integer of 1, 2 or 4 ; and when n is 1, R<sub>2</sub> represents a hydrogen atom,

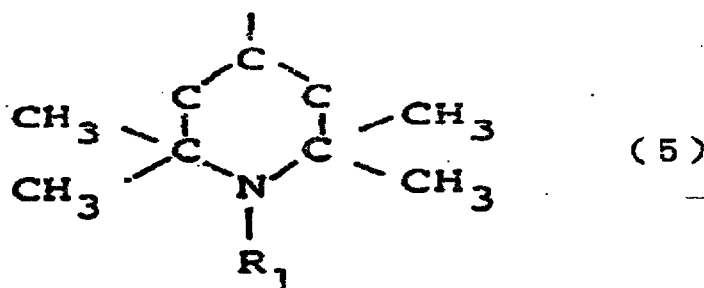
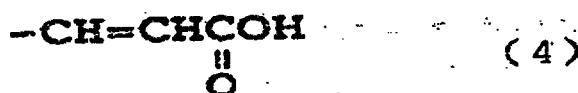


an alkyl group having 1-8 carbon atoms or  $\begin{array}{c} \text{O} \\ \parallel \\ -\text{C}-(\text{P})-(\text{Q})-\text{C}-\text{OH} \\ \parallel \\ \text{O} \end{array}$  in

which P and Q represent one selected from the group consisting of an alkylene group, an alkylidene group, a vinyl group and a vinylidene group, and  $R_3$  represents a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group, a benzyl group, an aryl group or a group represented by the following formula (6);



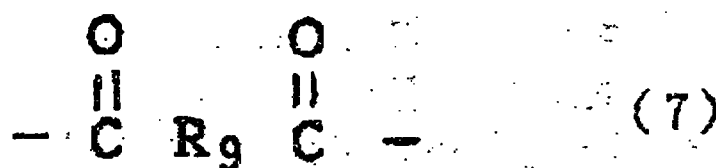
in which Y represents a direct bond,  $-\text{O}-$ ,  $-\text{NR}_8-$ ,  $R_6$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and  $R_7$  and  $R_8$  each represents a hydroxyl group, an alkyl group, an aryl group or a group represented by the following formula (4) or (5),



; when n is 2, R<sub>2</sub> represents a hydrogen atom, an alkyl group having 1-8 carbon atoms or -C-(P)-(Q)-C-OH in which P and Q

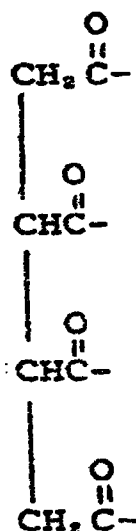


represent one selected from the group consisting of an alkylene group an alkylidene group, a vinyl group and a vinxylydene group, and R<sub>3</sub> represents an alkylene group or a group represented by the following formula (7),



in which R<sub>9</sub> represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group;

and when n is 4, R<sub>2</sub> represents a hydrogen atom or an alkyl group having 1-8 carbon atoms, and R<sub>3</sub> represents a group represented by the following formula (9) or (10),



(9)

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More preferably, in the formula (1),  $R_1$  and  $R_2$  each represents a hydrogen atom or an alkyl group having 1-8 carbon atoms, and

(i) when  $n$  is 1,  $R_3$  is a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group or a group represented by the formula (6) in which  $Y$  represents a direct bond,  $-O-$  or

$-\text{NR}_4-$ ,  $R_4$  represents a direct bond, a straight or branched

alkylene group having 1-10 carbon atoms, an alkylidene group

or arylene group, and  $R_5$  and  $R_6$  each represent a hydroxyl

group, an alkyl group having 1-18 carbon atoms, an aryl group having 6-21 carbon atoms or a group represented by the formula

(4) or (5),

and (ii) when  $n$  is 2,  $R_3$  represents an alkylene group or a

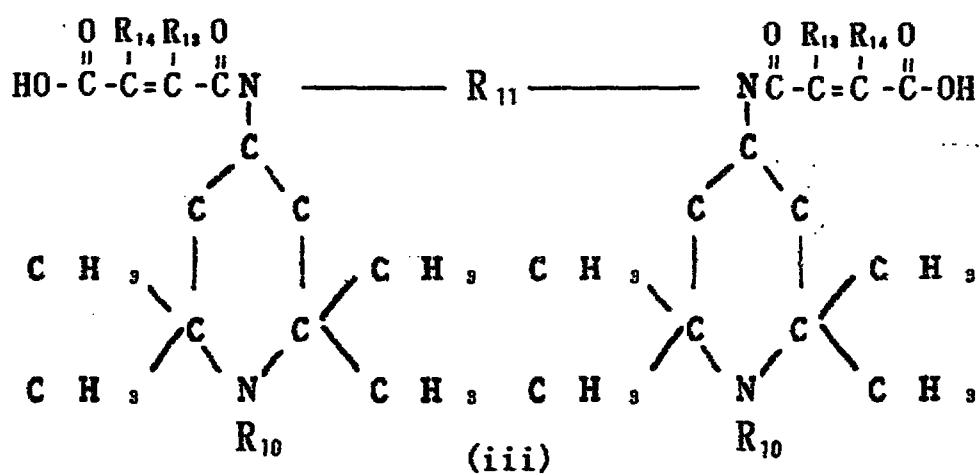
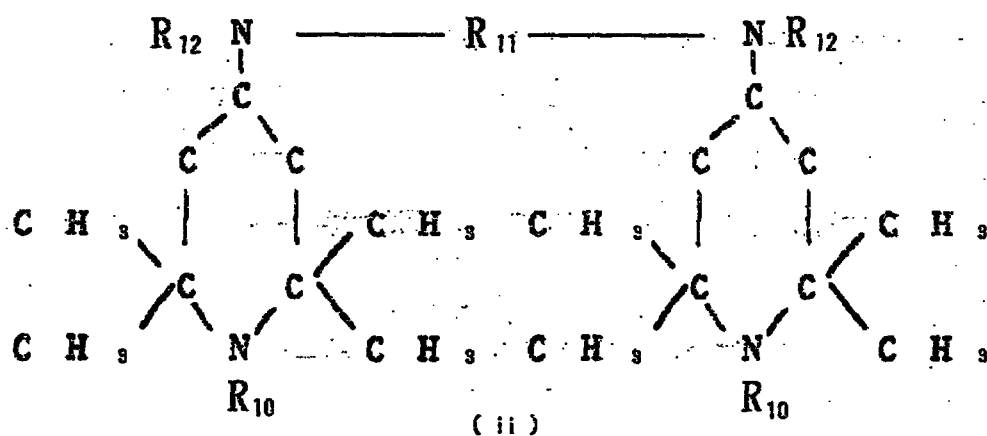
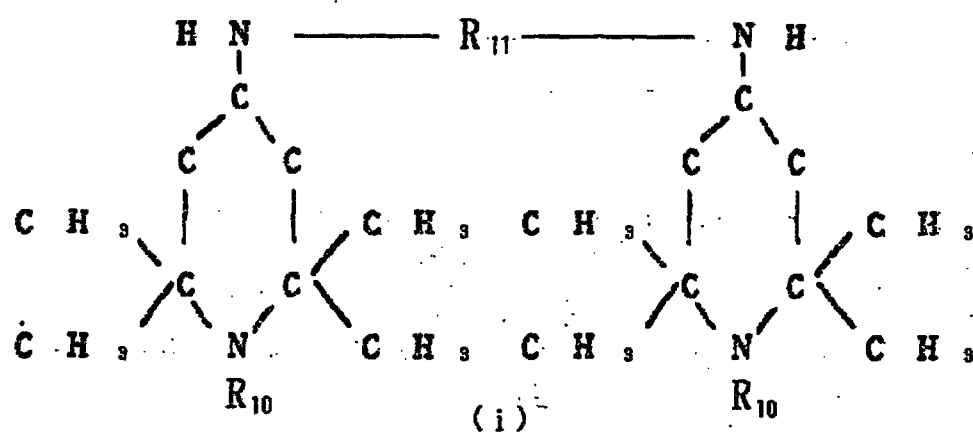
group represented by the formula (7) in which  $R_7$  represents a

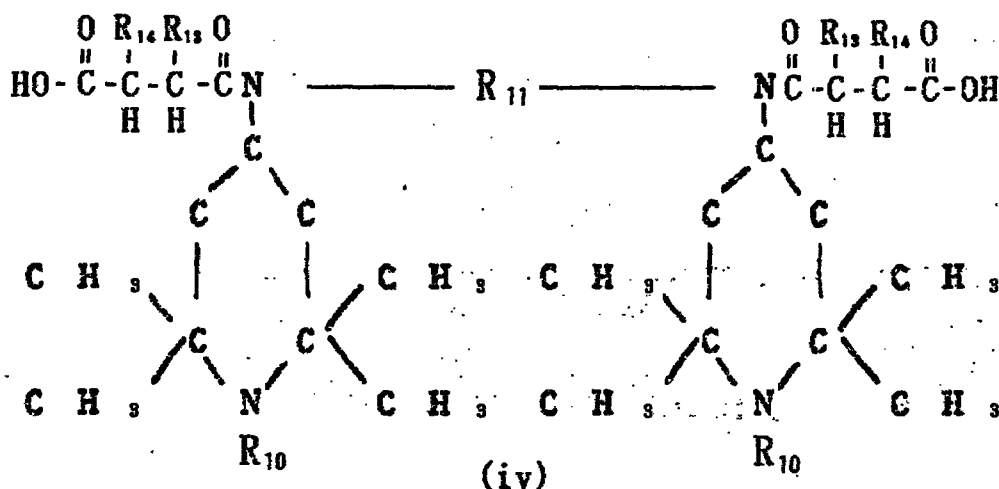
direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group or an arylene group.

Further, at least one compound represented by the following formulas (i)-(iv) is also more preferable ;

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wherein  $\text{R}_{10}$  represents a hydrogen atom, an alkyl group or an alkyloxy group,  $\text{R}_{11}$  represents an alkylene group,  $\text{R}_{12}$  represents an alkyl group, and  $\text{R}_{13}$  and  $\text{R}_{14}$  each represents a hydrogen atom or a methyl group.

In the formula (i), preferably,  $\text{R}_{10}$  represents a hydrogen atom, a hydroxyl group, an alkyl group having 1-10 carbon atoms or an alkyloxy group having 1-10 carbon atoms, and  $\text{R}_{11}$  represents an alkylene group having 2-8 carbon atoms, and more preferably,  $\text{R}_{10}$  represents a hydrogen atom or a methyl group and  $\text{R}_{11}$  represents a hexamethylene group.

In the formula (ii), preferably,  $\text{R}_{10}$  represents a hydrogen atom, a hydroxyl group, an alkyl group having 1-10 carbon atoms or an alkyloxy group having 1-10 carbon atoms,  $\text{R}_{11}$  represents an alkylene group having 2-8 carbon atoms, and  $\text{R}_{12}$  represents an alkyl group having 1-18 carbon atoms, and more preferably,  $\text{R}_{10}$  represents a hydrogen atom or a methyl group.

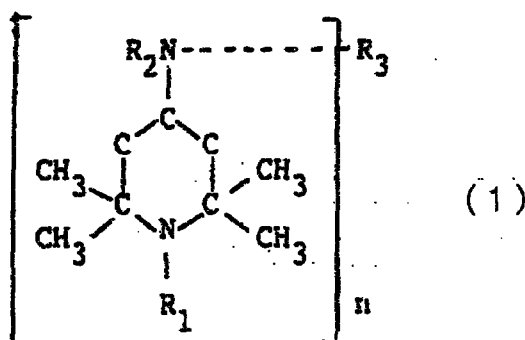


In the formula (iii), preferably,  $R_{10}$  represents a hydrogen atom, a hydroxyl group, an alkyl group having 1-10 carbon atoms or an alkyloxy group having 1-10 carbon atoms, and  $R_{11}$  represents an alkylene group having 2-8 carbon atoms, and more preferably,  $R_{10}$  represents a hydrogen atom or a methyl group.

In the formula (iv), preferably,  $R_{10}$  represents a hydrogen atom, a hydroxyl group, an alkyl group having 1-10 carbon atoms or an alkyloxy group having 1-10 carbon atoms, and  $R_{11}$  represents an alkylene group having 2-8 carbon atoms, and more preferably,  $R_{10}$  represents a hydrogen atom or a methyl group.

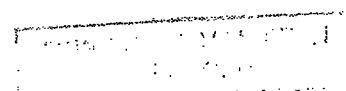
[VI] The compound represented by the following formula

(1) ;

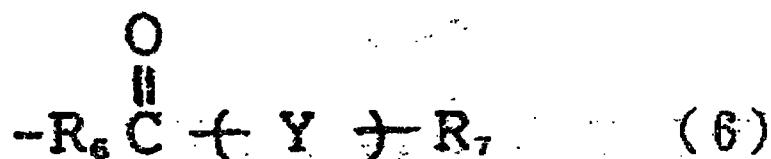


wherein  $R_1$  represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group having 1-8 carbon atoms, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group,  $R_2$  represents a hydrogen atom or alkyl group, ; and

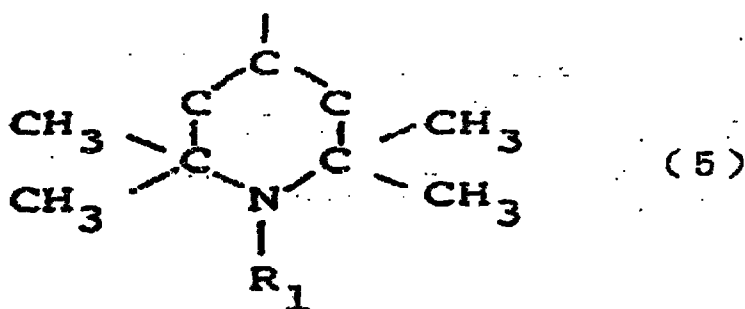
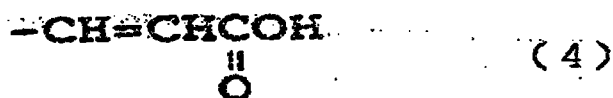
(i) when  $n$  is 1,  $R_3$  represents a hydrogen atom, an alkyl group,



a cycloalkyl group, an alkenyl group, a benzyl group, an allyl group or a group represented by the following formula (6),



in which Y represents a direct bond, -O- -NR<sub>8</sub>-, R<sub>6</sub> represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and R<sub>7</sub> and R<sub>8</sub> each represents a hydroxyl group, an alkyl group, an aryl group or a group represented by the following formula (4) or (5),

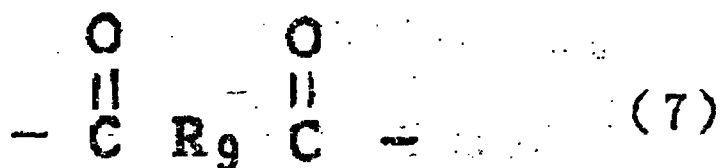


(ii) when n is 2, R<sub>3</sub> represents a group represented by the following formula (7),

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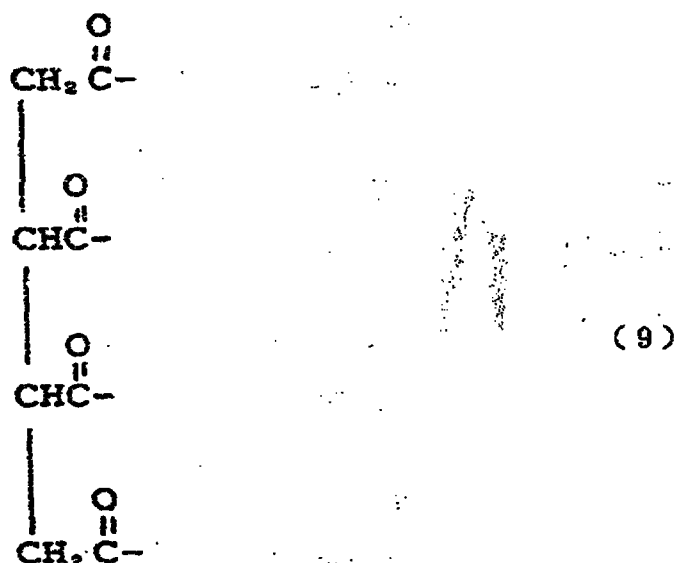


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in which  $\text{R}_9$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and

(iv) when  $n$  is 4,  $\text{R}_3$  represents a group represented by the following formula (9) or (10).

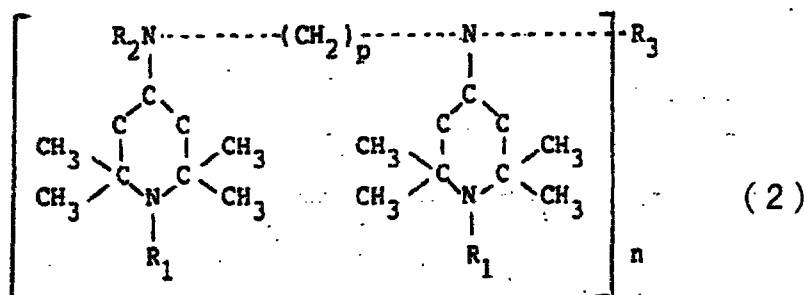


More preferably, in the formula (1),  $\text{R}_1$  represents a hydrogen atom or an alkyl group having 1-8 carbon atoms,  $\text{R}_2$  represents a hydrogen atom or an alkyl group having 1-18 carbon atoms ; and when  $n$  is 1,  $\text{R}_3$  represents a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group or a group represented by



the formula (6) in which Y represents a direct bond, -O- or -NR<sub>6</sub>-, R<sub>6</sub> represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group having 1-10 carbon atoms or arylene group, and R<sub>7</sub> and R<sub>8</sub> each represent a hydrogen atom, an alkyl group having 1-18 carbon atoms, an aryl group having 6-21 carbon atoms or a group represented by the formula (4) or (5); and when n is 2, R<sub>3</sub> represents a group represented by the formula (7) in which R<sub>9</sub> represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group having 1-10 carbon atoms or an arylene group.

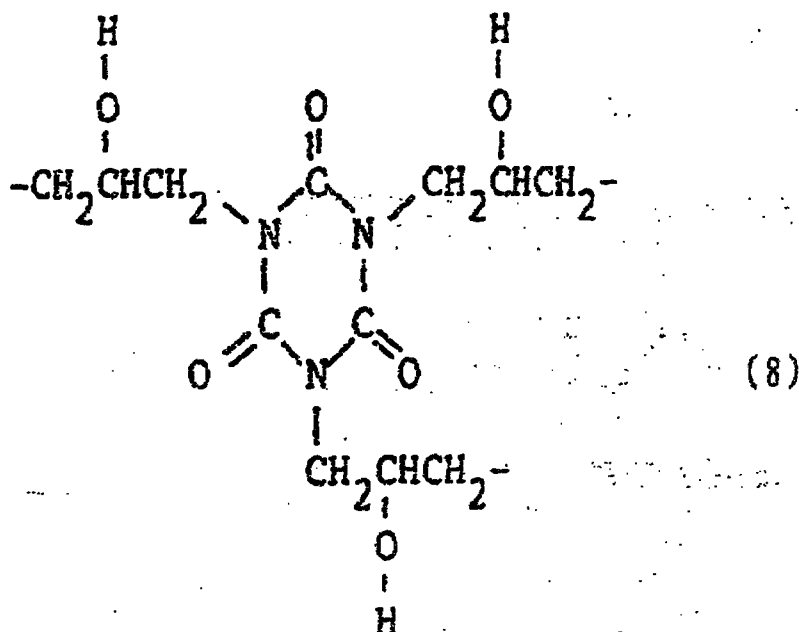
[VII] The compound represented by the following formula (2);



wherein R<sub>1</sub> represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group having 1-8 carbon atoms, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group, R<sub>2</sub> represents a hydrogen atom or an alkyl group having 1-8 carbon atoms, n is an integer of 3, p is an integer of 2 to 8, and R<sub>3</sub> represents a group

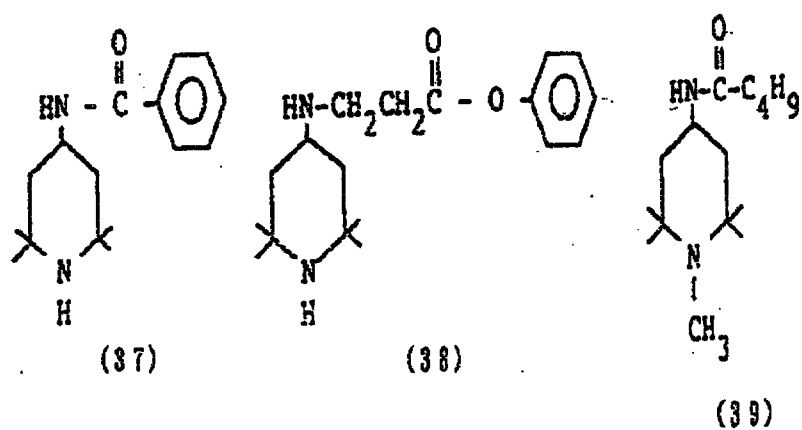
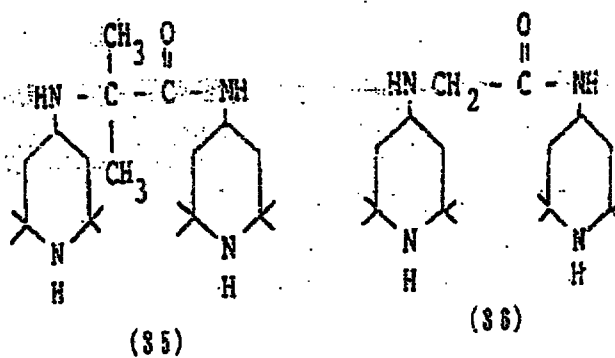
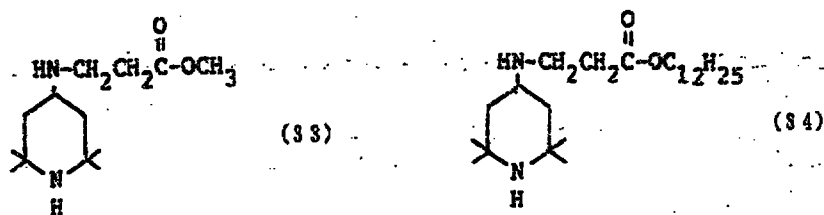
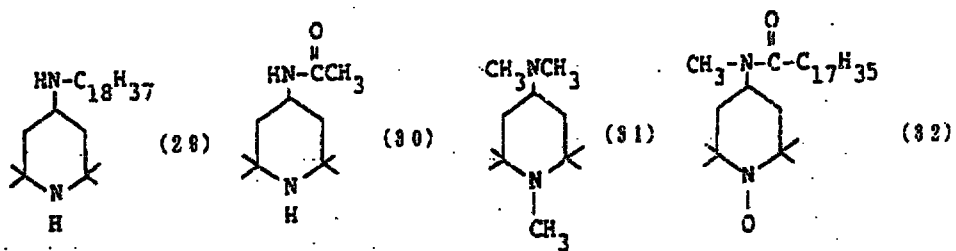


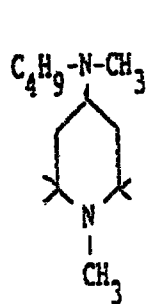
represented by the following formula (8) ;



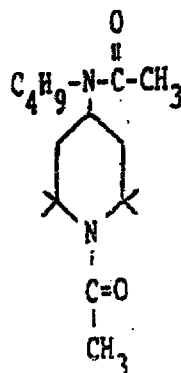
Examples of [n=1] of [I], [V] and [VI] of the compound (c) of the present invention include 4-Amino-2,2,6,6-tetramethyl-piperidine, 4-methyl-amino-2,2,6,6-tetramethyl-4-piperidine, 4-ethylamino-2,2,6,6-tetra-methylpiperidine, 4-propylamino-2,2,6,6-tetramethylpiperidine, 4-butylamino-2,2,6,6-tetra-methylpiperidine, 4-hexylamino-2,2,6,6-tetramethylpiperidine, 4-octylamino-2,2,6,6-tetra-methylpiperidine, 4-dodecanyl-amino-2,2,6,6-tetramethyl-piperidine, 4-diethylamino-2,2,6,6-tetramethylpiperidine, N-(2,2,6,6-tetramethyl-4-piperidyl) piperidine, N-(2,2,6,6-tetramethyl-4-piperidyl)acetamide and N-(2,2,6,6-tetramethyl-4-piperidyl)stearoylamide. Besides, mention may be made of the compounds represented by the following formulas (29) to (47).



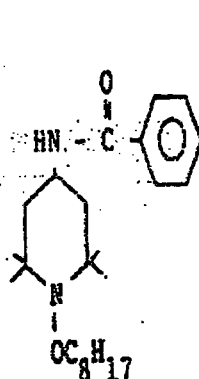




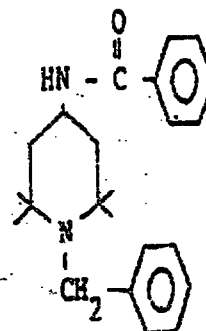
(40)



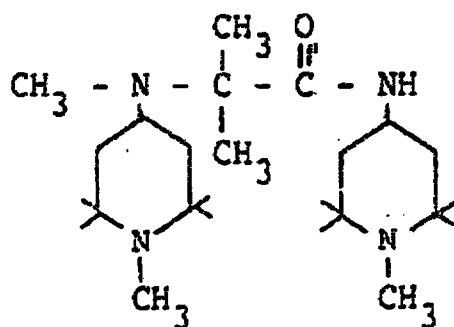
(41)



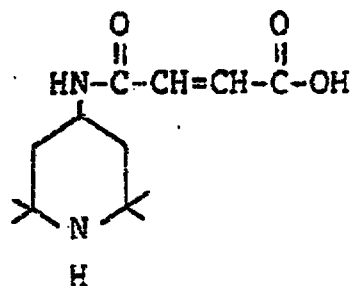
(42)



(43)



(44)

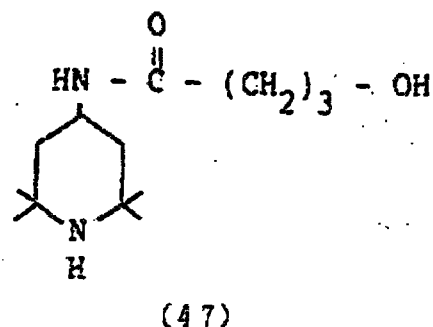
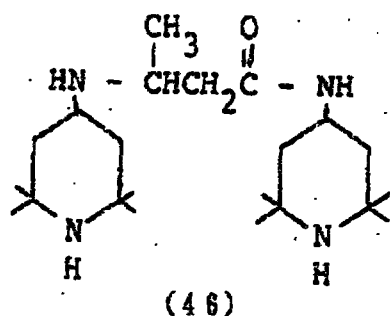


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Examples of [n=2] of [II], [V] and [VI] of the compound (c) of the present invention include compounds below.

In the formula (1) or (2),  $R_1$  includes, for example, a hydrogen atom, a hydroxyl group, an alkyl group of 1-10 carbon atoms such as methyl, ethyl or propyl, or an alkyloxy group of 1-10 carbon atoms and further includes cyclic ones such as cyclohexylmethyl and cyclohexylethyl in addition to straight or branched ones when the carbon number is 3 or more.  $R_1$  is preferably a hydrogen atom or a methyl group.  $R_2$  is preferably an alkyl group having 1-18 carbon atoms or an alkyloxy group having 1-18 carbon atoms, such as methyl, ethyl, propyl, isopropyl, n-butyl, isobutyl, t-butyl, sec-butyl, n-pentyl, n-hexyl, 2-ethylhexyl, n-octyl, 1,1,3,3-tetramethylbutyl, n-decyl, n-dodecyl, n-octadodecyl, cyclohexyl, 3,3,5-trimethyl-cyclohexyl, cyclooctyl and cyclodecyl.  $R_3$  includes an alkylene group having 2-8 carbon atoms and alkylene groups represented by  $-\text{C}_n\text{H}_{2n}-$  such as ethylene, 1,2- or 1,3-propylene, 1,2-, 1,3- or 1,4-butylene, pentamethylene, 2,2-dimethyl-1,3-propylene, hexamethylene and octamethylene,



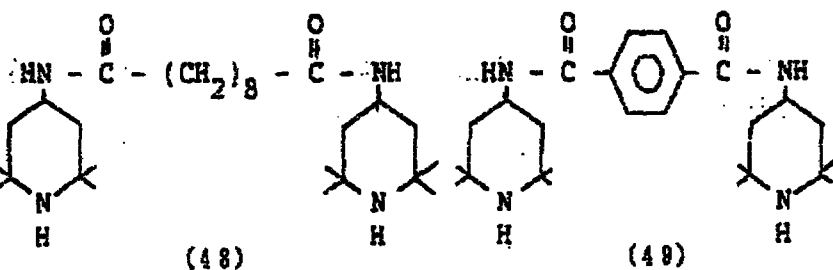
and cyclohexylene and cyclohexylenedimethylene.  $R_3$  is preferably a hexamethylene group having 6 carbon atoms.

In the formula (3),  $R_4$  is a hydrogen atom or an alkyl group having 1-18 carbon atoms such as methyl, ethyl, propyl, isopropyl, n-butyl, isobutyl, t-butyl, sec-butyl, n-pentyl, n-hexyl, 2-ethylhexyl, n-octyl, 1,1,3,3-tetramethylbutyl, n-decyl, n-dodecyl, n-octadodecyl, cyclohexyl, 3,3,5-trimethylcyclohexyl, cyclooctyl and cyclodecyl.

In the formula (7),  $R_5$  represents a direct bond, a straight or branched alkylene group of 1-10 carbon atoms, an alkylidene group or an arylene group such as, for example, ethylene, 1,2-, 1,3- or 1,4-propylene, 1,2-, 1,3- or 1,4-butylene,

pentamethylene, 2,2-dimethyl-1,3-propylene, hexamethylene, octamethylene, cyclohexylene and cyclohexylene-dimethylene.

Specific examples of the compounds include N,N'-bis(2,2,6,6-tetra-methyl-4-piperidyl)-1,6-hexanediamine, N,N'-bis(2,2,6,6-tetra-methyl-4-piperidyl)ethylenediamine, N,N'-bis(2,2,6,6-tetra-methyl-4-piperidyl)-1,8-octanediamine and a compound represented by the following formulas (48) to (53).



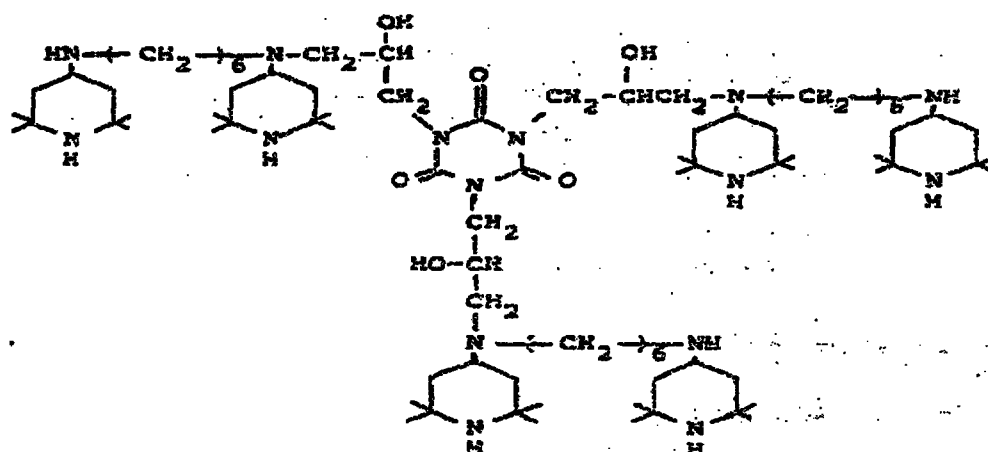


of the present invention include compounds below.

Specific examples of the compounds include a compound represented by the following formulas (54) and (55).



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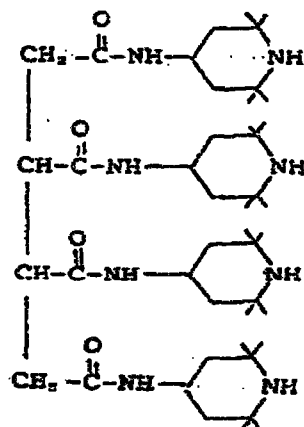


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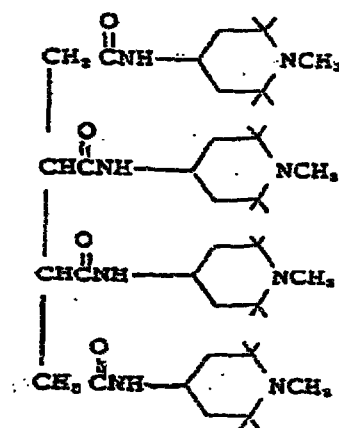
Examples of [n=4] of [I] and [IV] to [VI] of the compound (c) of the present invention include compounds below.

In the formula (1) or (2),  $R_1$  is the group represented by the formula (9) or (10).

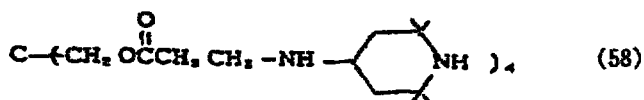
Specific examples of the compounds include a compound represented by the following formulas (56) to (58).



(56)



(57)



(58)



The amount ratio of the components (a) and (b) is ;

(a) 1 to 99 % by weight of a polyphenylene ether resin and

(b) 99 to 1 % by weight of at least one non-crystalline thermoplastic resin, preferably ;

(a) 10 to 90 % by weight of a polyphenylene ether resin and

(b) 90 to 10 % by weight of at least one non-crystalline thermoplastic resin, more preferably ;

(a) 20 to 90 % by weight of a polyphenylene ether resin and

(b) 80 to 10 % by weight of at least one non-crystalline thermoplastic resin, and most preferably ;

(a) 40 to 85 % by weight of a polyphenylene ether resin,

(b) 60 to 15 % by weight of at least one non-crystalline thermoplastic resin.

The amount of the component (c) is 0.001-15 parts by weight, preferably 0.1-10 parts by weight based on 100 parts by weight of the sum of (a) the polyphenylene ether resin and (b) the non-crystalline thermoplastic resin. When it is smaller than 0.001 part by weight, no effect to inhibit gelation is seen and when it is larger than 15 parts by weight, problems such as foaming occur at the time of actual processing owing to volatile matters.

In the present invention, fillers may be added in order to improve mechanical properties such as strength, stiffness and hardness to the thermoplastic resin composition.



The fillers include calcium carbonate, magnesium carbonate, aluminium hydroxide, magnesium hydroxide, zinc oxide, titanium oxide, magnesium oxide, magnesium sulfate, aluminium silicate, magnesium silicate, calcium silicate, silicic acid, calcium silicate hydrate, aluminium silicate hydrate, mineral fiber, zonotrite, potassium titanate whisker, magnesium oxysulfate, glass balloon, glass fiber, glass bead, inorganic fiber such as carbon fiber, stainless fiber, aluminum fiber and aramide fiber, carbon black, inorganic fillers such as a metal whisker, silica, alumina, talc, mica, clay, kaolinite,  $TiO_2$ ,  $ZnO$  and  $Sb_2O_3$ , and one or more kind of these fillers can be blended. Besides, the fillers used in the present invention are not restricted by these.

The thermoplastic resin composition of the present invention is obtained by blending and melt kneading the components (a), (b) and (c) with conventional methods. And the components (b) and (c) may be added in polymerization process of the component (a).

The thermoplastic resin composition of the present invention can be molded by various methods such as an injection molding, an extrusion molding, a blow molding, a sheet molding and film molding, which are most suitable since the composition is highly prevented from deterioration due to heat.

Besides, a molded article provided by the present



invention can be widely used as a packaging material, household appliances, an exterior material for automobiles parts and an interior decoration material such as trim parts of automobiles.

#### EXAMPLE

The Example of the present invention is illustrated in detail according to Examples and Comparative Examples as follows, but those are simple exemplifications, and the present invention is not limited to them.

Each Example and Comparative Example was obtained by blending the components shown in Tables 1 to 3. That is, those components were extruded with a twin-screw kneading machine (TEM-50A manufactured by Toshiba Machine Co., Ltd.) at a cylinder temperature of 260 °C, and pelletized to obtain the composition by a strand cutter after cooling with a water basin. Thus obtained pellets were dried at 130 °C for 2 hours under vacuum drying, and molded to obtain a test piece with an injection molding machine (IS220EN manufactured by Toshiba Machine Co., Ltd.) at a cylinder temperature of 290 °C, an injection pressure of 1200kg/cm<sup>2</sup> and a mold temperature of 80 °C.

Thus obtained pellets and the test piece were evaluated by the following method, and the results were obtained.



[The molecular weight of the polyphenylene ether]

The pellets provided were dissolved in chloroform and measured with a gel permeation chromatograph (GPC) (HLC-8020 manufactured by Toso Co., Ltd.; columns of K-806, K-805, K-804 and K-803; a detector of UV having a wave length of 290 nm; a temperature of 40 °C; a flow rate of 1.0 ml/min), and the molecular weight converted with that of the standard polystyrene was determined.

[Thermal resistance stability]

After a thermal hysteresis was given to the test piece having 3.2 mm in thickness for Izod impact test, obtained by the above-mentioned injection molding, by letting alone in a constant temperature of 150 °C for 100 and 500 hours, it was notched in accordance with ASTM D256, and Izod impact test was conducted at a temperature of 23 °C.

The materials illustrated as follows were prepared in order to obtain the compositions of Examples and Comparative Examples.

(Polyphenylene ether ; PPE)

The polyphenylene ether (PPE) was obtained by homo-polymerization of 2,6- dimethylphenol and having a logarithmic viscosity of 0.46 in chloroform solution (0.5g/dl) at 30°C. The molecular weight (Mw) of PPE before melt-kneaded was 46,100.

(Polycarbonate ; PC)

The trade name of CALIBRE 300-10 manufactured by



Sumitomo-Dow Co., Ltd., was used.

(Polymethyl methacrylate ; PMMA)

The trade name of Sumipex LG manufactured by Sumitomo Chemical Co., Ltd.

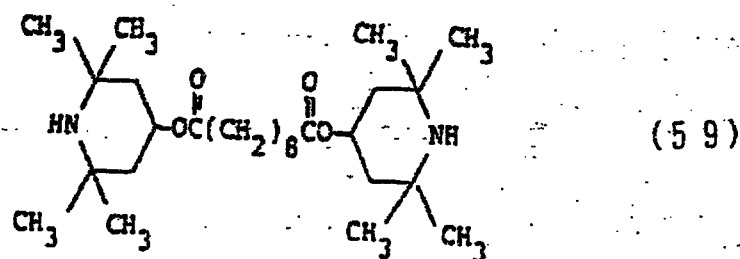
(Polystyrene)

Polystyrene 1 (PS-1) : The trade name of Sumibright E183N manufactured by Sumitomo Chemical Co., Ltd.

Polystyrene 2 (PS-2) : The trade name of Sumibright UG3000 manufactured by Sumitomo Chemical Co., Ltd.

(Additive)

Additive 1 : The trade name of Sanol LS-770 manufactured by Sankyo Co., Ltd. It is represented by the following chemical formula (59).



Additive 2 : The trade name of Tinuvin 622 manufactured by Ciba-Geigy Corp. It is represented by the following chemical formula (60).

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The compounds used in Examples are prepared by the Reference Examples shown below. Besides, % in those Reference Examples is by weight unless otherwise notified.

#### Reference Example 1

Preparation of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 1)

In a 5 liter autoclave were charged 981 g (6.32 moles) of 2,2,6,6-tetramethyl-4-piperidine, 350 g (3.01 moles) of hexamethylenediamine, 1500 g of methanol and 5.0 g of carbon containing 5 % platinum thereon with keeping the temperature of at 45 to 55°C. The inside of the autoclave was pressurized to 40 bars with hydrogen and heated to 70-80 °C to carry out hydrogenation. The hydrogenation was completed after 4 to 5 hours. The catalyst was removed by filtration at 70 to 80 °C under 2 to 3 bars, and 1070 g of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 1) was obtained by distillation. Yield : 90 %. Boiling point : 188 to 190 °C/0.80 mbar.

#### Reference Example 2

Preparation of N,N'-bis( 2-methoxycarbonyl ethyl )-N,N'-bis(2, 2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 2)

In a 1 liter four-necked flask were charged 80 g (0.20 moles) of N,N'-bis(2, 2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 1) obtained in Reference Example 1 and 200 g of methanol, and they were completely dissolved.

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Thereto was added dropwise of a solution prepared by dissolving 35 g (0.41 moles) of methyl acrylate in 100 g of methanol at room temperature. Then, reaction was allowed to proceed for 12 hours under reflux. After completion of the reaction, the solvent was distilled off and the residue was recrystallized from hexane to obtain 98 g (0.17 mol) of the Compound 2 as a white solid. Yield: 85 %. Melting point: 50-52 °C.

#### Reference Example 3

Preparation of N,N'-bis(2-octadecyloxycarbonylethyl)-N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 3)

In a four-necked flask provided with a stirrer and a reflux condenser were charged 20 g (35 mmoles) of N,N'-bis(2-methoxycarbonylethyl)-N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 2) obtained in Reference Example 2, 21 g (77 mmoles) of stearyl alcohol and 200 ml of toluene, followed by stirring under reflux. Thereto was added dropwise of a solution of 0.1 g (4 mmoles) of lithium amide in 3 g of methanol. Thereafter, the solvent was distilled off with addition of toluene and the reaction was completed. After the reaction was allowed to proceed for 4 hours, toluene was added, followed by cooling to room temperature. The organic layer was washed with water three times and then concentrated to obtain 23 g (22 mmol) of the Compound 3. Yield: 63 %. Melting



point : 45-46 °C.

#### Reference Example 4

Preparation of N,N'-bis( 2-t-butoxycarbonylethyl)-N,N'-bis(2, 2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 4)

The procedure of Reference Example 3 was repeated except that t-butyl alcohol was used instead of stearyl alcohol, thereby obtaining 13 g of the Compound 4. Melting point : 72 to 73 °C.

#### Reference Example 5

Preparation of N,N'-diacetyl-N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 5)

In a 300-ml four-necked flask were charged 20.7 g (50.7 mmols) of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 1) obtained in Reference Example 1 and 70 g of pyridine, and they were completely dissolved. Thereto was added dropwise of 11.64 g (0.114 moles) of acetic anhydride over a period of 30 minutes, followed by 1 hour at 40 °C. After cooling, 100 g 20 % aqueous NaOH and 50 ml of toluene were added and the product was extracted and washed with 50 ml of water. The solvent in the toluene layer was distilled off and the residue was recrystallized from 20 ml of n-hexane to obtain 20.6 g of the Compound 5. Yield : 85 %. Melting point : 152-154 °C.

#### Reference Example 6

Preparation of N,N'-bis(n-hexylcarbonyl)-N,N'-bis(2,2,6,6-

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tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 6)

The procedure of Reference Example 5 was repeated except that heptanoic anhydride was used in place of acetic anhydride, thereby obtaining 18.8 g of the Compound 6. Melting point : 123-125 °C.

Reference Example 7

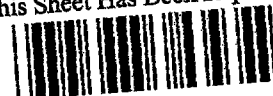
Preparation of N,N'-bis(ethoxycarbonyl)-N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 7)

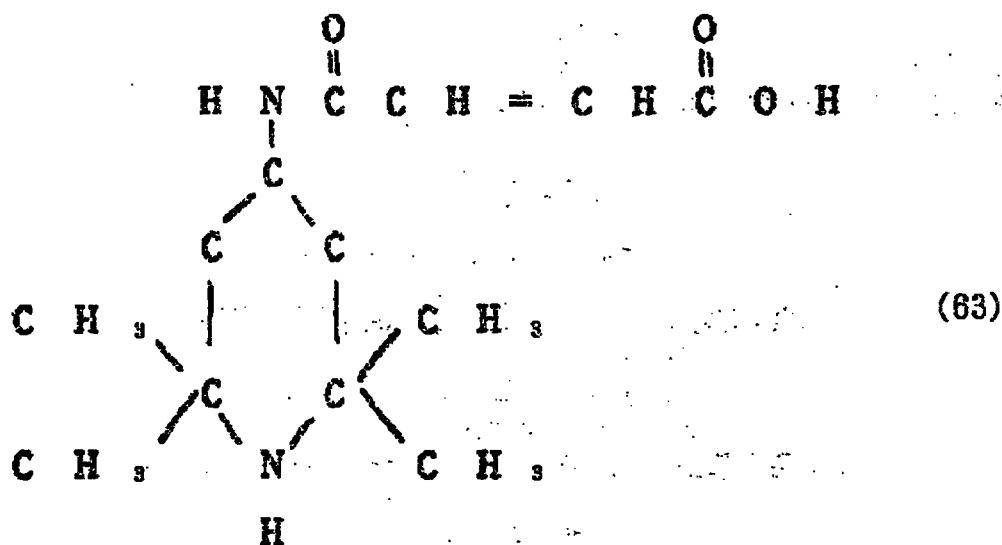
At a temperature of 0 °C or lower, 22.8 g (0.21 moles) of ethyl chlorocarbonate was added to solution of 39.4 g (0.1 moles) of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 1) obtained in Reference Example 1 in 200 ml of 1,2-dichloroethane which was cooled to -10 °C. Then, thereto was added slowly 8.4 g of sodium hydroxide dissolved in 50 ml of water with keeping the temperature of at 0 °C. Then, the temperature was elevated to 20 °C, and the reactant was separated to a water layer and an organic layer, and the organic layer was washed with water and dried over anhydrous sodium sulfate to remove the solvent. Then, the residue was crystallized from octane to obtain 44.1 g of the Compound 7. Yield : 82 %. Melting point : 125 °C.

Reference Example 8

Preparation of Compound 8 [the chemical formula (63)]

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In a four-necked flask provided with a thermometer and a stirrer were charged 156 g of 4-amino-2,2,6,6-tetramethyl-piperidine, 400 ml of toluene and 98g of maleic anhydride and the atmosphere in the flask was replaced with nitrogen, followed by keeping the temperature of at 60 °C for 6 hours with stirring. After completion of the reaction, the produced white crystal was filtrated and washed with toluene to obtain 243 g of the Compound 8.

#### Reference Example 9

The 2,2,6,6-tetramethyl-4-piperidylamine was used as Compound 9.

#### Reference Example 10

Preparation of methyl 2,2,6,6-tetramethyl-4-piperidyl-amino-propionate (Compound 10)

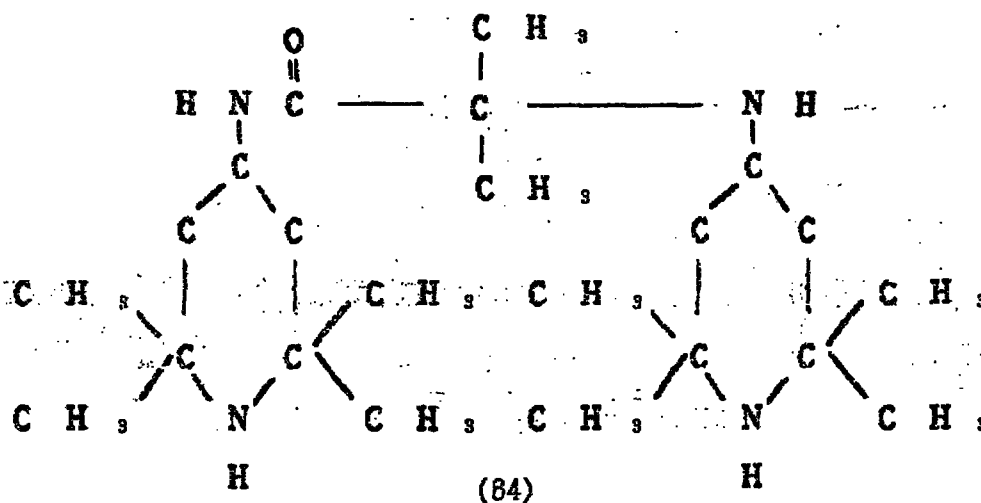
The 7.8 g (0.05 mol) of 2,2,6,6-tetramethyl-4-piperidine and 5.6 g (0.065 mol) of methyl acrylate were allowed to react



in 50 g of methanol under reflux for 3 hours. The solvent was distilled to obtain 8.0 g of colorless liquid of methyl 2,2,6,6-tetramethyl-4-piperidyl-aminopropionate (Compound 10) as a fraction of 130-136 °C/1.5mmHg.

Reference Example 11

Preparation of Compound 11 [the chemical formula (64)]



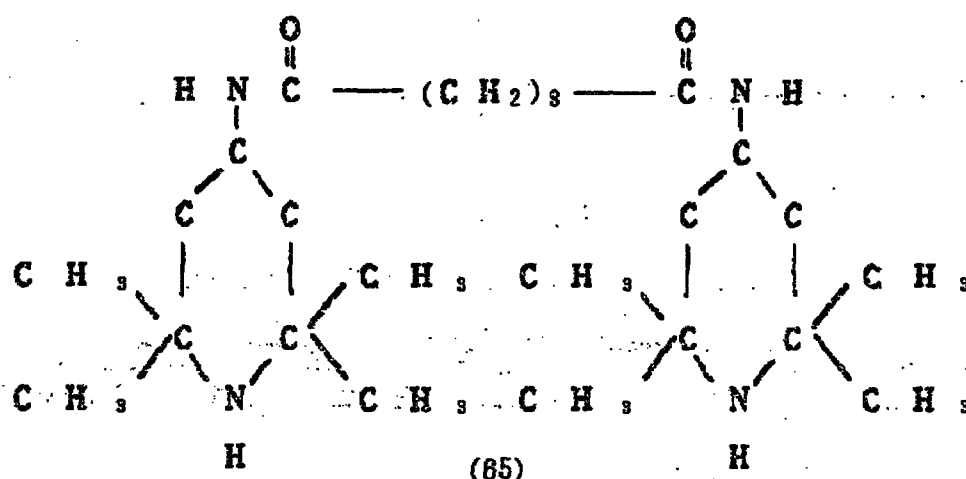
In a four-necked flask provided with a thermo-meter and a stirrer were charged 156 g of 4-amino-2,2,6,6-tetramethyl-piperidine, 32 g of acetone, 239 g of chloroform and 0.4 g of benzyltrimethylammonium chloride, and cooled to 5 °C with stirring. Then, thereto was added dropwise of 257.6 g of a 50 % aqueous potassium hydroxide solution over a period of 1 hour with keeping the temperature of at 5 to 10 °C, and thereafter, the reaction was allowed to proceed for 5 hours at 0 to 10 °C. After completion of the reaction, the aqueous layer was separated to a water layer and an organic layer, and the water



layer was removed. Then, the excess chloroform in the organic layer was distilled off to obtain 176 g of a white crystal as Compound 11. Yield : 92.6 %. Melting point : 127-128 °C. A parent ion-peak of 380 was confirmed by FD-mass spectrometry.

#### Reference Example 12

Preparation of Compound 12 [the chemical formula(65)]



In a four-necked flask provided with a thermo-meter and a stirrer were charged 156 g of 4-amino-2,2,6,6-tetramethylpiperidine, 400 ml of toluene and 121 g of triethylamine, and the atmosphere in the flask was replaced with nitrogen. After the content was cooled to 0 °C, thereto was added dropwise of 119 g of sebacoyl chloride under stirring with keeping the temperature of at 0 to 5 °C. After completion of the addition, 200 ml of 20% aqueous sodium hydroxide solution was added to dissolve the hydrochloride of triethylamine produced by the reaction. After the aqueous was separated to a water layer and an organic layer, the water layer was removed, and the



organic layer was washed with 100 ml of water. The organic layer was concentrated and the residue was crystallized from n-hexane to obtain 203 g of a white crystal as Compound 12.

#### Reference Example 13

Preparation of tris[2-hydroxy-3-(2,2,6,6-tetramethyl-4-piperidylamino)-propyl]-isocyanurate (Compound 13)

Tris(2,3-epoxypropyl)isocyanurate (29.7 g) and 4-amino-2,2,6,6-tetramethylpiperidine (47.5 g) were slowly heated in a hot bath under stirring. Heat was generated at about 80 °C. The temperature was raised up to  $100 \pm 5$  °C and the temperature was held for about two hours. After cooling, white powders of tris[2-hydroxy-3-(2,2,6,6-tetramethyl-4-piperidyl-amino)-propyl]-isocyanurate (74.5 g) (Compound 13) was obtained. Yield : 97.4 %. Melting point : 55-60 °C.

#### Examples 1 to 15

The blend ratios of Examples and the molecular weights of the polyphenylene ether in the obtained compositions (after melt-kneaded) were shown in Table 1.

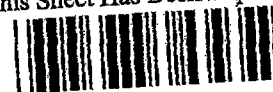
#### Comparative Examples 1 to 7

The compositions without the additives and Comparative Examples using the additives 1 to 4 were shown in Table 2.

#### Examples 16, 17 and Comparative Example 8

Thermal resistance stability of the compositions are shown in Table 3.

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EFFECT OF THE PRESENT INVENTION

Deterioration of polyphenylene ether caused by the heat at the time of processing can be markedly improved by adding a specific hindered piperidine compound to the composition comprising the polyphenylene ether and the non-crystalline thermoplastic resin.

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[Table 1]

|            | Composition |                                         | Molecular weight of<br>polyphenylene ether Mw |
|------------|-------------|-----------------------------------------|-----------------------------------------------|
|            |             |                                         |                                               |
| Example 1  | PPE<br>70   | Polystyrene 1<br>30<br>Compound 1<br>1  | 61,000                                        |
| Example 2  | PPE<br>70   | Polystyrene 1<br>30<br>Compound 2<br>1  | 51,000                                        |
| Example 3  | PPE<br>70   | Polystyrene 1<br>30<br>Compound 3<br>1  | 62,000                                        |
| Example 4  | PPE<br>70   | Polystyrene 1<br>30<br>Compound 4<br>1  | 65,000                                        |
| Example 5  | PPE<br>70   | Polystyrene 1<br>30<br>Compound 5<br>1  | 68,000                                        |
| Example 6  | PPE<br>70   | Polystyrene 1<br>30<br>Compound 6<br>1  | 68,000                                        |
| Example 7  | PPE<br>70   | Polystyrene 1<br>30<br>Compound 7<br>1  | 52,000                                        |
| Example 8  | PPE<br>70   | Polystyrene 1<br>30<br>Compound 8<br>1  | 64,000                                        |
| Example 9  | PPE<br>70   | Polystyrene 1<br>30<br>Compound 9<br>1  | 62,000                                        |
| Example 10 | PPE<br>70   | Polystyrene 1<br>30<br>Compound 10<br>1 | 68,000                                        |
| Example 11 | PPE<br>70   | Polystyrene 1<br>30<br>Compound 11<br>1 | 70,000                                        |
| Example 12 | PPE<br>70   | Polystyrene 1<br>30<br>Compound 12<br>1 | 69,000                                        |
| Example 13 | PPE<br>70   | Polystyrene 1<br>30<br>Compound 13<br>1 | 53,000                                        |
| Example 14 | PPE<br>70   | Polycarbonate<br>30<br>Compound 1<br>1  | 65,000                                        |
| Example 15 | PPE<br>70   | PMMA<br>30<br>Compound 2<br>1           | 58,000                                        |

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[Table 2]

|                          | Composition |                                        | Molecular weight of<br>polyphenylene ether Mw |
|--------------------------|-------------|----------------------------------------|-----------------------------------------------|
| Comparative<br>Example 1 | PPE<br>70   | Polystyrene 1<br>30                    | 76,000                                        |
| Comparative<br>Example 2 | PPE<br>70   | Polystyrene 1<br>30<br>Additive 1<br>1 | 77,000                                        |
| Comparative<br>Example 3 | PPE<br>70   | Polystyrene 1<br>30<br>Additive 2<br>1 | 79,000                                        |
| Comparative<br>Example 4 | PPE<br>70   | Polystyrene 1<br>30<br>Additive 3<br>1 | 75,000                                        |
| Comparative<br>Example 5 | PPE<br>70   | Polystyrene 1<br>30<br>Additive 4<br>1 | 77,000                                        |
| Comparative<br>Example 6 | PPE<br>70   | Polycarbonate<br>30                    | 82,000                                        |
| Comparative<br>Example 7 | PPE<br>70   | PMMA<br>30                             | 75,000                                        |

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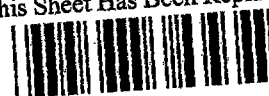
[Table 3]

Izod impact strength with a notch at 23 °C  
aging at 150 °C for

|                          |           | Initial             |                 | 100hours |     | 500hours |  |
|--------------------------|-----------|---------------------|-----------------|----------|-----|----------|--|
| Comparative<br>Example 8 | PPE<br>60 | Component           |                 | 7.2      | 2.1 | 2.3      |  |
|                          |           | Polystyrene 2<br>40 | Compound 1      |          |     |          |  |
| Example 16               | PPE<br>60 | Polystyrene 2<br>40 | 1               | 10.1     | 5.5 | 5.7      |  |
|                          |           | Polystyrene 2<br>30 | Compound 9<br>1 |          |     |          |  |
| Example 17               | PPE<br>70 |                     |                 | 8.8      | 4.6 | 4.8      |  |

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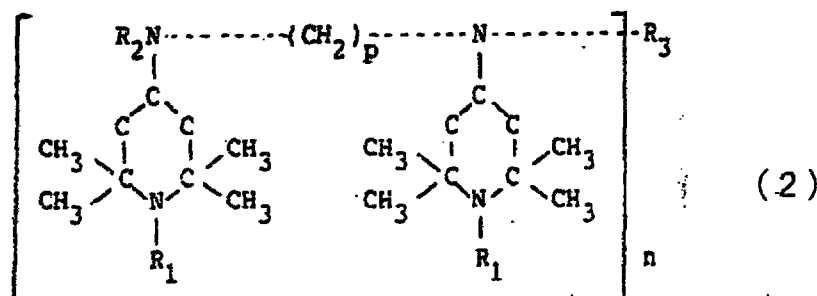
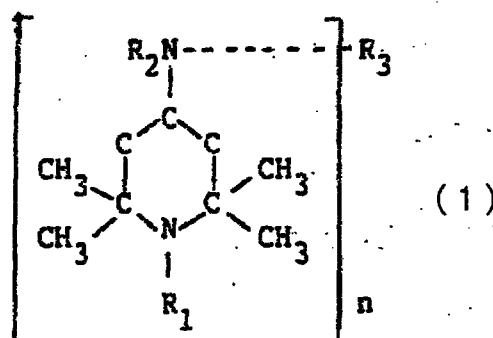


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WHAT IS CLAIMED IS:

1. A thermoplastic resin composition which comprises
- (a) 1 to 99 % by weight of a polyphenylene ether resin,
  - (b) 99 to 1 % by weight of at least one non-crystalline thermoplastic resin selected from an alkenyl-aromatic resin, a non-crystalline aromatic polyester resin, a methacrylate resin, a polyether imide resin and an aromatic polysulfone resin, and per 100 parts by weight of the sum of (a) and (b),
  - (c) 0.001 to 15 parts by weight of at least one 4-amino-2,2,6,6-tetramethylpiperidyl compound represented by the following formula (1) or (2):



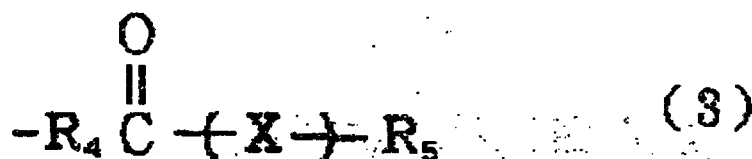
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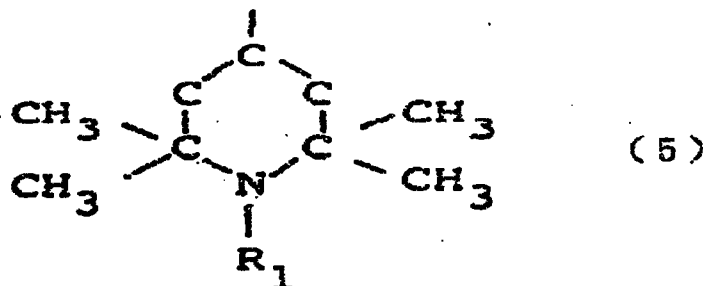
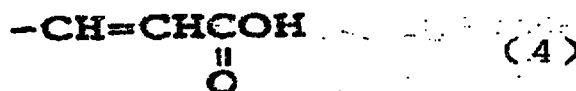
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; wherein  $R_1$  represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group.  $R_2$  represents a hydrogen atom, an alkyl group or a group represented by the following formula (3),



in which X represents a direct bond, -O- or -NH-,  $R_4$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and  $R_5$  represents a hydrogen atom, an alkyl group, an aryl group or a group represented by the following formula (4) or (5),



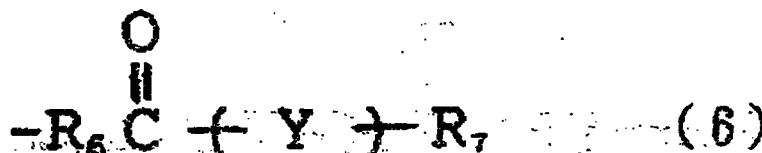
; and wherein n represents an integer of 1 to 4, p represents





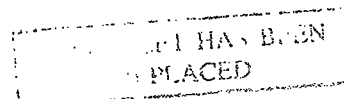
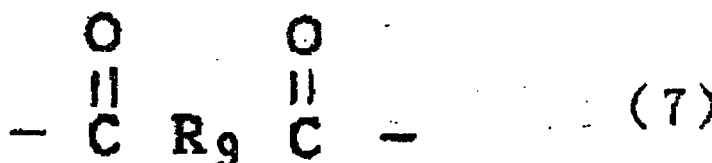
an integer of 2 to 6, and  $R_3$  represents an n valent organic group.

2. A thermoplastic resin composition according to claim 1, wherein in the formula (1) or (2), n is 1 and  $R_3$  is a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group, a benzyl group, an aryl group or a group represented by the following formula (6);



, wherein Y represents a direct bond,  $-O-$  or  $-NR_8-$ ,  $R_6$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and  $R_7$  and  $R_8$  each represents a hydrogen atom, a hydroxyl group, an alkyl group or an aryl group.

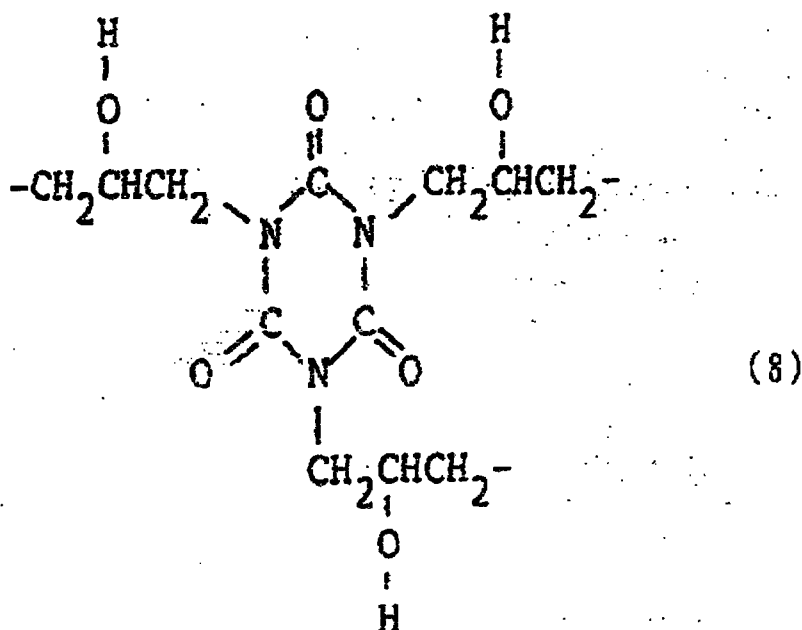
3. A thermoplastic resin composition according to claim 1, wherein in the formula (1) or (2), n is 2 and  $R_3$  is an alkylene group or a group represented by the following formula (7);





, wherein  $R_2$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group.

4. A thermoplastic resin composition according to claim 1, wherein in the formula (1) or (2),  $n$  is 3 and  $R_2$  is a group represented by the following formula (8);



5. A thermoplastic resin composition according to claim 1, wherein in the formula (1) or (2),  $n$  is 4 and  $R_2$  is a group represented by the following formula (9) or (10);

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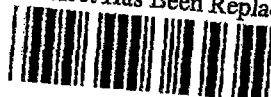
(8)



(10)

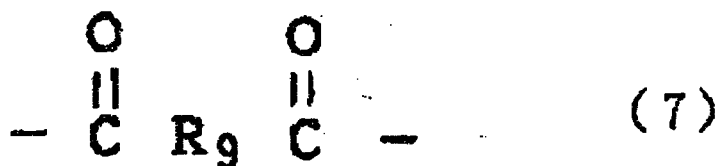
6. A thermoplastic resin composition according to claim 1, wherein in the formula (1),  $R_1$  and  $R_2$  each represents a hydrogen atom or an alkyl group having 1-18 carbon atoms and  $n$  is 1 or 2, and

(I) when  $n$  is 1,  $R_3$  is a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group or the group represented by the formula (3) in which  $X$  represents a direct bond,  $-O-$  or  $-NR_6-$ ,  $R_4$  represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group or arylene group, and  $R_7$  and  $R_8$  each represent a hydrogen atom, an alkyl group having 1-18 carbon atoms, an aryl group having 6-21 carbon atoms or a group represented by the formula (4) or (5), and (II) when  $n$  is 2,  $R_3$  is a group represented by the





following formula (7);



, wherein  $\text{R}_9$  represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group or an arylene group.

7. A thermoplastic resin composition according to claim 1, wherein in the formula (1),  $n$  is 2 and  $\text{R}_3$  is an alkylene group.

8. A thermoplastic resin composition according to claim 1, wherein in the formula (1),  $n$  is 2,  $\text{R}_2$  is a hydrogen atom and  $\text{R}_3$  is an alkylene group.

9. A thermoplastic resin composition according to claim 1, wherein in the formula (1),  $n$  is 2,  $\text{R}_1$  is a hydrogen atom or a methyl group,  $\text{R}_2$  is a hydrogen atom and  $\text{R}_3$  is a hexamethylene group.

10. A thermoplastic resin composition according to claim 1, wherein in the formula (1),  $n$  is 2,  $\text{R}_2$  is an alkyl group and  $\text{R}_3$  is an alkylene group.

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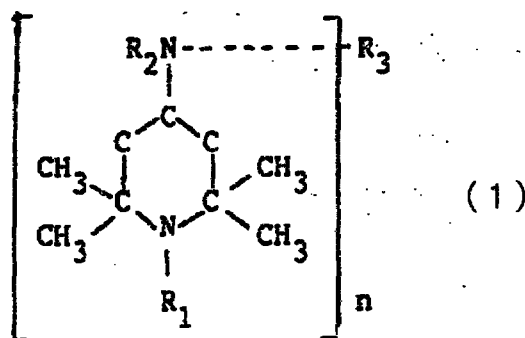




11. A thermoplastic resin composition according to claim 1, wherein in the formula (1),  $n$  is 2,  $R_2$  is a group represented by the formula (3) in which  $X$  represents a direct bond,  $-O-$  or  $-NH-$ ,  $R_4$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group,  $R_5$  represents a hydrogen atom, an alkyl group, an aryl group or a group represented by the formula (4) or (5), and  $R_3$  is an alkylene group.

12. A thermoplastic resin composition according to claim 1, wherein the non-crystalline thermoplastic resin (b) is an alkenylaromatic resin.

13. A thermoplastic resin composition according to claim 1, wherein the 4-amino-2,2,6,6-tetramethylpiperidyl compound (c) is a compound represented by the following formula (1) :



wherein  $R_1$  represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group having 1-8 carbon atoms, a cycloalkyl group, an allyl group, a benzyl group, an

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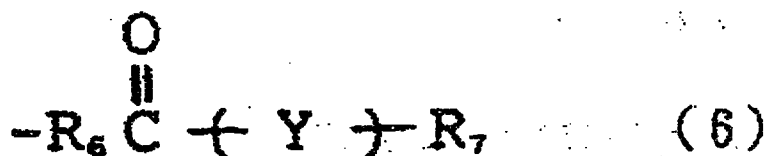


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aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group, and n is an integer of 1, 2, or 4 ; and when n is 1, R<sub>2</sub> represents a hydrogen atom, an alkyl group having 1-8 carbon atoms or  $\begin{array}{c} \text{O} \\ || \\ -\text{C}-(\text{P})-(\text{Q})-\text{C}-\text{OH} \\ || \\ \text{O} \end{array}$  in which P

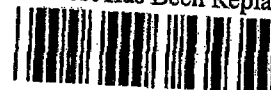
and Q represent one selected from the group consisting of an alkylene group, an alkylidene group, a vinyl group and a vinylidene group, and R<sub>3</sub> represents a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group, a benzyl group, an aryl group or a group represented by the following formula (6),



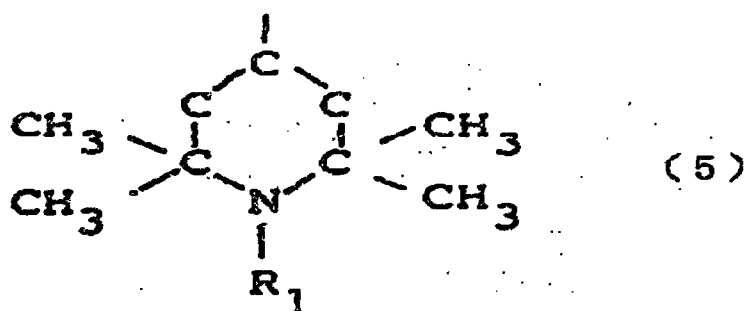
in which Y represents a direct bond, -O- -NR<sub>8</sub>-, R<sub>6</sub> represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and R<sub>7</sub> and R<sub>8</sub> each represents a hydroxyl group, an alkyl group, an aryl group or a group represented by the following formula (4) or (5),



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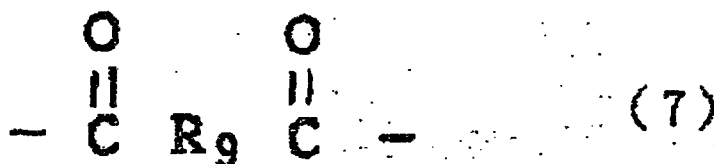


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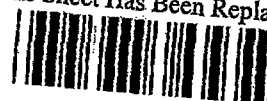
; when n is 2,  $\text{R}_2$  represents a hydrogen atom, an alkyl group having 1-8 carbon atoms or  $-\text{C}(=\text{O})-(\text{P})-(\text{Q})-\text{C}(=\text{O})-\text{OH}$  in which P:

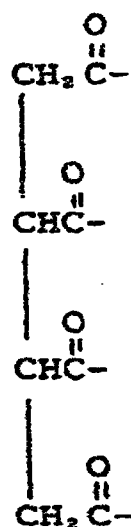
and Q represent one selected from the group consisting of an alkylene group, an alkylidene group, a vinyl group and a vinylylidene group, and  $\text{R}_3$  represents an alkylene group or a group represented by the following formula (7),



in which  $\text{R}_9$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group; and when n is 4,  $\text{R}_2$  represents a hydrogen atom or an alkyl group having 1-8 carbon atoms, and  $\text{R}_3$  represents a group represented by the following formula (9) or (10),

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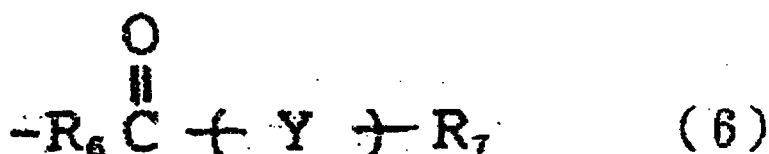
(9)



(10)

14. A thermoplastic resin composition according to claim 13, wherein in the formula (1),  $R_1$  and  $R_2$  each represents a hydrogen atom or an alkyl group having 1-8 carbon atoms and  $n$  is 1 or 2, and ;

(i) when  $n$  is 1,  $R_3$  is a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group or a group represented by the following formula (6),



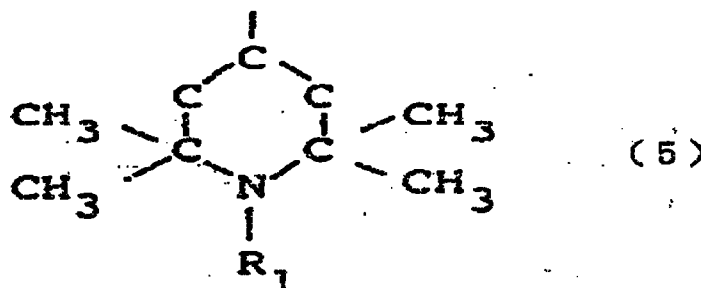
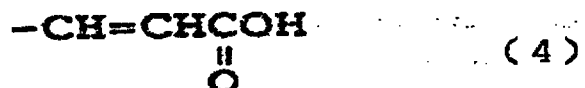
in which  $Y$  represents a direct bond,  $-O-$  or  $-NR_9-$ ,  $R_6$  represents a direct bond, a straight or branched alkylene

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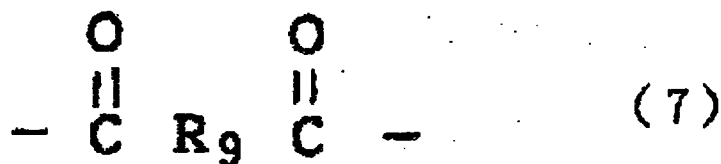




group having 1-10 carbon atoms, an alkylidene group or arylene group, and  $R_7$  and  $R_8$  each represent a hydroxyl group, an alkyl group having 1-18 carbon atoms, an aryl group having 6-21 carbon atoms or a group represented by the following formula (4) or (5),



and (ii) when  $n$  is 2,  $R_9$  represents a group represented by the following formula (7),



in which  $R_9$  represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group or an arylene group.

15. A thermoplastic resin composition according to claim 13, wherein in the formula (1),  $n$  is 2 and  $R_9$  represents an





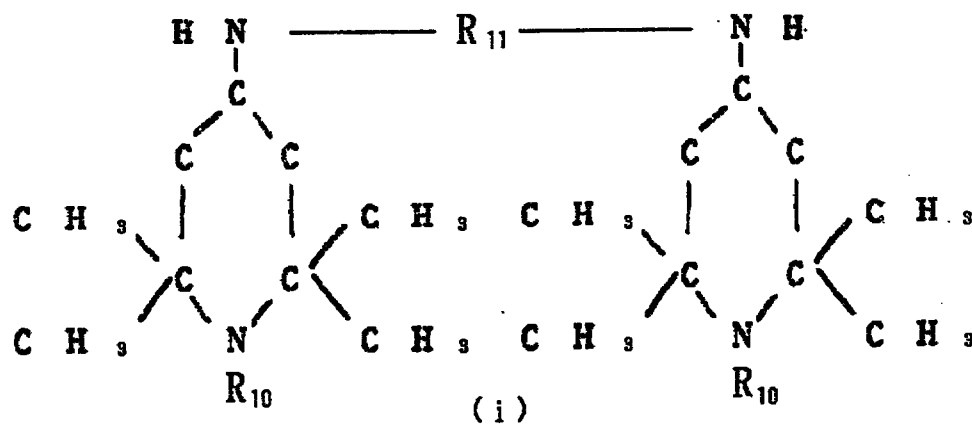
alkylene group.

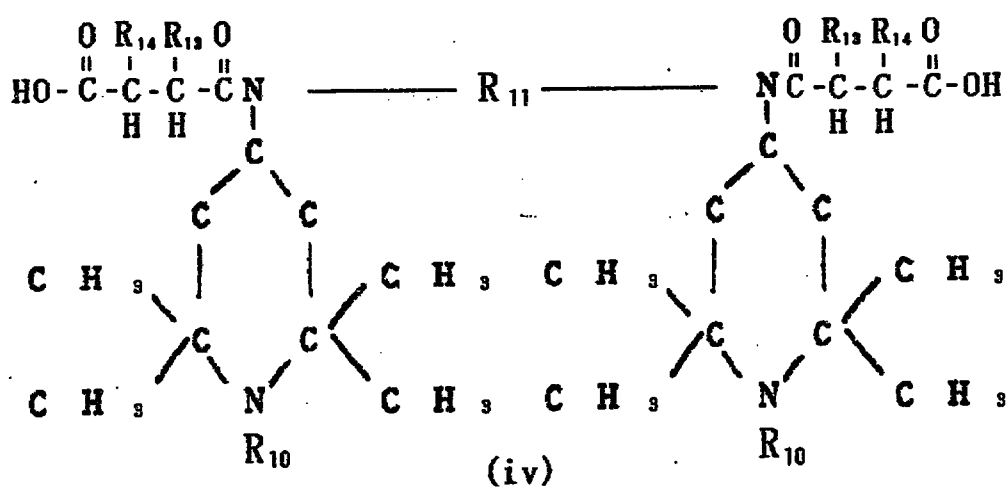
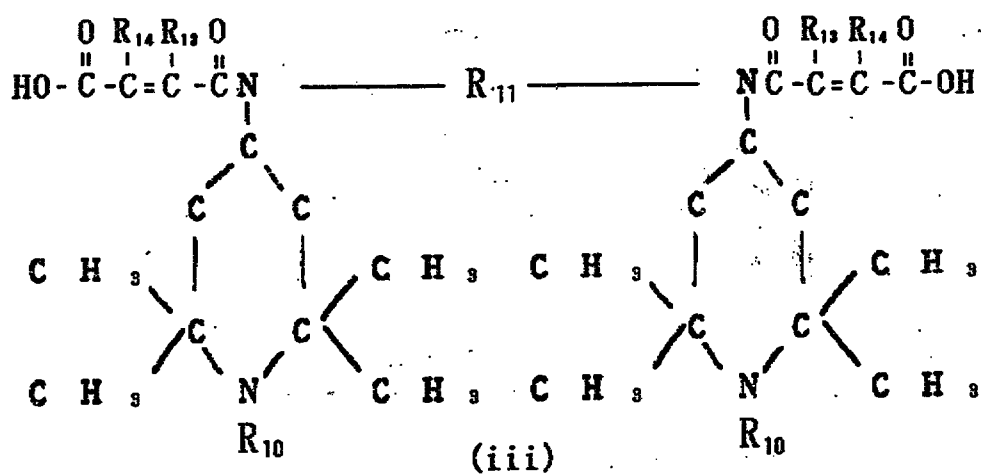
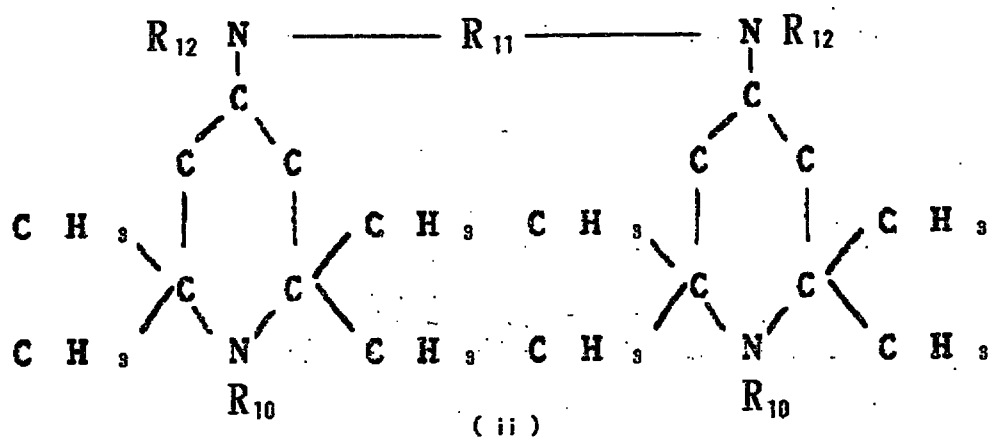
16. A thermoplastic resin composition according to claim 13, wherein in the formula (1),  $n$  is 2,  $R_2$  represents a hydrogen atom and  $R_3$  represents an alkylene group.

17. A thermoplastic resin composition according to claim 13, wherein in the formula (1),  $n$  is 2,  $R_1$  represents a hydrogen atom or a methyl group,  $R_2$  represents a hydrogen atom and  $R_3$  represents a hexamethylene group.

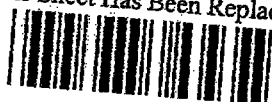
18. A thermoplastic resin composition according to claim 13, wherein in the formula (1),  $n$  is 2,  $R_2$  represents an alkyl group and  $R_3$  represents an alkylene group.

19. A thermoplastic resin composition according to claim 13, wherein the compound (b) represented by the formula (1) is at least one compound represented by the following formulas (i)-(iv):





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wherein  $R_{10}$  represents a hydrogen atom, an alkyl group or an alkyloxy group,  $R_{11}$  represents an alkylene group,  $R_{12}$  represents an alkyl group, and  $R_{13}$  and  $R_{14}$  each represents a hydrogen atom or a methyl group.

20. A thermoplastic resin composition according to claim 19, wherein in the formula (i),  $R_{10}$  represents a hydrogen atom, a hydroxyl group, an alkyl group having 1-10 carbon atoms or an alkyloxy group having 1-10 carbon atoms, and  $R_{11}$  represents an alkylene group having 2-8 carbon atoms.

21. A thermoplastic resin composition according to claim 19, wherein in the formula (i),  $R_{10}$  represents a hydrogen atom or a methyl group and  $R_{11}$  represents a hexamethylene group.

22. A thermoplastic resin composition according to claim 19, wherein in the formula (ii),  $R_{10}$  represents a hydrogen atom, a hydroxyl group, an alkyl group having 1-10 carbon atoms or an alkyloxy group having 1-10 carbon atoms,  $R_{11}$  represents an alkylene group having 2-8 carbon atoms, and  $R_{12}$  represents an alkyl group having 1-18 carbon atoms.

23. A thermoplastic resin composition according to claim 19, wherein in the formula (ii),  $R_{10}$  represents a hydrogen atom or a methyl group.

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24. A thermoplastic resin composition according to claim 19, wherein in the formula (iii),  $R_{10}$  represents a hydrogen atom, a hydroxyl group, an alkyl group having 1-10 carbon atoms or an alkyloxy group having 1-10 carbon atoms, and  $R_{11}$  represents an alkylene group having 2-8 carbon atoms.

25. A thermoplastic resin composition according to claim 19, wherein in the formula (iii),  $R_{10}$  represents a hydrogen atom or a methyl group.

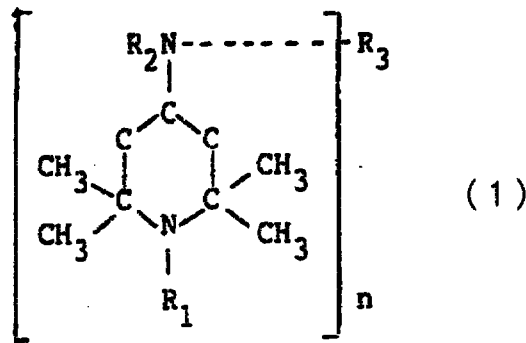
26. A thermoplastic resin composition according to claim 19, wherein in the formula (iv),  $R_{10}$  represents a hydrogen atom, a hydroxyl group, an alkyl group having 1-10 carbon atoms or an alkyloxy group having 1-10 carbon atoms, and  $R_{11}$  represents an alkylene group having 2-8 carbon atoms.

27. A thermoplastic resin composition according to claim 19, wherein in the formula (iv),  $R_{10}$  represents a hydrogen atom or a methyl group.

28. A thermoplastic resin composition according to claim 1, wherein the 4-amino-2,2,6,6-tetramethylpiperidyl compound (c) is a compound represented by the following formula (1) :

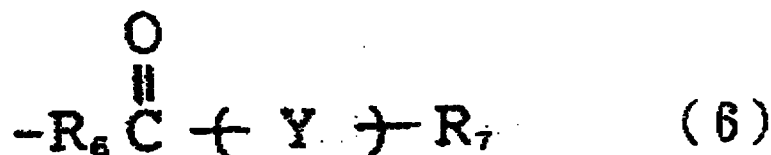
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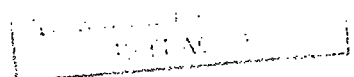
wherein  $\text{R}_1$  represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group having 1-8 carbon atoms, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group,  $\text{R}_2$  represents a hydrogen atom or alkyl group, and  $n$  is an integer of 1, 2 or 4 ; and

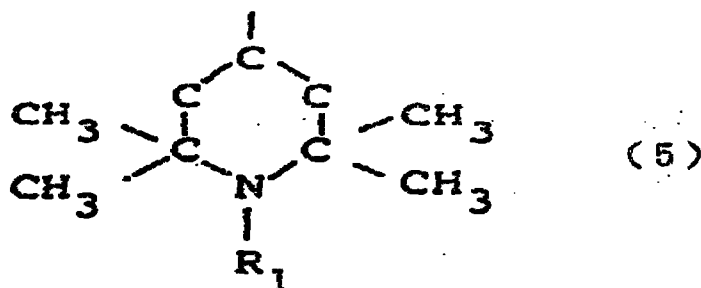
(I) when  $n=1$ ,  $\text{R}_3$  is a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group, a benzyl group, an allyl group or a group represented by the following formula (6),



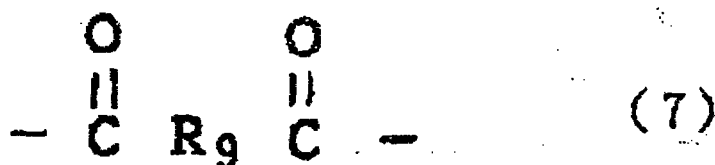
in which  $\text{Y}$  represents a direct bond,  $-\text{O}-$   $-\text{NR}_8-$ ,  $\text{R}_6$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and  $\text{R}_7$  and  $\text{R}_8$  each represents a hydroxyl group, an alkyl group, an aryl group or a group represented by the following formula (4) or (5),

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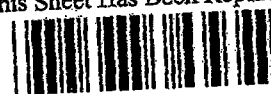
(II) when  $n=2$ ,  $R_2$  represents a group represented by the following formula (7),



in which  $R_9$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and

(IV) when  $n$  is 4,  $R_3$  represents a group represented by the following formula (9) or (10).

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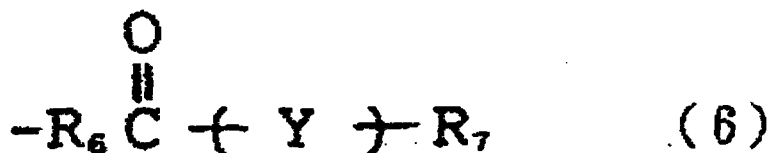


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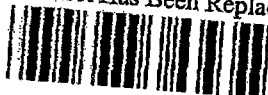


(10)

29. A thermoplastic resin composition according to claim 28, wherein in the formula (1),  $R_1$  represents a hydrogen atom or an alkyl group having 1-8 carbon atoms,  $R_2$  represents a hydrogen atom or an alkyl group having 1-18 carbon atoms, and  $n$  is an integer of 1 or 2 ; and when  $n$  is 1,  $R_3$  represents a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group or a group represented by the following formula (6),

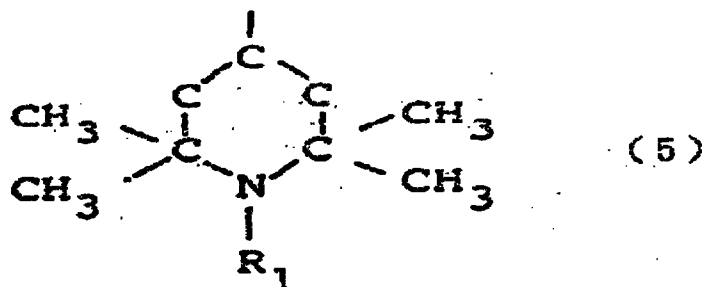


in which  $Y$  represents a direct bond,  $-O-$  or  $-NR_8-$ ,  $R_6$

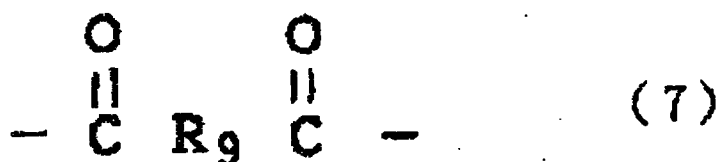




represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group having 1-10 carbon atoms or arylene group, and  $R_7$  and  $R_8$  each represent a hydrogen atom, an alkyl group having 1-18 carbon atoms, an aryl group having 6-21 carbon atoms or a group represented by the following formula (4) or (5),



; and when  $n$  is 2,  $R_9$  represents a group represented by the following formula (7),



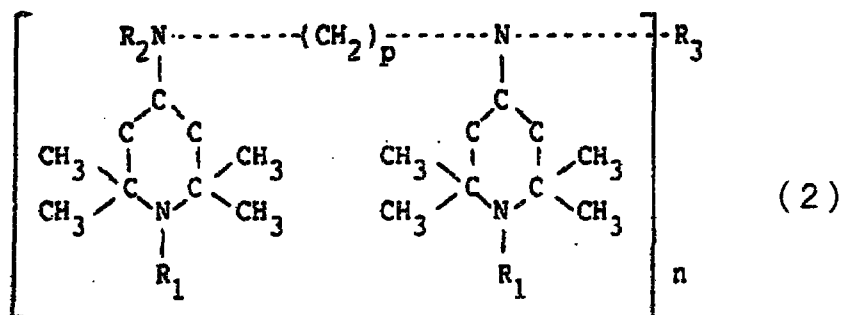
in which  $R_9$  represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group having 1-10 carbon atoms or an arylene group.

30. A thermoplastic resin composition according to claim



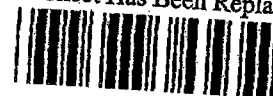


1, wherein the 4-amino-2,2,6,6-tetramethylpiperidyl compound (c) is a compound represented by the following formula (2) :

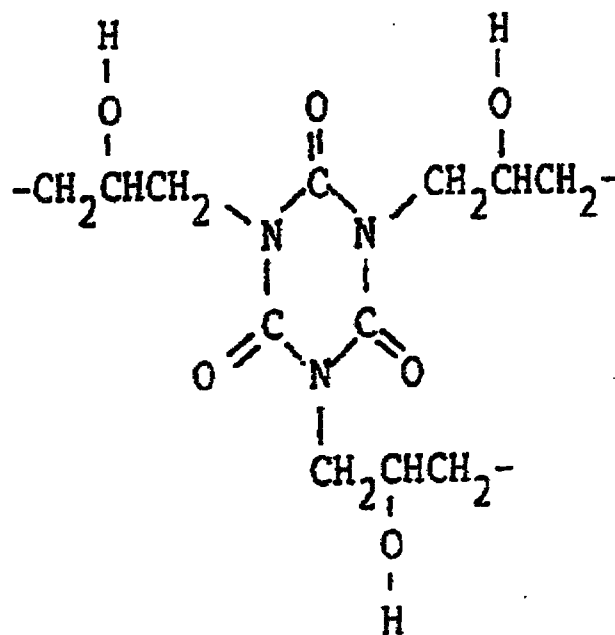


wherein  $\text{R}_1$  represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group having 1-8 carbon atoms, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group, and  $n$  is an integer of 3; and wherein  $\text{R}_2$  represents a hydrogen atom or an alkyl group having 1-8 carbon atoms and  $p$  is an integer of 2 to 8, and  $\text{R}_3$  represents a group represented by the following formula (8) ;

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2,2



(8)

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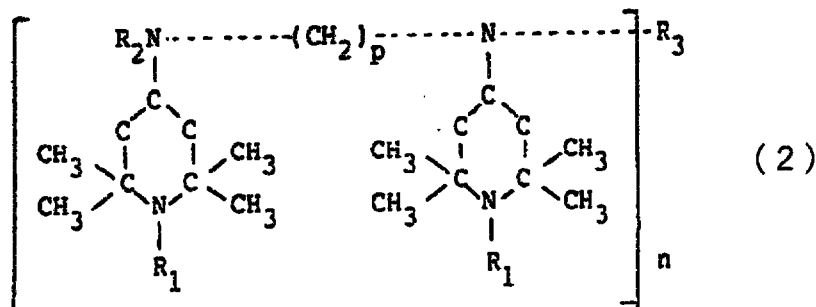
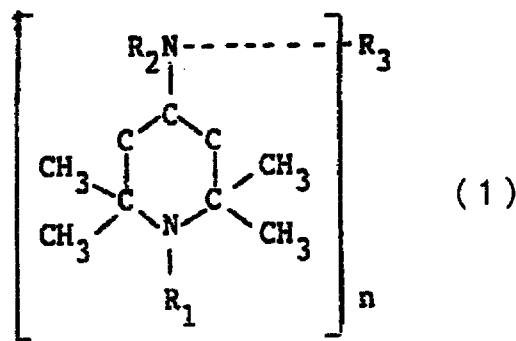
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# ABSTRACT

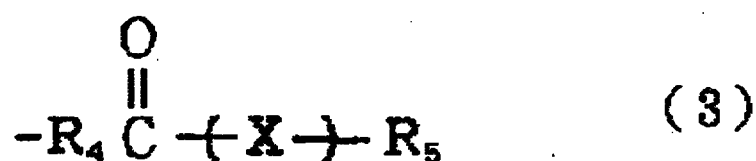
## THERMOPLASTIC RESIN COMPOSITION

The present invention provides a thermoplastic resin composition improved in process stability and in heat stability which comprises :

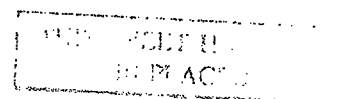
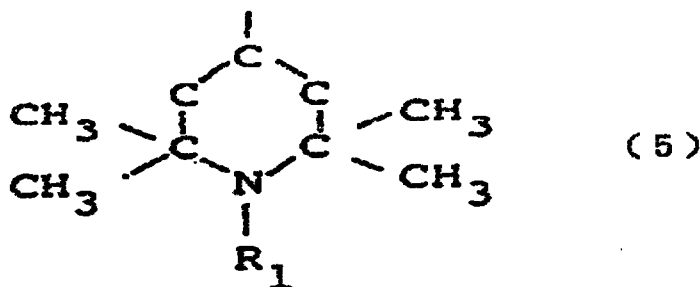
- (a) 1 to 99 % by weight of a polyphenylene ether resin,
- (b) 99 to 1 % by weight of at least one non-crystalline thermoplastic resin selected from an alkenyl aromatic resin, a non-crystalline aromatic polyester resin, a methacrylate resin, a polyether imide and an aromatic polysulfone resin, and per 100 parts by weight of the sum of (a) and (b),
- (c) 0.001 to 15 parts by weight of at least one 4-amino-2,2,6,6-tetramethylpiperidyl compound represented by the following formula (1) or (2) ;



; wherein  $R_1$  represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group.  $R_2$  represents a hydrogen atom, an alkyl group or a group represented by the following formula (3),



in which X represents a direct bond, -O- or -NH-,  $R_4$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and  $R_5$  represents a hydrogen atom, an alkyl group, an aryl group or a group represented by the following formula (4) or (5),



; and wherein  $n$  represents an integer of 1 to 4,  $p$  represents an integer of 2 to 6, and  $R$ , represents an  $n$  valent organic group.

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A THERMOPLASTIC RESIN COMPOSITION

BACKGROUND OF THE INVENTION

Field of The Invention

The present invention relates to a thermoplastic resin composition which comprises a polyphenylene ether and a non-crystalline thermoplastic resin, having improved properties in process stability and in heat stability on practically using.

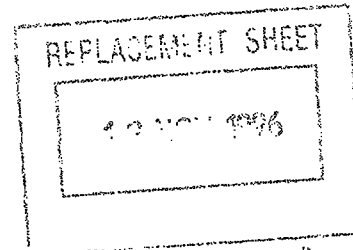
Description of Related Art

A polyphenylene ether resin is a thermoplastic resin superior in various characteristics such as mechanical properties, heat resistance and dimensional stability. However, it is apt to undergo coloration and deteriorations such as cross-linking when molded at high temperatures. Especially, in the presence of oxygen, it undergoes serious deterioration due to the heat to cause gelation, increasing in molecular weight or sometimes produce black specks. Furthermore, polyphenylene ether resin readily undergoes discoloration with light.

In order to offset these defects of the resin, it has been proposed to add various additives. For example, phosphorus additives are proposed in Japanese Patent Kokoku No. 44-29751 and phenolic additives are proposed in Japanese Patent Kokoku No. 46-24782. Furthermore, many attempts such

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as use of various stabilizers containing steric hindrance type amines or containing steric hindrance type amines and phenolic additives have been made mainly for inhibition of the discoloration with light as disclosed in Japanese Patent Kokai Nos. 60-149646 and 60-203150, U.S. Patent Nos. 4232131, 4233410, 4233412, 4234699, 4260689, 4260691 and 4299926, EP 094048A2, EP 134523A1, EP 146879A2, EP 146878A2, EP 151961A1, EP 149454A2, EP 233153A2, EP 240723A2, EP 229363A2 and EP 313118A1. Many of these attempts may be effective for improving (resistance) evaporation of stabilizers at the time of processing or actual use, and various compounds containing steric hindrance type amines alone or copolymers (oligomers) with other components have been proposed. However, none of these proposals have provided sufficient effects to inhibit deterioration at heating.

#### SUMMARY OF THE INVENTION

The present invention relates to a thermoplastic resin composition comprising a polyphenylene ether resin and a non-crystalline thermoplastic resin, which is inhibited from deterioration at the time of processing, especially with heat.

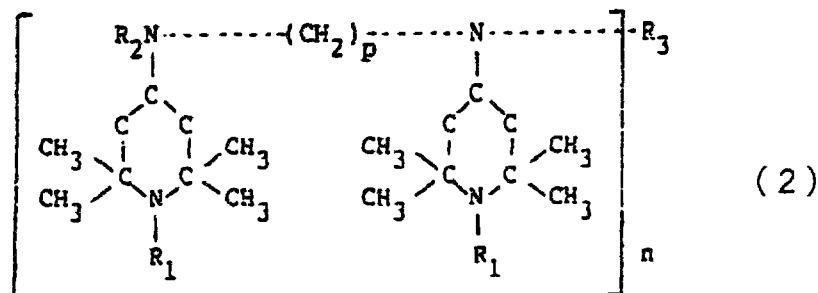
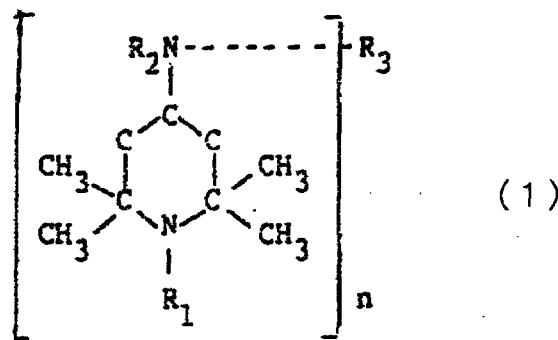
The present inventors have found that a conspicuous effect to inhibit the deterioration with heat can be obtained by adding a compound containing steric hindrance type amine having a specific structure to a thermoplastic resin composition comprising a polyphenylene ether resin and a non-



crystalline thermoplastic resin.

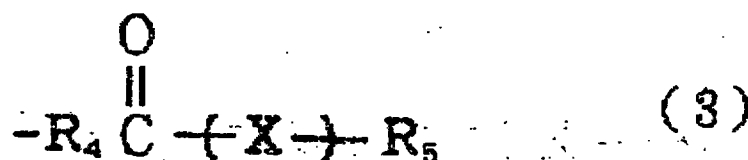
The present invention relates to a thermoplastic resin composition which comprises :

- (a) 1 to 99 % by weight of a polyphenylene ether resin,
- (b) 99 to 1 % by weight of at least one non-crystalline thermoplastic resin selected from an alkenyl aromatic resin, a non-crystalline aromatic polyester resin, a methacrylate resin, a polyetherimide resin and an aromatic polysulfone resin, and per 100 parts by weight of the sum of (a) and (b),
- (c) 0.001 to 15 parts by weight of at least one 4-amino-2,2,6,6-tetramethylpiperidyl compound represented by the following formula (1) or (2) ;

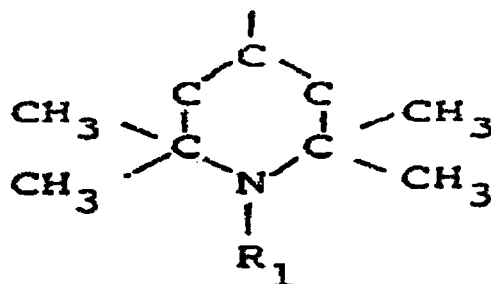
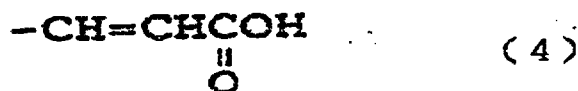


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; wherein  $R_1$  represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group, and  $R_2$  represents a hydrogen atom, an alkyl group or a group represented by the following formula (3),



in which X represents a direct bond, -O- or -NH-,  $R_4$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and  $R_5$  represents a hydrogen atom, an alkyl group, an aryl group or a group represented by the following formula (4) or (5),



(5)

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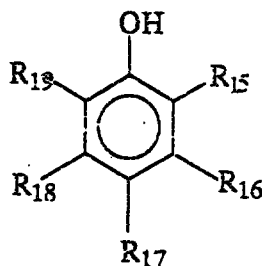
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; and wherein  $n$  represents an integer of 1 to 4,  $p$  represents an integer of 2 to 6, and  $R_j$  represents an  $n$  valent organic group.

#### DETAILED DESCRIPTION OF THE PRESENT INVENTION

The polyphenylene ether resin (a) of the present invention is a polymer obtained by oxidation polymerization of at least one phenol compound represented by the following formula with oxygen or a gas containing oxygen using an oxidation coupling catalyst ;



wherein  $R_{15}$ ,  $R_{16}$ ,  $R_{17}$ ,  $R_{18}$  and  $R_{19}$  each represents a hydrogen atom, a halogen atom or a substituted or unsubstituted hydrocarbon group and at least one of them is a hydrogen atom.

Examples of  $R_{15}$ ,  $R_{16}$ ,  $R_{17}$ ,  $R_{18}$  and  $R_{19}$  are hydrogen, chlorine, bromine, fluorine, iodine, methyl, ethyl, *n*- or *iso*-propyl, *pri*-, *sec*- or *t*-butyl, chloroethyl, hydroxyethyl, phenylethyl, benzyl, hydroxymethyl, carboxyethyl, methoxycarbonyl, cyanoethyl, phenyl, chlorophenyl, methylphenyl, dimethylphenyl, ethylphenyl and allyl.

Examples of the compounds represented by the above

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formula are phenol, o-, m- or p-cresol, 2,6-, 2,5-, 2,4- or 3,5-dimethylphenol, 2-methyl-6-phenylphenol, 2,6-diphenylphenol, 2,6-diethylphenol, 2-methyl-6-ethylphenol, 2,3,5-, 2,3,6- or 2,4,6-trimethylphenol, 3-methyl-6-t-butylphenol, thymol and 2-methyl-6-allylphenol.

The phenol compounds of the above formula can also be copolymerized with other phenol compounds, for example, polyhydric aromatic compounds such as bisphenol A, tetrabromobisphenol A, resorcin, hydroquinone and novolak resins.

As preferable examples of the polyphenylene ether resin (a), mention may be made of homopolymers of 2,6-dimethylphenol, homopolymers of 2,6-diphenylphenol, copolymers of a larger amount of 2,6-dimethylphenol and a smaller amount of 3-methyl-6-t-butylphenol or 2,3,6-trimethylphenol.

Any oxidation coupling catalysts may be used for oxidizing polymerization of the phenol compound as far as they have polymerization ability.

The non-crystalline thermoplastic resin (b) of the present invention is at least one selected from an alkenyl aromatic resin, a non-crystalline aromatic polyester resin, a methacrylate resin, a polyether imide resin and an aromatic polysulfone resin.

The alkenyl aromatic resin includes a homopolymer and a copolymer containing an aromatic vinyl compound represented by

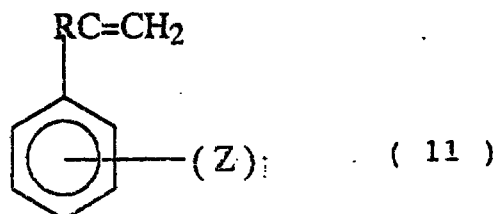
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the following formula (11) ;



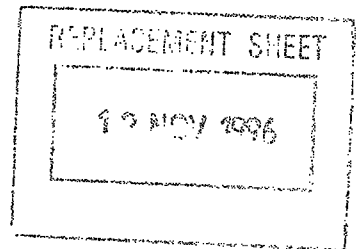
wherein R represents a hydrogen atom, an alkyl group having 1 to 6 carbon atoms or a halogen atom, Z represents a hydrogen atom, a vinyl group, a halogen atom, an amino group or an alkyl group having 1-6 carbon atoms, and  $l$  is 0 or an integer of 1 to 5.

The example of the alkenyl aromatic resin includes a polymer containing one kind or plural kinds of polymerization unit selected from styrene,  $\alpha$ -methylstyrene and  $p$ -methylstyrene, polystyrene (PS resin), polychlorostyrene, poly- $\alpha$ -methylstyrene, styrene-acrylonitrile copolymer (AS resin), styrene- $\alpha$ -methylstyrene copolymer, styrene-4-methylstyrene copolymer, styrene-maleic anhydride copolymer and styrene-methylmethacrylate copolymer (MS resin).

Besides, it includes a copolymer of a compound containing 2 or more vinyl groups such as divinyl benzene and ethyleneglycol dimethacrylate, with the above-mentioned aromatic vinyl compound.

Further, the alkenyl aromatic resin includes a rubber-modified polymer containing a dispersed rubber phase in the matrix of the alkenyl aromatic resin phase. The rubber used





here includes a homopolymer or copolymer containing a diene component such as polybutadiene rubber (BR), polyisoprene rubber (IR), polychlorobutadiene rubber, a butadiene-styrene copolymer rubber, an isoprene-styrene copolymer rubber, a butadiene-acrylonitrile copolymer rubber (NBR), an isobutylene-butadiene or -isoprene copolymer rubber, an ethylene-propylene copolymer rubber (EPM) and an ethylene-propylene-non-conjugated diene copolymer rubber (EPDM).

Various methods are known as a method for dispersing the rubber into the matrix of the alkenyl aromatic resin phase, and for example, a method by blending mechanically, a method by solution-blending, and a method by dissolving a rubber into an alkenyl aromatic monomer and graft-copolymerizing said monomer, are mentioned, and any one of them may be used.

Besides, a polymerization method may be any one of polymerization methods such as an emulsion polymerization process, a bulk polymerization process (Japanese Examined Patent Kokoku No.42-662 and USP 3435096), a solution polymerization process (USP 3538190 and USP 3538191) and a suspension polymerization process (Japanese Examined Patents Kokoku No.49-10831, No.57-40166 and No.62-10565). The ratio of the rubber to the alkenyl aromatic monomer in the rubber-modified alkenyl aromatic resin is preferably 0.5 to 80 %, and more preferably 0.5 to 50% by weight of the rubber as total amount of 100.

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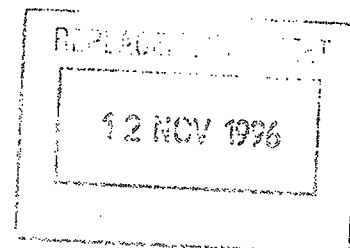
As the examples of the rubber-modified alkenyl aromatic resin, a high impact polystyrene (HIPS) such as polystyrene modified with polybutadiene rubber, polystyrene modified with styrene-butadiene copolymer rubber and polystyrene modified with ethylene-propylene-non-conjugated diene copolymer rubber, an acrylonitrile-butadiene-styrene copolymer resin known as ABS resin and an acrylonitrile-(ethylene-propylene-non-conjugated diene copolymer rubber)-styrene resin known as AES resin are mentioned.

The non-crystalline aromatic polyester resin is polycarbonate resin and polyarylate resin.

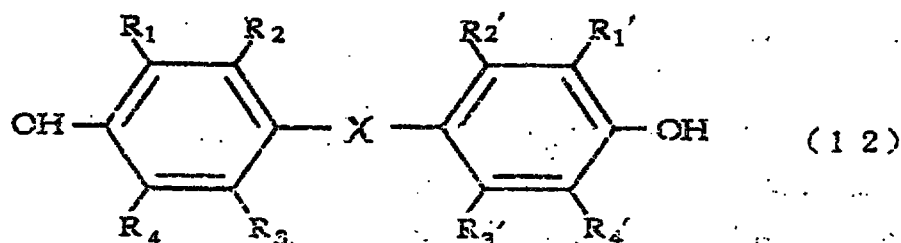
The polycarbonate resin of the present invention is 4,4-dioxydialkanic polycarbonate including 4,4-dihydroxydiphenyl-2,2-propane (bisphenol A), and among them, 4,4-dihydroxydiphenyl-2,2-propane of polycarbonate having number average molecular weight of 15,000 to 80,000 is especially preferred. These polycarbonates are prepared with an arbitrary method. For example, a method of preparing by using 4,4-dihydroxydiphenyl-2,2-propane and phosgene in the presence of sodium hydroxide aqueous solution and a solvent, and a method of preparing by ester interchanging of 4,4-dihydroxydiphenyl-2,2-propane with carbonic diester in the presence of a catalyst are mentioned.

The polyarylate resin of the present invention is a polyester resin obtained from bisphenols and terephthalic acid





and/or isophthalic acid. As the bisphenols, those having a structure represented by the following formula (12) are used ;



In the formula (12), X is a group selected from -O-, -S-, -CO- and a hydrocarbon group having 1 to 10 carbon atoms, and R<sub>1</sub> to R<sub>4</sub> and R'<sub>1</sub> to R'<sub>4</sub> represent a group selected from a hydrogen atom, a halogen atom and a hydrocarbon group. When X represents a hydrocarbon group having 1 to 10 carbon atoms, an alkylene group, a branched alkylene group and a halogen-substituted alkyl group are included. On the other hand, each terephthalic acid and isophthalic acid can be used alone or in combination. It is obtained by polymerizing the above-mentioned bisphenols with terephthalic acid and/or isophthalic acid, and as a polymerization method, either of usual interfacial polymerization process or solution polymerization process may be used. Besides, these polyarylate resins having a molecular weight of 5,000 to 70,000 are preferred.

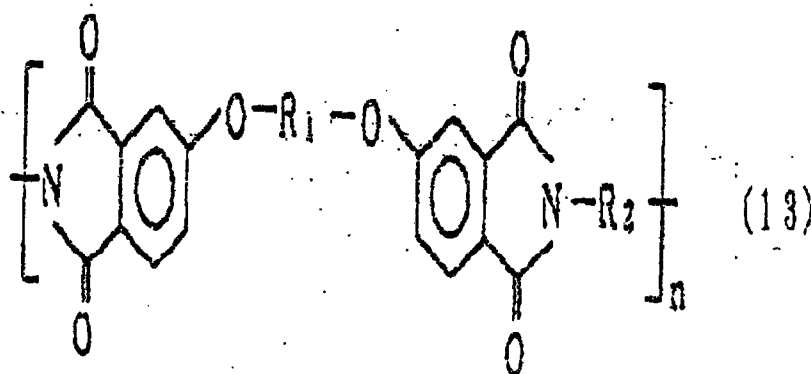
The methacrylate resin of the present invention is a homopolymer of methyl methacrylate or a copolymer of methyl methacrylate with acrylate, methacrylate, acrylic acid, methacrylic acid, styrene, acrylonitrile, glutaric anhydride, glutarimide or a diene rubber.



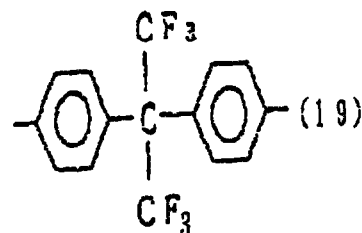
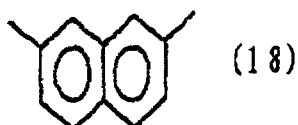
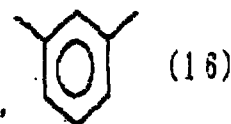
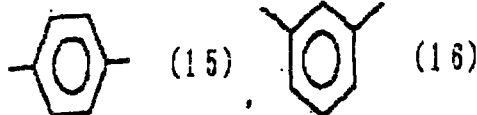
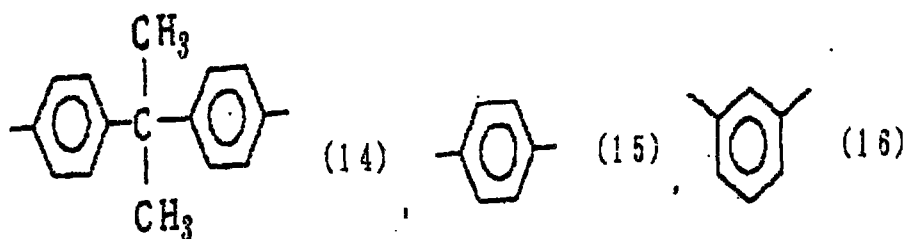
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Acrylate includes methyl acrylate, ethyl acrylate, and butyl acrylate, and methacrylate includes methyl methacrylate, ethyl methacrylate, butyl methacrylate and cyclohexyl methacrylate.

The polyetherimide resin of the present invention is a resin represented by the following formula (13),

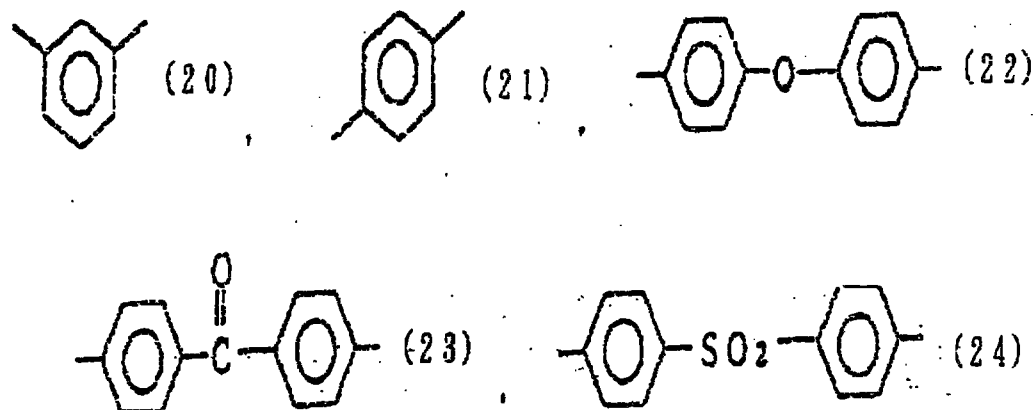


In the formula (13), n represents an integer of 1 to 1000, and R<sub>1</sub> represents a group represented by the following formulas (14) to (19).

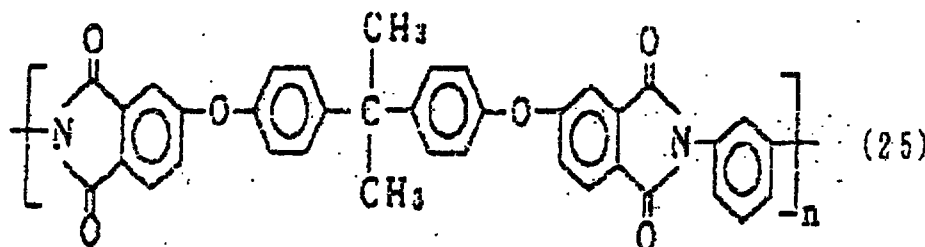


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R<sub>2</sub> represents a group represented by the following formulas (20) to (24).



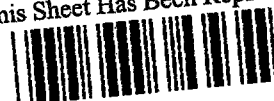
Preferable example from the view points of property and production cost includes the structure represented by the following formula (25),



wherein n represents an integer of 1 to 1000.

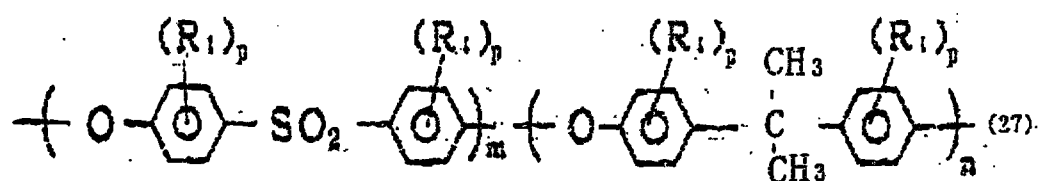
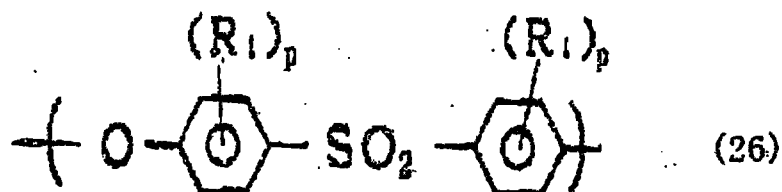
The aromatic polysulfone resin used in the present invention is defined as the polyarylene compound wherein arylene units are located in disorder or order with ether bond and sulfone bond. Those having a repeating unit represented by the following formulas (26) to (28) are mentioned as typical examples, but it is not limited to these.

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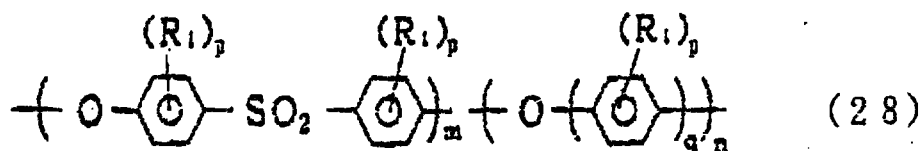


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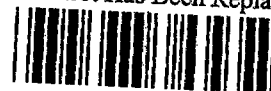


The compound of the formula (26) includes a homopolymer and the compound of the formula (27) includes a random copolymer, wherein  $\text{R}_1$  represents an alkyl group having 1 to 6 carbon atoms, an alkenyl group having 3 to 10 carbon atoms, a phenyl group or a halogen atom,  $p$  is an integer of 0 to 4, besides,  $m$  and  $n$  represent mean repeating unit numbers and are integers of 1 to 1000, each  $\text{R}_1$  on the same or different nucleus may be different mutually, and each  $p$  may be different mutually.



The compound of the formula (28) includes a random copolymer, wherein  $\text{R}_1$  represents an alkyl group having 1 to 6 carbon atoms, an alkenyl group having 3 to 10 carbon atoms, a

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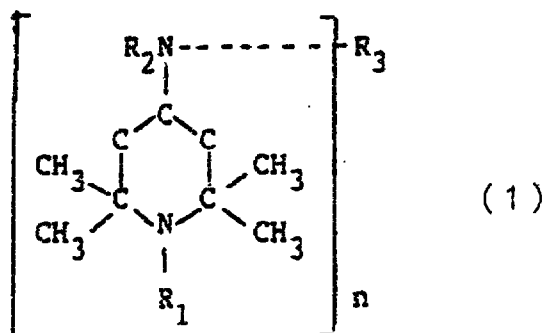
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phenyl group or halogen atom,  $p$  is an integer of 0 to 4, besides,  $q$ ,  $m$  and  $n$  represent mean repeating unit numbers,  $q$  is an integer of 1 to 3,  $m$  and  $n$  are an integer of 0 to 100, each  $R_1$  on the same or different nucleus may be different mutually, and each  $p$  may be different mutually.

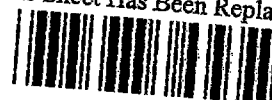
As the aromatic polysulfone resin used in the present invention,  $m/(m+n)$  in the structure unit of the formula (27) is preferably 0.8 or more. Besides, it is preferred that  $m/(m+n)$  in the structure unit of the formula (28) is 0.3 or more and  $q$  is 1.

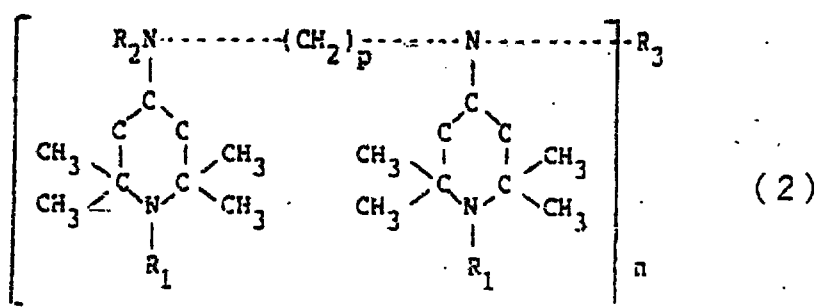
Among these, the one having the repeating unit of the formula (26) or (27) is preferred.

The 4-amino-2,2,6,6-tetramethylpiperidyl compound (c) having a steric hindrance type amino group as a basic skeleton is represented by the following formula (1) or (2):

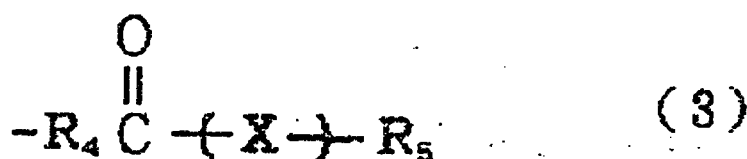


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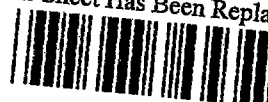


; wherein  $R_1$  represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group.  $R_2$  represents a hydrogen atom, an alkyl group or a group represented by the following formula (3),



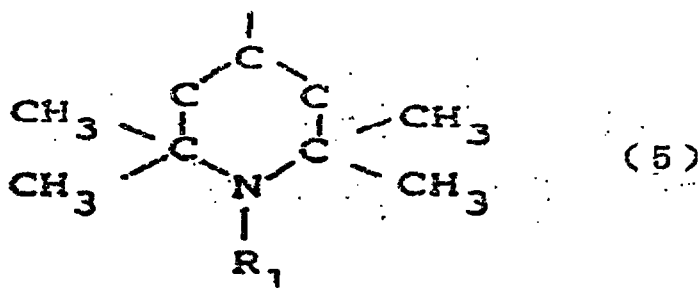
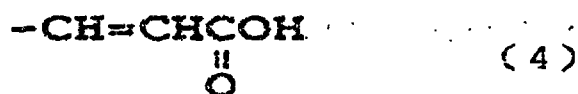
in which X represents a direct bond, -O- or -NH-,  $R_4$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and  $R_5$  represents a hydrogen atom, an alkyl group, an aryl group or a group represented by the following formula (4) or (5),

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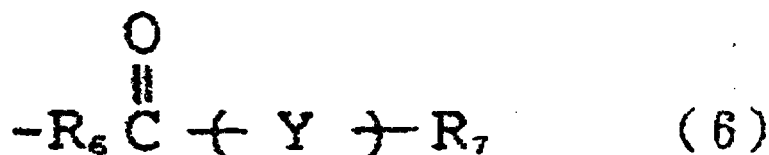
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; and wherein n represents an integer of 1 to 4, p represents an integer of 2 to 6, and R<sub>1</sub> represents an n valent organic group.

Preferable examples of the 4-amino-2,2,6,6-tetramethylpiperidyl compound represented by the formula (1) or (2) are [I] to [VII] shown below.

[I] When n is 1 and R<sub>1</sub> is a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group, a benzyl group, an aryl group or a group represented by the following formula (6);



, wherein Y represents a direct bond, -O- or -NR<sub>8</sub>-, R<sub>6</sub> represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and R<sub>7</sub> and R<sub>8</sub>

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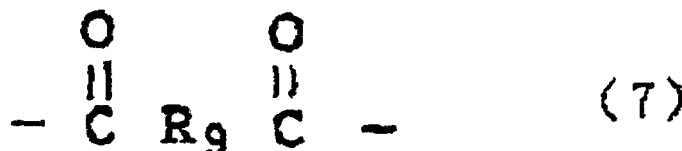


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each represents a hydrogen atom, a hydroxyl group, an alkyl group or an aryl group.

More preferably, when n is 1, R<sub>1</sub> and R<sub>2</sub> each represents a hydrogen atom or an alkyl group having 1-18 carbon atoms and R<sub>3</sub> represents a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group or the group represented by the formula  $\begin{smallmatrix} 6 \\ \text{---} \end{smallmatrix}$  in which  $\begin{smallmatrix} Y \\ \text{---} \end{smallmatrix}$  represents a direct bond, -O- or -NR<sub>8</sub>, R<sub>4</sub> represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group or arylene group, and R<sub>5</sub> and R<sub>6</sub> each represent a hydrogen atom, an alkyl group having 1-18 carbon atoms, an aryl group having 6-21 carbon atoms or a group represented by the formula (4) or (5).

[II] When n is 2, R<sub>1</sub> represents an alkylene group or a group represented by the following formula (7);



wherein R<sub>1</sub> represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group.

More preferably, when n is 2, R<sub>1</sub> and R<sub>2</sub> each represents a hydrogen atom or an alkyl group having 1-18 carbon atoms and R<sub>3</sub> represents an alkylene group or a group represented by the formula (7) in which R<sub>1</sub> represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group or an arylene group.

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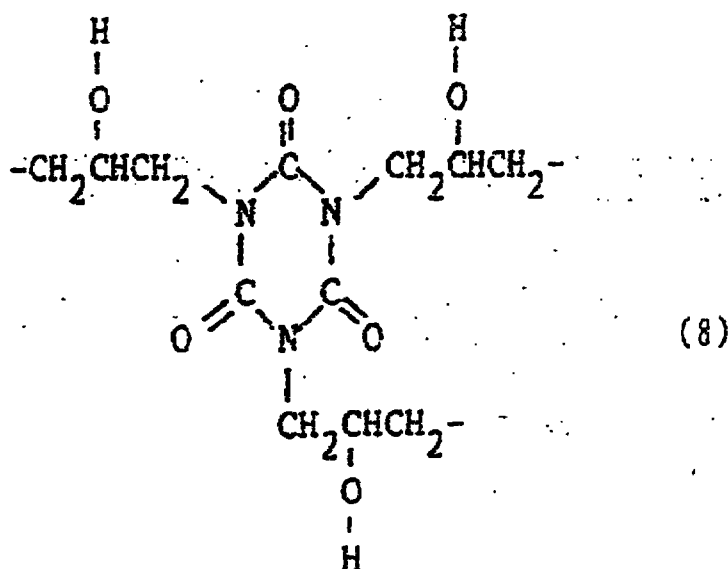


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The compound in the formula (1), when  $n$  is 2,  $R_2$  represents a group represented by the formula (3) in which  $X$  represents a direct bond,  $-O-$  or  $-NH-$ ,  $R_1$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group,  $R_3$  represents a hydrogen atom, an alkyl group, an aryl group or a group represented by the formula (4) or (5), and  $R_4$  represents an alkylene group, is also more preferable.

[III] When  $n$  is 3,  $R_2$  is a group represented by the following formula (8);



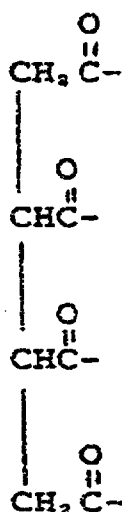
[IV] When  $n$  is 4,  $R_2$  is a group represented by the following formula (9) or (10);

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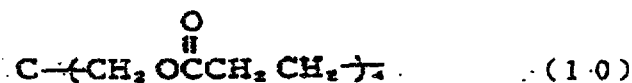


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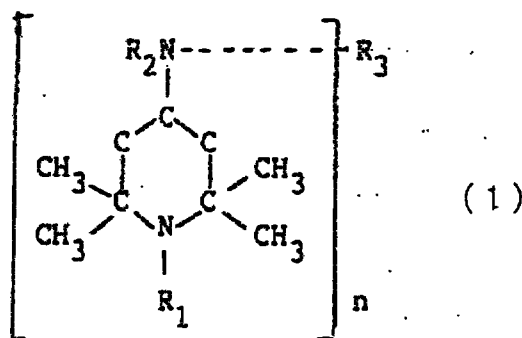


(9)



(10)

[V] Further, the compound represented by the following formula (1) is also preferable.



(1)

In the formula (1),  $\text{R}_1$  represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group having 1-8 carbon atoms, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group, and  $n$  is an integer of 1, 2 or 4 ; and when  $n$  is 1,  $\text{R}_2$  represents a hydrogen atom,

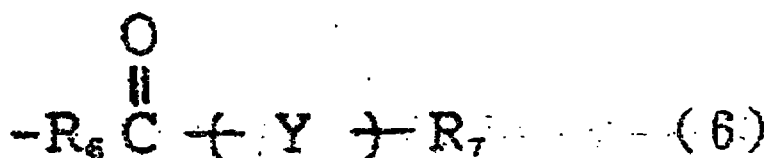


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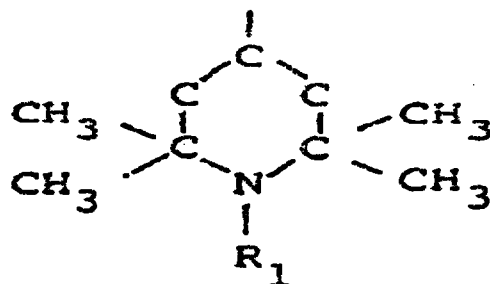
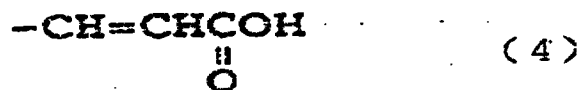
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an alkyl group having 1-8 carbon atoms or  $-\overset{\overset{\text{O}}{\parallel}}{\text{C}}-(\text{P})-(\text{Q})-\overset{\overset{\text{O}}{\parallel}}{\text{C}}-\text{CH}$  in

which P and Q represent one selected from the group consisting of an alkylene group, an alkylidene group, a vinyl group and a vinylidene group, and  $\text{R}_3$  represents a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group, a benzyl group, an aryl group or a group represented by the following formula (5);

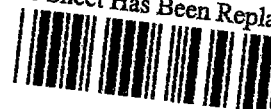


in which Y represents a direct bond,  $-\text{O}-$   $-\text{NR}_8-$ ,  $\text{R}_6$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and  $\text{R}_7$  and  $\text{R}_8$  each represents a hydroxyl group, an alkyl group, an aryl group or a group represented by the following formula (4) or (5),



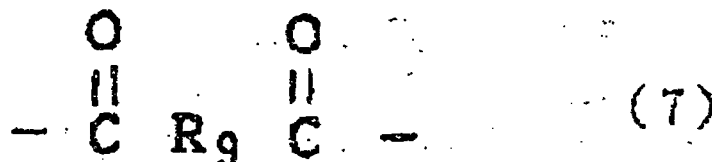
(5)

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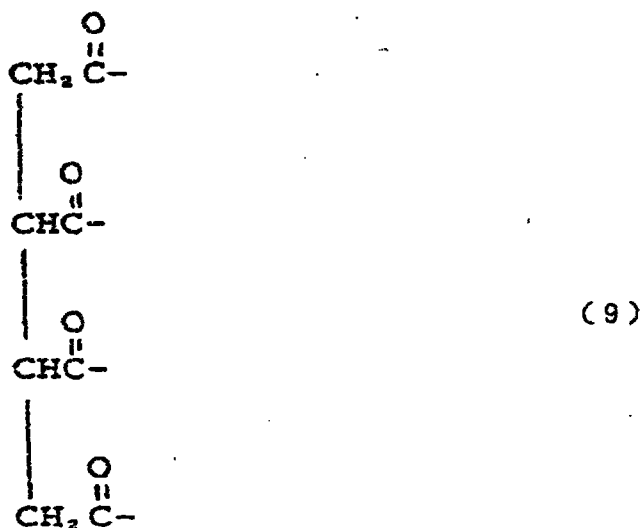


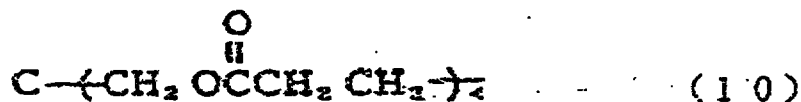
; when n is 2, R<sub>2</sub> represents a hydrogen atom, an alkyl group having 1-8 carbon atoms or  $\begin{array}{c} \text{O} \\ || \\ -\text{C}-(\text{P})-(\text{Q})-\text{C}-\text{OH} \\ || \\ \text{O} \end{array}$  in which P and Q

represent one selected from the group consisting of an alkylene group an alkylidene group, a vinyl group and a vinylylidene group, and R<sub>3</sub> represents an alkylene group or a group represented by the following formula (7),



in which R<sub>9</sub> represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group; and when n is 4, R<sub>2</sub> represents a hydrogen atom or an alkyl group having 1-8 carbon atoms, and R<sub>3</sub> represents a group represented by the following formula (9) or (10),



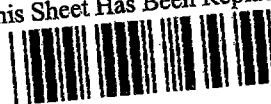


More preferably, in the formula (1),  $R_1$  and  $R_2$  each represents a hydrogen atom or an alkyl group having 1-8 carbon atoms, and (i) when  $n$  is 1,  $R_3$  is a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group or a group represented by the formula (6) in which  $Y$  represents a direct bond,  $-O-$  or  $-NR_4-$ ,  $R_4$  represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group or arylene group, and  $R_5$  and  $R_6$  each represent a hydroxyl group, an alkyl group having 1-18 carbon atoms, an aryl group having 6-21 carbon atoms or a group represented by the formula (4) or (5),

and (ii) when  $n$  is 2,  $R_3$  represents an alkylene group or a group represented by the formula (7) in which  $R_7$  represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group or an arylene group.

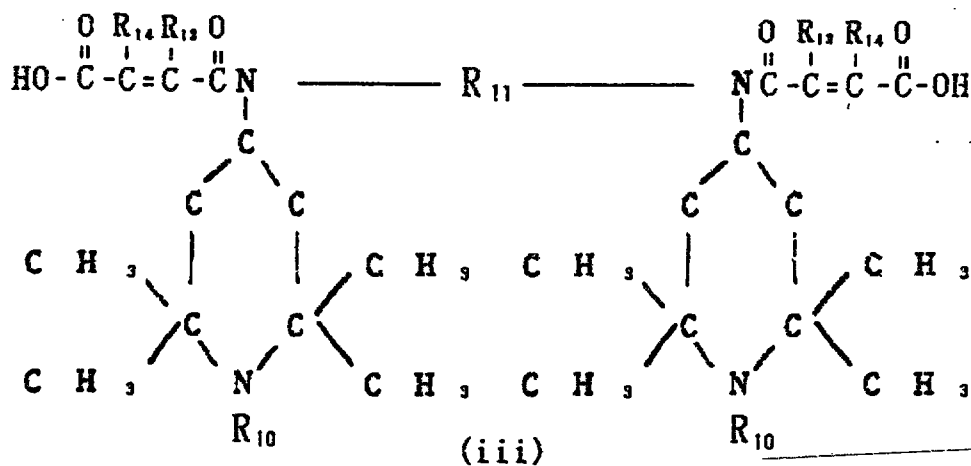
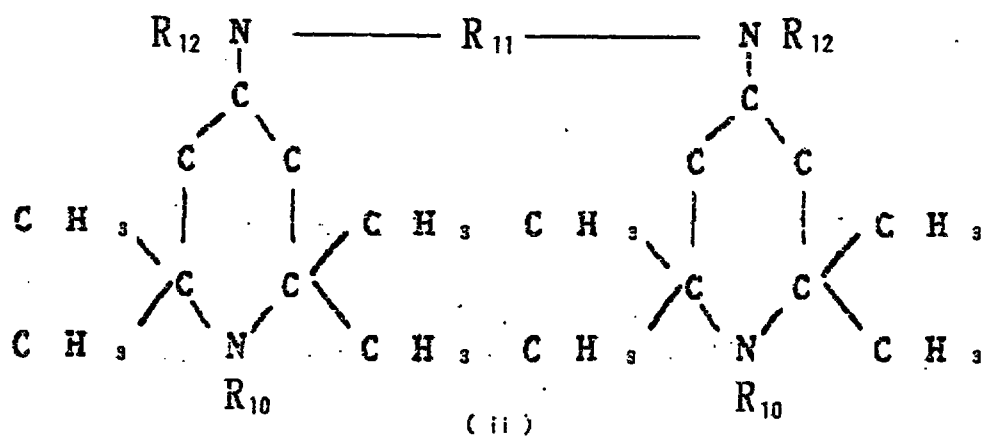
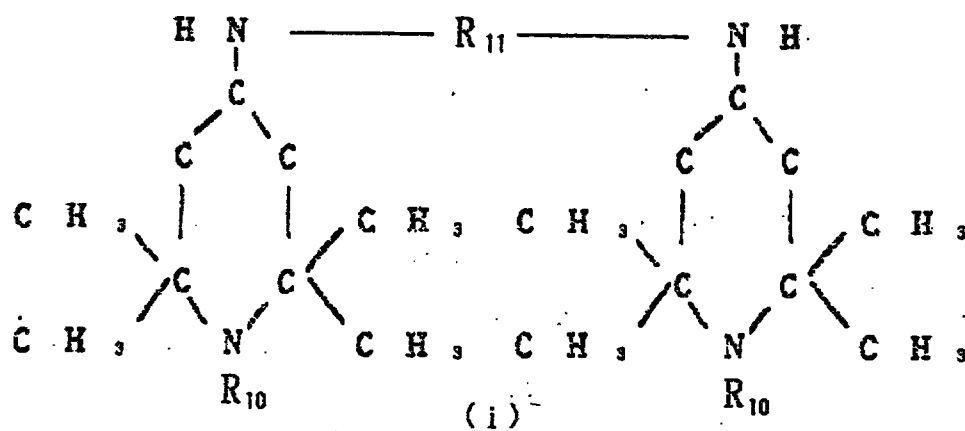
Further, at least one compound represented by the following formulas (i)-(iv) is also more preferable ;

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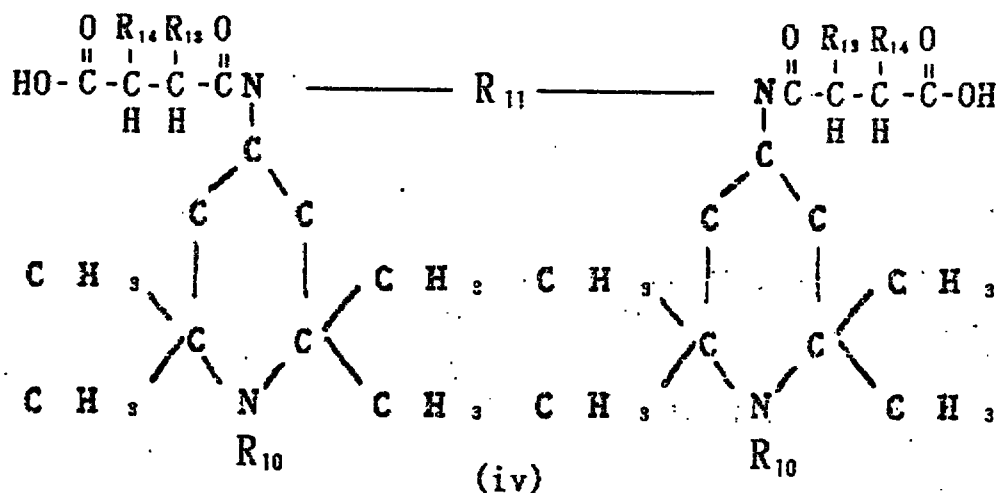
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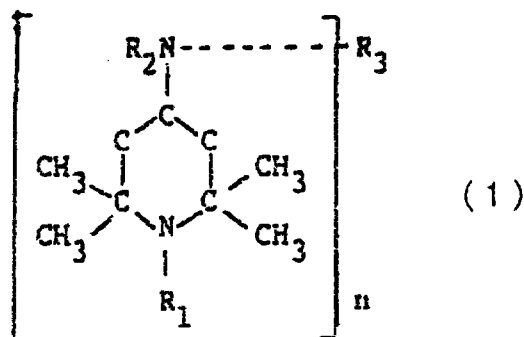
wherein  $R_{10}$  represents a hydrogen atom, an alkyl group or an alkyloxy group,  $R_{11}$  represents an alkylene group,  $R_{12}$  represents an alkyl group, and  $R_{13}$  and  $R_{14}$  each represents a hydrogen atom or a methyl group.

In the formula (i), preferably,  $R_{10}$  represents a hydrogen atom, a hydroxyl group, an alkyl group having 1-10 carbon atoms or an alkyloxy group having 1-10 carbon atoms, and  $R_{11}$  represents an alkylene group having 2-8 carbon atoms, and more preferably,  $R_{10}$  represents a hydrogen atom or a methyl group and  $R_{11}$  represents a hexamethylene group.

In the formula (ii), preferably,  $R_{10}$  represents a hydrogen atom, a hydroxyl group, an alkyl group having 1-10 carbon atoms or an alkyloxy group having 1-10 carbon atoms,  $R_{11}$  represents an alkylene group having 2-8 carbon atoms, and  $R_{12}$  represents an alkyl group having 1-18 carbon atoms, and more preferably,  $R_{10}$  represents a hydrogen atom or a methyl group.

In the formula (iii), preferably,  $R_{10}$  represents a hydrogen atom, a hydroxyl group, an alkyl group having 1-10 carbon atoms or an alkyloxy group having 1-10 carbon atoms, and  $R_{11}$  represents an alkylene group having 2-8 carbon atoms, and more preferably,  $R_{10}$  represents a hydrogen atom or a methyl group. In the formula (iv), preferably,  $R_{10}$  represents a hydrogen atom, a hydroxyl group, an alkyl group having 1-10 carbon atoms or an alkyloxy group having 1-10 carbon atoms, and  $R_{11}$  represents an alkylene group having 2-8 carbon atoms, and more preferably,  $R_{10}$  represents a hydrogen atom or a methyl group.

[VI] The compound represented by the following formula (1) ;

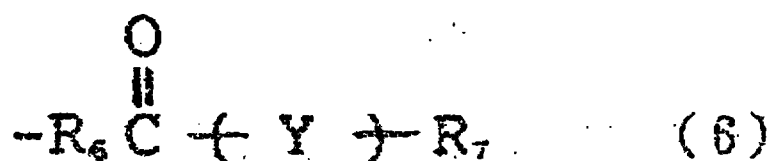


wherein  $R_1$  represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group having 1-8 carbon atoms, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group,  $R_2$  represents a hydrogen atom or alkyl group, ; and

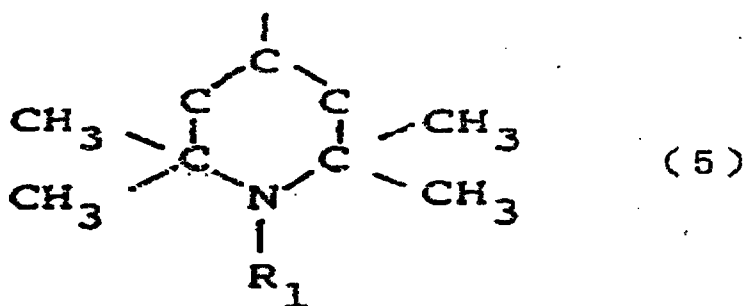
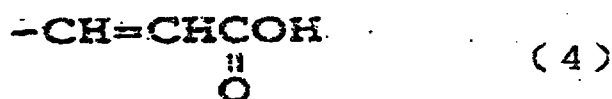
(i) when  $n$  is 1,  $R_3$  represents a hydrogen atom, an alkyl group,



a cycloalkyl group, an alkenyl group, a benzyl group, an allyl group or a group represented by the following formula (6),



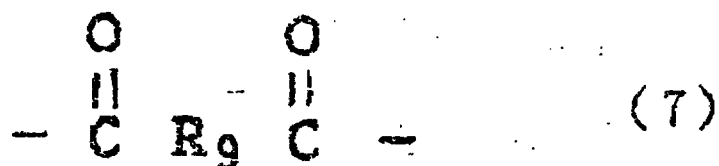
in which Y represents a direct bond, -O- -NR<sub>8</sub>-, R<sub>6</sub> represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and R<sub>7</sub> and R<sub>8</sub> each represents a hydroxyl group, an alkyl group, an aryl group or a group represented by the following formula (4) or (5),



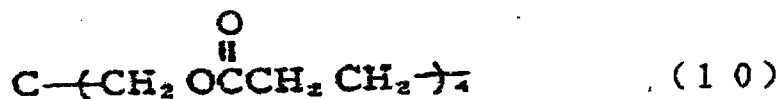
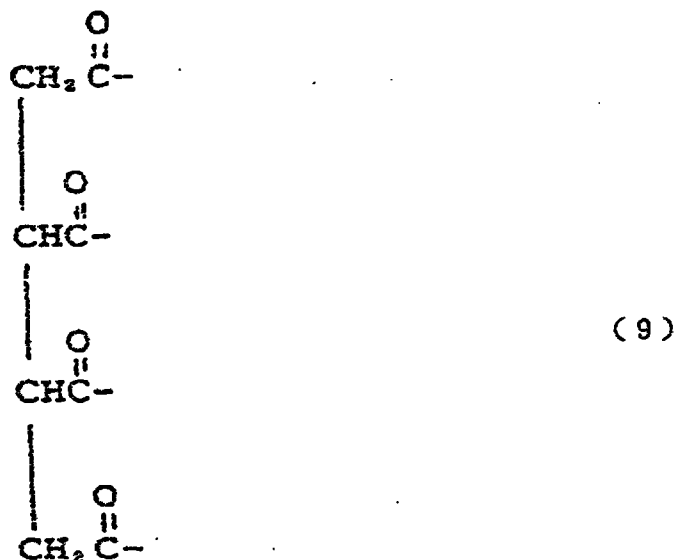
(ii) when n is 2, R<sub>3</sub> represents a group represented by the following formula (7),

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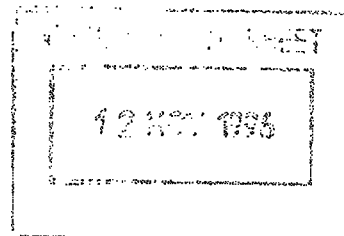




in which R<sub>9</sub> represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and (iv)-when n is 4, R<sub>9</sub> represents a group represented by the following formula (9) or (10).

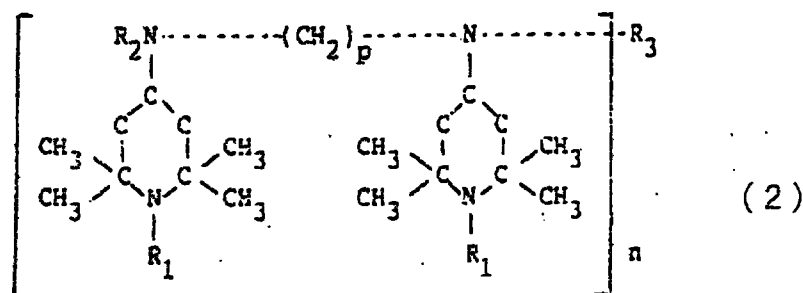


More preferably, in the formula (1), R<sub>1</sub> represents a hydrogen atom or an alkyl group having 1-8 carbon atoms, R<sub>2</sub> represents a hydrogen atom or an alkyl group having 1-18 carbon atoms ; and when n is 1, R<sub>3</sub> represents a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group or a group represented by



the formula (6) in which Y represents a direct bond, -O- or -NR<sub>8</sub>-, R<sub>5</sub> represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group having 1-10 carbon atoms or arylene group, and R<sub>6</sub> and R<sub>7</sub> each represent a hydrogen atom, an alkyl group having 1-18 carbon atoms, an aryl group having 6-21 carbon atoms or a group represented by the formula (4) or (5); and when n is 2, R<sub>8</sub> represents a group represented by the formula (7) in which R<sub>9</sub> represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group having 1-10 carbon atoms or an arylene group.

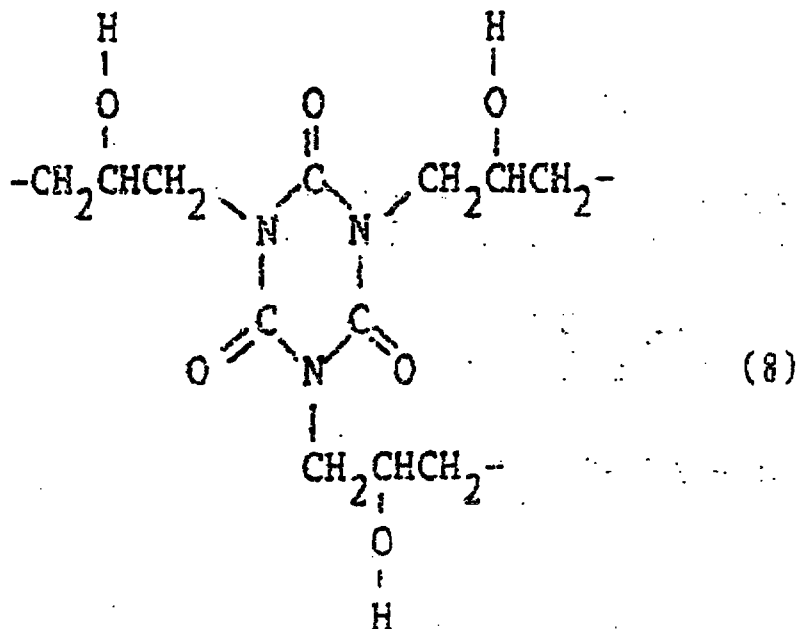
[VII] The compound represented by the following formula (2);



wherein R<sub>1</sub> represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group having 1-8 carbon atoms, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group, R<sub>2</sub> represents a hydrogen atom or an alkyl group having 1-8 carbon atoms, n is an integer of 3, p is an integer of 2 to 8, and R<sub>3</sub> represents a group



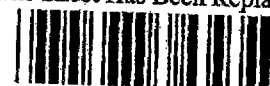
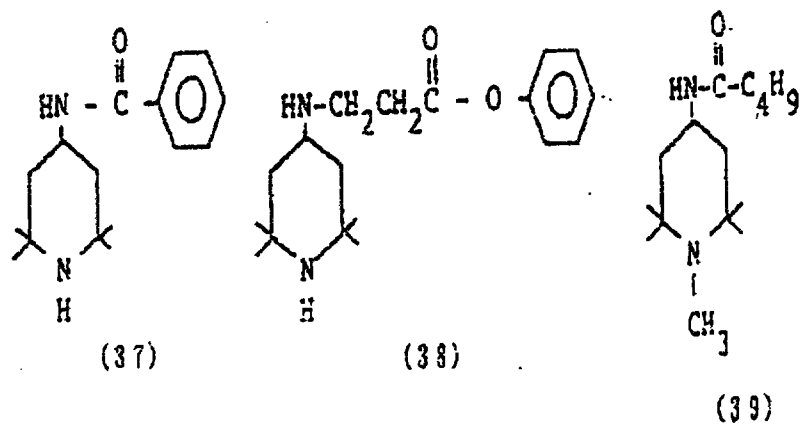
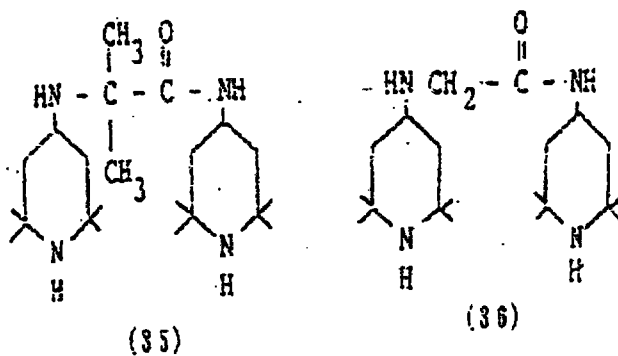
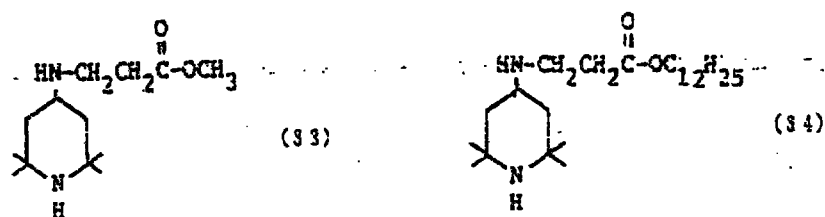
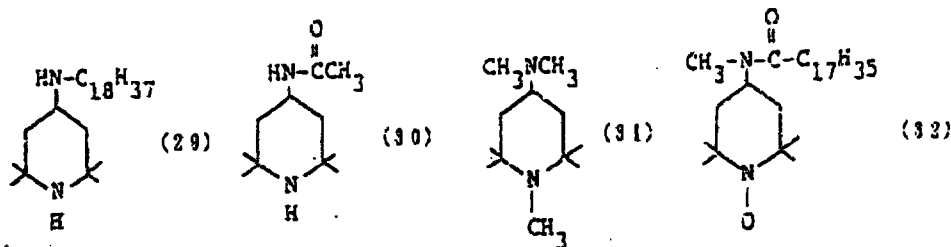
represented by the following formula (8) ;



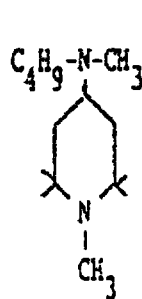
Examples of [n=1] of [I], [V] and [VI] of the compound (c) of the present invention include 4-Amino-2,2,6,6-tetramethyl-piperidine, 4-methyl-amino-2,2,6,6-tetramethyl-4-piperidine, 4-ethylamino-2,2,6,6-tetra-methylpiperidine, 4-propylamino-2,2,6,6-tetramethylpiperidine, 4-butylamino-2,2,6,6-tetra-methylpiperidine, 4-hexylamino-2,2,6,6-tetramethylpiperidine, 4-octylamino-2,2,6,6-tetra-methylpiperidine, 4-dodecanyl-amino-2,2,6,6-tetramethyl-piperidine, 4-diethylamino-2,2,6,6-tetramethylpiperidine, N-(2,2,6,6-tetramethyl-4-piperidyl) piperidine, N-(2,2,6,6-tetramethyl-4-piperidyl)acetamide and N-(2,2,6,6-tetramethyl-4-piperidyl)stearoylamide. Besides, mention may be made of the compounds represented by the following formulas (29) to (47).



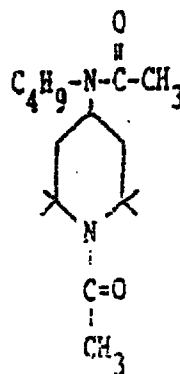
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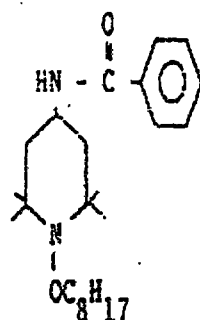
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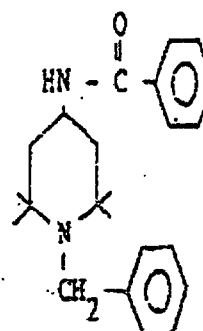
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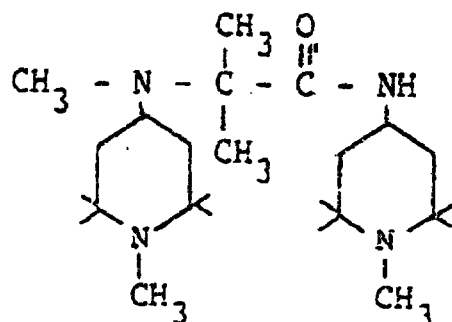
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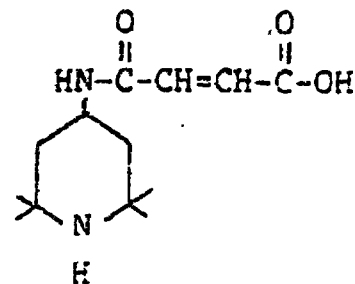
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(43)



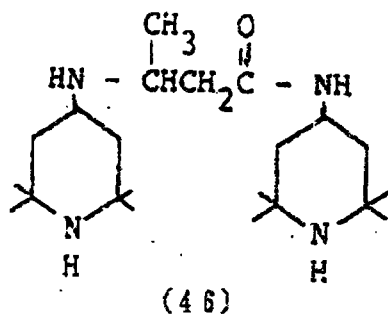
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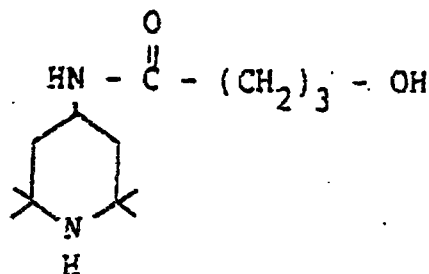
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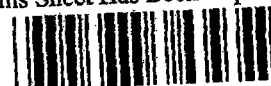
(47)

Examples of  $\{n=2\}$  of [II], [V] and [VI] of the compound (c) of the present invention include compounds below.

In the formula (1) or (2),  $R_1$  includes, for example, a hydrogen atom, a hydroxyl group, an alkyl group of 1-10 carbon atoms such as methyl, ethyl or propyl, or an alkyloxy group of 1-10 carbon atoms and further includes cyclic ones such as cyclohexylmethyl and cyclohexylethyl in addition to straight or branched ones when the carbon number is 3 or more.  $R_1$  is preferably a hydrogen atom or a methyl group.

$R_2$  is preferably an alkyl group having 1-18 carbon atoms or an alkyloxy group having 1-18 carbon atoms, such as methyl, ethyl, propyl, isopropyl, n-butyl, isobutyl, t-butyl, sec-butyl, n-pentyl, n-hexyl, 2-ethylhexyl, n-octyl, 1,1,3,3-tetramethylbutyl, n-decyl, n-dodecyl, n-octadodecyl, cyclohexyl, 3,3,5-trimethyl-cyclohexyl, cyclooctyl and cyclodecyl.

$R_3$  includes an alkylene group having 2-8 carbon atoms and alkylene groups represented by  $-\text{C}_n\text{H}_{2n}-$  such as ethylene, 1,2- or 1,3-propylene, 1,2-, 1,3- or 1,4-butylene, pentamethylene, 2,2-dimethyl-1,3-propylene, hexamethylene and octamethylene,

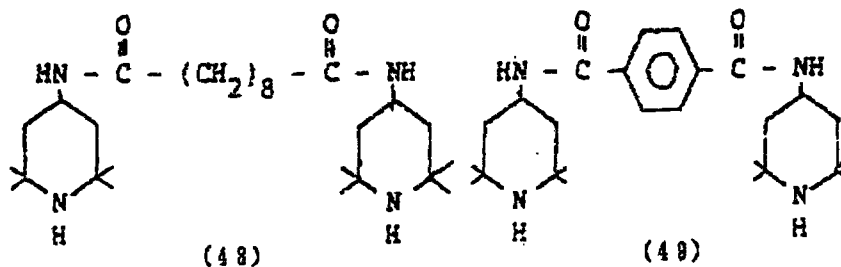


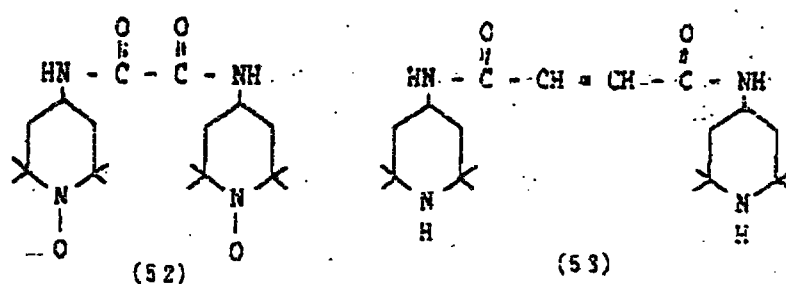
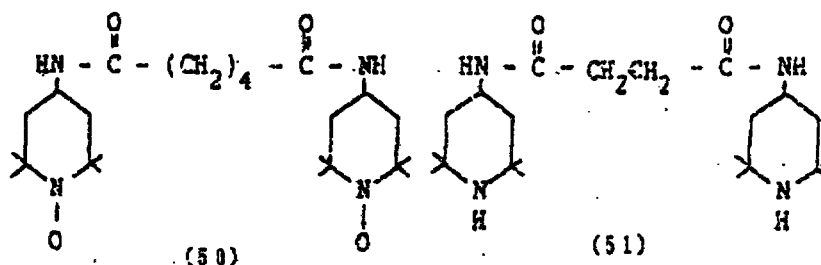
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and cyclohexylene and cyclohexylenedimethylene.  $R_3$  is preferably a hexamethylene group having 6 carbon atoms.

In the formula (3),  $R_4$  is a hydrogen atom or an alkyl group having 1-18 carbon atoms such as methyl, ethyl, propyl, isopropyl, n-butyl, isobutyl, t-butyl, sec-butyl, n-pentyl, n-hexyl, 2-ethylhexyl, n-octyl, 1,1,3,3-tetramethylbutyl, n-decyl, n-dodecyl, n-octadodecyl, cyclohexyl, 3,3,5-trimethylcyclohexyl, cyclooctyl and cyclodecyl.

In the formula (7),  $R_5$  represents a direct bond, a straight or branched alkylene group of 1-10 carbon atoms, an alkylidene group or an arylene group such as, for example, ethylene, 1,2-, 1,3- or 1,4-propylene, 1,2-, 1,3- or 1,4-butylene, pentamethylene, 2,2-dimethyl-1,3-propylene, hexamethylene, octamethylene, cyclohexylene and cyclohexylene-dimethylene. Specific examples of the compounds include N,N'-bis(2,2,6,6-tetra-methyl-4-piperidyl)-1,6-hexanediamine, N,N'-bis(2,2,6,6-tetra-methyl-4-piperidyl)ethylenediamine, N,N'-bis(2,2,6,6-tetra-methyl-4-piperidyl)-1,8-octanediamine and a compound represented by the following formulas (48) to (53).

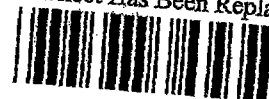
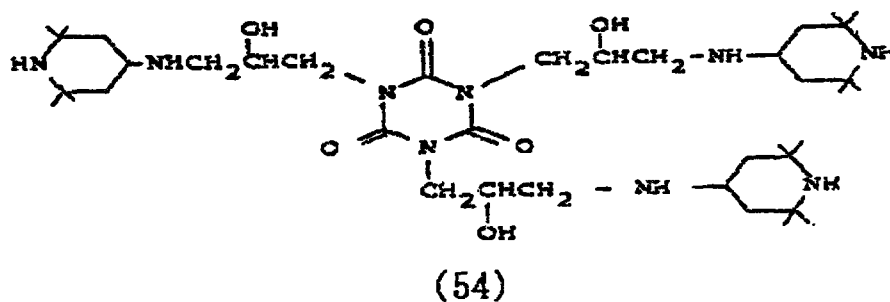




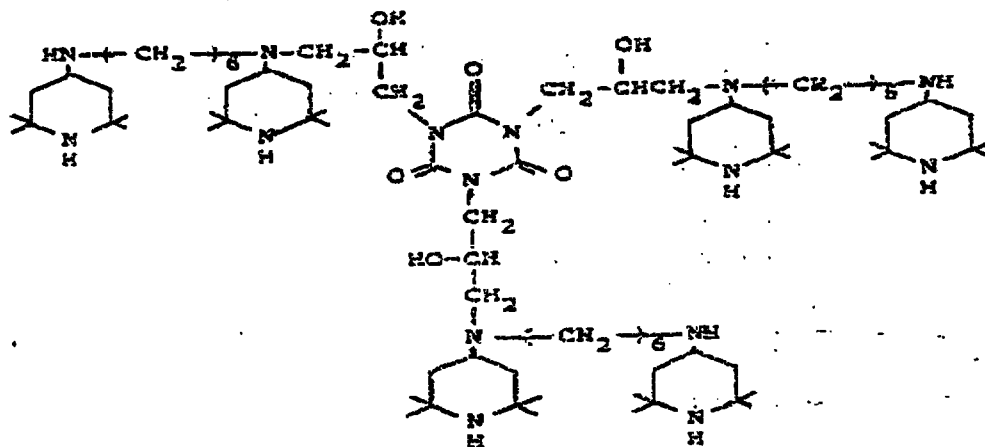
Examples of [n=3] of [III] and [VII] of the compound (c) of the present invention include compounds below.

In the formula (2), R<sub>2</sub> is the group represented by the formula (8).

Specific examples of the compounds include a compound represented by the following formulas (54) and (55).



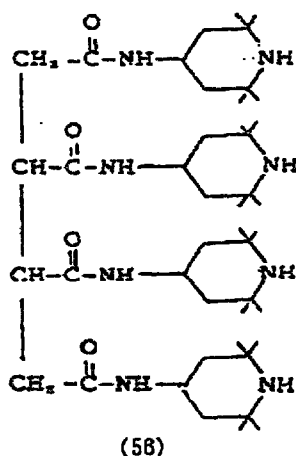
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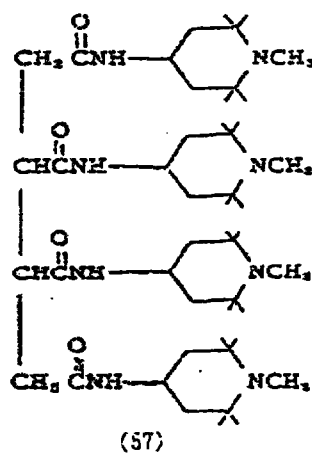
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Examples of [n=4] of [I] and [IV] to [VI] of the compound (c) of the present invention include compounds below. In the formula (1) or (2), R, is the group represented by the formula (9) or (10).

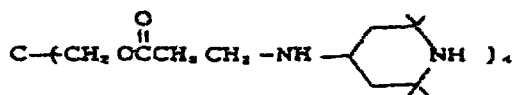
Specific examples of the compounds include a compound represented by the following formulas (56) to (58).



(58)



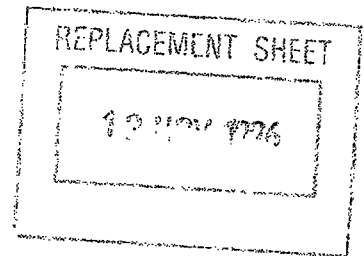
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The amount ratio of the components (a) and (b) is ;

(a) 1 to 99 % by weight of a polyphenylene ether resin and

(b) 99 to 1 % by weight of at least one non-crystalline thermoplastic resin, preferably ;

(a) 10 to 90 % by weight of a polyphenylene ether resin and

(b) 90 to 10 % by weight of at least one non-crystalline thermoplastic resin, more preferably ;

(a) 20 to 90 % by weight of a polyphenylene ether resin and

(b) 80 to 10 % by weight of at least one non-crystalline thermoplastic resin, and most preferably ;

(a) 40 to 85 % by weight of a polyphenylene ether resin,

(b) 60 to 15 % by weight of at least one non-crystalline thermoplastic resin.

The amount of the component (c) is 0.001-15 parts by weight, preferably 0.1-10 parts by weight based on 100 parts by weight of the sum of (a) the polyphenylene ether resin and (b) the non-crystalline thermoplastic resin. When it is smaller than 0.001 part by weight, no effect to inhibit gelation is seen and when it is larger than 15 parts by weight, problems such as foaming occur at the time of actual processing owing to volatile matters.

In the present invention, fillers may be added in order to improve mechanical properties such as strength, stiffness and hardness to the thermoplastic resin composition.



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The fillers include calcium carbonate, magnesium carbonate, aluminium hydroxide, magnesium hydroxide, zinc oxide, titanium oxide, magnesium oxide, magnesium sulfate, aluminium silicate, magnesium silicate, calcium silicate, silicic acid, calcium silicate hydrate, aluminium silicate hydrate, mineral fiber, zirconite, potassium titanate whisker, magnesium oxysulfate, glass balloon, glass fiber, glass bead, inorganic fiber such as carbon fiber, stainless fiber, aluminum fiber and aramide fiber, carbon black, inorganic fillers such as a metal whisker, silica, alumina, talc, mica, clay, kaolinite,  $\text{TiO}_2$ ,  $\text{ZnO}$  and  $\text{Sb}_2\text{O}_3$ , and one or more kind of these fillers can be blended. Besides, the fillers used in the present invention are not restricted by these.

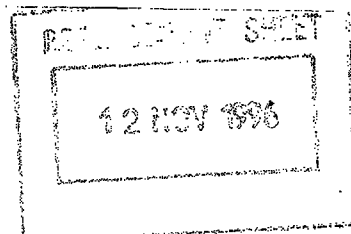
The thermoplastic resin composition of the present invention is obtained by blending and melt kneading the components (a), (b) and (c) with conventional methods. And the components (b) and (c) may be added in polymerization process of the component (a).

The thermoplastic resin composition of the present invention can be molded by various methods such as an injection molding, an extrusion molding, a blow molding, a sheet molding and film molding, which are most suitable since the composition is highly prevented from deterioration due to heat.

Besides, a molded article provided by the present

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invention can be widely used as a packaging material, household appliances, an exterior material for automobiles parts and an interior decoration material such as trim parts of automobiles.

#### EXAMPLE

The Example of the present invention is illustrated in detail according to Examples and Comparative Examples as follows, but those are simple exemplifications, and the present invention is not limited to them.

Each Example and Comparative Example was obtained by blending the components shown in Tables 1 to 3. That is, those components were extruded with a twin screw kneading machine (TEM-50A manufactured by Toshiba Machine Co., Ltd.) at a cylinder temperature of 260 °C, and pelletized to obtain the composition by a strand cutter after cooling with a water basin. Thus obtained pellets were dried at 130 °C for 2 hours under vacuum drying, and molded to obtain a test piece with an injection molding machine (IS220EN manufactured by Toshiba Machine Co., Ltd.) at a cylinder temperature of 290 °C, an injection pressure of 1200kg/cm<sup>2</sup> and a mold temperature of 80 °C.

Thus obtained pellets and the test piece were evaluated by the following method, and the results were obtained.



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[The molecular weight of the polyphenylene ether]

The pellets provided were dissolved in chloroform and measured with a gel permeation chromatograph (GPC) (HLC-8020 manufactured by Toso Co., Ltd.; columns of K-806, K-805, K-804 and K-803; a detector of UV having a wave length of 290 nm; a temperature of 40 °C; a flow rate of 1.0 ml/min), and the molecular weight converted with that of the standard polystyrene was determined.

[Thermal resistance stability]

After a thermal hysteresis was given to the test piece having 3.2 mm in thickness for Izod impact test, obtained by the above-mentioned injection molding, by letting alone in a constant temperature of 150 °C for 100 and 500 hours, it was notched in accordance with ASTM D256, and Izod impact test was conducted at a temperature of 23 °C.

The materials illustrated as follows were prepared in order to obtain the compositions of Examples and Comparative Examples.

(Polyphenylene ether ; PPE)

The polyphenylene ether (PPE) was obtained by homopolymerization of 2,6- dimethylphenol and having a logarithmic viscosity of 0.46 in chloroform solution (0.5g/dl) at 30°C. The molecular weight (Mw) of PPE before melt-kneaded was 46,100.

(Polycarbonate ; PC)

The trade name of CALIBRE 300-10 manufactured by



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Sumitomo-Dow Co., Ltd., was used.

(Polymethyl methacrylate ; PMMA)

The trade name of Sumipex LG manufactured by Sumitomo Chemical Co., Ltd.

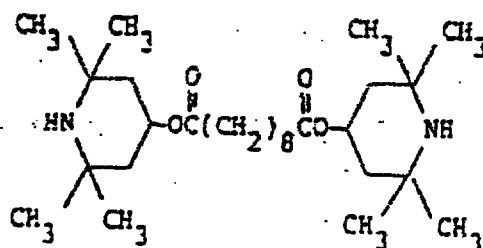
(Polystyrene)

Polystyrene 1 (PS-1) : The trade name of Sumibright E183N manufactured by Sumitomo Chemical Co., Ltd.

Polystyrene 2 (PS-2) : The trade name of Sumibright UG3000 manufactured by Sumitomo Chemical Co., Ltd.

(Additive)

Additive 1 : The trade name of Sanol LS-770 manufactured by Sankyo Co., Ltd. It is represented by the following chemical formula (59).



(59)

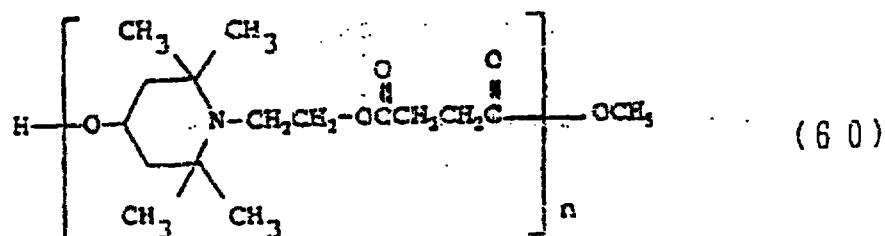
Additive 2 : The trade name of Tinuvin 622 manufactured by Ciba-Geigy Corp. It is represented by the following chemical formula (60).

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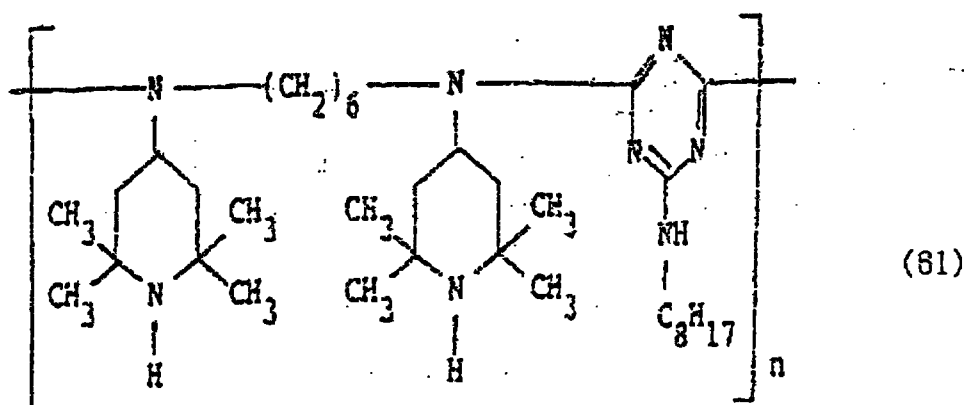


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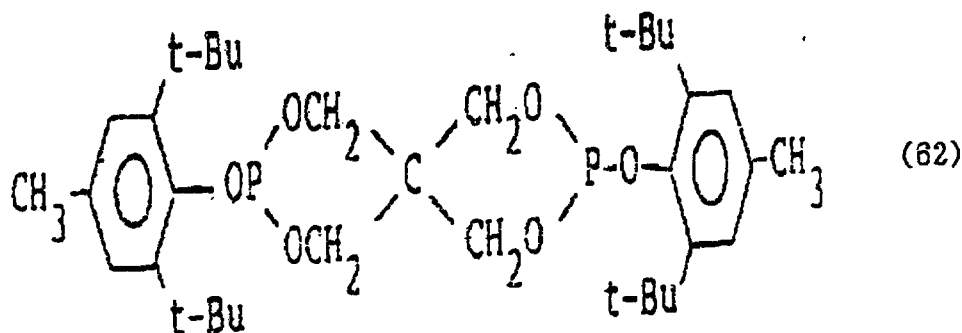
- 41 -



Additive 3 : The trade name of Chimasorb 944 manufactured by Ciba-Geigy Corp. It is represented by the following chemical formula (61).

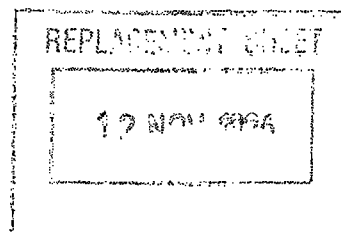


Additive 4 : The trade name of Adekastab PEP-36 manufactured by Asahi Denka Kogyo K.K. It is represented by the following chemical formula (62).



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The compounds used in Examples are prepared by the Reference Examples shown below. Besides, % in those Reference Examples is by weight unless otherwise notified.

Reference Example 1

Preparation of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 1)

In a 5 liter autoclave were charged 981 g (6.32 moles) of 2,2,6,6-tetramethyl-4-piperidine, 350 g (3.01 moles) of hexamethylenediamine, 1500 g of methanol and 5.0 g of carbon containing 5 % platinum thereon with keeping the temperature of at 45 to 55°C. The inside of the autoclave was pressurized to 40 bars with hydrogen and heated to 70-80 °C to carry out hydrogenation. The hydrogenation was completed after 4 to 5 hours. The catalyst was removed by filtration at 70 to 80 °C under 2 to 3 bars, and 1070 g of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 1) was obtained by distillation. Yield : 90 %. Boiling point : 188 to 190 °C/0.80 mbar.

Reference Example 2

Preparation of N,N'-bis( 2-methoxycarbonyl ethyl )-N,N'-bis(2, 2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 2)

In a 1 liter four-necked flask were charged 80 g (0.20 moles) of N,N'-bis(2, 2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 1) obtained in Reference Example 1 and 200 g of methanol, and they were completely dissolved.



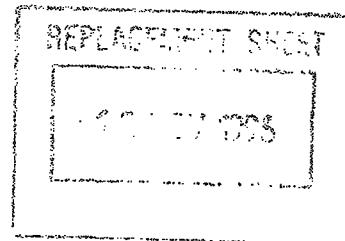
Thereto was added dropwise of a solution prepared by dissolving 35 g (0.41 moles) of methyl acrylate in 100 g of methanol at room temperature. Then, reaction was allowed to proceed for 12 hours under reflux. After completion of the reaction, the solvent was distilled off and the residue was recrystallized from hexane to obtain 98 g (0.17 mol) of the Compound 2 as a white solid. Yield: 85 %. Melting point: 50-52 °C.

#### Reference Example 3

Preparation of N,N'-bis(2-octadecyloxycarbonylethyl)-N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 3)

In a four-necked flask provided with a stirrer and a reflux condenser were charged 20 g (35 mmoles) of N,N'-bis(2-methoxycarbonylethyl)-N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 2) obtained in Reference Example 2, 21 g (77 mmoles) of stearyl alcohol and 200 ml of toluene, followed by stirring under reflux. Thereto was added dropwise of a solution of 0.1 g (4 mmoles) of lithium amide in 3 g of methanol. Thereafter, the solvent was distilled off with addition of toluene and the reaction was completed. After the reaction was allowed to proceed for 4 hours, toluene was added, followed by cooling to room temperature. The organic layer was washed with water three times and then concentrated to obtain 23 g (22 mmol) of the Compound 3. Yield: 63 %. Melting





point : 45-46 °C.

#### Reference Example 4

Preparation of N,N'-bis( 2-t-butoxycarbonyl-ethyl)-N,N'-bis(2, 2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 4)

The procedure of Reference Example 3 was repeated except that t-butyl alcohol was used instead of stearyl alcohol, thereby obtaining 13 g of the Compound 4. Melting point : 72 to 73 °C.

#### Reference Example 5

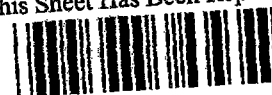
Preparation of N,N'-diacetyl-N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 5)

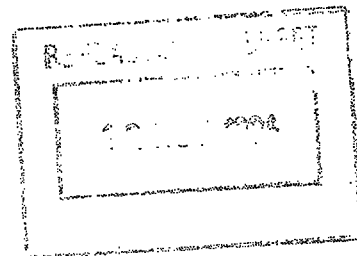
In a 300 ml four-necked flask were charged 20.7 g (50.7 mmoles) of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 1) obtained in Reference Example 1 and 70 g of pyridine, and they were completely dissolved. Thereto was added dropwise of 11.64 g (0.114 moles) of acetic anhydride over a period of 30 minutes, followed by 1 hour at 40 °C. After cooling, 100 g 20 % aqueous NaOH and 50 ml of toluene were added and the product was extracted and washed with 50 ml of water. The solvent in the toluene layer was distilled off and the residue was recrystallized from 20 ml of n-hexane to obtain 20.6 g of the Compound 5. Yield : 85 %. Melting point : 152-154 °C.

#### Reference Example 6

Preparation of N,N'-bis(n-hexylcarbonyl)-N,N'-bis(2,2,6,6-

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tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 6)

The procedure of Reference Example 5 was repeated except that heptanoic anhydride was used in place of acetic anhydride, thereby obtaining 18.8 g of the Compound 6. Melting point : 123-125 °C.

Reference Example 7.

Preparation of N,N'-bis(ethoxycarbonyl)-N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 7)

At a temperature of 0 °C or lower, 22.8 g (0.21 moles) of ethyl chlorocarbonate was added to solution of 39.4 g (0.1 moles) of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 1) obtained in Reference Example 1 in 200 ml of 1,2-dichloroethane which was cooled to -10 °C. Then, thereto was added slowly 8.4 g of sodium hydroxide dissolved in 50 ml of water with keeping the temperature of at 0 °C. Then, the temperature was elevated to 20 °C, and the reactant was separated to a water layer and an organic layer, and the organic layer was washed with water and dried over anhydrous sodium sulfate to remove the solvent. Then, the residue was crystallized from octane to obtain 44.1 g of the Compound 7. Yield : 82 %. Melting point : 125 °C.

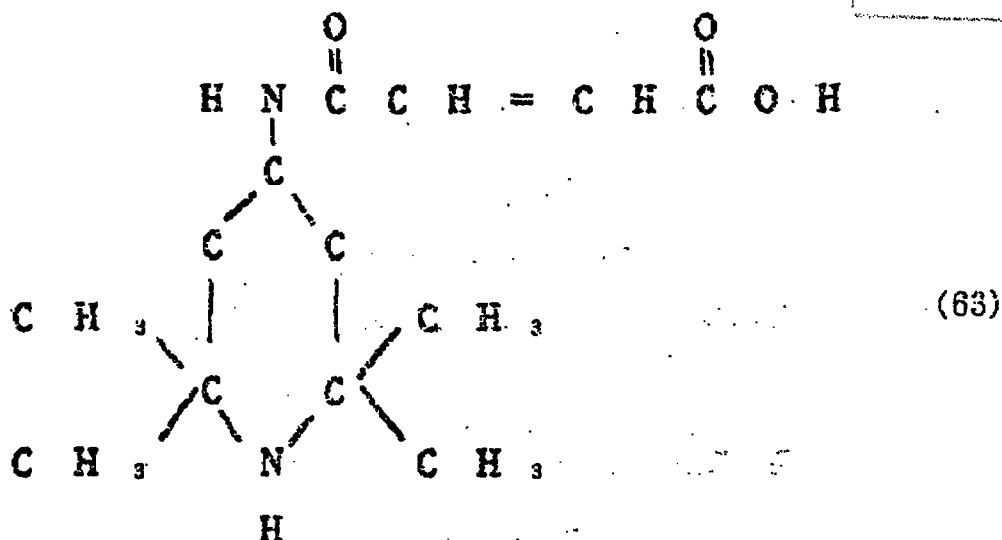
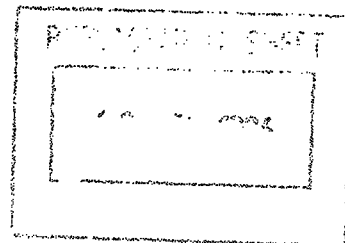
Reference Example 8

Preparation of Compound 8 [the chemical formula (63)]

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In a four-necked flask provided with a thermometer and a stirrer were charged 156 g of 4-amino-2,2,6,6-tetramethylpiperidine, 400 ml of toluene and 98g of maleic anhydride and the atmosphere in the flask was replaced with nitrogen, followed by keeping the temperature of at 60 °C for 6 hours with stirring. After completion of the reaction, the produced white crystal was filtrated and washed with toluene to obtain 243 g of the Compound 8.

#### Reference Example 9

The 2,2,6,6-tetramethyl-4-piperidylamine was used as Compound 9.

#### Reference Example 10

Preparation of methyl 2,2,6,6-tetramethyl-4-piperidyl-amino-propionate (Compound 10)

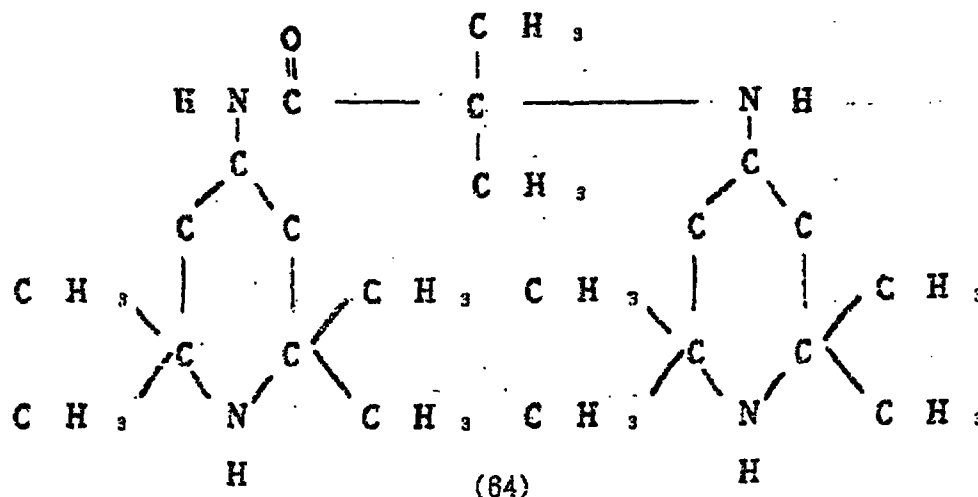
The 7.8 g (0.05 mol) of 2,2,6,6-tetramethyl-4-piperidine and 5.6 g (0.065 mol) of methyl acrylate were allowed to react



in 50 g of methanol under reflux for 3 hours. The solvent was distilled to obtain 8.0 g of colorless liquid of methyl 2,2,6,6-tetramethyl-4-piperidyl-aminopropionate (Compound 10) as a fraction of 130-136 °C/1.5mmHg.

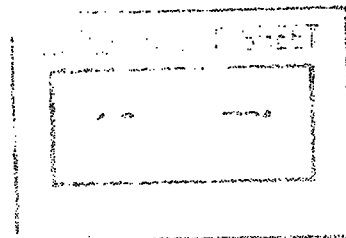
Reference Example 11

Preparation of Compound 11: [the chemical formula (64)]



In a four-necked flask provided with a thermo-meter and a stirrer were charged 156 g of 4-amino-2,2,6,6-tetramethyl-piperidine, 32 g of acetone, 239 g of chloroform and 0.4 g of benzyltrimethylammonium chloride, and cooled to 5 °C with stirring. Then, thereto was added dropwise of 257.6 g of a 50 % aqueous potassium hydroxide solution over a period of 1 hour with keeping the temperature of at 5 to 10 °C, and thereafter, the reaction was allowed to proceed for 5 hours at 0 to 10 °C. After completion of the reaction, the aqueous layer was separated to a water layer and an organic layer, and the water

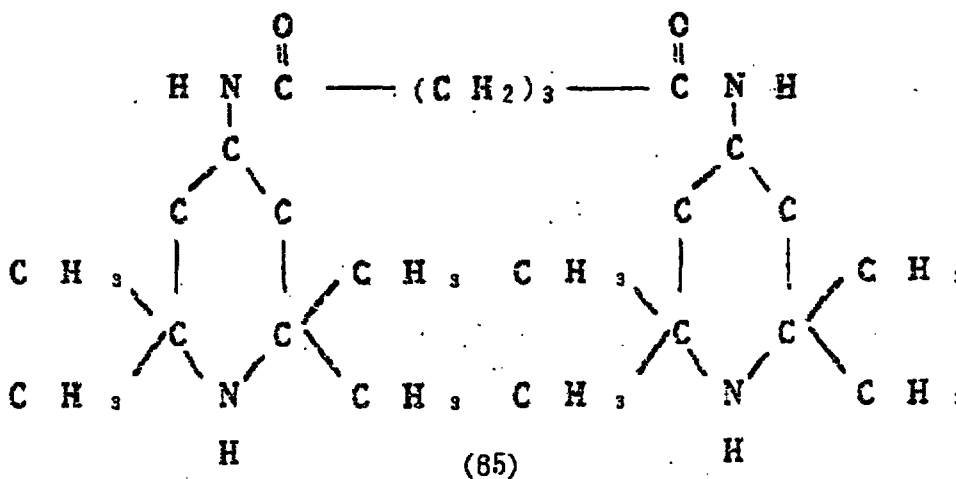




layer was removed. Then, the excess chloroform in the organic layer was distilled off to obtain 176 g. of a white crystal as Compound 11. Yield : 92.6 %. Melting point : 127-128 °C. A parent ion peak of 380 was confirmed by FD-mass spectrometry.

#### Reference Example 12

Preparation of Compound 12 [the chemical formula(65)]



In a four-necked flask provided with a thermo-meter and a stirrer were charged 156 g. of 4-amino-2,2,6,6-tetramethylpiperidine, 400 ml of toluene and 121 g. of triethylamine, and the atmosphere in the flask was replaced with nitrogen. After the content was cooled to 0 °C, thereto was added dropwise of 119 g of sebacyl chloride under stirring with keeping the temperature of at 0 to 5 °C. After completion of the addition, 200 ml of 20% aqueous sodium hydroxide solution was added to dissolve the hydrochloride of triethylamine produced by the reaction. After the aqueous was separated to a water layer and an organic layer, the water layer was removed, and the



organic layer was washed with 100 ml of water. The organic layer was concentrated and the residue was crystallized from n-hexane to obtain 203 g of a white crystal as Compound 12.

#### Reference Example 13

Preparation of tris[2-hydroxy-3-(2,2,6,6-tetramethyl-4-piperidylamino)-propyl]-isocyanurate (Compound 13)

Tris(2,3-epoxypropyl)isocyanurate (29.7 g) and 4-amino-2,2,6,6-tetramethylpiperidine (47.5 g) were slowly heated in a hot bath under stirring. Heat was generated at about 80 °C. The temperature was raised up to  $100 \pm 5$  °C and the temperature was held for about two hours. After cooling, white powders of tris[2-hydroxy-3-(2,2,6,6-tetramethyl-4-piperidyl-amino)-propyl]-isocyanurate (74.5 g) (Compound 13) was obtained. Yield : 97.4 %. Melting point : 55-60 °C.

#### Examples 1 to 15

The blend ratios of Examples and the molecular weights of the polyphenylene ether in the obtained compositions (after melt-kneaded) were shown in Table 1.

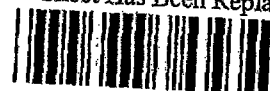
#### Comparative Examples 1 to 7

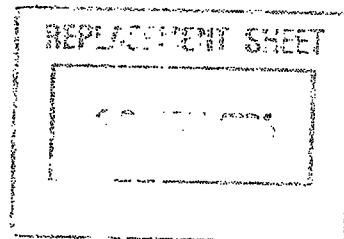
The compositions without the additives and Comparative Examples using the additives 1 to 4 were shown in Table 2.

#### Examples 16, 17 and Comparative Example 8

Thermal resistance stability of the compositions are shown in Table 3.

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EFFECT OF THE PRESENT INVENTION

Deterioration of polyphenylene ether caused by the heat at the time of processing can be markedly improved by adding a specific hindered piperidine compound to the composition comprising the polyphenylene ether and the non-crystalline thermoplastic resin.

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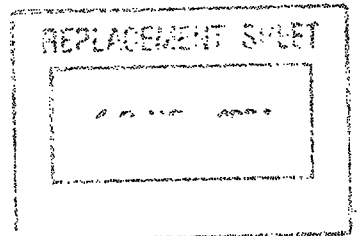


REPLACEMENT SHEET

[Table 1]

|            |        | Composition         |                  | Molecular weight of<br>polyphenylene ether Mw |
|------------|--------|---------------------|------------------|-----------------------------------------------|
|            |        |                     |                  |                                               |
| Example 1  | PPE 70 | Polystyrene 1<br>30 | Compound 1<br>1  | 61,000                                        |
| Example 2  | PPE 70 | Polystyrene 1<br>30 | Compound 2<br>1  | 51,000                                        |
| Example 3  | PPE 70 | Polystyrene 1<br>30 | Compound 3<br>1  | 62,000                                        |
| Example 4  | PPE 70 | Polystyrene 1<br>30 | Compound 4<br>1  | 65,000                                        |
| Example 5  | PPE 70 | Polystyrene 1<br>30 | Compound 5<br>1  | 68,000                                        |
| Example 6  | PPE 70 | Polystyrene 1<br>30 | Compound 6<br>1  | 68,000                                        |
| Example 7  | PPE 70 | Polystyrene 1<br>30 | Compound 7<br>1  | 52,000                                        |
| Example 8  | PPE 70 | Polystyrene 1<br>30 | Compound 8<br>1  | 64,000                                        |
| Example 9  | PPE 70 | Polystyrene 1<br>30 | Compound 9<br>1  | 62,000                                        |
| Example 10 | PPE 70 | Polystyrene 1<br>30 | Compound 10<br>1 | 68,000                                        |
| Example 11 | PPE 70 | Polystyrene 1<br>30 | Compound 11<br>1 | 70,000                                        |
| Example 12 | PPE 70 | Polystyrene 1<br>30 | Compound 12<br>1 | 69,000                                        |
| Example 13 | PPE 70 | Polystyrene 1<br>30 | Compound 13<br>1 | 53,000                                        |
| Example 14 | PPE 70 | Polycarbonate<br>30 | Compound 1<br>1  | 65,000                                        |
| Example 15 | PPE 70 | PMMA<br>30          | Compound 2<br>1  | 58,000                                        |



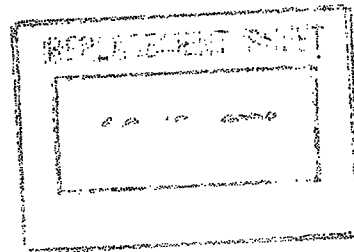


[Table 2]

|                          |           | Composition         |                 | Molecular weight of<br>polyphenylene ether Mw |
|--------------------------|-----------|---------------------|-----------------|-----------------------------------------------|
| Comparative<br>Example 1 | PPE<br>70 | Polystyrene 1<br>30 |                 | 76,000                                        |
| Comparative<br>Example 2 | PPE<br>70 | Polystyrene 1<br>30 | Additive 1<br>1 | 77,000                                        |
| Comparative<br>Example 3 | PPE<br>70 | Polystyrene 1<br>30 | Additive 2<br>1 | 79,000                                        |
| Comparative<br>Example 4 | PPE<br>70 | Polystyrene 1<br>30 | Additive 3<br>1 | 75,000                                        |
| Comparative<br>Example 5 | PPE<br>70 | Polystyrene 1<br>30 | Additive 4<br>1 | 77,000                                        |
| Comparative<br>Example 6 | PPE<br>70 | Polycarbonate<br>30 |                 | 82,000                                        |
| Comparative<br>Example 7 | PPE<br>70 | PMMA<br>30          |                 | 75,000                                        |

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[Table 3]

|                          |                     | Izod impact strength with a notch at 23 °C |          |          |
|--------------------------|---------------------|--------------------------------------------|----------|----------|
|                          |                     | aging at 150 °C for                        |          |          |
|                          |                     | Initial                                    | 100hours | 500hours |
| Comparative<br>Example 8 | PPE 60              | 7.2                                        | 2.1      | 2.3      |
|                          | Polystyrene 2<br>40 |                                            |          |          |
| Example 16               | PPE 60              | 10.1                                       | 5.5      | 5.7      |
|                          | Polystyrene 2<br>40 |                                            |          |          |
| Example 17               | PPE 70              | 8.8                                        | 4.6      | 4.8      |
|                          | Polystyrene 2<br>30 |                                            |          |          |

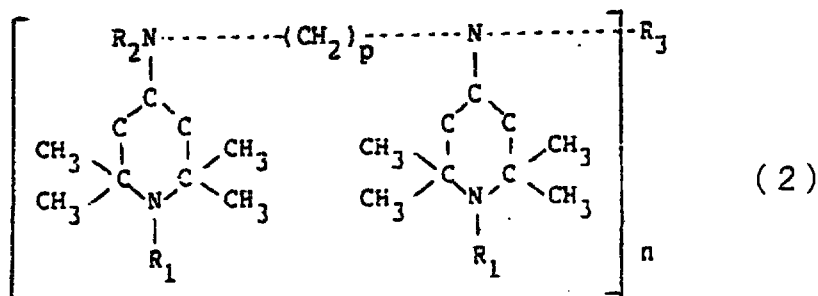
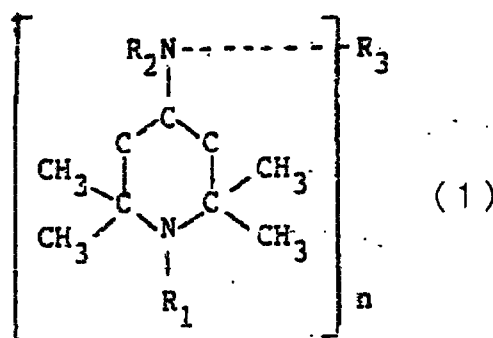
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WHAT IS CLAIMED IS:

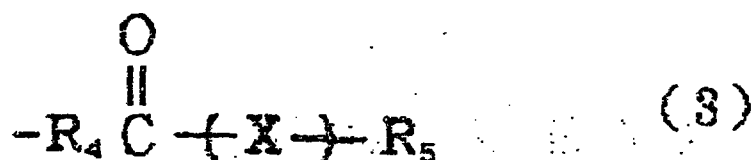
1. A thermoplastic resin composition which comprises
- (a) 1 to 99 % by weight of a polyphenylene ether resin,
- (b) 99 to 1 % by weight of at least one non-crystalline thermoplastic resin selected from an alkenyl-aromatic resin, a non-crystalline aromatic polyester resin, a methacrylate resin, a polyether imide resin and an aromatic polysulfone resin, and per 100 parts by weight of the sum of (a) and (b),
- (c) 0.001 to 15 parts by weight of at least one 4-amino-2,2,6,6-tetramethylpiperidyl compound represented by the following formula (1) or (2):



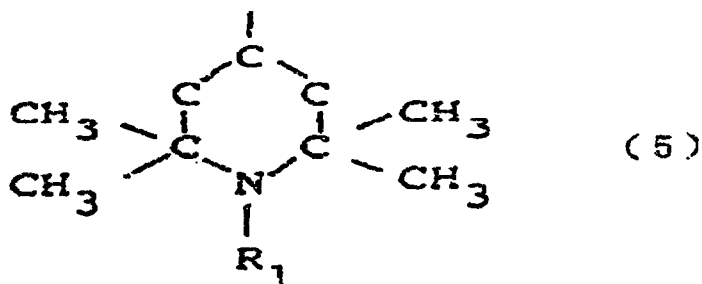
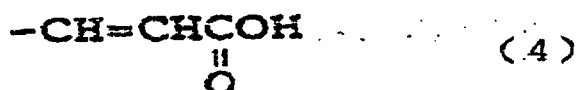


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; wherein  $R_1$  represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group.  $R_2$  represents a hydrogen atom, an alkyl group or a group represented by the following formula (3),



in which X represents a direct bond, -O- or -NH-,  $R_4$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and  $R_5$  represents a hydrogen atom, an alkyl group, an aryl group or a group represented by the following formula (4) or (5),



; and wherein n represents an integer of 1 to 4, p represents

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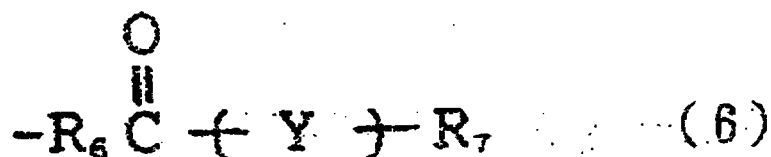




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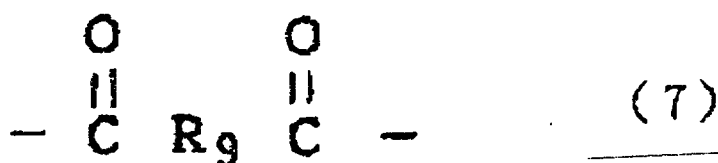
an integer of 2 to 6, and  $R_1$  represents an  $n$  valent organic group.

2. A thermoplastic resin composition according to claim 1, wherein in the formula (1) or (2),  $n$  is 1 and  $R_1$  is a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group, a benzyl group, an aryl group or a group represented by the following formula (6);



, wherein  $Y$  represents a direct bond,  $-O-$  or  $-NR_8-$ ,  $R_6$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and  $R_7$  and  $R_8$  each represents a hydrogen atom, a hydroxyl group, an alkyl group or an aryl group.

3. A thermoplastic resin composition according to claim 1, wherein in the formula (1) or (2),  $n$  is 2 and  $R_1$  is an alkylene group or a group represented by the following formula (7);



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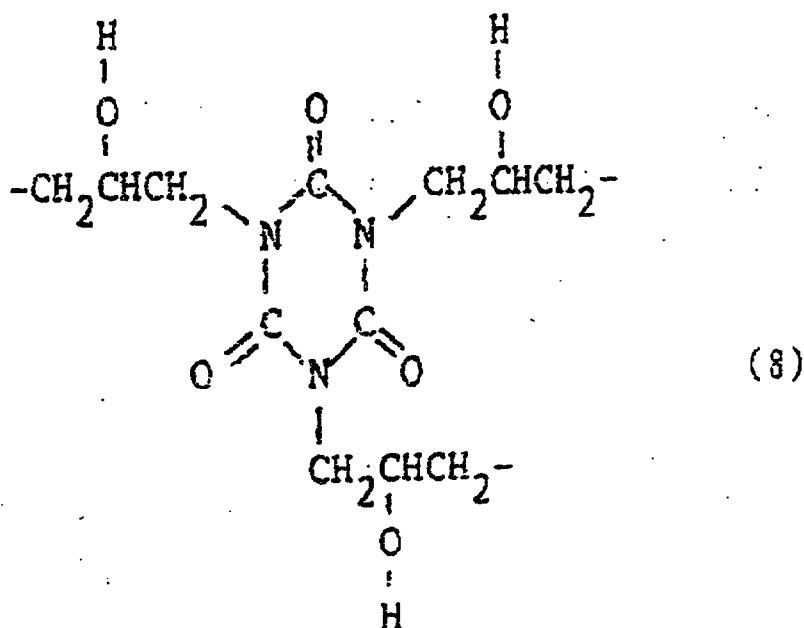




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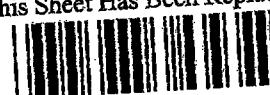
, wherein  $R_1$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group.

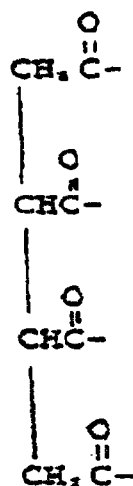
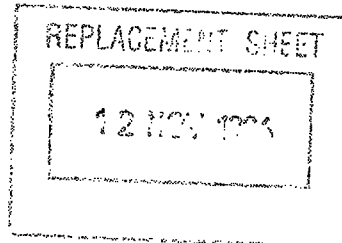
4. A thermoplastic resin composition according to claim 1, wherein in the formula (1) or (2),  $n$  is 3 and  $R_1$  is a group represented by the following formula (8);



5. A thermoplastic resin composition according to claim 1, wherein in the formula (1) or (2),  $n$  is 4 and  $R_1$  is a group represented by the following formula (9) or (10);

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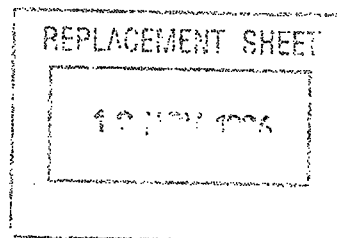
(9)



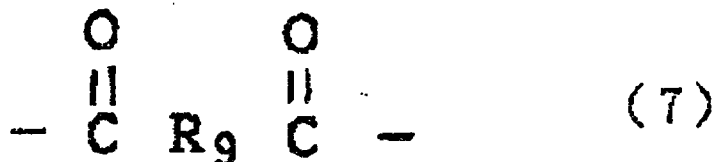
(10)

6. A thermoplastic resin composition according to claim 1, wherein in the formula (1),  $R_1$  and  $R_2$  each represents a hydrogen atom or an alkyl group having 1-18 carbon atoms and  $n$  is 1 or 2, and (I) when  $n$  is 1,  $R_1$  is a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group or the group represented by the formula (6) in which  $\text{Y}$  represents a direct bond, -O- or -NR<sub>3</sub>-,  $R_3$  represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group or arylene group, and  $R_4$  and  $R_5$  each represent a hydrogen atom, an alkyl group having 1-18 carbon atoms, an aryl group having 6-21 carbon atoms or a group represented by the formula (4) or (5), and (II) when  $n$  is 2,  $R_1$  is a group represented by the





following formula (7);



, wherein  $\text{R}_9$  represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group or an arylene group.

7. A thermoplastic resin composition according to claim 1, wherein in the formula (1),  $n$  is 2 and  $\text{R}_2$  is an alkylene group.

8. A thermoplastic resin composition according to claim 1, wherein in the formula (1),  $n$  is 2,  $\text{R}_2$  is a hydrogen atom and  $\text{R}_3$  is an alkylene group.

9. A thermoplastic resin composition according to claim 1, wherein in the formula (1),  $n$  is 2,  $\text{R}_1$  is a hydrogen atom or a methyl group,  $\text{R}_2$  is a hydrogen atom and  $\text{R}_3$  is a hexamethylene group.

10. A thermoplastic resin composition according to claim 1, wherein in the formula (1),  $n$  is 2,  $\text{R}_2$  is an alkyl group and  $\text{R}_3$  is an alkylene group.

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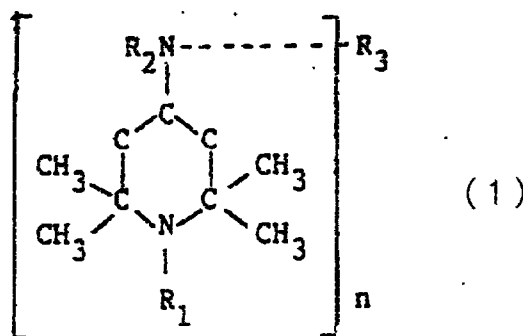


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11. A thermoplastic resin composition according to claim 1, wherein in the formula (1),  $n$  is 2,  $R_2$  is a group represented by the formula (3) in which  $X$  represents a direct bond,  $-O-$  or  $-NH-$ ,  $R_4$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group,  $R_5$  represents a hydrogen atom, an alkyl group, an aryl group or a group represented by the formula (4) or (5), and  $R_3$  is an alkylene group.

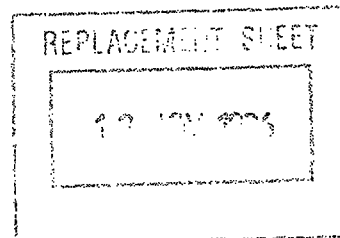
12. A thermoplastic resin composition according to claim 1, wherein the non-crystalline thermoplastic resin (b) is an alkenyl aromatic resin..

13. A thermoplastic resin composition according to claim 1, wherein the 4-amino-2,2,6,6-tetramethylpiperidyl compound (c) is a compound represented by the following formula (1) :

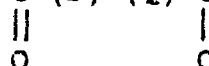


wherein  $R_1$  represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group having 1-8 carbon atoms, a cycloalkyl group, an allyl group, a benzyl group, an

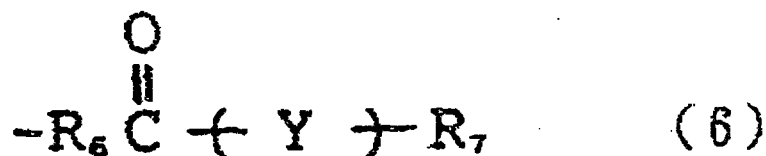




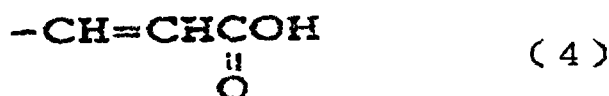
aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group, and n is an integer of 1, 2 or 4 ; and when n is 1, R<sub>2</sub> represents a hydrogen atom, an alkyl group having 1-8 carbon atoms or -C-(P)-(Q)-C-OH in which P



and Q represent one selected from the group consisting of an alkylene group, an alkylidene group, a vinyl group and a vinylidene group, and R<sub>3</sub> represents a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group, a benzyl group, an aryl group or a group represented by the following formula (6),

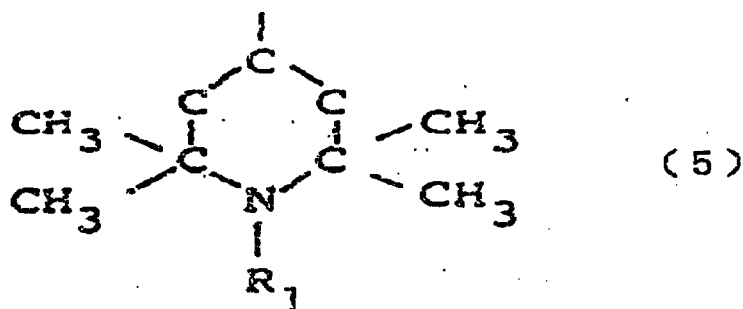
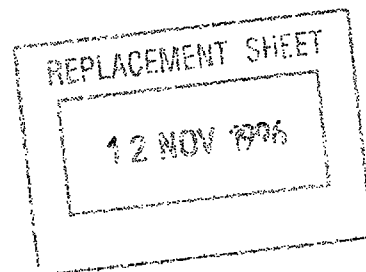


in which Y represents a direct bond, -O- -NR<sub>8</sub>-, R<sub>6</sub> represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and R<sub>7</sub> and R<sub>8</sub> each represents a hydroxyl group, an alkyl group, an aryl group or a group represented by the following formula (4) or (5),



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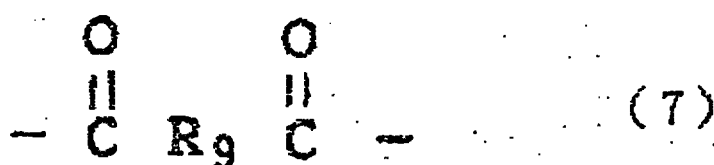




; when n is 2,  $R_1$  represents a hydrogen atom, an alkyl group having 1-8 carbon atoms or  $-\text{C}(\text{P})-\text{C}(\text{Q})-\text{OH}$  in which P

$$\begin{array}{c}
 \text{O} \\
 || \\
 \text{O}
 \end{array}
 \quad
 \begin{array}{c}
 \text{O} \\
 || \\
 \text{O}
 \end{array}$$

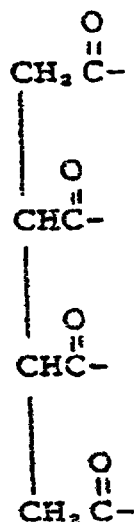
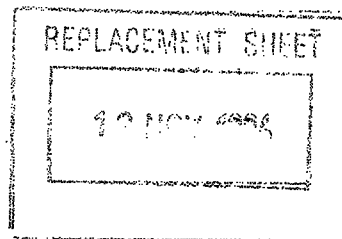
and Q represent one selected from the group consisting of an alkylene group an alkylidene group, a vinyl group and a vinylylidene group, and  $R_1$  represents an alkylene group or a group represented by the following formula (7),



in which  $R_1$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group; and when n is 4,  $R_1$  represents a hydrogen atom or an alkyl group having 1-8 carbon atoms, and  $R_2$  represents a group represented by the following formula (9) or (10),

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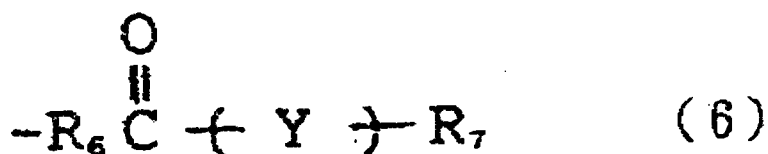


(9)



14. A thermoplastic resin composition according to claim 13, wherein in the formula (1),  $R_1$  and  $R_2$  each represents a hydrogen atom or an alkyl group having 1-8 carbon atoms and  $n$  is 1 or 2, and ;

(i) when  $n$  is 1,  $R_3$  is a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group or a group represented by the following formula (6),

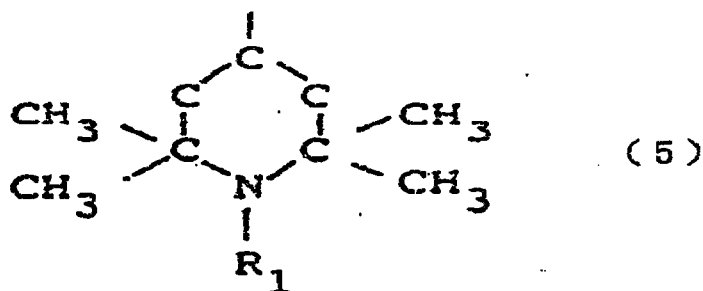
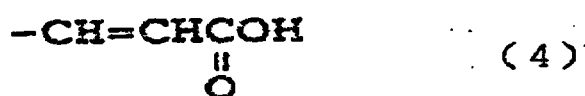


in which  $Y$  represents a direct bond,  $-O-$  or  $-NR_8-$ ,  $R_6$  represents a direct bond, a straight or branched alkylene

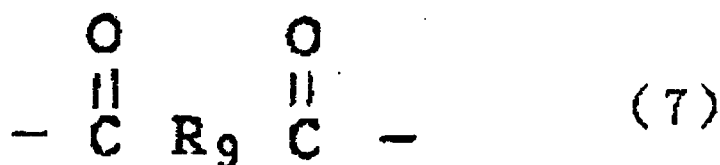




group having 1-10 carbon atoms, an alkylidene group or arylene group, and  $R_7$  and  $R_8$  each represent a hydroxyl group, an alkyl group having 1-18 carbon atoms, an aryl group having 6-21 carbon atoms or a group represented by the following formula (4) or (5),



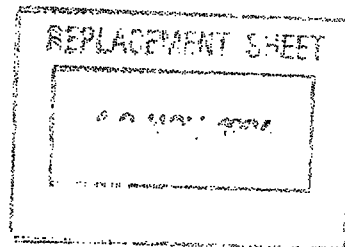
and (ii) when  $n$  is 2,  $R_9$  represents a group represented by the following formula (7),



in which  $R_9$  represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group or an arylene group.

15. A thermoplastic resin composition according to claim 13, wherein in the formula (1),  $n$  is 2 and  $R_7$  represents an





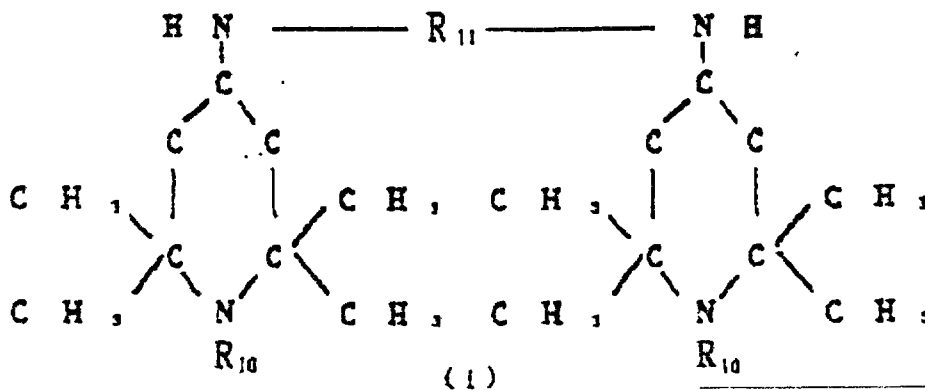
alkylene group.

16. A thermoplastic resin composition according to claim 13, wherein in the formula (1),  $n$  is 2,  $R_1$  represents a hydrogen atom and  $R_2$  represents an alkylene group.

17. A thermoplastic resin composition according to claim 13, wherein in the formula (1),  $n$  is 2,  $R_1$  represents a hydrogen atom or a methyl group,  $R_2$  represents a hydrogen atom and  $R_3$  represents a hexamethylene group.

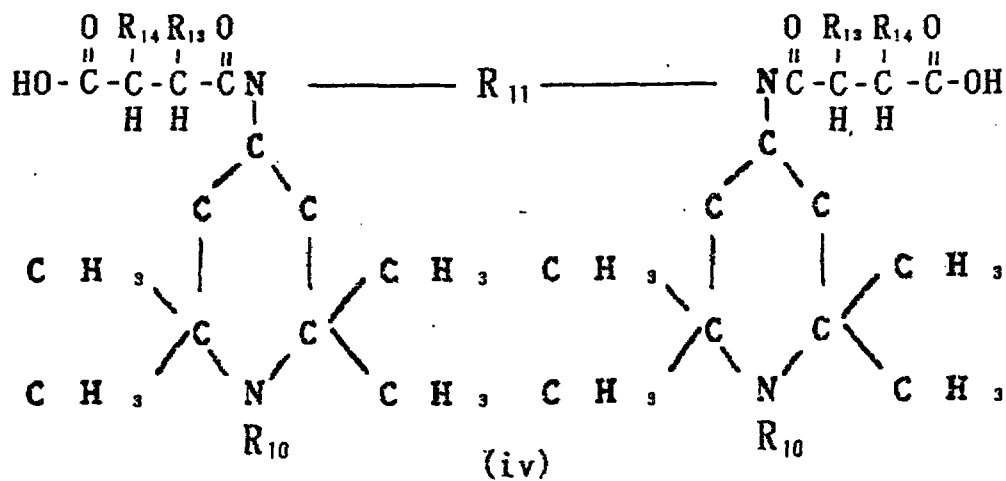
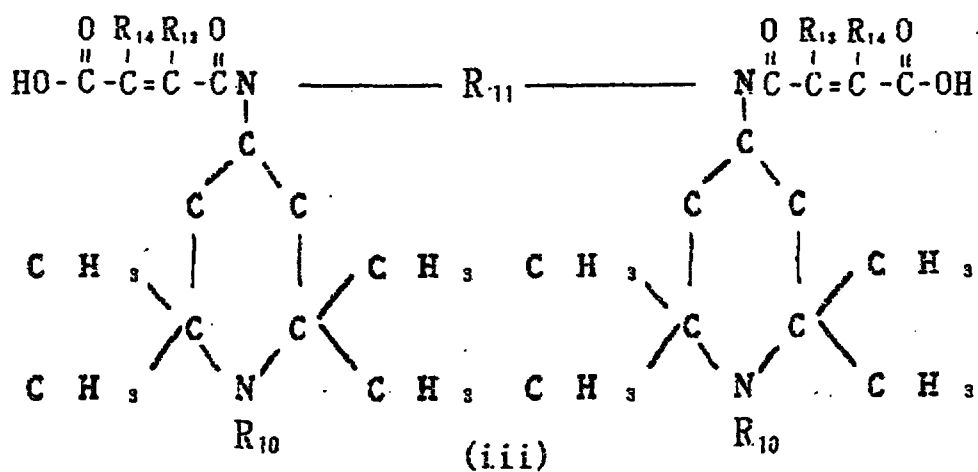
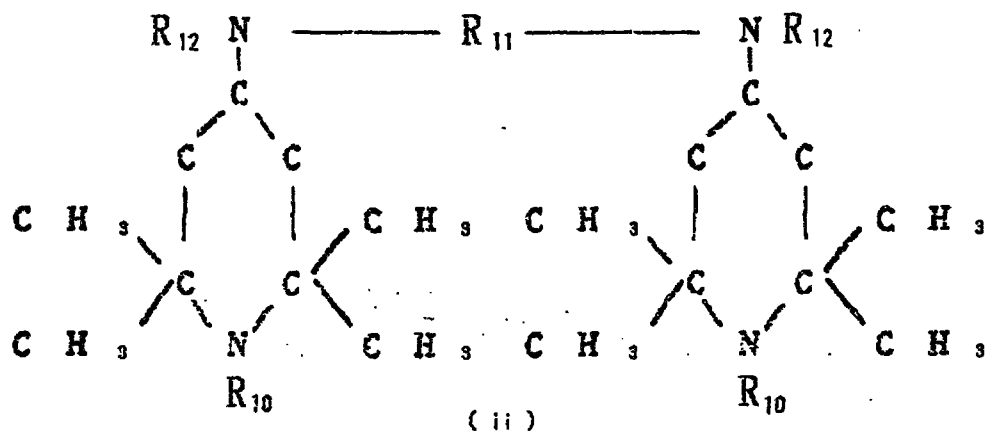
18. A thermoplastic resin composition according to claim 13, wherein in the formula (1),  $n$  is 2,  $R_1$  represents an alkyl group and  $R_2$  represents an alkylene group.

19. A thermoplastic resin composition according to claim 13, wherein the compound  $\text{(I)}$  represented by the formula (1) is at least one compound represented by the following formulas (i)-(iv):



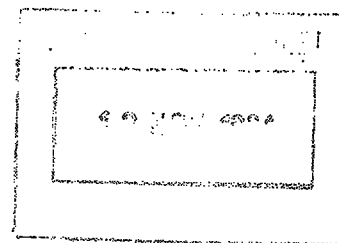
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a hydroxyl group.

wherein  $R_{10}$  represents a hydrogen atom, an alkyl group or an alkyloxy group,  $R_{11}$  represents an alkylene group,  $R_{12}$  represents an alkyl group, and  $R_{13}$  and  $R_{14}$  each represents a hydrogen atom or a methyl group.

20. A thermoplastic resin composition according to claim 19, wherein in the formula (i),  $R_{10}$  represents a hydrogen atom, a hydroxyl group, an alkyl group having 1-10 carbon atoms or an alkyloxy group having 1-10 carbon atoms, and  $R_{11}$  represents an alkylene group having 2-8 carbon atoms.

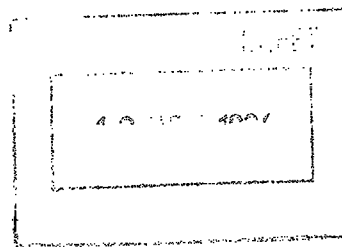
21. A thermoplastic resin composition according to claim 19, wherein in the formula (i),  $R_{10}$  represents a hydrogen atom or a methyl group and  $R_{11}$  represents a hexamethylene group.

22. A thermoplastic resin composition according to claim 19, wherein in the formula (ii),  $R_{10}$  represents a hydrogen atom, a hydroxyl group, an alkyl group having 1-10 carbon atoms or an alkyloxy group having 1-10 carbon atoms,  $R_{11}$  represents an alkylene group having 2-8 carbon atoms, and  $R_{12}$  represents an alkyl group having 1-18 carbon atoms.

23. A thermoplastic resin composition according to claim 19, wherein in the formula (ii),  $R_{10}$  represents a hydrogen atom or a methyl group.

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24. A thermoplastic resin composition according to claim 19, wherein in the formula (iii),  $R_{10}$  represents a hydrogen atom, a hydroxyl group, an alkyl group having 1-10 carbon atoms or an alkyloxy group having 1-10 carbon atoms, and  $R_{11}$  represents an alkylene group having 2-8 carbon atoms.

25. A thermoplastic resin composition according to claim 19, wherein in the formula (iii),  $R_{10}$  represents a hydrogen atom or a methyl group.

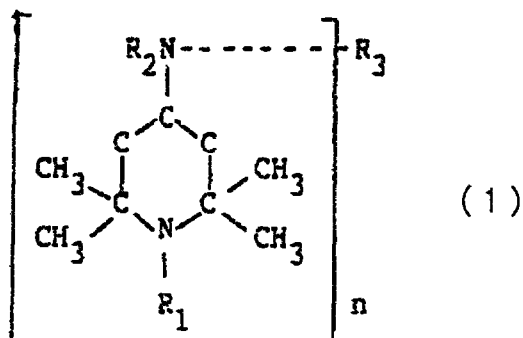
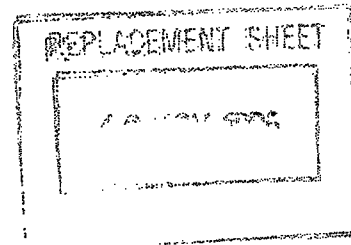
26. A thermoplastic resin composition according to claim 19, wherein in the formula (iv),  $R_{10}$  represents a hydrogen atom, a hydroxyl group, an alkyl group having 1-10 carbon atoms or an alkyloxy group having 1-10 carbon atoms, and  $R_{11}$  represents an alkylene group having 2-8 carbon atoms.

27. A thermoplastic resin composition according to claim 19, wherein in the formula (iv),  $R_{10}$  represents a hydrogen atom or a methyl group.

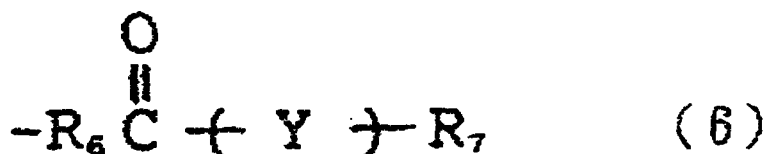
28. A thermoplastic resin composition according to claim 1, wherein the 4-amino-2,2,6,6-tetramethylpiperidyl compound (c) is a compound represented by the following formula (1) :

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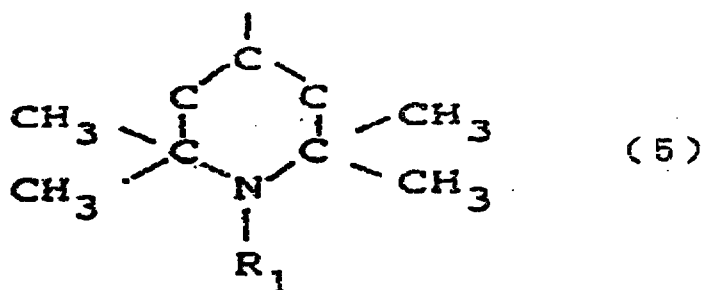
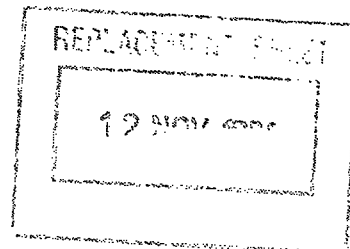
wherein  $R_1$  represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group having 1-8 carbon atoms, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group,  $R_2$  represents a hydrogen atom or alkyl group, and  $n$  is an integer of 1, 2 or 4 ; and  
(I) when  $n=1$ ,  $R_3$  is a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group, a benzyl group, an allyl group or a group represented by the following formula (6),



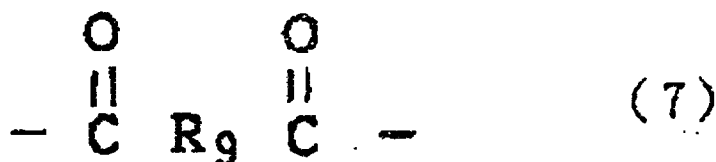
in which  $Y$  represents a direct bond,  $-O-$   $-NR_8-$ ,  $R_6$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and  $R_7$  and  $R_8$  each represents a hydroxyl group, an alkyl group, an aryl group or a group represented by the following formula (4) or (5),

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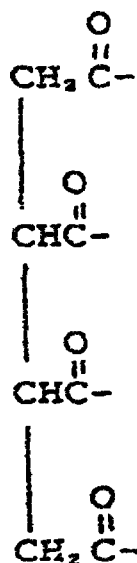
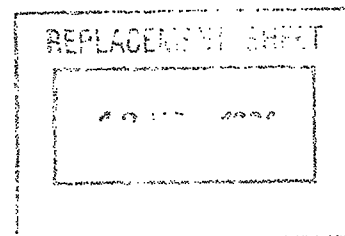
(II) when  $n=2$ ,  $R_3$  represents a group represented by the following formula (7),



in which  $R_9$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and

(IV) when  $n$  is 4,  $R_3$  represents a group represented by the following formula (9) or (10).

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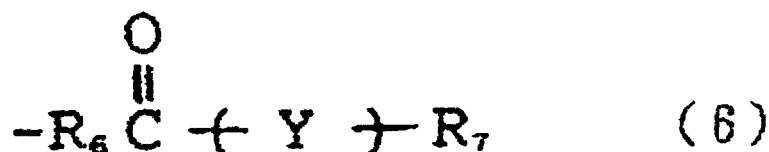


(9)



(10)

29. A thermoplastic resin composition according to claim 28, wherein in the formula (1),  $R_1$  represents a hydrogen atom or an alkyl group having 1-8 carbon atoms,  $R_2$  represents a hydrogen atom or an alkyl group having 1-18 carbon atoms, and  $n$  is an integer of 1 or 2 ; and when  $n$  is 1,  $R_3$  represents a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group or a group represented by the following formula (6),



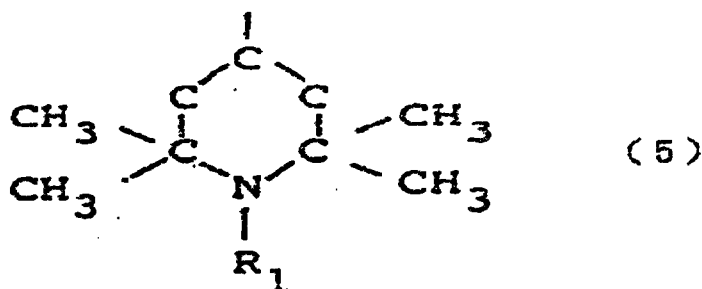
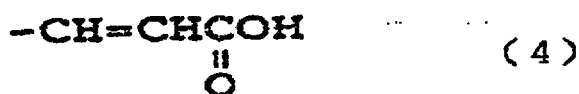
in which  $Y$  represents a direct bond,  $-O-$  or  $-NR_8-$ ,  $R_5$



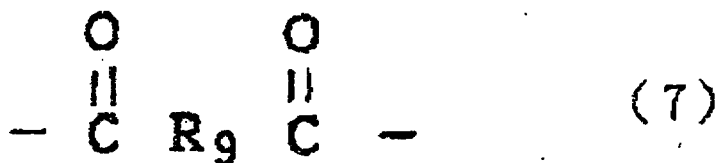


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represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group having 1-10 carbon atoms or arylene group, and  $R_7$  and  $R_8$  each represent a hydrogen atom, an alkyl group having 1-18 carbon atoms, an aryl group having 6-21 carbon atoms or a group represented by the following formula (4) or (5),



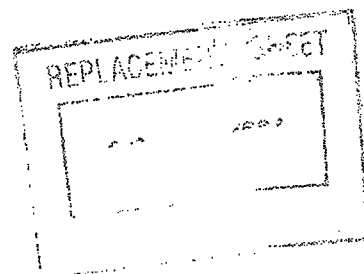
; and when  $n$  is 2,  $R_9$  represents a group represented by the following formula (7),



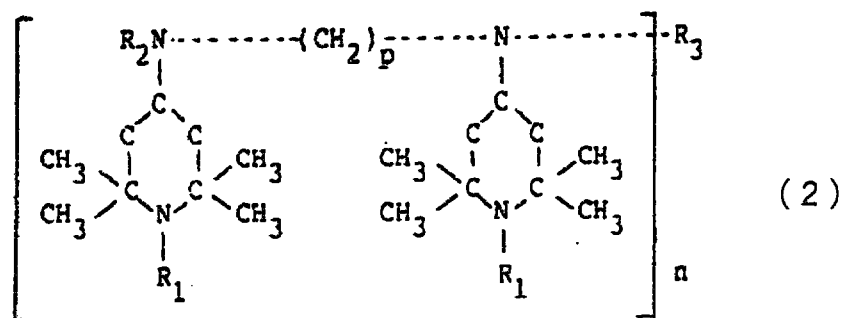
in which  $R_9$  represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group having 1-10 carbon atoms or an arylene group.

30. A thermoplastic resin composition according to claim





1, wherein the 4-amino-2,2,6,6-tetramethylpiperidyl compound (c) is a compound represented by the following formula (2) :



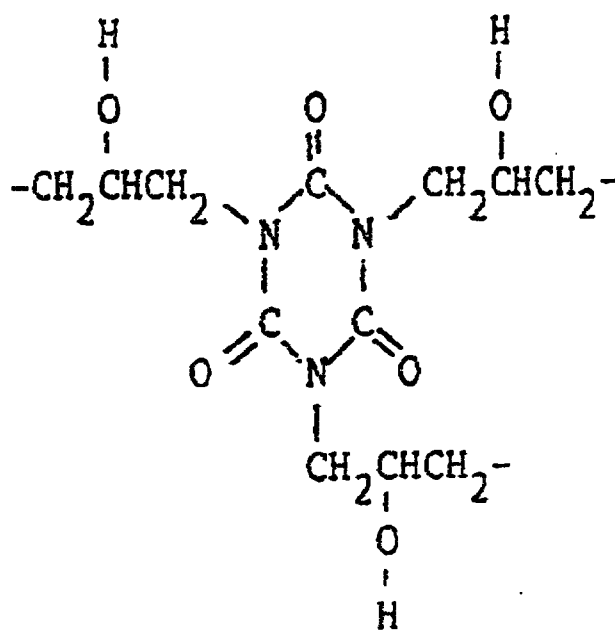
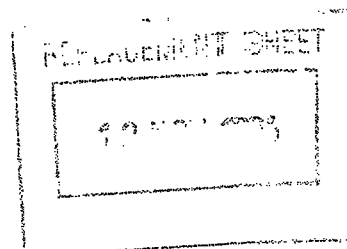
wherein  $\text{R}_1$  represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group having 1-8 carbon atoms, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group, and  $n$  is an integer of 3; and wherein  $\text{R}_2$  represents a hydrogen atom or an alkyl group having 1-8 carbon atoms and  $p$  is an integer of 2 to 8, and  $\text{R}_3$  represents a group represented by the following formula (8) ;

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A THERMOPLASTIC RESIN COMPOSITIONBACKGROUND OF THE INVENTIONField of The Invention

The present invention relates to a thermoplastic resin composition which comprises a polyphenylene ether and a non-crystalline thermoplastic resin, having improved properties in process stability and in heat stability on practically using.

Description of Related Art

A polyphenylene ether resin is a thermoplastic resin superior in various characteristics such as mechanical properties, heat resistance and dimensional stability. However, it is apt to undergo coloration and deteriorations such as cross-linking when molded at high temperatures. Especially, in the presence of oxygen, it undergoes serious deterioration due to the heat to cause gelation, increasing in molecular weight or sometimes produce black specks. Furthermore, polyphenylene ether resin readily undergoes discoloration with light.

In order to offset these defects of the resin, it has been proposed to add various additives. For example, phosphorus additives are proposed in Japanese Patent Kokoku No. 44-29751 and phenolic additives are proposed in Japanese Patent Kokoku No. 46-24782. Furthermore, many attempts such

as use of various stabilizers containing steric hindrance type amines or containing steric hindrance type amines and phenolic additives have been made mainly for inhibition of the discoloration with light as disclosed in Japanese Patent Kokai Nos. 60-149646 and 60-203150, U.S. Patent Nos. 4232131, 4233410, 4233412, 4234699, 4260689, 4260691 and 4299925, EP 094048A2, EP 134523A1, EP 146879A2, EP 146878A2, EP 151961A1, EP 149454A2, EP 233153A2, EP 240723A2, EP 229363A2 and EP 313118A1. Many of these attempts may be effective for improving (resistance) evaporation of stabilizers at the time of processing or actual use, and various compounds containing steric hindrance type amines alone or copolymers (oligomers) with other components have been proposed. However, none of these proposals have provided sufficient effects to inhibit deterioration at heating.

#### SUMMARY OF THE INVENTION

The present invention relates to a thermoplastic resin composition comprising a polyphenylene ether resin and a non-crystalline thermoplastic resin, which is inhibited from deterioration at the time of processing, especially with heat.

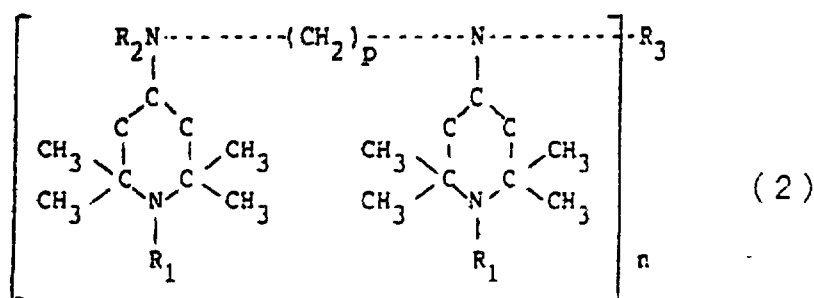
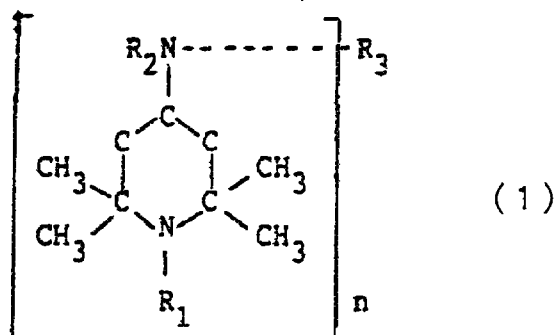
The present inventors have found that a conspicuous effect to inhibit the deterioration with heat can be obtained by adding a compound containing steric hindrance type amine having a specific structure to a thermoplastic resin composition comprising a polyphenylene ether resin and a non-

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crystalline thermoplastic resin.

The present invention relates to a thermoplastic resin composition which comprises :

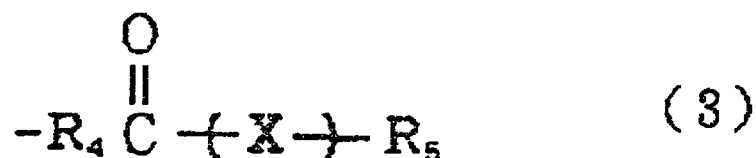
- (a) 1 to 99 % by weight of a polyphenylene ether resin,
- (b) 99 to 1 % by weight of at least one non-crystalline thermoplastic resin selected from an alkenyl aromatic resin, a non-crystalline aromatic polyester resin, a methacrylate resin, a polyetherimide resin and an aromatic polysulfone resin, and per 100 parts by weight of the sum of (a) and (b),
- (c) 0.001 to 15 parts by weight of at least one 4-amino-2,2,6,6-tetramethylpiperidyl compound represented by the following formula (1) or (2) ;



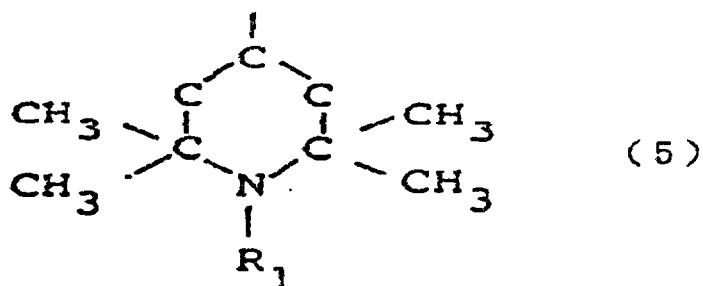
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; wherein  $R_1$  represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group, and  $R_2$  represents a hydrogen atom, an alkyl group or a group represented by the following formula (3),



in which X represents a direct bond, -O- or -NH-,  $R_4$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and  $R_5$  represents a hydrogen atom, an alkyl group, an aryl group or a group represented by the following formula (4) or (5),

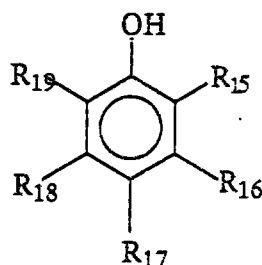


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; and wherein n represents an integer of 1 to 4, p represents an integer of 2 to 6, and R<sub>i</sub> represents an n valent organic group.

DETAILED DESCRIPTION OF THE PRESENT INVENTION

The polyphenylene ether resin (a) of the present invention is a polymer obtained by oxidation polymerization of at least one phenol compound represented by the following formula with oxygen or a gas containing oxygen using an oxidation coupling catalyst ;



wherein R<sub>15</sub>, R<sub>16</sub>, R<sub>17</sub>, R<sub>18</sub> and R<sub>19</sub> each represents a hydrogen atom, a halogen atom or a substituted or unsubstituted hydrocarbon group and at least one of them is a hydrogen atom.

Examples of R<sub>15</sub>, R<sub>16</sub>, R<sub>17</sub>, R<sub>18</sub> and R<sub>19</sub> are hydrogen, chlorine, bromine, fluorine, iodine, methyl, ethyl, n- or isopropyl, pri-, sec- or t-butyl, chloroethyl, hydroxyethyl, phenylethyl, benzyl, hydroxymethyl, carboxyethyl, methoxycarbonyl, cyanoethyl, phenyl, chlorophenyl, methylphenyl, dimethylphenyl, ethylphenyl and allyl.

Examples of the compounds represented by the above

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formula are phenol, o-, m- or p-cresol, 2,6-, 2,5-, 2,4- or 3,5-dimethylphenol, 2-methyl-6-phenylphenol, 2,6-diphenylphenol, 2,6-diethylphenol, 2-methyl-6-ethylphenol, 2,3,5-, 2,3,6- or 2,4,6-trimethylphenol, 3-methyl-6-t-butylphenol, thymol and 2-methyl-6-allylphenol.

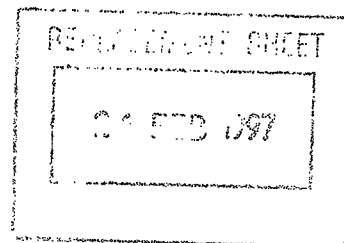
The phenol compounds of the above formula can also be copolymerized with other phenol compounds, for example, polyhydric aromatic compounds such as bisphenol A, tetrabromobisphenol A, resorcin, hydroquinone and novolak resins.

As preferable examples of the polyphenylene ether resin (a), mention may be made of homopolymers of 2,6-dimethylphenol, homopolymers of 2,6-diphenylphenol, copolymers of a larger amount of 2,6-dimethylphenol and a smaller amount of 3-methyl-6-t-butylphenol or 2,3,6-trimethylphenol.

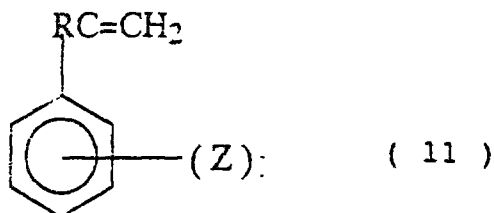
Any oxidation coupling catalysts may be used for oxidizing polymerization of the phenol compound as far as they have polymerization ability.

The non-crystalline thermoplastic resin (b) of the present invention is at least one selected from an alkenyl aromatic resin, a non-crystalline aromatic polyester resin, a methacrylate resin, a polyether imide resin and an aromatic polysulfone resin.

The alkenyl aromatic resin includes a homopolymer and a copolymer containing an aromatic vinyl compound represented by



the following formula (11) ;



wherein R represents a hydrogen atom, an alkyl group having 1 to 6 carbon atoms or a halogen atom, Z represents a hydrogen atom, a vinyl group, a halogen atom, an amino group or an alkyl group having 1-6 carbon atoms, and l is 0 or an integer of 1 to 5.

The example of the alkenyl aromatic resin includes a polymer containing one kind or plural kinds of polymerization unit selected from styrene,  $\alpha$ -methylstyrene and p-methylstyrene, polystyrene (PS resin), polychlorostyrene, poly- $\alpha$ -methylstyrene, styrene-acrylonitrile copolymer (AS resin), styrene- $\alpha$ -methylstyrene copolymer, styrene-4-methylstyrene copolymer, styrene-maleic anhydride copolymer and styrene-methylmethacrylate copolymer (MS resin).

Besides, it includes a copolymer of a compound containing 2 or more vinyl groups such as divinyl benzene and ethyleneglycol dimethacrylate, with the above-mentioned aromatic vinyl compound.

Further, the alkenyl aromatic resin includes a rubber-modified polymer containing a dispersed rubber phase in the matrix of the alkenyl aromatic resin phase. The rubber used

here includes a homopolymer or copolymer containing a diene component such as polybutadiene rubber (BR), polyisoprene rubber (IR), polychlorobutadiene rubber, a butadiene-styrene copolymer rubber, an isoprene-styrene copolymer rubber, a butadiene-acrylonitrile copolymer rubber (NBR), an isobutylene-butadiene or -isoprene copolymer rubber, an ethylene-propylene copolymer rubber (EPM) and an ethylene-propylene-non-conjugated diene copolymer rubber (EPDM).

Various methods are known as a method for dispersing the rubber into the matrix of the alkenyl aromatic resin phase, and for example, a method by blending mechanically, a method by solution-blending, and a method by dissolving a rubber into an alkenyl aromatic monomer and graft-copolymerizing said monomer, are mentioned, and any one of them may be used.

Besides, a polymerization method may be any one of polymerization methods such as an emulsion polymerization process, a bulk polymerization process (Japanese Examined Patent Kokoku No.42-662 and USP 3435096), a solution polymerization process (USP 3538190 and USP 3538191) and a suspension polymerization process (Japanese Examined Patents Kokoku No.49-10831, No.57-40166 and No.62-10565). The ratio of the rubber to the alkenyl aromatic monomer in the rubber-modified alkenyl aromatic resin is preferably 0.5 to 80 %, and more preferably 0.5 to 50% by weight of the rubber as total amount of 100.

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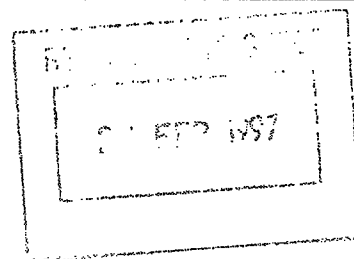
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As the examples of the rubber-modified alkenyl aromatic resin, a high impact polystyrene (HIPS) such as polystyrene modified with polybutadiene rubber, polystyrene modified with styrene-butadiene copolymer rubber and polystyrene modified with ethylene-propylene-non-conjugated diene copolymer rubber, an acrylonitrile-butadiene-styrene copolymer resin known as ABS resin and an acrylonitrile-(ethylene-propylene-non-conjugated diene copolymer rubber)-styrene resin known as AES resin are mentioned.

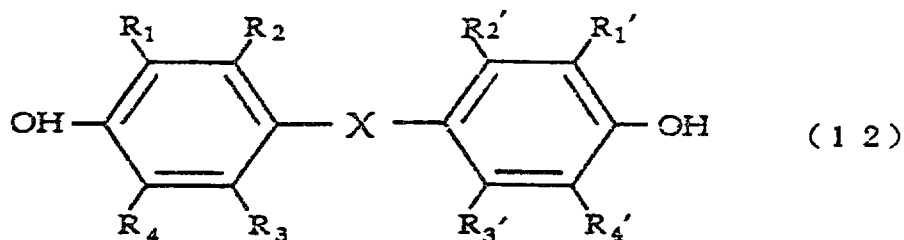
The non-crystalline aromatic polyester resin is polycarbonate resin and polyarylate resin.

The polycarbonate resin of the present invention is 4,4-dioxydialkanic polycarbonate including 4,4-dihydroxydiphenyl-2,2-propane (bisphenol A), and among them, 4,4-dihydroxydiphenyl-2,2-propane of polycarbonate having number average molecular weight of 15,000 to 80,000 is especially preferred. These polycarbonates are prepared with an arbitrary method. For example, a method of preparing by using 4,4-dihydroxydiphenyl-2,2-propane and phosgene in the presence of sodium hydroxide aqueous solution and a solvent, and a method of preparing by ester interchanging of 4,4-dihydroxydiphenyl-2,2-propane with carbonic diester in the presence of a catalyst are mentioned.

The polyarylate resin of the present invention is a polyester resin obtained from bisphenols and terephthalic acid



and/or isophthalic acid. As the bisphenols, those having a structure represented by the following formula (12) are used ;



In the formula (12), X is a group selected from -O-, -S-, -CO- and a hydrocarbon group having 1 to 10 carbon atoms, and R<sub>1</sub> to R<sub>4</sub> and R'<sub>1</sub> to R'<sub>4</sub> represent a group selected from a hydrogen atom, a halogen atom and a hydrocarbon group. When X represents a hydrocarbon group having 1 to 10 carbon atoms, an alkylene group, a branched alkylene group and a halogen-substituted alkyl group are included. On the other hand, each terephthalic acid and isophthalic acid can be used alone or in combination. It is obtained by polymerizing the above-mentioned bisphenols with terephthalic acid and/or isophthalic acid, and as a polymerization method, either of usual interfacial polymerization process or solution polymerization process may be used. Besides, these polyarylate resins having a molecular weight of 5,000 to 70,000 are preferred.

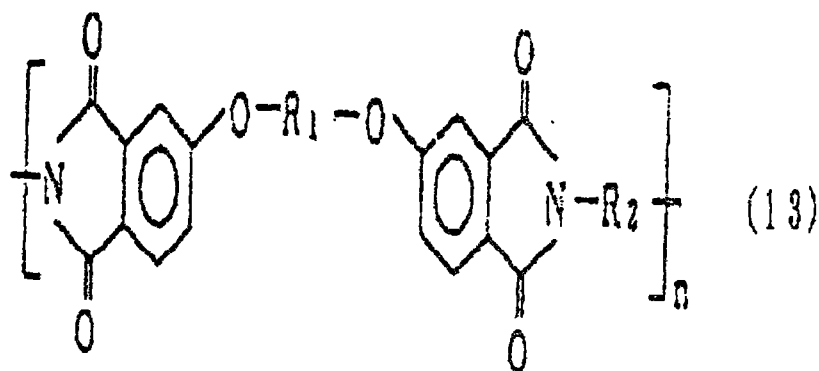
The methacrylate resin of the present invention is a homopolymer of methyl methacrylate or a copolymer of methyl methacrylate with acrylate, methacrylate, acrylic acid, methacrylic acid, styrene, acrylonitrile, glutaric anhydride, glutarimide or a diene rubber.

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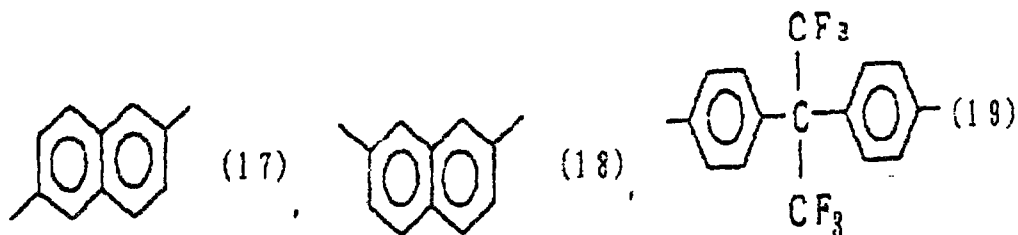
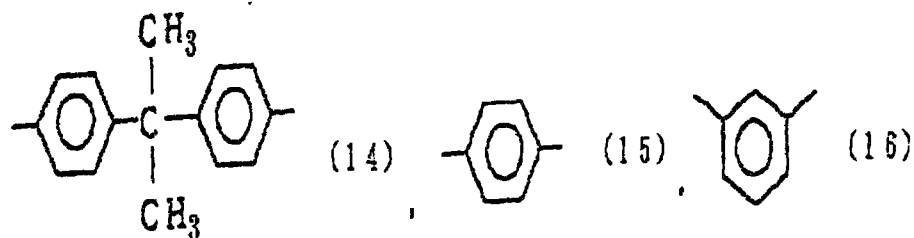
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Acrylate includes methyl acrylate, ethyl acrylate and butyl acrylate, and methacrylate includes methyl methacrylate, ethyl methacrylate, butyl methacrylate and cyclohexyl methacrylate.

The polyetherimide resin of the present invention is a resin represented by the following formula (13),



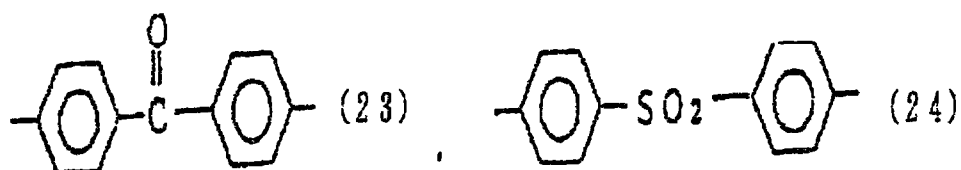
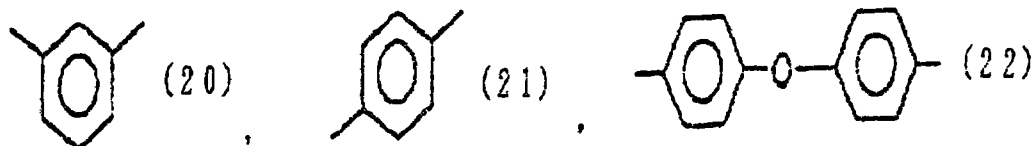
In the formula (13),  $n$  represents an integer of 1 to 1000, and  $R_1$  represents a group represented by the following formulas (14) to (19).



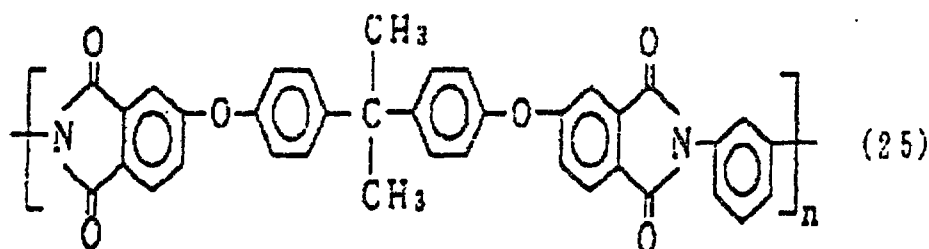
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$R_2$  represents a group represented by the following formulas (20) to (24).



Preferable example from the view points of property and production cost includes the structure represented by the following formula (25),

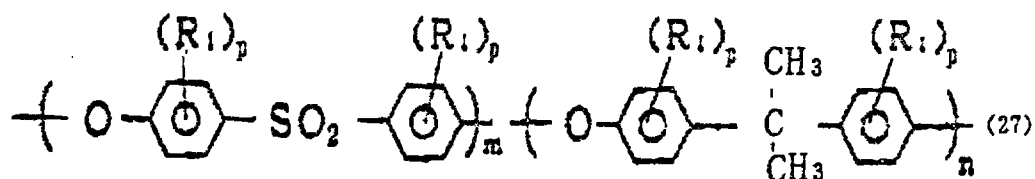
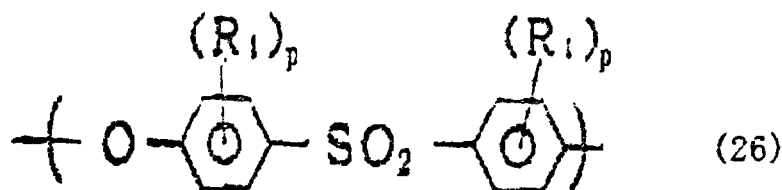


wherein  $n$  represents an integer of 1 to 1000.

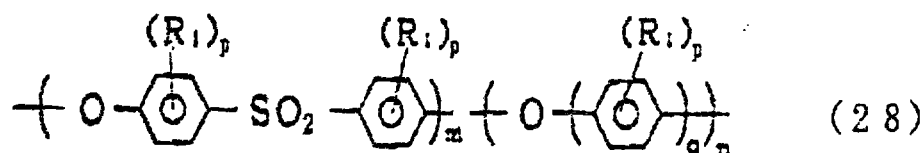
The aromatic polysulfone resin used in the present invention is defined as the polyarylene compound wherein arylene units are located in disorder or order with ether bond and sulfone bond. Those having a repeating unit represented by the following formulas (26) to (28) are mentioned as typical examples, but it is not limited to these.

3 FEB 1997

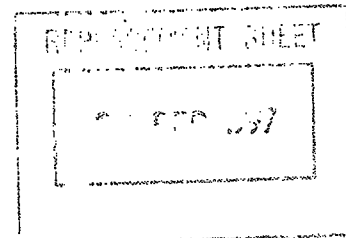
- 13 -



The compound of the formula (26) includes a homopolymer and the compound of the formula (27) includes a random copolymer, wherein  $\text{R}_1$  represents an alkyl group having 1 to 6 carbon atoms, an alkenyl group having 3 to 10 carbon atoms, a phenyl group or a halogen atom,  $p$  is an integer of 0 to 4, besides,  $m$  and  $n$  represent mean repeating unit numbers and are integers of 1 to 1000, each  $\text{R}_1$  on the same or different nucleus may be different mutually, and each  $p$  may be different mutually.



The compound of the formula (28) includes a random copolymer, wherein  $\text{R}_1$  represents an alkyl group having 1 to 6 carbon atoms, an alkenyl group having 3 to 10 carbon atoms, a

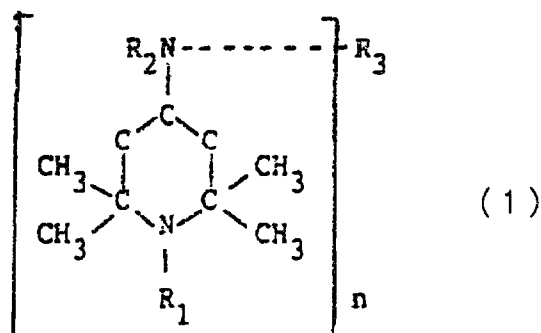


phenyl group or halogen atom,  $p$  is an integer of 0 to 4, besides,  $q$ ,  $m$  and  $n$  represent mean repeating unit numbers,  $q$  is an integer of 1 to 3,  $m$  and  $n$  are an integer of 0 to 100, each  $R_1$  on the same or different nucleus may be different mutually, and each  $p$  may be different mutually.

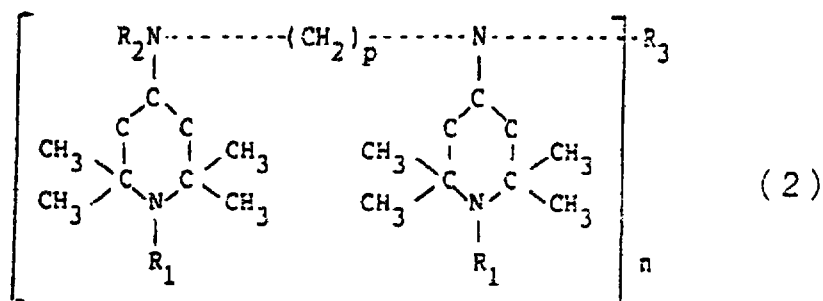
As the aromatic polysulfone resin used in the present invention,  $m/(m+n)$  in the structure unit of the formula (27) is preferably 0.8 or more. Besides, it is preferred that  $m/(m+n)$  in the structure unit of the formula (28) is 0.8 or more and  $q$  is 1.

Among these, the one having the repeating unit of the formula (26) or (27) is preferred.

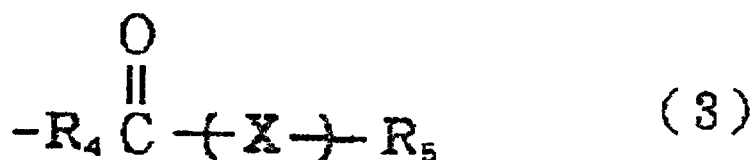
The 4-amino-2,2,6,6-tetramethylpiperidyl compound (c) having a steric hindrance type amino group as a basic skeleton is represented by the following formula (1) or (2):



24 FEB 1991

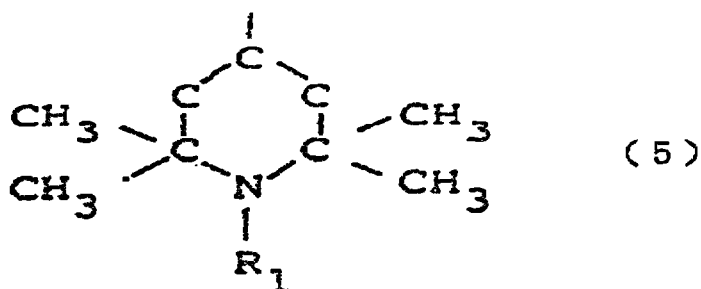
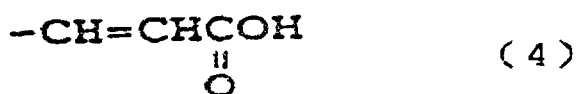


; wherein  $R_1$  represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group.  $R_2$  represents a hydrogen atom, an alkyl group or a group represented by the following formula (3),



in which X represents a direct bond, -O- or -NH-,  $R_4$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and  $R_5$  represents a hydrogen atom, an alkyl group, an aryl group or a group represented by the following formula (4) or (5),

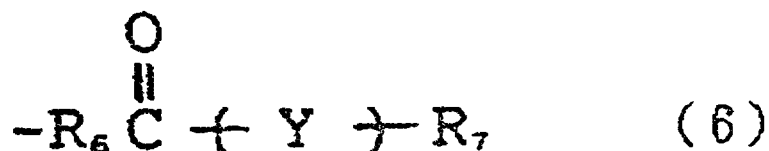
24 FEB 1997



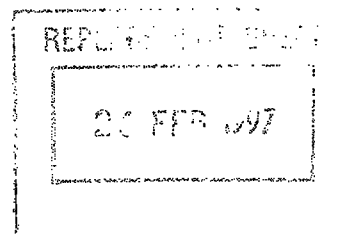
; and wherein n represents an integer of 1 to 4, p represents an integer of 2 to 6, and R<sub>1</sub> represents an n valent organic group.

Preferable examples of the 4-amino-2,2,6,6-tetramethylpiperidyl compound represented by the formula (1) or (2) are [I] to [VII] shown below.

[I] When n is 1 and R<sub>1</sub> is a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group, a benzyl group, an aryl group or a group represented by the following formula (6);



, wherein Y represents a direct bond, -O- or -NR<sub>8</sub>-, R<sub>6</sub> represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and R<sub>7</sub> and R<sub>8</sub>



each represents a hydrogen atom, a hydroxyl group, an alkyl group or an aryl group.

More preferably, when  $n$  is 1,  $R_1$  and  $R_2$  each represents a hydrogen atom or an alkyl group having 1-18 carbon atoms and  $R_3$  represents a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group or the group represented by the formula  $\text{---}\overset{\text{6}}{\text{C}}\text{---}$  in which  $\text{---}\overset{\text{Y}}{\text{C}}\text{---}$  represents a direct bond,  $\text{---O---}$  or  $\text{---NR}_2\text{---}$ ,  $R_4$  represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group or arylene group, and  $R_5$  and  $R_6$  each represent a hydrogen atom, an alkyl group having 1-18 carbon atoms, an aryl group having 6-21 carbon atoms or a group represented by the formula (4) or (5).

[II] When  $n$  is 2,  $R_7$  represents an alkylene group or a group represented by the following formula (7);



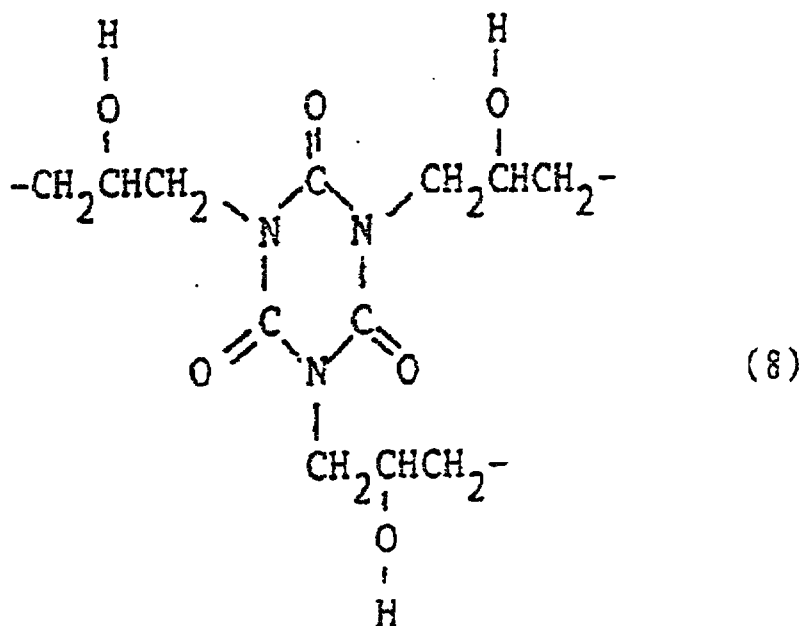
wherein  $R_9$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group.

More preferably, when  $n$  is 2,  $R_1$  and  $R_2$  each represents a hydrogen atom or an alkyl group having 1-18 carbon atoms and  $R_3$  represents an alkylene group or a group represented by the formula (7) in which  $R_9$  represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group or an arylene group.

2 FEB 1977

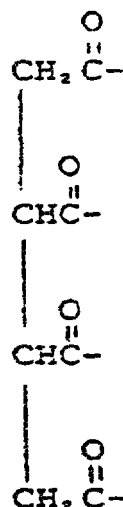
The compound in the formula (1), when n is 2, R<sub>2</sub> represents a group represented by the formula (3) in which X represents a direct bond, -O- or -NH-, R<sub>1</sub> represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, R<sub>3</sub> represents a hydrogen atom, an alkyl group, an aryl group or a group represented by the formula (4) or (5), and R<sub>4</sub> represents an alkylene group, is also more preferable.

[III] When n is 3, R<sub>3</sub> is a group represented by the following formula (8);

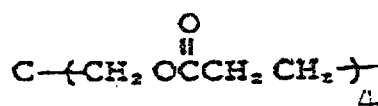


[IV] When n is 4, R<sub>3</sub> is a group represented by the following formula (9) or (10);

23 FEB 1997

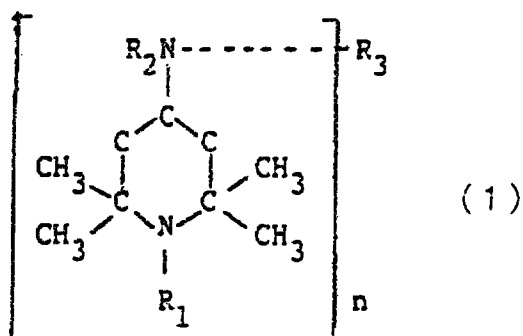


(9)



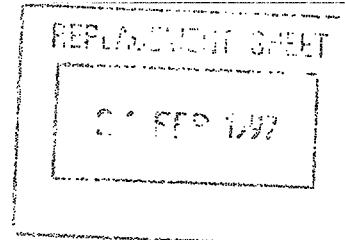
(10)

[V] Further, the compound represented by the following formula (1) is also preferable.



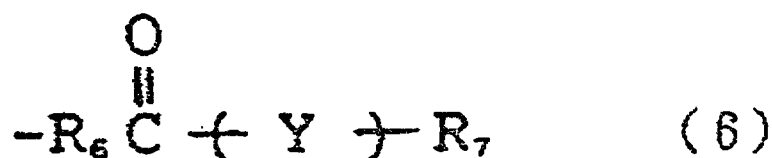
(1)

In the formula (1),  $\text{R}_1$  represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group having 1-8 carbon atoms, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group, and  $n$  is an integer of 1, 2 or 4 ; and when  $n$  is 1,  $\text{R}_2$  represents a hydrogen atom,

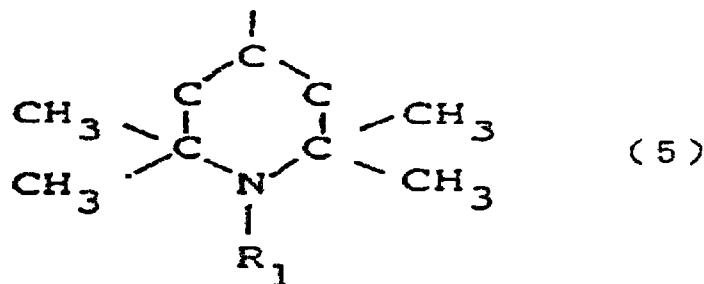
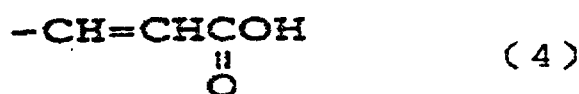


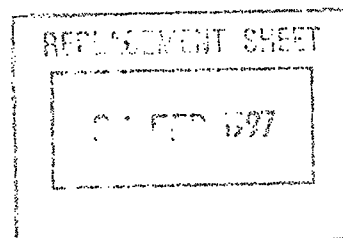
an alkyl group having 1-8 carbon atoms or  $\begin{array}{c} \text{O} \\ \parallel \\ -\text{C}-(\text{P})-(\text{Q})-\text{C}-\text{OH} \\ \parallel \\ \text{O} \end{array}$  in

which P and Q represent one selected from the group consisting of an alkylene group, an alkylidene group, a vinyl group and a vinylidene group, and R<sub>3</sub> represents a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group, a benzyl group, an aryl group or a group represented by the following formula (6);



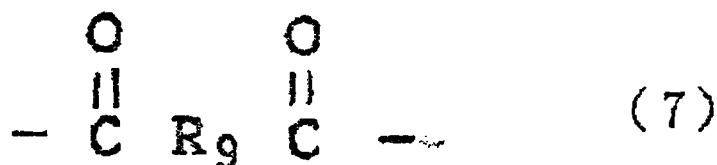
in which Y represents a direct bond, -O- -NR<sub>8</sub>-, R<sub>6</sub> represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and R<sub>7</sub> and R<sub>8</sub> each represents a hydroxyl group, an alkyl group, an aryl group or a group represented by the following formula (4) or (5),



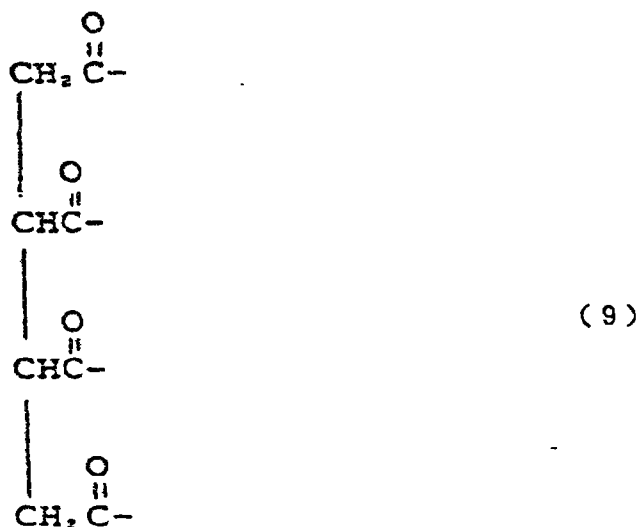


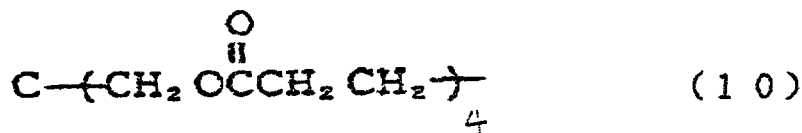
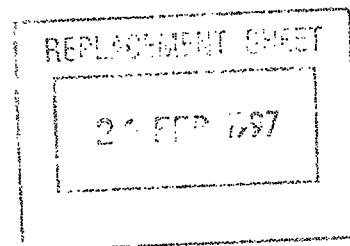
; when n is 2, R<sub>2</sub> represents a hydrogen atom, an alkyl group having 1-8 carbon atoms or  $\begin{array}{c} \text{O} \\ || \\ -\text{C}-(\text{P})-(\text{Q})-\text{C}-\text{OH} \\ || \\ \text{O} \end{array}$  in which P and Q

represent one selected from the group consisting of an alkylene group an alkylidene group, a vinyl group and a vinylylidene group, and R<sub>3</sub> represents an alkylene group or a group represented by the following formula (7),



in which R<sub>9</sub> represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group; and when n is 4, R<sub>2</sub> represents a hydrogen atom or an alkyl group having 1-8 carbon atoms, and R<sub>3</sub> represents a group represented by the following formula (9) or (10),

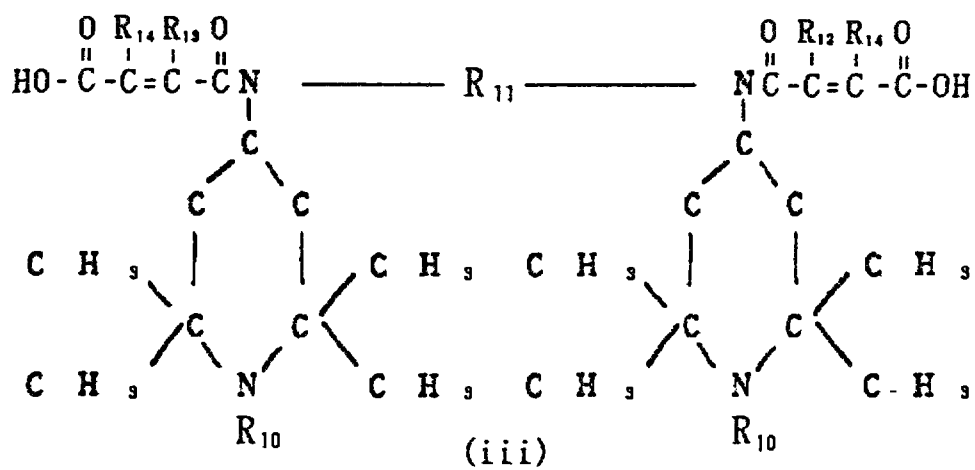
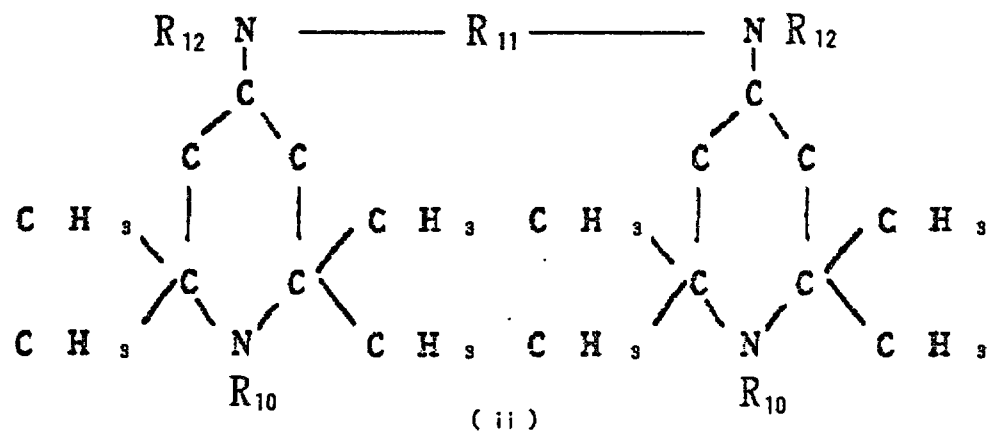




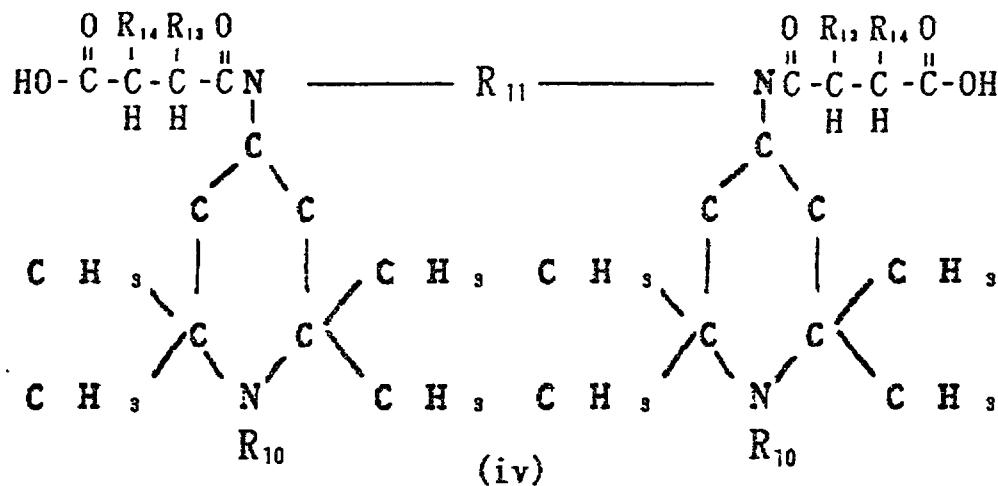
More preferably, in the formula (1),  $R_1$  and  $R_2$  each represents a hydrogen atom or an alkyl group having 1-8 carbon atoms, and (i) when  $n$  is 1,  $R_3$  is a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group or a group represented by the formula (6) in which  $Y$  represents a direct bond,  $-O-$  or  $-NR_6-$ ,  $R_6$  represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group or arylene group, and  $R_7$  and  $R_8$  each represent a hydroxyl group, an alkyl group having 1-18 carbon atoms, an aryl group having 6-21 carbon atoms or a group represented by the formula (4) or (5),

and (ii) when  $n$  is 2,  $R_3$  represents an alkylene group or a group represented by the formula (7) in which  $R_9$  represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group or an arylene group.

Further, at least one compound represented by the following formulas (i)-(iv) is also more preferable ;



23 FEB 1957



wherein  $R_{10}$  represents a hydrogen atom, an alkyl group or an alkyloxy group,  $R_{11}$  represents an alkylene group,  $R_{12}$  represents an alkyl group, and  $R_{13}$  and  $R_{14}$  each represents a hydrogen atom or a methyl group.

In the formula (i), preferably,  $R_{10}$  represents a hydrogen atom, a hydroxyl group, an alkyl group having 1-10 carbon atoms or an alkyloxy group having 1-10 carbon atoms, and  $R_{11}$  represents an alkylene group having 2-8 carbon atoms, and more preferably,  $R_{10}$  represents a hydrogen atom or a methyl group and  $R_{11}$  represents a hexamethylene group.

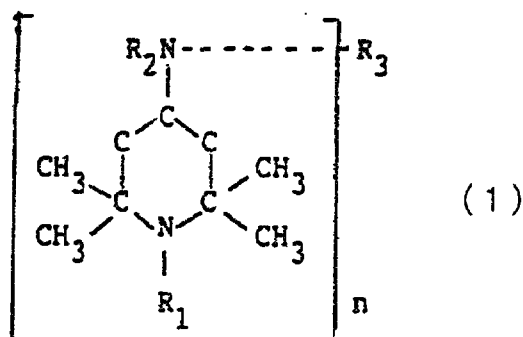
In the formula (ii), preferably,  $R_{10}$  represents a hydrogen atom, a hydroxyl group, an alkyl group having 1-10 carbon atoms or an alkyloxy group having 1-10 carbon atoms,  $R_{11}$  represents an alkylene group having 2-8 carbon atoms, and  $R_{12}$  represents an alkyl group having 1-18 carbon atoms, and more preferably,  $R_{10}$  represents a hydrogen atom or a methyl group.

In the formula (iii), preferably,  $R_{10}$  represents a hydrogen atom, a hydroxyl group, an alkyl group having 1-10 carbon atoms or an alkyloxy group having 1-10 carbon atoms, and  $R_{11}$  represents an alkylene group having 2-8 carbon atoms, and more preferably,  $R_{10}$  represents a hydrogen atom or a methyl group.

In the formula (iv), preferably,  $R_{10}$  represents a hydrogen atom, a hydroxyl group, an alkyl group having 1-10 carbon atoms or an alkyloxy group having 1-10 carbon atoms, and  $R_{11}$  represents an alkylene group having 2-8 carbon atoms, and more preferably,  $R_{10}$  represents a hydrogen atom or a methyl group.

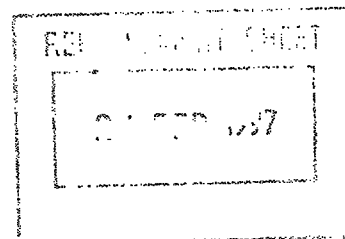
[VI] The compound represented by the following formula

(1) ;

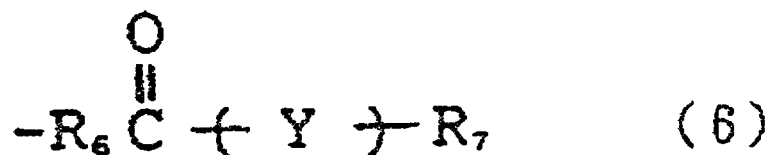


wherein  $R_1$  represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group having 1-8 carbon atoms, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group,  $R_2$  represents a hydrogen atom or alkyl group, ; and

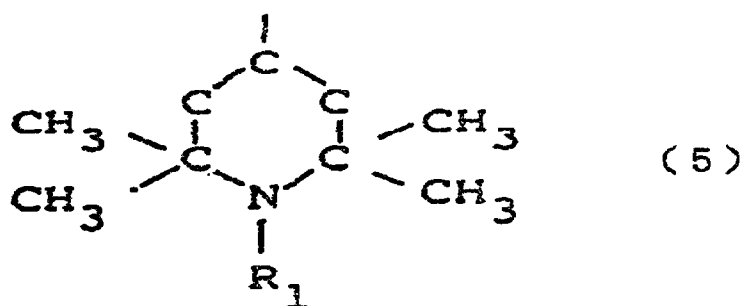
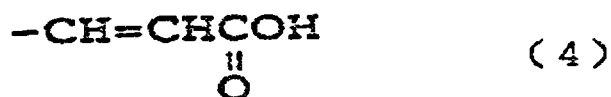
(i) when  $n$  is 1,  $R_3$  represents a hydrogen atom, an alkyl group,



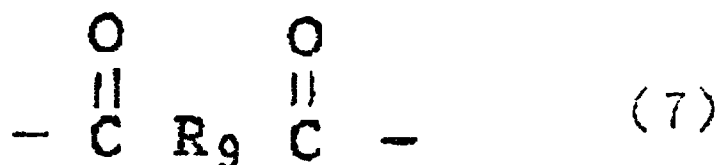
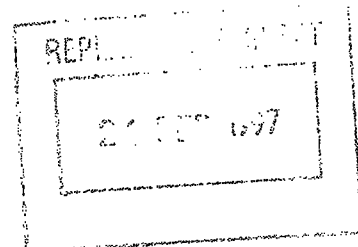
a cycloalkyl group, an alkenyl group, a benzyl group, an allyl group or a group represented by the following formula (6),



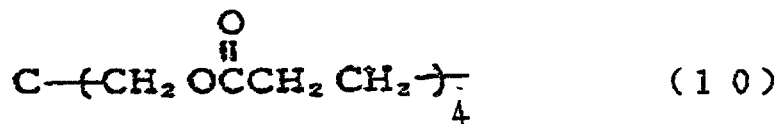
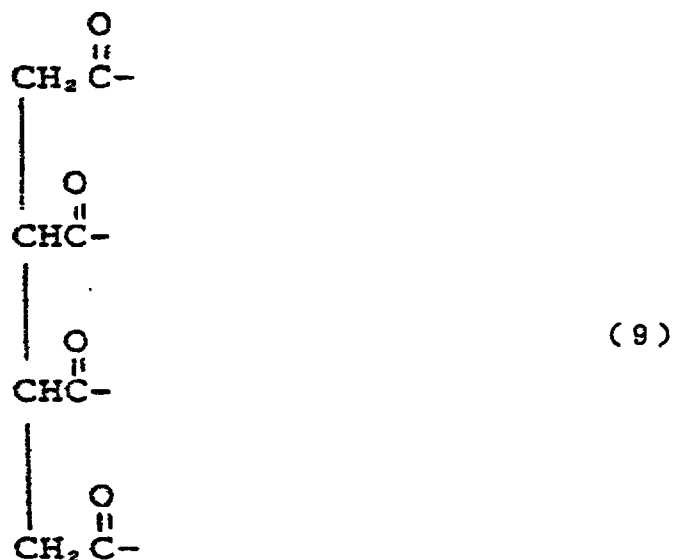
in which Y represents a direct bond, -O- -NR<sub>8</sub>-, R<sub>6</sub> represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and R<sub>7</sub> and R<sub>8</sub> each represents a hydroxyl group, an alkyl group, an aryl group or a group represented by the following formula (4) or (5),



(ii) when n is 2, R<sub>1</sub> represents a group represented by the following formula (7),



in which R<sub>9</sub> represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and (iv) when n is 4, R<sub>9</sub> represents a group represented by the following formula (9) or (10).



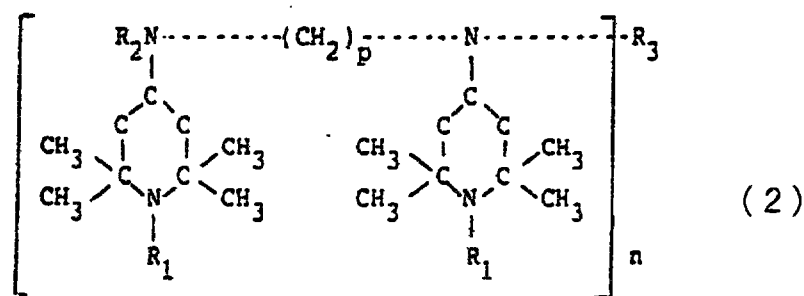
More preferably, in the formula (1), R<sub>1</sub> represents a hydrogen atom or an alkyl group having 1-8 carbon atoms, R<sub>2</sub> represents a hydrogen atom or an alkyl group having 1-18 carbon atoms ; and when n is 1, R<sub>3</sub> represents a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group or a group represented by

24 FEB 1972

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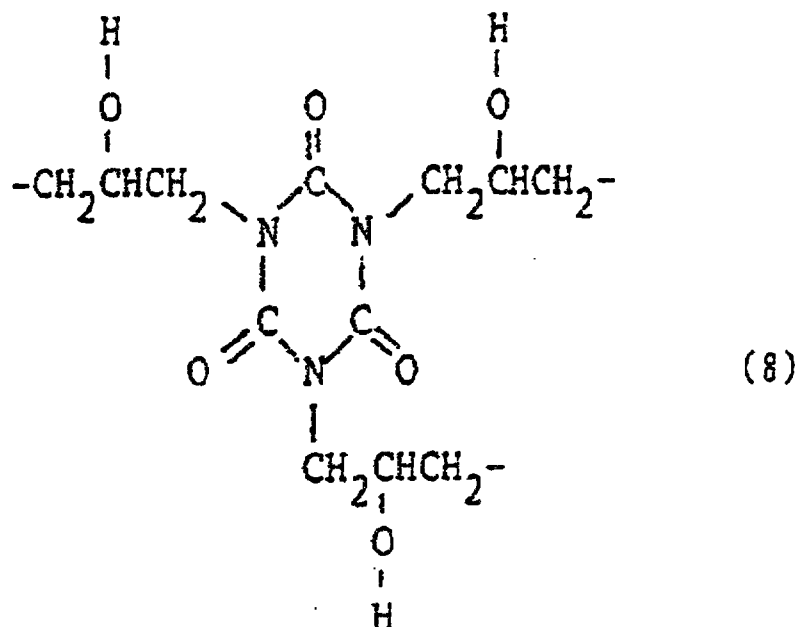
the formula (6) in which Y represents a direct bond, -O- or -NR<sub>9</sub>-, R<sub>5</sub> represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group having 1-10 carbon atoms or arylene group, and R<sub>7</sub> and R<sub>8</sub> each represent a hydrogen atom, an alkyl group having 1-18 carbon atoms, an aryl group having 6-21 carbon atoms or a group represented by the formula (4) or (5); and when n is 2, R<sub>3</sub> represents a group represented by the formula (7) in which R<sub>6</sub> represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group having 1-10 carbon atoms or an arylene group.

[VII] The compound represented by the following formula (2);



wherein R<sub>1</sub> represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group having 1-8 carbon atoms, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group, R<sub>2</sub> represents a hydrogen atom or an alkyl group having 1-8 carbon atoms, n is an integer of 3, p is an integer of 2 to 8, and R<sub>3</sub> represents a group

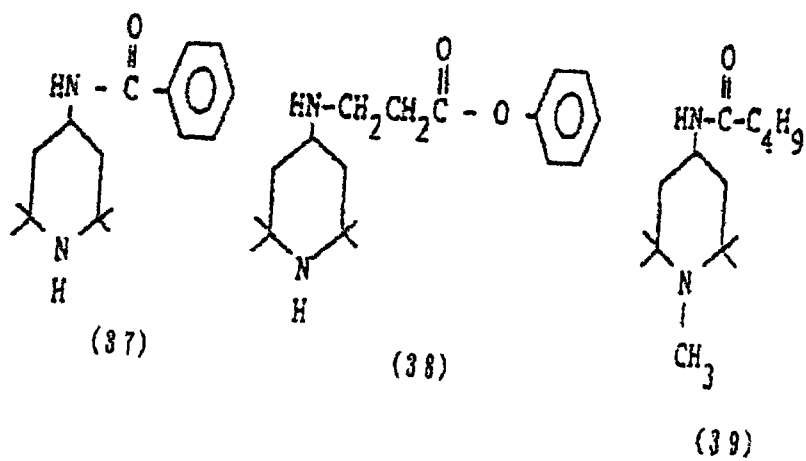
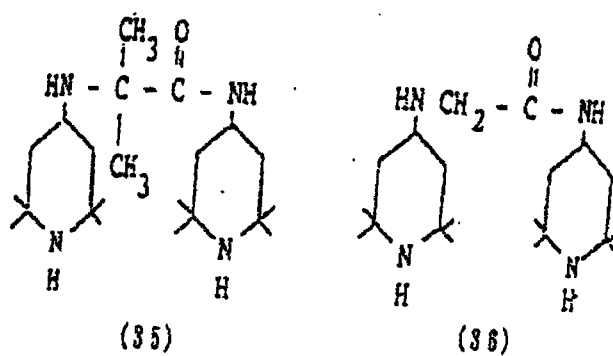
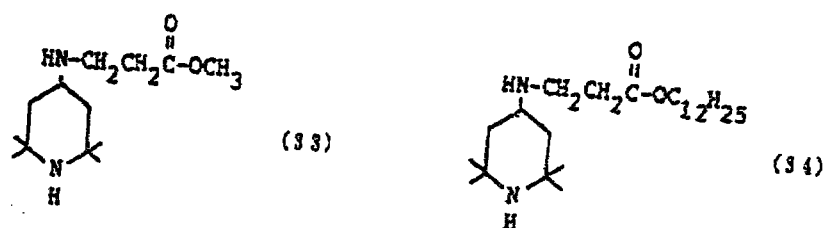
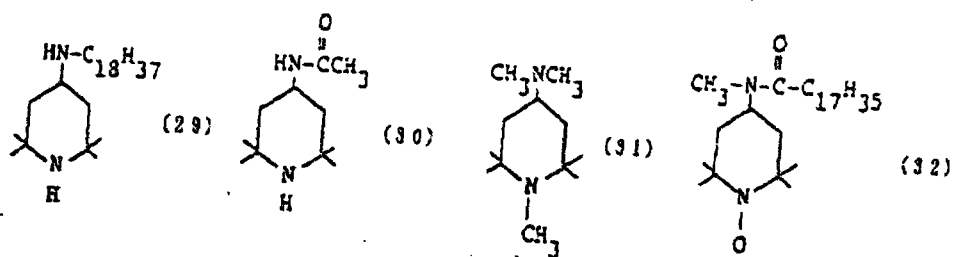
represented by the following formula (8) ;



Examples of [n=1] of [I], [V] and [VI] of the compound (c) of the present invention include 4-Amino-2,2,6,6-tetramethyl-piperidine, 4-methyl-amino-2,2,6,6-tetramethyl-4-piperidine, 4-ethylamino-2,2,6,6-tetra-methylpiperidine, 4-propylamino-2,2,6,6-tetramethylpiperidine, 4-butylamino-2,2,6,6-tetra-methylpiperidine, 4-hexylamino-2,2,6,6-tetra-methylpiperidine, 4-octylamino-2,2,6,6-tetra-methylpiperidine, 4-dodecanyl-amino-2,2,6,6-tetramethyl-piperidine, 4-diethylamino-2,2,6,6-tetramethylpiperidine, N-(2,2,6,6-tetramethyl-4-piperidyl) piperidine, N-(2,2,6,6-tetramethyl-4-piperidyl)acetamide and N-(2,2,6,6-tetramethyl-4-piperidyl)stearoylamide. Besides, mention may be made of the compounds represented by the following formulas (29) to (47).

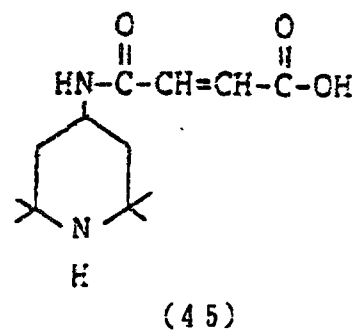
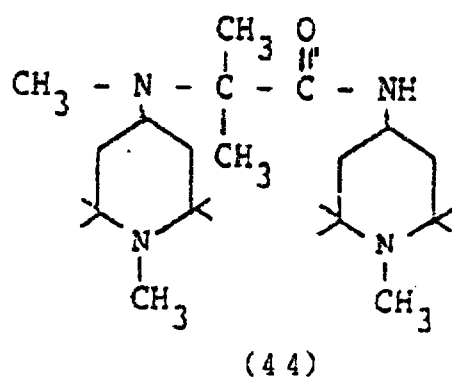
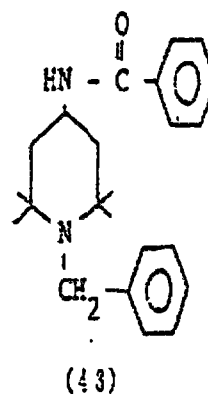
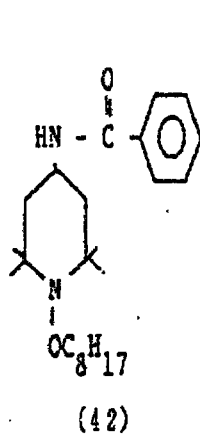
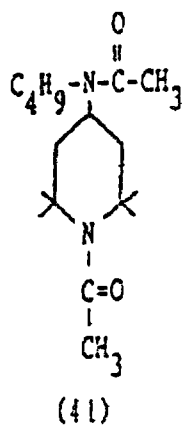
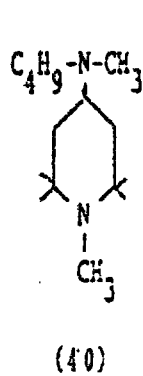
21 SEP 1997

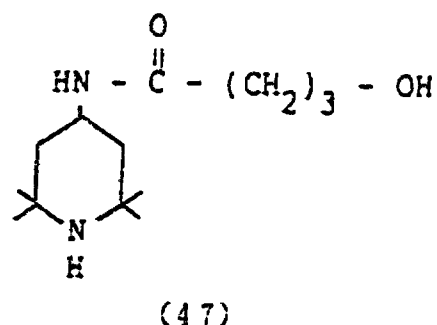
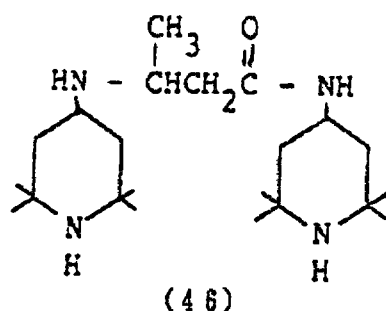
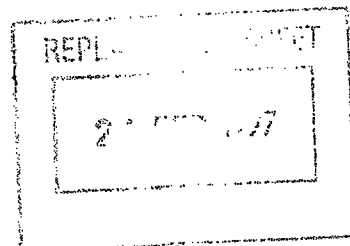
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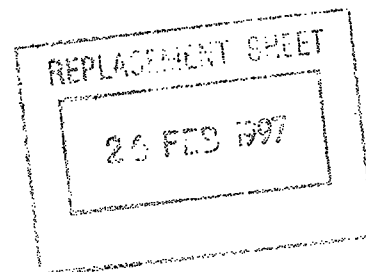


Examples of [n=2] of [II], [V] and [VI] of the compound (c) of the present invention include compounds below.

In the formula (1) or (2),  $R_1$  includes, for example, a hydrogen atom, a hydroxyl group, an alkyl group of 1-10 carbon atoms such as methyl, ethyl or propyl, or an alkyloxy group of 1-10 carbon atoms and further includes cyclic ones such as cyclohexylmethyl and cyclohexylethyl in addition to straight or branched ones when the carbon number is 3 or more.  $R_1$  is preferably a hydrogen atom or a methyl group.

$R_2$  is preferably an alkyl group having 1-18 carbon atoms or an alkyloxy group having 1-18 carbon atoms, such as methyl, ethyl, propyl, isopropyl, n-butyl, isobutyl, t-butyl, sec-butyl, n-pentyl, n-hexyl, 2-ethylhexyl, n-octyl, 1,1,3,3-tetramethylbutyl, n-decyl, n-dodecyl, n-octadodecyl, cyclohexyl, 3,3,5-trimethyl-cyclohexyl, cyclooctyl and cyclodecyl.

$R_3$  includes an alkylene group having 2-8 carbon atoms and alkylene groups represented by  $-\text{C}_n\text{H}_{2n}-$  such as ethylene, 1,2- or 1,3-propylene, 1,2-, 1,3- or 1,4-butylenes, pentamethylene, 2,2-dimethyl-1,3-propylene, hexamethylene and octamethylene,

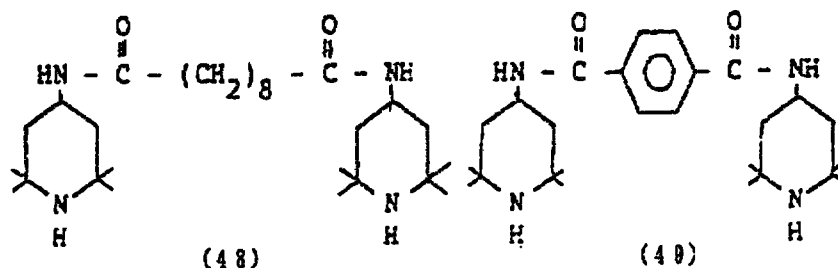


and cyclohexylene and cyclohexylenedimethylene.  $R_1$  is preferably a hexamethylene group having 6 carbon atoms.

In the formula (3),  $R_1$  is a hydrogen atom or an alkyl group having 1-18 carbon atoms such as methyl, ethyl, propyl, isopropyl, n-butyl, isobutyl, t-butyl, sec-butyl, n-pentyl, n-hexyl, 2-ethylhexyl, n-octyl, 1,1,3,3-tetramethylbutyl, n-decyl, n-dodecyl, n-octadodecyl, cyclohexyl, 3,3,5-trimethylcyclohexyl, cyclooctyl and cyclodecyl.

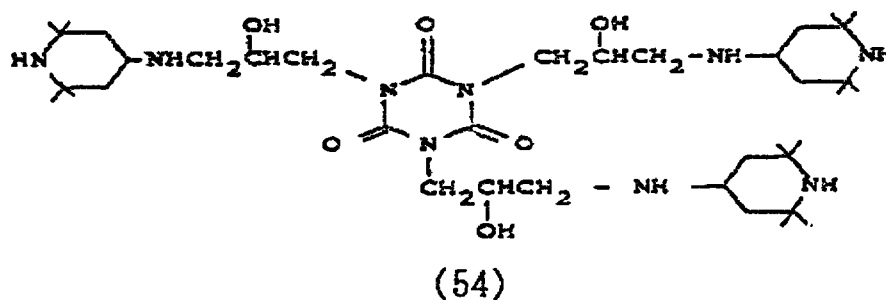
In the formula (7),  $R_2$  represents a direct bond, a straight or branched alkylene group of 1-10 carbon atoms, an alkylidene group or an arylene group such as, for example, ethylene, 1,2-, 1,3- or 1,4-propylene, 1,2-, 1,3- or 1,4-butylene, pentamethylene, 2,2-dimethyl-1,3-propylene, hexamethylene, octamethylene, cyclohexylene and cyclohexylene-dimethylene.

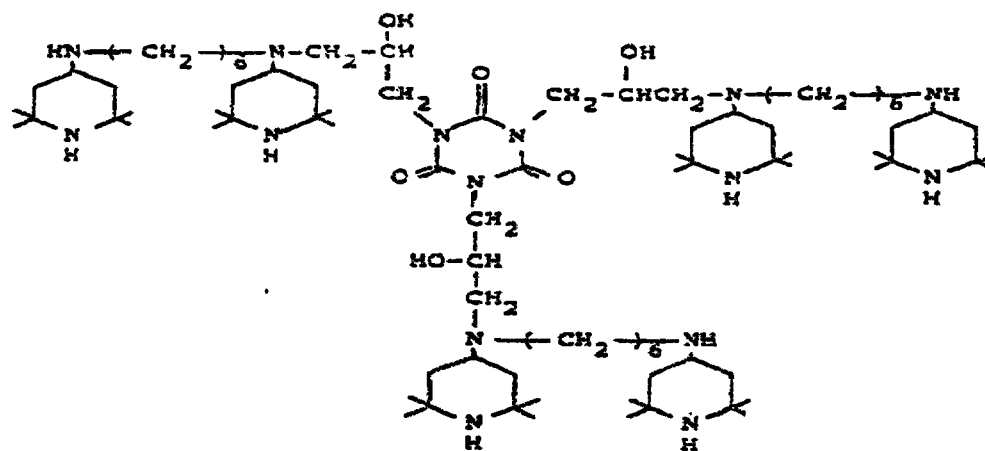
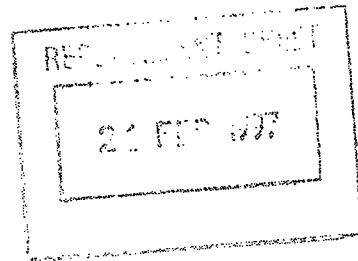
Specific examples of the compounds include N,N'-bis(2,2,6,6-tetra-methyl-4-piperidyl)-1,6-hexanediamine, N,N'-bis(2,2,6,6-tetra-methyl-4-piperidyl)ethylenediamine, N,N'-bis(2,2,6,6-tetra-methyl-4-piperidyl)-1,8-octanediamine and a compound represented by the following formulas (48) to (53).





Specific examples of the compounds include a compound represented by the following formulas (54) and (55).

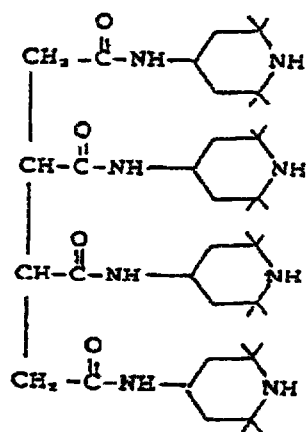




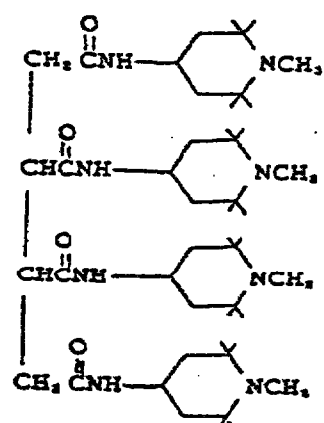
(55)

Examples of [n=4] of [I] and [IV] to [VI] of the compound (c) of the present invention include compounds below. In the formula (1) or (2), R<sub>1</sub> is the group represented by the formula (9) or (10).

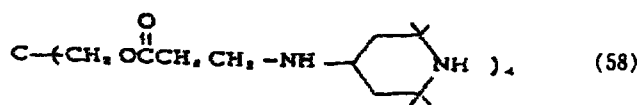
Specific examples of the compounds include a compound represented by the following formulas (56) to (58).



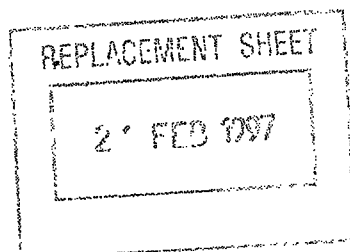
(56)



(57)



(58)



The amount ratio of the components (a) and (b) is ;

(a) 1 to 99 % by weight of a polyphenylene ether resin and  
(b) 99 to 1 % by weight of at least one non-crystalline thermoplastic resin, preferably ;

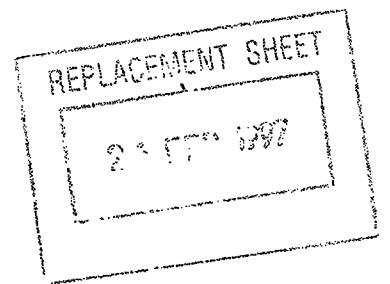
(a) 10 to 90 % by weight of a polyphenylene ether resin and  
(b) 90 to 10 % by weight of at least one non-crystalline thermoplastic resin, more preferably ;

(a) 20 to 90 % by weight of a polyphenylene ether resin and  
(b) 80 to 10 % by weight of at least one non-crystalline thermoplastic resin, and most preferably ;

(a) 40 to 85 % by weight of a polyphenylene ether resin,  
(b) 60 to 15 % by weight of at least one non-crystalline thermoplastic resin.

The amount of the component (c) is 0.001-15 parts by weight, preferably 0.1-10 parts by weight based on 100 parts by weight of the sum of (a) the polyphenylene ether resin and (b) the non-crystalline thermoplastic resin. When it is smaller than 0.001 part by weight, no effect to inhibit gelation is seen and when it is larger than 15 parts by weight, problems such as foaming occur at the time of actual processing owing to volatile matters.

In the present invention, fillers may be added in order to improve mechanical properties such as strength, stiffness and hardness to the thermoplastic resin composition.

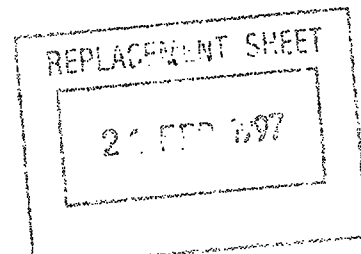


The fillers include calcium carbonate, magnesium carbonate, aluminium hydroxide, magnesium hydroxide, zinc oxide, titanium oxide, magnesium oxide, magnesium sulfate, aluminium silicate, magnesium silicate, calcium silicate, silicic acid, calcium silicate hydrate, aluminium silicate hydrate, mineral fiber, zonotrite, potassium titanate whisker, magnesium oxysulfate, glass balloon, glass fiber, glass bead, inorganic fiber such as carbon fiber, stainless fiber, aluminum fiber and aramide fiber, carbon black, inorganic fillers such as a metal whisker, silica, alumina, talc, mica, clay, kaolinite,  $TiO_2$ ,  $ZnO$  and  $Sb_2O_3$ , and one or more kind of these fillers can be blended. Besides, the fillers used in the present invention are not restricted by these.

The thermoplastic resin composition of the present invention is obtained by blending and melt kneading the components (a), (b) and (c) with conventional methods. And the components (b) and (c) may be added in polymerization process of the component (a).

The thermoplastic resin composition of the present invention can be molded by various methods such as an injection molding, an extrusion molding, a blow molding, a sheet molding and film molding, which are most suitable since the composition is highly prevented from deterioration due to heat.

Besides, a molded article provided by the present



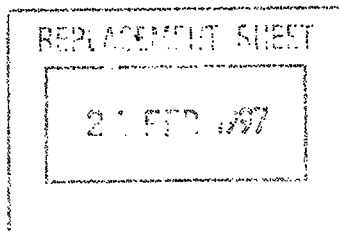
invention can be widely used as a packaging material, household appliances, an exterior material for automobiles parts and an interior decoration material such as trim parts of automobiles.

#### EXAMPLE

The Example of the present invention is illustrated in detail according to Examples and Comparative Examples as follows, but those are simple exemplifications, and the present invention is not limited to them.

Each Example and Comparative Example was obtained by blending the components shown in Tables 1 to 3. That is, those components were extruded with a twin screw kneading machine (TEM-50A manufactured by Toshiba Machine Co., Ltd.) at a cylinder temperature of 260 °C, and pelletized to obtain the composition by a strand cutter after cooling with a water basin. Thus obtained pellets were dried at 130 °C for 2 hours under vacuum drying, and molded to obtain a test piece with an injection molding machine (IS220EN manufactured by Toshiba Machine Co., Ltd.) at a cylinder temperature of 290 °C, an injection pressure of 1200kg/cm<sup>2</sup> and a mold temperature of 80 °C.

Thus obtained pellets and the test piece were evaluated by the following method, and the results were obtained.



[The molecular weight of the polyphenylene ether]

The pellets provided were dissolved in chloroform and measured with a gel permeation chromatograph (GPC) (HLC-8020 manufactured by Toso Co., Ltd.; columns of K-806, K-805, K-804 and K-803; a detector of UV having a wave length of 290 nm; a temperature of 40 °C; a flow rate of 1.0 ml/min), and the molecular weight converted with that of the standard polystyrene was determined.

[Thermal resistance stability]

After a thermal hysteresis was given to the test piece having 3.2 mm in thickness for Izod impact test, obtained by the above-mentioned injection molding, by letting alone in a constant temperature of 150 °C for 100 and 500 hours, it was notched in accordance with ASTM D256, and Izod impact test was conducted at a temperature of 23 °C.

The materials illustrated as follows were prepared in order to obtain the compositions of Examples and Comparative Examples.

(Polyphenylene ether ; PPE)

The polyphenylene ether (PPE) was obtained by homo-polymerization of 2,6- dimethylphenol and having a logarithmic viscosity of 0.46 in chloroform solution (0.5g/dl) at 30°C. The molecular weight (Mw) of PPE before melt-kneaded was 46,100.

(Polycarbonate ; PC)

The trade name of CALIBRE 300-10 manufactured by

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Sumitomo-Dow Co., Ltd., was used.

(Polymethyl methacrylate ; PMMA)

The trade name of Sumipex LG manufactured by Sumitomo Chemical Co., Ltd.

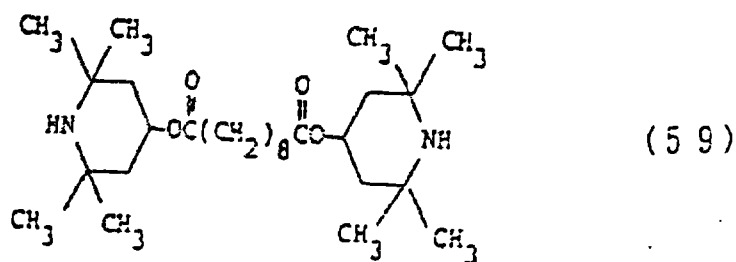
(Polystyrene)

Polystyrene 1 (PS-1) : The trade name of Sumibright E183N manufactured by Sumitomo Chemical Co., Ltd.

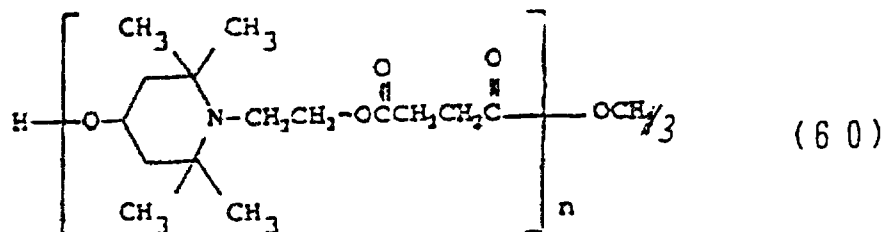
Polystyrene 2 (PS-2) : The trade name of Sumibright UG3000 manufactured by Sumitomo Chemical Co., Ltd.

(Additive)

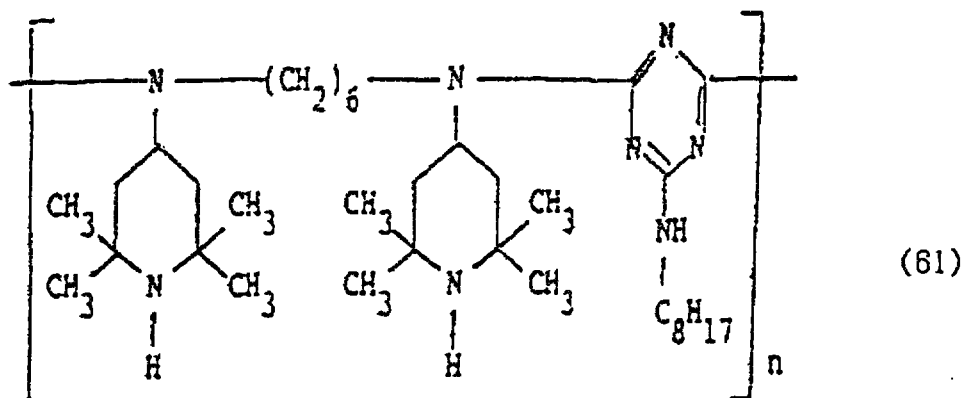
Additive 1 : The trade name of Sanol LS-770 manufactured by Sankyo Co., Ltd. It is represented by the following chemical formula (59).



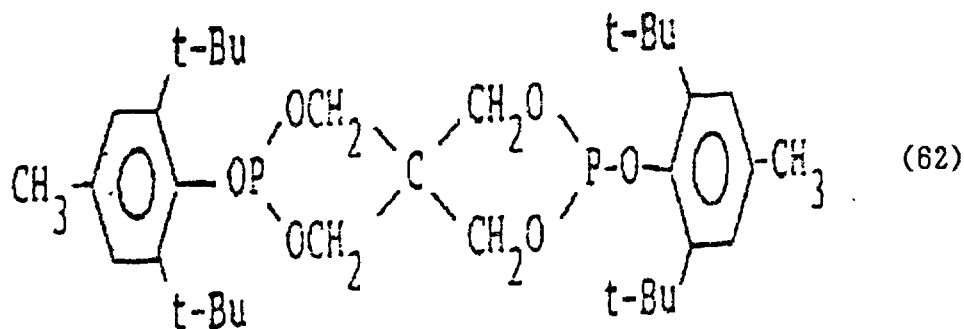
Additive 2 : The trade name of Tinuvin 622 manufactured by Ciba-Geigy Corp. It is represented by the following chemical formula (60).

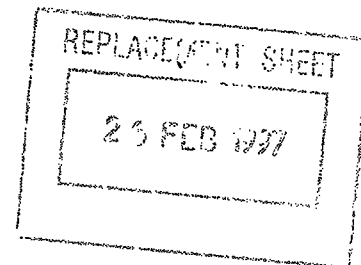


Additive 3 : The trade name of Chimasorb 944 manufactured by Ciba-Geigy Corp. It is represented by the following chemical formula (61).



Additive 4 : The trade name of Adekastab PEP-36 manufactured by Asahi Denka Kogyo K.K. It is represented by the following chemical formula (62).





The compounds used in Examples are prepared by the Reference Examples shown below. Besides, % in those Reference Examples is by weight unless otherwise notified.

#### Reference Example 1

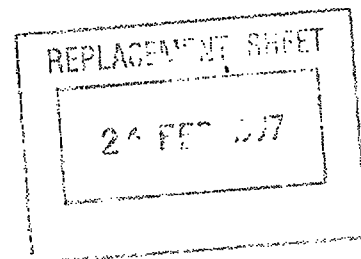
Preparation of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 1)

In a 5 liter autoclave were charged 981 g (6.32 moles) of 2,2,6,6-tetramethyl-4-piperidine, 350 g (3.01 moles) of hexamethylenediamine, 1500 g of methanol and 5.0 g of carbon containing 5 % platinum thereon with keeping the temperature of at 45 to 55°C. The inside of the autoclave was pressurized to 40 bars with hydrogen and heated to 70-80 °C to carry out hydrogenation. The hydrogenation was completed after 4 to 5 hours. The catalyst was removed by filtration at 70 to 80 °C under 2 to 3 bars, and 1070 g of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 1) was obtained by distillation. Yield : 90 %. Boiling point : 188 to 190 °C/0.80 mbar.

#### Reference Example 2

Preparation of N,N'-bis( 2-methoxycarbonyl ethyl )-N,N'-bis(2, 2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 2)

In a 1 liter four-necked flask were charged 80 g (0.20 moles) of N,N'-bis(2, 2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 1) obtained in Reference Example 1 and 200 g of methanol, and they were completely dissolved.

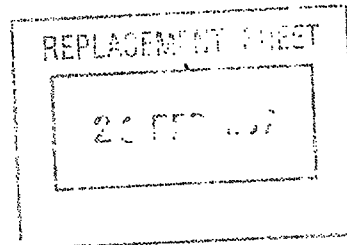


Thereto was added dropwise of a solution prepared by dissolving 35 g (0.41 moles) of methyl acrylate in 100 g of methanol at room temperature. Then, reaction was allowed to proceed for 12 hours under reflux. After completion of the reaction, the solvent was distilled off and the residue was recrystallized from hexane to obtain 98 g (0.17 mol) of the Compound 2 as a white solid. Yield: 85 %. Melting point: 50-52 °C.

#### Reference Example 3

Preparation of N,N'-bis(2-octadecyloxycarbonylethyl)-N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 3)

In a four-necked flask provided with a stirrer and a reflux condenser were charged 20 g (35 mmols) of N,N'-bis(2-methoxycarbonylethyl)-N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 2) obtained in Reference Example 2, 21 g (77 mmols) of stearyl alcohol and 200 ml of toluene, followed by stirring under reflux. Thereto was added dropwise of a solution of 0.1 g (4 mmols) of lithium amide in 3 g of methanol. Thereafter, the solvent was distilled off with addition of toluene and the reaction was completed. After the reaction was allowed to proceed for 4 hours, toluene was added, followed by cooling to room temperature. The organic layer was washed with water three times and then concentrated to obtain 23 g (22 mmol) of the Compound 3. Yield: 63 %. Melting



point : 45-46 °C.

#### Reference Example 4

Preparation of N,N'-bis( 2-t-butoxycarbonylethyl)-N,N'-bis(2, 2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 4)

The procedure of Reference Example 3 was repeated except that t-butyl alcohol was used instead of stearyl alcohol, thereby obtaining 13 g of the Compound 4. Melting point : 72 to 73 °C.

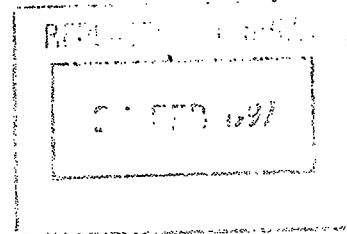
#### Reference Example 5

Preparation of N,N'-diacetyl-N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 5)

In a 300 ml four-necked flask were charged 20.7 g (50.7 mmoles) of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 1) obtained in Reference Example 1 and 70 g of pyridine, and they were completely dissolved. Thereto was added dropwise of 11.64 g (0.114 moles) of acetic anhydride over a period of 30 minutes, followed by 1 hour at 40 °C. After cooling, 100 g 20 % aqueous NaOH and 50 ml of toluene were added and the product was extracted and washed with 50 ml of water. The solvent in the toluene layer was distilled off and the residue was recrystallized from 20 ml of n-hexane to obtain 20.6 g of the Compound 5. Yield : 85 %. Melting point : 152-154 °C.

#### Reference Example 6

Preparation of N,N'-bis(n-hexylcarbonyl)-N,N'-bis(2,2,6,6-



tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 6)

The procedure of Reference Example 5 was repeated except that heptanoic anhydride was used in place of acetic anhydride, thereby obtaining 18.8 g of the Compound 6. Melting point : 123-125 °C.

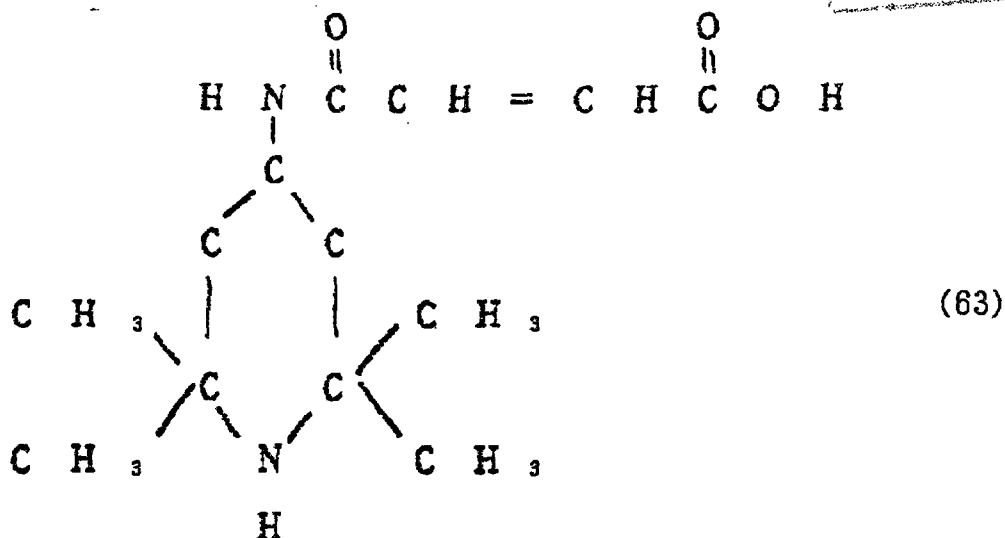
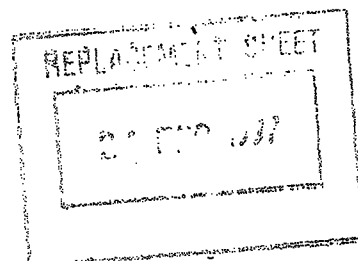
#### Reference Example 7

Preparation of N,N'-bis(ethoxycarbonyl)-N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 7)

At a temperature of 0 °C or lower, 22.8 g (0.21 moles) of ethyl chlorocarbonate was added to solution of 39.4 g (0.1 moles) of N,N'-bis(2,2,6,6-tetramethyl-4-piperidyl)-1,6-hexanediamine (Compound 1) obtained in Reference Example 1 in 200 ml of 1,2-dichloroethane which was cooled to -10 °C. Then, thereto was added slowly 8.4 g of sodium hydroxide dissolved in 50 ml of water with keeping the temperature of at 0 °C. Then, the temperature was elevated to 20 °C, and the reactant was separated to a water layer and an organic layer, and the organic layer was washed with water and dried over anhydrous sodium sulfate to remove the solvent. Then, the residue was crystallized from octane to obtain 44.1 g of the Compound 7. Yield : 82 %. Melting point : 125 °C.

#### Reference Example 8

Preparation of Compound 8 [the chemical formula (63)]



In a four-necked flask provided with a thermometer and a stirrer were charged 156 g of 4-amino-2,2,6,6-tetramethylpiperidine, 400 ml of toluene and 98g of maleic anhydride and the atmosphere in the flask was replaced with nitrogen, followed by keeping the temperature of at 60 °C for 6 hours with stirring. After completion of the reaction, the produced white crystal was filtrated and washed with toluene to obtain 243 g of the Compound 8.

#### Reference Example 9

The 2,2,6,6-tetramethyl-4-piperidylamine was used as Compound 9.

#### Reference Example 10

Preparation of methyl 2,2,6,6-tetramethyl-4-piperidyl-amino-propionate (Compound 10)

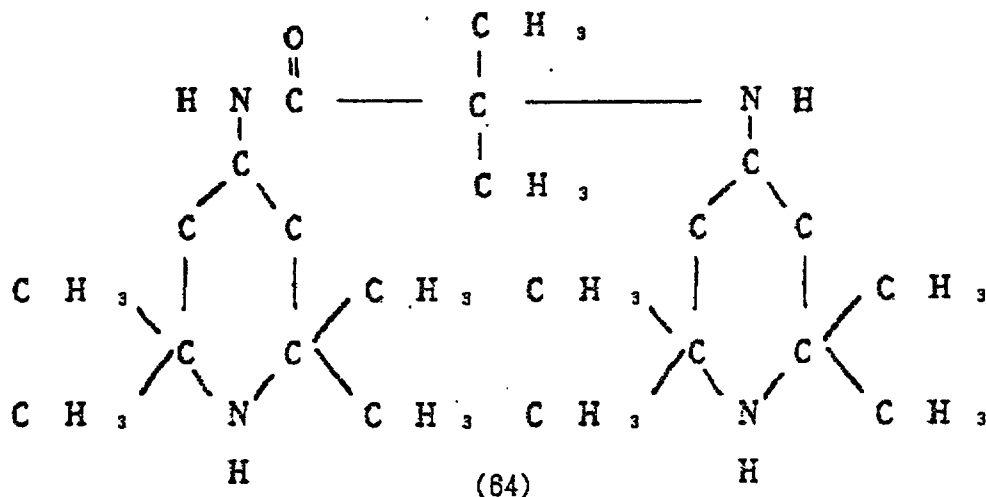
The 7.8 g (0.05 mol) of 2,2,6,6-tetramethyl-4-piperidine and 5.6 g (0.065 mol) of methyl acrylate were allowed to react

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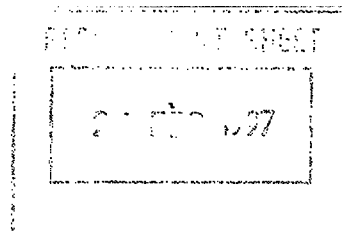
in 50 g of methanol under reflux for 3 hours. The solvent was distilled to obtain 8.0 g of colorless liquid of methyl 2,2,6,6-tetramethyl-4-piperidyl-aminopropionate (Compound 10) as a fraction of 130-136 °C/1.5mmHg.

#### Reference Example 11

Preparation of Compound 11 [the chemical formula (64)]



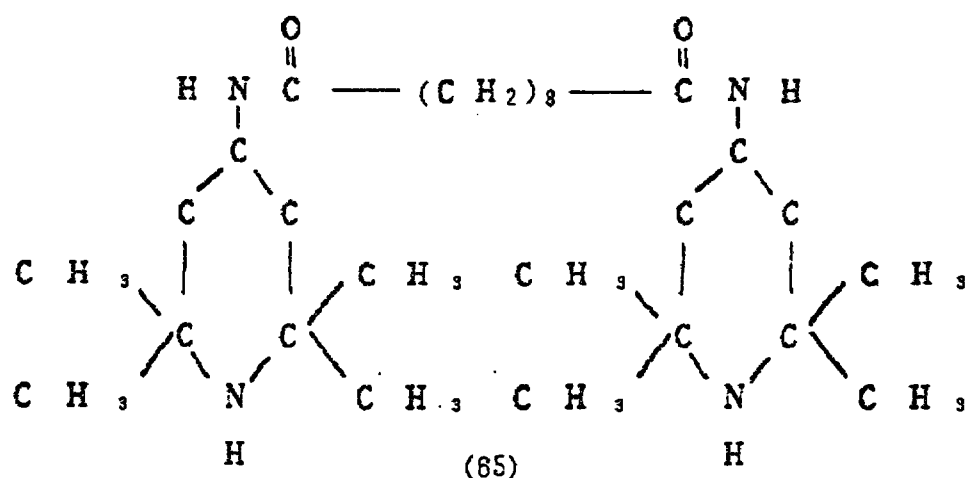
In a four-necked flask provided with a thermo-meter and a stirrer were charged 156 g of 4-amino-2,2,6,6-tetramethyl-piperidine, 32 g of acetone, 239 g of chloroform and 0.4 g of benzyltrimethylammonium chloride, and cooled to 5 °C with stirring. Then, thereto was added dropwise of 257.6 g of a 50 % aqueous potassium hydroxide solution over a period of 1 hour with keeping the temperature of at 5 to 10 °C, and thereafter, the reaction was allowed to proceed for 5 hours at 0 to 10 °C. After completion of the reaction, the aqueous layer was separated to a water layer and an organic layer, and the water



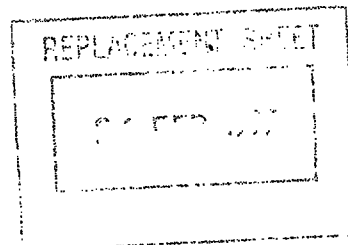
layer was removed. Then, the excess chloroform in the organic layer was distilled off to obtain 176 g of a white crystal as Compound 11. Yield : 92.6 %. Melting point : 127-128 °C. A parent ion peak of 380 was confirmed by FD-mass spectrometry.

#### Reference Example 12

Preparation of Compound 12 [the chemical formula(65)]



In a four-necked flask provided with a thermo-meter and a stirrer were charged 156 g of 4-amino-2,2,6,6-tetramethyl-piperidine, 400 ml of toluene and 121 g of triethylamine, and the atmosphere in the flask was replaced with nitrogen. After the content was cooled to 0 °C, thereto was added dropwise of 119 g of sebacoyl chloride under stirring with keeping the temperature of at 0 to 5 °C. After completion of the addition, 200 ml of 20% aqueous sodium hydroxide solution was added to dissolve the hydrochloride of triethylamine produced by the reaction. After the aqueous was separated to a water layer and an organic layer, the water layer was removed, and the



organic layer was washed with 100 ml of water. The organic layer was concentrated and the residue was crystallized from n-hexane to obtain 203 g of a white crystal as Compound 12.

#### Reference Example 13

Preparation of tris[2-hydroxy-3-(2,2,6,6-tetramethyl-4-piperidylamino)-propyl]-isocyanurate (Compound 13)

Tris(2,3-epoxypropyl)isocyanurate (29.7 g) and 4-amino-2,2,6,6-tetramethylpiperidine (47.5 g) were slowly heated in a hot bath under stirring. Heat was generated at about 80 °C. The temperature was raised up to  $100 \pm 5$  °C and the temperature was held for about two hours. After cooling, white powders of tris[2-hydroxy-3-(2,2,6,6-tetramethyl-4-piperidyl-amino)-propyl]-isocyanurate (74.5 g) (Compound 13) was obtained. Yield : 97.4 %. Melting point : 55-60 °C.

#### Examples 1 to 15

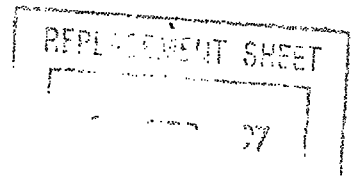
The blend ratios of Examples and the molecular weights of the polyphenylene ether in the obtained compositions (after melt-kneaded) were shown in Table 1.

#### Comparative Examples 1 to 7

The compositions without the additives and Comparative Examples using the additives 1 to 4 were shown in Table 2.

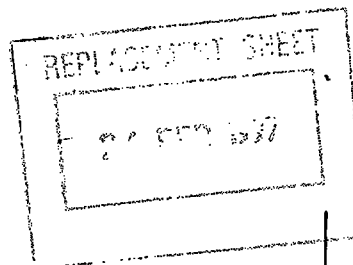
#### Examples 16, 17 and Comparative Example 8

Thermal resistance stability of the compositions are shown in Table 3.



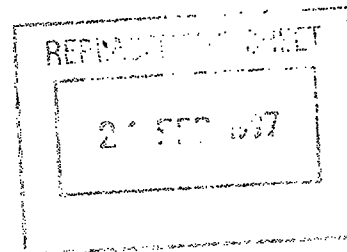
EFFECT OF THE PRESENT INVENTION

Deterioration of polyphenylene ether caused by the heat at the time of processing can be markedly improved by adding a specific hindered piperidine compound to the composition comprising the polyphenylene ether and the non-crystalline thermoplastic resin.



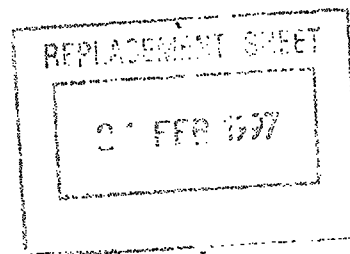
[Table 1]

|            |     | Composition |             | Molecular weight of<br>polyphenylene ether Mw |        |
|------------|-----|-------------|-------------|-----------------------------------------------|--------|
| Example    | PPE | Polystyrene | Compound    |                                               |        |
|            |     |             |             |                                               |        |
| Example 1  | 70  | 30          | Compound 1  | 1                                             | 61,000 |
| Example 2  | 70  | 30          | Compound 2  | 1                                             | 51,000 |
| Example 3  | 70  | 30          | Compound 3  | 1                                             | 62,000 |
| Example 4  | 70  | 30          | Compound 4  | 1                                             | 65,000 |
| Example 5  | 70  | 30          | Compound 5  | 1                                             | 68,000 |
| Example 6  | 70  | 30          | Compound 6  | 1                                             | 68,000 |
| Example 7  | 70  | 30          | Compound 7  | 1                                             | 52,000 |
| Example 8  | 70  | 30          | Compound 8  | 1                                             | 64,000 |
| Example 9  | 70  | 30          | Compound 9  | 1                                             | 62,000 |
| Example 10 | 70  | 30          | Compound 10 | 1                                             | 68,000 |
| Example 11 | 70  | 30          | Compound 11 | 1                                             | 70,000 |
| Example 12 | 70  | 30          | Compound 12 | 1                                             | 69,000 |
| Example 13 | 70  | 30          | Compound 13 | 1                                             | 53,000 |
| Example 14 | 70  | 30          | Compound 1  | 1                                             | 65,000 |
| Example 15 | 70  | 30          | Compound 2  | 1                                             | 58,000 |



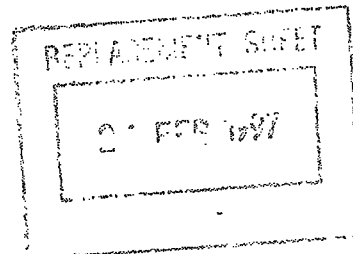
[Table 2]

|                       |        | Composition      |              | Molecular weight of polyphenylene ether Mw |  |
|-----------------------|--------|------------------|--------------|--------------------------------------------|--|
| Comparative Example 1 | PPE 70 | Polystyrene 1 30 |              | 76,000                                     |  |
| Comparative Example 2 | PPE 70 | Polystyrene 1 30 | Additive 1 1 | 77,000                                     |  |
| Comparative Example 3 | PPE 70 | Polystyrene 1 30 | Additive 2 1 | 79,000                                     |  |
| Comparative Example 4 | PPE 70 | Polystyrene 1 30 | Additive 3 1 | 75,000                                     |  |
| Comparative Example 5 | PPE 70 | Polystyrene 1 30 | Additive 4 1 | 77,000                                     |  |
| Comparative Example 6 | PPE 70 | Polycarbonate 30 |              | 82,000                                     |  |
| Comparative Example 7 | PPE 70 | PMMA 30          |              | 75,000                                     |  |



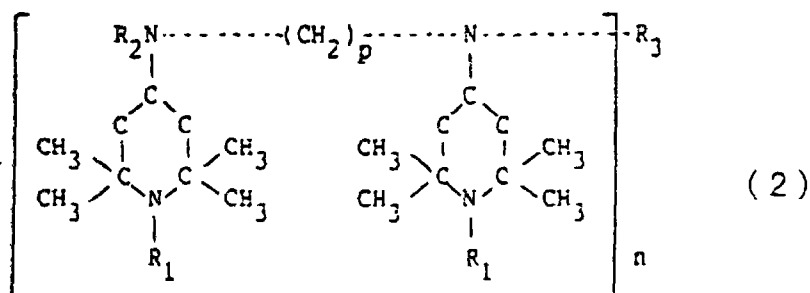
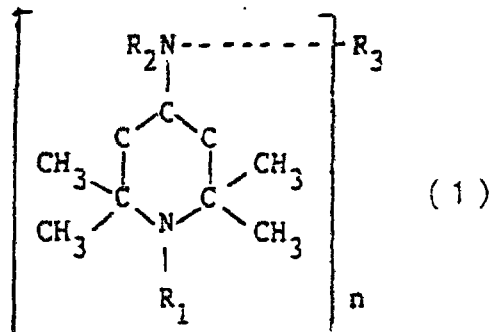
[Table 3]

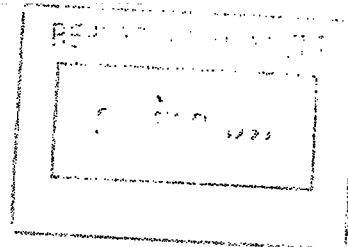
| Izod impact strength with a notch at 23 °C |                               |         |           |
|--------------------------------------------|-------------------------------|---------|-----------|
| aging at 150 °C for                        |                               |         |           |
|                                            |                               | Initial | 500 hours |
| Comparative                                | Component                     |         |           |
| Example 8                                  | PPE 60<br>Polystyrene 2<br>40 | 7.2     | 2.3       |
| Example 16                                 | PPE 60<br>Polystyrene 2<br>40 | 10.1    | 5.7       |
| Example 17                                 | PPE 70<br>Polystyrene 2<br>30 | 8.0     | 4.8       |



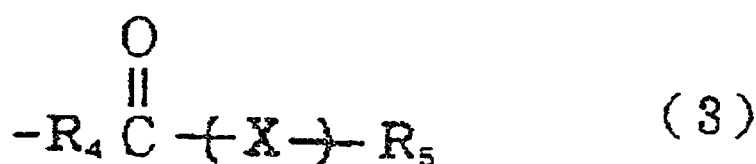
WHAT IS CLAIMED IS :

1. A thermoplastic resin composition which comprises
  - (a) 1 to 99 % by weight of a polyphenylene ether resin,
  - (b) 99 to 1 % by weight of at least one non-crystalline thermoplastic resin selected from an alkenyl aromatic resin, a non-crystalline aromatic polyester resin, a methacrylate resin, a polyether imide resin and an aromatic polysulfone resin, and per 100 parts by weight of the sum of (a) and (b),
  - (c) 0.001 to 15 parts by weight of at least one 4-amino-2,2,6,6-tetramethylpiperidyl compound represented by the following formula (1) or (2):

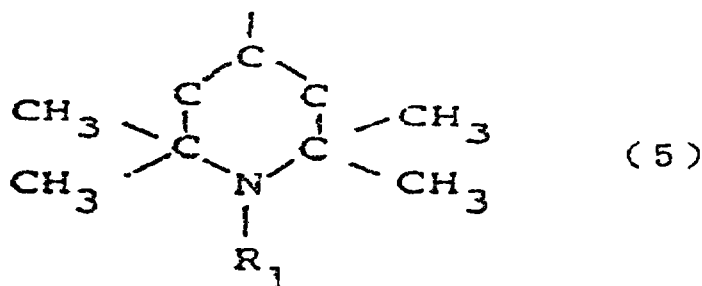
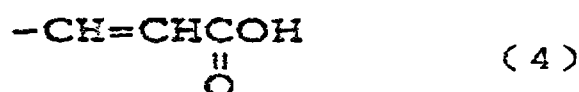




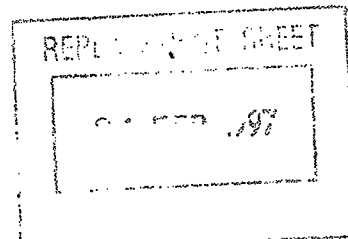
; wherein  $R_1$  represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group.  $R_2$  represents a hydrogen atom, an alkyl group or a group represented by the following formula (3),



in which X represents a direct bond, -O- or -NH-,  $R_4$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and  $R_5$  represents a hydrogen atom, an alkyl group, an aryl group or a group represented by the following formula (4) or (5),

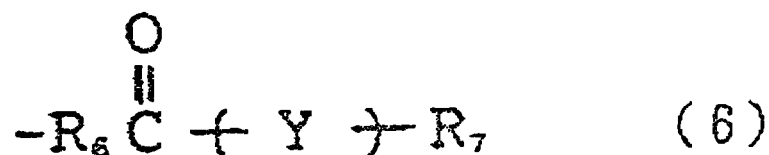


; and wherein n represents an integer of 1 to 4, p represents



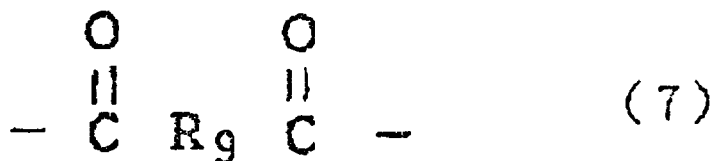
an integer of 2 to 6, and  $R_1$  represents an n valent organic group.

2. A thermoplastic resin composition according to claim 1, wherein in the formula (1) or (2), n is 1 and  $R_1$  is a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group, a benzyl group, an aryl group or a group represented by the following formula (6);



, wherein Y represents a direct bond, -O- or -NR<sub>3</sub>-,  $R_6$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and  $R_7$  and  $R_8$  each represents a hydrogen atom, a hydroxyl group, an alkyl group or an aryl group.

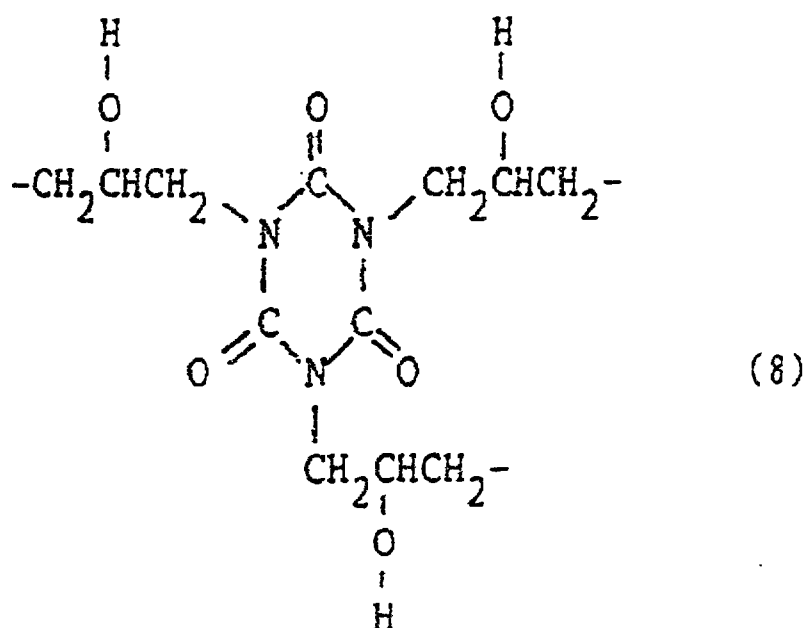
3. A thermoplastic resin composition according to claim 1, wherein in the formula (1) or (2), n is 2 and  $R_1$  is an alkylene group or a group represented by the following formula (7);



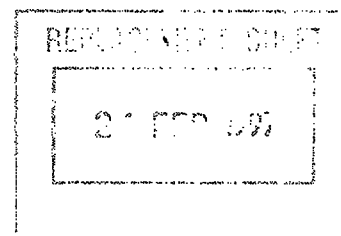


, wherein  $R_1$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group.

4. A thermoplastic resin composition according to claim 1, wherein in the formula (1) or (2),  $n$  is 3 and  $R_1$  is a group represented by the following formula (8);



5. A thermoplastic resin composition according to claim 1, wherein in the formula (1) or (2),  $n$  is 4 and  $R_1$  is a group represented by the following formula (9) or (10);



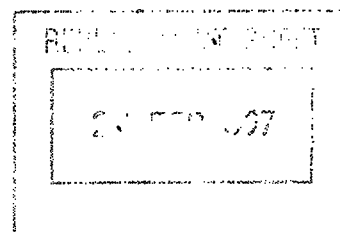
(9)



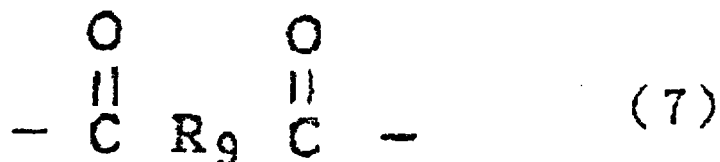
(10)

6. A thermoplastic resin composition according to claim 1, wherein in the formula (1),  $R_1$  and  $R_2$  each represents a hydrogen atom or an alkyl group having 1-18 carbon atoms and  $n$  is 1 or 2, and

(I) when  $n$  is 1,  $R_1$  is a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group or the group represented by the formula (6) in which  $\text{Y}$  represents a direct bond,  $-\text{O}-$  or  $-\text{NR}_3-$ ,  $R_3$  represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group or arylene group, and  $R_4$  and  $R_5$  each represent a hydrogen atom, an alkyl group having 1-18 carbon atoms, an aryl group having 6-21 carbon atoms or a group represented by the formula (4) or (5), and (II) when  $n$  is 2,  $R_1$  is a group represented by the



following formula (7);



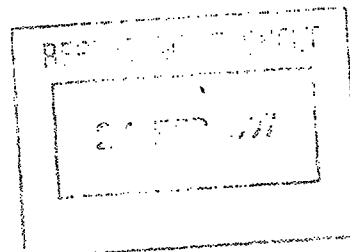
, wherein  $\text{R}_9$  represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group or an arylene group.

7. A thermoplastic resin composition according to claim 1, wherein in the formula (1),  $n$  is 2 and  $\text{R}_1$  is an alkylene group.

8. A thermoplastic resin composition according to claim 1, wherein in the formula (1),  $n$  is 2,  $\text{R}_2$  is a hydrogen atom and  $\text{R}_3$  is an alkylene group.

9. A thermoplastic resin composition according to claim 1, wherein in the formula (1),  $n$  is 2,  $\text{R}_1$  is a hydrogen atom or a methyl group,  $\text{R}_2$  is a hydrogen atom and  $\text{R}_3$  is a hexamethylene group.

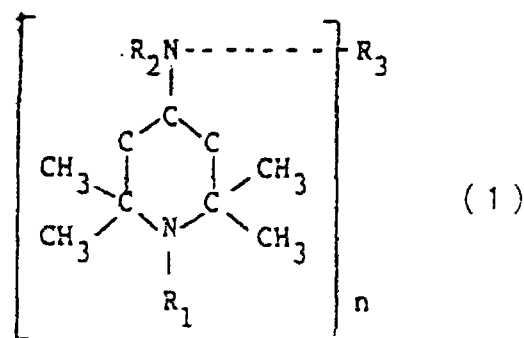
10. A thermoplastic resin composition according to claim 1, wherein in the formula (1),  $n$  is 2,  $\text{R}_2$  is an alkyl group and  $\text{R}_3$  is an alkylene group.



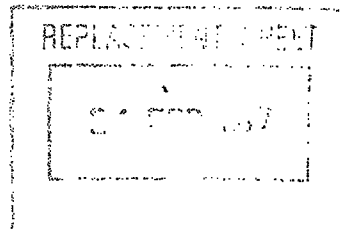
11. A thermoplastic resin composition according to claim 1, wherein in the formula (1), n is 2, R<sub>2</sub> is a group represented by the formula (3) in which X represents a direct bond, -O- or -NH-, R<sub>4</sub> represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, R<sub>5</sub> represents a hydrogen atom, an alkyl group, an aryl group or a group represented by the formula (4) or (5), and R<sub>3</sub> is an alkylene group.

12. A thermoplastic resin composition according to claim 1, wherein the non-crystalline thermoplastic resin (b) is an alkenyl aromatic resin.

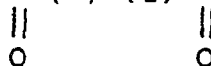
13. A thermoplastic resin composition according to claim 1, wherein the 4-amino-2,2,6,6-tetramethylpiperidyl compound (c) is a compound represented by the following formula (1) :



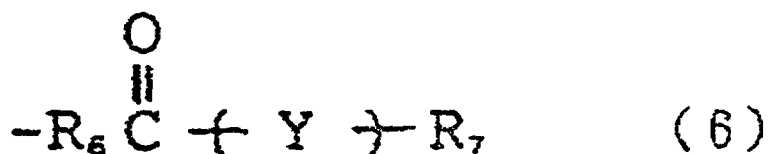
wherein R<sub>1</sub> represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group having 1-8 carbon atoms, a cycloalkyl group, an allyl group, a benzyl group, an



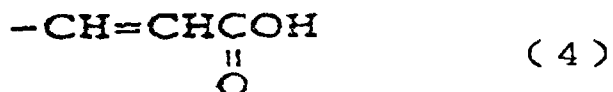
aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group, and n is an integer of 1, 2 or 4 ; and when n is 1, R<sub>1</sub> represents a hydrogen atom, an alkyl group having 1-8 carbon atoms or -C-(P)-(Q)-C-OH in which P

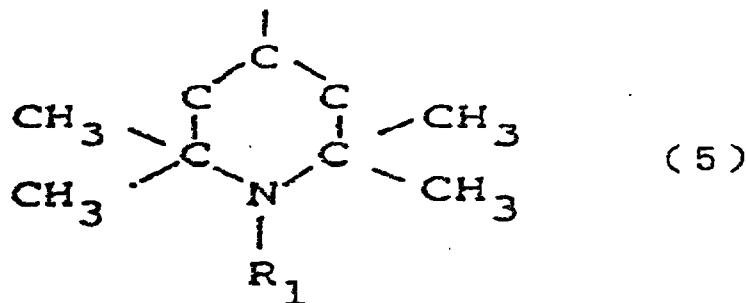
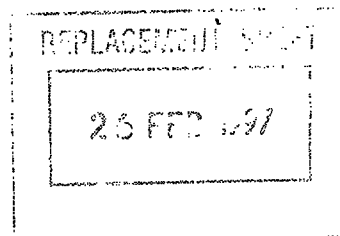


and Q represent one selected from the group consisting of an alkylene group, an alkylidene group, a vinyl group and a vinylidene group, and R<sub>1</sub> represents a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group, a benzyl group, an aryl group or a group represented by the following formula (6),



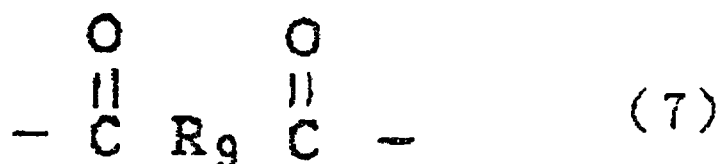
in which Y represents a direct bond, -O- -NR<sub>3</sub>-, R<sub>6</sub> represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and R<sub>7</sub> and R<sub>8</sub> each represents a hydroxyl group, an alkyl group, an aryl group or a group represented by the following formula (4) or (5),



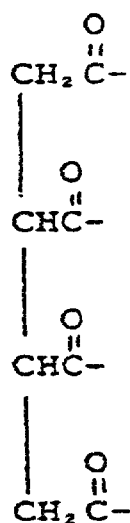
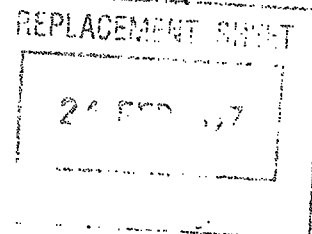


; when n is 2, R<sub>2</sub> represents a hydrogen atom, an alkyl group having 1-8 carbon atoms or  $\text{-C(=O)-(P)-(Q)-C(=O)-OH}$  in which P

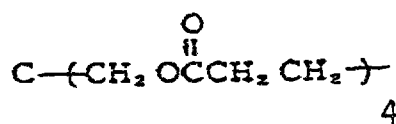
and Q represent one selected from the group consisting of an alkylene group an alkylidene group, a vinyl group and a vinxyldene group, and R<sub>1</sub> represents an alkylene group or a group represented by the following formula (7),



in which R<sub>9</sub> represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group; and when n is 4, R<sub>2</sub> represents a hydrogen atom or an alkyl group having 1-8 carbon atoms, and R<sub>1</sub> represents a group represented by the following formula (9) or (10),



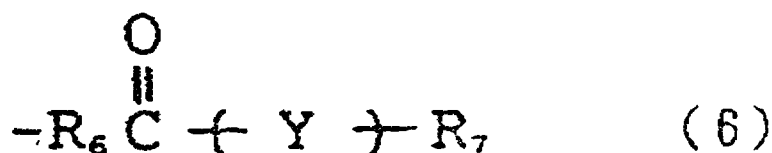
(9)



(10)

14. A thermoplastic resin composition according to claim 13, wherein in the formula (1),  $R_1$  and  $R_2$  each represents a hydrogen atom or an alkyl group having 1-8 carbon atoms and  $n$  is 1 or 2, and ;

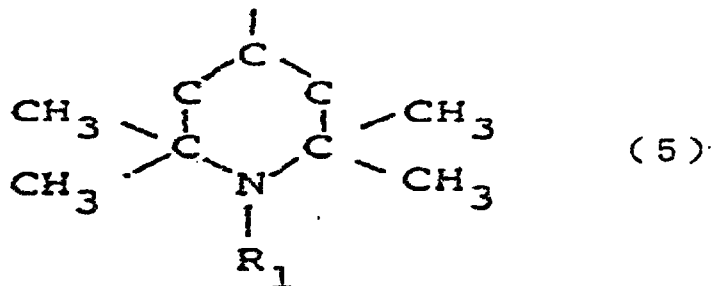
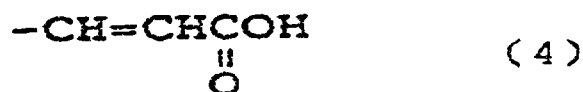
(i) when  $n$  is 1,  $R_2$  is a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group or a group represented by the following formula (6),



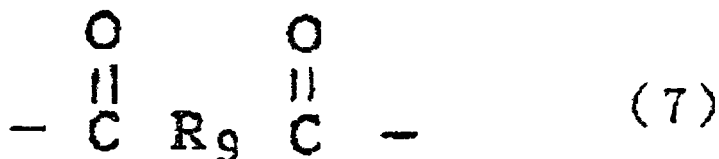
in which  $Y$  represents a direct bond,  $-O-$  or  $-NR_8-$ ,  $R_6$  represents a direct bond, a straight or branched alkylene



group having 1-10 carbon atoms, an alkylidene group or arylene group, and  $R_1$  and  $R_2$  each represent a hydroxyl group, an alkyl group having 1-18 carbon atoms, an aryl group having 6-21 carbon atoms or a group represented by the following formula (4) or (5),

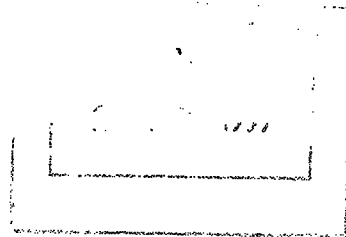


and (ii) when  $n$  is 2,  $R_3$  represents a group represented by the following formula (7),



in which  $R_3$  represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group or an arylene group.

15. A thermoplastic resin composition according to claim 13, wherein in the formula (1),  $n$  is 2 and  $R_3$  represents an



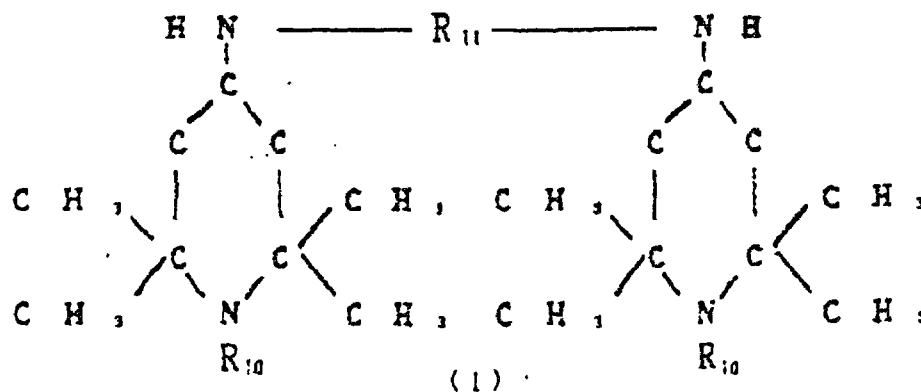
alkylene group.

16. A thermoplastic resin composition according to claim 13, wherein in the formula (1),  $n$  is 2,  $R_1$  represents a hydrogen atom and  $R_2$  represents an alkylene group.

17. A thermoplastic resin composition according to claim 13, wherein in the formula (1),  $n$  is 2,  $R_1$  represents a hydrogen atom or a methyl group,  $R_2$  represents a hydrogen atom and  $R_3$  represents a hexamethylene group.

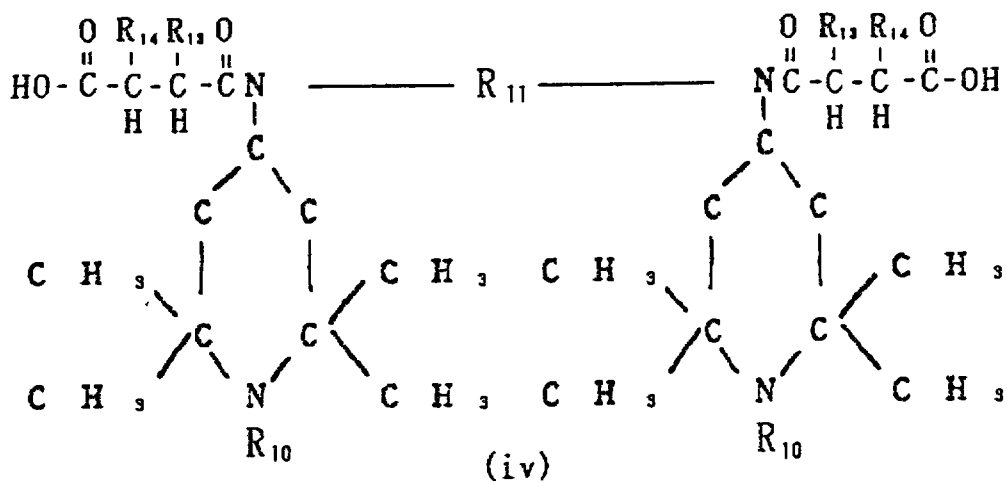
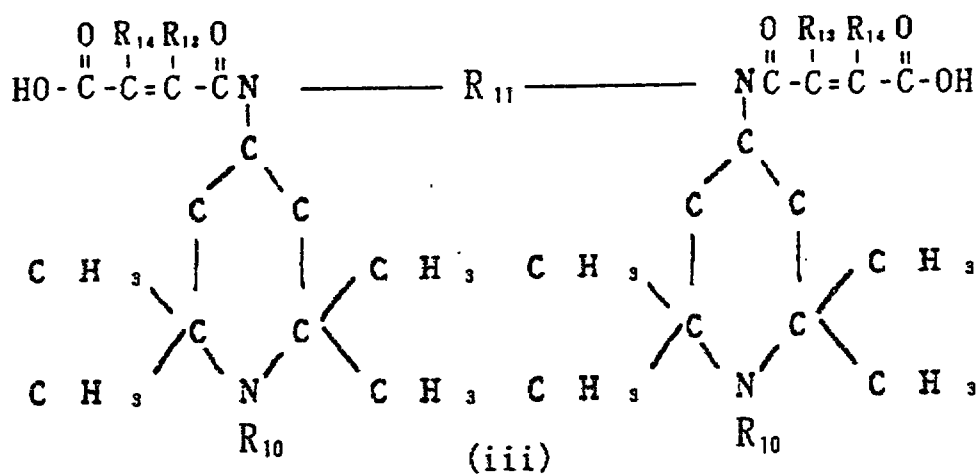
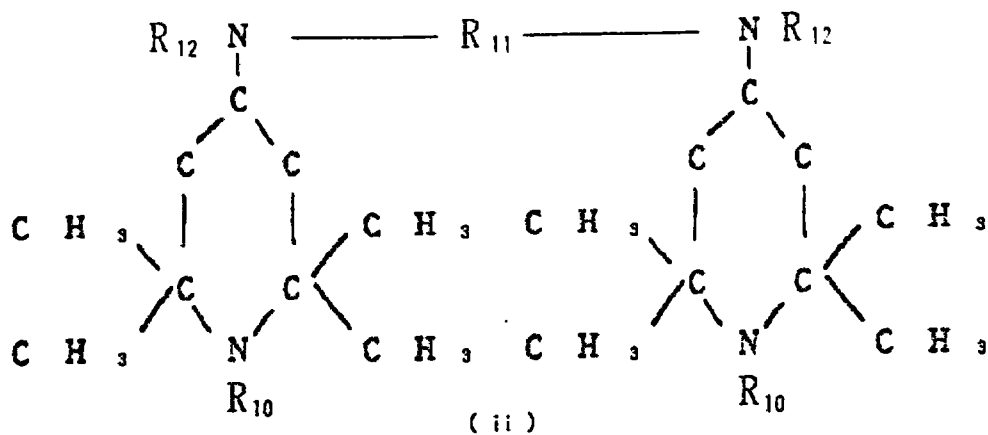
18. A thermoplastic resin composition according to claim 13, wherein in the formula (1),  $n$  is 2,  $R_1$  represents an alkyl group and  $R_2$  represents an alkylene group.

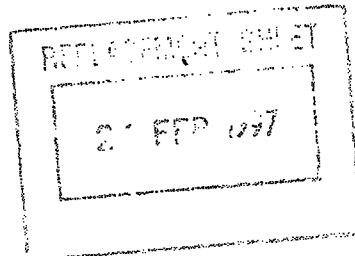
19. A thermoplastic resin composition according to claim 13, wherein the compound  $\text{C}$  represented by the formula (1) is at least one compound represented by the following formulas (i)-(iv):





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*a hydroxyl group,*

wherein  $R_{10}$  represents a hydrogen atom, an alkyl group or an alkyloxy group,  $R_{11}$  represents an alkylene group,  $R_{12}$  represents an alkyl group, and  $R_{13}$  and  $R_{14}$  each represents a hydrogen atom or a methyl group.

20. A thermoplastic resin composition according to claim 19, wherein in the formula (i),  $R_{10}$  represents a hydrogen atom, a hydroxyl group, an alkyl group having 1-10 carbon atoms or an alkyloxy group having 1-10 carbon atoms, and  $R_{11}$  represents an alkylene group having 2-8 carbon atoms.

21. A thermoplastic resin composition according to claim 19, wherein in the formula (i),  $R_{13}$  represents a hydrogen atom or a methyl group and  $R_{14}$  represents a hexamethylene group.

22. A thermoplastic resin composition according to claim 19, wherein in the formula (ii),  $R_{13}$  represents a hydrogen atom, a hydroxyl group, an alkyl group having 1-10 carbon atoms or an alkyloxy group having 1-10 carbon atoms,  $R_{14}$  represents an alkylene group having 2-8 carbon atoms, and  $R_{15}$  represents an alkyl group having 1-18 carbon atoms.

23. A thermoplastic resin composition according to claim 19, wherein in the formula (ii),  $R_{13}$  represents a hydrogen atom or a methyl group.



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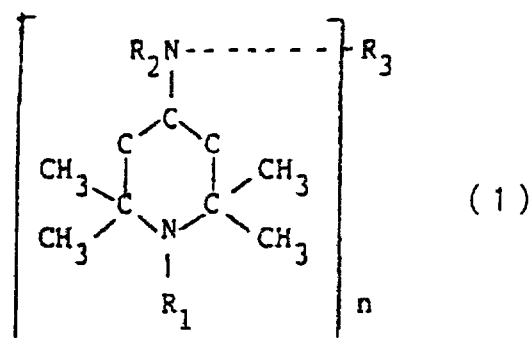
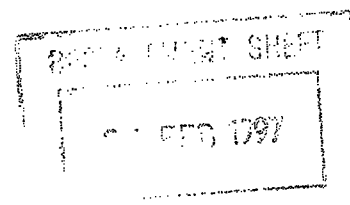
24. A thermoplastic resin composition according to claim 19, wherein in the formula (iii),  $R_{10}$  represents a hydrogen atom, a hydroxyl group, an alkyl group having 1-10 carbon atoms or an alkyloxy group having 1-10 carbon atoms, and  $R_{11}$  represents an alkylene group having 2-8 carbon atoms.

25. A thermoplastic resin composition according to claim 19, wherein in the formula (iii),  $R_{10}$  represents a hydrogen atom or a methyl group.

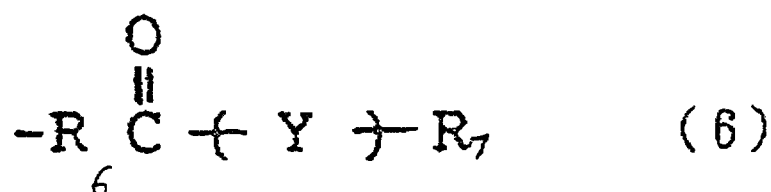
26. A thermoplastic resin composition according to claim 19, wherein in the formula (iv),  $R_{10}$  represents a hydrogen atom, a hydroxyl group, an alkyl group having 1-10 carbon atoms or an alkyloxy group having 1-10 carbon atoms, and  $R_{11}$  represents an alkylene group having 2-8 carbon atoms.

27. A thermoplastic resin composition according to claim 19, wherein in the formula (iv),  $R_{10}$  represents a hydrogen atom or a methyl group.

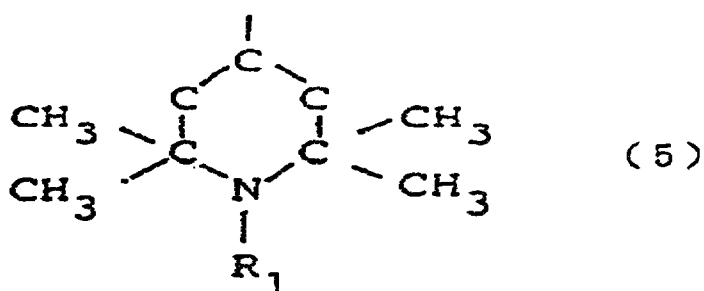
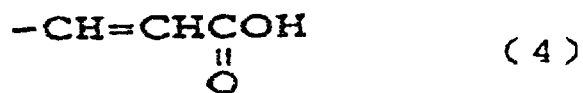
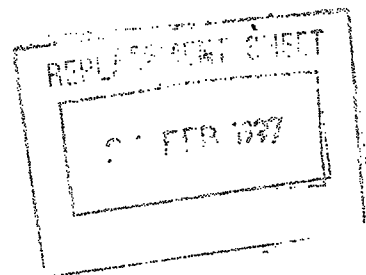
28. A thermoplastic resin composition according to claim 1, wherein the 4-amino-2,2,6,6-tetramethylpiperidyl compound (c) is a compound represented by the following formula (1) :



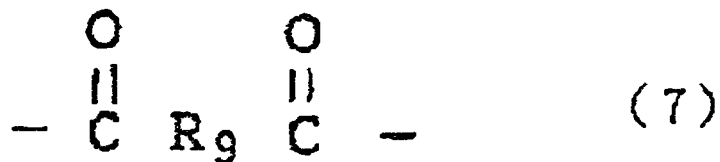
wherein  $\text{R}_1$  represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group having 1-8 carbon atoms, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group,  $\text{R}_2$  represents a hydrogen atom or alkyl group, and  $n$  is an integer of 1, 2 or 4 ; and  
(I) when  $n=1$ ,  $\text{R}_3$  is a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group, a benzyl group, an allyl group or a group represented by the following formula (6),



in which  $\text{Y}$  represents a direct bond,  $-\text{O}-$   $-\text{NR}_8-$ ,  $\text{R}_6$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and  $\text{R}_7$  and  $\text{R}_8$  each represents a hydroxyl group, an alkyl group, an aryl group or a group represented by the following formula (4) or (5),



(II) when  $n=2$ ,  $R_1$  represents a group represented by the following formula (7),



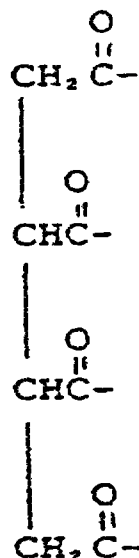
in which  $R_9$  represents a direct bond, a straight or branched alkylene group, an alkylidene group or an arylene group, and (IV) when  $n$  is 4,  $R_1$  represents a group represented by the following formula (9) or (10).



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REPLACEMENT SHEET

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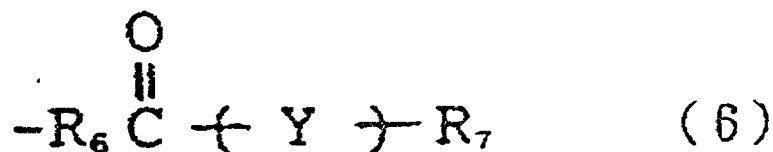


(9)



(10)

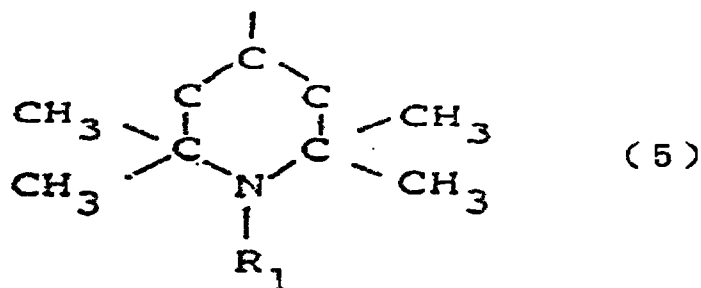
29. A thermoplastic resin composition according to claim 28, wherein in the formula (1),  $R_1$  represents a hydrogen atom or an alkyl group having 1-8 carbon atoms,  $R_2$  represents a hydrogen atom or an alkyl group having 1-18 carbon atoms, and  $n$  is an integer of 1 or 2 ; and when  $n$  is 1,  $R_3$  represents a hydrogen atom, an alkyl group, a cycloalkyl group, an alkenyl group or a group represented by the following formula (6),



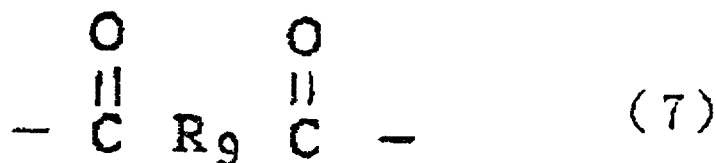
in which  $Y$  represents a direct bond,  $-O-$  or  $-NR_8-$ ,  $R_6$



represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group having 1-10 carbon atoms or arylene group, and R, and R, each represent a hydrogen atom, an alkyl group having 1-18 carbon atoms, an aryl group having 6-21 carbon atoms or a group represented by the following formula (4) or (5),

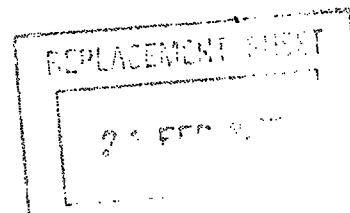


; and when n is 2, R, represents a group represented by the following formula (7),

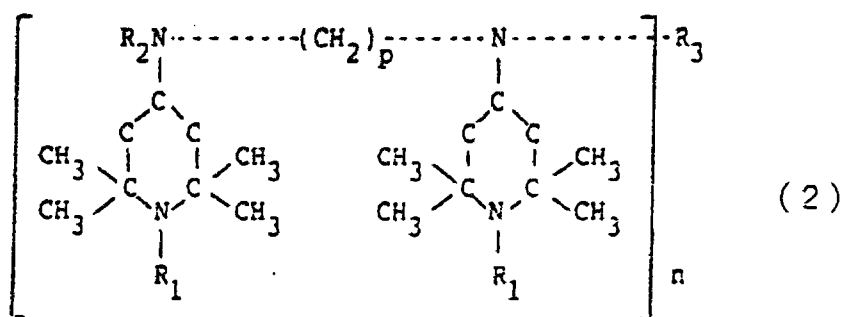


in which R, represents a direct bond, a straight or branched alkylene group having 1-10 carbon atoms, an alkylidene group having 1-10 carbon atoms or an arylene group.

30. A thermoplastic resin composition according to claim



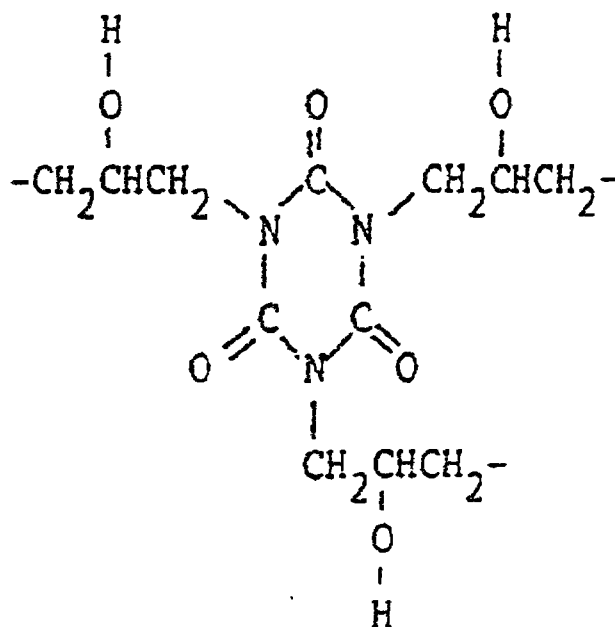
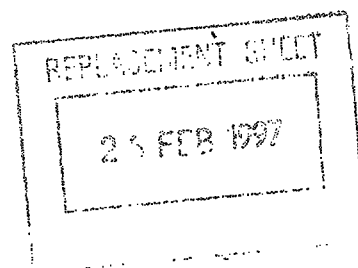
1, wherein the 4-amino-2,2,6,6-tetramethylpiperidyl compound (c) is a compound represented by the following formula (2) :



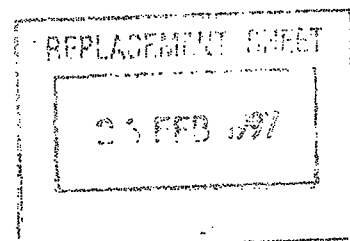
wherein  $\text{R}_1$  represents a hydrogen atom, an oxygen atom (oxy. radical), a hydroxyl group, an alkyl group having 1-8 carbon atoms, a cycloalkyl group, an allyl group, a benzyl group, an aryl group, an alkanoyl group, an alkenoyl group, an alkyloxy group or a cycloalkyloxy group, and  $n$  is an integer of 3; and wherein  $\text{R}_2$  represents a hydrogen atom or an alkyl group having 1-8 carbon atoms and  $p$  is an integer of 2 to 8, and  $\text{R}_3$  represents a group represented by the following formula (8) ;



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(8)

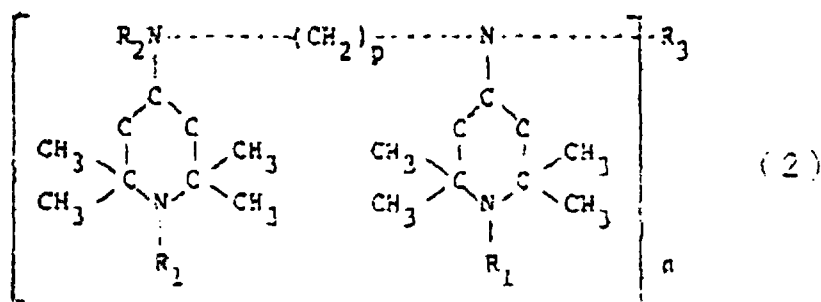
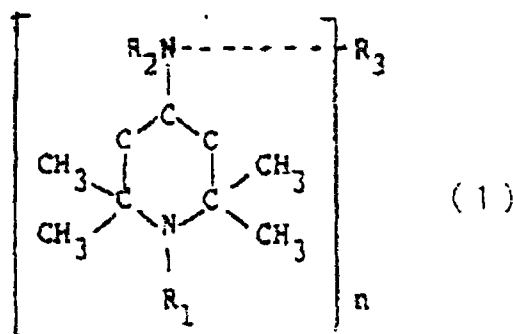


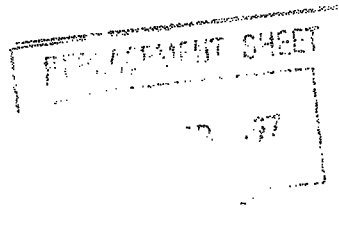
# ABSTRACT

## THERMOPLASTIC RESIN COMPOSITION

The present invention provides a thermoplastic resin composition improved in process stability and in heat stability which comprises :

- (a) 1 to 99 % by weight of a polyphenylene ether resin,
- (b) 99 to 1 % by weight of at least one non-crystalline thermoplastic resin selected from an alkenyl aromatic resin, a non-crystalline aromatic polyester resin, a methacrylate resin, a polyether imide and an aromatic polysulfone resin, and per 100 parts by weight of the sum of (a) and (b),
- (c) 0.001 to 15 parts by weight of at least one 4-amino-2,2,6,6-tetramethylpiperidyl compound represented by the following formula (1) or (2) ;





; wherein  $R_1$  represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group, a cycloalkyl group, *etc.*, ~~an aryl group, an alkyl group, an aryl group, an alkanoyl group, an alkenyl group, an alkynyl group or a cycloalkyl group~~;  $R_2$  represents a hydrogen atom, an alkyl group, *etc.*

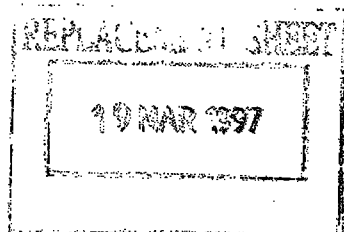
; ~~and wherein~~  $n$  represents an integer of 1 to 4;  $p$  represents an integer of 2 to 6; and  $R_3$  represents an  $n$  valent organic group.

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; wherein  $R_1$  represents a hydrogen atom, an oxygen atom (oxy radical), a hydroxyl group, an alkyl group, a cycloalkyl group, etc.;  $R_2$  represents a hydrogen atom, an alkyl group, etc.

;  $n$  represents an integer of 1 to 4;  $p$  represents an integer of 2 to 6; and  $R_3$  represents an  $n$  valent organic group.



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