



(51) International Patent Classification:

C09D 5/00 (2006.01)

C09D 5/20 (2006.01)

TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, WS, ZA, ZM, ZW.

(21) International Application Number:

PCT/CN2023/100060

(84) Designated States (unless otherwise indicated, for every

kind of regional protection available): ARIPO (BW, CV, GH, GM, KE, LR, LS, MW, MZ, NA, RW, SC, SD, SL, ST, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, RU, TJ, TM), European (AL, AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HR, HU, IE, IS, IT, LT, LU, LV, MC, ME, MK, MT, NL, NO, PL, PT, RO, RS, SE, SI, SK, SM, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, KM, ML, MR, NE, SN, TD, TG).

(22) International Filing Date:

14 June 2023 (14.06.2023)

(25) Filing Language:

English

(26) Publication Language:

English

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Published:

— with international search report (Art. 21(3))

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(81) Designated States (unless otherwise indicated, for every

kind of national protection available): AE, AG, AL, AM, AO, AT, AU, AZ, BA, BB, BG, BH, BN, BR, BW, BY, BZ, CA, CH, CL, CN, CO, CR, CU, CV, CZ, DE, DJ, DK, DM, DO, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, GT, HN, HR, HU, ID, IL, IN, IQ, IR, IS, IT, JM, JO, JP, KE, KG, KH, KN, KP, KR, KW, KZ, LA, LC, LK, LR, LS, LU, LY, MA, MD, MG, MK, MN, MU, MW, MX, MY, MZ, NA, NG, NI, NO, NZ, OM, PA, PE, PG, PH, PL, PT, QA, RO, RS, RU, RW, SA, SC, SD, SE, SG, SK, SL, ST, SV, SY, TH,

(54) Title: PEELABLE COATING COMPOSITION AND USE THEREOF

(57) Abstract: This disclosure relates to a peelable coating composition which dries/cures to a peelable coating. The composition comprises an aqueous styrene-(meth)acrylic emulsion copolymer binder in combination with a silicone polyether copolymer. The resulting peelable coating provides temporary protection to articles and/or substrates during a period of construction, storage or transportation or the like, and is designed to be water-resistant such that it is able to continue protecting substrates after periods of time in the open-air, exposed to the elements, e.g., rain.



PEELABLE COATING COMPOSITION AND USE THEREOF

This disclosure relates to a peelable coating composition which dries/cures to a peelable coating.

The composition comprises an aqueous styrene-(meth)acrylic emulsion copolymer binder in combination with a silicone polyether copolymer. The resulting peelable coating provides temporary protection to articles and/or substrates during a period of construction, storage or transportation or the like, and is designed to be water-resistant such that it is able to continue protecting substrates after periods of time in the open-air, exposed to the elements, e.g., rain.

In recent years, there has been an increasing demand for peelable coatings that can be applied to a wide range of substrates as described in US8440759, US6822012, US6620890 and CN102850923.

Peelable coatings, sometimes alternatively referred to as strippable coatings, are designed to adhere to substrates sufficiently well so that, whilst the coating does not spontaneously peel off from the substrate surface being protected, it remains durable and can be easily peeled off the substrate as and when required without cracking, tearing or breaking the coating in any other way. Hence, they are temporary protective coatings which are removeable by being peeled from the substrate surface after use. The peelable coating compositions need to provide a substrate on to which they have been applied with appropriate physical properties to provide peelable coatings with excellent film properties such as light resistance and thermal stability, whilst also providing chemical and/or physical protection and enabling the coatings to be peeled off in continuous and sizable sheets from the substrate after a period of use.

They are designed to be a cost-effective and time-efficient means for protection of a wide range of substrate surfaces such as metals, plastics, glass and construction materials such as concrete during periods of construction, storage or transportation. Hence, in the case of construction applications such peelable coatings may be used to protect glass such as façade glass, and window glass as well as metal, plastic and/or wooden door frames and window frames and indeed concrete articles etc. during construction whilst being easily removed after construction has been completed. Similarly, peelable coatings may be used to protect vehicles, machine parts, metallic household articles and other ferrous and non-ferrous articles, wooden articles, glass articles, rubber articles, and coated rubber articles during transportation or storage.

They are used, for the sake of example, to protect substrate surfaces from weathering caused by sunlight wind and rain as well as scratches, stains and discoloration damage and/or contamination due to sand, dust, iron powder, salts, alkalis, acids, soot and smoke, insects and bird excrement and the like.

Many commercial peelable coatings require the use of one or more release agents to prevent permanent adhesion of the film to the substrate and to enhance the peelability of the film from the substrate surface onto which it was applied after the period of protection is complete. The industry often prefers to avoid release agents as historically they were often based on materials containing fatty acids which are prone to undergo unwanted reactions with metal and stain masonry and wood.

That said it is known that many peelable coatings don't function well after having been subjected to weathering such as after being rained on and then drying out in the sun. Such situations for coatings in exterior situations can lead to a serious loss in coating durability with the coatings cracking and tearing when being removed from the substrate, potentially as a consequence of the coating being too strongly adhered to the substrate surface resulting in an inability to remove the coating by hand. This has been the situation with coatings prepared from peelable coating compositions having one or more binders or film formers (hereafter referred to as "binder(s)") comprising or consisting of aqueous emulsions or aqueous dispersions.

There is provided herein a peelable coating composition comprising

- 10 (a) an aqueous styrene-(meth)acrylic emulsion copolymer binder, wherein the copolymer is derived from at least monomers (a)(1), (a)(2) and (a)(3) wherein

(a)(1) is a styrene-based monomer in an amount of from 17.5 wt. % to 45 wt. %, of the solids content of the starting ingredients

- 15 (a)(2) is at least one (meth)acrylic acid monomer in an amount of from 0.1 wt. % to 4.5 wt. % of the solids content of the starting ingredients; and

(a)(3) at least one (meth)acrylate monomer;

Which copolymer is not derived from a vinyl acetate monomer as a starting ingredient and is not derived from a (meth)acrylic ester monomer having a reactive methylene group as a starting ingredient;

- 20 (b) a silicone polyether copolymer in an amount of from 0.75 to 10 wt. % of the peelable coating composition selected from one or both of

(i) $\text{RO}(\text{C}_3\text{H}_6\text{O})_m(\text{CH}_2\text{CH}_2\text{O})_{n''}\text{R}^{15}-((\text{CH}_3)_2\text{SiO})_x(\text{CH}_3)_2\text{Si}-\text{R}^{16}(\text{OCH}_2\text{CH}_2)_{n'}(\text{OC}_3\text{H}_6)_m\text{OR}^1$, where:-
R and R^1 may be the same or are different and are each selected from H, or an alkyl group,
($\text{C}_3\text{H}_6\text{O}$) is ($\text{CH}_2(\text{CH}_3)\text{CHO}$), ($\text{CH}_2\text{CH}_2\text{CH}_2\text{O}$), ($(\text{CH}_3)\text{CHCH}_2\text{O}$) or mixtures thereof;

- 25 (OC_3H_6) is ($\text{OCH}_2\text{CH}(\text{CH}_3)$), ($\text{OCH}_2\text{CH}_2\text{CH}_2$), ($\text{OCH}(\text{CH}_3)\text{CH}_2$) or mixtures thereof;
 R^{15} and R^{16} are the same or different and are alkylene groups having from 2 to 6 carbons,
x is from 2 to about 500, m and m' may be the same or are different and are each in a range of from 0 to about 50 and n'' and n' may be the same or are different and are in a range of from 3 to about 50;

- 30 or

(ii) $(\text{CH}_3)_3\text{SiO}-((\text{CH}_3)_2\text{SiO})_x-(\text{CH}_3\text{R}'\text{SiO})_y-\text{Si}(\text{CH}_3)_3$;

Wherein $\text{R}' = -\text{R}^{17}-(\text{OCH}_2\text{CH}_2)_{n'}(\text{OC}_3\text{H}_6)_m\text{OR}$;

where R, x, m' , n' and (OC_3H_6) are the same as above R^{17} is an alkylene group having from 2 to 6 carbons, and y is from 1 to about 100; and optionally

- 35 (c) one or more additives selected from aqueous solvent, coalescents, defoamers, rheology modifiers, wetting agents, plasticisers, pigments and coloring agents and/or a mixture thereof.

There is also provided a substrate coated with a temporary peelable coating which coating is the cured/dried product of the above peelable coating composition.

There is also provided a method of forming a peelable coating on a substrate, the method comprising the steps of:

(I') combining an aqueous styrene-(meth)acrylic emulsion copolymer binder (a) as hereinbefore described; with

a silicone polyether copolymer (b) in an amount of from 0.75 to 10 wt. % of the peelable coating composition selected from one or both of

- 10 (i) $\text{RO}(\text{C}_3\text{H}_6\text{O})_m(\text{CH}_2\text{CH}_2\text{O})_n\text{R}^{15}-((\text{CH}_3)_2\text{SiO})_x(\text{CH}_3)_2\text{Si}-\text{R}^{16}(\text{OCH}_2\text{CH}_2)_n'(\text{OC}_3\text{H}_6)_m'\text{OR}^1$, where:-
 R and R^1 may be the same or are different and are each selected from H, or an alkyl group,
 $(\text{C}_3\text{H}_6\text{O})$ is $(\text{CH}_2(\text{CH}_3)\text{CHO})$, $(\text{CH}_2\text{CH}_2\text{CH}_2\text{O})$, $((\text{CH}_3)\text{CHCH}_2\text{O})$ or mixtures thereof;
 (OC_3H_6) is $(\text{OCH}_2\text{CH}(\text{CH}_3))$, $(\text{OCH}_2\text{CH}_2\text{CH}_2)$, $(\text{OCH}(\text{CH}_3)\text{CH}_2)$ or mixtures thereof;
 R^{15} and R^{16} are the same or different and are alkylene groups having from 2 to 6 carbons,

- 15 x is from 2 to about 500, m and m' may be the same or are different and are each in a range of from 0 to about 50 and n'' and n' may be the same or are different and are in a range of from 3 to about 50;

or

- (ii) $(\text{CH}_3)_3\text{SiO}-((\text{CH}_3)_2\text{SiO})_x-(\text{CH}_3\text{R}'\text{SiO})_y-\text{Si}(\text{CH}_3)_3$;

- 20 Wherein $\text{R}' = -\text{R}^{17}-(\text{OCH}_2\text{CH}_2)_n'(\text{OC}_3\text{H}_6)_m'\text{OR}$;

where R, x, m' , n' and (OC_3H_6) are the same as above R^{17} is an alkylene group having from 2 to 6 carbons, and y is from 1 to about 100; and optionally

(c) one or more additives selected from aqueous solvent, coalescents, defoamers, rheology modifiers, wetting agents, plasticisers, pigments and coloring agents and/or a mixture thereof, to form a peelable coating composition,

(II') applying the peelable coating composition on a substrate surface;

(III') drying the peelable coating composition applied in step (II') to form a peelable coating on the substrate surface. The peelable coating resulting from this method is a temporary coating which protects the surface of the substrate to which the peelable coating composition has been applied.

30 There is also provided a substrate coated with a peelable coating obtained or obtainable in accordance with the above process. The substrate coated with a peelable coating is thereby provided with a temporary coating which protects the surface of the substrate to which the peelable coating composition has been applied. The peelable coating is removable from the substrate surface by peeling when required.

35

There is also provided a use of (a) an aqueous styrene-(meth)acrylic emulsion copolymer binder, wherein the copolymer is derived from at least monomers (a)(1), (a)(2) and (a)(3) wherein

(a)(1) is a styrene-based monomer in an amount of from 17.5 wt. % to 45 wt. % of the solids content of the starting ingredients

5 (a)(2) is at least one (meth)acrylic acid monomer in an amount of from 0.1wt. % to 4.5 wt. % of the solids content of the starting ingredients; and

(a)(3) at least one (meth)acrylate monomer;

Which copolymer is not derived from a vinyl acetate monomer as a starting ingredient and is not derived from a (meth)acrylic ester monomer having a reactive methylene group as a starting

10 ingredient;

; in a peelable coating composition

which peelable coating composition otherwise comprises

a silicone polyether copolymer (b) in an amount of from 0.75 to 10 wt. % of the peelable coating composition selected from one or both of

15 (i) $\text{RO}(\text{C}_3\text{H}_6\text{O})_m(\text{CH}_2\text{CH}_2\text{O})_n\text{R}^{15}-((\text{CH}_3)_2\text{SiO})_x(\text{CH}_3)_2\text{Si}-\text{R}^{16}(\text{OCH}_2\text{CH}_2)_{n'}(\text{OC}_3\text{H}_6)_{m'}\text{OR}^1$, where:-

R and R¹ may be the same or are different and are each selected from H, or an alkyl group,

(C₃H₆O) is (CH₂(CH₃)CHO), (CH₂CH₂CH₂O), ((CH₃)CHCH₂O) or mixtures thereof;

(OC₃H₆) is (OCH₂CH(CH₃)), (OCH₂CH₂CH₂), (OCH(CH₃)CH₂) or mixtures thereof;

R¹⁵ and R¹⁶ are the same or different and are alkylene groups having from 2 to 6 carbons,

20 x is from 2 to about 500, m and m' may be the same or are different and are each in a range of from 0 to about 50 and n'' and n' may be the same or are different and are in a range of from 3 to about 50;

or

(ii) $(\text{CH}_3)_3\text{SiO}-((\text{CH}_3)_2\text{SiO})_x-(\text{CH}_3\text{R}'\text{SiO})_y-\text{Si}(\text{CH}_3)_3$;

25 Wherein R' = -R¹⁷-(OCH₂CH₂)_{n'}(OC₃H₆)_{m'}OR;

where R, x, m', n' and (OC₃H₆) are the same as above R¹⁷ is an alkylene group having from 2 to 6 carbons, and y is from 1 to about 100

and optionally

(c) one or more additives selected from aqueous solvent, coalescents, defoamers, rheology

30 modifiers, wetting agents, plasticisers, pigments and coloring agents and/or a mixture thereof.

For the avoidance of doubt the total wt. % of all ingredients in the composition is 100 wt. %. The term "peelable" is intended to mean that the coating applied onto the substrate as described above is peelable, i.e., removably or temporarily adhered to a substrate. Hence the peelable coating is

adhered to the substrate but is removeable therefrom as or when required by peeling the film away

35 from the substrate surface. The use of the term "(meth)" in (meth)acrylates as used throughout the disclosure, is intended to mean acrylates or methacrylates or mixtures thereof.

The peelable coating is a temporary coating provided to protect a substrate surface. It is designed to be water resistant and functions as a protective coating over the substrate onto which it is applied. It is designed to be removeable from the substrate surface by peeling therefrom. Use of the term water-resistant is intended to mean that the peelable coating as described herein is able to resist water penetration to the extent that the peelability and adhesion of the peelable coating to the substrate is not noticeably negatively affected after being exposed to water in the form of rain or the like. It is intended to be used as a means of temporary protection for a period of up to a year but may be used for long term protection if desired.

The components of the peelable coating composition will hereafter be described in more detail.

10 **Binder (a)**

The binder (a) in the above peelable coating composition is an aqueous styrene-(meth)acrylic emulsion copolymer. The aqueous styrene-(meth)acrylic emulsion copolymer as herein described may be a random, block or alternating copolymer or a mixture thereof.

The aqueous styrene-(meth)acrylic emulsion copolymer as herein described is a copolymer of at least three monomers (a)(1), (a)(2) and (a)(3) wherein

(a)(1) is a styrene-based monomer in an amount of from 17.5 wt. % to 45 wt. %, of the monomer content of the starting ingredients

(a)(2) is at least one (meth)acrylic acid monomer in an amount of from 0.1 wt. % to 4.5 wt. % of the monomer content of the starting ingredients; and

20 (a)(3) at least one (meth)acrylate monomer;

For the avoidance of doubt the monomer content values given above are the wt. % of each monomer on a basis of the total monomer content being the total monomer content e.g., 100wt. % of the monomer content. The solids content is the non-solvent content, i.e., in this case non-water starting ingredients for making the aqueous styrene-(meth)acrylic emulsion copolymer.

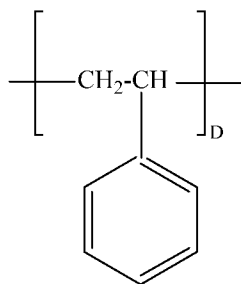
25 The aqueous styrene-(meth)acrylic emulsion copolymer does not contain any repeating vinyl acetate units. Vinyl acetate is not a starting ingredient for the aqueous styrene-(meth)acrylic emulsion copolymer as described herein.

Likewise, the aqueous styrene-(meth)acrylic emulsion copolymer does not contain any repeating derived from a (meth)acrylic ester monomer having a reactive methylene group as a starting ingredient such as acetoacetoxyethyl methacrylate which is sometimes referred as a “self-crosslinker”. No a (meth)acrylic ester monomer having a reactive methylene group is a starting ingredient for the aqueous styrene-(meth)acrylic emulsion copolymer as described herein.

Regarding the (a)(1), (a)(2) and (a)(3) monomers:

(a)(1) the styrene-based monomer

In the case of monomer (a)(1) the styrene-based monomer may be styrene (also known as vinyl benzene and ethenyl benzene) which upon polymerisation generates repeat units as follows:

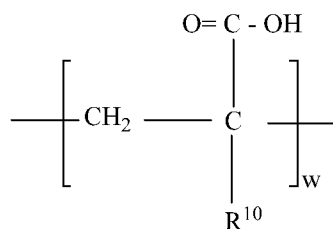


Where D is a whole number. Alternatively, the styrene-based monomer may be a substituted styrene such as an alkyl styrene/vinyl toluene such as 2-alkylstyrene, a 3-alkylstyrene or a 4-alkylstyrene where the alkyl groups contain 1 to 4 carbons, e.g., methyl, ethyl, propyl, butyl or tertiary butyl groups or alkoxy styrenes, specific examples of the substituted styrenes being alpha-methylstyrene, trans-beta-methylstyrene, 2,4-dimethylstyrene, ethylstyrene, butylstyrene, and p-methoxystyrene; o-, m-, and p-methoxystyrene; and p-trifluoromethylstyrene. Preferably the styrene-based monomer is styrene.

The styrene-based monomer is present in the copolymer in an amount of from 17.5 wt. % to 45 wt. %, based on the monomer content of the starting ingredients. In one embodiment the styrene-based monomer is present in the polymerisation starting ingredients in an amount of from 17.5 wt. % to 45 wt. % of the monomer content of the starting ingredients, alternatively of from 17.5 wt. % to 40 wt. % of the monomer content of the starting ingredients, alternatively of from 17.5 wt. % to 37.5 of the monomer content of the starting ingredients, alternatively of from 20.0 wt. % to 37.5 wt. % of the monomer content of the starting ingredients.

(a)(2) (meth)acrylic acid monomer

(a)(2) is at least one (meth)acrylic acid monomer in an amount of from 0.1 wt. % to 4.5 wt. % of the solids content of the starting ingredients, alternatively in an amount of from 0.1 wt. % to 4.0 wt. %, alternatively in an amount of from 0.1 wt. % to 3.75 wt. %. Typically (meth)acrylic acid is intended to extend to both acrylic acid (AA) and methacrylic acid (MAA) and as such component (a)(2) is either acrylic acid or methacrylic acid which has the following repeating units



Which comprise a carboxylic acid group and where R^{10} is either hydrogen (in the case of an acrylic acid monomer or a methyl group in the case of methacrylic acid as monomer.

(a)(3) at least one (meth)acrylate monomer.

The at least one (meth)acrylate monomer of component (a)(3) may be any suitable methacrylate monomer. For example, C₁-C₂₀-alkyl esters of (meth)acrylic acid, alternatively C₁-C₁₀-alkyl esters of (meth)acrylic acid or C₁-C₈-alkyl esters of (meth)acrylic acid, such as methyl (meth)acrylate, ethyl (meth)acrylate, butyl (meth)acrylate (BAA and BMAA), decyl (meth)acrylate, lauryl (meth)acrylate, isodecyl (meth)acrylate, hydroxyethyl methacrylate, hydroxypropyl methacrylate, 2-ethylhexyl (meth)acrylate (2-EHMA and 2-EHA), 2-propylheptyl (meth)acrylate, and neopentyl (meth)acrylate.

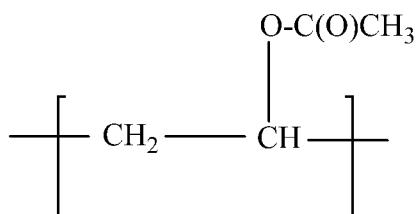
An aqueous styrene-(meth)acrylic emulsion copolymer may be made using more than one alkyl (meth)acrylate and may incorporate other organic monomers such as Aryl (meth)acrylate monomers such as phenyl (meth)acrylate and tolyl (meth)acrylate; Aralkyl (meth)acrylate monomers such as benzyl (meth)acrylate and phenethyl (meth)acrylate; cycloalkyl (meth)acrylates such as cyclohexyl (meth)acrylate (CHMA and CHA), 1-adamatyl (meth)acrylate; (meth)acrylamide; (meth)acrylonitrile; ureido-functional monomers such as uriedo (meth)acrylate (UMA and UA) and hydroxyethyl ethylene urea methacrylate; isobornyl methacrylate, isobornyl acrylate, and dihydrodicyclopentadienyl acrylate; and hydroxyalkyl(meth)acrylate such as hydroxyethyl (meth)acrylate HEMA or HEA) or combinations thereof.

Other monomers may be utilised in the aqueous styrene-(meth)acrylic emulsion copolymer in addition to (a)(1), (a)(2) and (a)(3), if desired. These may include organic diacids such as itaconic acid, butadiene; α -olefins such as ethylene, propylene, and 1-decene; ; glycidyl (meth)acrylate; or combinations thereof, as well as silicon containing monomers such as vinyltrialkoxysilanes such as vinyltrimethoxysilane, vinyltriethoxysilane, and vinyltris(2-methoxyethoxy)silane, vinyldimethylethoxysilane, vinylmethyldiethoxysilane, and (meth)acryloxyalkyltrialkoxysilanes such as (meth)acryloxyethyltrimethoxysilane and (meth)acryloxypropyltrimethoxysilane or combinations thereof.

However, in a preferred embodiment component (a)(3) makes up the difference between 100 wt. % and the cumulative wt. % present of components (a)(1) and (a)(2), other than the catalyst used for polymerisation.

In one embodiment of the invention the aqueous styrene-(meth)acrylic emulsion copolymer comprises more than one monomer from component (a)(2) and/or (a)(3). In a further embodiment the aqueous styrene-(meth)acrylic emulsion copolymer is a ter-polymer consisting of one component (a)(1) monomer, one component (a)(2) monomer and one component (a)(3) monomer.

As previously mentioned herein the aqueous styrene-(meth)acrylic emulsion copolymer is not prepared using vinyl acetate monomer ($\text{CH}_2=\text{CH}-\text{O}-\text{C}(\text{O})\text{CH}_3$) and as such does not contain polyvinyl acetate repeating units



- 5 Likewise, the aqueous styrene-(meth)acrylic emulsion copolymer does not contain any repeating derived from a (meth)acrylic ester monomer having a reactive methylene group as a starting ingredient e.g., acetoacetoxyethyl methacrylate (AAEMA) which has the structure:-



- 10 The aqueous styrene-(meth)acrylic emulsion copolymer as described herein further comprises water, typically deionized water, in an amount of from 30% to 90%, from 40% to 80%, from 50% to 70%, or from 55% to 60%, by weight based on the total weight of the aqueous dispersion.

- The aqueous styrene-(meth)acrylic emulsion copolymer as described herein may be prepared by emulsion polymerization of a mixture of the monomers described above. Total weight concentration of the mixture of monomers for preparing the emulsion polymer is equal to 100%. The dosage of
15 such monomer based on the total weight of the monomers, is substantially the same as the weight amount of each of these monomers as structural units in the emulsion polymer.

- The mixture of monomers may be added neat or as an emulsion in water; or added in one or more additions or continuously, linearly or nonlinearly, over the reaction period of preparing the polymer. Temperature suitable for free-radical polymerization process may be lower than 100° C., in the
20 range of from 10° C. to 95° C., or in the range of from 50° C. to 90° C. One or more surfactants may be used in preparing the polymer. In one embodiment an emulsion of monomers is prepared prior to the polymerisation process.

- The emulsion polymerisation may be initiated/catalysed by thermal, redox (using redox catalysts), photochemical, and electrochemical initiation, however, the polymerisation process is usually
25 initiated/catalysed using one or more conventional free radical initiators for example, peroxides, such as, for example, hydrogen peroxide, sodium or potassium hydroperoxide, t-alkyl peroxides, t-alkyl hydroperoxides e.g., dicumyl hydroperoxide, t-amyl hydroperoxide, t-butyl hydroperoxide; t-alkyl peresters, wherein the t-alkyl group includes at least 5 carbon atoms; perboric acids and their salts, such as, for example, sodium perborate; perphosphoric acids and salts thereof; ammonium
30 and/or alkali persulfates, potassium permanganate; and ammonium or alkali metal salts of peroxydisulfuric acid. Such initiators may be used in amounts ranging from 0.01 to 3.0 wt. % (weight percent), based on the total weight of monomers.

Redox systems comprising the above-described initiators coupled with a suitable reductant may be used in the polymerization process. Examples of suitable reductants include sodium sulfoxylate formaldehyde, ascorbic acid, isoascorbic acid, alkali metal and ammonium salts of sulfur-containing acids, such as sodium sulfite, bisulfite, thiosulfate, hydrosulfite, sulfide, hydrosulfide or dithionite, formadinesulfinic acid, acetone bisulfite, glycolic acid, hydroxymethanesulfonic acid, glyoxylic acid hydrate, lactic acid, glyceric acid, malic acid, tartaric acid and salts of the proceeding acids. Metal salts of iron, copper, manganese, silver, platinum, vanadium, nickel, chromium, palladium, or cobalt may be used to catalyze the redox reaction. Chelating agents for the metals may optionally be used.

Optionally, one or more chain transfer agents may be used in the polymerization process to control the molecular weight of the emulsion polymer. Examples of suitable chain transfer agents include 3-mercaptopropionic acid, methyl 3-mercaptopropionate, butyl 3-mercaptopropionate, n-dodecyl mercaptan, n-hexadecanethiol, tert-dodecyl mercaptan, n-octadecanethiol, benzenethiol, azelaic alkyl mercaptan, hydroxy group containing mercaptans such as hydroxyethyl mercaptan, mercaptopropionic acid, and mixtures thereof. When present the chain transfer agent may be used in an amount of up to 2%, alternatively up to 1.5%, alternatively up to 1%, alternatively up to 0.5% by weight based on the total weight of the monomers used for preparing the emulsion polymer.

Upon completion of the polymerization process, the resulting aqueous styrene-(meth)acrylic emulsion copolymer may be neutralized by one or more bases as neutralizers to a pH value, for example, at least 6, from 6 to 10, or from 7 to 9.

The bases may lead to partial or complete neutralization of the ionic or latently ionic groups of the emulsion polymer. Examples of suitable bases include ammonia; alkali metal or alkaline earth metal compounds such as sodium hydroxide, potassium hydroxide, calcium hydroxide, zinc oxide, magnesium oxide, sodium carbonate; primary, secondary, and tertiary amines, such as triethyl amine, ethylamine, propylamine, monoisopropylamine, monobutylamine, hexylamine, ethanolamine, diethyl amine, dimethyl amine, tributylamine, triethanolamine, dimethoxyethylamine, 2-ethoxyethylamine, 3-ethoxypropylamine, dimethylethanolamine, diisopropanolamine, morpholine, ethylenediamine, 2-diethylaminoethylamine, 2,3-diaminopropane, 1,2-propylenediamine, neopentanediamine, dimethylaminopropylamine, hexamethylenediamine, 4,9-dioxadodecane-1,12-diamine, polyethyleneimine or polyvinylamine; aluminum hydroxide; or mixtures thereof.

The aqueous styrene-(meth)acrylic emulsion copolymer herein may be of any suitable weight average molecular weight e.g., an amount of from 10,000 to 5,000,000 g/mol or more, i.e. in the range of from 10,000 to 5,000,000 g/mol, for example from 250,000 to 1,000,000 g/mol, alternatively from 50,000 to 1,000,000 g/mol, alternatively from 50,000 to 1,000,000 g/mole, alternatively from 100,000 to 900,000 g/mol, alternatively from 100,000 to 750,000 g/mol, alternatively from 150,000 to 500,000 g/mole, alternatively from 150,000 to 400,000 g/mole.

As used herein, unless otherwise indicated, the phrase “molecular weight” with respect to the aqueous styrene-(meth)acrylic emulsion copolymer refers to the weight average molecular weight as measured by gel permeation chromatography (GPC) against polystyrene (PS) standards.

In one embodiment the aqueous styrene-(meth)acrylic emulsion copolymer as disclosed herein has a glass transition temperature (T_g) in the range of from 5 to 40° C. As used herein, unless otherwise indicated, the term “ T_g ” or “glass transition temperature” of a polymer, with regard to the aqueous styrene-(meth)acrylic emulsion copolymer and components thereof, refers to the T_g of a polymer calculated by using the Fox equation (T. G. Fox, Bull. Am. Physics Soc., Volume 1, Issue No. 3, page 123 (1956)).

If desired, a mixture of aqueous styrene-(meth)acrylic emulsion copolymers may be present with the copolymer (mixture) being an emulsion of liquid in water and/or small polymer particles in water.

The aqueous styrene-(meth)acrylic emulsion copolymer is present in the peelable coating composition in an amount of from 70 wt. % to 99.25 wt. % of the peelable coating composition.

Silicone polyether copolymer (b)

The silicone polyether copolymer (b) is present in the peelable coating composition in an amount of from 0.75 to 10 wt. % of the peelable coating composition. It is selected from one or both of

(i) $\text{RO}(\text{C}_3\text{H}_6\text{O})_m(\text{CH}_2\text{CH}_2\text{O})_n\text{R}^{15}-((\text{CH}_3)_2\text{SiO})_x(\text{CH}_3)_2\text{Si}-\text{R}^{16}(\text{OCH}_2\text{CH}_2)_n(\text{OC}_3\text{H}_6)_m\text{OR}^1$, where:-
R and R^1 may be the same or are different and are each selected from H, or an alkyl group,

$(\text{C}_3\text{H}_6\text{O})$ is $(\text{CH}_2(\text{CH}_3)\text{CHO})$, $(\text{CH}_2\text{CH}_2\text{CH}_2\text{O})$, $((\text{CH}_3)\text{CHCH}_2\text{O})$ or mixtures thereof;

(OC_3H_6) is $(\text{OCH}_2\text{CH}(\text{CH}_3))$, $(\text{OCH}_2\text{CH}_2\text{CH}_2)$, $(\text{OCH}(\text{CH}_3)\text{CH}_2)$ or mixtures thereof;

R^{15} and R^{16} are the same or different and are alkylene groups having from 2 to 6 carbons,

x is from 2 to about 500, m and m' may be the same or are different and are each in a range of from 0 to about 50 and n'' and n' may be the same or are different and are in a range of from 3 to about 50;

or

(ii) $(\text{CH}_3)_3\text{SiO}-((\text{CH}_3)_2\text{SiO})_x-(\text{CH}_3\text{R}'\text{SiO})_y-\text{Si}(\text{CH}_3)_3$;

Wherein $\text{R}' = -\text{R}^{17}-(\text{OCH}_2\text{CH}_2)_n(\text{OC}_3\text{H}_6)_m\text{OR}$;

where R, x, m' , n' and (OC_3H_6) are the same as above R^{17} is an alkylene group having from 2 to 6 carbons, and y is from 1 to about 100.

It will be appreciated that structurally the $(\text{C}_3\text{H}_6\text{O})$ and (OC_3H_6) units are the same as are

$(\text{CH}_2(\text{CH}_3)\text{CHO})$ and $(\text{OCH}(\text{CH}_3)\text{CH}_2)$;

$(\text{CH}_2\text{CH}_2\text{CH}_2\text{O})$ and $(\text{OCH}_2\text{CH}_2\text{CH}_2)$; and

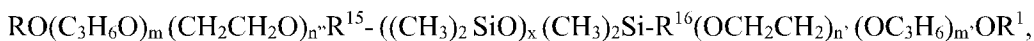
$((\text{CH}_3)\text{CHCH}_2\text{O})$ and $(\text{OCH}_2\text{CH}(\text{CH}_3))$;

In one embodiment the $(\text{C}_3\text{H}_6\text{O})$ and (OC_3H_6) units are respectively $(\text{CH}_2(\text{CH}_3)\text{CHO})$ and

$(\text{OCH}(\text{CH}_3)\text{CH}_2)$ which may alternatively be referred to as propylene oxide (PO) units and the

$(\text{CH}_2\text{CH}_2\text{O})$ and $(\text{OCH}_2\text{CH}_2)$ units may alternatively be referred to as ethylene oxide (EO) units.

In the case of alternative (i)



R and R¹ may be the same or are different and are each selected from is H, or an alkyl group.

In a most preferred embodiment of alternative (i) (C₃H₆O) is (CH₂(CH₃)CHO) and (OC₃H₆) is

5 (OCH(CH₃)CH₂);

In another embodiment of alternative (i) (C₃H₆O) is (CH₂(CH₃)CHO) and (OC₃H₆) is

(OCH₂CH(CH₃)).

In a further embodiment of alternative (i) (C₃H₆O) is (CH₂CH₂CH₂O) and (OC₃H₆) is

(OCH₂CH₂CH₂).

10

When one or both of R and R¹ is an alkyl group, the alkyl group may comprise from 1 to 12 carbons, alternatively from 1 to 10 carbons, alternatively from 1 to 6 carbons, alternatively is methyl or ethyl.

In one embodiment at least one of R and R¹ is H or a methyl group; alternatively, both R and R¹ is H or a methyl group. In one embodiment both R and R¹ are either H or a methyl group.

15 R¹⁵, R¹⁶ and R¹⁷ are each alkylene groups having from 2 to 6 carbons. The alkylene groups may be linear or branched but are preferably linear and are for example -(CH₂)_k- where k is from 2 to 6, alternatively from 2 to 5, alternatively from 2 to 4, alternatively are -(CH₂)₃- groups.

Subscript x is from 2 to 500, alternatively subscript x is from 3 to 450, alternatively subscript x is from 4 to 400, alternatively subscript x is from 5 to 375.

20 Subscripts m and m' may be the same or are different and are each selected from 0 to about 50, alternatively one or both of subscripts m and m' is from 0 to 45, alternatively one or both of subscripts m and m' is from 0 to 40. In one embodiment both subscripts m and m' have the same value of from 0 to 40.

Subscript n'' and n' may be the same or are different and are each selected from 3 to about 50,

25 alternatively each of subscript n'' and n' is from 3 to 40, alternatively each of subscript n'' and n' is from 4 to 35, alternatively each of subscript n'' and n' is from 5 to 30. In one embodiment subscript n'' and n' are the same and are from 5 to 30.

In one embodiment with respect to alternative (i) of silicone polyether copolymer (b) (i.e. (b)(i)) both R and R¹ are hydrogen, each of m and m' from 0 to 40 with preferably m and m' being equal;

30 each of subscript n'' and n' from 5 to 30 with preferably n'' and n' being equal and x is from 5 to 375. In such an embodiment preferably R¹⁵ and R¹⁶ are the same and are -(CH₂)₃-

In the case of alternative (ii) is (CH₃)₃SiO-((CH₃)₂SiO)_x-(CH₃R'SiO)_y-Si(CH₃)₃;

Wherein R' = -R¹⁷-(OCH₂CH₂)_n-(OC₃H₆)_mOR

i.e., where Wherein R' = -R¹⁷-(OCH₂CH₂)_n-(OCH(CH₃)CH₂)_mOR or

35 R' = -R¹⁷-(OCH₂CH₂)_n-(OCH₂CH(CH₃)CH₂)_mOR or

R' = -R¹⁷-(OCH₂CH₂)_n-(OCH₂CH₂CH₂)_mOR.

Preferably in the case of alternative (ii) R' = -R¹⁷-(OCH₂CH₂)_n-(OCH(CH₃)CH₂)_mOR;

where R, R¹⁷, x, m' and n' are the same as above, and

y is from 1 to about 100, alternatively y is from 1 to 75 alternatively y is from 1 to 50, alternatively y is from 1 to 40, alternatively y is from 1 to 30, alternatively y is from 1 to 25. In one embodiment for (b)(ii) m' is from 0 to 45, y is from 1 to 25 and n' is from 4 to 35. In a preferred embodiment of each of the above R¹⁷ is $-(CH_2)_3-$

Additives (c)

The peelable coating composition may additionally comprise one or more additives (c). Additives (c) are all optional and may be selected from aqueous solvent, coalescents, plasticizer, defoamers (otherwise referred to as antifoams or antifoaming agents), rheology modifiers wetting agents, plasticisers and pigments or colouring agents. In one embodiment the peelable coating composition comprises at least one of the additives (c).

Aqueous Solvent

As discussed previously, binder (a) i.e., aqueous styrene-(meth)acrylic emulsion copolymer comprises an aqueous liquid continuous phase. If desired additional aqueous solvent may be introduced into the peelable coating composition during its preparation but this is not usually necessary. When introduced, this may again be solely water, but it may additionally contain small amounts of other solvents, such as alcohols such as methanol, ethanol, isopropanol, butanol and/or hexanol. When present the additional aqueous solvent is added in an amount of up to 10 wt. % of the peelable coating composition, alternatively up to 5.0 wt. % of the peelable coating composition.

Coalescents

Optionally the peelable coating composition herein may comprise one or more coalescents to assist in the forming of a continuous peelable coating on the substrate surface. "Coalescents" herein refer to slow-evaporating solvents that fuse polymer particles into a continuous film under ambient condition. The presence of the coalescent(s) herein helps prevent crack formation on the film surface as the peelable coating forms on the substrate surface. The coalescent(s) may include partially hydrophobic organic solvents which are less volatile than water including glycols, ester alcohols and ethers such as butoxydiglycol, butyl glycol, glycol ethyl ether, diethylene glycol ethyl ether, alkylene glycol ethers such as ethylene glycol monomethyl ether, ethylene glycol monohexyl ether, ethylene glycol monoethyl ether, propylene glycol n-butyl ether, ethylene glycol mono-n-butyl ether, diethylene glycol monomethyl ether, diethylene glycol mono-n-butyl ether, propylene glycol monomethyl ether, triethylene glycol monobutyl ether, dipropylene glycol monomethyl ether, tripropylene glycol monomethyl ether, ethylene glycol monoisobutyl ether, diethylene glycol monoisobutyl ether, propylene glycol monoisobutyl ether, ethylene glycol monophenyl ether, propylene glycol monophenyl ether, propylene glycol n-propyl ether, dipropylene glycol n-propyl ether ethylene glycol monomethyl ether acetate, 2-n-butoxyethanol, n-butyl ether and mixtures of any two or more thereof. Preferred coalescents include dipropylene glycol n-butyl ether, ethylene glycol monobutyl ether, diethylene glycol monobutyl ether, n-butyl ether, or mixtures thereof. When

present, the coalescents may be present in an amount of up to 12 wt. % of the peelable coating composition, alternatively in an amount of up to 10 wt. % of the peelable coating composition, alternatively in an amount of up to 7.5 wt. % of the peelable coating composition, alternatively in an amount of up to 5.0 wt. % of the peelable coating composition.

5 **Plasticisers**

Any suitable plasticisers may be incorporated, for example phthalate esters such as di n-octyl phthalate (DOP), di-isononyl phthalate (DINP), di-2-ethylhexyl phthalate (DEHP) and di isodecyl phthalate (DIDP); citrates such as acetyl tributyl citrate (ATBC); adipates such as dioctyl adipate (DOA), di 2-ethylhexyl adipate (DEHA), di isononyl adipate (DINA); and di-isononyl-1,2-

10 cyclohexanedicarboxylate and mixtures thereof.

Defoamers

The peelable coating composition may comprise one or more defoamers. "Defoamers" herein refers to chemical additives that reduce and hinder the formation of foam. Defoamers may be ethylene oxide/propylene oxide-based defoamers, silicone-based defoamers including silicone polyethers
15 (SPE), polyoxyalkylene-substituted silicones, silicone alkanolamides, silicone esters and silicone glycosides; mineral oil-based defoamers, alkyl polyacrylates, or mixtures thereof.

A silicone polyether defoamer, when present may have a rake type structure wherein the polyoxyethylene or polyoxyethylene-polyoxypropylene copolymeric units are grafted onto the siloxane backbone, or the SPE can have an ABA block copolymeric structure wherein A represents
20 the polyether portion and B the siloxane portion of an ABA structure. Suitable SPE's include DOWSIL™ OFX-5329 Fluid from Dow Silicones Corporation of Midland, Michigan, USA. When an SPE is used as the defoamer herein, typically it has a hydrophilic-lipophilic balance (HLB) of from about 2 to 3. The defoamer may be an emulsion of a polyether siloxane copolymer in combination with fumed silica such as TEGO™ Airex 902 W and TEGO™ Foamex 1488 polyether
25 siloxane copolymer emulsions supplied by Evonik and BYK-024 silicone defoamer available from BYK, and mixtures thereof. For the avoidance of doubt the defoamer is different from component (b) herein.

When present the defoamer can be incorporated into the composition in an amount of up to 2 wt. % of the peelable coating composition, alternatively in an amount of up to 1.5 wt. % of the peelable
30 coating composition alternatively in an amount of up to 1.0 wt. % of the peelable coating composition e.g., in a range of from 0.1 wt.% to 1 wt. % of the peelable coating composition.

Rheology modifiers

The peelable coating composition may comprise one or more rheology modifiers, sometimes referred to as thickeners. These may include one or more clay materials, acid derivatives, naturally
35 occurring polymers having e.g., polysaccharide or amino acid building blocks, such as starch, modified starch, proteins, and modified proteins, dimeric and trimeric fatty acids and/or imidazolines. Alternatively, they may comprise polyvinyl alcohol (PVA), acid copolymers,

urethane associate thickeners (UAT), polyether urea polyurethanes (PEUPU), polyether polyurethanes (PEPU), alkali swellable emulsions (ASE) such as sodium or ammonium neutralized acrylic acid polymers; hydrophobically modified alkali swellable emulsions (HASE) such as hydrophobically modified acrylic acid copolymers; associative thickeners such as hydrophobically modified ethoxylated urethanes (HEUR); and cellulosic thickeners such as methyl cellulose ethers, hydroxymethyl cellulose (HMC), hydroxyethyl cellulose (HEC), hydrophobically-modified hydroxyethyl cellulose (HMHEC), styrene-maleic anhydride terpolymer (SMAT), sodium carboxymethyl cellulose (SCMC), sodium carboxymethyl 2-hydroxyethyl cellulose, 2-hydroxypropyl methyl cellulose, 2-hydroxyethyl methyl cellulose, 2-hydroxybutyl methyl cellulose, 2-hydroxyethyl ethyl cellulose, and 2-hydroxypropyl cellulose.

ASE-rheology modifiers are similar in polymer structure to HASE rheology modifiers but do not contain the hydrophobe groupings, i.e., they are dispersions of insoluble acrylic polymers in water which have a high percentage of acid groups distributed throughout their polymer chains. When the acid groups are neutralized, the salt that is formed is 'hydrated' the salt either swells in aqueous solutions or becomes completely water soluble. As the concentration of neutralized polymer in an aqueous formulation increases, the swollen polymer chains start to overlap, until they 'tangle up'. It is this overlapping and tangling that causes viscosity to increase. Again, the concentration of acid groups, the molecular weight and degree of crosslinking of the polymer are important in determining rheology and thickening efficiency. Examples include ACRY SOL™ ASE-75 from Dow. HASE polymers are commercially important as associative rheology modifier type rheology modifiers in aqueous paints and coatings. They are dispersions of water-insoluble acrylic polymers in water which may be rendered water soluble by neutralizing acid groups on the polymer chain and also contain long-chain hydrophobic groups, sometimes referred to as "hydrophobes". Typically, they are aqueous dispersion of copolymers of

- (i) acrylate ester or methacrylate ester monomers such as methyl methacrylate ethyl acrylate, butyl acrylate, or ethylhexyl acrylate);
- (ii) methacrylic acid, acrylic acid, or itaconic acid; and
- (iii) monomers containing long chain hydrophobic groups such as an ethylenically unsaturated polyethylene oxide (poly EO) macromonomer, e.g., an alkylated ethoxylate monomer, preferably an alkylated ethoxylate acrylate or methacrylate.

The alkylated chains may be in the range of C10 to C25, alternatively C12 to C20.

For example, the following commercially available HASEs from the Dow Chemical Company contain polymerized units of ethyl acrylate and methacrylic acid monomers with hydrophobes attached, ACRY SOL™ DR-6600, ACRY SOL™ DR-5500, ACRY SOL™ RM-7 ACRY SOL™ TT-615, ACRY SOL™ DR-72 and ACRY SOL™ TT-935. Other commercially available HASEs include ACRY SOL™ Primal HT-400, ACULYN™ 88, ACULYN™ 28, ACULYNL™ 88 and Romax™ 7011 from the Dow Chemical Company, and RHEOTECH™ 4800 from Coatex.

Hydrophobe modified ethoxylated urethanes (HEURs) associative rheology modifier type rheology modifiers are widely used in water-borne coatings for their desirable rheological and application properties. The hydrophobically modified alkylene oxide urethane polymer is a polyethylene oxide, polypropylene oxide, or polybutylene oxide urethane polymer, preferably a polyethylene oxide urethane polymer modified with suitable the hydrophobes and may be prepared by e.g., reacting a diisocyanate; a water soluble polyalkylene glycol; and a capping agent comprising the hydrophobe. The hydrophobes are then introduced by end-capping this isocyanate terminated prepolymer with e.g., hydrophobic alcohols or amines. Commercially available HEURs include ACRY SOL™ RM-8W Rheology Modifier and ACRY SOL™ RM-5000 Rheology Modifier both of which are available from the Dow Chemical Company.

Hydroxyethyl cellulose polymers (HEC) are non-ionic, water-soluble polymer that can thicken, suspend, bind, emulsify, form films, stabilize, disperse, retain water, and provide protective colloid action. They are readily soluble in hot or cold water and can be used to prepare solutions with a wide range of viscosities. Examples include Natrosol™ 250 HBR (a water-soluble, non-ionic hydroxyethyl cellulose surface-treated with glyoxal from Ashland Specialty Chemical). Preferably the rheology modifiers chosen when present, are chosen from HECs, HUERS or a mixture thereof.

When present in the peelable coating composition the rheology modifier(s) may be present in an amount of up to 5 wt. % of the peelable coating composition, alternatively of up to 4.0 wt. % of the peelable coating composition alternatively in a range of from 0.01 to 4 wt. % of the peelable coating composition, alternatively from 0.05% to 3%.wt. % of the peelable coating composition.

Wetting Agents

The peelable coating composition as hereinbefore described may further comprise one or more wetting agents. "Wetting agents" herein refer to chemical additives that reduce the surface tension of a coating composition, causing the peelable coating composition to spread across more easily or penetrate the surface of a substrate. Wetting agents may be polycarboxylates, anionic, zwitterionic, or non-ionic.

Anionic wetting agents may include but are not limited to, alkali metal alkyl sulphates e.g., sodium Lauryl sulfate; Fatty Alcohol Ether Sulfates (FAES); Alkyl Phenol Ether Sulfates (APES); carboxylic, phosphoric and sulfonic acids and their salt derivatives; alkyl carboxylates; acyl lactylates; alkyl ether carboxylates; n-acyl sarcosinate; n-acyl glutamates; fatty acid-polypeptide condensates; alkali metal sulfosuccinates; sulfonated glycerol esters of fatty acids, such as sulfonated monoglycerides of coconut oil acids; salts of sulfonated monovalent alcohol esters, such as sodium oleylisethionate; amides of amino sulfonic acids, such as the sodium salt of oleyl methyl tauride; sulfonated products of fatty acids nitriles, such as palmitonitrile sulfonate; sulfonated aromatic hydrocarbons, such as sodium alpha-naphthalene monosulfonate; condensation products of naphthalene sulfonic acids with formaldehyde; sodium octahydroanthracene sulfonate; ether sulphates having alkyl groups of 8 or more carbon atoms; alkylarylsulfonates having 1 or more alkyl

groups of 8 or more carbon atoms. Sodium dodecyl benzene sulfonate, dioctylsulfosuccinate, sodium polyoxyethylene lauryl ether sulfate, diphenyl sulfonate derivatives, e.g., sodium dodecyl diphenyl oxide disulfonate, sodium salt of tert-octylphenoxyethoxypoly(39)ethoxyethyl sulfate.

Anionic wetting agents which are commercially available and useful herein may include but are not limited to, for the sake of example, POLYSTEP™ A4, A7, A11, A15, A15-30K, A16, A16-22, A18, A13, A17, B1, B3, B5, B11, B12, B19, B20, B22, B23, B24, B25, B27, B29, C-OP3S; ALPHA-STEP™ ML40, MC48; STEPANOL™ MG; all produced by STEPAN CO., Chicago, IL; HOSTAPUR™ SAS produced by HOECHST CELANESE; HAMPOSYL™ C30 and L30 produced by W.R.GRACE & CO., Lexington, MA. Silicone polyether wetting agents may include

DOWSIL™ OFX-5329 Fluid from Dow Silicones Corporation of Midland, Michigan, USA or BYK-346 commercially available from Byk-Chemie GmbH. Non-ionic wetting agents include polyethoxylates, such as ethoxylated alkyl polyethylene glycol ethers; polyoxyalkylene alkyl ethers; polyoxyalkylene sorbitan esters; polyoxyalkylene esters; polyoxyalkylene alkylphenyl ethers, ethoxylated amides; ethoxylated alcohols; ethoxylated esters; polysorbate esters; polyoxypropylene compounds, such as propoxylated alcohols; ethoxylated/propoxylated block polymers and propoxylated esters; alkanolamides; amine oxides; fatty acid esters of polyhydric alcohols, such as ethylene glycol esters, diethylene glycol esters, propylene glycol esters, glyceryl esters, polyglyceryl fatty acid esters, sorbitan esters, sucrose esters and glucose esters. Commercial non-ionic wetting agents include, for the sake of example, TERGITOL™ TMN-6, TERGITOL™ 15S40, TERGITOL™ 15S9, TERGITOL™ 15S12, TERGITOL™ 15S15 and TERGITOL™ 15S20, and TRITON™ X405 produced by The Dow Chemical Company of Midland, Michigan; BRIJ™ 30 and BRIJ™ 35 produced by Croda (UK); MAKON™ 10 produced by STEPAN COMPANY, (Chicago, IL); and ETHOMID™ O/17 produced by Akzo Nobel Surfactants (Chicago, IL).

The wetting agent may alternatively or additionally comprise a silicone polyether (SPE). The silicone polyether as a wetting agent may have a rake type structure wherein the polyoxyethylene or polyoxyethylene-polyoxypropylene copolymeric units are grafted onto the siloxane backbone, or the SPE can have an ABA block copolymeric structure wherein A represents the polyether portion and B the siloxane portion of an ABA structure. Alternatively, the wetting agent may be selected from polyoxyalkylene-substituted silicones, silicone alkanolamides, silicone esters and silicone glycosides. When an SPE is used as the wetting agent herein, typically it has a hydrophilic-lipophilic balance (HLB) of from about 7 to 9 determined by the method described above.

For the avoidance of doubt the SPEs utilised as wetting agents are different from component (b) herein. A commercial example of an SPE wetting agent is BYK-346 commercially available from Byk-Chemie GmbH.

The wetting agent, when present, may be present based on the total weight of the peelable coating composition, in an amount of up to 5 wt. %, alternatively from 0.01 wt. % to 4 wt. % of the peelable coating composition, or alternatively of from 0.1 wt. % to 3 wt. % of the peelable coating

composition.

Pigments and colouring Agents

Examples of pigments include titanium dioxide, chromium oxide, bismuth vanadium oxide, iron oxides and mixtures thereof.

- 5 Examples of colouring agents for which may be utilized herein include pigments, vat dyes, reactive dyes, acid dyes, chrome dyes, disperse dyes, cationic dyes and mixtures thereof. The pigments and/or coloring agents may be coloured, white, black, metal effect, and luminescent e.g., fluorescent and phosphorescent. Pigments are utilized to colour the composition as required. Any suitable pigment may be utilized providing it is compatible with the composition herein. In two-part
- 10 moisture cure organopolysiloxane compositions pigments and/or coloured (non-white) fillers e.g., carbon black may be utilized in the catalyst package to colour the end sealant product. Suitable white pigments and/or coloring agents include titanium dioxide, zinc oxide, lead oxide, zinc sulfide, lithophone, zirconium oxide, and antimony oxide. Suitable non-white inorganic pigments and/or coloring agents include, but are not limited to, iron
- 15 oxide pigments such as goethite, lepidocrocite, hematite, maghemite, and magnetite black iron oxide, yellow iron oxide, brown iron oxide, and red iron oxide; blue iron pigments; chromium oxide pigments; cadmium pigments such as cadmium yellow, cadmium red, and cadmium cinnabar; bismuth pigments such as bismuth vanadate and bismuth vanadate molybdate; mixed metal oxide pigments such as cobalt titanate green; chromate and molybdate pigments such as chromium yellow,
- 20 molybdate red, and molybdate orange; ultramarine pigments; cobalt oxide pigments; nickel antimony titanates; lead chrome; carbon black; lampblack, and metal effect pigments such as aluminium, copper, copper oxide, bronze, stainless steel, nickel, zinc, and brass. Suitable organic non-white pigments and/or coloring agents include phthalocyanine pigments, e.g., phthalocyanine blue and phthalocyanine green; monoarylide yellow, diarylide yellow,
- 25 benzimidazolone yellow, heterocyclic yellow, DAN orange, quinacridone pigments, e.g., quinacridone magenta and quinacridone violet; organic reds, including metallized azo reds and nonmetallized azo reds and other azo pigments, monoazo pigments, diazo pigments, azo pigment lakes, β -naphthol pigments, naphthol AS pigments, benzimidazolone pigments, diazo condensation pigment, isoindolinone, and isoindoline pigments, polycyclic pigments, perylene and perinone
- 30 pigments, thioindigo pigments, anthrapyrimidone pigments, flavanthrone pigments, anthanthrone pigments, dioxazine pigments, triarylcarbonium pigments, quinophthalone pigments, and diketopyrrolo pyrrole pigments.

- In addition to the components described above, the peelable coating composition may further comprise any one or combination of the following additives: buffers, neutralizers, humectants,
- 35 mildewcides, biocides, anti-skinning agents, antioxidants, leveling agents, adhesion promoters and/or anti-flash rust additives, if desired or required. These may be introduced into the composition as part of the binder (a) or maybe mixed into the composition at a suitable stage in

mixing.

The peelable coating composition may be prepared by a process comprising: admixing the aqueous styrene-(meth)acrylic emulsion copolymer with other optional components, e.g., pigments and/or plasticisers as described above. Components in the peelable coating composition may be mixed in any order to provide the peelable coating composition. Any of the above-mentioned optional components may also be added to the composition during or prior to the mixing to form the peelable coating composition. When the peelable coating composition comprises pigment and/or plasticiser, the pigments and/or plasticisers are preferably mixed with the dispersant to form a slurry of pigments and/or plasticiser.

Cumulatively the total amount of additives (c) present in the peelable coating composition may be from 0 (zero) to 19.25 wt. % of the peelable coating composition, alternatively from 0 (zero) to 15.0 wt. % of the peelable coating composition.

The total wt. % of the peelable coating composition described herein is 100wt. % and the total wt. % of additional additives © present is the difference between 100 wt. % and the cumulative wt. % of components (a) and (b) of the peelable coating composition.

Hence, there is provided herein a peelable coating composition comprising

(a) an aqueous styrene-(meth)acrylic emulsion copolymer binder as herein described in an amount of from 70 wt. % to 99.25 wt. % of the peelable coating composition; and

(b) a silicone polyether copolymer in an amount of from 0.75 to 10 wt. % of the peelable coating composition selected from one or both of

(i) $\text{RO}(\text{C}_3\text{H}_6\text{O})_m(\text{CH}_2\text{CH}_2\text{O})_n\text{R}^{15}-((\text{CH}_3)_2\text{SiO})_x(\text{CH}_3)_2\text{Si}-\text{R}^{16}(\text{OCH}_2\text{CH}_2)_n(\text{OC}_3\text{H}_6)_m\text{OR}^1$, where:-

R and R¹ may be the same or are different and are each selected from H, or an alkyl group,

(C₃H₆O) is (CH₂(CH₃)CHO), (CH₂CH₂CH₂O), ((CH₃)CHCH₂O) or mixtures thereof;

(OC₃H₆) is (OCH₂CH(CH₃)), (OCH₂CH₂CH₂), (OCH(CH₃)CH₂) or mixtures thereof;

R¹⁵ and R¹⁶ are the same or different and are alkylene groups having from 2 to 6 carbons, preferably -(CH₂)₃-; x is from 2 to about 500, m and m' may be the same or are different and are each in a range of from 0 to about 50 and n' and n' may be the same or are different and are in a range of from 3 to about 50;

or

(ii) $(\text{CH}_3)_3\text{SiO}-((\text{CH}_3)_2\text{SiO})_x-(\text{CH}_3\text{R}'\text{SiO})_y-\text{Si}(\text{CH}_3)_3$;

Wherein R' = -R¹⁷-(OCH₂CH₂)_n-(OC₃H₆)_mOR;

where R, x, m', n' and (OC₃H₆) are the same as above R¹⁷ is an alkylene group having from 2 to 6 carbons, and y is from 1 to about 100. Preferably in the case of alternative (ii) R' = -

(CH₂)₃(OCH₂CH₂)_n(OCH(CH₃)CH₂)_mOR;

and optionally

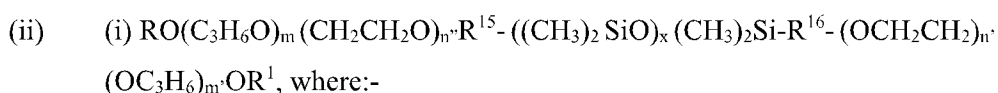
one or more additives (c) selected from aqueous solvent, coalescents, defoamers, rheology modifiers, wetting agents, plasticisers, pigments and/or coloring agents and/or a mixture thereof, in an amount of from 0 (zero) to 19.25 wt. % of the peelable coating composition.

The peelable coating formed after the application of the peelable coating composition, is provided to temporarily protect a substrate surface for a predetermined period of time and then can be removed by being peeled off the substrate surface.

The method for forming the peelable coating on a substrate comprises the steps of:

(I') combining the aqueous styrene-(meth)acrylic emulsion copolymer binder (a) as hereinbefore described; with

a silicone polyether copolymer (b) in an amount of from 0.75 to 10 wt. % of the peelable coating composition selected from one or bot



R and R¹ may be the same or are different and are each selected from H, or an alkyl group,

(C₃H₆O) is (CH₂(CH₃)CHO), (CH₂CH₂CH₂O), ((CH₃)CHCH₂O) or mixtures thereof;

(OC₃H₆) is (OCH₂CH(CH₃)), (OCH₂CH₂CH₂), (OCH(CH₃)CH₂) or mixtures thereof;

R¹⁵ and R¹⁶ are the same or different and are alkylene groups having from 2 to 6 carbons,

x is from 2 to about 500, m and m' may be the same or are different and are each in a range of from 0 to about 50 and n'' and n' may be the same or are different and are in a range of from 3 to about

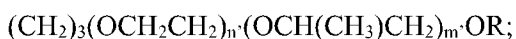
50;

or



Wherein R' = -R¹⁷-(OCH₂CH₂)_{n'}(OC₃H₆)_{m'}OR;

where R, x, m', n' and (OC₃H₆) are the same as above R¹⁷ is an alkylene group having from 2 to 6 carbons, and y is from 1 to about 100. Preferably in the case of alternative (ii) R' = -



and optionally

one or more additives (c) selected from aqueous solvent, coalescents, defoamers, rheology modifiers, wetting agents, plasticisers, pigments and coloring agents and/or a mixture thereof, to form a peelable coating composition,

(II') applying the peelable coating composition on a substrate surface;

(III') drying the peelable coating composition applied in step (II') to form a peelable coating on the substrate.

The peelable coating composition may be prepared in step (I) of the above in any suitable manner.

For example, component (a) may be introduced into a suitable mixer and is then stirred as any optional additives are introduced into the mixture. Subsequent to the addition of any optional additives, the desired amount of the silicone polyether copolymer (b) may be added and is then

thoroughly mixed with the other component(s) to ensure the silicone polyether copolymer (b) is consistently dispersed throughout the peelable coating composition.

In one embodiment component (a) is stirred in the mixer until all the optional additives are added and then the silicone polyether copolymer (b) was added and was mixed in at 700rpm for a period of up to 10 minutes. Any type of mixing equipment may be used such as Terrell™, Neulander™ or Ross™ mixers or a FlackTek SpeedMixer™ from FlackTek of Landrum, South Carolina, USA.

Once prepared, in step (II') of the process said peelable coating composition is applied onto a substrate surface by any suitable method, for example it may be spray-applied, brushed, rolled, dipped or otherwise coated onto a substrate although spraying techniques are preferred. Typically,

the peelable coating composition is applied as a single layer but, if desired, the peelable coating composition coating can be applied in multiple layers.

Typically, the target wet coating thickness of the peelable coating composition on the substrate is from 100µm to 750µm, alternatively a wet coating thickness of from 200 to 700µm, alternatively a wet coating thickness from 250 µm to 600 µm. The wet thickness was determined using a coating bar.

In step (III') of the process the peelable coating composition is then left to dry/cure to a peelable coating on the substrate surface. The resulting peelable coating is visually clear (see-through) unless a suitable pigment or coloring agent additive has been incorporated as an additive (c). This can be generally completed at room temperature and standard conditions, e.g., atmospheric pressure at 50 % relative humidity. Step (III') can be allowed to take place for anything from 2 to 12 hours, alternatively between 2 and 6 hours.

Substrates to which the peelable coating composition is intended to be applied include materials which are used in construction or are used to protect substrate surfaces during transportation, storage or construction and may include metal, plastic, glass, cloth, ceramic, clay, fiber, concrete, brick, rock or wood or painted walls and painted metals. Surprisingly, however, the current compositions have proved to be excellent means of providing peelable coatings for two particularly problematic situations for peelable coatings of this type:

- (1) they function well at low temperatures, e.g., below 10°C moreover, below 5°C so they can be used in situations where construction materials require peelable coating protection in weather situations where temperatures dip below say 5°C in storage or in cold weather or overnight; and
- (2) they have been found to be surprisingly adhesive to polyvinylidene fluoride or polyvinylidene difluoride (PVDF) surfaces to which such peelable coatings are usually difficult to adhere.

Polyvinylidene fluoride or polyvinylidene difluoride (PVDF) made of repeating units of $-\text{[C}_2\text{H}_2\text{F}_2\text{]}-$ is a thermoplastic material widely used in architecture and construction applications such as cladding, coatings and paints for exterior surfaces such as metals, e.g., aluminium because of, for

example, their outstanding colour retention, fade resistance, film integrity, and protection against chalking etc., Whilst with the use of suitable primers they can be adhered to the substrate surfaces, the resulting cladding, coatings and paints are renowned for their unreactiveness and as such are difficult substrates to which to adhere protective peelable coatings thereto. Unexpectedly the coating compositions defined herein are not only excellent for use at low temperatures but are also suitable for use with Polyvinylidene fluoride or polyvinylidene difluoride (PVDF) substrate surfaces.

The peelable coatings formed, after the application of the peelable coating composition, are provided as a temporary means of protection of the materials before use and/or temporarily after use to protect the surface until for example the substrate is ready for use, e.g., after the completion of the construction of a building or once transportation has been completed.

The peelable coatings resulting from the peelable coating composition proved to be a temporarily protective coating which is removeable by peeling from substrates to which they were applied without the presence of an intervening layer. The peelable coatings which are temporarily applied to substrate surfaces and are removeable by peeling were peeled away from the substrate surface by hand as a complete film proving that they had a suitable tensile strength given no or minimal fracturing or tearing occurred during the peeling operation even after simulated weathering. Subsequent to the application of the peelable coating on to the substrate the method additionally comprises the step of removing the peelable coating by peeling it off the substrate surface onto which it has been applied.

Hence, there may alternatively be provided a method of temporarily protecting a substrate by preparing and applying a peelable coating composition as hereinbefore described onto a substrate surface; forming a peelable coating on said substrate, transporting, storing and/or using said substrate in construction; and subsequently removing said peelable coating by peeling same from the substrate surface. In such a process the peelable coating is a temporary removeable coating which is removed by peeling the coating from the substrate surface as and when desired.

While not being tested quantitatively, the fact that the peelable coatings could be removably adhered to the substrates means they also retained a sufficiently low peel strength to enable the act of peeling to be carried out manually by hand. It is the general consensus of the industry that for peelable coatings to be peeled manually the peel strength must be at the very most no more than about 400 N/m, but preferably no more than about 200 N/m and preferably less than about 100 N/m so that the peel strength of the coating to the surface is sufficiently low, and the act of peeling the coating can be carried out by hand manually. Surprisingly it was found that when using silicone polyether copolymers other than component (b) herein, much poorer results were observed such that in many instances the resulting coatings could not be peeled manually or were seen to crack or tear, especially after simulated weathering. These results were determined using the naked eye.

Hence the peelable coatings resulting from application of the peelable coating compositions described herein can be considered to be weather resistant and/or rain resistant and/or waterproof enabling them to be used in both exterior and interior situations. In the case of exterior situations, where the peelable coating composition is applied on an exterior glass, the resulting peelable coating herein may protect exterior facing glass substrates because it has good water resistance, it is not damaged or washed away after exposure to rain even long-term exposure to rain and the peelable coating can then dry but not get damaged, even in the case of direct exposure to the sun, as it remains peelable. Indeed, it would appear to be water resistant because even after harsh testing rain/water fails to affect the peelable performance of the peelable coating.

The peelable coating herein is provided as a protective coating derived from a visually clear (see-through) peelable coating composition which may be applied to a wide range of substrates such as metal, plastic, glass, cloth, ceramic, clay, fiber, concrete, brick, rock or wood. Substrate surfaces may also include painted walls and painted and/or coated metals such as polyvinyl difluoride (PVDF) coated surfaces e.g., PVDF coated metal surfaces such as aluminium surfaces.

For example, peelable coating compositions as described herein may be utilised to provide peelable coatings to protect paint, glass, plastic, or metal portions of a vehicle such as an automobile, aeroplane, boat, snowmobile or motorcycle during storage and transportation. Other applications may include temporarily protective coatings for bathroom fixtures, plumbing fixtures, kitchen fittings, white goods such as refrigerators, microwave ovens, plated or chromed parts, instrument panels or the like. The peelable coating may be peeled off the substrate as and when required together with any dirt and contaminants which have impacted the peelable coating during its presence of the substrate surface.

It is particularly designed to provide a peelable coating on interior and exterior glass to provide the glass substrate with a water-resistant protective coating during transportation or construction or the like which can be removed manually as a single film without cracks and/or tears. The glass substrate onto which the peelable coating composition is applied may be virtually any glass substrate for example, borosilicate glass, soda lime glass, silica glass, alkali barium glass, aluminosilicate glass, lead glass, phosphate glass, alkali borosilicate glass, xena glass fluorosilicate glass or a pre-treated glass, for example, vacuum-deposited reflective metallic-coated plate glass which may be used in e.g., commercial building and architectural spandrel applications. It can also protect glass substrates for use in a wide variety of indoor and exterior applications, e.g., in or for optical glass, architectural glass, façade glass, glass for shadow boxes, decorative glass, technical glass, construction glass such as structural glass, float glass, shatterproof glass, laminated glass, extra clean glass, chromatic glass, tinted glass, toughened glass, glass bricks, frosted glass and/or bulletproof glass, elevator glass.glass products such as windows and the like as well as fixtures and fittings for buildings such as bathroom and kitchen fixtures e.g., chrome plated or brass surfaces

must be protected not only during shipping, but also during installation or assembly. to avoid scratching and marring before or during construction taking place.

Examples

Multiple coating compositions were prepared with a view to comparing their peelability after application on a suitable substrate, mainly concentrating on the effects caused by the variation in the component (a) aqueous styrene-(meth)acrylic emulsion binder, copolymers as described herein. Several combinations of aqueous styrene-(meth)acrylic emulsion copolymers were tested for their suitability as binders herein. The binders assessed are described below.

Comparative binder 1 (CB. 1) was a vinyl acrylic emulsion commercially available under the trade name ROVACE™ 662 from the Dow Chemical Company made using vinyl acetate monomer excluded from the compositions described herein.

Comparative binder 2 (CB. 2) was an acrylate copolymer emulsion prepared, as described below, using the following monomers 2 wt. % acrylic acid, 57 wt. % methyl methacrylate, 37 wt. % butyl acrylate, 4 wt. % hydroxyethyl methacrylate and 0 wt. % styrene (based on 100 wt. % on monomer content).

Comparative Binder 3 (CB. 3) was an acrylate copolymer emulsion prepared, as described below, using the following monomers 2 wt. % acrylic acid, 52 wt. % methyl methacrylate, 46 wt. % butyl acrylate and 0 wt. % styrene (based on 100 wt. % on monomer content).

Comparative Binder 4 (CB. 4) was an aqueous styrene-(meth)acrylic emulsion copolymer outside the scope of the binders herein, prepared, using a similar emulsion polymerisation process to those described below for CB. 2 and 3 with the following monomers 38.5 wt. % styrene, 2.8 wt. % methacrylic acid, 32 wt. % ethylhexyl acrylate, 23.3 wt. % cyclohexyl methacrylate and 2.8 wt. % acetoacetoxyethyl methacrylate (based on 100 wt. % on monomer content). It was found to have a T_g of 38°C using the test method described above.

Comparative Binder 5 (CB. 5) was an aqueous styrene-(meth)acrylic emulsion copolymer outside the scope of the binders herein, prepared, in an analogous process to that of CB. 4 using a similar emulsion polymerisation process to those described below for CB. 2 and 3 with the following monomers 58.1 wt. % styrene, 2.2 wt.% acrylic acid and 39.7 wt.% butyl acrylate (based on 100 wt. % on monomer content). It was found to have a T_g of 37°C using the test method described above.

Binder 1 was an aqueous styrene-(meth)acrylic emulsion copolymer as described herein, using a similar emulsion polymerisation process to those described below for CB. 2 and 3 using the following monomers, 34.7 wt. % styrene, 3.3 wt. % of methacrylic acid and 42 wt. % butyl acrylate and 20 wt. % methyl methacrylate (based on 100 wt. % on monomer content). It was found to have a T_g of 33°C using the test method described above.

Comparative Binder 6 (CB. 6) was an aqueous styrene-(meth)acrylic emulsion copolymer outside the scope of the binders herein, prepared, using a similar emulsion polymerisation process to those

described below for CB. 2 and 3 using the following monomers 14.3 wt. % styrene, 1.1 wt. % acrylic acid, 77.5 wt. % butyl acrylate and 7 wt. % acrylonitrile (based on 100 wt. % on monomer content). It was found to have a T_g of -12°C using the test method described above.

Binder Preparations

- 5 The binders utilised in the present application can be prepared in several ways via emulsion polymerisation. In the following preparations DISPONILTM FES-32 was used as the surfactant. It is a commercially available surfactant available from BASF. However other suitable surfactants may be used.

10 Preparation of Comparative 2 acrylate copolymer emulsion

A monomer emulsion (ME1) was first prepared by mixing the following together to produce a stable monomer emulsion:

Deionized water (370.0g), a surfactant DISPONILTM FES-32 (29.8g, 30.5% active), butyl acrylate (563.4g), acrylic acid (30.7g), hydroxyethyl methacrylate (60.9g) and methyl methacrylate (865.4g).

- 15 Deionized water (900.0g) was introduced into a glass reactor and then the glass reactor was heated to 88°C , after which was added surfactant DISPONILTM FES-32 (9.9g), a 6.5% Na_2CO_3 solution (46.8g), monomer emulsion ME1 (60.0g) and 13.0% ammonium persulfate solution (47.2g). After a further 5 minutes the remainder of the monomer emulsion ME1 was gradually introduced into the glass reactor over about 70 minutes, with a view to maintaining the reactor temperature at $87-89^{\circ}\text{C}$.
- 20 The resulting mixture was continuously stirred for a further 10 minutes after the introduction of the monomer emulsion was complete. Subsequently, the reactor was cooled to 70°C , $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ (0.01g) (a promoter for monomer polymerization), was mixed with ethylenediaminetetraacetic acid (EDTA) (0.02g) (chelator) in water (12.0g) and added into the reactor. A solution of tertiary butyl hydroperoxide (t-BHP) (0.9g dissolved in 18.0g of distilled water) and a solution of
- 25 isoascorbic acid (IAA) (0.7g) which functions as a reductant in deionized water (18.9g) were fed into the reactor over 30 minutes. After this 14.0g ammonia solution (26%) was added to adjust pH and the resulting acrylate copolymer emulsion polymer emulsion was cooled to room temperature.

Preparation of Comparative 3 acrylate copolymer emulsion

- 30 A monomer emulsion (ME2) was first prepared by mixing the following together to produce a stable monomer emulsion:

Deionized water (370.0g), a surfactant DISPONILTM FES-32 (29.8g, 30.5% active), butyl acrylate (700.5g), acrylic acid (30.7g), and methyl methacrylate (789.5g).

Deionized water (860.0g) was introduced into a glass reactor and then the glass reactor was heated to 88°C , after which was added surfactant DISPONILTM FES-32 (9.9g), a 6.5% Na_2CO_3 solution

(46.8g), monomer emulsion ME2 (60.0g) and 13.0% ammonium persulfate solution (47.2g). After a further 5 minutes the remainder of the monomer emulsion ME2 was gradually introduced into the glass reactor over about 70 minutes, with a view to maintaining the reactor temperature at 87-89°C. The resulting mixture was continuously stirred for a further 10 minutes after the introduction of the monomer emulsion was complete. Subsequently, the reactor was cooled to 70°C, $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ (0.01g) was mixed with ethylenediaminetetraacetic acid (EDTA) (0.02g) in water (12.0g) and added into the reactor. A solution of tertiary butyl hydroperoxide (t-BHP) (0.9g dissolved in 25.0g of deionized water) and a solution of isoascorbic acid (0.7g) in deionized water (26.0g) were fed into the reactor over 30 minutes. After this 14.0g ammonia solution (26%) was added to adjust pH and the resulting acrylate copolymer emulsion polymer emulsion was cooled to room temperature.

A Preparation for Comparative 4 acrylate copolymer emulsion

A monomer emulsion (ME3) is first prepared by mixing the following together to produce a stable monomer emulsion:

Deionized water (370.0g), a surfactant DISPONILTM FES-32 (49.7g, 30.5% active), ethylhexyl acrylate (499.5g), methacrylic acid (42.9g), acetoacetoxyethyl methacrylate (39.8g), cyclohexyl methacrylate (353.8g) and styrene (584.6g).

Deionized water (860.0g) is introduced into a glass reactor and then the glass reactor is heated to 88°C, after which is added surfactant DISPONILTM FES-32 (11.9g), a 6.5% Na_2CO_3 solution (46.8g), monomer emulsion ME3 (60.0g) and 13.0% ammonium persulfate solution (47.2g). After a further 5 minutes the remainder of the monomer emulsion ME3 is gradually introduced into the glass reactor over about 120 minutes, with a view to maintaining the reactor temperature at 87-89°C. The resulting mixture is continuously stirred for a further 10 minutes after the introduction of the monomer emulsion is complete. Subsequently, the reactor is cooled to 70°C, $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ (0.01g) is mixed with ethylenediaminetetraacetic acid (EDTA) (0.02g) in water (12.0g) and added into the reactor. A solution of tertiary butyl hydroperoxide (t-BHP) (1.4g dissolved in 25.0g of deionized water) and a solution of isoascorbic acid (1.0g) in deionized water (26.0g) are fed into the reactor over 50 minutes. After this 18.0g ammonia solution (26%) is added to adjust pH and the resulting acrylate copolymer emulsion polymer emulsion is cooled to room temperature.

A method for making Binder 1

A monomer emulsion (ME4) is first prepared by mixing the following together to produce a stable monomer emulsion: water (370.0g), surfactant DISPONILTM FES-32 (49.7g, 30.5% active), butyl acrylate (639.5g), methacrylic acid (50.6g), methyl methacrylate (303.7g) and styrene (526.9g).

Deionized water (860.0g) is introduced into a glass reactor and then the glass reactor is heated to 88°C, after which is added surfactant DISPONILTM FES-32 (11.9g), a 6.5% Na₂CO₃ solution (46.8g), monomer emulsion ME4 (60.0g) and 13.0% ammonium persulfate solution (47.2g). After a further 5 minutes the remainder of the monomer emulsion ME4 is gradually introduced into the glass reactor over about 120 minutes, with a view to maintaining the reactor temperature at 87-89°C. The resulting mixture is continuously stirred for a further 10 minutes after the introduction of the monomer emulsion is complete. Subsequently, the reactor is cooled to 70°C, FeSO₄·7H₂O (0.01g) is mixed with ethylenediaminetetraacetic acid (EDTA) (0.02g) in water (12.0g) and added into the reactor. A solution of tertiary butyl hydroperoxide (t-BHP) (1.4g dissolved in 25.0g of deionized water) and a solution of isoascorbic acid (1.0g) in deionized water (26.0g) are fed into the reactor over 50 minutes. After this 18.9g ammonia solution (26%) is added to adjust pH and the resulting acrylate copolymer emulsion polymer emulsion is cooled to room temperature.

A Preparation for Comparative 5 acrylate copolymer emulsion

A monomer emulsion (ME5) is first prepared by mixing the following together to produce a stable monomer emulsion: water (33.7g), surfactant DISPONILTM FES-32 (49.7g, 30.5% active), butyl acrylate (604.5g), acrylic acid (33.7g), and styrene (882.2g).

Deionized water (860.0g) is introduced into a glass reactor and then the glass reactor is heated to 88°C, after which is added surfactant DISPONILTM FES-32 (11.9g), a 6.5% Na₂CO₃ solution (46.8g), monomer emulsion ME5 (60.0g) and 13.0% ammonium persulfate solution (47.2g). After a further 5 minutes the remainder of the monomer emulsion ME5 is gradually introduced into the glass reactor over about 120 minutes, with a view to maintaining the reactor temperature at 87-89°C. The resulting mixture is continuously stirred for a further 10 minutes after the introduction of the monomer emulsion is complete. Subsequently, the reactor is cooled to 70°C, FeSO₄·7H₂O (0.01g) is mixed with ethylenediaminetetraacetic acid (EDTA) (0.02g) in water (12.0g) and added into the reactor. A solution of tertiary butyl hydroperoxide (t-BHP) (1.4g dissolved in 25.0g of deionized water) and a solution of isoascorbic acid (1.0g) in deionized water (26.0g) are fed into the reactor over 50 minutes. After this 15g ammonia solution (26%) is added to adjust pH and the resulting acrylate copolymer emulsion polymer emulsion is cooled to room temperature.

A Preparation for Comparative 6 acrylate copolymer emulsion

A monomer emulsion (ME6) is first prepared by mixing the following together to produce a stable monomer emulsion: water (33.7g), surfactant DISPONILTM FES-32 (29.8g, 30.5% active), butyl acrylate (1181.5g), acrylic acid (16.9g), % acrylonitrile (106.6g) and styrene (217.2g).

Deionized water (820.0g) is introduced into a glass reactor and then the glass reactor is heated to 85°C, after which is added surfactant DISPONIL™ FES-32 (1.1g), a 6.5% Na₂CO₃ solution (46.8g), monomer emulsion ME6 (60.0g) and 13.0% ammonium persulfate solution (47.2g). After a further 5 minutes the remainder of the monomer emulsion ME6 is gradually introduced into the glass reactor over about 180 minutes, with a view to maintaining the reactor temperature at 82-84°C. The resulting mixture is continuously stirred for a further 10 minutes after the introduction of the monomer emulsion is complete. Subsequently, the reactor is cooled to 70°C, FeSO₄·7H₂O (0.01g) is mixed with ethylenediaminetetraacetic acid (EDTA) (0.02g) in water (12.0g) and added into the reactor. A solution of tertiary butyl hydroperoxide (t-BHP) (2.1g dissolved in 30.0g of deionized water) and a solution of isoascorbic acid (1.5g) in deionized water (26.0g) are fed into the reactor over 60 minutes. After this 7.5g ammonia solution (26%) is added to adjust pH and the resulting acrylate copolymer emulsion polymer emulsion is cooled to room temperature.

The binders used in the following Table were respectively mixed with a standard set of ingredients as the basis for the coating compositions. The different compositions tested are depicted in Table 1 below:

Table 1. Compositions of Example 1 (Ex. 1) and Comparatives 1 to 6 (C. 1 to C. 6).

	C. 1	C. 2	C. 3	C. 4	Ex. 1	C. 5	C. 6
CB. 1	92						
CB. 2		92					
CB. 3			92				
CB. 4				92			
binder 1					92		
CB. 5						92	
CB. 6							92
SPE	1	1	1	1	1	1	1
Coalescent	4	4	4	4	4	4	4
Wetting agent	0.3	0.3	0.3	0.3	0.3	0.3	0.30
Defoamer	0.3	0.3	0.3	0.3	0.3	0.3	0.3
Rheology modifier	0.5	0.5	0.5	0.5	0.5	0.5	0.5
Water	1.9	1.9	1.9	1.9	1.9	1.9	1.9

In Table 1:

20 The silicone polyether copolymer (SPE) used was:

$R^2Me_2SiO(Me_2SiO)_{14}Me_2SiR^2$ where $R^2 = -(CH_2)_3(EO)_{12}OH$

The coalescent used in the examples was Texanol™ Ester Alcohol commercially available from the Eastman Chemical Company.

The defoamer utilised was TEGO™ Airex 902 W commercially available from Evonik Operations GmbH.

The wetting agent used was BYK-346 commercially available from Byk-Chemie GmbH

Rheology Modifier was ACRY SOL™ RM-8W Rheology Modifier commercially available from

5 The Dow Chemical Company.

The different compositions were prepared by introducing all ingredients other than the SPE into the binder and stirring for a few minutes in a FlackTek SpeedMixer™ from FlackTek of Landrum, South Carolina, USA at 700rpm. Subsequently the selected SPE was added and mixing continued for a further 10 minutes again at 700 rpm to ensure the SPE was dispersed uniformly in the

10 composition.

Each resulting composition was then applied onto a glass, ceramic or polyvinylidene fluoride (PVDF) panel (unless otherwise indicated). The wet coating thickness of each applied liquid coating was an average of about 300µm measured using a Myers coating bar. The resulting coating was then allowed to cure for 4 hours at room temperature and 50% relative humidity.

15 After drying/curing, the peelability of each coating relative to the substrate on which it was applied was assessed:

1) At room temperature

peelability was tested shortly after completion of the 4 hours drying/curing process;

2) At 5°C

20 the coated sample underwent drying/curing in a 5°C temperature fridge for 24 hours in order to mimic winter temperatures, after which peelability was assessed;

In both cases the peelability of a sample peelable coatings was graded from 1 (worst) to 5 (best) by visually inspecting the peelable coatings for defects in accordance with the following definitions:

25 **Grade 5:** Whole peelable coating is easily peeled off, no residual peelable coating left on the substrate with no splitting cracking during peeling and no damage to substrate.

Grade 4: Almost completely peels off, but slightly more force required. Minimal (5% or less) residue left on the substrate. The peelable coating may have a slight split or crack.

30 **Grade 3:** Greater force needed to peel peelable coating from substrate. The peelable coating is more easily torn than (4 or 5 above) and more than 30% residual peelable coating may be left on the substrate.

Grade 2: Can be peeled off, but the peelable coating is easily torn. Greater than 50% of the peelable coating may be left on the substrate and the substrate can be damaged.

Grade 1: Hardly peels off, too good adhesion necessitating solvent or tools to be removed.

35 Table 2 provides an indication of the target grade for each test undertaken. For the avoidance of doubt the symbol $\geq$ means equal to or greater than.

Table 2. Acceptable coating performance in clear coat formulation:

Performance	Target
Peelability at room temperature (RT) on glass	≥ 4
Peelability at RT on ceramic	≥ 4
Peelability at RT on polyvinylidene fluoride (PVDF)	≥ 4
Peelability at 5°C on glass	≥ 4
Peelability at 5°C on PVDF	≥ 4
Peelability after water spraying 7 hrs. then drying at 50°C overnight (Glass, Ceramic)	≥ 4
Peelability after water spraying 7 hrs. then drying at 50°C overnight on PVDF	≥ 3

5

The results for room temperature peelability are provided in Table 3a and the results for cold temperature peelability are provided in Table 3b:

Table 3a. Room temperature peelability

	C. 1	C. 2	C. 3	C. 4	Ex. 1	C. 5	C. 6
glass	5	5	4	5	5	5	3
powder coated Al panel	5	na	na	5	5	na	na
anodized coated AL panel	5	na	na	5	5	na	na
PVDF coated AL panel	2	2	3	5	5	3	2

10

In the above and Tables below the term “na” stands for not available.

Table 3b. Low Temp. peelability

	C. 1	C. 2	C. 3	C. 4	Ex. 1	C. 5	C. 6
Glass	4	na	na	3	5	na	na
PVDF coated AL panel	2	na	na	3	5	na	na

15 Samples were also assessed after being sprayed with water

After curing/drying the coated substrates were put into a suitable laboratory fog box and sprayed with water for 7 hours to mimic rainy weather. The coated substrates were then dried at 50°C in an oven, after which the peelability of the peelable coating was assessed. This we believe is a harsher regime than any natural weather the peelable coatings are likely to be exposed to from natural weather even in hot climates such as in the Southeast Asia region.

20

The water resistance was assessed in accordance with the following standard:

5: No whitening, no wrinkle and no sliding for wet film and easily peeled off as a whole film after drying

4: Whitening but no wrinkles or sliding and easily peeled off as a whole film

3: Wet films whiten and bubble but no wrinkles or sliding. It can be peeled off as a whole film.

5 2: Wet films whiten, wrinkle and slide down a little. Hard to peel off after drying.

1: Wet films fully slide down from glass.

The water resistance results and an assessment of stickiness to the touch are provided for each sample in Table 3c:-

Table 3c. Hand Feel and Water resistance

	C. 1	C. 2	C. 3	C. 4	Ex. 1	C. 5	C. 6
Hand feeling of film surface	non sticky	non sticky	non sticky	non sticky	non sticky	non sticky	Sticky
Water Resistance	3	na	na	5	5	5	na

10 It was found that compositions using binder Ex. 1 in combination with SPEs in accordance with the definitions herein provided excellent peelability after water resistance results with the spraying as can be seen in the tables above.

Comparative C. 1 containing binder CB. 1 contained a comonomer binder mainly consisting of vinyl acetate repeating units and no styrene based repeating units. It was found such a coating gave poor water resistance & peelability results, particularly on a PVDF substrate.

Comparative binder 4 (CB. 4) contained an amount of a “self cross-linker, i.e., 2.8 wt. % acetoacetoxyethyl methacrylate and was considered to have poor low temperature peelability results.

Comparative binders 2, 3, 5 and 6 were all outside the scope of the invention and were found to have poor peelability results particularly on PVDF substrates.

CLAIMS

1. A peelable coating composition comprising

(a) an aqueous styrene-(meth)acrylic emulsion copolymer binder, wherein the copolymer is derived from at least monomers (a)(1), (a)(2) and (a)(3) wherein

5 (a)(1) is a styrene-based monomer in an amount of from 17.5 wt. % to 45 wt. %, of the solids content of the starting ingredients

(a)(2) is at least one (meth)acrylic acid monomer in an amount of from 0.1 wt. % to 4.5 wt. % of the solids content of the starting ingredients; and

(a)(3) at least one (meth)acrylate monomer;

10 Which copolymer is not derived from a vinyl acetate monomer as a starting ingredient and is not derived from a (meth)acrylic ester monomer having a reactive methylene group as a starting ingredient;

(b) a silicone polyether copolymer in an amount of from 0.75 to 10 wt. % of the peelable coating composition selected from one or both of

15 (i) $\text{RO}(\text{C}_3\text{H}_6\text{O})_m(\text{CH}_2\text{CH}_2\text{O})_{n''}\text{R}^{15}-((\text{CH}_3)_2\text{SiO})_x(\text{CH}_3)_2\text{Si}-\text{R}^{16}-(\text{OCH}_2\text{CH}_2)_{n'}(\text{OC}_3\text{H}_6)_m\text{OR}^1$, where:-
R and R¹ may be the same or are different and are each selected from H, or an alkyl group,
(C₃H₆O) is (CH₂(CH₃)CHO), (CH₂CH₂CH₂O), ((CH₃)CHCH₂O) or mixtures thereof;
(OC₃H₆) is (OCH₂CH(CH₃)), (OCH₂CH₂CH₂), (OCH(CH₃)CH₂) or mixtures thereof;
R¹⁵ and R¹⁶ are the same or different and are alkylene groups having from 2 to 6 carbons,

20 x is from 2 to about 500, m and m' may be the same or are different and are each in a range of from 0 to about 50 and n'' and n' may be the same or are different and are in a range of from 3 to about 50;
or

(ii) $(\text{CH}_3)_3\text{SiO}-((\text{CH}_3)_2\text{SiO})_x-(\text{CH}_3\text{R}'\text{SiO})_y-\text{Si}(\text{CH}_3)_3$;

Wherein R' = -R¹⁷-(OCH₂CH₂)_{n'}-(OC₃H₆)_mOR;

25 where R, x, m', n' and (OC₃H₆) are the same as above R¹⁷ is an alkylene group having from 2 to 6 carbons, and y is from 1 to about 100; and optionally

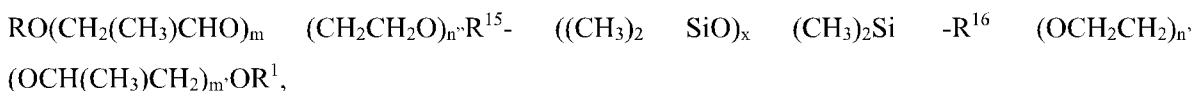
(c) one or more additives selected from aqueous solvent, coalescents, defoamers, rheology modifiers, wetting agents, plasticisers and coloring agents and/or a mixture thereof

2. A peelable coating composition in accordance with claim 1 wherein silicone polyether
30 copolymer (b)(i) has the formula

$\text{RO}(\text{C}_3\text{H}_6\text{O})_m(\text{CH}_2\text{CH}_2\text{O})_{n''}\text{R}^{15}-((\text{CH}_3)_2\text{SiO})_x(\text{CH}_3)_2\text{Si}-\text{R}^{16}-(\text{OCH}_2\text{CH}_2)_{n'}(\text{OC}_3\text{H}_6)_m\text{OR}^1$

Wherein R is hydrogen, m and m' are the same or different and are from 0 to 40, n'' and n' are from 5 to 30 and x is from 5 to 375.

3. A peelable coating composition in accordance with claim 1 or 2 wherein silicone polyether
35 copolymer (b)(i) has the formula



Wherein R and R¹ is hydrogen, m and m' are the same or different and are from 0 to 40, n'' and n' are the same or different and are from 5 to 30 and x is from 5 to 375.

- 5 4. A peelable coating composition in accordance with claim 4 wherein silicone polyether copolymer (b)(ii) has the formula $(\text{CH}_3)_3\text{SiO}-((\text{CH}_3)_2\text{SiO})_x-(\text{CH}_3 \text{R}'\text{SiO})_y-\text{Si}(\text{CH}_3)_3$;

Wherein R' = $-\text{R}^{17}-(\text{OCH}_2\text{CH}_2)_{n'}(\text{OC}_3\text{H}_6)_{m'}\text{OR}$;

where m' is from 0 to 45, y is from 1 to 25 and n' is from 4 to 35.

- 10 5. A peelable coating composition in accordance with any preceding claim wherein the styrene-based monomer (a)(1) is present in an amount 20 wt. % to 40 wt. % of the solids content of the starting ingredients of component (a).

6. A peelable coating composition in accordance with any preceding claim wherein monomer (a)(3) of aqueous styrene-(meth)acrylic emulsion copolymer binder (a) comprises at least one of
15 methyl (meth)acrylate, ethyl (meth)acrylate, butyl (meth)acrylate, decyl (meth)acrylate, lauryl (meth)acrylate, isodecyl (meth)acrylate, hydroxyethyl methacrylate, hydroxypropyl methacrylate, 2-ethylhexyl (meth)acrylate, 2-propylheptyl (meth)acrylate, and neopentyl (meth)acrylate.

7. A peelable coating composition in accordance with any preceding claim wherein aqueous styrene-(meth)acrylic emulsion copolymer binder (a) has a glass transition temperature (T_g) in the
20 range of from 5 to 40° C.

8. A peelable coating composition in accordance with any preceding claim wherein aqueous styrene-(meth)acrylic emulsion copolymer binder (a) is present in the amount of from 70 wt. % to 99.25 wt. % of the peelable coating composition and the additive(s) of component (c), when present, are cumulatively present in an amount of up to 19.25 wt. % of the peelable coating composition.

- 25 9. A peelable coating composition in accordance with any preceding claim wherein at least one additive (c) is present in the composition.

10. A peelable coating composition in accordance with any preceding claim wherein in the aqueous styrene-(meth)acrylic emulsion copolymer binder (a), the styrene-based monomer (a)(1) is present in an amount of from 17.5 wt. % to 37.5 wt. % of the solids content of the starting ingredients
30 thereof.

11. A substrate coated with a peelable coating which coating is the cured product of the peelable coating composition in accordance with any one of claims 1 to 10.

12. A method of forming a peelable coating on a substrate, the method comprising the steps of:
35 (I') combining an aqueous styrene-(meth)acrylic emulsion copolymer binder comprising

(a) an aqueous styrene-(meth)acrylic emulsion copolymer binder, wherein the copolymer is derived from at least monomers (a)(1), (a)(2) and (a)(3) wherein

(a)(1) is a styrene-based monomer in an amount of from 17.5 wt. % to 45 wt. %, of the solids content of the starting ingredients;

5 (a)(2) is at least one (meth)acrylic acid monomer in an amount of from 0.1wt. % to 4.5 wt. % of the solids content of the starting ingredients; and

(a)(3) at least one (meth)acrylate monomer;

Which copolymer is not derived from a vinyl acetate monomer as a starting ingredient and is not derived from a (meth)acrylic ester monomer having a reactive methylene group as a starting
10 ingredient;

with a silicone polyether copolymer (b) in an amount of from 0.75 to 10 wt. % of the peelable coating composition selected from one or both of

Wherein $R' = -R^{17} - (OCH_2CH_2)_n(OC_3H_6)_m$ OR;

and optionally

15 one or more additives (c) selected from aqueous solvent, coalescents, defoamers, rheology modifiers, wetting agents plasticisers, pigments and coloring agents and/or a mixture thereof, to form a peelable coating composition,

(II') applying the peelable coating composition on a substrate surface;

(III') drying the peelable coating composition applied in step (II') to form a peelable coating on the
20 substrate surface.

13. A method of forming a peelable coating on a substrate, in accordance with claim 12 wherein the wet coating thickness of the peelable coating composition once applied onto the substrate is from 100μm to 750μm measured using a coating bar.

14. A method of forming a peelable coating on a substrate, in accordance with claim 12 or
25 13 wherein the substrate is metal, plastic, glass, cloth, ceramic, clay, fiber, concrete, brick, rock or wood, painted walls and painted metals or substrates coated with polyvinyl difluoride (PVDF).

15. A method of forming a peelable coating on a substrate in accordance with claim 12, 13 or 14 wherein the method additionally comprises the step of removing the peelable coating by peeling it off the substrate surface onto which it has been applied.

30 16. A substrate coated with a peelable coating obtained or obtainable using the method in accordance with claim 12, 13 or 14.

17. Use of (a) an aqueous styrene-(meth)acrylic emulsion copolymer binder, in a peelable coating composition wherein the copolymer binder is derived from at least monomers (a)(1), (a)(2) and (a)(3) wherein

(a)(1) is a styrene-based monomer in an amount of from 17.5 wt. % to 45 wt. %, of the solids content of the starting ingredients

(a)(2) is at least one (meth)acrylic acid monomer in an amount of from 0.1 wt. % to 4.5 wt. % of the solids content of the starting ingredients; and

5 (a)(3) at least one (meth)acrylate monomer;

Which copolymer is not derived from a vinyl acetate monomer as a starting ingredient and is not derived from a (meth)acrylic ester monomer having a reactive methylene group as a starting ingredient;

10 which peelable coating composition otherwise comprises a silicone polyether copolymer (b) in an amount of from 0.75 to 10 wt. % of said peelable coating composition, which silicone polyether copolymer (b) is selected from one or both of

(i) $\text{RO}(\text{C}_3\text{H}_6\text{O})_m(\text{CH}_2\text{CH}_2\text{O})_n\text{R}^{15}-((\text{CH}_3)_2\text{SiO})_x(\text{CH}_3)_2\text{Si}-\text{R}^{16}-(\text{OCH}_2\text{CH}_2)_n'(\text{OC}_3\text{H}_6)_m'\text{OR}^1$, where:-

R and R¹ may be the same or are different and are each selected from H, or an alkyl group,

(C₃H₆O) is (CH₂(CH₃)CHO), (CH₂CH₂CH₂O), ((CH₃)CHCH₂O) or mixtures thereof;

15 (OC₃H₆) is (OCH₂CH(CH₃)), (OCH₂CH₂CH₂), (OCH(CH₃)CH₂) or mixtures thereof;

R¹⁵ and R¹⁶ are the same or different and are alkylene groups having from 2 to 6 carbons,

x is from 2 to about 500, m and m' may be the same or are different and are each in a range of from 0 to about 50 and n'' and n' may be the same or are different and are in a range of from 3 to about 50;

or

20 (ii) $(\text{CH}_3)_3\text{SiO}-((\text{CH}_3)_2\text{SiO})_x-(\text{CH}_3\text{R}'\text{SiO})_y-\text{Si}(\text{CH}_3)_3$;

Wherein R' = -R¹⁷-(OCH₂CH₂)_{n'}(OC₃H₆)_{m'}OR;

where R, x, m', n' and (OC₃H₆) are the same as above R¹⁷ is an alkylene group having from 2 to 6 carbons, and y is from 1 to about 100; and optionally

25 (c) one or more additives selected from aqueous solvent, coalescents, defoamers, rheology modifiers, wetting agents, plasticisers, pigments and coloring agents and/or a mixture thereof.

18. Use of (a) an aqueous styrene-(meth)acrylic emulsion copolymer binder, in a peelable coating composition in accordance with claim 17 wherein in the aqueous styrene-(meth)acrylic emulsion copolymer binder (a), the styrene-based monomer (a)(1) is present in an amount of from 17.5 wt. %
30 to 37.5 wt. % of the solids content of the starting ingredients thereof.

19. Use of a peelable coating composition in accordance with any one of claims 1 to 10 in the preparation of a peelable coating on a substrate wherein the substrate is selected from metal, plastic, glass, cloth, ceramic, clay, fiber, concrete, brick, rock or wood, painted walls and painted metals or
35 substrates coated with polyvinyl difluoride (PVDF).

INTERNATIONAL SEARCH REPORT

International application No
PCT/CN2023/100060

A. CLASSIFICATION OF SUBJECT MATTER INV. C09D5/00 C09D5/20 ADD.		
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) 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 practicable, search terms used) EPO-Internal		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2014/158043 A1 (WOODHALL EDWARD WILLIAM [US] ET AL) 12 June 2014 (2014-06-12) paragraph [0011] - paragraph [0017] paragraph [0045] - paragraph [0066] abstract; claims 1-6; figures 1, 2; tables 1-3 -----	1-19
X	WO 2018/067859 A1 (CAL WEST SPECIALTY COATINGS INC [US]) 12 April 2018 (2018-04-12) paragraph [0009] - paragraph [0034] paragraph [0065] - paragraph [0075] abstract; claims 1-17; example 1; tables 1, 2 ----- -/--	1-19
☒ Further documents are listed in the continuation of Box C. ☒ 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 application or patent but published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "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 "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance;; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "Y" document of particular relevance;; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art "&" document member of the same patent family		
Date of the actual completion of the international search		Date of mailing of the international search report
18 October 2023		27/10/2023
Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016		Authorized officer Glomm, Bernhard

INTERNATIONAL SEARCH REPORT

International application No

PCT/CN2023/100060

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT		
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