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(54) **Title**: COMPRESSIBLE MICRON-SIZED POLYMER PARTICLE AND USE THEREOF, CURABLE COMPOSITION AND PREPARATION METHOD THEREOF, PRESSURE-SENSITIVE ADHESIVE, AND ADHESIVE TAPE

(57) **Abstract**: The present invention provides a compressible micron-sized polymer particle and use thereof, a curable composition and preparation method thereof, a pressure-sensitive adhesive, and an adhesive tape. The compressible micron-sized polymer particle provided by the present invention comprises a polymer particle having a shell formed by a polymer material and a cavity surrounded by the shell and a metallic layer covering the external surface of the shell. The compressible micron-sized polymer particle provided by the present invention is applied to polyacrylate pressure-sensitive adhesive, capable of enabling the pressure-sensitive adhesive to exhibit sufficient black color without reducing the strength of the pressure-sensitive adhesive.

COMPRESSIBLE MICRON-SIZED POLYMER PARTICLE AND USE THEREOF, CURABLE COMPOSITION AND PREPARATION METHOD THEREOF, PRESSURE-SENSITIVE ADHESIVE, AND ADHESIVE TAPE

Technical Field

The present invention belongs to the technical field of pressure-sensitive adhesives, specifically relating to a compressible micron-sized polymer particle and its use thereof, a curable composition and preparation method thereof, a pressure-sensitive adhesive, and an adhesive tape.

Background

Acrylate adhesive tape, as one type of pressure-sensitive adhesive tapes, has compressibility and adhesion similar to foam tapes (such as polyurethane foam tape); thus, it is also known as “Foam-like Tape”. However, unlike foam tape, acrylate adhesive tape is not composed of a foam core or an adhesive layer on the surface of the core, but exclusively composed of polyacrylate pressure-sensitive adhesive (i.e. the polyacrylate pressure-sensitive adhesive serves as the core and the adhesive layer at the same time), thus effectively protecting the adhesive joint through dissipating stress at the adhesive joint by overall mechanical relaxation of the tape. Besides, the polyacrylate pressure-sensitive adhesive has a strong resistance against heat, ultraviolet and chemicals; thus, the acrylate adhesive tape is excellent in terms of weather-resistance and durability.

The acrylate adhesive tape is suitable for realizing adhesion and sealing of consumer electronics, building, automobile and household appliances, such as the adhering of a display panel of mobile phone to a shell or adhering automobile Logo to a car body. In practical application, the acrylate adhesive tape is expected to be black in many cases to conceal its existence in the background. Polyacrylate pressure-sensitive adhesive is intrinsically colorless; thus, black pigment or black dye is required to be added into polyacrylate pressure-sensitive adhesive to blacken the adhesive. However, acrylate adhesive tape is usually cured with ultraviolet, and the black pigment or black dye is liable to absorb a large amount of ultraviolet, which is liable to affect the curing of the acrylate adhesive tape and reduce the cohesive strength and adhesive strength. Alternatively, the use amount of the black pigment or black dye is required to be decreased for reducing the influence on the curing of the acrylate adhesive tape, which is liable to make the adhesive tape become light (i.e. grey); thus, the appearance is unable to meet the requirement.

Disclosure of the Invention

One purpose of the present invention is to provide a compressible micron-sized polymer particle which can be applied in pressure-sensitive adhesives and enable the pressure-sensitive adhesives to exhibit sufficient black color without reducing the strength of the pressure-sensitive adhesives.

The compressible micron-sized polymer particle provided by the present invention comprises:

a polymer particle having a shell formed by a polymer material and a cavity surrounded by the shell; and

a metallic layer covering the external surface of the shell.

The polymer particle is basically of a spherical shape.

The thickness of the metallic layer ranges from 5 nm to 80 nm, most preferably 20 nm to 60 nm.

Another purpose of the present invention is to provide a curable composition containing the compressible micron-sized polymer particle and capable of forming a pressure-sensitive adhesive after curing.

The curable composition provided by the present invention comprises:

a slurry polymer which comprises a mixture formed of at least two polymerizable monomers by partial copolymerization, wherein the said mixture comprises the said polymerizable monomers unpolymerized and a copolymer formed of the said polymerizable monomers by copolymerization;

a photoinitiator; and

the compressible micron-sized polymer particle.

The polymerizable monomers comprise acrylate monomers serving as first monomers, and non-ester unsaturated monomers serving as second monomers and having at least one olefinic bond; the first monomers are more preferably non-tertiary alcohol (meth)acrylate monomers, and the second monomers are more preferably acid-functional non-ester unsaturated monomers with at least one olefinic bond.

Another purpose of the present invention is to provide a preparation method of the curable composition, which comprises the following steps:

partially copolymerizing the polymerizable monomers to obtain the slurry polymer; and

mixing the photoinitiator and the compressible micron-sized polymer particle with the said slurry polymer.

Another purpose of the present invention is to provide a pressure-sensitive adhesive which can be applied in adhesive tapes and enable the adhesive tapes to exhibit sufficient black color without reducing the strength of the adhesive tapes.

The pressure-sensitive adhesive provided by the present invention is formed of the curable composition through curing.

Another purpose of the present invention is to provide an adhesive tape which can be sufficiently black without reducing the strength.

The adhesive tape provided by the present invention comprises the pressure-sensitive adhesive.

Another purpose of the present invention is to provide a use of the compressible micron-sized polymer particle in polyacrylate pressure-sensitive adhesive.

Brief Description of the Drawings

Fig. 1 is a structural schematic diagram of a vacuum sputtering device for forming a metallic layer on the external surface of an expanded polymer microsphere.

In the present invention, marks in the drawing are as follows:

10. Vacuum Sputtering Device; 12. Housing; 14. Vacuum Cavity; 16. Particle Agitating Fluidizer; 18. Bottom; 20. Interface; 28. Top; 34. Opening; 40. Shaft; 42. Agitating Blade; 44. Hole

Specific Embodiments

To enable technical personnel in the art understand the technical scheme better, the present invention will be further described in detail with the drawing and specific embodiments.

Definition of Terms

In the present invention, the meanings of the following terms or descriptions are as follows:

"A and/or B" refers to any one case or both cases are available, i.e. involving three cases such as "A and B", "A", and "B".

"A to B" refers to value A, value B and any value larger than A but smaller than B. For instance, "1 to 10" refers to 1, 10, and any value larger than 1 but smaller than 10, such as 2, 3, 4, 5, 6, 7, 8, 9, 2.3, 3.5, 5.26, 7.18, 9.999.

"A is basically B" refers to A basically conforms to characteristics of B based on regarding the intrinsic dimensions of A as a reference, but may be slightly different from B in some details. For instance, "A is basically of a spherical shape" means that A is like a sphere in view of the intrinsic dimensions of A, but the said sphere may have no strict requirement on shape, which may have a rough surface, or have fine recesses or bulges on the surface, or have a small deformation overall (i.e. ellipsoidal or pyriform shape).

"Be about A" refers to that A is a target value, but an inevitable and allowable error may appear between an actual value and the target value under existing technical conditions.

"Viscosity": If not specified otherwise, viscosity involved in the present invention is measured by an Ubbelohde viscometer.

"Molecular weight": If not specified otherwise, molecular weight involved in the present invention refers to the weight-average molecular weight, measured by Gel Permeation Chromatography (GPC). A method for measuring the weight-average molecular weight comprises specific steps as follows: taking 0.1g of sample to place into a 5 mL sample bottle, adding 3mL of tetrahydrofuran (TEDIA Co., Ltd., Ohio, U.S.) for dissolution; filtering the solution with a filtration film with the aperture of 0.45 μm and then adding into the sample bottle; testing with a chromatographic instrument (Waters Co., Ltd., Maryland, U.S.); calibrating the chromatographic column with polystyrene having an known molecular weight; and establishing a calibration curve through linear least-squares analysis to obtain the molecular weight.

"Substance use amount": If not specified otherwise, use amount or use amount ratio involved in the present invention refers to the weight or weight ratio.

"(meth)acrylic acid" refers to two cases, i.e. acrylic acid and methacrylic acid.

"Tertiary alcohol" refers to that other three groups connected on carbon atoms of hydroxyl are alcohol of substituent groups of non-hydrogen atoms. "Non-tertiary alcohol" refers to the alcohol not belonging to tertiary alcohol.

"Micron-sized polymer particle" refers to that the particle size of polymer particle is in micron

dimension, i.e. larger than or equal to 1 μm but smaller than 1,000 μm.

"Compressible polymer particle" refers to that the polymer particle can be compressed and deformed without being damaged in case of suffering from external stress, and can recover the original shape and dimensions after the stress is removed.

"Shell formed by a polymer material" refers to an unbroken and enclosed shell formed by a polymer. The shell isolates the internal space from the external space, and the space inside the shell is the "cavity surrounded by the shell".

"Partial copolymerization" refers to that some polymerizable monomers have been polymerized to form a copolymer, while some are still in the form of monomers rather than being polymerized.

"Copolymer" refers to a substance formed of at least two different monomers by copolymerization, comprising random copolymers, block copolymers, grafted copolymers, alternating copolymers, or mixtures thereof.

"Pressure-sensitive adhesive" refers to a substance formed of two bonded matrixes, at least meeting the following requirements at an ambient temperature (5°C to 40°C): (1) long-lasting stickiness; (2) capability of realizing adhesion under finger-press stress; (3) capability of changing the shape to attach to the matrix; and (4) enough cohesive strength for completely removing from the bonded matrix.

"Curing" refers to a process of transforming liquid substances from the liquid to the viscoelastic solid (due to the fact that what formed by curing in the present invention is the pressure-sensitive adhesive) through polymerization and crosslinking of components.

"Adhesive tape" refers to a basically tape-like product adhesive to the matrix or capable of bonding two matrixes.

Compressible Micron-sized Polymer Particle

The present invention provides a compressible micron-sized polymer particle, which comprises: a polymer particle having a shell formed by a polymer material and a cavity surrounded by the shell; and

a metallic layer covering the external surface of the shell.

The compressible micron-sized polymer particle is applicable to pressure-sensitive adhesives, particularly to polyacrylate pressure-sensitive adhesive, enabling the pressure-sensitive adhesive to exhibit sufficient black color without reducing the strength of the pressure-sensitive adhesive.

The compressible micron-sized polymer particle provided by the present invention enables the pressure-sensitive adhesive to be black due to the following possible reasons (not to restrict its mechanism of action): the metallic layer on the surface of the compressible micron-sized polymer particle is excellent in light reflection, capable of reflecting (scattering) most light radiated on the compressible micron-sized polymer particle; besides, the pressure-sensitive adhesive comprises a plurality of compressible micron-sized polymer particles; thus, light radiated onto the pressure-sensitive adhesive is liable to be reflected (scattered) among the particles and gradually absorbed by the pressure-sensitive adhesive, and thus, light is unable to be emitted from the

pressure-sensitive adhesive, and thus enabling the pressure-sensitive adhesive to appear black. The compressible micron-sized polymer particle provided by the present invention is provided with the metallic layer on the surface, so the compressible micron-sized polymer particle basically does not absorb light. Therefore, the pressure-sensitive adhesive is unlikely to be affected by the absorption of ultraviolet light, and capable of appearing sufficiently black at the same time.

Besides, the shell of the polymer particle of the compressible micron-sized polymer particle is formed of a soft polymer material rather than a hard material, so the compressible micron-sized polymer particle provided by the present invention is compressible itself. Therefore, compressible micron-sized polymer particle is likely to improving the compressibility, stickiness and cutting properties (representing whether the particle is liable to be cut and to be sticky to knife during cutting), instead of causing adverse effects on the said properties.

Preferably, the polymer particle is basically of a spherical shape. Thus, the compressible micron-sized polymer particles can be distributed in the pressure-sensitive adhesive more uniformly and stably, and can uniformly reflect (scatter) the light in different directions.

Preferably, the polymer forming the shell of the polymer particle is a thermoplastic polymer, and the cavity is filled with gas. In other words, the polymer particle can be an "expanded polymer microsphere". The expanded polymer microsphere is commercial filler usually added to compositions for reducing the density of compositions. The expanded polymer microsphere is formed through heating and expanding an "expandable microsphere" which comprises a thermoplastic polymer sphere wall and gas inside the sphere wall. The internal gas is liable to expand during heating, thus expanding the sphere wall to different dimensions to form the expanded polymer microsphere. Specific dimensions of the expanded polymer microsphere are determined upon the heating temperature and the heating time, etc.

More preferably, the thermoplastic polymer forming the shell of the polymer particle is thermoplastic polyacrylic resin, and the gas is alkane (such as methane and ethane). On one hand, the preceding material is the most common material for the expanded polymer microsphere in the market; on the other hand, the compressible micron-sized polymer particle provided by the present invention is preferably applied to polyacrylate pressure-sensitive adhesive. Therefore, if the shell of the polymer particle is formed of acrylates, the material of the shell will be compatible well with the pressure-sensitive adhesive in case the metallic layer on the shell is incomplete.

To guarantee the reflection, compressibility and dispersibility, the particle size (i.e. outer diameter) of the preceding polymer particle is preferably 10 μm to 100 μm, and the thickness (i.e. wall thickness) of the shell is preferably 2 μm to 15 μm.

The polymer particle is also provided with the metallic layer on the external surface of the shell, which means that the said expanded polymer particle is "metalized".

Preferably, the metallic layer is made of any or the alloy of tungsten, titanium, aluminum, chromium, niobium and zirconium. The preceding metal is low in cost, capable of enabling the pressure-sensitive adhesive to have adequate corrosion resistance and become sufficiently black.

The thickness of the metallic layer is liable to obviously affect the reflection and compressibility of the compressible micron-sized polymer particle. According to researches, in case the polymer particle is used as the matrix, the metallic layer is preferably 5 nm to 80 nm thick, more preferably 6 nm to 70 nm thick, even more preferably 12 nm to 60 nm thick, most preferably 20 nm to 60 nm thick. The reason is that the metallic layer within the said thickness range has enough reflection performance without causing adverse effects on the compressibility.

The metallic layer is formed on the external surface of the polymer particle preferably by vacuum sputtering.

Generally, the methods for forming the metallic layer include vacuum sputtering, chemical vapor deposition, electroplating and electroless plating. The material for forming the substrate of the metallic layer in the present invention is the particulate polymer material, so some methods are unsuitable, and the vacuum sputtering is preferably applicable.

For specific vacuum sputtering, the following American patents US8664148, US8518854, US7989384, US7727931, US7458693, US7156528 and US6767745 authorized to 3M Co., Ltd can be referenced to. The vacuum sputtering can be conducted in a vacuum sputtering device 10 as shown in Fig. 1. The vacuum sputtering device 10 comprises a housing 12 and a vacuum cavity 14 in the housing 12. The vacuum cavity 14 is internally provided with a particle agitating fluidizer 16 for containing particles; the particle agitating fluidizer is internally provided with agitating blades 42 with holes 44; and the agitating blades 42 are connected to a rotatable shaft 40. The particle agitating fluidizer 16 is provided with an opening 34 at the top, the housing 12 is provided with a round opening at the top 28 opposite to the opening 34; and a direct-current magnetron sputtering cathode and a metallic sputtering target (unmarked in the drawing) are arranged at the round opening. The housing 12 is also provided with an interface 20 at the bottom 18 to connect with a vacuum pump. During sputtering, particulate materials (such as the expanded polymer microspheres) are placed in the particle agitating fluidizer 16, sputtered particles deposit on the surface of the materials through the opening 34, and the materials in the particle agitating fluidizer 16 are overturned continuously due to rotation of the agitating blades 42, such that the particles accepting the sputtered particles change continuously, and the surface of each particle is uniformly covered with the metallic layer.

Curable Composition

The present invention also provides a curable composition, which can form pressure-sensitive adhesives or adhesive tapes after ultraviolet curing.

1. Components of the Curable Composition

The curable composition provided by the present invention comprises:

a slurry polymer;

the preceding compressible micron-sized polymer particle; and

a photoinitiator.

Preferably, the curable composition provided by the present invention may also comprise any one or more of the following components:

a crosslinking agent;

hydrophobic fumed silica;

an expanded polymer microsphere (excluding the metallic layer);

a tackifying resin;

a glass bead;

a chain transfer agent; and

other additives (i.e. plasticizer, dye, antioxidant, dispersant, suspending agent and ultraviolet stabilizer) and functional components.

The function, specific substance available and content of each component will be introduced one by one in the following paragraphs.

1) Slurry Polymer

The slurry polymer comprises a mixture formed of at least two polymerizable monomers by partial copolymerization, wherein the said mixture comprises the said polymerizable monomers unpolymerized and a copolymer formed of the said polymerizable monomers by copolymerization. In other words, the slurry polymer comprises at least two polymerizable monomers, some of which have been polymerized to form the copolymer, while some are still in the form of monomers.

The polymerizable monomers in the slurry polymer are partially polymerized, so no polymerizable monomers are subjected to crosslinking or only a few polymerizable monomers are subjected to crosslinking, and generated copolymers are still dissoluble in the unpolymerized polymerizable monomers; therefore, the slurry polymer still belongs to liquid homogeneous system overall, which is flowable and can be coated. Besides, the slurry polymer is partially polymerized, so the slurry polymer has a high viscosity, liable to form a product (such as the adhesive tape) having a certain thickness and shape.

The viscosity of the slurry polymer at 22°C is preferably 500 cPS to 10,000 cPs, more preferably 2,000 cPs to 6,000 cPs. Within the preceding viscosity range, the compressible micron-sized polymer particle is likely to be mixed, convenient to be coated and capable of forming the adhesive tape, meeting a thickness requirement (such as 5 mm).

The weight-average molecular weight of the copolymer in the slurry polymer is preferably 500,000 D (Daltons, g/mol) to 10,000,000 D. In other words, the weight-average molecular weight of the polymerized substance (i.e. the copolymer excluding the unpolymerized monomers) in the slurry polymer is 500,000 D to 10,000,000 D, more preferably 750,000 D to 6,000,000 D, more preferably 1,000,000 D to 5,000,000 D.

Besides, in the slurry polymer, the polymerized polymerizable monomers account for 1% to 30% of the total weight of the polymerized and unpolymerized polymerizable monomers. In other words, when the overall polymerizable monomers (i.e. the polymerized and unpolymerized monomers) in the slurry polymer are 100 parts by weight, the proportion of the polymerizable monomers polymerized to form the copolymer is 1-30 parts by weight, or the conversion ratio of the polymerizable monomers is 1-30%. The conversion ratio of the polymerizable monomers is more preferably 2-20%, even more

preferably 5-15%, most preferably 7-12%.

Based on the preceding molecular weight and conversion ratio, the slurry polymer with appropriate viscosity and strength overall is likely to be mixed with the compressible micron-sized polymer particle.

The polymerizable monomers comprise acrylate monomers serving as first monomers; and non-ester unsaturated monomers serving as second monomers and having at least one olefinic bond. The polymerizable monomers optionally comprise nonacid-functional olefinically unsaturated polar monomers and/or vinyl monomers. Each polymerizable monomer will be further introduced in the following paragraphs.

(1) Acrylate Monomer

The acrylate monomer is more preferably non-tertiary alcohol (meth)acrylate monomer, i.e. ester formed of (meth)acrylic acid and non-tertiary alcohol.

The non-tertiary alcohol for forming the non-tertiary alcohol (meth)acrylate monomer preferably contains 1-20 carbon atoms, more preferably 2-18 carbon atoms, even more preferably 4-12 carbon atoms; the chain segment of the non-tertiary alcohol may be straight chain, branched chain or a combination of the two. Specifically, the non-tertiary alcohol includes but not limited to any one or more of methanol, ethanol, 1-propanol, 2-propanol, 1-butanol, 2-butanol, 1-pentanol, 2-pentanol, 3-pentanol, 2-methyl-1-butanol, 3-methyl-1-butanol, 1-hexanol, 2-hexanol, 2-methyl-1-pentanol, 3-methyl-1-pentanol, 2-ethyl-1-butanol, 3,5,5-trimethyl-1-hexanol, 3-heptanol, 1-octanol, 2-octanol, isooctanol, 2-ethyl-1-hexanol, 1-decanol, 2-propyl heptanol, 1-dodecanol, 1-tridecanol and 1-tetradecanol.

The preceding non-tertiary alcohols are applicable. However, in some preferable embodiments, the non-tertiary alcohol preferably is one or more of butanol, isooctanol and 2-ethyl hexanol (corresponding esters: isooctyl acrylate, 2-ethylhexyl acrylate and butyl acrylate); in some other preferable embodiments, the non-tertiary alcohol is derived from renewable alcohol, such as any one or more of 2-octanol, citronellol and 3,7-dimethyl-1-octanol.

For per 100 parts by weight of the overall polymerizable monomers (i.e. all the monomers for forming the slurry polymer, including the polymerized and unpolymerized monomers), the non-tertiary alcohol (meth)acrylate monomers preferably account for 80-99.5 parts, more preferably accounting for 85-99.5 parts, even more preferably accounting for 87-98 parts.

Among the non-tertiary alcohol (meth)acrylate monomers, some may have a high glass transition temperature (Tg), which means that the Tg of homopolymers thereof is at least 25°C, preferably at least 50°C. The non-tertiary alcohol (meth)acrylate monomer with high Tg includes but not limited to any one or more of methyl methacrylate, ethyl methacrylate, isopropyl methacrylate, n-butyl methacrylate, isobutyl methacrylate, sec-butyl methacrylate, octadecyl methacrylate, phenyl methacrylate, cyclohexyl methacrylate, isobornyl acrylate, isobornyl methacrylate, phenylmethyl methacrylate, 3,3,5-trimethylcyclohexyl acrylate, cyclohexyl acrylate, N-octyl acrylamide and propyl methacrylate.

For per 100 parts by weight of the overall polymerizable monomers (i.e. all the monomers for forming the slurry polymer, including the polymerized and unpolymerized monomers), the non-tertiary alcohol (meth)acrylate monomers with high Tg preferably account for 0-40 parts (0 representing exclusion), more preferably accounting for 1-35 parts, even more preferably accounting for 5-35 parts. The content of the non-tertiary alcohol (meth)acrylate monomers with high Tg is included in the content of the non-tertiary alcohol (meth)acrylate monomers. For instance, the content of the non-tertiary alcohol (meth)acrylate monomers with high Tg is 35 parts in case the content of the non-tertiary alcohol (meth)acrylate monomers is 85 parts. In other words, the non-tertiary alcohol (meth)acrylate monomers with the total content of 85 parts comprise 35 parts of the non-tertiary alcohol (meth)acrylate monomers with high Tg and 50 parts of other non-tertiary alcohol (meth)acrylate monomers with low Tg.

(2) Non-ester Unsaturated Monomer with At Least One Olefinic Bond

The non-ester unsaturated monomer with at least one olefinic bond is more preferably acid-functional non-ester unsaturated monomer with at least one olefinic bond, i.e. comprising olefinic bond and acid functional group which can be acid (like carboxylic acid) or salt of acid (like alkali metal salt of carboxylic acid) but ester. The acid-functional non-ester unsaturated monomer with at least one olefinic bond may be olefinically unsaturated carboxylic acid, olefinically unsaturated sulfonic acid, or olefinically unsaturated phosphonic acid. Specifically, the acid-functional non-ester unsaturated monomer with at least one olefinic bond includes but not limited to any one or more of acrylic acid, methacrylic acid, itaconic acid, fumaric acid, crotonic acid, citraconic acid, maleic acid, oleic acid, styrene sulfonic acid, 2-acrylamido-2-methylpropane sulfonic acid and vinyl phosphonic acid. In view of the feasibility, the acid-functional non-ester unsaturated monomer with at least one olefinic bond is more preferably olefinically unsaturated carboxylic acid (such as acrylic acid or methacrylic acid).

For per 100 parts by weight of the overall polymerizable monomers (i.e. all the monomers for forming the slurry polymer, including the polymerized and unpolymerized monomers), the acid-functional non-ester unsaturated monomers with at least one olefinic bond preferably account for 0.5-20 parts, more preferably accounting for 0.5-15 parts, even more preferably accounting for 2-12.5 parts.

(3) Nonacid-functional Olefinically Unsaturated Polar Monomer

Preferably, the slurry polymer may also comprise the nonacid-functional olefinically unsaturated polar monomers (possibly including ester). The nonacid-functional olefinically unsaturated polar monomer includes but not limited to any one or more of 2-hydroxyethyl methacrylate, N-vinylpyrrolidone, N-vinyl caprolactam, acrylamide, mono-N-alkyl-substituted acrylamide, di-N-alkyl-substituted acrylamide, tert-butyl acrylamide, dimethyl aminoethyl acrylamide, N-octyl acrylamide and poly(alkoxy alkyl)(meth)acrylate. The poly(alkoxy alkyl)(meth)acrylate comprises any one or more of 2-(2-ethoxyethoxy) ethyl (meth)acrylate, 2-ethoxy ethyl (meth)acrylate, 2-methoxyethoxy ethyl (meth)acrylate, 2-methoxyethyl methacrylate and polyglycol

mono(meth)acrylate. Most preferably, the nonacid-functional olefinically unsaturated polar monomer is 2-hydroxyethyl methacrylate and/or N-vinylpyrrolidone.

For per 100 parts by weight of the overall polymerizable monomers (i.e. all the monomers for forming the slurry polymer, including the polymerized and unpolymerized monomers), the nonacid-functional olefinically unsaturated polar monomers preferably account for 0-10 parts (0 representing exclusion), more preferably accounting for 0.5-8 parts, even more preferably accounting for 1-6 parts.

(4) Vinyl Monomer

Preferably, the slurry polymer may also comprise the vinyl monomers (i.e. monomers regarding vinyl groups as an essential element) involving but not limited to any one or more of vinyl ester (i.e. vinyl acetate and vinyl propionate), styrene, substituted styrene (i.e. α-methyl styrene) and vinyl halide. Obviously, the said vinyl monomer does not involve other monomers described in the preceding paragraphs.

For per 100 parts by weight of the overall polymerizable monomers (i.e. all the monomers for forming the slurry polymer, including the polymerized and unpolymerized monomers), the vinyl monomers preferably account for 0-5 parts (0 representing exclusion), more preferably accounting for 0.5-5 parts, even more preferably accounting for 1-4 parts.

2) Compressible Micron-sized Polymer Particle

After being added into the curable composition, the preceding compressible micron-sized polymer particle can enable the pressure-sensitive adhesive to exhibit sufficient black color without reducing the strength of the pressure-sensitive adhesive formed of the said curable composition.

The compressible micron-sized polymer particles account for 0.1% to 7% of the total weight of polymerized and unpolymerized polymerizable monomers. In other words, for per 100 parts of the overall polymerizable monomers (i.e. all the monomers for forming the slurry polymer, including the polymerized and unpolymerized monomers) in the curable composition, the use amount of the compressible micron-sized polymer particles is preferably 0.1-7 parts, more preferably 1-7 parts, even more preferably 1-6 parts. Obviously, the component described herein and hereinafter is not part of the slurry polymer, so the use amount of the said component is the added amount relative to the slurry polymer.

Obviously, insufficient use amount of the compressible micron-sized polymer particles cannot be effective; however, researches have proven that excessive use amount is liable to affect the curing of the curable composition. Therefore, the use amount of the compressible micron-sized polymer particles shall be preferably within the said range.

3) Photoinitiator

The photoinitiator is applied to generating free radicals under ultraviolet illumination, thus enabling the curable composition to form the pressure-sensitive adhesive. The photoinitiator available includes but not limited to any one or more of benzoin ether (such as benzoin methyl ether, benzoin isopropyl ether), substituted acetophenon (such as 2,2-dimethoxyacetophenon and dimethoxy

hydroxyacetophenone), substituted α-ketol (such as 2-hydroxy-2-methylpropiophenone), aromatic sulfonyl chloride (such as 2-naphthalenesulfonyl chloride, photosensitive oxime (such as 1-phenyl-1,2-propanedione-2-(o-ethoxy-carbonyl)-oxime), 1-hydroxycyclohexyl phenyl ketone, 1-[4-(2-hydroxyethoxy)phenyl]-2-hydroxy-2-methyl-1-propan-1-one, (4-methylthio benzoyl)-1-methyl-1-morpholino ethane, (4-morpholino benzoyl)-1-benzyl-1-dimethylaminopropylamine, (4-morpholino benzoyl)-1-(4-methylbenzyl)-1-dimethylaminopropylamine, bis (2,4,6-trimethyl benzoyl) phenylphosphine oxide, and 1-hydroxycyclohexyl benzophenone. Particularly preferably, the photoinitiator is substituted acetophenone.

For per 100 parts of the overall polymerizable monomers (i.e. all the monomers for forming the slurry polymer, including the polymerized and unpolymerized monomers) in the curable composition, the use amount of the photoinitiator is 0.001-3 parts, preferably 0.005-1 parts, more preferably 0.01-0.5 parts.

What should be noted is that the use amount of the photoinitiator refers to the total content in the curable composition. However, the photoinitiator may not be added at one time. A part of the photoinitiator may be added to the polymerizable monomers to partially copolymerize the polymerizable monomers to form the slurry polymer, and the left photoinitiator may be added to the slurry polymer to form the pressure-sensitive adhesive. Generally, the photoinitiator added to the polymerizable monomers in advance accounts for about 1/10 to 1/5 of the total content.

4) Crosslinking Agent

Preferably, the curable composition may also comprise the crosslinking agent which can improve the cohesive strength of the prepared pressure-sensitive adhesive. The crosslinking agent applied in the present invention is a photosensitive crosslinking agent which can be activated by ultraviolet illumination. General photosensitive crosslinking agent includes but not limited to benzophenone, polymerizable aromatic ketone and triazine such as 2,4-bis (trichloromethyl)-6-(4-metoxybenzene)-triazine. Another crosslinking agent suitable for the present invention is multifunctional (meth)acrylate such as di(meth)acrylate, tri(meth)acrylate and tetra(meth)acrylate. The specific crosslinking agent includes but not limited to any one or more of 1,6-hexanediol dimethacrylate, poly(glycol) di (meth)acrylate, polybutadiene di(meth)acrylate, polyurethane di(meth)acrylate, propoxylated glyceryl tri(meth) acrylate and pentaerythritol tri(meth)acrylate. Another crosslinking agent suitable for the present invention is thermally activated crosslinking agent such as multifunctional aziridine, isocyanate and epoxy resin, specifically 1,1'-isophthaloyl bis(2-methylaziridine). The crosslinking agent suitable for the present invention also comprises aziridine crosslinking agent.

Most preferably, the crosslinking agent for the present invention is multifunctional (meth)acrylate and/or triazine.

For per 100 parts of the overall polymerizable monomers (i.e. all the monomers for forming the slurry polymer, including the polymerized and unpolymerized monomers) in the curable composition,

the use amount of the crosslinking agent is preferably less than or equal to 5 parts, more preferably 0.01-2 parts, even more preferably 0.03-1 parts.

5) Hydrophobic Fumed Silica

Preferably, the hydrophobic fumed silica can be added to the curable composition to improve the cohesive strength (such as high-temperature shearing performance) and stickiness of the pressure-sensitive adhesive. The hydrophobic fumed silica refers to silicon dioxide, especially porous material, which is formed by a vapor-phase process and also known as vapor-phase white carbon black.

In other words, for per 100 parts of the overall polymerizable monomers (i.e. all the monomers for forming the slurry polymer, including the polymerized and unpolymerized monomers) in the curable composition, the use amount of the hydrophobic fumed silica is preferably 0.1-15 parts, more preferably 0.5-15 parts, even more preferably 1.5-10 parts.

6) Expanded Polymer Microsphere (Excluding the Metallic Layer)

Preferably, the curable composition may also comprise the expanded polymer microspheres without the sputtered metallic layer. The expanded polymer microspheres can improve the compressibility of the adhesive tape.

For per 100 parts of the overall polymerizable monomers (i.e. all the monomers for forming the slurry polymer, including the polymerized and unpolymerized monomers) in the curable composition, the use amount of the expanded polymer microspheres is preferably less than or equal to 10 parts, more preferably less than or equal to 8 parts, even more preferably less than or equal to 6 parts.

7) Tackifying Resin

Preferably, the curable composition may also comprise the tackifying resin for improving the adhesive strength of the pressure-sensitive adhesive. Ordinary tackifying resin includes rosin resin, polyterpene phenolic resin, petroleum resin and aromatic resin. Obviously, the tackifying resin is applied provided that the curing of the curable composition and the performance of the final adhesive tape are not affected. However, during ultraviolet curing process, most tackifying resins are liable to seriously obstruct the progress of curing reaction, such as resulting in phase splitting between the pressure-sensitive adhesive and the tackifying resin, or reducing the molecular weight of the polymer to weaken the cohesive strength of the adhesive, or resulting in incomplete curing. Available tackifying resin for the present invention includes hydrogenated rosin resin Foral 85 (Pinova Co., Ltd., Georgia, U.S.), and hydrogenated polyterpene phenolic resin UH115 (Yasuhara Chemical Co., Ltd., Hiroshima, Japan).

For per 100 parts of the overall polymerizable monomers (i.e. all the monomers for forming the slurry polymer, including the polymerized and unpolymerized monomers) in the curable composition, the use amount of the tackifying resin is preferably less than or equal to 40 parts, more preferably less than or equal to 30 parts, even more preferably less than or equal to 25 parts.

8) Glass Bead

Preferably, the glass beads with appropriate particle size can be added to the curable composition

to enhance the foam property of the adhesive tape. The particle size of the glass beads is preferably 5 μm to 200 μm.

For per 100 parts of the overall polymerizable monomers (i.e. all the monomers for forming the slurry polymer, including the polymerized and unpolymerized monomers) in the curable composition, the use amount of the glass beads is preferably less than or equal to 10 parts, more preferably less than or equal to 9 parts, even more preferably less than or equal to 8 parts.

9) Chain Transfer Agent

Preferably, the chain transfer agent can be added to the curable composition to control the molecular weight of the copolymer in the pressure-sensitive adhesive. The chain transfer agent for the present invention includes but not limited to any one or more of carbon tetrabromide, alcohol and thiol. Most preferably, the chain transfer agent is isooctyl thioglycolate and/or carbon tetrabromide.

For per 100 parts of the overall polymerizable monomers (i.e. all the monomers for forming the slurry polymer, including the polymerized and unpolymerized monomers) in the curable composition, the use amount of the chain transfer agent is preferably 0.01-0.5 parts, more preferably 0.05-0.2 parts.

10) Other Additives (i.e. Plasticizer, Dye, Antioxidant, Dispersant, Suspending Agent and Ultraviolet Stabilizer) and Functional Components

Preferably, to improve different properties of the adhesive tape, other known additives including but not limited to any one or more of plasticizer, dye, antioxidant, dispersant, suspending agent and ultraviolet stabilizer can be added to the curable composition.

Even more preferably, to realize specific functions of the adhesive tape, functional components such as specific electricity-conductive, magnetic and heat-conductive particles, staple fibers and sheets can be added to the curable composition. For instance, Chinese patent CN103320037A discloses adding electricity-conductive particles to obtain foam-shaped conductive adhesive tape.

Of course, each optional component described in the preceding paragraphs shall be added based on not affecting the intrinsic performance of the adhesive tape. In addition, the technical personnel in the art may also add other known components to the curable composition if necessary.

2. Preparation Method of the Curable Composition

The preparation method of the curable composition comprises the following steps:

partially copolymerizing at least two of the said polymerizable monomers to obtain the slurry polymer; and

mixing the photoinitiator and the compressible micron-sized polymer particle with the said slurry polymer.

In other words, the curable composition is obtained through mixing the slurry polymer with other components (the compressible micron-sized polymer particle, the photoinitiator and so on) according to a conventional process. Therefore, the slurry polymer shall be prepared in advance for preparing the curable composition. The slurry polymer is formed through partial copolymerization of polymerizable monomers, specifically prepared by two methods, i.e. ultraviolet-initiated mass polymerization (ultraviolet initiation method for short) and heat-initiated solution polymerization

(solution method for short). The two methods will be introduced in sequence in the following paragraphs.

1) Ultraviolet Initiation Method

The ultraviolet initiation method does not employ any additional solvent, just directly polymerizing the polymerizable monomers (mass polymerization) and using unpolymerized polymerizable monomers as the solvent to dissolve the copolymer generated by polymerization, thus forming the slurry polymer.

The ultraviolet initiation method comprises specific steps of:

(1) Mixing the polymerizable monomers with the photoinitiator

In this step, the non-tertiary alcohol (meth)acrylate monomer with high Tg (if any) can be completely added or partially added, or added after other polymerizable monomers are partially copolymerized. Since the copolymerization of the non-tertiary alcohol (meth)acrylate monomer with high Tg is usually slow, the non-tertiary alcohol (meth)acrylate monomer with high Tg added in this step is usually unable to be polymerized on a large scale.

The photoinitiator added in this step is only a portion of the photoinitiator in the final curable composition, only accounting for 1/10 to 1/5 of the total content.

(2) Irradiating the mixture of the polymerizable monomers with ultraviolet, thus partially copolymerizing the polymerizable monomers to form the slurry polymer

Available ultraviolet light source includes low-intensity light source and high-intensity light source. For the low-intensity light source such as black light, the wavelength ranges from 280 nm to 400 nm, and the intensity is usually below 10 mw/cm2; for the high-intensity light source such as medium mercury lamp, the intensity is usually higher than 10 mw/cm2, more preferably ranging from 15 mw/cm2 to 450 mw/cm2. The intensity is measured with UVIMAPTM UM 365 L-S radiometer (General Electronic Instrumentation & Technology Inc., Virginia, U.S.) as per specifications of the United States National Institute of Standards and Technology (NIST). In this step, the intensity of ultraviolet is preferably 0.1 mw/cm2 to 150 mw/cm2, more preferably 0.5 mw/cm2 to 100 mw/cm2, even more preferably 0.5 mw/cm2 to 50 mw/cm2. The time of ultraviolet irradiation can be regulated according to the light intensity and the polymerization, which is usually about several minutes.

During polymerization, the viscosity of the slurry polymer can be measured constantly with the Ubbelohde viscometer, and the refractive index (i.e. the proportion of polymerized polymerizable monomers to the overall polymerizable monomers) of the slurry polymer shall be measured constantly to judge the monomer conversion ratio. The ultraviolet can be removed after the index meets the predetermined standard, and then the slurry polymer shall be filled with air or oxygen to quench the free radicals to terminate the polymerization.

The ultraviolet initiation method does not employ any additional organic solvent (i.e. the solvent except for the polymerizable monomers) during preparation, so the whole production process is basically free of wastewater and exhaust emission, friendlier to the environment and more efficient. Besides, the ultraviolet initiation method employing the ultraviolet for initiating the polymerization is

advantageous no heating, simple and easy operation, low energy consumption and high efficiency, capable of terminating the reaction instantly after turning off the ultraviolet light source and filling the air. Therefore, the viscosity, monomer conversion ratio and weight-average molecular weight of the product can be controlled precisely.

Therefore, the ultraviolet initiation method is a preferable method for preparing the slurry polymer in the present invention. In each embodiment of the present invention, the slurry polymer is prepared by the ultraviolet initiation method.

2) Solution Method

The slurry polymer may also be prepared by the solution method, i.e. dissolving the polymerizable monomers in other organic solvents for polymerization and then removing the said organic solvents after the polymerization to obtain the residual which is the slurry polymer.

The solution method comprises specific steps of:

(1) Dissolving the polymerizable monomers and a thermal initiator into the solvent, and filling nitrogen for full purification

Available solvent includes but not limited to any one or more of methanol, tetrahydrofuran, ethanol, isopropanol, acetone, methyl ethyl ketone, methyl acetate, ethyl acetate, toluene, dimethylbenzene and ethylene glycol alkyl ether.

For the solution method, the thermal initiator such as organic peroxide, organic hydroperoxide and azo compound, which are capable of generating the free radicals, is required. Available organic peroxide includes but not limited to any one or more of benzoyl peroxide, lauroyl peroxide, di-tert-amyl peroxide, tert-butyl peroxybenzoate, 2,5-dimethyl-2,5-di(tert-butylperoxy)hexane, 2,5-dimethyl-2,5-di(tert-butylperoxy)hexyne-3 and dicumyl peroxide. Available organic hydroperoxide includes but not limited to tert-cyclopentadienyl hydroperoxide and/or tert-butyl hydroperoxide. Available azo compound includes but not limited to any one or more of 2,2'-azobis (isobutyronitrile), 2,2'-azobis(2-methylbutyronitrile) and 2,2'-azobis(2,4-dimethylvaleronitrile).

For per 100 parts of the overall polymerizable monomers (i.e. all the monomers for forming the slurry polymer, including the polymerized and unpolymerized monomers), the use amount of the thermal initiator is preferably 0.001-3 parts, more preferably 0.005-1 parts, even more preferably 0.1-0.5 parts.

(2) Heating the solution to partially polymerize the polymerizable monomers

The heating temperature usually ranges from 40°C to 100°C, and the heating time usually ranges from 1h to 20h, both of which are determined upon the total substance use amount and the needed monomer conversion ratio.

(3) Removing the additional solvent through vacuum distillation to obtain the slurry polymer

Notes: the solution method for preparing the slurry polymer is feasible, but the method is not the best one due to high cost and intricate process because of employing the additional solvent and requiring a step of removing the solvent, high energy consumption because of heating, and long time

(usually several hours in comparison with the ultraviolet initiation method only taking several minutes) and low efficiency because of slower polymerization of monomers in the solution.

The curable composition can be obtained through uniformly mixing the formed slurry polymer with other components of the curable composition in conventional manner (such as agitation). Therefore, it is no need to discuss here.

Pressure-sensitive Adhesive and Adhesive Tape

The embodiment of the present invention also provides a pressure-sensitive adhesive formed of the said curable composition through curing.

The curable composition can be fully cured with ultraviolet irradiation. The curing preferably employs the low-intensity ultraviolet light source, and the ultraviolet irradiation time is determined upon fully curing the curable composition, which is usually several minutes.

The embodiment of the present invention also provides an adhesive tape, which comprises the said pressure-sensitive adhesive.

As one embodiment of the present invention, the adhesive tape can be completely formed of the said pressure-sensitive adhesive, which means that the pressure-sensitive adhesive is used as a core of the adhesive tape as well as an adhesive layer.

Alternatively, as one embodiment of the present invention, the adhesive tape may also use the said pressure-sensitive adhesive as the core and employ other adhesive layers on one or two sides of the core to realize more functions.

Available material for the adhesive layer includes ordinary acrylate adhesive, rubber adhesive and organic silicon adhesive, and the thickness of the adhesive layer ranges from 0.001mm to 0.5mm. The components and thickness of the adhesive layers on two sides of the pressure-sensitive adhesive core may be the same or different.

Besides, to improve the bonding strength between the core and the adhesive layer, the core and/or the adhesive layer can be subjected to conventional pretreatment such as corona treatment, plasma treatment and primer coating treatment before the adhesive layer is applied.

The adhesive tape can be adhered with a release film or release paper on one or two sides for practical application. Available release film or release paper includes but not limited to organosilicone-coated kraft paper, glassine paper, PE-coated kraft paper and poly(ethylene terephthalate).

During preparation of the adhesive tape, the curable composition can be coated on the release film or release paper to form specific shape with specific thickness, and then the curable composition can be cured to form the pressure-sensitive adhesive.

Available method for coating the curable composition includes but not limited to roll coating, flow coating, dip coating, spin coating, spray coating, blade coating and mold coating, chosen according to the coating thickness. Generally, the pressure-sensitive adhesive (dry adhesive) in the adhesive tape is preferably 0.05 mm to 5 mm thick, more preferably 0.1 mm to 2.5 mm thick, even more preferably 0.1 mm to 2 mm thick. With the compressible micron-sized polymer particle, the

adhesive tape provided by the present invention is easy to be prepared and formed, and can meet the requirement on strength and compressibility even if the tape is thick or thin. Therefore, the adhesive tape (accurately the pressure-sensitive adhesive therein) provided by the present invention has a larger allowable thickness range which is beyond the thickness range of existing adhesive tape, with the minimum thickness of 0.05 mm and the maximum thickness of 5mm.

The adhesive tape provided by the present invention comprises the compressible micron-sized polymer monomers, so the adhesive tape can be sufficiently black without affecting the strength thereof. Thus, in a color system represented by L-value, a-value and b-value (i.e. L*a*b), the L-value of the tape color is preferably 0-30, more preferably 0-25, even more preferably 0-20. The color system represented by L-value, a-value and b-value is also called Lab color space, which was developed in 1976 by the International Commission on Illumination (CIE for short in French). L-value, a-value and b-value are three coordinate axes perpendicular to each other, representing one color space. L represents the brightness, with 0 representing the most black, 100 representing the most white. Positive a represents magenta, and negative a represents green. Positive b represents yellow, and negative b represents blue. Generally, a-value as well as b-value of the adhesive tape provided by the present invention ranges from -5 to 5, so a-value and b-value are generally inconsiderable, and L-value is required to be considered to confirm whether the blackness meets the standard or not.

Embodiments

According to the preceding description, different formulations and parameters are employed to prepare different adhesive tapes as embodiments and comparative examples to exemplarily illustrate the present invention.

1. Material List

The materials employed in each embodiment are illustrated in the following table.

Table 1. List of Materials Employed in Each Embodiment

Description	Category	Component and Relevant Parameter	Origin
IOA	Non-tertiary alcohol (meth)acrylate monomer	Isooctyl acrylate	3M Co., Ltd., Minnesota, U.S.
2-EHA	Non-tertiary alcohol (meth)acrylate monomer	2-ethylhexyl acrylate	Huayi Acrylic Acid Co., Ltd., Shanghai, China
BA	Non-tertiary alcohol (meth)acrylate monomer	Butyl acrylate	Huayi Acrylic Acid Co., Ltd., Shanghai, China
IBxA	Non-tertiary alcohol (meth)acrylate monomer with high Tg	Isobornyl acrylate	San Ester Co., Ltd., Osaka, Japan
AA	Acid-functional non-ester unsaturated monomers with at least one olefinic bond	Acrylic acid	Huayi Acrylic Acid Co., Ltd., Shanghai, China
N,N-DMAA	Nonacid-functional olefinically unsaturated polar monomer	N,N-dimethyl acrylamide	Boruilong Petroleum Science & Technology Development Co., Ltd., Beijing, China
PETA	Crosslinking agent	Pentaerythritol triacrylate	Sartomer Co., Inc., Pennsylvania, U.S.
HDDA	Crosslinking agent	1,6-hexanediol diacrylate	Cytec, New Jersey, U.S.
Triazine	Crosslinking agent	2,4-bis(trichloro methyl)-6-(4-met oxybenzene)-triaz ine	3M Co., Ltd., Minnesota, U.S.
Irgacure 651	Photoinitiator	2,2 -dimethoxy-2-phe nyl-1-acetopheno ne	BASF Corporation, New Jersey, U.S.
Foral 85	Tackifying resin	Hydrogenated rosin resin	Pinova Co., Ltd., Georgia, U.S.
UH115	Tackifying resin	Hydrogenated polyterpene phenolic resin	Yasuhara Chemical Co., Ltd., Hiroshima, Japan
FN-80SDE	Expanded polymer microsphere	Average particle size of 40 μm	Matsumoto Yushi-Seiyaku Co., Ltd., Osaka, Japan
461DE20d70	Expanded polymer microsphere	Average particle size of 20 μm	AkzoNobel Pulp and Performance Chemicals Co., Ltd., Sundsvall, Sweden

461DET80d 25	Expanded polymer microsphere	Average particle size of 75 μm	AkzoNobel Pulp and Performance Chemicals Co., Ltd., Sundsvall, Sweden
Aerosil R972	Hydrophobic fumed silica.	Fumed silica	EVONIK, Essen, Germany
K15	Glass bead	Average particle size of 60 μm	3M Co., Ltd., Minnesota, U.S.
Penncolor 9B117	Black dye		Penn Color Inc., Pennsylvania, U.S.

2. Test Method

The adhesive tape prepared in each comparative example and embodiment is tested as follows:

1) Test of 90° Stripping Adhesive Strength

Carefully cut the adhesive tape with a knife along the glue coating direction to obtain a sample of about 25.4 mm wide and 115 mm long, remove the release film from one side, center the sample and adhere to a stainless steel plate (Chemsultants International Inc., Ohio, U.S.), align the upper end of the adhesive tape with the edge of the steel plate, wipe the stainless steel plate with isopropanol (Sinopharm Chemical Reagent Co., Ltd., Shanghai, China) for three times before the adhesion step, and ensure the isopropanol to be volatilized completely.

Peel off the release film from the other side, and then adhere a 0.13 mm-thick anodic alumina foil (Lawerence and Fredrick, Illinois, U.S.), and attach the oxidized side of the anodic alumina foil to the glue surface.

Conduct rolling with a 6.8 kg rubber roller for a round at a speed of 305 mm/min, lay aside for a certain time and then employ Instron 3343 tester (Chemsultants International Inc., Ohio, U.S.) to measure the force for stripping the adhesive tape from the stainless plate along the 90° direction (i.e. the direction perpendicular to the adhesion surface of the adhesive tape) at a stripping speed of 305 mm/min, and take the average of measured results of three samples of each adhesive tape.

The storage conditions are as follows:

First condition: Lay aside for 20 min at 23±2°C with the relative humidity of 50±5%.

Second condition: Lay aside for 30 days at 23±2°C with the relative humidity of 50±5%.

Third condition: For the adhesive tape with release film (i.e. un-adhered adhesive tape), lay aside for 7 days at 65°C with the relative humidity of 95% firstly to test the anti-aging performance of the adhesive tape at high temperature with high humidity, and then lay aside for 1 day at 23±2°C with the relative humidity of 50±5%, conduct the shearing and adhesion operations, and then lay the adhered adhesive tape aside for 20min at 23±2°C with the relative humidity of 50±5%.

2) Test of T-type Stripping Strength

Carefully cut the adhesive tape with a knife along the glue coating direction to obtain a sample of about 25.4 mm wide and 127 mm long, remove the release film from one side, and make the sample adhere to the oxidized side of the said anodic alumina foil.

Peel off the release film from the other side and attach the sample to the oxidized side of

another anodic alumina foil (i.e. respectively adhering two sides of the adhesive tape to the anodic alumina foil), and guarantee that the edge of the adhesive tape is within the edge scope of the alumina foil.

Conduct rolling with a 6.8 kg rubber roller for a round at a speed of 305 mm/min, lay aside for a certain time and then slightly bend the extension end (i.e. the end not adhered to the adhesive tape) of the anodic alumina foil on two sides to form an angle of 90° with the adhesive tape, so that the test sample is in the shape of T (the two transverse parts of the T shape are two anodic alumina foils, and the longitudinal part is the adhesive tape provided with the anodic alumina foils on two sides).

Assemble the T-shaped test sample on the test clip of Instron 3343 tester, guarantee that the adhesive tape is centered (i.e. equal length of anodic alumina foils on two sides) and not distorted or deformed, and move the test clip at a speed of 305mm/min to measure the tensile force for stripping the alumina foil from the adhesive tape.

The storage conditions are the same as those in the test of 90° stripping adhesive strength.

3) Measurement of Tape Color

The tape color is measured with HunterLab colorimeter (HunterLab Corporation, Virginia, U.S.).

Employ 10° reflection angle during measurement, and only record the L-value. The measured thickness of the adhesive tape shall be at least 0.8mm. In case the thickness of monolayer adhesive tape is not enough, a plurality of adhesive tapes shall be overlapped to meet the thickness requirement.

Observed by naked eye, the tape color (i.e. black, grey) is regarded as a reference.

4) Measurement of Tape Thickness

The tape thickness is measured with a digital thickness gauge (Mitutoyo Corporation, Tokyo, Japan).

Completely remove the release film from the adhesive tape before the test and then place the adhesive tape between two measurement heads of the digital thickness gauge, guarantee flatness and no extension of the adhesive tape, slightly settle down presser feet of the digital thickness gauge (without collision), read the value 2 seconds later, test each adhesive tape respectively at left, middle and right points in the width direction, and take the average as the measured result.

3. Preparation of Compressible Micron-sized Polymer Particle

The compressible micron-sized polymer particle MA to ML is obtained through forming the metallic layer on the surface of the expanded polymer microspheres by vacuum sputtering according to the substances and parameters as shown in Table 2.

The housing of the vacuum sputtering device is a cylinder with the height of 45cm and the diameter of 50 cm; the particle agitating fluidizer is a transverse cylinder with the length of 12cm and the diameter of 9.5 cm; with the dimensions of 6.5 cm × 7.5 cm, the opening of the particle agitating fluidizer is 7cm away from the metallic sputtering target; the shaft is arranged on the central axis of the particle agitating fluidizer and provided with four orthogonal agitating blades

with the dimensions of 11 cm × 4 cm; each agitating blade is provided with two holes with the diameter of 2 cm, and the terminal of each agitating blade is 2-3 mm away from the internal wall of the particle agitating fluidizer; the magnetron sputtering cathode has a diameter of 3 inches, and the metallic sputtering target has a diameter of 7.6cm and a thickness of 0.48 cm; and the power supply for the magnetron sputtering target is MDX-10 power supply (Advanced Energy Industries Co., Ltd., Colorado, U.S.) matched with Sparc-le 20 glow stabilizer (Advanced Energy Industries Co., Ltd., Colorado, U.S.).

In Table 2, the thickness of the metallic layer is calculated with the weight of sputtered metal and the total surface area of polymer particles. Obviously, the thickness of the metallic layer is obtained through dividing the volume of sputtered metal by the total surface area of polymer particles with ignoring the surface area change caused by the intrinsic thickness of the metallic layer (due to the fact that the thickness of the metallic layer is much less than the particle size of the polymer particle). The volume of sputtered metal can be obtained through dividing the weight of sputtered metal by the density of metal, and the weight of sputtered metal is the weight difference of expanded polymer microsphere before and after the sputtering. The total surface area of polymer particles is calculated as follows:

The object of sputtering is the expanded polymer microspheres, so the object can be regarded as spheres. According to the principles of closing packing of spheres, the maximum space utilization rate when the space is filled with isometric spheres is 74.05%, so the space per unit volume can be filled with as many as 0.7405/(4πR3/3) spheres. The surface area of each sphere is 4πR2, so the theoretical total surface area of spheres in the space per unit volume is represented by Formula:

$$\frac{0.7405}{4\pi R^3/3} \times (4\pi R^2) = 2.2215/R$$

wherein R refers to the radius of sphere.

The expanded polymer microsphere is compressible and not perfectly spherical, with the particle size having a certain distribution range, so the preceding area is required to be revised. We sputter the metallic layer respectively on 20 μm-sized expanded polymer microspheres, 40 μm-sized expanded polymer microspheres and 75μm-sized expanded polymer microspheres, observe the thickness of the actually formed metallic layer with Quanta 600 scanning electron microscope (FEI Co., Ltd., Oregon, U.S.), and then elicit the total surface area of the expanded polymer microspheres according to the thickness of the metallic layer. As illustrated by the results, the total surface area of the expanded polymer microspheres calculated according to the thickness of the actually formed metallic layer is about 3 times of the theoretical value. Therefore, the estimated total surface area of the expanded polymer microspheres in the space per unit volume can be 3 × 2.2215/R.

The experiment comprises steps as follows: using a vessel capable of weighing the volume to contain the expanded polymer microspheres and weigh the volume; conducting vacuum sputtering to the expanded polymer microspheres at different times to obtain the compressible micron-sized

polymer particles (i.e. the expanded polymer microspheres with the metallic layer), measuring the weight again to obtain the weight of sputtered metal; and then calculating the thickness of the metallic layer according to the weight of sputtered metal, the density of metal, the volume of expanded polymer microspheres and the particle size of expanded polymer microspheres as per the preceding steps.

Table 2. Parameters of Compressible Micron-sized Polymer Particle

SN	Type of Expanded Polymer Microsphere	Category of Metal	Density of Metal (g/cm^2)	Volume of Microsphere (ml)	Weight of Sputtered Metal (g)	Estimated Total Surface Area of Microspheres (cm^2)	Thickness of Metallic Layer (nm)
MA	FN-80SDE	Titanium	4.51	241	9.22	803072.3	25.46
MB	461DET80d25	Titanium	4.51	241	5.94	428305.2	30.75
MC	FN-80SDE	Titanium	4.51	241	3.46	803072.3	9.55
MD	FN-80SDE	Tungsten	19.35	241	117.76	803072.3	75.78
ME	FN-80SDE	Tungsten	19.35	241	87.76	803072.3	56.48
MF	461DE20d70	Tungsten	19.35	241	64.43	1606145	20.73
MG	FN-80SDE	Tungsten	19.35	241	32.42	803072.3	20.86
MH	FN-80SDE	Aluminum	2.70	276	6.77	919701	27.26
MI	FN-80SDE	Zirconium	6.51	242	14.55	806404.5	27.72
MJ	FN-80SDE	Chromium	7.19	240	25.00	799740	43.48
MK	FN-80SDE	Niobium	8.57	241	36.15	803072.3	52.53
ML	FN-80SDE	Alloy comprising 65 wt% nickel and 35 wt% chromium	8.25	276	53.34	919701	70.30

4. Preparation of Slurry Polymer

The slurry polymer is prepared through the following steps: adding each polymerizable monomer (with the total use amount of polymerizable monomers in each example being 100 parts by weight) according to the proportion as shown in Table 3 into a 1-quart wide-mouthed jar, adding 0.04 parts by weight of photoinitiator Irgacure 651, purifying with nitrogen for 15min under magnetic stirring, and then exposing the jar under low-intensity ultraviolet with the wavelength of 365 nm and the intensity of 1.5 mw/cm2 to irradiate for several minutes, employing the Ubbelohde

viscometer to constantly measure the viscosity during irradiation, not stopping irradiation until the viscosity at 22°C reaches 2,000 cPs to 6,000 cPs, and filling air to terminate the polymerization to obtain the slurry polymer S1-S4.

Blank cell in the Table indicates the comparative example or the embodiment lacks corresponding substance, similarly hereinafter.

Table 3. Parameters of Slurry Polymer (Parts by Weight)

SN	Use amount of IOA	Use amount of BA	Use amount of 2-EHA	Use amount of IBxA	Use amount of N,N-DMAA	Use amount of AA	Total Use amount
S1			87.5			12.5	100
S2	90					10	100
S3		72		23		5	100
S4			82		17	1	100

5. Specific Embodiment and Performance Test Results

1) Comparative Examples C1 to C3 and Embodiments 1 to 7

The curable composition in each embodiment and comparative example is obtained through the following steps: adding 100 parts of the slurry polymer S1 as well as adding Penncolor 9B117, Irgacure 651 (the use amount illustrated in the Table excludes 0.04 parts by weight added in advance) and HDDA according to the formulation as shown in Table 4 into the 1-quart wide-mouthed jar under a high-speed stirring condition in the absence of light, obtaining clear and colorless solution after all the components are fully dissolved; adding the expanded polymer microspheres FN-80SDE (without the metallic layer), the compressible micron-sized polymer particles (MA, MC) and Aerosil R972, stirring fully; and then adding the glass beads K15, stirring uniformly at a low speed in the absence of light to obtain the curable composition. The adhesive tape in each embodiment and comparative example is obtained through the following steps: coating the curable composition between release sides of two 0.051mm-thick CP Film T10 PET transparent release films (Solutia Co., Ltd., Tennessee, U.S.), controlling the adhesive film with specific thickness, irradiating the curable composition with low-intensity ultraviolet with the wavelength of 365 nm and the intensity of 1.5 mw/cm2 for 5-10min to fully polymerize and cure the curable composition (i.e. the monomer conversion ratio reaching or approaching to 100%), thereby obtaining the adhesive tape. The performance is tested as per the preceding method, and the results are shown in Table 5.

Table 4. Components of Curable Composition in Comparative Examples C1-C3 and Embodiments 1-7 (Parts by Weight)

SN	Use amount of S1	Use amount of HDDA	Use amount of Irgacure 651	Use amount of Penncolor 9B117	Use amount of FN-80SDE	Use amount of MA	Use amount of MC	Use amount of Aerosil R972	Use amount of K15
C1	100	0.08	0.15		4.0				
C2	100	0.20	0.20	0.27	4.0			1.5	
C3	100	0.30	0.26	1.00	4.0			3.0	
Embodiment 1	100	0.20	0.20			1.5		4.5	5.0
Embodiment 2	100	0.20	0.20			3.5		1.5	
Embodiment 3	100	0.26	0.26			3.5		7.0	5.0
Embodiment 4	100	0.26	0.26				3.5	7.0	5.0
Embodiment 5	100	0.26	0.26		4.0	2.0		6.0	
Embodiment 6	100	0.26	0.26		2.0	2.0		6.0	3.0
Embodiment 7	100	0.30	0.28			8.0		10.0	3.0

Table 5. Performance of Adhesive Tape in Comparative Examples C1-C3 and Embodiments 1-7

SN	Thickness (mm)	90° Stripping Adhesive Strength (N/mm)			T-type Stripping Strength (N/mm)		L-value	Color
		First condition	Second condition	Third condition	First condition	Second condition		
C1	0.75	2.543	4.634	2.609	4.394	4.280	88.2	White
C2	0.74	2.482	4.458	1.930	2.921	3.325	42.6	Light grey
C3	0.71	2.458	2.673	1.857	1.853	1.838	22.1	Dark grey-black
Embodiment 1	0.76	3.122	4.609	3.018	3.317	3.530	22.5	Dark grey-black
Embodimen	0.73	2.150	3.302	2.259	3.353	3.204	12.1	Black

t 2								
Embodiment 3	0.77	1.743	2.697	1.888	1.533	1.560	19.6	Black
Embodiment 4	0.78	1.652	2.659	1.599	1.420	1.431	25.6	Grey
Embodiment 5	0.79	2.324	3.264	1.921	1.876	1.975	30.5	Grey
Embodiment 6	0.80	1.597	1.784	1.145	0.907	0.961	26.2	Grey
Embodiment 7	The curable composition is unable to be fully cured.							

2) Embodiments 8 to 16

The curable composition in each embodiment is obtained through the following steps: adding 100 parts of the slurry polymer S2 as well as adding Irgacure 651 (the use amount illustrated in the Table excludes 0.04 parts by weight added in advance), HDDA and Triazine according to the formulation as shown in Table 6 into the 1-quart wide-mouthed jar under a high-speed stirring condition in the absence of light, obtaining clear and colorless solution after all the components are fully dissolved; adding the compressible micron-sized polymer particles (MA, MD, ME, MG) and Aerosil R972, stirring fully; and then adding the glass beads K15, stirring uniformly at a low speed in the absence of light to obtain the curable composition. The adhesive tape in each embodiment is obtained through the following steps: coating the curable composition between release sides of two 0.051 mm-thick CP Film T10 PET transparent release films (Solutia Co., Ltd., Tennessee, U.S.), controlling the adhesive film with specific thickness, irradiating the curable composition with low-intensity ultraviolet with the wavelength of 365 nm and the intensity of 1.5 mw/cm2 for 5-10 min to fully polymerize and cure the curable composition (i.e. the monomer conversion ratio reaching or approaching to 100%), thereby obtaining the adhesive tape. The performance is tested as per the preceding method, and the results are shown in Table 7.

Table 6. Components of Curable Composition in Embodiments 8-16 (Parts by Weight)

SN	Use amount of S2	Use amount of HDDA	Use amount of Triazine	Use amount of Irgacure 651	Use amount of MA	Use amount of MD	Use amount of ME	Use amount of MG	Use amount of Aerosil R972	Use amount of K15
Embodiment 8	100	0.28		0.24	0.8				4.0	5.0
Embodiment 9	100	0.20		0.20		1.0			4.0	
Embodiment 10	100	0.20		0.20			1.0		4.0	
Embodiment 11	100	0.20		0.20				1.0	4.0	
Embodiment 12	100	0.32		0.26	1.0		2.0		6.5	4.0
Embodiment 13	100	0.26		0.26			2.5		7.0	
Embodiment 14	100	0.08	0.12	0.26	1.5		2.5		8.5	2.0
Embodiment 15	100	0.30		0.28			4.0		9.0	6.0
Embodiment 16	100	0.34		0.30			6.0		9.0	5.0

Table 7. Performance of Adhesive Tape in Embodiments 8-16

SN	Thickness (mm)	90° Stripping Adhesive Strength (N/mm)			T-type Stripping Strength (N/mm)		L-value	Color
		First condition	Second condition	Third condition	First condition	Second condition		
Embodiment 8	0.74	1.864	3.236	1.784	2.503	2.840	17.5	Black
Embodiment 9	0.38	1.602	2.659	1.610	3.356	3.347	19.1	Black
Embodiment 10	0.39	1.596	2.729	1.628	3.407	3.123	18.9	Black
Embodimen	0.39	1.507	2.655	1.526	3.349	3.238	29.1	Grey

t 11								
Embodiment 12	1.05	2.692	5.375	2.763	3.004	3.026	24.6	Grey
Embodiment 13	0.22	1.539	2.038	1.497	2.028	2.450	14.9	Black
Embodiment 14	0.45	1.580	2.724	1.506	2.351	2.617	22.1	Dark grey-black
Embodiment 15	0.78	2.279	5.707	1.734	3.104	3.265	25.8	Grey
Embodiment 16	0.78	1.762	3.868	1.705	3.135	2.858	17.7	Black

3) Embodiments 17 to 23

The curable composition in each embodiment is obtained through the following steps: adding 100 parts of the slurry polymer S3 as well as adding Irgacure 651 (the use amount illustrated in the Table excludes 0.04 parts by weight added in advance), HDDA, PETA, Triazine and tackifying resin according to the formulation as shown in Table 8 into the 1-quart wide-mouthed jar under a high-speed stirring condition in the absence of light, obtaining clear and colorless solution after all the components are fully dissolved; adding the compressible micron-sized polymer particles (MB, MF) and Aerosil R972, stirring fully; and then adding the glass beads K15, stirring uniformly at a low speed in the absence of light to obtain the curable composition. The adhesive tape in each embodiment is obtained through the following steps: coating the curable composition between release sides of two 0.051 mm-thick CP Film T10 PET transparent release films (Solutia Co., Ltd., Tennessee, U.S.), controlling the adhesive film with specific thickness, irradiating the curable composition with low-intensity ultraviolet with the wavelength of 365 nm and the intensity of 1.5 mw/cm2 for 5-10 min to fully polymerize and cure the curable composition (i.e. the monomer conversion ratio reaching or approaching to 100%), thereby obtaining the adhesive tape. The performance is tested as per the preceding method, and the results are shown in Table 9.

Table 9. Components of Curable Composition in Embodiments 17-23 (Parts by Weight)

SN	Use amount of S3	Use amount of HDDA	Use amount of PETA	Use amount of Triazine	Use amount of Irgacure 651	Use amount of MF	Use amount of MB	Use amount of Aerosil R972	Use amount of UH115	Use amount of Foral 85	Use amount of K15
Embodiment 17	100	0.25			0.24	3.0		2.5			
Embodiment 18	100	0.36			0.30	6.0		12.0	30.0		
Embodiment 19	100	0.50			0.35	9.0		15.0			3.0
Embodiment 20	100		0.30		0.24		2.5	0.5			1.0
Embodiment 21	100	0.32			0.24		4.5	7.0			1.5
Embodiment 22	100	0.56			0.32	3.0	1.5	13.0		15.0	3.0
Embodiment 23	100			0.24	0.32	4.0	2.0	15.0	15.0	10.0	3.0

Table 9. Performance of Adhesive Tape in Embodiments 17-23

SN	Thickness (mm)	90° Stripping Adhesive Strength (N/mm)			T-type Stripping Strength (N/mm)		L-value	Color
		First condition	Second condition	Third condition	First condition	Second condition		
Embodiment 17	0.11	0.725	0.923	0.718	1.103	1.081	24.7	Grey
Embodiment 18	0.16	1.245	1.428	1.201	1.689	1.623	17.9	Black
Embodiment 19	The curable composition is unable to be fully cured.							
Embodiment 20	1.49	2.213	3.126	2.081	1.517	1.647	26.8	Grey
Embodiment 21	1.56	2.538	3.864	2.481	2.897	2.752	19.1	Black

Embodiment 22	2.01	3.115	4.587	3.022	3.642	3.705	18.6	Black
Embodiment 23	1.98	3.232	4.642	3.217	3.801	3.829	19.7	Black

4) Embodiments 24 to 27

The curable composition in each embodiment is obtained through the following steps: adding 100 parts of the slurry polymer S4 as well as adding Irgacure 651 (the use amount illustrated in the Table excludes 0.04 parts by weight added in advance) and HDDA according to the formulation as shown in Table 10 into the 1-quart wide-mouthed jar under a high-speed stirring condition in the absence of light, obtaining clear and colorless solution after all the components are fully dissolved; adding the compressible micron-sized polymer particles (MH, MI, MJ, MK) and Aerosil R972, stirring fully; and then adding the glass beads K15, stirring uniformly at a low speed in the absence of light to obtain the curable composition. The adhesive tape in each embodiment is obtained through the following steps: coating the curable composition between release sides of two 0.051 mm-thick CP Film T10 PET transparent release films (Solutia Co., Ltd., Tennessee, U.S.), controlling the adhesive film with specific thickness, irradiating the curable composition with low-intensity ultraviolet with the wavelength of 365 nm and the intensity of 1.5 mw/cm2 for 5-10 min to fully polymerize and cure the curable composition (i.e. the monomer conversion ratio reaching or approaching to 100%), thereby obtaining the adhesive tape. The performance is tested as per the preceding method, and the results are shown in Table 11.

Table 10. Components of Curable Composition in Embodiments 24-27 (Parts by Weight)

SN	Use amount of S4	Use amount of HDDA	Use amount of Irgacure 651	Use amount of MH	Use amount of MI	Use amount of MJ	Use amount of MK	Use amount of Aerosil R972	Use amount of K15
Embodiment 24	100	0.26	0.26	2.0				6.0	3.0
Embodiment 25	100	0.26	0.26		2.0			6.0	3.0
Embodiment 26	100	0.26	0.26			2.0		6.0	3.0
Embodiment 27	100	0.26	0.26				2.0	6.0	3.0

Table 11. Performance of Adhesive Tape in Embodiments 24-27

SN	Thickness (mm)	90° Stripping Adhesive Strength (N/mm)			T-type Stripping Strength (N/mm)		L-value	Color
		First condition	Second condition	Third condition	First condition	Second condition		
Embodiment 24	0.79	0.721	0.978	0.675	1.019	1.183	2.72	Grey
Embodiment 25	0.81	0.764	0.996	0.723	1.123	1.087	2.68	Grey
Embodiment 26	0.76	0.697	0.921	0.703	0.998	1.052	2.23	Dark grey-black
Embodiment 27	0.78	0.706	0.963	0.721	1.106	1.074	2.19	Dark grey-black

5) Embodiments 28 to 29

The curable composition in each embodiment is obtained through the following steps: adding 100 parts of the slurry polymer S2 as well as adding Irgacure 651 (the use amount illustrated in the Table excludes 0.04 parts by weight added in advance) and HDDA according to the formulation as shown in Table 12 into the 1-quart wide-mouthed jar under a high-speed stirring condition in the absence of light, obtaining clear and colorless solution after all the components are fully dissolved; adding the compressible micron-sized polymer particles (ML) and Aerosil R972, stirring fully; and then adding the glass beads K15, stirring uniformly at a low speed in the absence of light to obtain the curable composition.

Unlike the adhesive tape in the preceding embodiments, the adhesive tape in this embodiment uses the pressure-sensitive adhesive as a core and is provided with other adhesive layers on two sides. The preparation method of the adhesive tape in this step comprises specific steps of:

coating the curable composition on a matrix, controlling the adhesive film within specific thickness, irradiating the curable composition with low-intensity ultraviolet with the wavelength of 365 nm and the intensity of 1.5 mw/cm2 for 5-10 min to fully polymerize and cure the curable composition (i.e. the monomer conversion ratio reaching or approaching to 100%) to obtain the pressure-sensitive adhesive core;

coating a primer coating on each side of the core, wherein the primer coating comprises:

a solute comprising 10 parts by weight of Macromelt 6240 (Henkel Co., Ltd., Dusseldorf, Germany), and a solvent comprising 47.5 parts by weight of isopropanol (Sinopharm Chemical Reagent Co., Ltd., Shanghai, China),47.5 parts by weight of n-propanol (Sinopharm Chemical Reagent Co., Ltd., Shanghai, China) and 5 parts by weight of deionized water;

standing for 10min at room temperature to obtain a primary coating with the thickness of 8.3 μm; and

respectively covering a 50 μm-thick double-sized adhesive film 467MP (3M Co., Ltd., Minnesota, U.S.) on the primary coating on both sides, laminating with the rubber roller and then standing for one day at room temperature to obtain the adhesive tape. The performance is tested as per the preceding method, and the results are shown in Table 13.

Table 12. Components of Curable Composition in Embodiments 28-29 (Parts by Weight)

SN	Use amount of S2	Use amount of HDDA	Use amount of Irgacure 651	Use amount of ML	Use amount of Aerosil R972	Use amount of K15
Embodiment 28	100	0.29	0.20	0.1	0.1	0.5
Embodiment 29	100	0.34	0.27	7.0	11.0	

Table 13. Performance of Adhesive Tape in Embodiments 28-29

SN	Thickness of Core (mm)	Overall Thickness of Adhesive Tape (mm)	90° Stripping Adhesive Strength (N/mm)			T-type Stripping Strength (N/mm)		L-value	Color
			First condition	Second condition	Third condition	First condition	Second condition		
Embodiment 28	0.49	0.60	1.932	3.123	1.819	1.697	1.664	28.1	Grey
Embodiment 29	0.82	0.92	2.620	4.985	2.576	3.261	3.329	16.9	Black

6. Performance Analysis

According to comparison and analysis of the test data in each comparative example and embodiment, the conclusions are as follows:

(1) For the adhesive tape (added with the expanded polymer microspheres without the metallic layer) realizing black with the black dye, each strength performance index of the adhesive tape is liable to be obviously poor as the color becomes darker. However, the adhesive tape provided by the present invention (added with the compressible micron-sized polymer particles) can realize superior cohesive strength and stripping adhesive strength based on guaranteeing dark color.

(2) In case other conditions are relatively constant, the more the compressible micron-sized

polymer particles are, the darker the tape color is; however, the strength performance of the adhesive tape is improved rather than degraded. It is thus clear that the compressible micron-sized polymer particle can enable the adhesive tape to become black, simultaneously with improving the strength rather than degrading the strength.

(3) In presence of the compressible micron-sized polymer particle, other components (i.e. the tackifying resin, the glass bead and the expanded polymer microsphere) are still available, which indicates that the application of the compressible micron-sized polymer particle does not have adverse effects on the application of other components.

(4) In case other conditions are relatively constant, the thicker the metallic layer in the compressible micron-sized polymer particle is, the darker the tape color is. It is thus clear that the selection of the thickness of the metallic layer has an important impact on the color of the product.

(5) In case the use amount of the compressible micron-sized polymer particles exceeds the upper limit specified in the present invention (i.e. exceeding 7 parts by weight with respect to 100 parts of the polymerizable monomers), the curable composition is unable to be fully cured. It is thus clear that the use amount of the compressible micron-sized polymer particles shall not exceed the range specified in the present invention.

(6) For the adhesive tape provided by the present invention, the pressure-sensitive adhesive can be directly used as the core and the adhesive; alternatively, the pressure-sensitive adhesive can be used as the core and other adhesive layers can be employed. Both methods are feasible. Therefore, adhesive layers with different properties can be selected to realize more functions of the adhesive tape if necessary.

It can be understood that, the preceding embodiments are exemplary embodiments for illustrating principles of the present invention, and the present invention is not restricted by the embodiments. A person skilled in the art can make some modifications and improvements based on and not deviating from the spirit and scope of the present invention, and these modifications and improvements are within the protection scope of the present invention.

Claims:

1. A compressible micron-sized polymer particle, comprising:
a polymer particle having a shell formed by a polymer material and a cavity surrounded by the shell; and
a metallic layer covering the external surface of the shell.

2. The compressible micron-sized polymer particle according to Claim 1, wherein the polymer particle is basically of a spherical shape.

3. The compressible micron-sized polymer particle according to Claim 1, wherein the particle size of the polymer particle ranges from 10 μm to 100 μm; and the thickness of the shell of the polymer particle ranges from 2 μm to 15 μm.

4. The compressible micron-sized polymer particle according to Claim 1, wherein the polymer material is a thermoplastic polymer, and the cavity is filled with gas.

5. The compressible micron-sized polymer particle according to Claim 4, wherein the thermoplastic polymer is thermoplastic polyacrylic resin, and the gas is alkane.

6. The compressible micron-sized polymer particle according to Claim 1, wherein the material of metallic layer is selected from any one of tungsten, titanium, aluminum, chromium, niobium and zirconium, or the alloy thereof.

7. The compressible micron-sized polymer particle according to Claim 1, wherein the thickness of the metallic layer ranges from 5 nm to 80 nm.

8. The compressible micron-sized polymer particle according to Claim 7, wherein the thickness of the metallic layer ranges from 6 nm to 70 nm.

9. The compressible micron-sized polymer particle according to Claim 8, wherein the thickness of the metallic layer ranges from 12 nm to 60 nm.

10. The compressible micron-sized polymer particle according to Claim 9, wherein the thickness of the metallic layer ranges from 20 nm to 60 nm.

11. The compressible micron-sized polymer particle according to Claim 1, wherein the metallic layer is formed on the external surface of the polymer particle by vacuum sputtering.

12. A curable composition, comprising:
a slurry polymer which comprises a mixture formed of at least two polymerizable monomers by partial copolymerization, wherein the said mixture comprises the said polymerizable monomers unpolymerized and a copolymer formed of the said polymerizable monomers by copolymerization;
a photoinitiator; and
a compressible micron-sized polymer particle which comprises a polymer particle having a shell formed by a polymer material and a cavity surrounded by the shell; and
a metallic layer covering the external surface of the shell.

13. The curable composition according to Claim 12, wherein the polymerizable monomers comprise:
acrylate monomers serving as first monomers; and
non-ester unsaturated monomers serving as second monomers and with at least one olefinic bond.

14. The curable composition according to Claim 13, wherein
the first monomers are non-tertiary alcohol (meth)acrylate monomers; and
the second monomers are acid-functional non-ester unsaturated monomers with at least one olefinic bond.

15. The curable composition according to Claim 12, wherein the viscosity of the slurry polymer at 22°C ranges from 500 cPS to 10,000 cPs.

16. The curable composition according to Claim 12, wherein
the weight-average molecular weight of the copolymer ranges from 500,000 D to 10,000,000 D; and
the ratio of the weight of the polymerized polymerizable monomers to the total weight of the polymerized and unpolymerized polymerizable monomers is between 1% and 30%.

17. The curable composition according to Claim 12, wherein the ratio of the weight of the compressible micron-sized polymer particles to the total weight of the polymerized and unpolymerized polymerizable monomers is between 0.1% and 7%.

18. The curable composition according to Claim 17, wherein the ratio of the weight of the compressible micron-sized polymer particles to the total weight of the polymerized

and unpolymerized polymerizable monomers is between 1% and 7%.

19. The curable composition according to Claim 18, wherein the ratio of the weight of the compressible micron-sized polymer particles to the total weight of the polymerized and unpolymerized polymerizable monomers is between 1% and 6%.

20. The curable composition according to Claim 12, wherein it also comprises a crosslinking agent.

21. The curable composition according to Claim 20, wherein the crosslinking agent comprises multifunctional (meth)acrylate and/or triazine.

22. The curable composition according to Claim 12, wherein it also comprises hydrophobic fumed silica.

23. A preparation method of a curable composition, comprising the following steps:
partially copolymerizing at least two polymerizable monomers to obtain the slurry polymer; and
mixing a photoinitiator and a compressible micron-sized polymer particle with the said slurry polymer, wherein the compressible micron-sized polymer particle comprises a polymer particle having a shell formed by a polymer material and a cavity surrounded by the shell; and a metallic layer covering the external surface of the shell.

24. A pressure-sensitive adhesive, formed of a curable composition which comprises:
a slurry polymer which comprises a mixture formed of at least two polymerizable monomers by partial copolymerization, wherein the said mixture comprises the said polymerizable monomers unpolymerized and a copolymer formed of the said polymerizable monomers by copolymerization;
a photoinitiator; and
a compressible micron-sized polymer particle which comprises a polymer particle having a shell formed by a polymer material and a cavity surrounded by the shell; and a metallic layer covering the external surface of the shell.

25. An adhesive tape, comprising a pressure-sensitive adhesive formed of a curable composition which comprises:
a slurry polymer which comprises a mixture formed of at least two polymerizable monomers by partial copolymerization, wherein the said mixture comprises the said polymerizable monomers unpolymerized and a copolymer formed of the said

polymerizable monomers by copolymerization;

a photoinitiator; and

a compressible micron-sized polymer particle which comprises a polymer particle having a shell formed by a polymer material and a cavity surrounded by the shell; and a metallic layer covering the external surface of the shell.

26. The adhesive tape according to Claim 25, wherein

the adhesive tape is formed of a pressure-sensitive adhesive;

or

the adhesive tape comprises a core and an adhesive layer positioned on at least one surface of the core which is formed of the pressure-sensitive adhesive.

27. The adhesive tape according to Claim 25, wherein the L-value of the tape color ranges from 0 to 30 in a color system indicated by an L value, an a value and a b value

28. The adhesive tape according to Claim 27, wherein the L value of the tape color ranges from 0 to 25.

29. The adhesive tape according to Claim 28, wherein the L value of the tape color ranges from 0 to 20.

30. The adhesive tape according to Claim 25, wherein the pressure-sensitive adhesive of the adhesive tape is 0.05 mm to 5 mm thick.

31. A compressible micron-sized polymer particle used for a pressure-sensitive adhesive, wherein

the compressible micron-sized polymer particle comprises: a polymer particle having a shell formed by a polymer material and a cavity surrounded by the shell; and a metallic layer covering the external surface of the shell; and

the pressure-sensitive adhesive is polyacrylate pressure-sensitive adhesive.

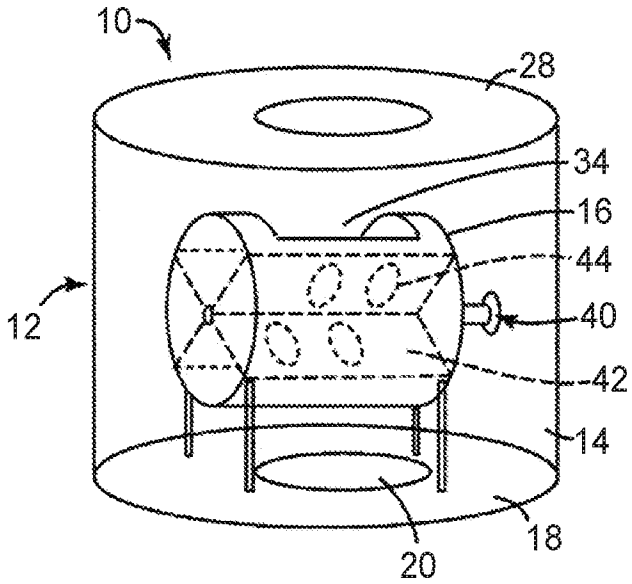

FIG. 1

INTERNATIONAL SEARCH REPORT

International application No
PCT/US2015/066569

A. CLASSIFICATION OF SUBJECT MATTER
INV. C09J4/06 C09J9/02 C09J133/06 C09J151/06 C23C14/35
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)
C09J C23C

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, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 2013/118773 A1 (LIU JUNKANG J [US] ET AL) 16 May 2013 (2013-05-16) paragraphs [0041], [0045] claims -----	1,2, 11-14, 23-25,31
X	WO 2014/075304 A1 (3M INNOVATIVE PROPERTIES CO [US]; LIU WEIDE [CN]; VEERARAGHAVAN BADRI) 22 May 2014 (2014-05-22) claims -----	1,2, 11-14, 23-25,31
X	EP 2 662 428 A1 (NITTO DENKO CORP [JP]) 13 November 2013 (2013-11-13) claims ----- -/--	1,2, 11-14, 23-25,31

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

[X] See patent family annex.

* Special categories of cited documents :

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"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

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Date of the actual completion of the international search	Date of mailing of the international search report
15 March 2016	21/03/2016

Name and mailing address of the ISA/	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Fax: (+31-70) 340-3016	Kaumann, Edgar

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

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	CN 103 320 037 A (3M SHANGHAI SPECIALTY MATERIALS CO LTD) 25 September 2013 (2013-09-25) cited in the application claims	1-31
Y	WO 2014/009552 A2 (CONPART AS [NO]) 16 January 2014 (2014-01-16) page 11, line 16 - line 23 page 14, line 27 - page 15, line 1 claims	1-31
A	WO 2011/087664 A1 (3M INNOVATIVE PROPERTIES CO [US]) 21 July 2011 (2011-07-21) claims	1-31

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No
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