

AUSTRALIA
PATENTS ACT 1990

636794

PATENT REQUEST/NOTICE OF ENTITLEMENT

We MINISTERO DELL'UNIVERSITA' E DELLA RICERCA SCIENTIFICA E
TECNOLOGICA

of 76 Lungotevere Thaon di Revel, 00196 ROME, ITALY

being the Applicant and Nominated Person, request the grant of a patent for an
invention entitled SELF-EXTINGUISHING POLYMERIC COMPOSITIONS which is
described in the accompanying standard complete specification.

Convention priority is claimed from the following basic application

Basic Applicant	Application Number	Application Date	Country	Country Code
Ministero Dell 'Universita' e Della Ricerca Scientifica e Tecnologica	21421 A/90	11 September 1990	Italy	IT

Roberto Cipolli; Gilberto Nucida; Enrico Masarati; Roberto Oriani and Mario
Pirozzi are the actual inventors of the invention.

Entitlement is by virtue of a contract of employment wherein the inventors made
the invention for and on behalf of the applicant in the course of their duties as
employees of the applicant.

The basic application was the first application made in a Convention Country in
respect of the invention the subject of this request.

DATED this 10th day of September 1991

Our address for Service:

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GH

MINISTERO DELL'UNIVERSITA' E DELLA
RICERCA SCIENTIFICA E TECNOLOGICA
By their Patent Attorney

J.H.A.

5024280 11/09/91

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GH&CO REF: 21463-J:RPW:RK

2571A:rk



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SELF-EXTINGUISHING POLYMERIC COMPOSITIONS

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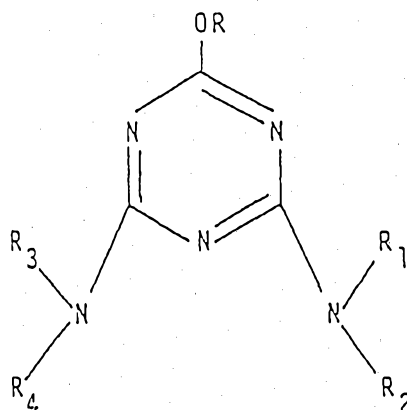
(56) Prior Art Documents
AU 83840/91 C08K 5/3492 C08L 23/12
AU 83839/91 C08K 5/5353 C08L 101/00
AU 627615 61365/90 C08K 13/02 C08L 23/02

(57) Claim 1. Self-extinguishing polymeric compositions consisting essentially of:

a) from 91 to 40 parts by weight of a thermoplastic polymer or of a polymer showing elastomeric properties;

b) from 6 to 33 parts by weight of ammonium phosphates, ammonium phosphonates, amine phosphates, amine phosphonates or mixtures thereof;

c) from 3 to 27 parts by weight of one or more compounds derived from the 2,4-diamino-6-hydroxy-1,3,5-triazine, having the general formula (I):



(I)

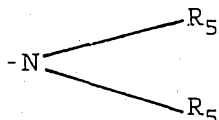
wherein:

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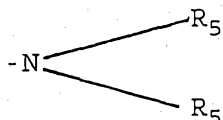
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(1) R is (a) H (b) $-\text{[C}_n\text{H}_{2n}\text{]}-\text{X}$,

wherein n is an integer from 1 to 8 and X=H; CN; -O-(C₁-C₄)-alkyl; -O-(C₂-C₄)-alkenyl; (C₆-C₁₂)-cycloalkyl or alkylcycloalkyl; -O-(C₆-C₁₂)-aryl;



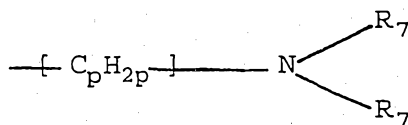
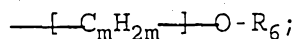
wherein radicals R₅, are the same or different and are (C₁-C₄)-alkyl, (C₃-C₄)-alkenyl; or the group:



is replaced by a N-containing heterocyclic radical bound to the alkyl chain through the nitrogen atom of said N-containing heterocyclic radical, said N-containing heterocyclic radical optionally containing another heteroatom selected from O, S and N;

(c) (C₂-C₆)-alkenyl, (d) (C₆-C₁₂)-cycloalkyl or (e) alkylcycloalkyl; (f) (C₆-C₁₂)-aryl or aralkyl;

(2) at least one of radicals from R₁ to R₄ is:



with:

m = integer from 2 to 8;

p = integer from 2 to 6;

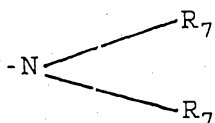
R₆=H; (C₁-C₈)-alkyl; (C₂-C₆)-alkenyl; $[\text{C}_q\text{H}_{2q}]-\text{O}-\text{R}_8$,

wherein q is an integer from 1 to 4 and R₈ is hydrogen or (C₁-C₄)-alkyl; (C₆-C₁₂)-cycloalkyl or alkylcycloalkyl; radicals R₇ are the same or different and are H; (C₁-C₈)-alkyl; (C₂-C₆)-alkenyl; (C₁-C₄)-hydroxyalkyl; (C₆-C₁₂)-cycloalkyl or alkylcycloalkyl;

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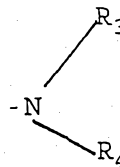
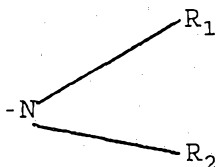
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or the group:



is replaced by a N-containing heterocyclic radical bound to the alkyl chain through the nitrogen atom of said N-containing heterocyclic radical, said N-containing heterocyclic radical optionally containing another heteroatom selected from O, S, and N;

(3) or in the general formula (I) at least one of groups:



is replaced by a N-containing heterocyclic radical bound to the triazinic ring through the nitrogen atom of said

N-containing heterocyclic radical, said N-containing heterocyclic radical optionally containing another heteroatom selected from O, S and N

(4) the remaining groups from R₁ to R₄, the same or different, have the above specified meaning or are:

H; (C₁-C₁₈)-alkyl; (C₂-C₈)-alkenyl; (C₆-C₁₆) cycloalkyl or alkylcycloalkyl, optionally substituted by a hydroxyl or (C₁-C₄)-hydroxyalkyl function, wherein the parts by weight of components a), b) and c) add up to 100 parts by weight.

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Regulation 3.2

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ORIGINAL
COMPLETE SPECIFICATION
STANDARD PATENT

Invention Title: SELF-EXTINGUISHING POLYMERIC COMPOSITIONS

The following statement is a full description of this invention, including the best method of performing it known to us:

GH&CO REF: 21463-J:RPW:RK

The present invention relates to self-extinguishing compositions based on thermoplastic polymers or on polymers endowed with elastomeric properties, especially olefinic polymers and copolymers, containing particular triazinic compounds combined with phosphates and/or phosphonates of ammonium or of an amine.

In the art there are known different solutions to reduce or remove the combustibility of polymers. Some of these solutions are based on the use of metal compounds, especially antimonium, bismuth or arsenic compounds, in combination with organic compounds partially halogenated and thermally unstable, such as chlorinated paraffinic waxes.

Other solutions are based on the use of substances able to produce intumescence. Formulations of intumescent type are generally consisting of the polymer and of at least three mainly additives: one essentially phosphorated, the purpose of which is to form during the combustion an impermeable, semisolid, vitreous layer, essentially consisting of polyphosphoric acid, and to activate the formation process of the intumescence, a second containing nitrogen which acts as foaming agent and a third containing carbon which acts as carbon donor to form an insulating cellular carbon layer (char) between the



polymer and the flame.

Examples of intumescent formulations of this type are those reported in US Patent 3,810,862 (Phillips Petroleum Co.) consisting of melamine, pentaerhythritol and ammonium polyphosphate, US Patent 4,727,102 (Vamp s.r.l.) consisting of melamine cyanurate, an hydroxyalkyl derivative of isocyanuric acid and ammonium polyphosphate, and in Published Patent Application WO 85/05626 (Plascoat U.K. Ltd.) consisting of different phosphorus and nitrogen compounds among which, in particular, a combination of melamine phosphate, pentaerhythritol and ammonium polyphosphate.

In more recent formulations, together with the use of an organic or inorganic phosphorus compound an organic compound has been used containing nitrogen, especially an aminoplastic resin obtained by condensing urea melamine or dicyandiamide with formaldehyde.

Examples of formulations consisting of two additives are those reported in US Patent 4,504,610 (Montedison S.p.A.), consisting of oligomeric derivatives of 1,3,5-triazine and ammonium polyphosphate, and European Patent 14,463 (Montedison S.p.A.) consisting of organic compounds selected from benzylguanamine and reaction products between aldehydes and several nitrogenous cyclic compounds, in particular benzylguanamine-formaldehyde copolymers, and ammonium polyphosphate.

It is also possible to obtain self-extinguishing compositions using monocomponent additives, containing in the organic molecule both nitrogen and phosphorus, as described in US Patent 4,201,705 (Borg-Warner Corp.).

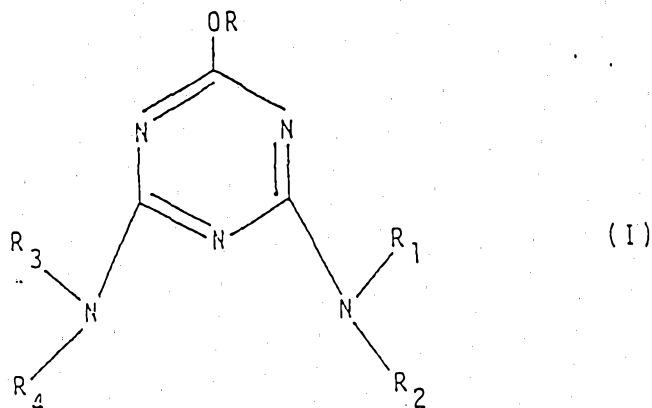
These intumescent retarding systems give the property of forming a carbon residue to the polymer containing them following a fire or application of a flame. Retarding systems of this type show many advantages: absence of corrosion phenomena in apparatus in which polymers are processed, lower emission of smokes in comparison with systems containing metal compounds and halogenated hydrocarbons and, in particular, the possibility of giving satisfactory antifiame properties to polymers using a lower quantity of total additive and therefore without an excessive decay of the mechanical properties of polymers themselves.

The Applicant have now found that it is possible to give very good anti-flame properties to the above mentioned polymers using a class of compounds derived from the 2,4-diamino-6-hydroxy-1,3,5-triazine, the effectiveness of which results to be superior also to that of the products known from the art.

More specifically the compositions of the present invention comprise:

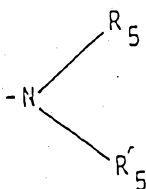
- a) from 91 to 40 parts by weight of a thermoplastic polymer or of a polymer having elastomeric properties;
- b) from 6 to 33, preferably from 8 to 30, parts by weight of ammonium phosphates, ammonium phosphonates, amine phosphates, amine phosphonates or mixtures thereof;
- c) from 3 to 27, preferably from 4 to 20, parts by weight of one or more compounds derived from the 2,4-diamino-6-hydroxy-1,3,5-triazine having the general formula (I):



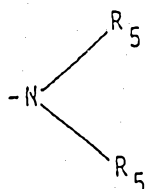


wherein:

- (1) R is (a) H, (b) $-\{C_nH_{2n}\}-X$, wherein n is an integer from 1 to 8, preferably between 1 and 4 and wherein X=H, CN, $-O-(C_1-C_4)$ -alkyl, $O-(C_2-C_4)$ -alkenyl, (C_6-C_{12}) -cycloalkyl or alkylcycloalkyl, $O-(C_6-C_{12})$ -aryl, preferably hydrogen,



wherein said R_5 groups are the same or different and are (C_1-C_4) -alkyl, (C_3-C_4) -alkenyl, or the group:

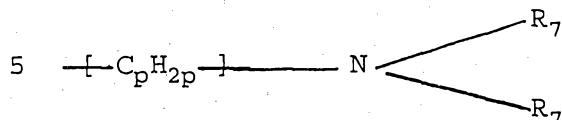
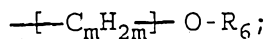


is replaced by a N-containing heterocyclic radical bound to the alkyl chain through the nitrogen atom of said N-containing heterocyclic radical, said N-containing heterocyclic radical optionally containing another heteroatom selected from O, S, N;

- (c) (C_2-C_6) -alkenyl, (d) (C_6-C_{12}) -cycloalkyl, (e) alkylcycloalkyl; (f) (C_6-C_{12}) -aryl or

-aralkyl;

(2) at least one of radicals from R_1 to R_4 is:

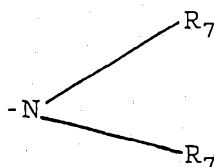


with : m =integer from 2 to 8, preferably from 2 to 4;

p = integer from 2 to 6

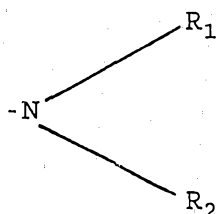
R_6 = H; (C_1-C_8) -alkyl, preferably H or alkyl C_1-C_4 ;
 10 (C_2-C_6) -alkenyl; $-[-C_qH_{2q}]O-R_8$, wherein q is an integer
 from 1 to 4 and R_8 is hydrogen or (C_1-C_4) -alkyl; (C_6-C_{12}) -
 cycloalkyl or alkylcycloalkyl;

radicals R_7 , are the same or different and are H;
 15 (C_1-C_8) -alkyl; (C_2-C_6) -alkenyl; (C_1-C_4) -hydroxyalkyl;
 (C_6-C_{12}) -cycloalkyl or alkylcycloalkyl;
 or the group:

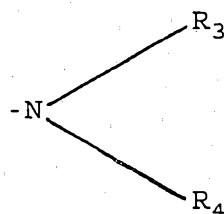


20 is replaced by a N-containing heterocyclic radical bound
 to the alkyl chain through the nitrogen atom of said N-
 containing heterocyclic radical, said N-containing
 heterocyclic radical optionally containing another
 heteroatom selected from O, S, N;

25 (3) or in the general formula (I) at least one of
 groups:



;



is replaced by a N-containing heterocyclic radical bound to the triazinic ring through the nitrogen atom of said N-containing heterocyclic radical, said N-containing heterocyclic radical optionally containing another heteroatom selected from O, S, N;

(4) the remaining groups from R_1 to R_4 are the same or different and have the above mentioned meaning or are:

H; (C_1-C_{18}) -alkyl; (C_6-C_{16}) -cycloalkyl or alkylcycloalkyl, optionally substituted by a hydroxy or (C_1-C_4) hydroxyalkyl function, wherein the parts by weight of compounds (a), (b) and (c) add up to 100 parts by weight.

The above mentioned compounds of general formula (I) obtained by the insertion on the molecule of the 2,4,-diamino-6-hydroxy,1,3,5-triazine, which however is unable to give self-extinguishing property to the above mentioned polymers, of a suitable substituent selected from those hereinafter described, have a simple structure and are particularly stable when subjected to heat and therefore maintain a high activity as flame retarders even after polymeric compositions containing them have been subjected to heat.

Furthermore, the compositions of the present invention show the advantage of giving, in the event of fire, a very poor and not darkening emission of smoke.

Examples of radical R in the general formula (I) are:

methyl; ethyl; propyl; isopropyl; n-butyl; isobutyl; tert-butyl; n-pentyl; isopentyl; n-hexyl; ter-hexyl; octyl; ter-octyl; ethenyl; propenyl; butenyl; isobutenyl; hexenyl; cyclohexyl; propylcyclohexyl; butylcyclohexyl; phenyl; benzyl; 2-phenylethyl; cyanomethyl; 2-cyanoethyl; 2-methoxyethyl; 2-methoxypropyl; 3-methoxypropyl; 4-methoxy-

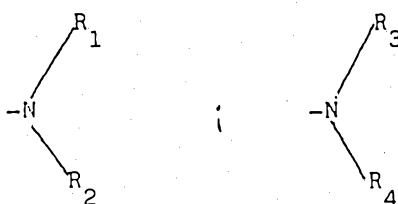
butyl; 6-methoxyhexyl; 7-methoxyheptyl; 7-methoxyoctyl; 2-ethoxyethyl;
3-ethoxypropyl; 4-ethoxybutyl; 5-ethoxypentyl; 2-cyclohexyloxyethyl;
2-ethenyloxyethyl; 2-phenoxyethyl; 2-(N,N-dimethylamino)ethyl; 3-(N,N-
dimethylamino)propyl; 4-(N,N-dimethylamino)butyl; 5-(N,N-dimethyla-
mino)pentyl; 6-(N,N-dimethylamino)hexyl; 2-(N,N-diethylamino)ethyl;
3-(N,N-diethylamino)propyl; 4-(N,N-diethylamino)butyl; 2-(N,N-dipro-
pylamino)ethyl; 2-[N-methyl-N-(1-propenyl)amino]ethyl; 2-[N,N-di(1-pro-
penyl)amino]ethyl; 4-[N,N-di(1-propenyl)amino]butyl; etc.

Examples of radicals from R_1 to R_4 are:

methyl; ethyl; propyl; isopropyl; n-butyl; isobutyl; terbutyl;
n-pentyl; isopentyl; n-hexyl; ter-hexyl; octyl; ter-octyl; decyl;
dodecyl; octadecyl; ethenyl; propenyl; butenyl; isobutenyl; hexenyl;
octenyl; cyclohexyl; propylcyclohexyl; butylcyclohexyl; decylcyclo-
hexyl; hydroxycyclohexyl; hydroxyethylcyclohexyl; 2-hydroxyethyl;
2-hydroxypropyl; 3-hydroxypropyl; 3-hydroxybutyl; 4-hydroxybutyl;
3-hydroxypentyl; 5-hydroxypentyl; 6-hydroxyhexyl; 3-hydroxy-2,5-
dimethylhexyl; 7-hydroxyheptyl; 7-hydroxyoctyl; 2-methoxyethyl;
2-methoxypropyl; 3-methoxypropyl; 4-methoxybutyl; 6-methoxyhexyl;
7-methoxyheptyl; 7-methoxyoctyl; 2-ethoxyethyl; 3-ethoxypropyl;
4-ethoxybutyl; 3-propoxypropyl; 3-butoxypropyl; 4-butoxybutyl;
4-isobutoxybutyl; 5-propoxypentyl; 2-cyclohexyloxyethyl; 2-ethenyl-
oxyethyl; 2-(N,N-dimethylamino)ethyl; 3-(N,N-dimethylamino)propyl;
4-(N,N-dimethylamino)butyl; 5-(N,N-dimethylamino)pentyl; 4-(N,N-die-
thylamino)butyl; 5-(N,N-diethylamino)pentyl; 5-(N,N-diidropro-
pylamino)pentyl; 3-(N-ethylamino)propyl; 4-(N-methylamino)butyl;

4-(N,N-dipropylamino)butyl; 2-(N,N-diisopropylamino)ethyl; 6-(N-hexenylamino)hexyl; 2-(N-ethenylamino)ethyl; 2-(N-cyclohexylamino)ethyl; 2-(N-hydroxyethylamino)ethyl; 2-(2-hydroxyethoxy)ethyl; 2-(2-methoxyethoxy)ethyl; 6-(N-propylamino)hexyl; etc.

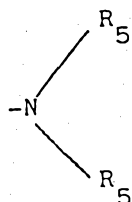
Examples of heterocyclic radical which may replace groups:



in the general formula (II) are:

aziridine; pyrrolidine; piperidine; morpholine; thiomorpholine; piperazine; 4-methylpiperazine; 4-ethylpiperazine; 2-methylpiperazine; 2,5-dimethylpiperazine; 2,3,5,6-tetramethylpiperazine; 2,2,5,5-tetramethylpiperazine; 2-ethylpiperazine; 2,5-diethylpiperazine; etc.

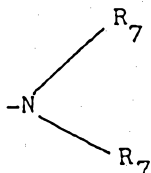
Examples of heterocyclic radical which may replace the group:



are:

pyrrolidine; piperidine, morpholine; thiomorpholine; piperazine; 4-methylpiperazine; 4-ethylpiperazine; etc.

Examples of heterocyclic radical which may replace the group:

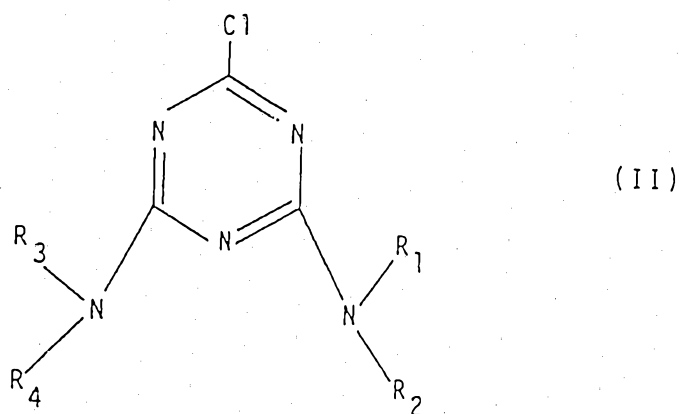


are:

aziridine, pyrrolidine; piperidine; morpholine; thiomorpholine; piperazine; 4-methylpiperazine; 4-ethylpiperazine; etc.

Particularly preferred are the compounds of general formula (I) wherein R is hydrogen.

Compounds of general formula (I) can be prepared from the intermediates of general formula (II):



wherein radicals from R_1 to R_4 have the previously defined meaning, according to the following modalities:

- a) when R is hydrogen by hydrolysis either with an acid (such as for instance hydrochloric acid, hydrobromic acid, sulfuric acid, phosphoric acid, etc.) at temperatures comprised between 60 and 100°C, or with a base (such as for instance sodium hydroxide, potassium hydroxide, etc.), at temperatures comprised between 100 and 180°C;
- b) when R is different from hydrogen by condensation reaction with a reactive of general formula (III):



wherein R has the previously defined meaning, in a solvent (such as for instance toluene, xylene, orthodichlorobenzene, etc.) or in

excess of reactive (III) if it may act as solvent (such as for instance methyl alcohol, ethyl alcohol, etc.) in the presence of a base (such as for instance sodium hydroxide, potassium hydroxide, metal sodium, etc.) at temperatures comprised between 60 and 150°C.

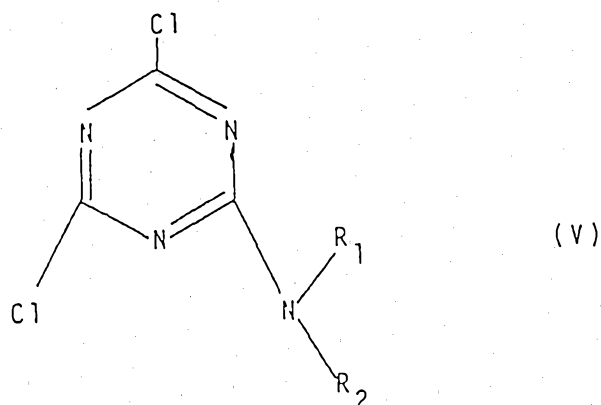
The product formed can be easily separated from the reaction mass by filtration.

Generally products of general formula (I) are obtained, having a good quality, in form of white crystalline powder, useable in self-extinguishing compositions without any further purification.

Intermediates of general formula (II) can be easily synthesized by allowing to react a halide of the cyanuric acid, for instance, the chloride, at a temperature comprised between 0 and 10°C, in a solvent (such as for instance, acetone, water, methylene chloride, etc.) with an amine of general formula (IV):



wherein R_1 and R_2 have the previously defined meaning, in the presence or not (according to the molar ratio used) of an acidity acceptor (such as for instance NaOH , NaHCO_3 , Na_2CO_3 , triethylamine, etc.) thus obtaining the intermediate of general formula (V):

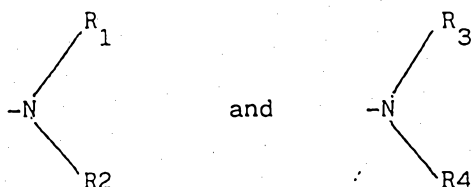


This intermediate, either separated or not, is then allowed to react under conditions analogous to the preceding ones, but working at higher temperature, for instance between 10 and 50°C, with an amine of general formula (VI):



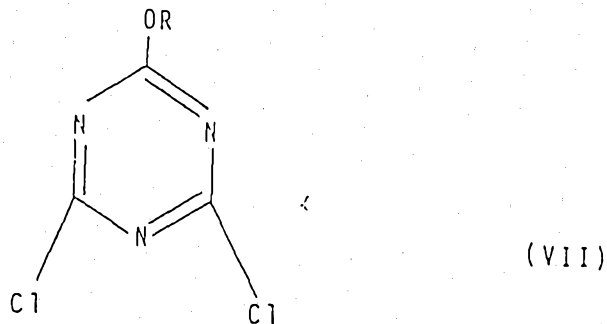
wherein R_3 and R_4 have the previously defined meaning.

In the event that it is desired to obtain derivatives of the general formula (I) wherein groups :



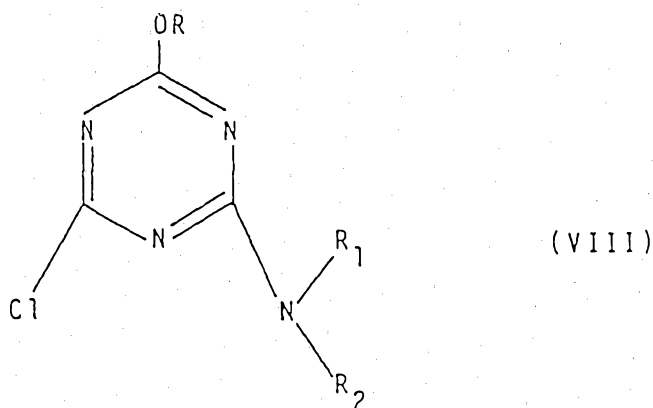
are equal between them, one mol of the halide of cyanuric acid is allowed to react with two mols, if in the presence of an acidity acceptor or with four mols if in the absence of the acidity acceptor, of an amine of the general formula (IV) under working conditions analogous to those previously described.

An alternative method for obtaining compounds of the general formula (I), wherein R is different from hydrogen, consists in allowing halide of cyanuric acid, for instance the chloride, to react with a derivative of general formula (III), at temperature comprised between 10 and 110°C, in a suitable solvent (such as for instance, acetone, methylene chloride, toluene, xylene etc.) or in an excess of the reagent of general formula (III) if it has solvent characteristics (such as methyl alcohol, ethyl alcohol, etc.) in the presence of an acidity acceptor (such as for instance NaHCO_3 , Na_2CO_3 , NaOH , triethylamine, collidine, etc.) thus obtaining the intermediate of general formula (VII):



wherein R has the previously defined meaning.

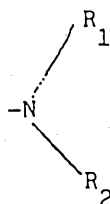
This intermediate, either separated or not, is preferably allowed to react with an amine of general formula (IV) under conditions analogous to the preceding ones, but at temperatures comprised between -5 and 30°C, to give the intermediate of general formula (VIII):



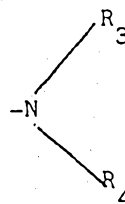
wherein R, R₁ and R₂ have the previously defined meaning.

gain under conditions analogous to the preceding ones, working at higher temperature, for instance from 40 to 120°C, and therefore using a solvent compatible also with these temperatures (such as for instance, water, toluene, xylene, etc.) with an amine of general formula (VI) thus obtaining finally compounds of general formula (I).

In the event that compounds of general formula (I) are desired wherein R is different also in this case from hydrogen, and groups :



and



are equal between them, one mol of the intermediate of general formula (VII) is allowed to react with two mols, if in the presence of an acidity acceptor, or with four mols, if in absence, of an amine of general formula (IV) under conditions analogous to those previously described.

From compounds of general formula (I) wherein R is different from

hydrogen, and is preferably (C₁-C₄) alkyl, it is possible to obtain derivatives of general formula (I) wherein R is equal to hydrogen, by hydrolysis reaction either with an acid, by working at temperatures comprised between 80 and 140°C, or with a base, by working at temperatures comprised between 100 and 180°C, using the same reagents indicated for the hydrolysis of intermediates of general formula (II).

Among phosphates ammonium polyphosphates are preferred which are comprised in the general formula $(\text{NH}_4)_{n+2} \text{P}_n \text{O}_{3n+1}$ wherein n represents an integer equal or higher than 2; preferably the molecular weight of polyphosphates must be enough high to secure a low water solubility. As example, n varies preferably between 2 and 500.

The composition of polyphosphates having the above specified formula, wherein n is a number enough great and preferably comprised between 5 and 500, is practically that corresponding to the formula of metaphosphates $(\text{NH}_4\text{PO}_3)_n$.

An example of these polyphosphates is that known under the trade name "Exolit 422" (manufactured and sold by Hoechst Society) and having the composition $(\text{NH}_4\text{PO}_3)_n$ wherein n is higher than 50; another example is the product known under the trademark "Phos-Chek P/30" (Monsanto Chemical) and having analogous composition.

Another polyphosphate which can be advantageously used especially because the reduced water solubility is that known under the trade name "Exolit 462" (manufactured and sold by Hoechst) and corresponding to Exolit 422 microincapsulated in melamina-formaldehyde resin.

Other useable phosphates are those deriving from amines, such as for instance dimethylammonium phosphate or diethylammonium phosphate, ethylenediamine phosphate, melamine ortho or pyrophosphate.

Among phosphonates very good results are obtained when ammonium phosphonates (mono or poly substituted) derived from mono or poly-phosphonic acids are used.

Examples of these phosphonates are:

ethane-1,1,2-triphosphonic acid; 2-hydroxyethane-1,1,2-triphosphonic acid; propane-1,2,3-triphosphonic acid; methylphosphonic acid; ethylphosphonic acid; n-propylphosphonic acid; n-butylphosphonic acid; phenylphosphonic acid; 1-aminoethane-1,1-diphosphonic acid; 1-hydroxyethane-1,1-diphosphonic acid; 1-hydroxydodecane-1,1-diphosphonic acid, phosphonoacetic acid; 2-phosphonopropionic acid; 3-phosphonopropionic acid; 2-phosphonobutyric acid; 4-phosphonobutyric acid; aminotris(methylenphosphonic) acid; ethylenediaminetetra(methylenphosphonic) acid; hexamethylenediaminetetra(methylenphosphonic) acid; diethylenetriaminopenta(methylenphosphonic) acid; etc.

Among polymers useable in compositions of the present invention olefine polymers or copolymers are preferred having general formula $R-CH=CH_2$ wherein R is a hydrogen atom or a (C_1-C_8) -alkyl or (C_1-C_8) -aryl radical, in particular:

- 1) isotactic or prevailing isotactic polypropylene;
- 2) HDPE, LLDPE, LDPE polyethylene;
- 3) crystalline propylene copolymers with lower proportions of ethylene and/or other alpha-olefins, such as for instance 1-butene, 1-hexene, 1-octene, 4-methyl-1-pentene;

- 4) heterophasic compositions comprising (A) a homopolymeric fraction of propylene or one of copolymers specified under (3) and (B) a copolymeric fraction consisting of elastomeric copolymers of ethylene with an alpha-olefin, optionally containing lower proportion of diene, wherein the alpha-olefin is preferably selected from propylene and 1-butene;
- 5) elastomeric copolymers of ethylene with alpha-olefins optionally containing lower proportions of a diene.

Examples of dienes among those more commonly present in the above mentioned elastomeric copolymers are butadiene, ethylidene-norbornene, hexadiene-1,4.

Among polymers of olefins of the general formula $R-CH=CH_2$ wherein R is an aryl radical, "crystal" and antishock polystyrene are preferred.

Other examples of commonly useable polymers are the ABS and SAN copolymers; the polyurethane (polyester and polyether); the polyethyleneterephthalate; the polybutylenterephthalate; polyamides; etc.

Self-extinguishing compositions of the present invention can be prepared according to known methods: for instance the phosphate and/or the phosphonate of ammonium or of an amine are first intimately mixed with one or more nitrogenous compounds of the general formula (I) finely milled (preferably with particle sizes lower than 70 microns) and the thus obtained mixture is added to the polymer in a turbomixer to form a homogeneous blend which is extruded and granulated. The

granulated product thus obtained can be transformed in different articles according to anyone of known moulding techniques.

Antiflame additives of the present invention can be used also in the field of antifire paints.

Amelinic compounds comprised in the general formula (I), besides those exemplified, which can be advantageously used in polymeric compositions of the present invention are those listed in Table 1.

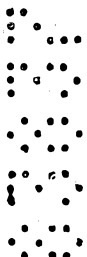


TABLE 1

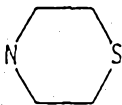
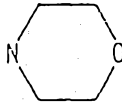
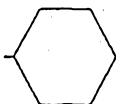
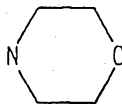
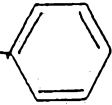
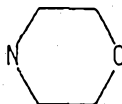
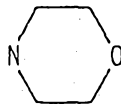
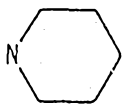
Pro- ducts	R	R_1-N-R_2	R_3-N-R_4
1	H		H H
2	C ₂ H ₅	CH ₂ CHOH CH ₃	H H
3	CH ₃		C ₂ H ₅ H
4	CH ₃	CH ₂ CH ₂ OH C ₂ H ₅	H H
5			H H
6	H	CH ₂ CH ₂ OH	H 
7	CH ₂ - 	CH ₂ CH ₂ OCH ₃	H CH ₂ CH ₂ OCH ₃
8	CH ₂ CH ₂ CN	CH ₂ CH ₂ CH ₂ OH	H CH ₂ CH ₂ CH ₂ OH
9	CH ₃		
10	CH ₃		H H

TABLE 1

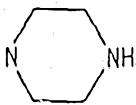
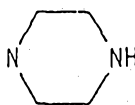
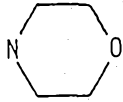
Products	R	$R_1 - N - R_2$	$R_3 - N - R_4$
11	H	$(CH_2)_3OC_2H_5$ H	$(CH_2)_3OC_2H_5$ H
12	C_2H_5	CH_2CHOH CH_3 H	CH_2CHOH CH_3 H
13	$n-C_4H_9$	CH_2CHOH CH_3 H	H H
14	CH_3	$(CH_2)_3OCH_3$ H	$(CH_2)_3OCH_3$ H
15	$CH_2CH_2N(CH_3)_2$	CH_2CH_2OH H	CH_2CH_2OH H
16	H	 N NH	 N NH
17	H	CH_2CH_2OH CH_2CH_2OH H	H H
18	H	$(CH_2)_4OCH_3$ H	$(CH_2)_4OCH_3$ H
19	C_2H_5	$CH_2CH_2CH_2N$  H	H H
20	CH_3	$CH_2-C=CH_2$ CH_3 H	H H

TABLE 1

Pro- ducts	R	R_1-N-R_2	R_3-N-R_4
21	$CH_2CH_2OCH_3$	CH_2CH_2OH H	
22	H	$(CH_2)_2O(CH_2)_2OH$ H	$(CH_2)_2O(CH_2)_2OH$ H

1000

1000

1000

Examples reported hereinbelow illustrate the characteristics of the invention without limiting them in any way.

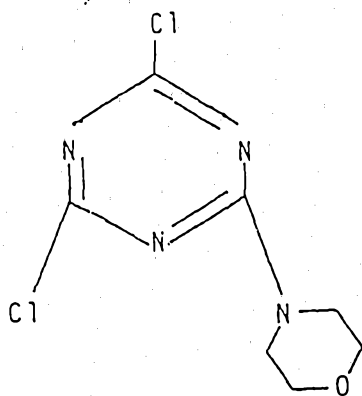
EXAMPLE 1

Into a 2 litres reactor equipped with stirrer, thermometer, dropping funnel, condenser and heating bath, there are introduced 184.5 g of cyanuric acid chloride and 1300 cc of methylene chloride.

By cooling from the outside, 87.2 g of morpholine and 40 g of sodium hydroxide dissolved in 150 g of water are fed contemporarily, within 3 hours, by keeping the pH comprised between 5 and 7 and the temperature comprised between 0 and 3°C.

The mixture is kept at the temperature of 0-3°C for further 3 hours and thereafter the aqueous phase is separated.

By distillation of the methylene chloride 230 g of the intermediate (IX):



(IX)

are obtained in form of white crystalline powder having m.p.=155-157°C (m.p.= melting point) and a chlorine content equal to 30.12% (theor.=30.12%).

In 1 liter reactor, equipped with stirrer, thermometer, feeding funnel, cooler and heating bath, there are introduced 300 cc of water,

30.5 g of 2-hydroxyethylamine and, while agitating, 117.5 g of the intermediate (IX).

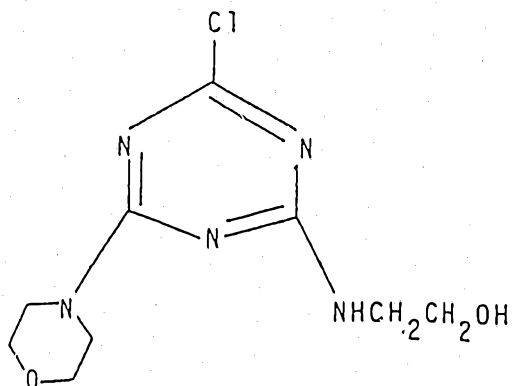
The temperature is gradually raised to 40°C; after 30 minutes the whole is heated at 45°C and is kept at this temperature for about 3 hours.

The temperature is raised again to 50°C and within 3 hours a solution consisting of 20 g of sodium hydroxide dissolved in 100 cc of water is added.

The whole is kept for further 2 hours at 50°C and thereafter is heated to 70°C and is allowed to further react at this new temperature for 30 minutes.

After cooling to room temperature, the product formed is filtered off and washed on the filter with water.

After drying of the cake in oven at 100°C, 120.3 g of the intermediate (X):



(X)

are obtained in form of white crystalline powder the m.p. of which is 172-173°C and the chlorine content is equal to 13.51% (theor.:13.68%).

The structure of intermediates (IX) and (X) has been further con-

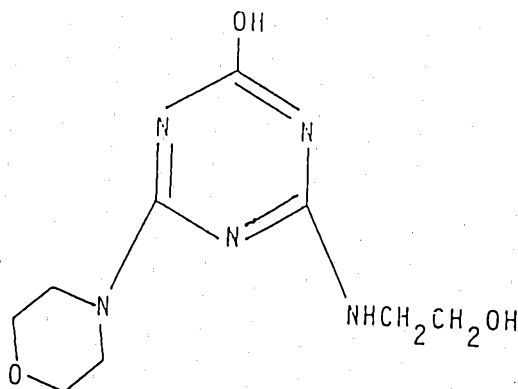
firmed by spectroscopic IR analysis.

In the same 1 liter reactor there are fed 500 cc of water, 103.8 g of the intermediate (X) and 79 g of a 37% by weight solution of hydrochloric acid.

The mass is heated to 90°C and is kept at this temperature for 3 hours.

The solution is then cooled to 50°C and is neutralized by adding 48 g of sodium hydroxide dissolved in 80 cc of water.

By drying the cake in oven at 100°C 84.7 g of the product:



are obtained in form of white crystalline powder; m.p.=251-253°C.

EXAMPLE 2

Into a 2 litres reactor, equipped as in example 1, 800 cc of methyl alcohol, 100 cc of water and 151.2 g of sodium bicarbonate are fed.

The mixture is cooled to 10°C and then 166 g of the cyanuric acid chloride are introduced.

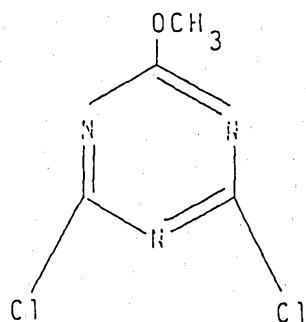
The temperature is allowed to raise up to 30°C and is kept at this value for about 1 hour, until the carbon dioxide release is ended.

The exothermy itself of the reaction is enough to keep the desired

temperature.

The whole is cooled to 5°C and thereafter 800 cc of cold water are added. The product formed is filtered off and washed on the filter with cold water.

By drying the cake in oven under vacuum at 60°C, 123.8 g of the intermediate (XI):



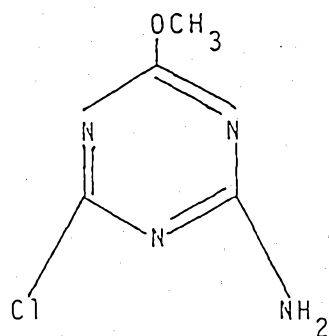
(XI)

are obtained in form of white crystalline powder; m.p.= 90-92°C; chlorine content equal to 39.27% (theor. 39.44%).

In a 1 liter reactor, equipped as in example 1, 400 cc of water and 108 g of the intermediate (XI) are introduced.

After having cooled from the outside to 0-5°C, 100 g of a 30% by weight ammonia solution are fed within about 1 hour while keeping the temperature at 0-5°C. The temperature is allowed to raise spontaneously to room temperature and this value is maintained for 2 hours.

The whole is cooled to 10°C, the product formed is filtered off and washed with cold water. By drying the cake in oven at 100°C, 82.3 g of the intermediate (XII):



(XII)

are obtained in form of white crystalline powder; m.p. higher than 300°C; chlorine content equal to 21.96% (theor.: 22.12%).

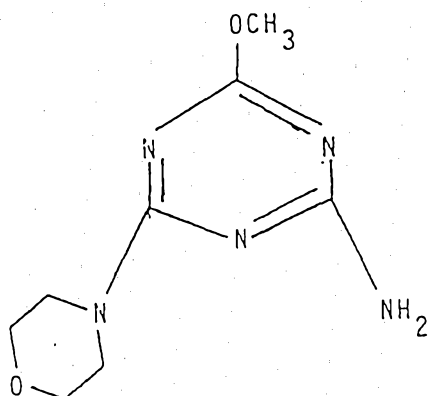
The structure of intermediates (XI) and (XII) was further confirmed by NMR analysis.

In the same 1 liter apparatus, but provided with heating bath, 300 cc of toluene, 80.2 g of the intermediate (XII) and 90 g of morpholine are fed.

The whole is heated to 60-65°C and is kept at this temperature for 2 hours; thereafter the mixture is heated to boiling and is kept under reflux for 1 hour.

After having allowed the mixture to cool to room temperature, the product formed is separated by filtration.

The cake is abundantly washed with water and, after drying, 90.3 g of the product:



are obtained in form of white crystalline powder; m.p. = 182-184°C.

EXAMPLE 3

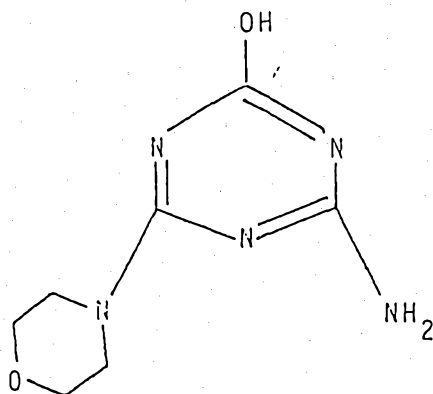
In a 1 liter reactor, equipped as in example 2, 400 cc of water, 63.3 g of the product of example 2 as well as 59.1 g of a 37% by weight hydrochloric acid solution are fed.

The mixture is heated to boiling and is kept under reflux for 2 hours.

After cooling to 80°C, 24 g of sodium hydroxide dissolved in 100 cc of water are added.

The whole is allowed to cool to room temperature, then the product formed is filtered off and washed on the filter with water.

By drying the cake in oven at 100°C, 54.7 g of the product:



are obtained in form of white crystalline powder; m.p. higher than 300°C.

EXAMPLE 4

600 cc of water and 184.5 g of the cyanuric acid chloride are fed in a 2 litres reactor equipped as in example 1.

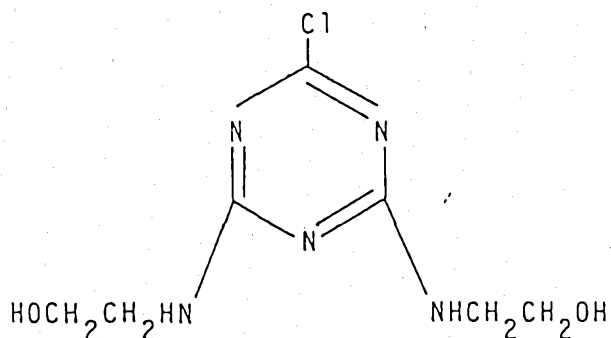
By cooling from the outside to 2°C, 122.5 g of 2-hydroxyethylamine in 100 cc of water are fed within 2 hours; during the addition the temperature is allowed to gradually raise up to 5-7°C.

The temperature is raised to 20°C and is kept at this value for 1 hour; then the whole is heated to 35-40°C and 80 g of sodium hydroxide dissolved in 200 cc of water are added within about 3 hours.

The reaction mass is heated to 60°C and is kept at this temperature for 2 hours.

The reaction mass is cooled to room temperature, the product formed is filtered off and washed on the filter with water.

By drying the cake in oven at 100°C, 203.1 g of the intermediate (XIII):



(XIII)

are obtained in form of white crystalline powder; m.p.= 188-190°C; chlorine content equal to 15.33% (theor. 15.20%).

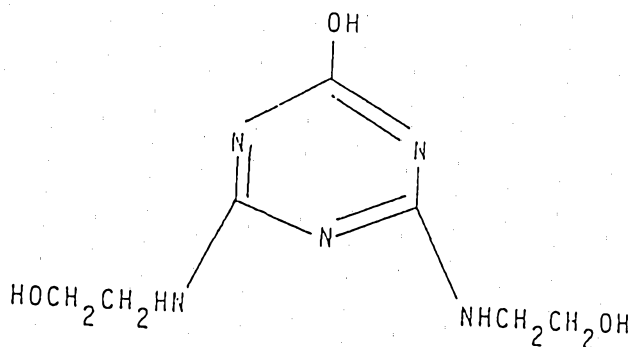
The structure of the intermediate (XIII) has been further confirmed by IR spectroscopic analysis.

400 cc of water, 13 g of sodium hydroxide and 70.1 g of the intermediate (XIII) are fed in a 1 liter stainless reactor equipped as in example 1.

The mixture is then heated to 150°C and is kept at this temperature for about 10 hours.

The whole is cooled to room temperature, the product formed is filtered and washed on the filter with water.

By drying the cake in oven at 100°C, 57.7 g of the product:

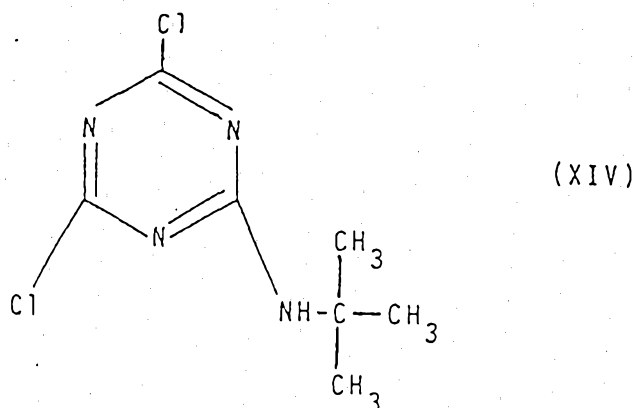


are obtained in form of white crystalline powder; m.p. higher than 300°C.

EXAMPLE 5

1000 cc of methylene chloride, 129.1 g of cyanuric acid chloride and 51.2 g of t.butyl amine are fed in the same 2 litres apparatus of example 1.

By working then as described in example 1, after having distilled the solvent, 148 g of the intermediate (XIV):



are obtained in form of white crystalline powder; m.p.= 129-130°C; chlorine content equal to 31.87% (theor. 32.13%).

500 cc of chloroform, 110.5 g of the intermediate (XIV) and 30.5 g of 2-hydroxyethylamine dissolved in 80 cc of water are fed in a 1 liter reactor equipped as in the preceding examples.

The mixture is heated to boiling and kept under reflux for 3 hours; thereafter, a solution consisting of 20 g of sodium hydroxide in 70 cc of water is fed within 2 hours.

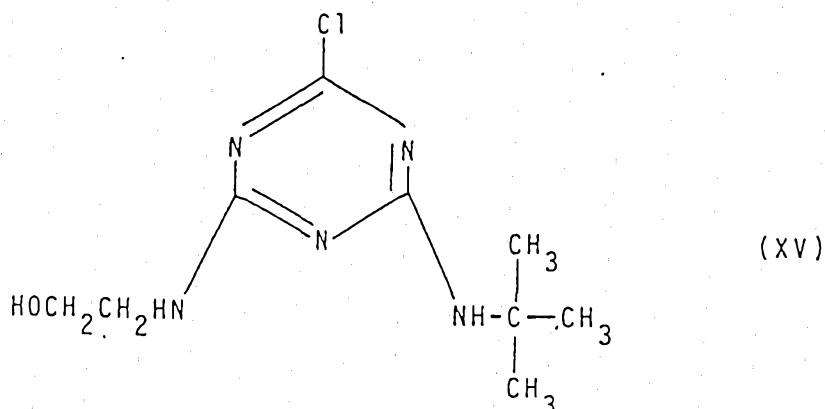
The mixture is kept for further 1 hour to boiling and then is cooled to room temperature by separating the organic phase.

The solvent is distilled off and the product which remains after the distillation is treated with 500 cc of water in the same 1 liter reactor.

The product is heated to 50-60°C until a good dispersion is obtained and thereafter the whole is cooled to room temperature and the product formed is separated by filtration.

The cake is washed with water and dried in oven at 80°C.

106.7 g of the intermediate (XV):



are obtained in form of white crystalline powder; m.p.=134-135°C; chlorine content equal to 14.32% (theor. 14.4%).

The structure of intermediates (XIV) and (XV) has been further confirmed by NMR analysis.

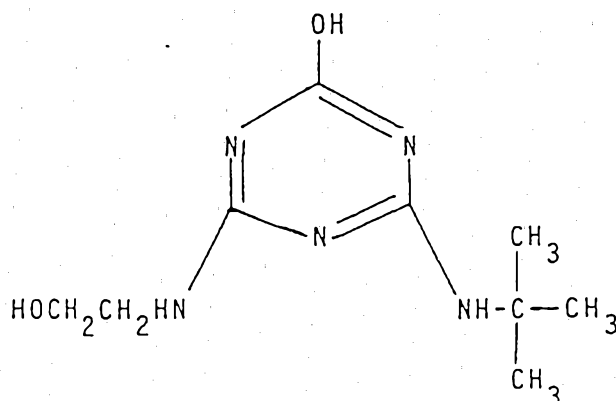
500 cc of water, 98.2 g of the intermediate (XV) and 20.5 g of 96% by weight sulfuric acid are fed in the same 1 liter reactor.

The reaction mass is heated to 85°C and is kept at this temperature for 2 hours.

Thereafter, 32 g of sodium hydroxide dissolved in 100 cc of water are fed within 30 minutes.

The whole is kept for further 30 minutes at 85°C and thereafter is cooled to room temperature; the product formed is filtered and washed on the filter.

By drying the cake in oven at 100°C, 83.2 g of the product:



are obtained in form of white crystalline powder; m.p. higher than 300°C.

EXAMPLE 6

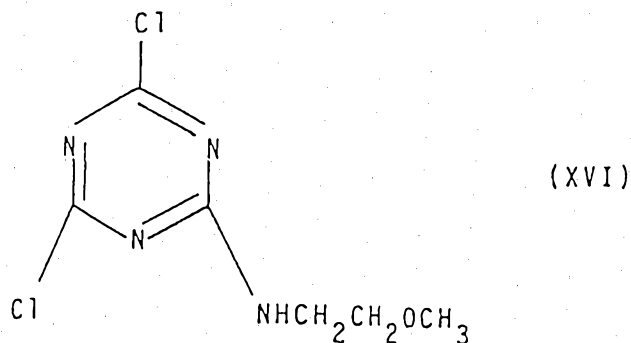
600 cc of water and 184.5 g of the cyanuric acid chloride are introduced in a 2 litres reactor, equipped as in example 1.

While cooling from the outside to 0-2°C, 75 g of 2-methoxyethylamine are fed within 1 hour and 30 minutes.

Subsequently, 40 g of sodium hydroxide dissolved in 250 cc of water are fed within 2 hours, always maintaining the temperature at 0-2°C.

The mass is kept under agitation for further 1 hour at the same temperature, then the product formed is separated by filtration and washed on the filter with water.

By drying in oven under vacuum at 60°C, 178.9 g of the intermediate (XVI):



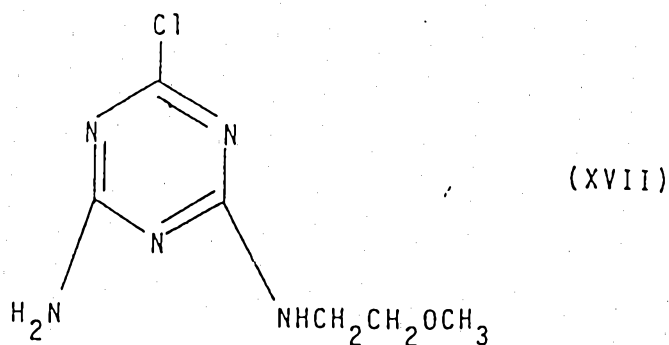
are obtained in form of white crystalline powder; m.p.=73-75°C; chlorine content equal to 31.68% (theor. 31.84%).

In a 1 liter reactor equipped as in example 1, there are introduced 85 g of a 30% by weight ammonia solution, 250 cc of water and 111.5 g of the intermediate (XVI).

The mixture is first heated to 40°C keeping this temperature for 4 hours, then to 55°C for 2 hours.

The mass is cooled to 10°C and the product formed is filtered and washed on the filter with water.

By drying the cake in oven at 100°C 98 g of the intermediate (XVII):



are obtained in form of white crystalline powder; m.p.=195-197°C; chlorine content equal to 17.21% (theor. 17.44%).

The structure of intermediates (XVI) and (XVII) has been further confirmed by IR spectroscopic analysis.

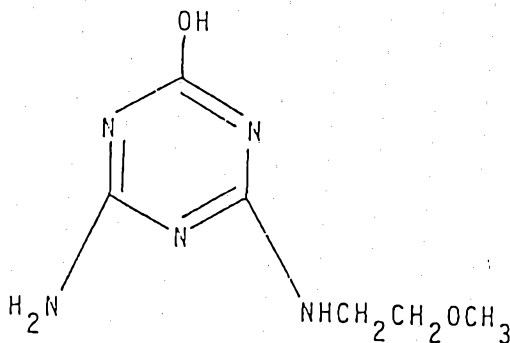
400 cc of water, 81.4 g of the intermediate (XVII) and 42.3 g of 37% by weight hydrochloric acid are introduced in the same 1 liter reactor.

The mixture is heated to 80°C and is kept at this temperature for 2 hours.

Thereafter, 44.2 g of sodium carbonate dissolved in 200 cc of water are added, always at 80°C.

The whole is then cooled to room temperature and the product formed is filtered and washed on the filter with water.

By drying the cake in oven at 100°C, 68.1 g of the product:



are obtained in form of white crystalline powder having a melting point higher than 300°C.

EXAMPLE 7

400 cc of methyl alcohol are introduced in a 1 liter reactor equipped as in the preceding examples and, while keeping the temperature at 15-20°C, 17.6 g of sodium hydroxide are added.

The mixture is kept under agitation until the solution is comple-

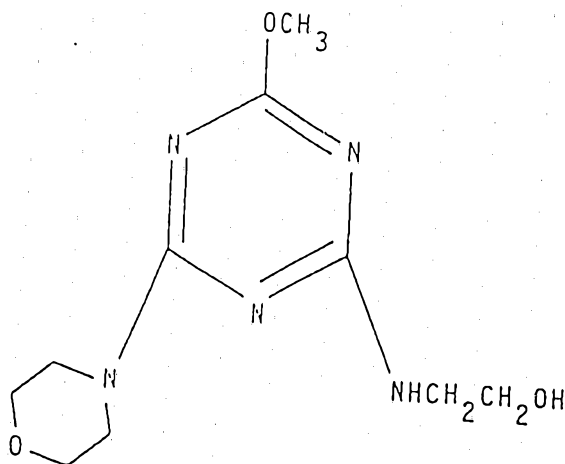
ted, thereafter 103.8 g of the intermediate (X) are introduced.

The mixture is heated to boiling and is kept under reflux for 4 hours.

Then the most part of the solvent (about 350 cc) are distilled off and the residue of the distillation is treated with 200 cc of water.

The product formed is filtered and washed on the filter with water.

By drying the cake in oven at 100°C 88.7 g of the product:

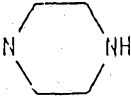


are obtained in form of white crystalline powder; m.p. = 136-138°C.

EXAMPLES 8-24

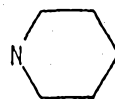
By working under conditions analogous to those described in examples from 1 to 7, product of general formula (I) listed in Table 2 are prepared.

TABLE 2

EX.	R	R_1-N-R_2	R_3-N-R_4	m.p.
No.				(°C)
8	H	$(CH_2)_3OH$	H $(CH_2)_3OH$	> 300
9	H	CH_2CH_2OH	H $CH_2-CH=CH_2$	> 300
10	H	$CH_2CH_2OCH_3$	H $CH_2CH_2OCH_3$	> 300
11	H		H	> 300
12	H	CH_2CH_2OH	CH ₃ CH_2CH_2OH	207-209
13	H	$(CH_2)_5OH$	H H	254-257
14	H			> 300
15	n-C ₄ H ₉		H	109-113
16	H	CH_2CHOH CH ₃	H CH_2CHOH CH ₃	> 300
17		CH_2CH_2OH	H CH_2CH_2OH	238-242

T A B L E . 2 (follows)

EX.	R	R_1 —N—	R_3 —N— R_4	m.p.
Nö.				(°C)
18	H	CH ₂ CH ₂ OH	CH ₂ CH ₂ OH	200-203
19	H	CH ₂ CH ₂ CH ₂ N <chem>C1CCOCC1</chem>	H	> 300
20	CH ₂ -CH=CH ₂	<chem>C1CCNCC1</chem>	H	149-152
21	H	CH ₂ CH ₂ OCH ₃	H	250-253
22	<chem>C1CCNCC1</chem>	CH ₂ CH ₂ OH	H	227-231
23	H	CH ₂ CH ₂ OCH=CH ₂	H	> 300
24	H	CH ₂ CH ₂ OH	H	> 300



Tables 3 and 4

Tests reported in the above specified tables relates to compositions containing products of general formula (I) prepared according to the preceding examples.

Specimens in form of little plates having 3 mm thickness have been prepared by molding the mixtures of granulated polymer and additives in a MOORE plate press, by working for 7 minutes at a pressure of 40 kg/cm².

On the thus obtained plates the self-extinguishing level has been determined by measuring the oxygen index (L.O.I. according to ASTM D-2863/77) in a Stanton Redcroft apparatus and applying the "Vertical Burning Test" which allows to classify the material at three levels 94V-0, 94V-1 and 94V-2 according to rules UL 94 (edited by "Underwriters Laboratories" USA).

In Table 3 values obtained using an isotactic polypropylene in flakes having a Melt Flow Index equal to 12 and a fraction insoluble in boiling n-heptane equal to 96% by weight are reported.

In table 4 values are reported obtained using a low density polyethylene in granules, having a Melt Flow Index equal to 7; a polystyrene in granules containing 5% by weight of butadiene rubber and having Melt Flow Index equal to 9; a thermoplastic polyurethane either polyester (ESTANE 54600^R by Goodrich) or polyether (ESTANE 58300^R by Goodrich) in granules having specific gravity equal to 1.19 and 1.10 g/cm³ respectively; an elastomeric ethylene-propylene copolymer having a propylene by weight percentage content equal to 45; an acrylo-

nitrile-butadiene-styrene terpolymer having specific gravity equal to 1.06 g/cm³, Melt Flow Index equal to 1.6 and containing about 40% of acrylonitrile and styrene and 20% of butadiene.

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T A B L 3

Example No.	Product Example No.	P A R T S Y W E I G H T				L.O.I. (ASTM D2863)	UL 94 3 mm
		Product	PP (1)	AO (2)	APP (1)		
25	1	7,4	73	1	18,6	33,8	VO
26	2	8,3	70	1	20,7	34,6	VO
27	3	16,0	75	1	8,0	29,5	V1
28	4	4,2	78	1	16,8	31,8	VO
29	5	6,9	75	1	17,1	31,4	V1
30	6	8,0	75	1	16,0	33,3	VO
31	7	8,3	70	1	20,7	34,7	VO
32	8	7,2	70	1	21,8	37,1	VO
33	9	8,3	70	1	20,7	33,8	VO
34	10	7,2	70	1	21,8	36,5	VO
35	11	6,9	75	1	17,1	30,8	VO
36	12	6,8	75	1	17,2	31,4	VO
37	13	9,7	70	1	19,4	32,1	V1
38	14	6,8	75	1	17,2	34,6	VO
39	15	8,3	70	1	20,7	31,8	V1
40	16	6,0	75	1	18,0	34,7	VO
41	17	8,3	70	1	20,7	32,3	V1
42	18	6,0	75	1	18,0	34,3	VO
43	19	12,0	75	1	12,0	31,3	VO
44	20	8,3	70	1	20,7	32,4	VO
45	21	6,8	75	1	17,2	32,7	VO
46	22	8,3	70	1	20,7	34,7	VO
47	23	8,0	75	1	16,0	33,6	VO
48	24	7,2	70	1	21,8	32,9	VO
49	3	8,0	75	1	16,0*	31,5	VO
50	6	6,0	75	1	18,0*	34,1	VO
51	4	6,0	75	1	18,0 ^③	30,5	VO
52	16	6,8	75	1	17,2 ^④	31,8	VO

(1) PP = polypropylene

APP = ammonium polyphosphate

Exolit 422[®] (Hoechst)

* APP = microincapsulated with melamine-formaldehyde resin

Exolit 462[®] (Hoechst)

(2) = antioxidant

Mixture consisting of two parts of dilaurylthiopropionate and
1 part of tetra[3-(3,5-di-terbutyl-4-hydroxyphenyl)propionate]
of pentaerythritol.

(3) monoammonium salt from 1-aminoethane-1,1-diphosphonic acid

(4) monammonium salt from 1-hydroxyethane-1,1-diphosphonic acid.

TABLE 4

Ex. No.	Supp. Polymeric (1)	Product Example No	PARTS BY WEIGHT				L.O.I. (ASTM-D2863)	UL94 3 mm
			Polymer	Product	A0 (2)	APP (1)		
53	LDPE	1	70	7,2	1	21,8	30,0	V1
54		4	65	8,5	1	25,5	35,7	V0
55		16	60	9,0	1	30,0	40,7	V0
56		18	65	8,5	1	25,5	32,1	V1
57	H	3	65	11,3	1	22,7	29,2	V1
58	I	4	65	13,6	1	20,4	32,7	V0
59	P	8	65	11,3	1	22,7	31,5	V1
60	S	16	65	8,5	1	25,5	30,5	V0
61		18	63	12,0	1	24,0	31,8	V0
62		4	64	8,8	1	26,2 ^③	29,8	V0
63	PU	1	70	7,2	1	21,8	33,8	V0
64	(e-	3	70	7,2	1	21,8	35,2	V0
65	ste-	4	70	8,3	1	20,7	36,1	V0
66	re)	10	70	7,2	1	21,8	34,4	V0
67		16	70	8,3	1	20,7	34,5	V0
68	PU etere	3	65	8,5	1	25,5	28,9	V0
69	PP/PE	1	65	8,5	1	25,5	30,4	V1
70		4	65	9,7	1	24,3	32,5	V0
71		16	65	8,5	1	25,5	30,8	V0
72	ABS	4	65	13,6	1	20,4	29,7	V0
73		16	65	13,6	1	20,4	28,5	V0

(1) APP = ammonium polyphosphate- Exolit 422^R (Hoechst)

LDPE = low density polyethylene

HIPS = polystyrene containing 5% of butadiene rubber

PU (ester) = polyurethane polyester

PU (ether) = polyurethane polyether

PP/PE = propylene-ethylene copolymer

ABS = acrylonitrile-butadiene-styrene terpolymer

(2) AO = antioxidant

Mixture consisting of 2 parts of dilaurylthiopropionate and

1 part of tetra [3-(3, 5-di-terbutyl-4-hydroxyphenyl)pro_pionate] of pentaerythritol

(3) monoammonium salt from phenylphosphonic acid.

EXAMPLE 74 (COMPARISON EXAMPLE)

By working according to the modalities used in examples from 25 to 52, but using as nitrogenous compound 2,4-diamino-6-hydroxy-1,3,5-triazine, the following composition is prepared:

Polypropylene:	72	parts by weight
Antioxidant	1	part by weight
Ammonium polyphosphate	19.3	parts by weight
2,4-diamino-6-hydroxy-1,3,5-triazine	7.7	parts by weight

Using the above specified compositions, specimens have been prepared which have been subjected to self-extinguishing tests according to the previously described modalities.

The following results have been obtained:

L.O.I. =23.8

UL 94 (3 mm): Class B (the specimen burns).

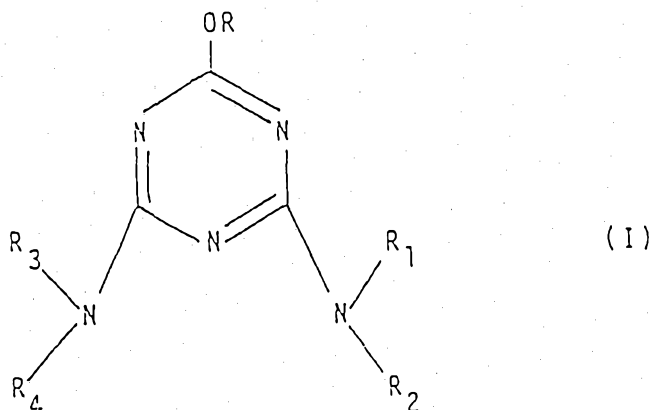
THE CLAIMS DEFINING THE INVENTION ARE AS FOLLOWS:

1. Self-extinguishing polymeric compositions consisting essentially of:

5 a) from 91 to 40 parts by weight of a thermoplastic polymer or of a polymer showing elastomeric properties;

b) from 6 to 33 parts by weight of ammonium phosphates, ammonium phosphonates, amine phosphates, amine phosphonates or mixtures thereof;

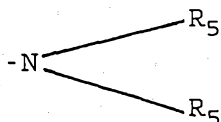
10 c) from 3 to 27 parts by weight of one or more compounds derived from the 2,4-diamino-6-hydroxy-1,3,5-triazine, having the general formula (I):



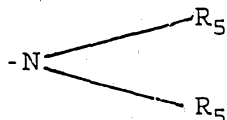
wherein:

(1) R is (a) H (b) $-\text{[C}_n\text{H}_{2n}\text{]}-\text{X}$,

15 wherein n is an integer from 1 to 8 and X=H; CN; -O-(C₁-C₄)-alkyl; -O-(C₂-C₄)-alkenyl; (C₆-C₁₂)-cycloalkyl or alkylcycloalkyl; -O-(C₆-C₁₂)-aryl;



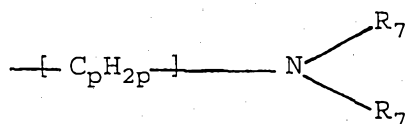
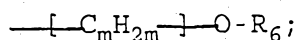
20 wherein radicals R₅, are the same or different and are (C₁-C₄)-alkyl, (C₃-C₄)-alkenyl; or the group:



is replaced by a N-containing heterocyclic radical bound to the alkyl chain through the nitrogen atom of said N-containing heterocyclic radical, said N-containing heterocyclic radical optionally containing another heteroatom selected from O, S and N;

(c) (C₃-C₆)-alkenyl, (d) (C₆-C₁₂)-cycloalkyl or (e) alkylcycloalkyl; (f) (C₆-C₁₂)-aryl or aralkyl;

(2) at least one of radicals from R₁ to R₄ is:



with:

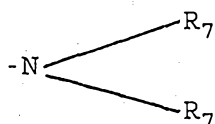
m = integer from 2 to 8;

p = integer from 2 to 6;

R₆=H; (C₁-C₈)-alkyl; (C₂-C₆)-alkenyl; [C_qH_{2q}]-O-R₈,

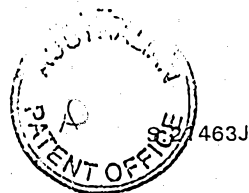
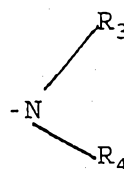
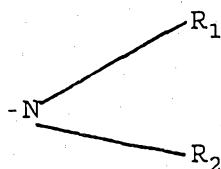
wherein q is an integer from 1 to 4 and R₈ is hydrogen or (C₁-C₄)-alkyl; (C₆-C₁₂)-cycloalkyl or alkylcycloalkyl; radicals R₇ are the same or different and are H; (C₁-C₈)-alkyl; (C₂-C₆)-alkenyl; (C₁-C₄)-hydroxyalkyl; (C₆-C₁₂)-cycloalkyl or alkylcycloalkyl;

or the group:



is replaced by a N-containing heterocyclic radical bound to the alkyl chain through the nitrogen atom of said N-containing heterocyclic radical, said N-containing heterocyclic radical optionally containing another heteroatom selected from O, S, and N;

(3) or in the general formula (I) at least one of groups:



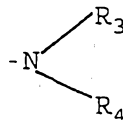
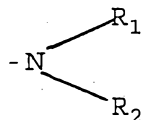
is replaced by a N-containing heterocyclic radical bound to the triazinic ring through the nitrogen atom of said

N-containing heterocyclic radical, said N-containing
5 heterocyclic radical optionally containing another heteroatom selected from O, S and N

(4) the remaining groups from R_1 to R_4 , the same or different, have the above specified meaning or are:

H; (C_1-C_{18}) -alkyl; (C_2-C_8) -alkenyl; (C_6-C_{16})
10 cycloalkyl or alkylcycloalkyl, optionally substituted by a hydroxyl or (C_1-C_4) -hydroxyalkyl function, wherein the parts by weight of components a), b) and c) add up to 100 parts by weight.

2. Self-extinguishing polymeric compositions
15 according to claim 1, wherein at least one of groups:

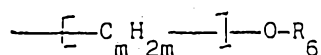


in the general formula (I) is replaced by a heterocyclic

radical selected from:

aziridine; pyrrolidine; piperidine; morpholine; thiomorpholine; piperazine; 4-methylpiperazine; 4-ethylpiperazine; 2-methylpiperazine; 2,5-dimethylpiperazine; 2,3,5,6-tetramethylpiperazine; 2,2,5,5-tetramethylpiperazine; 2-ethylpiperazine; 2,5-diethylpiperazine.

3. Self-extinguishing polymeric compositions according to claims 1 or 2, wherein at least one of radicals from R_1 to R_4 in the general formula (I) is replaced by a group:

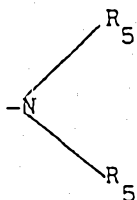


wherein:

m is an integer from 2 to 4 and

R_6 is hydrogen or (C_1-C_4) -alkyl.

4. Self-extinguishing polymeric compositions according to any of claims from 1 to 3, wherein the group:

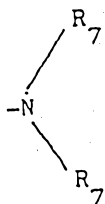


is replaced by a heterocyclic radical selected from:

pyrrolidine; piperidine; morpholine; thiomorpholine; piperazine; 4-methylpiperazine; 4-ethylpiperazine.

5. Self-extinguishing polymeric compositions according to any of claims from 1 to 4, wherein the group:



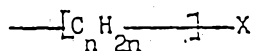


is replaced by a heterocyclic radical selected from:

aziridine; pyrrolidine; piperidine; morpholine; thiomorpholine; piperazine; 4-methylpiperazine; 4-ethylpiperazine.

6. Self-extinguishing polymeric compositions according to claims 1, 2, 3 or 5 wherein R, in the general formula (I), is equal to hydrogen.

7. Self-extinguishing polymeric compositions according to claim 1, 2, 3 or 5 wherein R in the general formula (I) is replaced by a group:



wherein:

n is an integer from 1 to 4 and

X is hydrogen.

8. Self-extinguishing polymeric compositions according to any of claims from 1 to 7, wherein the ammonium phosphate or phosphates (b) have the general formula $(\text{NH})_{4n+2} \text{P} \text{O}_{3n+1}$ wherein n is an integer equal to or higher than 2.

9. Self-extinguishing polymeric compositions according to any of claims from 1 to 7, wherein the ammonium phosphate or phosphates (b) have the general formula $(\text{NH}_4 \text{PO}_3)_n$ wherein n is an integer from 50 to 500.

10. Self-extinguishing polymeric compositions according to any of



claims from 1 to 7, wherein the amine phosphate or phosphates

(b) are selected from dimethylammonium or diethylammonium phosphate; ethylenediamine phosphate; melamine ortho- or pyrophosphate.

11. Self-extinguishing polymeric compositions according to any of claims from 1 to 7 wherein the ammonium phosphonate or phosphonates (b) are those mono or polysubstituted and selected among salts derived from mono or polyphosphonic acids.
12. Self-extinguishing polymeric compositions according to any of claims from 1 to 11, wherein the polymer (a) is selected among polymers and copolymers of olefins of the general formula $R-CH=CH_2$ wherein R is a hydrogen atom or a (C_1-C_8) -alkyl or -aryl radical; acrylonitrile/butadiene/styrene copolymers; styrene/acrylonitrile copolymers; polyurethane; polyethyleneterephthalate; polybutyleneterephthalate; polyamides.
13. Self-extinguishing polymeric compositions according to claim 12 wherein polymers and copolymers of olefins are selected from:
 - a) isotactic or prevailing isotactic polypropylene;
 - b) HDPE, LLDPE, LDPE polyethylene;
 - c) propylene crystalline copolymers with lower proportions of ethylene and/or other alpha-olefins, such as 1-butene; 1-hexene; 1-octene; 4-methyl-1-pentene;
 - d) heterophasic compositions comprising (A) a homopolymeric fraction of propylene or one of copolymers indicated under (c) and (B) a copolymeric fraction consisting of elastomeric copolymers

of ethylene with an alpha-olefin, optionally containing lower proportions of a diene, wherein the alpha-olefin is preferably selected from propylene and 1-butene;

5 e) elastomeric copolymers of ethylene with alpha-olefins containing lower proportions of a diene.

14. Molded manufactured articles, obtained from the compositions of any one of the preceding claims.

10 15. Self extinguishing polymeric compositions as defined in claim 1 and incorporating a compound of general formula (I) as herein described with reference to any one of Examples 1 to 24.

Dated this 18th day of February 1993

MINISTERO DELL'UNIVERSITA' E DELLA
RICERCA SCIENTIFICA E TECNOLOGICA

15 By their Patent Attorney
GRIFFITH HACK & CO

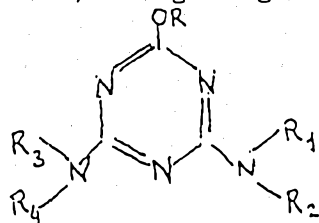


"SELF-EXTINGUISHING POLYMERIC COMPOSITIONS"

Abstract

Polymeric compositions endowed with high characteristics of self-extinguishing to flame, based on thermoplastic polymers or having elastomeric properties, especially olefin polymers and copolymers, comprising:

- a) from 91 to 40 parts by weight of a thermoplastic polymer or having elastomeric properties;
- b) from 6 to 33 parts by weight of one or more phosphates and/or phosphonates of ammonium or of an amine;
- c) from 3 to 27 parts by weight of one or more compounds derived from 2,4-diamino-6-hydroxy-1,3,5-triazine, having the general formula (I):



(I)