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(54) Title: WATER BASED COMPOSITION COMPRISING LIGHT DIFFUSION PARTICLES AND A SILOXANE-MODIFIED ACRYLIC RESIN

(57) Abstract: The present invention then concerns a water slurry composition comprising a light diffuser and a siloxane-modified acrylic resin allowing production of a coating on a transparent substrate. The invention also concerns a method for coating at least a part of a surface, preferably the surface of a transparent substrate, comprising at least the step of a) applying the composition of the invention on at least a part of the surface and b) carrying out the coating forming on the surface.



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Water based composition comprising light diffusion particles and a
siloxane-modified acrylic resin

The present invention then concerns a water slurry composition comprising a light diffuser and a siloxane-modified acrylic resin allowing production of a coating on a transparent substrate. The invention also concerns a method for coating at least a part of a surface, preferably the surface of a transparent substrate, comprising at least the step of a) applying the composition of the invention on at least a part of the surface and b) carrying out the coating forming on the surface.

PRIOR ART

The following discussion of the prior art is provided to place the invention in an appropriate technical context and enable the advantages of it to be more fully understood. It should be appreciated, however, that any discussion of the prior art throughout the specification should not be considered as an express or implied admission that such prior art is widely known or forms part of common general knowledge in the field.

Light emitting diode (LED) systems are becoming more prevalent as replacements for older lighting systems. LED systems have indeed the advantages over traditional lighting solutions such as incandescent and fluorescent lighting as they use less energy, are more durable, operate longer, can be combined in multi-color arrays that can be controlled to deliver virtually any color light, and generally contain no lead or mercury.

In recent years, LED devices have been deployed in various applications, including indicators, light sensors, traffic lights, broadband data transmission, and illumination devices. For example, LED devices are often used in illumination devices provided to replace conventional incandescent light bulbs, such as those used in a troffer light.

However, LED devices typically are highly directional by nature. Common LED devices are flat and emit light from only one side. They produce intense light within the beam of their output, but dim light outside of that beam. Using multiple LEDs does not fully alleviate this problem, as there are then interference patterns in the light. Thus, although superior in performance, many

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commercially-available LED lamps cannot achieve intensity distribution of incandescent lamps.

It is then known to provide a light-transparent or light-filtering coating on a transparent substrate close to the LEDs, usually added to their shells or bodies, in order to improve the luminous intensity distribution by spreading out the light from the LED. However the known coating compositions do not permit to allow a scratch resistant homogeneous coating without letting appear bubble on the surface coating. Another method has been to roughen the surface of the LED package. Neither of these methods accomplishes uniform light distribution for an LED light source, and may lower luminous efficiency. Methods of accomplishing approximate angular uniformity may also involve partially absorptive processes, further lowering luminous efficacy.

Therefore, while conventional LEDs have been generally adequate for their intended purposes, they have not been entirely satisfactory in every aspect. It is hence desired to provide LED assemblies that distribute light in more uniform fashion across all directions, similar to that of an incandescent light bulb.

INVENTION

The present invention then concerns a water slurry composition comprising at least a light diffuser and a siloxane-modified acrylic resin allowing production of a coating on a transparent substrate having not the above identified drawbacks, with a sufficient transparency. The improvement notably concerns on construction of a diffuser element for LEDs that can disperse light from the light sources to generate a light intensity pattern, for instance similar to incandescent light sources; then overcoming the non-uniform light distribution issues associated with conventional LEDs. It also appears that the compositions for coating of the invention permit to obtain a surface coating with an excellent coating yield and a very good balance of anti-scratch, transparency and shading properties. Moreover, the compositions for coating of the present invention permit to obtain a very efficient anti-scratch and anti-UV properties.

The invention notably concerns a composition comprising at least light diffusion particles, a siloxane-modified acrylic resin and water.

The present invention concerns a method for coating at least a part of a surface, preferably the surface of a transparent substrate, comprising at least the step of :

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- a) applying the composition of the invention on at least a part of the surface;
and
- b) carrying out the coating forming on the surface.

The present invention also concerns a LED assembly comprising at least a
5 LED light source component and a transparent substrate having at least a part of
the surface coated by the above identified method. The invention also relates to
a LED assembly comprising at least a LED light source component and a
transparent substrate having at least a part of the surface coated by a composition
comprising at least light diffusion particles, and a siloxane-modified acrylic resin.
10 Other characteristics, details and advantages of the invention will emerge
even more fully upon reading the description which follows.

DETAILS OF THE INVENTION

Throughout the description, including the claims, the term "comprising
15 one" should be understood as being synonymous with the term "comprising at
least one", unless otherwise specified, and "between" should be understood as
being inclusive of the limits.

It is perfectly possible in the composition of the invention one, two or more
different siloxane-modified acrylic resins.

20 Acrylic resins are usually derived from acrylic acid, methacrylic acid or
other related compounds, such as for instance polymethyl acrylate, polymethyl
methacrylate, polyethyl acrylate and polybutyl acrylate polymers and
copolymers.

Poly(methyl methacrylate) (PMMA) or polymethyl methacrylate is notably
25 produced by emulsion polymerization, solution polymerization, and bulk
polymerization.

Polymethyl methacrylates are generally obtained via free-radical
polymerization of mixtures which comprise methyl methacrylate. These
mixtures generally comprise at least 40 % by weight, preferably at least 60 % by
30 weight, and particularly preferably at least 80 % by weight, of methyl
methacrylate, based on the weight of the monomers.

Alongside this, these mixtures for preparing polymethyl methacrylates may
comprise other (meth)acrylates which are copolymerizable with methyl
methacrylate. The term (meth)acrylates encompasses methacrylates and
35 acrylates, and also mixtures of the two.

These monomers are well known and notably include (meth)acrylates derived from saturated alcohols, for example methyl acrylate, ethyl (meth)acrylate, propyl (meth)acrylate, n-butyl (meth)acrylate, tert-butyl (meth)acrylate, pentyl (meth)acrylate and 2-ethylhexyl (meth)acrylate; (meth)acrylates derived from unsaturated alcohols, for example oleyl (meth)acrylate, 2-propynyl (meth)acrylate, allyl (meth)acrylate, vinyl (meth)acrylate; aryl (meth)acrylates, such as benzyl (meth)acrylate or phenyl (meth)acrylate, where in each case the aryl radicals may be unsubstituted or have up to four substituents; cycloalkyl (meth)acrylates, such as 3-vinylcyclohexyl (meth)acrylate, bornyl (meth)acrylate; hydroxyalkyl (meth)acrylates, such as 3-hydroxypropyl (meth)acrylate, 3,4-dihydroxybutyl (meth)acrylate, 2-hydroxyethyl (meth)acrylate, 2-hydroxypropyl (meth)acrylate; glycol di(meth)-acrylates, such as 1,4-butanediol (meth)acrylate, (meth)acrylates of ether alcohols, for example tetrahydrofurfuryl (meth)acrylate, vinyloxyethoxyethyl (meth)acrylate; amides and nitriles of (meth)acrylic acid, for example N-(3-dimethylaminopropyl)-(meth)acrylamide, N-(diethylphosphono)(meth) acrylamide, 1-methacryloylamido-2-methyl-2-propanol; sulphur-containing methacrylates, such as ethylsulphinylethyl (meth)acrylate, 4-thiocyanatobutyl (meth)acrylate, ethylsulphonylethyl (meth)acrylate, thiocyanatomethyl (meth)acrylate, methylsulphinylmethyl (meth)acrylate, bis((meth)acryloyloxyethyl) sulphide; polyfunctional (meth)acrylates, such as trimethyloxypropane tri(meth)acrylate.

Besides the abovementioned (meth)acrylates, the compositions to be polymerized may also comprise other unsaturated monomers copolymerizable with methyl methacrylate and the abovementioned (meth)acrylates. They include 1-alkenes, such as 1-hexene, 1-heptene; branched alkenes, such as vinylcyclohexane, 3,3-dimethyl-1-propene, 3-methyl-1-diisobutylene, 4-methyl-1-pentene; acrylonitrile; vinyl esters, such as vinyl acetate; styrene, substituted styrenes having an alkyl substituent in the side chain, e.g. α -methylstyrene and α -ethylstyrene, substituted styrenes having an alkyl substituent on the ring, such as vinyltoluene and p-methylstyrene, halogenated styrenes, such as monochlorostyrenes, dichlorostyrenes, tribromostyrenes and tetrabromostyrenes; heterocyclic vinyl compounds, such as 2-vinylpyridine, 3-vinylpyridine, 2-methyl-5-vinylpyridine, 3-ethyl-4-vinylpyridine, 2,3-dimethyl-5-vinylpyridine, vinylpyrimidine, vinylpiperidine, 9-vinylcarbazole, 3-vinylcarbazole, 4-vinylcarbazole, 1-vinylimidazole, 2-methyl-1-vinylimidazole, N-vinyl-

pyrrolidone, 2-vinylpyrrolidone, N-vinylpyrrolidine, 3-vinylpyrrolidine, N-vinylcaprolactam, N-vinylbutyrolactam, vinyloxolane, vinylfuran, vinylthiophene, vinylthiolane, vinylthiazoles and hydrogenated vinylthiazoles, vinyloxazoles and hydrogenated vinyloxazoles; vinyl and isoprenyl ethers; maleic acid derivatives, such as maleic anhydride, methylmaleic anhydride, maleimide, methylmaleimide; and dienes, such as divinylbenzene.

The amount generally used of these co-monomers is from 0 to 60 % by weight, preferably from 0 to 40 % by weight, and particularly preferably from 0 to 20 % by weight, based on the weight of the monomers, and these compounds may be used individually or in the form of a mixture.

As used herein, the term "siloxane" refers to straight-chain compounds having silicon atoms single-bonded to oxygen atoms and so arranged that each silicon atom is linked to at least one oxygen atom. Preferably, siloxanes of the present invention will be silicones (i.e. siloxane polymers based upon a structure consisting of alternating silicon and oxygen atoms with various organic radicals attached to the silicon atoms).

Siloxane-modified acrylic resins used in the present invention are well known products. Siloxane-modified acrylic resins in the invention may be notably used as a coating forming binder in the compositions of the invention.

Siloxane-modified poly(methyl methacrylate) may notably be for instance :

- a blend of siloxane and poly(methyl methacrylate).
- a crosslinked network of siloxane and poly(methyl methacrylate)
- a copolymer of poly(methyl methacrylate) and siloxane compound, notably by using functionalized silane or siloxane able to react with methyl methacrylate or poly(methyl methacrylate).

Siloxane-modified poly(methyl methacrylate) may notably be obtained by reaction of poly(methyl methacrylate) with silane compounds carrying an unsaturated organic substitution, such as acrylate and methacrylate functionalized silanes. It is also possible to obtain siloxane-modified poly(methyl methacrylate) by reaction of poly(methyl methacrylate) and alkyl aromatic silane. Siloxane-modified poly(methyl methacrylate) may also be the result of a crosslinking between siloxane polymer and poly(methyl methacrylate) polymer.

Different types of crosslinkings are notably possible between the siloxane and poly(methyl methacrylate), such as for instance

- (i) covalent crosslinking, which is regarded as the most stable,

- (ii) ionic bonds, and/or
- (iii) physical crosslinking, such as via Van der Waals, hydrogen bonds or other interactions.

Preferably, the siloxane-modified acrylic resin is a copolymer of
 5 poly(methyl methacrylate) and a silane compound and/or a siloxane compound.

Siloxane-modified acrylic resins may notably be obtained by reaction, notably copolymerization, of poly(methyl methacrylate) (PMMA) with silane having unsaturated organic substitution, such as vinyl-triethoxysilane or methacryloxysilane, for instance methacryloxypropyl-trimethoxysilane, as
 10 described in *Acta Biomaterialia* 1, 671-676 (2005).

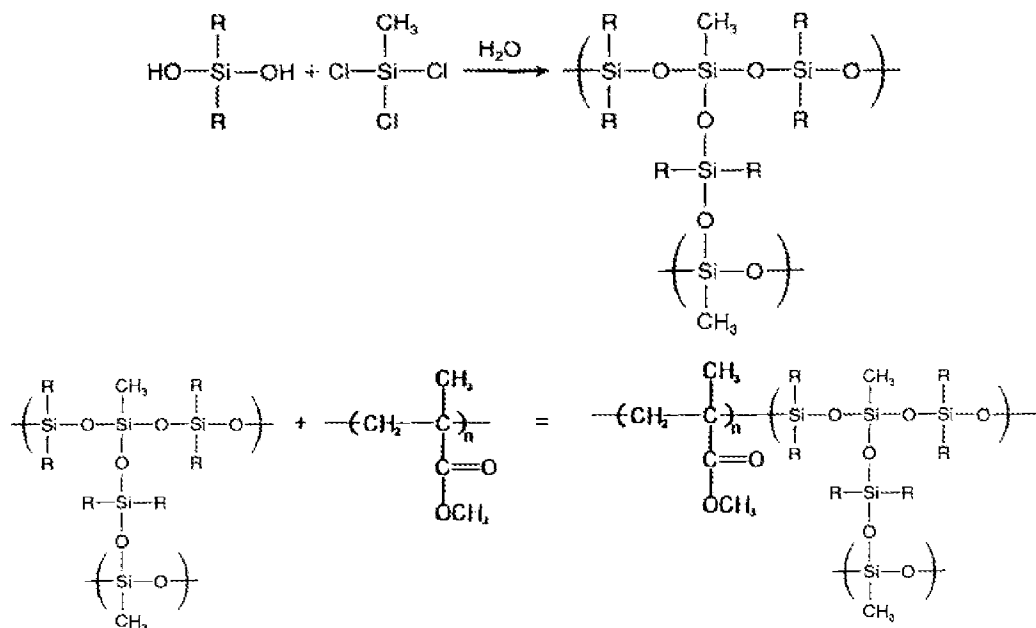
It is also possible to produce siloxane-modified acrylic resins by emulsion copolymerization of acrylic resin monomers, such as methylmethacrylate, butylacrylate, and/or methacrylic acid, with silane having unsaturated organic substitution, such as vinyltriethoxysilane, as described in *Iranian Polymer*
 15 *Journal*, 14 (3), 2005, 211-222.

Siloxane-modified acrylic resins may also be obtained via sequential or simultaneous polymerization in order to produce semi-interpenetrating polymer networks (IPNs), for instance composed of poly(methyl methacrylate) and aromatic and/or aliphatic siloxanes, as notably disclosed in *Journal of Polymer*
 20 *Science: Part A: Polymer Chemistry*, Vol 34, 1025-1037 (1996).

Acrylate and methacrylate functionalized silanes may be selected for instance in the group consisting of : (3-acryloxypropyl)trimethoxy-silane, methacryloxypropyltrimethoxy-silane, n-(3-acryloxy-2-hydroxypropyl)-3-aminopropyltriethoxysilane, o-(methacryloxyethyl)-n-(triethoxy-
 25 silylpropyl)urethane, n-(3-methacryloxy-2-hydroxypropyl)-3-aminopropyltriethoxysilane, methacryloxymethyltriethoxysilane, methacryloxypropyltriethoxysilane, (3-acryloxypropyl)methyldimethoxy silane, (methacryloxymethyl)methyl- diethoxysilane, (methacryloxymethyl)methyl-
 30 dimethoxysilane, methacryloxypropylmethyldi- ethoxysilane, methacryloxypropylmethyldi- methoxysilane, methacryloxypropyldimethylethoxy-silane, and methacryloxypropyldimethyl- methoxysilane.

Siloxane-modified acrylic resins may also be a crosslinking polymer obtained by reaction between siloxane polymer and poly(methyl methacrylate)
 35 polymer in order to obtain a three-dimensional polymer network as expressed as follows :

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It notably permits to obtain a film having a strong water resistance and adhesion; as described in Journal of Applied Polymer Science, Vol. 86, 1736-1740 (2002) and Journal of Polymer Science: Part A: Polymer Chemistry, Vol. 34, 1025-1037 (1996).

Siloxane-modified acrylic resins may notably be used as a water emulsions for the production of the composition of the invention. Water emulsions may be water-in-oil or oil-in-water emulsions, preferably oil-in-water. More preferably siloxane-modified acrylic resin is a oil in water emulsion comprising from 35-45 %wt of resin and from 55-65 %wt of water.

Light diffusion particles are preferably inorganic particles, usually chosen in the group consisting of : LaPO₄, CaCO₃, Y₂O₃, YVO₃, TiO₂, ZrO₂, MgO, CaO, SmTi₂O₇, LaZr₂O₇, CeTi₂O₇, CeO₂, La₂O₃, LaHf₂O₇, HfO₂, SnO₂, Ca₃(PO₄)₂, BaSO₄, Al₂O₃ and/or ZnO.

The average size D50 of the primary particles (crystallites) of the light diffusion particles may be comprised between 0.1 μm and 10 μm, preferably between 0.2 μm and 6 μm, more preferably between 0.4 μm and 5 μm. The average value of the size of the primary particles may be determined by the SEM technique.

Secondary particles are aggregates made from other, finer particles called primary particles or crystallites. The secondary particle size D50 of the light diffusion particles may be comprised between 0.1 μm and 100 μm, preferably between 0.5 μm and 20 μm, more preferably between 1 μm and 10 μm. The size

of the secondary particles may be measured by using a Horiba LA920 laser particle sizer or a Malvern mastersizer S.

It is perfectly possible in the composition of the invention one, two or more different light diffusing particles.

5 Composition of the invention may comprise between 10 and 40 % by weight, preferably between 20 and 40 % by weight of light diffusion particles. Composition of the invention may also comprise between 5 and 30 % by weight, preferably between 10 and 20 % by weight of a siloxane-modified acrylic resin. Composition of the invention may notably comprise between 20 and 70 % by
10 weight of water. Percent by weight are expressed in comparison with the total weight of the composition.

Composition of the present invention preferably comprises at least :

- between 10 and 40 % by weight, preferably between 20 and 40 % by weight of light diffusion particles,
- 15 - between 5 and 30 % by weight, preferably between 10 and 20 % by weight of a siloxane-modified acrylic resin, and
- between 30 and 85 % by weight of water, preferably between 20 and 70 % by weight of water.

Composition of the present invention may also comprise one or more
20 additives, such as for instance an additive chosen in the group consisting of : polyether polyol, binders, dispersants, anti-scratch additives, and thickening agents.

Polyether polyols usually refer to polyols comprising poly(alkylene oxide) chains, which polyols are normally obtained by reacting a polyhydroxy initiator
25 compound with at least one alkylene oxide and optionally other compounds.

Methods for preparing polyether polyols, also sometimes referred to as poly(oxyalkylene) polyols, are well known in the art. Typically, such methods involve reacting a starting compound having a plurality of active hydrogen atoms with one or more alkylene oxides, such as ethylene oxide, propylene oxide,
30 butylene oxide or mixtures of two or more of these. Suitable starting compounds include polyfunctional alcohols, generally containing 2 to 6 hydroxyl groups. Examples of such alcohols are glycol, such as diethylene glycol, dipropylene glycol, glycerol, di- and polyglycerols, pentaerythritol, trimethylolpropane, triethanolamine, sorbitol, sucrose, mannitol, etc. Usually a strong base like an
35 alkali metal hydroxide (typically potassium hydroxide, cesium hydroxide or sodium hydroxide) is used as a catalyst in this type of reaction.

Polyether Polyol may be defined as a compound formed through the polymerization of ethylene oxide (EO) or propylene oxide (PO) or other cyclic ethers with compounds having one or more reactive hydrogens (i.e., a hydrogen atom bonded to nitrogen, oxygen, phosphorus, sulfur, etc.) to form polyethers.

5 Preferably, polyether polyols of the invention may be poloxamers, that are nonionic triblock copolymers composed of a central hydrophobic chain of polyoxypropylene (poly(propylene oxide)) flanked by two hydrophilic chains of polyoxyethylene (poly(ethylene oxide)). Poloxamers are also known by the trade names Synperonics, Plurionics, and Kolliphor.

10 Composition of the present invention preferably comprises between 0.01 and 2 % by weight, preferably between 0.05 and 0.1 % by weight of a polyether polyol.

Dispersants may be for instance ethoxylated tristyrilphenol phosphate potassium salt. Anti-scratch additives may be for instance silicone, paraffin and
15 silane. Thickening agents may be for instance cellulose and acrylic acid.

Composition of the present invention preferably comprises a polyether polyols siloxane copolymer.

This polyether polyols siloxane copolymer may be the polymerization product from polyether polyols siloxane. Polyether polyols are mainly PPG and
20 POP types, PPG is synthesized by monohydric or organic amine with propylene oxide (PO). POP is PPG polymerization with acrylonitrile (AN) or styrene (SN). Siloxane may be selected in the cyclic siloxane group consisting of: hexamethylcyclotrisiloxane, octamethylcyclotetrasiloxane, decamethylcyclopentasiloxane, and dodecamethylcyclohexasiloxane. Siloxane
25 also may be selected in the linear siloxane group consisting of: octamethyltrisiloxane, decamethyltetrasiloxane, dodecamethylpentasiloxane, and tetradecamethylhexasiloxane.

Preferably polyether polyols siloxane copolymers are selected in the group constituted of: WSG-125 from Wanzhao chemical, DF 805,1587 from Defeng,
30 Tego Foamex 4200, Tego Foamex 1488, Tego Foamex 825 and Tego Foamex 843 from Evonik.

Composition of the present invention may provide a viscosity comprised between 13S and 22S preferably between 15S and 20S according to C4 cup methodology described in the GB/T 1723-1993 standard permitting
35 determination of viscosity.

Composition of the present invention may notably be obtained by mixing at least light diffusion particles, a siloxane-modified acrylic resin and water, optionally with additives.

Composition of the present invention may be notably used for the coating forming on a substrate, notably a transparent substrate.

The present invention concerns a method for coating at least a part of a surface comprising at least the step of :

- a) applying the composition of the invention on at least a part of the surface; and
- b) carrying out the coating forming on the surface.

A part of the surface is usually defined as a portion of the total area of the surface. Composition of the invention may notably be applied on a part of the surface, for instance between 80 and 100 % of the total area of the surface, preferably between 95 and 100 % of the total area of the surface. Composition of the invention may notably be applied on one or two faces of the surface.

The composition can be applied in a liquid state on a surface by dipping, spraying, roller coating, flooding, ink jet, pad printing, flexographic printing, and screen printing.

Coating forming is the generic term for the transition of a coating layer from the liquid state to the solid state, or hardening. The composition of the invention may be hardened in particular by a thermal method, usually by drying or UV hardening. The term "drying" is related to the process engineering used for drying the liquid layer, notably at a temperature comprised between 40 and 90°C, preferably between 50 and 80°C. Drying may notably occur under a hot air gas. The residual content of water in the coating is usually comprised between 20 and 70 % by weight of the weight of the overall water.

The coating may notably be a layer of a thickness comprised between 5 and 30 μm , preferably comprised between 10 and 20 μm .

A transparent substrate, may be for instance a glass substrate or a plastic substrate such as poly(methyl methacrylate), polycarbonate, or polyvinyl chloride polymer.

The composition of the substrate glass, for instance, may be composed of or comprises the following oxides in various compositions : SiO_2 , B_2O_3 , Bi_2O_3 , P_2O_5 , K_2O , Cs_2O , SrO , GeO_2 , Al_2O_3 , Li_2O , Na_2O , CaO , BaO , ZnO , La_2O_3 , Gd_2O_3 , Y_2O_3 , Ta_2O_5 , Nb_2O_5 , TiO_2 , ZrO_2 , WO_3 , As_2O_3 , Sb_2O_3 , TiO_2 and/or ZrO_2 .

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Transparent substrate preferably has an index of refraction comprised between 1.4 and 1.7. Once coated by the composition of the present invention, the transparent substrate has an index of refraction higher in comparison with non-coated transparent substrate; notably comprised between 1.45 and 3. Refraction index Value may be measured at 435 nm from Handbook of Optics. Lamp lumen flux test methods are standard according to GB/T9468-2008. Optical transmission data may correspond to the flux of T8 LED strip with coated transparent substrate divided by the flux T8 LED strip without coating.

The invention also concerns a LED assembly comprising at least a LED light source component and a transparent substrate having at least a part of the surface coated by the above identified method.

The term "LED light source component" refers to a light source component which includes at least one light emitting diode. The LED light source component may also concern lighting modules which include one or more such light source components. The LED light source component may also concern a lighting system which includes a plurality of such lighting modules. A LED lighting system may include, for example, a packaged light emitting device including one or more light emitting diodes (LEDs), which may include inorganic LEDs, which may include semiconductor layers forming p-n junctions and/or organic LEDs (OLEDs), which may include organic light emission layers.

Some light emitting diodes may have a diffusion lens, such as for instance LED of SMD type.

Usually a LED assembly may have a round or bulb shape or a tube shape, such as for instance straight tube type lamp.

LED straight tube type lamp mainly comprises a LED light source component, usually in a shape of a LED holder, also referred as belt or strip, which is arranged in a transparent tube body. The LED holder and the transparent tube body are usually adhered by using chemical adhesives, such as silicon adhesives.

The composition of the invention is usually applied on at least a part of the inner surface of the transparent tube body, preferably on the total inner surface of the transparent tube body.

The following examples are included to illustrate embodiments of the invention. Needless to say, the invention is not limited to described examples.

EXPERIMENTAL PART

Raw materials used in this experimental part as are follows :

- Water emulsion of siloxane-modified poly(methyl methacrylate) : MK2272 grade from Tianjin Yining Meike Fine Chemical Co., Ltd
- 5 - PAA : water emulsion of acrylic resin. AC261P grade from Dow Chemical
- PAA-Silica hybrid. DV-961 grade from JP DIC., Ltd.
- Calcium carbonate (CaCO_3). Super 4S grade from JP Maruo.,Ltd
- Polyether siloxane copolymer. Tego Foamex 843 grade from EVONIK
- Triethanolamine. AR grade from Guo Yao group

10

Example 1 : preparation of the water slurry

144 g of deionized water was introduced into a tank, in which 144 g CaCO_3 and 0.4 g potassium salt of ethoxylated tristyrilphenol phosphate was added. The mixture is blended by an agitator at 180rpm for 30mins.

- 15 Then 143.6 g of water emulsions of siloxane-modified poly(methyl methacrylate) was added to the previous mixture. Mixture was stirred with an agitator at 180 rpm for 10mins.

- 20 30 g of 10 % by weight of an acrylic emulsion thickener in deionized water, was added to the water slurry. Mixture was then stirred with an agitator at 180 rpm for 2 h. 0.1 g polyether siloxane copolymer and 4.7 g dipropylene glycol butyl ether were and added and the resulting mixture was stirred with an agitator at 180 rpm for 2 h. 4.7 g triethanolamine was added and the mixture was stirred with an agitator at 180 rpm for 0.5 h. Resulting water slurry is passed then through a 200 mesh filter.

- 25 Viscosity of the water slurry as obtained is 18S, according to C4 cup methodology described in the GB/T 1723-1993 standard

Example 2 : Coating process

- 30 A diaphragm pump was used to load up the water slurry composition as obtained in Example 1 on T8 tube auto coating machine (YD-T8 from Shangyu Yuandong). Water slurry composition was sprayed into T8 tubes, and then a 50-80°C hot air gas was sent into the tubes in order to dry the water slurry to form a homogeneous coating layer inside T8 tubes. Thickness of the coating is about 10-15 μm , for a coating total weight of 3.0 g per tube.

- 35 Some properties were tested and results are expressed in Table 1

Table 1

Components (% wt.)	C1	C2	1	2	3
CaCO ₃	30.5 %	30.5 %	30.5 %	30.5 %	30.5 %
Resin	PAA : 11.9 %	PAA- Silica hybrid : 11.9 %	Siloxane PMMA : 11.9 %	Siloxane PMMA : 11.9 %	Siloxane PMMA : 11.9 %
Additive	Polyether siloxane copolymer : 0.1 %	Polyether siloxane copolymer : 0.1 %	Polyether siloxane copolymer : 0.1 %	Polyether siloxane copolymer : 0.1 %	-
Additive	-	-	-	Triethanolamine 0.1 %	-
Water	qsp	qsp	qsp	qsp	qsp
Properties					
Anti-scratch	2/5	4.5/5	4.5/5	4.5/5	4.5/5
Glass wetting	5/5	5/5	5/5	5/5	5/5
Anti-bubble	4/5	3/5	4/5	4/5	2/5
Transparency	4/5	4/5	4/5	5/5	4/5
Shading	5/5	4/5	5/5	5/5	5/5
Auto coating yield	5/5	5/5	5/5	5/5	3/5
Anti-UV	3/5	4/5	5/5	5/5	5/5
Anti-bacteria	2/5	2/5	2/5	4/5	2/5

Tested properties are as follows :

1) **Anti-scratch property** : Finger scratch test (one pass) :

5= tube coating has no scratch

5 4= coating only has scratch at tube top

3= coating has scratch at tube top and bottom

2= coating at tube top is removed

1= both tube top and bottom is removed

10 2) **Glass wetting property** : Quality of the coating on the T8 tube

5= coating is homogeneous and no defect

4= coating is homogeneous and less than two hole (<2mm)

3= coating is homogeneous and more than 2 hole (<2mm), or have more than one big hole (>2mm)

15 2= less 10 % tube surface is not covered by coating

1= more than 10 % tube surface is not covered by coating

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3) **Anti-bubble property** : Visual aspect of the coating

5= both slurry and coating on T8 tube has no bubble

4= only slurry has some bubble and coating T8 tube has no bubble

3= coating on T8 tube has less than 10 bubbles

5 2= coating on T8 tube has more than 10 bubbles

1= coating on T8 tube having too much bubbles to count them properly

4) **Transparency property** : Optical transmission

10 Optical transmission is measured by integrative sphere (SL-300 from HangZHou Zhejiang University sensing Instruments company). Lamp lumen flux test method according to GB/T9468-2008.

5=optical transmission of T8 tube coating is higher than 88 % (included)

4=optical transmission of T8 tube coating is comprised between 86 % (included) and 88 % (non included)

15 3=optical transmission of T8 tube coating is comprised between 84 % (included) and 86 % (non included)

2=optical transmission of T8 tube coating is comprised between 82 % (included) and 84 % (non included),

1=optical transmission of T8 tube coating is below 82 % (non included)

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5) **Shading property** : Light intensity difference on the same tube coating at 2 different points, detected by light sensor (PM200T from Chuanghui instrument company)

5=shading data is higher than 95 % (included)

25 4=shading data is comprised between 85 % (included) and 95 % (non included)

3=shading data is comprised between 75 % (included) and 85 % (non-included)

2=shading data is comprised between 65 % (included) and 75 % (non-included)

30 1=shading data is comprised between 55 % (included) and 65 % (non-included)

6) **Auto coating yield property** : Number of T8 coating having a glass wetting rate of 4 or 5 and anti-bubble rate of 4 or 5

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5= yield is higher than 85 % (included)

4= yield is comprised between 75 % (included) and 85 % (non included)

3= yield is comprised between 60 % (included) and 75 % (non included)

2= yield is comprised between 50 % (included) and 60 % (non included)

1= yield is comprised between 40 % (included) and 50 % (non included)

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7) **Anti-UV property** : A 100 µm wet layer on glass pane is made by using the wet layer maker as previously described. Wet layer is then dried in an oven at 80°C for 20 min. Optical transmission (OP0) of the glass is tested with PM-200T (from Inventfine, Ltd) before ageing (OP0) and after ageing (OP1) with an UV aging machine (WFH-203B from SHJK, Ltd), switch on 365nm lights for 72 h.

Anti-UV performance = OP1/OP0

5= OP1/OP0 is comprised between 95 % (included) and 100 % (included)

4= OP1/OP0 is comprised between 90 % (included) and 95 % (excluded)

15 3= OP1/OP0 is comprised between 85 % (included) and 90 % (excluded)

2= OP1/OP0 is comprised between 80 % (included) and 85 % (excluded)

1= OP1/OP0 is inferior to 80 % (excluded)

8) **Anti-bacteria property** : An anti-bacteria testing paper (Microbiology Cult Dip Combi M from Merck) is put inside the coating and aged for 30 days at 25°C.

5=no mildew

4= 1 small mildew covers less than 5 % (excluded) of testing paper surface area

25 3= more than 1 small mildew covers between 5 % (included) and 10 % (excluded) of testing paper surface area

2= more than 1 mildew covers between 10 % (included) and 20 % (excluded) of testing paper surface area

30 1= more than 1 mildew covers more than 20 % (excluded) of testing paper surface area

It appears then that the composition of the present invention permits obtaining of a tube coating with a very good balance of properties, notably anti-scratch, anti-UV, transparency, glass wetting and shading properties, in comparison with comparative formulations that are mainly deficient in the anti-scratch and anti-UV properties.

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CLAIMS

1. A composition comprising at least light diffusion particles, a siloxane-modified acrylic resin and water.
2. Composition according to claim 1 wherein the siloxane-modified
5 acrylic resin is a copolymer of poly(methyl methacrylate) and a silane compound and/or a siloxane compound.
3. Composition according to claim 1 or 2 wherein light diffusion particles are chosen in the group consisting of : LaPO_4 , CaCO_3 , Y_2O_3 , YVO_3 , TiO_2 , ZrO_2 , MgO , CaO , SmTi_2O_7 , LaZr_2O_7 , CeTi_2O_7 , CeO_2 , La_2O_3 , LaHf_2O_7 ,
10 HfO_2 , SnO_2 , $\text{Ca}_3(\text{PO}_4)_2$, BaSO_4 , Al_2O_3 and/or ZnO .
4. Composition according to anyone of claims 1 to 3 wherein the average size D50 of the primary particles of the light diffusion particles is comprised between $0.1\text{ }\mu\text{m}$ and $10\text{ }\mu\text{m}$.
5. Composition according to anyone of claims 1 to 4 wherein the average
15 size D50 of the secondary particle of the light diffusion particles is comprised between $0.1\text{ }\mu\text{m}$ and $100\text{ }\mu\text{m}$.
6. Composition according to anyone of claims 1 to 5 wherein the composition comprises between 10 and 40 % by weight of light diffusion particles, expressed in comparison with the total weight of the composition.
- 20 7. Composition according to anyone of claims 1 to 6 wherein the composition comprises between 5 and 30 % by weight of a siloxane-modified acrylic resin, expressed in comparison with the total weight of the composition.
8. Composition according to anyone of claims 1 to 7 wherein the composition comprises between 20 and 70 % by weight of water, expressed in
25 comparison with the total weight of the composition.
9. Composition according to anyone of claims 1 to 8 wherein the composition comprises at least one additive chosen in the group consisting of : polyether polyol, binders, dispersants, anti-scratch additives, and thickening agents.

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10. Composition according to anyone of claims 1 to 9 wherein the composition comprises between 0.01 and 2 % by weight of a polyether polyol.

11. A method for coating at least a part of a surface comprising at least the step of :

- 5 a) applying the composition according to anyone of claims 1 to 10 on at least a part of the surface; and
- b) carrying out the coating forming on the surface.

12. Method according to claim 11 wherein the coating forming on the surface in step b) is a hardening carried out by drying at a temperature comprised
10 between 40 and 90°C.

13. Method according to claim 11 or 12 wherein the coating is a layer with a thickness comprised between 5 and 30 µm.

14. A LED assembly comprising at least a LED light source component and a transparent substrate having at least a part of the surface coated by the
15 method according to anyone of claims 11 to 13.

15. A LED assembly comprising at least a LED light source component and a transparent substrate having at least a part of the surface coated by a composition comprising at least light diffusion particles and a siloxane-modified acrylic resin.

20 16. LED assembly according to claims 14 or 15 wherein the LED assembly is a straight tube type lamp.

INTERNATIONAL SEARCH REPORT

International application No.

PCT/CN2015/076133

A. CLASSIFICATION OF SUBJECT MATTER

C09D 133/06(2006.01)i; C09D 5/33(2006.01)i; C09D 7/12(2006.01)i; C09D 143/04(2006.01)i

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; C09J

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)

CNABS;VEN;CNKI;CNTXT:acrylate, acrylic, silane, siloxane, light emitting diode, LED, water, light, diffusion, scatter, particle, refraction, LaPO₄, CaCO₃, Y₂O₃, YVO₃, TiO₂, ZrO₂, MgO, CaO, SmTi₂O₇, LaZr₂O₇, CeTi₂O₇, CeO₂, La₂O₃, LaHf₂O₇, HfO₂, SnO₂, Ca₃(PO₄)₂, BaSO₄, Al₂O₃, ZnO**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	JP 2007224187 A (TOYO INK MFG CO) 06 September 2007 (2007-09-06) see description, paragraphs [0001]-[0002],[0041] and example 1	1-16
X	CN 102627893 A (JIANGSU TONGHUI LIGHTING SCI & TECHNOLOG) 08 August 2012 (2012-08-08) see description, paragraphs [0002]-[0005]	1,3-7,9-16
X	CN 101121771 A (HENKEL KGAA) 13 February 2008 (2008-02-13) see claims 1-40	1-13
X	KR 1183892 B1 (KCC CORP) 19 September 2012 (2012-09-19) see claims 1-7	1-13
X	CN 103360890 A (CHINESE ACAD SCI GUANGZHOU CHEM RES INST) 23 October 2013 (2013-10-23) see claims 1-10	1-13
X	CN 101864235 A (NEIMENGGU XINGTAI CONSTR CO LTD) 20 October 2010 (2010-10-20) see claims 1-2	1-13



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

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INTERNATIONAL SEARCH REPORT

International application No.

PCT/CN2015/076133**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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X	DE 102007061876 A1 (BAYER MATERIALSCIENCE AG) 25 June 2009 (2009-06-25) see claims 1-15	1-13
X	DE 102007061871 A1 (BAYER MATERIALSCIENCE AG) 25 June 2009 (2009-06-25) see claims 1-15	1-13

INTERNATIONAL SEARCH REPORT
Information on patent family members

International application No.

PCT/CN2015/076133

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