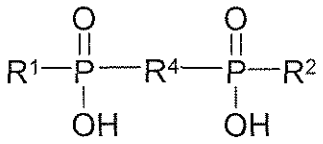
It is an object of the present invention to modify polymers for prepregs, printed circuit boards and laminates such that they are subject only to very low thermal expansion – if any at all – and the necessary dimensional stability is fulfilled.

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



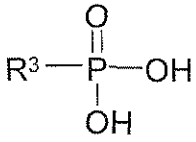
(I)

in which

R^1, R^2 are each H, C_1-C_{18}-alkyl, C_2-C_{18}-alkenyl, C_6-C_{18}-aryl, C_7-C_{18}-alkylaryl

R^4 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 alkylphosphonic acid of the formula (II)



(II)

in which

R^3 is H, 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 and R^3 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, and R^4 is ethylene, butylene, hexylene or octylene.

The mixtures preferably comprise 0.1 to 99.9% by weight of diphosphinic acid of the formula (I) and 99.9 to 0.1% by weight of alkylphosphonic acid of the formula (II).

The mixtures more preferably comprise 40 to 99.9% by weight of diphosphinic acid of the formula (I) and 60 to 0.1% by weight of alkylphosphonic acid of the formula (II). Preference is likewise given to mixtures comprising 60 to 99.9% by weight of

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

The initiator is preferably a free-radical initiator having a nitrogen-nitrogen or an oxygen-oxygen bond.

The free-radical initiator is more 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 reaction temperature is preferably 50 to 150°C.

~~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 alkylphosphonic 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~~Another aspect relates to the use of mixtures of at least one diphosphinic acid of the formula (I) and at least one alkylphosphonic acid of the formula (II) as ~~claimed in at least one of claims 1 to 13~~disclosed herein 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 encompasses~~Another aspect relates to flame-retardant thermoplastic or thermoset polymer molding compositions, moldings, films, filaments and fibers comprising 0.5 to 99.5% by weight of mixtures as ~~claimed in at least one of claims 1 to 13~~disclosed herein, 0.5 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 relates to flame-retardant thermoplastic or thermoset polymer molding compositions, moldings, films, filaments and fibers comprising 1 to 30% by weight of mixtures as ~~claimed in at least one of claims 1 to 13~~disclosed herein, 10 to 95% 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.

Preferably, R^1 and 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 is (independently of R^1 and R^2) preferably methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, tert-butyl, n-pentyl, isopentyl, n-hexyl, isohexyl and/or phenyl, and R^4 is ethylene, butylene, hexylene or octylene; this means the C_2, C_4, C_6 or C_8 group which connects the two phosphorus atoms.

Preference is also given to mixtures comprising 98 to 99.9% by weight of ethylene-1,2-bis(ethylphosphinic acid) and 2 to 0.1% by weight of ethylphosphonic acid.

Preferred two-component mixtures of at least one diphosphinic acid of the formula (I) and at least one alkylphosphonic acid of the formula (II) are composed of
ethylene-1,2-bis(ethylphosphinic acid) and ethylphosphonic acid,
ethylene-1,2-bis(ethylphosphinic acid) and propylphosphonic acid,
ethylene-1,2-bis(ethylphosphinic acid) and butylphosphonic acid,
ethylene-1,2-bis(ethylphosphinic acid) and pentylphosphonic acid,
ethylene-1,2-bis(ethylphosphinic acid) and hexylphosphonic acid,
ethylene-1,2-bis(propylphosphinic acid) and ethylphosphonic acid,
ethylene-1,2-bis(propylphosphinic acid) and propylphosphonic acid,
ethylene-1,2-bis(propylphosphinic acid) and butylphosphonic acid,
ethylene-1,2-bis(propylphosphinic acid) and pentylphosphonic acid,
ethylene-1,2-bis(propylphosphinic acid) and hexylphosphonic acid,