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(54) AQUEOUS ALUMINOSILICATE GEL-FORMING COMPOSITION

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(57) ABSTRACT

An aqueous gel-forming composition, e.g. a fire retardant coating composition, comprises an aluminosilicate and an organic liquid having a boiling point greater than 110° C., e.g. silicone oil, which enhances the integrity of films formed by application of the composition as a coating to a surface then followed by drying.

AQUEOUS ALUMINOSILICATE GEL-FORMING COMPOSITION

[0001] The present invention relates to aqueous compositions, precursor systems for producing such compositions and also application systems for applying said compositions, e.g. as coatings, to a surface or substrate.

[0002] Soluble silicates are widely used as adhesives, coatings and bondings. Whilst their inherent solubility is an asset in many of the applications for which they are used, it is disadvantageous for applications where, for example, water resistance, integrity and strength of structure are deemed essential.

[0003] Considerable efforts have been made to minimise the solubility of silicates in compositions of the kind referred to above, for example by addition of metal (such as calcium and magnesium) salts. However, addition of such salts tends to result in a precipitated form rather than a product with a continuous network-like structure. The soluble salt formed in the precipitation reaction is deleterious to the physical integrity of the applied film and hence ultimately, strength of the resultant product.

[0004] Factors such as these are a deterrent to the use of silicates in for example the production of fire retardant coating composition. Fire retardant coating compositions find wide application in the construction and building maintenance industries, for example for application to flammable building materials before, or after, their incorporation in a building structure. Examples of flammable materials are polymer tiling and sheeting, e.g. of expanded polystyrene or urethane plastics and composites containing such plastics. Wood, wood chip and paper based materials can also benefit from application of such coatings. Included within the class of fire retardant coating compositions are so-called intumescent coating which exert their protectant action partly by swelling when exposed to heat or fire.

[0005] In some cases, flammable materials are sold with the fire retardant coating pre-applied. For example, an intumescent fire retardant coating known as SafeCoat $E84^{TM}$ is pre-applied to expanded polystyrene/polyurethane foam articles prior to sale.

[0006] U.S. Pat. No. 5,462,699 relates to a fire retardant composition for application to, inter alia, building materials, the composition containing a silicate, water and surfactant.

[0007] The problem with using a silicate based fire retardant composition for building insulation materials such as expanded polystyrene (e.g. roofing insulation) is that, until needed, such materials are often left exposed to the adverse weather conditions after delivery to the building site, Unless precautions are taken to protect them against the wet, such exposure can result in significant loss of the fire retardancy properties of the silicate-based composition.

[0008] We have now found that improved aqueous coatings can be produced from silicates in such a way that the solubility problem discussed above is significantly counteracted thus making compositions according to the present invention suitable for use as fire retardant compositions. However, the invention is not limited to aqueous compositions for fire retardancy use; other applications such as use of the composition as an adhesive or binder are within the ambit of the present invention. A further advantage of the compositions of the invention is that they may be used to prepare fire retardant systems that are substantially free of halogen-containing compounds. Halogen-containing compounds are undesirable because of their potential environmental risk.

[0009] Thus, the present invention provides an aqueous gel-forming composition comprising an aluminosilicate and an organic liquid which enhances the integrity of films formed by application of the composition as a coating to a surface followed by drying.

[0010] In a first aspect, the invention provides an aqueous gel-forming composition comprising:

- [0011] (a) from 5% to 40%, preferably 5% to 30%, of an aluminosilicate;
- **[0012]** (b) from 0.1% to 10% of an organic liquid,

wherein the organic liquid has a boiling point of greater than 110° C.

[0013] Boiling points in this specification are to be measured at standard atmospheric pressure.

[0014] By aqueous it is meant that the balance of the composition comprises water and optionally one or more other ingredients. Typically, the compositions of the invention comprise at least 20% by weight of water, preferably at least 30%, more preferably at least 40%.

[0015] As used herein, the term "gel" refers to a substance that contains a continuous solid skeleton (in the present invention based on the aluminosilicate) enclosing a continuous liquid phase (in the present case, predominantly water)—see for example Sol-Gel Science, The Physics and Chemistry of Sol-Gel Processing (C J Brinker and G W Scheer) published by Academic Press Inc., 1990, for example page 8. These materials may also be referred to as co-gels or coagels. Initially, the solid may be in the form of dispersed, discontinuous solid particles (a sol), but these individual particles coalesce to form a continuous solid network. The compositions of the invention are initially in the form of a sol which converts over time to a gel.

[0016] A preferred class of compositions according to the first aspect of the present invention consists of those which comprise:

- [0017] (a) from 5% to 40%, preferably from 5 to 30%, more preferably from 10% to 25% by weight of the aluminosilicate;
- **[0018]** (b) from 0.1% to 10%, preferably from 0.3% to 5% by weight of the organic liquid; and
- [0019] (c) the balance being the water, optionally further including one or more other ingredients.

[0020] In the absence of the organic liquid, we have found that for a given drying temperature, the more extensive the drying (i.e. loss of water), the more prone a film coating produced using the aluminosilicate composition is to result in a weak and powdery coating in a relatively short time. When the organic liquid is present however, the integrity of the coating in terms of its strength and non-powdery nature is considerably improved.

[0021] Any reference herein to a liquid means a substance which is liquid, preferably pourable, at 25° C. at atmospheric pressure, unless explicitly stated to the contrary. Further, all viscosities stated herein which refer to non-Newtonian liquids or gels are viscosities measured at a shear rate of 23 s⁻¹ and at 25° C.

[0022] The organic liquid is preferably one which is substantially water-immiscible. Usually the degree of immiscibility is such that, at 25° C., the organic liquid dissolves to the extent of less than about 10% by weight (preferably less than about 5% by weight, more preferably less than 1% by weight) in water, or water dissolves to the extent of less than about 10% by weight (preferably less than about 5% by weight, more preferably less than 1% by weight) in the organic liquid. [0023] Although the composition of the present invention can be applied as a fire retardant coating to a surface or substrate, it is not limited to this particular application and may, for example, be used as a binder or adhesive or a waterresistant coating, irrespective of whether the composition serves to confer fire retardancy in such other applications.

[0024] The aluminosilicate as used in the present invention is typically formed by the sol-gel route and this can be effected in situ by forming the aluminosilicate at the point of use, by mixing precursor liquids. Thus, a second aspect of the present invention provides a precursor system for forming a coating composition according to the first aspect of the invention, the precursor system comprising:

[0025] (i) a metal aluminate, optionally in aqueous solution;

[0026] (ii) an aqueous solution of a metal silicate; and[0027] (iii) an organic liquid.

[0028] A sol-gel is basically a reaction product which is initially formed from the components of the precursor system as a liquid, but which subsequently forms a gel and ultimately solidifies. In order to form the gel-sol, either solid aluminate is admixed with an aqueous silicate solution or aqueous aluminate solution is admixed with an aqueous silicate solution.

[0029] At least part of the organic liquid may be incorporated in component (i) and/or component (ii). Alternatively, the organic liquid may initially be entirely separate from both of components (i) and (ii) and be admixed contemporaneously with, or subsequent to, the admixture of components (i) and (ii). Preferably, the organic liquid is incorporated into component (i) and/or (ii) prior to admixing the components to form the sol-gel system.

[0030] According to a third aspect of the present invention there is provided a composition comprising an aqueous solution of a metal aluminate and at least one organic liquid selected from the group consisting of polyhydroxy alcohols, mineral oils, liquid paraffin oils, glycol ethers, silicone oils and mixtures thereof. This composition is suitable as a part of the precursor system of the second aspect of the invention.

[0031] A fourth aspect of the invention provides an aqueous solution of a metal silicate and at least one organic liquid selected from the group consisting of polyhydroxy alcohols, mineral oils, liquid paraffin oils, glycol ethers, silicone oils and mixtures thereof. This composition is suitable as a part of the precursor system of the second aspect of the invention.

[0032] A further aspect of the present invention provides an application system for forming a coating composition according to the first aspect of the invention from a precursor system according to the second aspect of the invention and applying the coating composition so formed to a substrate, the application system comprising means for admixture of components (i), (ii) and (iii) and application means for effecting coating of the substrate with the resulting mixture. The application system for forming a coating composition and applying the coating composition so formed to a substrate suitable comprises a metal aluminate (i) stored in a first storage means, an aqueous solution of metal silicate (ii) in a second storage means and an organic liquid (iii) in a third storage means or in the first and/or second storage means, a means for admixture of components (i), (ii) and (iii) and application means for effecting coating of the substrate with the resulting mixture.

[0033] The organic liquid may be stored in its own, separate storage means, or may be admixed with either or both of the metal aluminate or the aqueous solution of metal aluminate in their respective storage means.

[0034] The metal aluminate may be in the form of an aqueous solution. Suitable storage means are tanks, containers or vessels in fluid connection with the means for admixture of the components. The transport of the components to and from the means for admixture may be effected by an arrangement of pumps and valves to meter the dosage of each component to the admixture means.

[0035] A further aspect of the present invention provides a method of making a coating composition according to the first aspect of the present invention, the method comprising admixture of the following components:

[0036] (i) a metal aluminate, optionally in aqueous solution;

[0037] (ii) an aqueous solution of a metal silicate; and [0038] (iii) an organic liquid.

[0039] Coatings formed from compositions according to the present invention exhibit superior physical integrity and long term stability, in comparison with conventional silicate systems. Without wishing to be bound by any theory, it is thought that this improvement arises by virtue of the aluminosilicate being present in the form of a network of bonded molecules that extends throughout the solution and by virtue of the presence of said organic liquid.

[0040] Typically the composition of the first aspect of the invention prior to application to a surface or substrate comprises at least 5% by weight of the aluminosilicate, and at least 0.1% by weight of said organic liquid.

[0041] For the sake of clarity, the preferred values for ingredients detailed below apply to all aspects of the invention. However the values are expressed in terms of the composition of the first aspect of the invention. When applied, for instance, to the precursor system or the application system or the precursor compositions, the preferred values apply to the values achieved in the resulting sol-gel composition.

[0042] The amount of water in compositions of the invention is preferably from 60% to 95%, more preferably from 70% to 90% by weight of the total composition.

[0043] The aluminosilicate is typically amorphous, which may be assessed by the absence of sharp peaks in the x-ray powder diffraction spectrum of the material. The mole ratio of Si:Al in the composition is typically from 3 to 30, preferably from 4 to 15 and more preferably from 5 to 10. In this context, the reference to mole ratio of Si:Al is based on the amount of silicon (in moles) in the silicate and aluminium (in moles) in the aluminate used for preparation of the compositions. The aluminosilicate is usually formed by the sol-gel route, preferably in situ from admixture of precursor components at the point of use. Compositions according the present invention preferably also comprise a metal or metal oxide to inhibit water absorption and/or to aid preservation of the film forming properties of the composition, especially film integrity, upon storage. The metal or oxide will usually be in particulate form and be sparingly soluble in water. Suitably the volume median particle diameter of the metal or metal oxide will be 50 µm or less. Preferably, less than 1% by volume of the metal or metal oxide particles will exceed 200 µm. Amphoteric or acidic oxides are typically employed for this purpose.

[0044] As used herein, the term "acidic oxide" means an oxide which reacts with either an alkali or base to form a salt plus water.

[0046] The metal oxide may, for example, be selected from amphoteric oxides of Group III elements, preferably aluminium boron and gallium oxides, or zinc oxide and mixtures thereof. Alternatively, the metal oxide may for example be selected from acidic oxides of Group IV elements, preferably silicon, tin oxides and germanium oxides, or zirconium oxide and mixtures thereof. Mixtures of one or more amphoteric oxides with one or more amphoteric oxides may also be used. Instead of introducing the metal in the form of an oxide, the oxide may alternatively be formed in situ as a result of adding the metal per se to the composition. Without wishing to be bound by theory, it is believed that the zinc or other metal oxide reacts with any residual silicate to reduce solubility of films formed by coating or otherwise applying the composition to substrates.

[0047] Preferably also, the amount of the metal oxide or metal is 0.1% to 10%, preferably from 0.3% to 5% by weight (e.g. from 0.3% to 3% by weight) of the total composition.

[0048] Compositions of the first aspect of the invention preferably comprise from 0.1% to 10%, preferably from 0.3% to 5% (e.g. 0.3% to 4%) by weight of the organic liquid.

[0049] Suitably, the organic liquid has a boiling point (at atmospheric pressure) greater than 110° C. Preferably, the organic liquid has a boiling point (at atmospheric pressure) of at least about 120° C., typically at least about 130° C. and typically up to about 500° C. Preferably the boiling point is no more than 500° C., preferably no more than 300° C.

[0050] The organic liquid is desirably one which is stable under alkaline conditions, by which is meant that it can withstand storage in an aqueous composition of pH 9 or more, preferably pH 12 or more without significant chemical degradation (i.e. less than 1% loss by weight of the liquid by degradation at 25° C. for 30 days storage) and also stable with respect to oxidation, heat and light.

[0051] The organic liquid is typically one having a viscosity of less than 5000 mPa·s, preferably less than 2000 mPa·s (e.g. less than 1000 mPa·s), at a temperature of 25° C. measured at a shear rate of 23 sec⁻¹.

[0052] The organic liquid may comprise one or more substantially water immiscible organic solvents selected from polyhydroxy alcohols, mineral oils, liquid paraffin oils, glycol ethers, silicone oils and mixtures thereof. Of these, silicone oils are especially preferred. It is preferred if the organic liquid is a silicone oil.

[0053] Suitable silicone oils for use in compositions according to the present invention and precursor systems therefore, are organosiloxanes, typically having the general formula (I):



wherein n is the number of repeating units in the polymer and can range from 2, e.g. from 10, up to 1,000,000, more preferably from 30, e.g. from 50, up to 500,000 and R_1 can be selected from hydrogen or methyl groups and R_2 can be

selected from hydrogen or SiR₅ in which R₅ can be either hydrogen, hydroxyl or methyl and wherein R₃ and R₄ can be independently selected from C₁ to C₁₂ straight chain or branched, saturated or unsaturated alkyl, alkenyl or phenyl moieties or from units according to formula (I) above or from substituted alkyl or substituted phenyl moieties in which substituents can be halogens, amino groups, sulphate groups, sulphonate groups, carboxy groups, hydroxy groups or nitro groups. Preferably, R₃ and R₄ are methyl groups. Preferably the silicone oils for use in the invention are free from halogen substituents.

[0054] One or more optional other ingredients may beneficially be incorporated in compositions according to any aspect of the present invention, e.g. in amounts from 0.001% to 5%, such as 0.01% to 2% by weight of the composition for any or each class, and may for example be selected from any of the classes:—

- **[0055]** (i) one or more surfactants, preferably selected from anionic, nonionic, cationic, amphoteric and zwitterionic surfactants and mixtures thereof, for example those which are known to be compatible with silicate and/or aluminate solutions, such as alkali caprybamphoproprionates;
- [0056] (ii) one or more phosphonates and/or phosphonic acids, such as triphenylphosphates and nitrilotric (methylene) triphosphoric acid;
- **[0057]** (iii) one or more slow proton releasing inorganic salts such as dihydrogen aluminium phosphates;
- **[0058]** (iv) one or more sequestrants such as EDTA or of the phosphonate type, eg those sold under the name Dequest; and
- [0059] (v) one or more isocyanates such as methylene di-isocyanate.

[0060] Compositions according to the present invention (which may optionally be prepared from a precursor system at the point of use) may for example be applied to the substrate by means of a spray gun (optionally air or gas pressurised), a roller system or a brush system. Alternatively the material to be treated may be coated or impregnated by immersion of the material in the coating composition while the coating composition is contained in a suitable vessel.

[0061] Compositions according to the present invention which are to be used as fire retardants may be applied to any appropriate flammable substrate but are especially suited to those which comprise an expanded or foamed polymer. Most preferably, that polymer is one which is substantially insoluble in the organic liquid at room temperature, i.e. the liquid component is selected with that requirement in mind.

[0062] Depending on the intended function of the cured composition, the composition may instead be applied to a substrate comprising one or more substances selected from wood, non-foamed polymer, metal, glass, ceramic, concrete, composite building material such as breezeblock, tile or brickwork, paper or china, or other vitreous ware.

[0063] When the composition of the invention is used to prepare fire retardant systems, it is preferred if the resulting system is substantially free of halogen-containing compounds, i.e. containing less than 1% by weight, preferably less than 0.5% by weight of such compounds.

[0064] Preferably, the moisture content of the resultant cured or dried composition film is no greater than 40%, more preferably no greater than 30%, still more preferably no greater than 25% and yet more preferably no greater than 20%

by weight. Even more preferably, the moisture content of the dried composition is 17% by weight or less.

[0065] The properties of the film resulting from the compositions of the invention may be improved by holding the composition at a temperature of 50° C. or more, while the composition is has a moisture content greater than 20% by weight, for at least 30 minutes.

[0066] In addition, the properties of the film, such as hydrophobicity or lubricity, may be enhanced by applying onto the film a low melting point wax, such as for example micronized polyethylene wax (a low molecular weight polyethylene polymer that is oxidized or non-oxidized and because of its low molecular weight has wax-like physical characteristics) or a stearate, such as glycol stearate (for example glycol tristrearate) or a metal stearate (for example Zn, Ca, Na, Mg Stearate) or a combination of one or more waxes and one or more stearates. The wax, stearate or mixture thereof should preferably have a melting point from 60° C. to 150° C., more preferably from 80° C. to 135° C. and most preferably from 90° C. to 130° C. For example Zinc Stearate, with a melting point of 120-130° C. can be applied onto the film to serve as a lubrication agent to facilitate further processing of the coated film when applied to a polymeric material.

[0067] Preferred coatings have a long term solubility of no greater than 25%, typically no greater than 20%, preferably no greater than 15%, and more preferably no greater than 10%, as determined by the water resistance/solubility methodology defined hereinafter, after oven drying the film at 80° C. to a water content of about 17% and then soaking in water at a temperature of about 22° C. for 7 days.

[0068] Another aspect of the invention provides a method of coating, impregnating or otherwise applying to a substrate which is other than an article or part of an article being substantially formed only of foamed polymer, the method comprising coating, impregnating or applying to said substrate, a composition according to the first aspect of the present invention.

[0069] The present invention will now be explained in more detail by way of the following non-limiting Examples.

EXAMPLE 1

Preparation of Aluminosilicate Sol-Gel with a Molar Ratio of Si/Al of 8.5 (Comparative)

[0070] 50 grams of sodium silicate solution (8.6% Na₂O, 29% SiO₂, balance water) was weighed directly into a plastic beaker. The silicate solution was vigorously stirred. To the vigorously stirred silicate solution, 40 grams of sodium aluminate solution (2.6% Na₂O, 3.6% Al₂O₃) was added dropwise over 3 to 4 minutes. After a further 10-20 seconds of mixing, stirring was stopped.

[0071] Approximately 25 grams of the resultant clear aluminosilicate sol formed was accurately weighed and poured onto a preweighed flat circular (~10 centimetre in diameter) plastic dish. The aluminosilicate sol was allowed to set for about 5 minutes to form a gel network. All of the foregoing steps were carried out under room temperature conditions (about 22° C.). The coated dish was placed in an oven at 80° C. for 24 hours and the plastic and the dried contents were then weighed. The dried aluminosilicate formed was a con-

tinuous and strong circular shaped sheet with about 1 mm thickness and its solid content was 74% by weight (i.e. water content of 26% by weight).

EXAMPLE 2

Comparative

[0072] The experiment of Example 1 was repeated but this time no aluminate solution was added and instead of the aluminate solution 37.5 grams of pure water was added. Once again, the resulting film (silicate only) was dried to a final moisture content of 26% by weight.

Water Resistance/Solubility Methodology

[0073] In order to test water resistance/solubility, the following procedure is adopted:

[0074] The dried film is first broken into large (about 2 cm across) pieces. 2.0 g of the pieces are put in a SterelinTM jar and 28 g of water added. The pieces are fully submerged in the water and left to stand for 24 hours at ambient temperature (about 22° C.). The contents of the solution are analysed (using titration and gravimetric methods) and the solubility of the pieces after 24 hours soaking, is determined using the following formula:

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Dissolved contents in the solution×100 2.0
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[0075] When this procedure was applied to the film obtained in Example 1 and also to the film as obtained in Example 2, the following solubility results were obtained.

	% Solubility
Example 1	5
Example 1 Example 2	100

[0076] These results clearly indicate that the formation of aluminosilicate films via the sol-gel process not only result in initially strong, solid films but also significantly enhances the water resistant property of the films produced.

[0077] The above results relate to a film which has been dried to a water content of 26% by weight. Elsewhere in this specification, the same methodology is employed except that the extent of drying and soaking may be varied as specified.

EXAMPLE 3

Effect of Organic Liquid

[0078] The film formed in Example 1 was strong and clear. However, when such film was oven dried beyond 24 hours, it gradually started to form white patches and within 72 hours, it had turned to a weak and white flaky/powdery material.

[0079] To enhance the integrity of the aluminosilicate film beyond 24 hours drying, a small amount of organic liquid was added to the silicate solution of Example 1. Addition of 0.5 g of silicone oil with a viscosity of 50 mPa·s, for example, maintained the integrity of the aluminosilicate film even after 168 hours of oven drying at 80° C. A number of organic liquids having various boiling points (BP) were tested (each added at a level of 0.5 g, corresponding to 0.55% by weight of the composition) and their impact on the integrity of aluminosilicate film after prolonged drying (168 hours of oven drying at 80° C.) are given below:

Organic Liquid tested	BP(° C.)	Film Integrity
Hexamethyl Silazane Hexamethyl Disiloxane (0.65 mPa · s) Silicone Oil (10 mPa · s) Silicone Oil (20 mPa · s) Silicone Oil (50 mPa · s) Silicone Oil (200 mPa · s) Silicone Oil (1000 mPa · s) Liquid Paraffin Oil	110 100 >150 >150 >150 >150 >150 >150 ~300	Powdery, white Powdery, white Clear, non-powdery Clear, non-powdery Clear, non-powdery Clear, non-powdery Clear, non-powdery

EXAMPLE 4

Effect of Film Moisture Content on Water Resistance

[0080] The dried film made according to Example 1 had a moisture content of 26%. When this film was soaked for 24 hours, it remained fairly intact. However, when the soaking in water was extended to 3 and then to 7 days, the solubility of the film increased proportionally. Reducing the moisture content of the film by for example prolonging drying time, would have minimised its water solubility. However, this is not an option for a film produced according to Example 1 as prolonged drying to minimise its moisture content, will result in weak and powdery film. However, the films made according to Example 3 and containing silicone oil had no such problem, and thus the silicone-oil-containing film of Example 3 using the 50 mPa·s silicone oil was dried to longer times to generate films with different moisture contents. The effect of film moisture content on the solubility of such film was conducted and the results are given below:

Solubility	y Results		
-	Days Soaked in Water		
% Moisture Content of Film	1	3	7
26	5%	20%	47%
22	4%	15%	30%
17	3%	4%	5%

[0081] The above table clearly indicate that an aluminosilicate film containing a water immiscible liquid such as silicone oil and having a moisture content of about 17% is significantly more resistant to water.

EXAMPLE 5

Long Term Water Resistance

[0082] The aluminosilicate film (with moisture content of 17%) made according to Example 4 and soaked for 7 days has good water resistance. However, when the same film is soaked for more than 7 days, for example 10 and 25 days, its water resistance decreases with increased soaking time.

[0083] To further improve the long-term water resistance of the aluminosilicate film, Example 4 was repeated but 1 gram of zinc oxide was added to and thereby suspended in the silicate and silicone oil mixture. The solubility of films made according to Examples 4 and 5 (with moisture contents of 17%) were assessed using the methodology defined hereinbefore using soaking times of 7, 10 and 25 days. The following results were obtained:

Solubility after Soaking for 7, 10 and 25 Days				
Sample	7 days	10 days	25 days	
Example 4 (without Zinc Oxide) Example 5 (with Zinc Oxide)	3% 1%	10% 1.3%	30% 4%	

[0084] As can be seen from the above results, addition of a small amount of zinc oxide enhances the long-term water resistance of films produced using aqueous aluminosilicate compositions obtained by the sol-gel route.

EXAMPLE 6

Production of Aluminosilicate Sol-Gel Film (Si/Al Ratio of 8.5) Using an In-Line Mixer

[0085] To stirred 1335 grams of sodium silicate solution (7.1% Na₂O, 23.9% SiO₂, balance water), 11 grams of silicone oil (viscosity mPa·s at 25° C.) was added. The silicone oil silicate mixture and sodium aluminate solution (4.6% Na₂O, 5.6% Al₂O₃) were simultaneously pumped to a high shear in-line mixer (with inlet ports adapted to suit the viscosity of the resultant sol) at the rate of 253 ml/minute and 107 mls/minute respectively. A clear aluminosilicate sol was formed and a sample of this sol was treated and characterised in the same way as in Example 1.

EXAMPLE 7

[0086] The same as Example 6 but with 22 grams of zinc oxide added to the silicate/silicone mixture

[0087] The results of solubility tests for samples of Examples 6 and 7 dried to moisture content of 17% and soaked in water for 7, 10 and 14 days were as follows:

Solubility after Soaking for 7, 10 and 14 Days					
Sample	7 days	10 days	14 days		
Example 6 (without Zinc Oxide) Example 7 (with Zinc Oxide)	2.7% 1%	9.5% 1.2%	13% 2.2%		

1. An aqueous gel-forming composition comprising:

(a) from 5% to 40% by weight of an aluminosilicate,

- (b) from 0.1% to 10% by weight of an organic liquid,
- wherein the organic liquid has a boiling point of greater than 110° C.

2. A composition according to claim **1** comprising from 5% to 30% by weight of an aluminosilicate.

3. A composition according to claim **1**, in which the organic liquid has a boiling point of at least 120° C.

4. A composition according to claim **1**, wherein the organic liquid has a boiling point of no more than 500° C.

5. A composition according to claim **1**, in which the organic liquid is substantially water immiscible.

6. A composition according to claim **1**, in which the organic liquid is stable under alkaline conditions.

7. A composition according to claim 1, in which the organic liquid has a viscosity of less than 5,000 mPa s at a temperature of 25° C.

8. A composition according to claim **1**, in which the organic liquid comprises a liquid selected from the group consisting of mineral oils, liquid paraffin oils, silicone oils and mixtures thereof.

9. A composition according to claim **1**, in which the organic liquid comprises a liquid selected from the group consisting of polyhydroxy alcohols, glycol ethers and mixtures thereof.

10. A composition according to claim **1** wherein the organic liquid is a silicone oil.

11. A composition according to claim **1**, further comprising at least one metal or metal oxide.

12. A composition according to claim **11**, in which the metal oxide comprises an amphoteric oxide.

13. A composition according to claim **12**, wherein the amphoteric oxide is selected from the group consisting of amphoteric oxides of Group III elements, zinc oxide and mixtures thereof.

14. A composition according to claim 11, in which the metal oxide comprises an acidic oxide.

15. A composition according to claim **14**, wherein the acidic oxide is selected from the group consisting of acidic oxides of Group IV elements, zirconium oxide and mixtures thereof.

16. A composition according to claim **11**, comprising up to 10% by weight of the metal or metal oxide.

17. A composition according to claim **1**, in which the aluminosilicate has a Si:Al mole ratio from **3** to **30**.

18-20. (canceled)

21. An application system for forming a coating composition and applying the coating composition so formed to a substrate, the application system comprising

- a metal aluminate (i) stored in a first storage means, an aqueous solution of metal silicate (ii) in a second storage means and an organic liquid (iii) in a third storage means or in the first and/or second storage means,
- a means for admixture of components (i), (ii) and (iii) to form an aqueous gel-forming coating composition according to claim **1**, and

application means for effecting coating of the substrate with the resulting coating composition.

22. An application system according to claim 21, in which the application means further comprises spray means, roller means, brush means or a vessel for containing the coating composition into which products to be coated or impregnated can be immersed.

23. A substrate being other than an article or part of an article substantially formed only of foamed polymer, the substrate being coated, impregnated or otherwise applied with a dried or cured composition according to claim **1**.

24. A substrate according to claim **23**, in which the moisture content of the dried or cured composition is no greater than 40% by weight.

25. A film of aluminosilicate produced from a composition according to claim **1**, the long term water solubility of the aluminosilicate film being no greater than 25%.

26. A method of making a coating composition according to claim **1**, the method comprising admixing the following components:

(i) a metal aluminate;

(ii) an aqueous solution of a metal silicate; and

(iii) an organic liquid.

27. A method of coating, impregnating or otherwise applying to a substrate which is other than an article or part of an article being substantially formed only of foamed polymer, the method comprising coating, impregnating or applying to said substrate, a composition according to claim **1**.

28. A method according to claim **27** comprising the subsequent step of drying the composition to a moisture content of 40% by weight or less.

29. A method according to claim **27**, comprising the step of gluing the substrate to a second substrate with the composition.

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