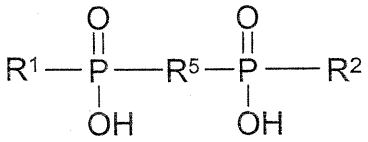
It is thus important to minimize the thermal expansion of test specimens in order to obtain a good, dimensionally stable product (finished printed circuit board).

It is an object of the present invention to modify polymers for prepregs, printed circuit boards and laminates such that they are subject only to very low thermal expansion – if any at all – and dimensional stability is fulfilled.

~~This object is achieved by~~In one aspect, there is provided a mixture~~s~~ of at least one diphosphinic acid of the formula (I)



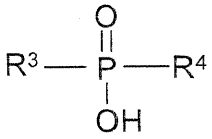
(I)

in which

R^1, R^2 are the same or different and are each independently H, C_1-C_{18}-alkyl, C_2-C_{18}-alkenyl, C_6-C_{18}-aryl, C_7-C_{18}-alkylaryl,

R^5 is C_1-C_{18}-alkylene, C_2-C_{18}-alkenylene, C_6-C_{18}-arylene, C_7-C_{18}-alkylarylene

with at least one dialkylphosphinic acid of the formula (II)



(II)

in which

R^3, R^4 are the same or different and are each independently C_1-C_{18}-alkyl, C_2-C_{18}-alkenyl, C_6-C_{18}-aryl and/or C_7-C_{18}-alkylaryl.

Preferably, R^1, R^2 are the same or different and are each H, methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, tert-butyl, n-pentyl, isopentyl, n-hexyl, isohexyl and/or phenyl; R^3, R^4 are the same or different and, independently of R^1 and R^2, are each methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, tert-butyl, n-pentyl,

phosphorus compounds such as phosphinic acids and salts thereof, phosphonic acids and salts thereof and/or phosphine oxides, phosphazenes and/or piperazine (pyro)phosphates;
carbodiimides, piperazines, (poly)isocyanates, styrene-acrylic polymers and/or carbonylbiscaprolactam;
nitrogen compounds from the group of oligomeric esters of tris(hydroxyethyl) isocyanurate with aromatic polycarboxylic acids, or benzoguanamine, acetoguanamine, tris(hydroxyethyl) isocyanurate, allantoin, glycoluril, cyanurates, cyanurate-epoxide compounds, urea cyanurate, dicyanamide, guanidine, guanidine phosphate and/or sulfate.

The mixtures preferably comprise 99 to 1% by weight of mixture of at least one diphosphinic acid of the formula (I) and at least one dialkylphosphinic acid of the formula (II) as claimed in at least one of claims 1 to 11 and 1 to 99% by weight of synergist.

~~The invention also~~Another aspect relates to a process for preparing the mixtures as ~~claimed in at least one of claims 1 to 11~~disclosed herein, which comprises reacting a phosphinic acid source with an alkyne in the presence of an initiator.

Preferably, the phosphinic acid source is ethylphosphinic acid and the alkyne is acetylene, methylacetylene, 1-butyne, 1-hexyne, 2-hexyne, 1-octyne, 4-octyne, 1-butyn-4-ol, 2-butyn-1-ol, 3-butyn-1-ol, 5-hexyn-1-ol, 1-octyn-3-ol, 1-pentyne, phenylacetylene, trimethylsilylacetylene and/or diphenylacetylene and the initiator is a free-radical initiator having a nitrogen-nitrogen or an oxygen-oxygen bond and the reaction temperature is between 50 and 150°C.

The free-radical initiator is preferably 2,2'-azobis(2-amidinopropane) dihydrochloride, 2,2'-azobis(N,N'-dimethyleneisobutyramidine) dihydrochloride, azobis(isobutyronitrile), 4,4'-azobis(4-cyanopentanoic acid) and/or 2,2'-azobis(2-methylbutyronitrile) or hydrogen peroxide, ammonium peroxodisulfate, potassium peroxodisulfate, dibenzoyl peroxide, di-tert-butyl peroxide, peracetic acid,

diisobutyryl peroxide, cumene peroxyneodecanoate, tert-butyl peroxyneodecanoate, tert-butyl peroxypivalate, tert-amyl peroxypivalate, dipropyl peroxydicarbonate, dibutyl peroxydicarbonate, dimyristyl peroxydicarbonate, dilauroyl peroxide, 1,1,3,3-tetramethylbutyl peroxy-2-ethylhexanoate, tert-amyl peroxy-2-ethylhexylcarbonate, tert-butyl peroxyisobutyrate, 1,1-di(tert-butylperoxy)cyclohexane, tert-butyl peroxybenzoate, tert-butyl peroxyacetate, tert-butyl peroxydiethylacetate, tert-butyl peroxyisopropylcarbonate, 2,2-di(tert-butylperoxy)butane, tert-amyl hydroperoxide and/or 2,5-dimethyl-2,5-di(tert-butylperoxy)hexane.

The solvent preferably comprises straight-chain or branched alkanes, alkyl-substituted aromatic solvents, water-immiscible or only partly water-miscible alcohols or ethers, water and/or acetic acid.

The alcohol is preferably methanol, propanol, i-butanol and/or n-butanol or comprises mixtures of these alcohols with water.

~~The invention also~~Another aspect relates to the use of mixtures of at least one diphosphinic acid of the formula (I) and at least one dialkylphosphinic acid of the formula (II) as ~~claimed in at least one of claims 1 to 11~~disclosed herein as an intermediate for further syntheses, as a binder, as a crosslinker or accelerator in the curing of epoxy resins, polyurethanes and unsaturated polyester resins, as polymer stabilizers, as crop protection compositions, as sequestrants, as a mineral oil additive, as an anticorrosive, in washing and cleaning composition applications and in electronics applications.

~~The invention additionally~~A further aspect relates to the use of mixtures of at least one diphosphinic acid of the formula (I) and at least one dialkylphosphinic acid of the formula (II) as disclosed herein ~~claimed in at least one of claims 1 to 13~~ as a flame retardant, especially as a flame retardant for clearcoats and intumescent coatings, as a flame retardant for wood and other cellulosic products, as a reactive and/or nonreactive flame retardant for polymers, for production of flame-retardant polymer molding compositions, for production of flame-retardant polymer moldings

and/or for rendering polyester and pure and blended cellulose fabrics flame-retardant by impregnation, and as a synergist.

~~The invention also~~Another aspect relates to a flame-retardant thermoplastic or thermoset polymer molding composition and to polymer moldings, films, filaments and fibers comprising 0.5 to ~~45~~50% by weight of mixtures as disclosed herein~~claimed in at least one of claims 1 to 13~~, 55 to 99.5% by weight of thermoplastic or thermoset polymer or mixtures thereof, 0 to 55% by weight of additives and 0 to 55% by weight of filler or reinforcing materials, where the sum of the components is 100% by weight.

~~The invention finally~~ Another aspect ~~also~~ relates to a flame-retardant thermoplastic or thermoset polymer molding composition and to polymer moldings, films, filaments and fibers comprising 2 to 30% by weight of mixtures as disclosed herein ~~claimed in at least one of claims 1 to 13~~, 60 to 94% by weight of thermoplastic or thermoset polymer or mixtures thereof, 2 to 30% by weight of additives and 2 to 30% by weight of filler or reinforcing materials, where the sum of the components is 100% by weight.

Preferred mixtures of diphosphinic acid of the formula (I) and dialkylphosphinic acid of the formula (II) are, for example:

ethylene-1,2-bis(ethylphosphinic acid) and diethylphosphinic acid,
ethylene-1,2-bis(ethylphosphinic acid) and butylethylphosphinic acid,
ethylene-1,2-bis(ethylphosphinic acid) and butylbutylphosphinic acid,
ethylene-1,2-bis(ethylphosphinic acid) and hexylethylphosphinic acid,
ethylene-1,2-bis(ethylphosphinic acid) and octylethylphosphinic acid,
ethylene-1,2-bis(ethylphosphinic acid) and hexylbutylphosphinic acid,
ethylene-1,2-bis(butylphosphinic acid) and diethylphosphinic acid,
ethylene-1,2-bis(butylphosphinic acid) and butylethylphosphinic acid,
ethylene-1,2-bis(butylphosphinic acid) and butylbutylphosphinic acid,
ethylene-1,2-bis(butylphosphinic acid) and hexylethylphosphinic acid,
ethylene-1,2-bis(butylphosphinic acid) and octylethylphosphinic acid,
ethylene-1,2-bis(butylphosphinic acid) and hexylbutylphosphinic acid,
butylene-1,2-bis(ethylphosphinic acid) and diethylphosphinic acid,