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(54) Title: FLAME RETARDANT RESINOUS COMPOSITIONS AND PROCESS

(57) Abstract: Disclosed is a flame-retardant composition comprising (i) 40-66 wt.% alkenyl aromatic resin, (ii) 9-33 wt.% ammonium polyphosphate and (iii) 14-40 wt.% cellulosic material, wherein all weights are based on the total weight of the composition and wherein ammonium polyphosphate and cellulosic material are present in a weight % ratio effective to provide molded articles exhibiting at least V-1 flame rating as determined according to the UL-94 protocol. A process to prepare the composition and articles comprising a composition of the invention and/or made by the process of the invention described herein are also disclosed.

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FLAME RETARDANT RESINOUS COMPOSITIONS AND PROCESS

BACKGROUND

[0001] The present invention relates to flame retardant resinous compositions comprising an alkenyl aromatic resin and a cellulosic material.

[0002] Flame retardant resinous compositions comprising an alkenyl aromatic resin, such as a styrenic resin, typically comprise a halogen-containing flame retardant additive. In order to minimize environmental, health and safety (EHS) issues, there is a great market need to develop flame retardant alkenyl aromatic resin compositions containing non-halogen flame retardant additives. Such compositions are known as eco-friendly flame retardant compositions. Typically, it has not been possible to develop flame retardant alkenyl aromatic resin compositions without halogen-containing additives with needed flammability rating while maintaining good mechanical properties and desirable processing characteristics. Hence, there is a need for eco-friendly flame retardant alkenyl aromatic resin compositions with suitable flame retardant properties which compositions also possess an attractive balance of mechanical properties.

BRIEF DESCRIPTION

[0003] The present inventors have discovered eco-friendly flame retardant alkenyl aromatic resin compositions which have flame retardant properties in combination with an attractive balance of mechanical properties. Articles made from the compositions of the present invention often exhibit V-1 flame rating or better as determined according to the UL-94 protocol. The articles are useful in applications requiring flame resistance, and particularly in applications requiring halogen-free (eco-friendly) compositions for flame resistance. This invention provides a unique solution for eco-friendly flame retardant polymer products using cost effective additives.

[0004] In one embodiment the present invention comprises a resinous, flame-retardant composition comprising (i) 40-66 wt.% alkenyl aromatic resin, (ii) 9-33 wt.% ammonium polyphosphate and (iii) 14-40 wt.% cellulosic material, wherein all weights are based on the total weight of the composition and wherein ammonium polyphosphate and cellulosic material are present in a weight % ratio effective to provide molded articles exhibiting at least V-1 flame rating as determined according to the UL-94 protocol.

[0005] In another embodiment the present invention comprises a resinous, flame-retardant composition comprising (i) 40-66 wt.% alkenyl aromatic resin, (ii) 9-33 wt.% ammonium polyphosphate and (iii) 14-40 wt.% cellulosic material, wherein ammonium

polyphosphate and cellulosic material are present in a weight % ratio in a range of 1:2 to 2:1 effective to provide molded articles exhibiting at least V-1 flame rating as determined according to the UL-94 protocol, and wherein the composition is prepared by an extrusion process wherein all or a portion of the ammonium polyphosphate is fed to the extruder down-stream from the alkenyl aromatic resin and the cellulosic material.

[0006] In still another embodiment the present invention comprises an extrusion process for preparing a resinous, flame-retardant composition comprising (i) 40-66 wt.% alkenyl aromatic resin, (ii) 9-33 wt.% ammonium polyphosphate and (iii) 14-40 wt.% cellulosic material, wherein ammonium polyphosphate and cellulosic material are present in a weight % ratio in a range of 1:2 to 2:1 effective to provide molded articles exhibiting at least V-1 flame rating as determined according to the UL-94 protocol, which process comprises the step of adding at least a portion of the ammonium polyphosphate to the extruder down-stream from the ABS and the cellulosic material. Articles comprising a composition of the invention and/or made by the process described are also disclosed. Various other features, aspects, and advantages of the present invention will become more apparent with reference to the following description and appended claims.

DETAILED DESCRIPTION

[0007] In the following specification and the claims which follow, reference will be made to a number of terms which shall be defined to have the following meanings. The singular forms "a", "an" and "the" include plural referents unless the context clearly dictates otherwise. The terminology “monoethylenically unsaturated“ means having a single site of ethylenic unsaturation per molecule. The terminology “polyethylenically unsaturated” means having two or more sites of ethylenic unsaturation per molecule. The terminology “(meth)acrylate” refers collectively to acrylate and methacrylate; for example, the term “(meth)acrylate monomers” refers collectively to acrylate monomers and methacrylate monomers. The term “(meth)acrylamide” refers collectively to acrylamides and methacrylamides.

[0008] The term "alkyl" as used in the various embodiments of the present invention is intended to designate linear alkyl, branched alkyl, aralkyl, cycloalkyl, bicycloalkyl, tricycloalkyl and polycycloalkyl radicals containing carbon and hydrogen atoms, and optionally containing atoms in addition to carbon and hydrogen, for example atoms selected from Groups 15, 16 and 17 of the Periodic Table. Alkyl groups may be saturated or unsaturated, and may comprise, for example, alkenyl, vinyl or allyl. The term “alkyl” also encompasses that alkyl portion of alkoxide groups. In various embodiments normal and

branched alkyl radicals are those containing from 1 to about 32 carbon atoms, and include as illustrative non-limiting examples C_1-C_{32} alkyl (optionally substituted with one or more groups selected from C_1-C_{32} alkyl, C_3-C_{15} cycloalkyl or aryl); and C_3-C_{15} cycloalkyl optionally substituted with one or more groups selected from C_1-C_{32} alkyl. Some particular illustrative examples comprise methyl, ethyl, n-propyl, isopropyl, n-butyl, sec-butyl, tertiary-butyl, pentyl, neopentyl, hexyl, heptyl, octyl, nonyl, decyl, undecyl and dodecyl. Some illustrative non-limiting examples of cycloalkyl and bicycloalkyl radicals include cyclobutyl, cyclopentyl, cyclohexyl, methylcyclohexyl, cycloheptyl, bicycloheptyl and adamantyl. In various embodiments aralkyl radicals are those containing from 7 to about 14 carbon atoms; these include, but are not limited to, benzyl, phenylbutyl, phenylpropyl, and phenylethyl. The term "aryl" as used in the various embodiments of the present invention is intended to designate substituted or unsubstituted aryl radicals containing from 6 to 20 ring carbon atoms. Some illustrative non-limiting examples of these aryl radicals include C_6-C_{20} aryl optionally substituted with one or more groups selected from C_1-C_{32} alkyl, C_3-C_{15} cycloalkyl, aryl, and functional groups comprising atoms selected from Groups 15, 16 and 17 of the Periodic Table. Some particular illustrative examples of aryl radicals comprise substituted or unsubstituted phenyl, biphenyl, tolyl, naphthyl and binaphthyl.

[0009] Compositions in embodiments of the present invention comprise at least one alkenyl aromatic resin. There is no particular limitation on the alkenyl aromatic resin which in some embodiments may comprise one or more homopolymers, copolymers, core-shell resins or rubber-modified resins. In some particular embodiments alkenyl aromatic resins comprise at least one polymeric material with structural units derived from styrene. In other particular embodiments alkenyl aromatic resins are selected from the group consisting of polystyrene, expandable polystyrene, acrylonitrile-butadiene-styrene copolymer (ABS), rubber-modified polystyrene, high-impact polystyrene (HIPS), styrene-acrylonitrile copolymer (SAN), alkyl methacrylate-styrene-acrylonitrile copolymer, methyl methacrylate-styrene-acrylonitrile copolymer, styrene-butadiene copolymer, styrene-maleic anhydride copolymer, styrene-methyl acrylate copolymer, styrene-methyl methacrylate copolymer, alpha-methylstyrene/acrylonitrile copolymer, alpha-methylstyrene/styrene/acrylonitrile copolymer, polychlorostyrene, polyvinyltoluene, and the like and mixtures thereof.

[0010] In some embodiments compositions of the present invention comprise at least one rubber modified thermoplastic resin comprising a discontinuous elastomeric phase dispersed in a rigid thermoplastic phase, wherein at least a portion of the rigid thermoplastic phase is grafted to the elastomeric phase. The rubber modified thermoplastic resin employs at

least one rubber substrate for grafting. The rubber substrate comprises the elastomeric phase of the composition. There is no particular limitation on the rubber substrate provided it is susceptible to grafting by at least a portion of a graftable monomer. In some embodiments suitable rubber substrates comprise butyl acrylate rubber, dimethyl siloxane/butyl acrylate rubber, or silicone/butyl acrylate composite rubber; polyolefin rubbers such as ethylene-propylene rubber or ethylene-propylene-diene (EPDM) rubber; diene-derived rubbers; or silicone rubber polymers such as polymethylsiloxane rubber. The rubber substrate typically has a glass transition temperature, Tg, in one embodiment less than or equal to 25°C, in another embodiment below about 0°C, in another embodiment below about minus 20°C, and in still another embodiment below about minus 30°C. As referred to herein, the Tg of a polymer is the T value of polymer as measured by differential scanning calorimetry (DSC; heating rate 20°C/minute, with the Tg value being determined at the inflection point).

[0011] In one embodiment the rubber substrate comprises a polymer having structural units derived from one or more unsaturated monomers selected from conjugated diene monomers and non-conjugated diene monomers. Suitable conjugated diene monomers include, but are not limited to, 1,3-butadiene, isoprene, 1,3-heptadiene, methyl-1,3-pentadiene, 2,3-dimethylbutadiene, 2-ethyl-1,3-pentadiene, 1,3-hexadiene, 2,4-hexadiene, dichlorobutadiene, bromobutadiene and dibromobutadiene as well as mixtures of conjugated diene monomers. In a particular embodiment the conjugated diene monomer is 1,3-butadiene. Suitable non-conjugated diene monomers include, but are not limited to, ethylidene norbornene, dicyclopentadiene, hexadiene and phenyl norbornene.

[0012] In some embodiments the rubber substrate may optionally comprise structural units derived from minor amounts of other unsaturated monomers, for example up to about 30 percent by weight ("wt.%") of structural units derived from one or more monomers selected from (C_2-C_8)olefin monomers, alkenyl aromatic monomers and monoethylenically unsaturated nitrile monomers. As used herein, the term "(C_2-C_8)olefin monomers" means a compound having from 2 to 8 carbon atoms per molecule and having a single site of ethylenic unsaturation per molecule. Suitable (C_2-C_8)olefin monomers include, e.g., ethylene, propene, 1-butene, 1-pentene, heptene. In other particular embodiments the rubber substrate may optionally include up to about 25 wt.% of structural units derived from one or more monomers selected from (meth)acrylate monomers, alkenyl aromatic monomers and monoethylenically unsaturated nitrile monomers. Suitable copolymerizable (meth)acrylate monomers include, but are not limited to, C_1-C_{12} aryl or haloaryl substituted acrylate, C_1-C_{12} aryl or haloaryl substituted methacrylate, or mixtures thereof; monoethylenically unsaturated

carboxylic acids, such as, for example, acrylic acid, methacrylic acid and itaconic acid; glycidyl (meth)acrylate, hydroxy alkyl (meth)acrylate, hydroxy(C_1-C_{12})alkyl (meth)acrylate, such as, for example, hydroxyethyl methacrylate; (C_4-C_{12})cycloalkyl (meth)acrylate monomers, such as, for example, cyclohexyl methacrylate; (meth)acrylamide monomers, such as, for example, acrylamide, methacrylamide and N-substituted-acrylamide or N-substituted-methacrylamides; maleimide monomers, such as, for example, maleimide, N-alkyl maleimides, N-aryl maleimides, N-phenyl maleimide, and haloaryl substituted maleimides; maleic anhydride; methyl vinyl ether, ethyl vinyl ether, and vinyl esters, such as, for example, vinyl acetate and vinyl propionate. Suitable alkenyl aromatic monomers include, but are not limited to, vinyl aromatic monomers, such as, for example, styrene and substituted styrenes having one or more alkyl, alkoxy, hydroxy or halo substituent groups attached to the aromatic ring, including, but not limited to, alpha-methyl styrene, p-methyl styrene, 3,5-diethylstyrene, 4-n-propylstyrene, 4-isopropylstyrene, vinyl toluene, alpha-methyl vinyl toluene, vinyl xylene, trimethyl styrene, butyl styrene, t-butyl styrene, chlorostyrene, alpha-chlorostyrene, dichlorostyrene, tetrachlorostyrene, bromostyrene, alpha-bromostyrene, dibromostyrene, p-hydroxystyrene, p-acetoxystyrene, methoxystyrene and vinyl-substituted condensed aromatic ring structures, such as, for example, vinyl naphthalene, vinyl anthracene, as well as mixtures of vinyl aromatic monomers and monoethylenically unsaturated nitrile monomers such as, for example, acrylonitrile, ethacrylonitrile, methacrylonitrile, alpha-bromoacrylonitrile and alpha-chloro acrylonitrile. Substituted styrenes with mixtures of substituents on the aromatic ring are also suitable. As used herein, the term "monoethylenically unsaturated nitrile monomer" means an acyclic compound that includes a single nitrile group and a single site of ethylenic unsaturation per molecule and includes, but is not limited to, acrylonitrile, methacrylonitrile, alpha-chloro acrylonitrile, and the like.

[0013] In a particular embodiment the elastomeric phase comprises from 60 to 100 wt.% repeating units derived from one or more conjugated diene monomers and from 0 to 40 wt.% repeating units derived from one or more monomers selected from alkenyl aromatic monomers and monoethylenically unsaturated nitrile monomers, such as, for example, a styrene-butadiene copolymer, an acrylonitrile-butadiene copolymer or a styrene-butadiene-acrylonitrile copolymer. In another particular embodiment the elastomeric phase comprises from 70 to 90 wt.% repeating units derived from one or more conjugated diene monomers and from 30 to 10 wt.% repeating units derived from one or more monomers selected from alkenyl aromatic monomers.

[0014] The rubber substrate may be present in the rubber modified thermoplastic resin in one embodiment at a level of from about 5 wt.% to about 80 wt.%, based on the weight of the rubber modified thermoplastic resin. In one particular embodiment the rubber substrate may be present in the rubber modified thermoplastic resin at a level of from about 10 wt.% to about 25 wt.%, based on the weight of the rubber modified thermoplastic resin. In another particular embodiment the rubber substrate may be present in the rubber modified thermoplastic resin at a level of from about 55 wt.% to about 80 wt.%, based on the weight of the rubber modified thermoplastic resin.

[0015] There is no particular limitation on the particle size distribution of the rubber substrate (sometimes referred to hereinafter as initial rubber substrate to distinguish it from the rubber substrate following grafting). In some embodiments the initial rubber substrate may possess a broad, essentially monomodal, particle size distribution with particles ranging in size from about 50 nanometers (nm) to about 1000 nm, and more particularly with particles ranging in size from about 200 nm to about 500 nm. In other embodiments the mean particle size of the initial rubber substrate may be less than about 100 nm. In still other embodiments the mean particle size of the initial rubber substrate may be in a range of between about 80 nm and about 400 nm. In other embodiments the mean particle size of the initial rubber substrate may be greater than about 400 nm. In still other embodiments the mean particle size of the initial rubber substrate may be in a range of between about 400 nm and about 750 nm. In still other embodiments the initial rubber substrate comprises particles which are a mixture of particle sizes with at least two mean particle size distributions. In a particular embodiment the initial rubber substrate comprises a mixture of particle sizes with each mean particle size distribution in a range of between about 80 nm and about 750 nm. In another particular embodiment the initial rubber substrate comprises a mixture of particle sizes, one with a mean particle size distribution in a range of between about 80 nm and about 400 nm; and one with a broad and essentially monomodal mean particle size distribution.

[0016] The rubber substrate may be made according to known methods, such as, but not limited to, a bulk, solution, or emulsion process. In one non-limiting embodiment the rubber substrate is made by aqueous emulsion polymerization in the presence of a free radical initiator, e.g., an azonitrile initiator, an organic peroxide initiator, a persulfate initiator or a redox initiator system, and, optionally, in the presence of a chain transfer agent, e.g., an alkyl mercaptan, to form particles of rubber substrate.

[0017] The rigid thermoplastic resin phase of the rubber modified thermoplastic resin comprises one or more thermoplastic polymers. In one embodiment of the present invention

monomers are polymerized in the presence of the rubber substrate to thereby form the rigid thermoplastic phase, at least a portion of which is chemically grafted to the elastomeric phase. The portion of the rigid thermoplastic phase chemically grafted to rubber substrate is sometimes referred to hereinafter as grafted copolymer. In some embodiments two or more different rubber substrates, each possessing a different mean particle size, may be separately employed in a polymerization reaction to prepare the rigid thermoplastic phase, and then the products blended together to make the rubber modified thermoplastic resin. In illustrative embodiments wherein such products each possessing a different mean particle size of initial rubber substrate are blended together, then the ratios of said substrates may be in a range of about 90:10 to about 10:90, or in a range of about 80:20 to about 20:80, or in a range of about 70:30 to about 30:70. In some embodiments an initial rubber substrate with smaller particle size is the major component in such a blend containing more than one particle size of initial rubber substrate.

[0018] The rigid thermoplastic phase comprises a thermoplastic polymer or copolymer that exhibits a glass transition temperature (Tg) in one embodiment of greater than about 25°C, in another embodiment of greater than or equal to 90°C, and in still another embodiment of greater than or equal to 100°C. In a particular embodiment the rigid thermoplastic phase comprises a polymer having structural units derived from one or more monomers selected from the group consisting of alkenyl aromatic monomers and monoethylenically unsaturated nitrile monomers. Suitable alkenyl aromatic monomers and monoethylenically unsaturated nitrile monomers include those set forth hereinabove in the description of the rubber substrate. In addition, the rigid thermoplastic resin phase may, provided that the Tg limitation for the phase is satisfied, optionally include up to about 10 wt.% of third repeating units derived from one or more other copolymerizable monomers.

[0019] The rigid thermoplastic phase typically comprises one or more alkenyl aromatic polymers. Suitable alkenyl aromatic polymers comprise at least about 20 wt.% structural units derived from one or more alkenyl aromatic monomers. In one embodiment the rigid thermoplastic phase comprises an alkenyl aromatic polymer having structural units derived from one or more alkenyl aromatic monomers and from one or more monoethylenically unsaturated nitrile monomers. Examples of such alkenyl aromatic polymers include, but are not limited to, styrene/acrylonitrile copolymers, alpha-methylstyrene/acrylonitrile copolymers, or alpha-methylstyrene/styrene/acrylonitrile copolymers. These copolymers may be used for the rigid thermoplastic phase either individually or as mixtures.

[0020] When structural units in copolymers are derived from one or more monoethylenically unsaturated nitrile monomers, then the amount of nitrile monomer added to form the copolymer comprising the grafted copolymer and the rigid thermoplastic phase may be in one embodiment in a range of between about 5 wt.% and about 40 wt.%, in another embodiment in a range of between about 5 wt.% and about 30 wt.%, in another embodiment in a range of between about 10 wt.% and about 30 wt.%, and in yet another embodiment in a range of between about 15 wt.% and about 30 wt.%, based on the total weight of monomers added to form the copolymer comprising the grafted copolymer and the rigid thermoplastic phase.

[0021] The rigid thermoplastic resin phase of the rubber modified thermoplastic resin may, provided that the Tg limitation for the phase is satisfied, optionally include up to about 10 wt.% of repeating units derived from one or more other copolymerizable monomers such as, e.g., monoethylenically unsaturated carboxylic acids such as, e.g., acrylic acid, methacrylic acid, and itaconic acid; hydroxy(C_1-C_{12})alkyl (meth)acrylate monomers such as, e.g., hydroxyethyl methacrylate; (C_4-C_{12})cycloalkyl (meth)acrylate monomers such as e.g., cyclohexyl methacrylate; (meth)acrylamide monomers such as e.g., acrylamide and methacrylamide; maleimide monomers such as, e.g., N-alkyl maleimides, N-aryl maleimides, maleic anhydride, vinyl esters such as, e.g., vinyl acetate and vinyl propionate. As used herein, the term "(C_4-C_{12})cycloalkyl" means a cyclic alkyl substituent group having from 4 to 12 carbon atoms per group.

[0022] The amount of grafting that takes place between the rubber substrate and monomers comprising the rigid thermoplastic phase varies with the relative amount and composition of the rubber substrate. In one embodiment, greater than about 10 wt.% of the rigid thermoplastic phase is chemically grafted to the rubber substrate, based on the total amount of rigid thermoplastic phase in the composition. In another embodiment, greater than about 15 wt.% of the rigid thermoplastic phase is chemically grafted to the rubber substrate, based on the total amount of rigid thermoplastic phase in the composition.. In still another embodiment, greater than about 20 wt.% of the rigid thermoplastic phase is chemically grafted to the rubber substrate, based on the total amount of rigid thermoplastic phase in the composition. In particular embodiments the amount of rigid thermoplastic phase chemically grafted to the rubber substrate may be in a range of between about 5 wt.% and about 90 wt.%; between about 10 wt.% and about 90 wt.%; between about 15 wt.% and about 85 wt.%; between about 15 wt.% and about 50 wt.%; or between about 20 wt.% and about 50 wt.%, based on the total amount of rigid thermoplastic phase in the composition. In yet other

embodiments, about 40 wt.% to 90 wt.% of the rigid thermoplastic phase is free, that is, non-grafted.

[0023] The rigid thermoplastic phase polymer may be made according to known processes, for example, mass polymerization, emulsion polymerization, suspension polymerization or combinations thereof, wherein at least a portion of the rigid thermoplastic phase is chemically bonded, i.e., "grafted" to the rubber substrate via reaction with unsaturated sites present in the rubber substrate in the case of the rigid thermoplastic phase. The grafting reaction may be performed in a batch, continuous or semi-continuous process.

[0024] In particular embodiments compositions of the invention comprise a rubber modified thermoplastic resin which is an ABS resin. Illustrative ABS resins suitable for use in compositions of the present invention comprise those available from SABIC Innovative Plastics™ under the tradename CYCLOLAC®. In some particular embodiments compositions of the invention comprise 5-50 wt.% rubber derived from at least one alkenyl aromatic resin such as ABS, the wt.% value being based on the weight of the entire composition, wherein the term "rubber" refers to the rubber substrate in ABS. In still other particular embodiments compositions of the invention comprise 5-35 wt.% rubber derived from at least one alkenyl aromatic resin such as ABS and based on the weight of the entire composition. In still other particular embodiments compositions of the invention comprise 5-30 wt.% rubber derived from at least one alkenyl aromatic resin such as ABS and based on the weight of the entire composition. In yet other particular embodiments compositions of the invention comprise less than 30 wt.% rubber derived from at least one alkenyl aromatic resin such as ABS and based on the weight of the entire composition. The rubber content may be varied by employing a single alkenyl aromatic resin with desired rubber content or by employing two or more alkenyl aromatic resins each with different rubber content. In some embodiments compositions of the invention comprise 40-70 wt.% alkenyl aromatic resin and in other embodiments 40-66 wt.% alkenyl aromatic resin based on the total weight of the composition

[0025] Compositions in embodiments of the invention also comprise at least one additive comprising cellulose, which additive is sometimes referred to herein after as cellulosic material. In various embodiments the cellulosic material comprises or is derived from cellulosic fiber, wood fiber, seed husks, ground rice hulls, newspaper, kenaf, coconut shell, or like materials. In some specific embodiments the cellulosic material may be wood fiber, which is available in different forms. In other illustrative examples cellulosic material comprises or is derived from sawdust, alfalfa, wheat pulp, wood chips, wood particles,

ground wood, wood flour, wood flakes, wood veneers, wood laminates, paper, cardboard, straw, cotton, peanut shells, bagass, plant fibers, bamboo fiber, palm fiber, or like materials. Those skilled in the art should recognize that the cellulosic material of the present invention may be any suitable combination of different types of cellulosic material. In some particular embodiments the cellulosic material is selected from cellulosic fibers and wood flour.

[0026] Compositions in embodiments of the invention also comprise at least one ammonium polyphosphate. Ammonium polyphosphates are known materials and may be prepared, for example, as exemplified in U.S. Patent Nos. 3,423,343 and 3,513,114. In some illustrative embodiments the ammonium polyphosphates have the general formula $(NH_4)_nH_2P_nO_{3n+1}$, wherein n is 1 or more, or $(NH_4PO_3)_n$ wherein n represents an integer equal to or greater than 2. Illustrative examples of commercially available ammonium polyphosphates comprise EXOLIT® ammonium polyphosphate produced and sold by Clariant, PHOS-CHECK® ammonium polyphosphate available from ICL Performance Products LP and FR CROS® ammonium polyphosphate available from Budenheim Iberica Comercial S.A. In one embodiment compositions of the invention comprise at least one "crystal phase II" ammonium polyphosphate which may be cross-linked and/or branched. Crystal phase II ammonium polyphosphates are known in the art. They are high molecular weight ammonium polyphosphates, and exhibit high thermal stability (for example, decomposition starting at about 300°C) and low water solubility. An illustrative example of a crystal phase II ammonium polyphosphate is EXOLIT® AP 423 available from Clariant. Coated ammonium polyphosphate may also be employed in compositions of the invention in some embodiments. Illustrative examples of coated ammonium polyphosphates comprise melamine-coated or melamine-formaldehyde-coated or surface-reacted melamine-coated ammonium polyphosphate. One illustrative coated ammonium polyphosphate is FR CROS® C40 available from Budenheim Iberica Comercial S.A.

[0027] In various embodiments of the invention ammonium polyphosphate and cellulosic material may be present together in compositions of the invention in total amount by weight effective to provide articles exhibiting at least V-1 flame rating or better as determined according to the UL-94 protocol. In particular embodiments ammonium polyphosphate may be present in compositions of the invention in an amount in a range of between about 5 wt.% and about 35 wt.%, or in an amount in a range of between about 7 wt.% and about 33 wt.%, or in an amount in a range of between about 9 wt.% and about 33 wt.%, or in an amount in a range of between about 12 wt.% and about 33 wt.%, based on the weight of the entire composition. In particular embodiments cellulosic material may be

present in compositions of the invention in an amount in a range of between about 5 wt.% and about 45 wt.%, or in an amount in a range of between about 10 wt.% and about 40 wt.%, or in an amount in a range of between about 14 wt.% and about 40 wt.%, based on the weight of the entire composition. In other particular embodiments of the invention ammonium polyphosphate and cellulosic material may be present in a weight % ratio effective to provide articles exhibiting V-0 or V-1 or V-2 flame rating as determined according to the UL-94 protocol. In other particular embodiments of the invention ammonium polyphosphate and cellulosic material may be present in a weight % ratio in a range of 1:10 to 10:1, often 1:8 to 8:1, more often 1:5 to 5:1, still more often 1:3 to 3:1 and still more often 1:2 to 2:1.

[0028] Compositions of the present invention may also optionally comprise additives known in the art including, but not limited to, stabilizers, such as color stabilizers, heat stabilizers, light stabilizers, antioxidants, UV screeners, and UV absorbers; adjunct flame retardants, anti-drip agents, lubricants, flow promoters or other processing aids; plasticizers, antistatic agents, mold release agents, impact modifiers, fillers, or colorants such as dyes or pigments which may be organic, inorganic or organometallic; visual effects additives and like additives. Illustrative additives include, but are not limited to, silica, silicates, zeolites, titanium dioxide, stone powder, glass fibers or spheres, carbon fibers, carbon black, graphite, calcium carbonate, talc, lithopone, zinc oxide, zirconium silicate, iron oxides, diatomaceous earth, calcium carbonate, magnesium oxide, chromic oxide, zirconium oxide, aluminum oxide, crushed quartz, clay, calcined clay, talc, kaolin, asbestos, cellulose, wood flour, cork, cotton or synthetic textile fibers, especially reinforcing fillers such as glass fibers, carbon fibers, metal fibers, and metal flakes, including, but not limited to aluminum flakes. Compositions of the invention do not contain cornstarch or a metal stearate such as zinc stearate. Often more than one additive is included in compositions of the invention, and in some embodiments more than one additive of one type is included.

[0029] In some particular embodiments compositions of the invention optionally comprise at least one organophosphorus compound as an adjunct flame retardant. Suitable organophosphorus flame retardant compounds are known in the art and include, but are not limited to, monophosphate esters such as triaryl phosphates, triphenyl phosphate, tricresyl phosphate, tritolyl phosphate, diphenylcresyl phosphate, phenyl bisdodecyl phosphate, and ethyl diphenyl phosphate, as well as diphosphate esters and oligomeric phosphates such as, for example, aryl diphosphates, resorcinol diphosphate, bisphenol A diphosphate, diphenyl hydrogen phosphate, and 2-ethylhexyl hydrogen phosphate. Suitable oligomeric phosphate compounds are set forth for example in U.S. Patent No. 5,672,645. An adjunct flame

retardant, when present, is typically present in an amount of about 4-16 wt.% and particularly 5-15 wt.% based on the weight of the entire composition.

[0030] Compositions of the invention and articles made therefrom may be prepared by known thermoplastic processing techniques. Known thermoplastic processing techniques which may be used include, but are not limited to, batch mixing, extrusion, calendering, kneading, profile extrusion, sheet extrusion, pipe extrusion, coextrusion, molding, extrusion blow molding, thermoforming, injection molding, co-injection molding, rotomolding, compression molding, and like processes and combinations of such processes. In a particular embodiment compositions are prepared by an extrusion process. In a particular embodiment articles are prepared from compositions of the invention by an injection molding process. The invention further contemplates additional fabrication operations on said articles, such as, but not limited to, in-mold decoration, baking in a paint oven, over-molding, co-extrusion, multilayer extrusion, surface etching, lamination, and/or thermoforming.

[0031] Novel aspects of the invention encompass processes for making compositions of the invention wherein in some embodiments ammonium polyphosphate and cellulosic material are included in the compositions in such a manner so as to minimize the contact time between ammonium polyphosphate and cellulosic material. Illustrative examples for minimizing said contact time include but are not limited to precompounding all or at least a portion of resinous components and cellulosic material before inclusion of ammonium polyphosphate. In other embodiments compositions of the invention are prepared in an extrusion process, and all or at least a portion of ammonium polyphosphate is fed to the extruder at a down-stream feed-port of the extruder wherein all or at least a portion of alkenyl aromatic resin and cellulosic material have been fed at the extruder feed-throat. Any compounding process for compositions of the invention, including but not limited to down-stream feeding of ammonium polyphosphate during extrusion, may be performed by feeding solid ammonium polyphosphate or by injecting a liquid mixture of ammonium polyphosphate optionally in combination with one or more additional blend components, for example, at least one adjunct flame retardant such as a liquid adjunct flame retardant. Optionally, any extrusion process may include vacuum venting at one or more appropriate points in the extruder. A particular embodiment of the invention is an extrusion process for preparing a resinous, flame-retardant composition comprising (i) 40-66 wt.% ABS, (ii) 9-33 wt.% ammonium polyphosphate and (iii) 14-40 wt.% cellulosic material, wherein ammonium polyphosphate and cellulosic material are present in a weight % ratio in a range of 1:2 to 2:1 effective to provide molded articles exhibiting at least V-1 flame rating as determined

according to the UL-94 protocol, and wherein the composition comprises 15-30 wt.% rubber derived from ABS, the rubber content being based on the weight of the entire composition, which process comprises the step of adding all or a portion of the ammonium polyphosphate to the extruder down-stream from the ABS and the cellulosic material. In other embodiments of the invention all or at least a portion of alkenyl aromatic resin may be precompounded with all or at least a portion of cellulosic material before combination with ammonium polyphosphate. Such precompounding may be performed using known methods, such as but not limited to extrusion or kneading.

[0032] The compositions of the present invention can be formed into useful articles. Useful articles comprise those which are commonly used in applications requiring flame resistance and particularly in applications requiring halogen-free (eco-friendly) compositions for flame resistance. In some embodiments the articles comprise unitary articles comprising a composition of the invention. In other embodiments articles comprise electrical housings, business machine internal and external parts, printers, computer housings, switches, profiles, window profiles and like articles. Multilayer articles comprising at least one layer derived from a composition of the invention are also contemplated. Articles, for example molded articles, made from the compositions of the present invention often exhibit V-1 flame rating or better (for example, V-1 or V-0 rating) as determined according to the UL-94 protocol.

[0033] Without further elaboration, it is believed that one skilled in the art can, using the description herein, utilize the present invention to its fullest extent. The following examples are included to provide additional guidance to those skilled in the art in practicing the claimed invention. The examples provided are merely representative of the work that contributes to the teaching of the present application. Accordingly, these examples are not intended to limit the invention, as defined in the appended claims, in any manner.

[0034] In the following examples (abbreviated "Ex.") and comparative examples ("C.Ex.") the amounts of components are expressed in wt.% unless noted. ABS in the following compositions comprised about 14-17% polybutadiene rubber and was obtained from SABIC Innovative Plastics™. High rubber ABS (abbreviated "HR-ABS") was BLENDEX® 338 comprising about 60-78% polybutadiene rubber and was obtained from SABIC Innovative Plastics™. High impact polystyrene (HIPS comprised less than about 10% rubber content. Styrene-acrylonitrile copolymer (SAN) comprised structural units derived from about 3:1 styrene:acrylonitrile. Ammonium polyphosphate (abbreviated "APP") was EXOLIT® AP 423 ammonium polyphosphate containing about 31-32 weight % phosphorus and was obtained from Clariant. Cellulose fiber was CreaTech TC180 obtained

from CreaFill Fibers Corp., Chestertown, Maryland. Wood flour was obtained from American Wood Flour Company, Schoefield, Wisconsin. Flame retardant properties were determined according to the UL-94 protocol. The notation "NC" for flame retardant rating indicates no flame retardancy was observed. Notched Izod impact strength (NII) values in units of joules per meter were determined according to ISO 180 at room temperature. Flexural strength values in units of megapascals and flexural modulus values in units of gigapascals were determined according to ISO 178.

[0035] In an embodiment, a resinous, flame-retardant composition comprises (i) 40-66 wt.% alkenyl aromatic resin, (ii) 9-33 wt.% ammonium polyphosphate and (iii) 14-40 wt.% cellulosic material, wherein all weights are based on the total weight of the composition and wherein ammonium polyphosphate and cellulosic material are present in a weight % ratio effective to provide molded articles exhibiting at least V-1 flame rating as determined according to the UL-94 protocol.

[0036] Various other embodiments, (i) the ammonium polyphosphate and cellulosic material are present in a weight % ratio in a range of 1:3 to 3:1; (ii) the ammonium polyphosphate and cellulosic material are present in a weight % ratio in a range of 1:2 to 2:1; (iii) the alkenyl aromatic resin is selected from the group consisting of polystyrene, acrylonitrile-butadiene-styrene copolymer (ABS), high-impact polystyrene (HIPS), styrene-acrylonitrile copolymer (SAN), alpha-methylstyrene/acrylonitrile copolymer, alpha-methylstyrene/styrene/acrylonitrile copolymer and mixtures thereof; (iv) the composition comprises 5-50 wt.% rubber derived from ABS or HIPS and based on the weight of the entire composition; (v) the composition comprises 5-35 wt.% rubber derived from ABS or HIPS and based on the weight of the entire composition; (vi) the type of ammonium polyphosphate is crystal phase II; (vii) the cellulosic material comprises or is derived from cellulosic fiber, wood fiber, seed husks, ground rice hulls, newspaper, kenaf, coconut shell, sawdust, alfalfa, wheat pulp, wood chips, wood particles, ground wood, wood flour, wood flakes, wood veneers, wood laminates, paper, cardboard, straw, cotton, peanut shells, bagass, plant fibers, bamboo fiber, or palm fiber; (viii) the composition further comprises at least one adjunct flame retardant; (ix) the adjunct flame retardant is selected from the group consisting of monophosphate esters, triaryl phosphates, triphenyl phosphate, tricresyl phosphate, tritolyl phosphate, diphenylcresyl phosphate, phenyl bisdodecyl phosphate, ethyl diphenyl phosphate, diphosphate esters, aryl diphosphates, resorcinol diphosphate, bisphenol A diphosphate, diphenyl hydrogen phosphate, 2-ethylhexyl hydrogen phosphate and oligomeric phosphates; (x) the composition is prepared by an extrusion process wherein at least a portion of the

ammonium polyphosphate is fed down-stream from the alkenyl aromatic resin and cellulosic material; and/or (xi) the composition is prepared by an extrusion process wherein at least a portion of the alkenyl aromatic resin and cellulosic material are precompounded together before combination with ammonium polyphosphate.

[0037] Also included herein are article(s) comprising any of the above compositions. The article can be an electrical housing, a business machine internal or external part, a printer housing, a computer housing, a switch, a profile or a window profile.

[0038] Another embodiment of a resinous, flame-retardant composition comprises (i) 40-66 wt.% alkenyl aromatic resin, (ii) 9-33 wt.% ammonium polyphosphate and (iii) 14-40 wt.% cellulosic material, wherein ammonium polyphosphate and cellulosic material are present in a weight % ratio in a range of 1:2 to 2:1 effective to provide molded articles exhibiting at least V-1 flame rating as determined according to the UL-94 protocol, and wherein the composition is prepared by an extrusion process wherein all or a portion of the ammonium polyphosphate is fed to the extruder down-stream from the alkenyl aromatic resin and the cellulosic material. This composition can further comprise at least one adjunct flame retardant selected from the group consisting of monophosphate esters, triaryl phosphates, triphenyl phosphate, tricresyl phosphate, tritolyl phosphate, diphenylcresyl phosphate, phenyl bisdodecyl phosphate, ethyl diphenyl phosphate, diphosphate esters, aryl diphosphates, resorcinol diphosphate, bisphenol A diphosphate, diphenyl hydrogen phosphate, 2-ethylhexyl hydrogen phosphate and oligomeric phosphates.

[0039] In one embodiment, an extrusion process for preparing a resinous, flame-retardant composition comprises (i) 40-66 wt.% alkenyl aromatic resin, (ii) 9-33 wt.% ammonium polyphosphate and (iii) 14-40 wt.% cellulosic material, wherein ammonium polyphosphate and cellulosic material are present in a weight % ratio in a range of 1:2 to 2:1 effective to provide molded articles exhibiting at least V-1 flame rating as determined according to the UL-94 protocol, which process comprises the step of adding at least a portion of the ammonium polyphosphate to the extruder down-stream from the ABS and the cellulosic material. In this process the composition can further comprise at least one adjunct flame retardant selected from the group consisting of monophosphate esters, triaryl phosphates, triphenyl phosphate, tricresyl phosphate, tritolyl phosphate, diphenylcresyl phosphate, phenyl bisdodecyl phosphate, ethyl diphenyl phosphate, diphosphate esters, aryl diphosphates, resorcinol diphosphate, bisphenol A diphosphate, diphenyl hydrogen phosphate, 2-ethylhexyl hydrogen phosphate and oligomeric phosphates. Also included herein are articles prepared from any of these processes.

COMPARATIVE EXAMPLES 1-8 AND EXAMPLES 1-6

[0040] Compositions comprising ABS in Table 1 were compounded in an extruder unless otherwise described. The compounded material was molded into test parts and the parts were tested for physical properties. The test results are shown in Table 1.

TABLE 1

Ex. or C.Ex. →	C.Ex. 1 [a]	C.Ex. 2 [a]	C.Ex. 3 [a]	C.Ex. 4	C.Ex. 5	C.Ex. 6	C.Ex. 7	Ex. 1 [b]	Ex. 2 [b]	Ex. 3 [b]	Ex. 4 [b]	Ex. 5 [d]	Ex. 6 [b]	C.Ex. 8 [c]
Component ↓														
ABS	80	70	60	80	70	60	70	50	55	50	55	63	57.7	57.5
Cellulose fiber	20	30	40	--	--	--	--	30	25	20	15	20	30	30
Wood flour	--	--	--	20	30	40	--	--	--	--	--	--	--	--
APP	--	--	--	--	--	--	30	20	20	30	30	17	12.5	12.5
FR rating	NC	NC	NC	NC	NC	NC	NC	V-0	V-1	V-0	V-1	V-1	V-0	NC
Strength, MPa	--	--	--	--	--	--	--	--	--	--	--	62.6	58.2	58.2
Modulus, GPa	--	--	--	--	--	--	--	--	--	--	--	3.40	3.08	3.06

[a] Prepared in an internal mixer

[b] APP fed downstream of other blend components

[c] APP fed in extruder feed-throat with other blend components

[d] ABS and cellulose precompounded before blending with APP

[0041] Comparative examples 1-3 show that cellulose fiber alone does not impart flame retardant properties to ABS. Comparative examples 4-6 show that wood flour alone does not impart flame retardant properties to ABS. Comparative example 7 shows that a high level (30 wt.%) of ammonium polyphosphate alone does not impart flame retardant properties to ABS. Surprisingly, examples 1-5 show that the combination of cellulosic material and ammonium polyphosphate imparts good flame resistance to ABS compositions either when ammonium polyphosphate is fed down-stream of the feed throat while ABS and cellulosic material are fed directly to the feed-throat of the extruder (example 1-4 and 6) or when ABS and cellulose are precompounded before blending with APP (example 5). Comparative example 8 shows that the combination of cellulose fiber and ammonium polyphosphate does not impart good flame resistance to ABS compositions when all three components are fed directly to the feed-throat of the extruder. Although the invention is in no way limited by any theory of operation, it is believed that feeding ammonium polyphosphate in the feed-throat along with cellulosic material leads to detrimental reaction between these two components. For example, it is believed that ammonium polyphosphate can promote cross-linking of at least some cellulosic material leading to poor dispersion of such material in a resinous matrix.

COMPARATIVE EXAMPLES 9-21 AND EXAMPLES 7-8

[0042] Compositions comprising ABS in Table 2 were prepared with a total rubber concentration of about 15 wt.%. The rubber level was achieved by combining two ABS grades with different rubber contents. The compositions were compounded in an extruder with downstream feeding of APP. The compounded material was molded into test parts and the parts were tested for physical properties. The test results are shown in Table 2. Example 7 and example 8 had similar amounts and ratio of ammonium polyphosphate and cellulosic material, but in addition example 8 had 10 wt.% adjunct flame retardant triphenyl phosphate. The data show that the addition of an adjunct flame retardant provided good flame resistance. The addition of adjunct flame retardant also provided molded parts with better color, better flow properties and allowed the use of lower processing temperature for the composition.

TABLE 2

Ex. or C.Ex. →	C.Ex. 9	C.Ex. 10	C.Ex. 11	C.Ex. 12	C.Ex. 13	C.Ex. 14	C.Ex. 15	C.Ex. 16	C.Ex. 17	C.Ex. 18	C.Ex. 19	C.Ex. 20	C.Ex. 21	Ex. 7	Ex. 8
Component ↓															
ABS	87.3	74.6	73.3	72.0	71.4	70.1	68.2	63.7	63.7	59.3	57.4	55.4	54.2	52.9	40.2
HR-ABS	2.7	5.4	5.7	6.0	6.1	6.4	6.8	8.0	7.8	8.7	9.1	9.6	9.8	10.1	12.8
cellulose	5	5	5	5	12.5	12.5	20	12.5	12.5	5	12.5	20	20	20	20
APP	5	5	11	17	5	11	5	11	11	17	11	5	11	17	17
TPP	--	10	5	--	5	--	--	5	5	10	10	10	5	--	10
FR rating	NC	NC	NC	NC	NC	NC	NC	NC	NC	NC	NC	NC	NC	NC	V-0
Strength, MPa	61.6	50.2	53.7	52.4	60.0	59.3	62.0	57.4	55.9	41.4	47.9	51.4	55.6	51.9	45.4
Modulus, GPa	2.30	2.31	2.23	2.21	2.68	2.62	2.74	2.65	2.63	2.29	2.77	2.99	2.98	2.87	3.29
NII, J/m^{-1}	43	32	27	32	27	32	32	27	27	21	21	21	21	21	27

COMPARATIVE EXAMPLES 22-23 AND EXAMPLES 9-12

[0043] Compositions comprising ABS in Table 3 were compounded in an internal mixer. The compounded material was molded into test parts and the parts were tested for flame resistance. The test results are shown in Table 3.

TABLE 3

Ex. or C.Ex. →	Ex. 9	Ex. 10	Ex. 11	Ex. 12	C.Ex. 22	C.Ex. 23
Component ↓						
ABS	65	60	60	57.5	65	60
cellulose	20	20	20	30	20	20
APP	15	20	10	12.5	--	--
TPP	--	--	10	--	15	20
FR rating	V-1	V-1	V-1	V-0	NC	NC

[0044] Compositions comprising alkenyl aromatic resin, cellulosic material and ammonium polyphosphate in examples 9-12 show good flame resistance. In contrast, the comparative examples 22-23 which contain ratios of styrenic polymer to cellulosic material similar to those ratios in examples of the invention but which also contain only organophosphorus flame retardant and no ammonium polyphosphate exhibit poor flame resistance.

COMPARATIVE EXAMPLES 24-26 AND EXAMPLES 13-18

[0045] Compositions comprising high impact polystyrene (HIPS) were compounded in an internal mixer. The compounded material was molded into test parts and the parts were tested for flame resistance. Blend compositions and test results are shown in Table 4.

TABLE 4

Ex. or C.Ex. →	C.Ex. 24	C.Ex. 25	C.Ex. 26	Ex. 13	Ex. 14	Ex. 15	Ex. 16	Ex. 17	Ex. 18
Component ↓									
HIPS	70.5	66.5	68	65	65.5	63	58	63	58
cellulose	12.5	12.5	15	15	12.5	15	15	20	20
APP	17	11	17	15	17	17	17	17	17
TPP	--	10	--	5	5	5	10	--	5
FR rating	NC	NC	NC	NC	V-1	V-1	V-0	V-0	V-0

COMPARATIVE EXAMPLES 27-30 AND EXAMPLES 19-26

[0046] Compositions comprising polystyrene (PS) were compounded in an internal mixer. The compounded material was molded into test parts and the parts were tested for flame resistance. Blend compositions and test results are shown in Table 5.

TABLE 5

Ex. or C.Ex. →	C.Ex. 27	C.Ex. 28	C.Ex. 29	C.Ex. 30	Ex. 19	Ex. 20	Ex. 21	Ex. 22	Ex. 23	Ex. 24	Ex. 25	Ex. 26
Component ↓												
PS	70.5	65.5	66.5	68	63	65	58	63	58	60	58	56
cellulose	12.5	12.5	12.5	15	15	15	15	20	20	25	25	25
APP	17	17	11	17	17	15	17	17	17	15	17	19
TPP	--	5	10	--	5	5	10	--	5	--	--	--
FR rating	NC	NC	NC	NC	NC	NC	V-0	V-1	V-0	V-1	V-0	V-0

COMPARATIVE EXAMPLES 31-34 AND EXAMPLES 27-34

[0047] Compositions comprising SAN were compounded in an internal mixer. The compounded material was molded into test parts and the parts were tested for flame resistance. Blend compositions and test results are shown in Table 6.

TABLE 6

Ex. or C.Ex. →	C.Ex. 31	C.Ex. 32	C.Ex. 33	C.Ex. 34	Ex. 27	Ex. 28	Ex. 29	Ex. 30	Ex. 31	Ex. 32	Ex. 33	Ex. 34
Component ↓												
SAN	70.5	65.5	66.5	68	63	65	58	63	58	60	58	56
cellulose	12.5	12.5	12.5	15	15	15	15	20	20	25	25	25
APP	17	17	11	17	17	15	17	17	17	15	17	19
TPP	--	5	10	--	5	5	10	--	5	--	--	--
FR rating	NC	NC	NC	NC	NC	NC	V-0	NC	V-0	V-1	V-1	V-0

[0048] While the invention has been illustrated and described in typical embodiments, it is not intended to be limited to the details shown, since various modifications and substitutions can be made without departing in any way from the spirit of the present invention. As such, further modifications and equivalents of the invention herein disclosed may occur to persons skilled in the art using no more than routine experimentation, and all such modifications and equivalents are believed to be within the spirit and scope of the invention as defined by the following claims. All Patents and published articles cited herein are incorporated herein by reference.

CLAIMS:

1. A resinous, flame-retardant composition comprising (i) 40-66 wt.% alkenyl aromatic resin, (ii) 9-33 wt.% ammonium polyphosphate and (iii) 14-40 wt.% cellulosic material, wherein all weights are based on the total weight of the composition and wherein ammonium polyphosphate and cellulosic material are present in a weight % ratio effective to provide molded articles exhibiting at least V-1 flame rating as determined according to the UL-94 protocol.

2. The composition of claim 1, wherein ammonium polyphosphate and cellulosic material are present in a weight % ratio in a range of 1:3 to 3:1.

3. The composition of claim 1, wherein ammonium polyphosphate and cellulosic material are present in a weight % ratio in a range of 1:2 to 2:1.

4. The composition of any of Claims 1 - 3, wherein the alkenyl aromatic resin is selected from the group consisting of polystyrene, acrylonitrile-butadiene-styrene copolymer (ABS), high-impact polystyrene (HIPS), styrene-acrylonitrile copolymer (SAN), alpha-methylstyrene/acrylonitrile copolymer, alpha-methylstyrene/styrene/acrylonitrile copolymer and mixtures thereof.

5. The composition of claim 4, comprising 5-50 wt.% rubber derived from ABS or HIPS and based on the weight of the entire composition.

6. The composition of claim 4, comprising 5-35 wt.% rubber derived from ABS or HIPS and based on the weight of the entire composition.

7. The composition of any of Claims 1 - 6, wherein the type of ammonium polyphosphate is crystal phase II.

8. The composition of any of Claims 1 - 10, wherein the cellulosic material comprises or is derived from cellulosic fiber, wood fiber, seed husks, ground rice hulls, newspaper, kenaf, coconut shell, sawdust, alfalfa, wheat pulp, wood chips, wood particles, ground wood, wood flour, wood flakes, wood veneers, wood laminates, paper, cardboard, straw, cotton, peanut shells, bagass, plant fibers, bamboo fiber, or palm fiber.

9. The composition of any of Claims 1 - 8, which further comprises at least one adjunct flame retardant.

10. The composition of claim 9, wherein the adjunct flame retardant is selected from the group consisting of monophosphate esters, triaryl phosphates, triphenyl phosphate, tricresyl phosphate, tritolyl phosphate, diphenylcresyl phosphate, phenyl bisdodecyl phosphate, ethyl diphenyl phosphate, diphosphate esters, aryl diphosphates, resorcinol

diphosphate, bisphenol A diphosphate, diphenyl hydrogen phosphate, 2-ethylhexyl hydrogen phosphate and oligomeric phosphates.

11. The composition of any of Claims 1 - 10, which is prepared by an extrusion process wherein at least a portion of the ammonium polyphosphate is fed down-stream from the alkenyl aromatic resin and cellulosic material.

12. The composition of any of Claims 1 - 10, which is prepared by an extrusion process wherein at least a portion of the alkenyl aromatic resin and cellulosic material are precompounded together before combination with ammonium polyphosphate.

13. An article comprising the composition of any of Claims 1 - 12.

14. The article of claim 13, which comprises an electrical housing, a business machine internal or external part, a printer housing, a computer housing, a switch, a profile or a window profile.

15. A resinous, flame-retardant composition comprising (i) 40-66 wt.% alkenyl aromatic resin, (ii) 9-33 wt.% ammonium polyphosphate and (iii) 14-40 wt.% cellulosic material, wherein ammonium polyphosphate and cellulosic material are present in a weight % ratio in a range of 1:2 to 2:1 effective to provide molded articles exhibiting at least V-1 flame rating as determined according to the UL-94 protocol, and wherein the composition is prepared by an extrusion process wherein all or a portion of the ammonium polyphosphate is fed to the extruder down-stream from the alkenyl aromatic resin and the cellulosic material.

16. The composition of claim 15, further comprising at least one adjunct flame retardant selected from the group consisting of monophosphate esters, triaryl phosphates, triphenyl phosphate, tricresyl phosphate, tritolyl phosphate, diphenylcresyl phosphate, phenyl bisdodecyl phosphate, ethyl diphenyl phosphate, diphosphate esters, aryl diphosphates, resorcinol diphosphate, bisphenol A diphosphate, diphenyl hydrogen phosphate, 2-ethylhexyl hydrogen phosphate and oligomeric phosphates.

17. An extrusion process for preparing a resinous, flame-retardant composition comprising (i) 40-66 wt.% alkenyl aromatic resin, (ii) 9-33 wt.% ammonium polyphosphate and (iii) 14-40 wt.% cellulosic material, wherein ammonium polyphosphate and cellulosic material are present in a weight % ratio in a range of 1:2 to 2:1 effective to provide molded articles exhibiting at least V-1 flame rating as determined according to the UL-94 protocol, which process comprises the step of adding at least a portion of the ammonium polyphosphate to the extruder down-stream from the ABS and the cellulosic material.

18. The extrusion process of claim 17, wherein the composition further comprises at least one adjunct flame retardant selected from the group consisting of monophosphate

esters, triaryl phosphates, triphenyl phosphate, tricresyl phosphate, tritolyl phosphate, diphenylcresyl phosphate, phenyl bisdodecyl phosphate, ethyl diphenyl phosphate, diphosphate esters, aryl diphosphates, resorcinol diphosphate, bisphenol A diphosphate, diphenyl hydrogen phosphate, 2-ethylhexyl hydrogen phosphate and oligomeric phosphates.

19. An article comprising the composition prepared by the process of claim 17 or 18.

INTERNATIONAL SEARCH REPORT

International application No
PCT/US2009/033078

A. CLASSIFICATION OF SUBJECT MATTER
INV. C08L25/00 C08L55/02 C08L51/04 C08L53/02 C08L1/00
C08K3/32 C09K21/04 C09K21/14

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
C08L C09K C08K C09D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)
EPO-Internal, WPI Data, CHEM ABS Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	GB 2 437 778 A (ENVIRONMENTAL SEALS LTD [GB]) 7 November 2007 (2007-11-07) the whole document	1-19
A	GB 1 184 878 A (MONSANTO CO [US]) 18 March 1970 (1970-03-18) examples	1-19
A	US 4 256 786 A (DIAS GIL M ET AL) 17 March 1981 (1981-03-17) examples	1-19

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[X] Further documents are listed in the continuation of Box C.

[X] See patent family annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

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"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

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Date of the actual completion of the international search	Date of mailing of the international search report
22 April 2009	12/05/2009

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Form PCT/ISA/210 (second sheet) (April 2005)

INTERNATIONAL SEARCH REPORT

International application No
PCT/US2009/033078

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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Information on patent family members

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