Title: MULTILAYER UV-CURABLE ADHESIVE FILM

Abstract: The present invention provides a multilayer UV-curable adhesive film, a multilayer UV-curable adhesive film which comprises an intermediate adhesive film layer and two surface adhesive film layers respectively disposed on two sides of the intermediate adhesive film layer, wherein a UV-curable composition I for forming the intermediate adhesive film layer and a UV-curable composition II for forming the two surface adhesive film layers independently contain a specific amount of ethylene-vinyl acetate copolymer, epoxy resin compound, hydroxy-containing compound, photoinitiator, and antioxidant. The multilayer UV-curable adhesive film provided by the present invention achieves a good balance between initial tack, high structural strength after curing, impact resistance, odorlessness or low odor, and long shelf life.
MULTILAYER UV-CURABLE ADHESIVE FILM

TECHNICAL FIELD

The present invention relates to the technical field of pressure-sensitive adhesives, and in particular to a UV-curable adhesive film.

BACKGROUND

For household appliances (such as televisions, refrigerators, washing machines and range hoods), automobile and the like, the aesthetic appeal thereof are currently increasing in importance. As a result, there is a trend that the use of bolts are replaced with adhesive tapes or adhesive films. In the meantime, it is important to make sure that the adhesives adopted in these products are low in odor. Furthermore, electronic products such as notebooks and hand-held mobile devices are also trending toward compactness, lightness, thinness and production efficiency. Such a trend necessitates that polymers (such as plastics and rubber) become more frequently used in these products. The use of adhesives with high bonding strength are thus desired in the production of these electronic products; the adhesives ought to be able to create firm bonds using a small bond area and ought to be low in odor.

For many of the products mentioned above, since the bonded parts will encounter strong impact during transportation or use, the adopted adhesive films ought to possess excellent impact resistance in addition to strong bonding performance.

Traditionally, single-component or two-component structural glue would meet the requirements for bonding components in these products. However, structural glue has many disadvantages: it cannot be removed or re-applied (i.e., re-assembled after removal); the bond area is hard to control, causing the bonding surface to be less attractive; some structural glue contains a large amount of solvents, which require a long time to cure (i.e., it is inefficient) or need to be cured at a high temperature (i.e., cannot be applied on non-thermostable plastic materials); structural glue often has a very high modulus, which means that the structural glue does not have good impact resistance. All these aspects greatly prevent structural glue from being widely adopted in the products mentioned above.

Pressure-sensitive tapes are also an option commonly adopted for bonding. They are convenient to use. A bond is created as soon as a small amount of pressure is applied to the pressure-sensitive tape. Curing for a long time or under high temperature over 160°C is not necessary. A continuous production line can be achieved because pressure-sensitive tapes can immediately form bonds following die-cutting. Bonding efficiency is thus very high. However, a major disadvantage of pressure-sensitive tapes are the weak bonding strength, which generally does not exceed 1 MPa. They are thus not suitable for small-area bonding.
Another widely used traditional product is foam adhesive tape, which is similar to pressure-sensitive adhesive tape. A bond is created as soon as a small amount of pressure is applied to the foam adhesive tape. Curing for a long time or under high temperature over 160°C is not necessary. A continuous production line can be achieved because foam adhesive tapes can immediately form bonds following die-cutting. Bonding efficiency is thus very high. Moreover, the bonding strength of foam adhesive tape is higher than that of pressure-sensitive adhesive tape. However, the biggest disadvantage of foam adhesive tape is the strong odor, making it unsuited for use in household appliances or automobile interiors.

Patent US2002/182955A1 (Weglewski) has disclosed a single-layer structural adhesive tape reinforced by using fibers. However, the structural adhesive tape requires that the adhesive film must be reinforced by using specific fibers to achieve good shape retention and bonding strength, which makes the manufacturing process significantly more complex and the cost significantly higher. In addition, the production process of this structural adhesive tape requires vacuum hot pressing to achieve good bonding results, which also makes the production process cost higher and the required conditions harsher. It is difficult to achieve large scale production using this structural adhesive tape.

Therefore, there is still a need in this field for an adhesive film or tape that has the following properties: good initial tack, high structural strength after curing, impact resistance, odorlessness or low odor, long shelf life, and can solve the common problem of adhesive overflow.

**SUMMARY**

In view of the above, the purpose of the present invention is to provide a novel UV-curable adhesive film which achieves a good balance between initial tack, high structural strength after curing, impact resistance, odorlessness or low odor, and long shelf life.

The present invention thus provides a multilayer UV-curable adhesive film; the multilayer UV-curable adhesive film comprises an intermediate adhesive film layer and two surface adhesive film layers respectively disposed on two sides of the intermediate adhesive film layer. The intermediate adhesive film layer is formed by a UV-curable composition I, and the surface adhesive film layers are formed by a UV-curable composition II, wherein based on a total weight of the UV-curable composition I, the UV-curable composition I comprises the following components:

- a) 70-85 wt. % of ethylene-vinyl acetate copolymer;
- b) 15-30 wt. % of epoxy resin compound;
- c) 0.3-8 wt. % of hydroxyl-containing compound;
- d) 0.5-5 wt. % of a photoinitiator; and
- e) 0.5-2 wt. % of an antioxidant, and wherein
based on a total weight of the UV-curable composition II, the UV-curable composition II comprises
the following components:
   a) 30-70 wt. % of ethylene-vinyl acetate copolymer;
   b) 30-70 wt. % of epoxy resin compound;
   c) 0.3-8 wt. % of hydroxyl-containing compound;
   d) 0.5-5 wt. % of a photoinitiator; and
   e) 0.5-2 wt. % of an antioxidant.

In one preferred embodiment, in the ethylene-vinyl acetate copolymers of the UV-curable
compositions I and II, the content of vinyl acetate units is 60-90 wt. %.

In one preferred embodiment, the ethylene-vinyl acetate copolymer is a pre-cross-linked polymer
or a non-cross-linked linear polymer, and the ethylene-vinyl acetate copolymers in the UV-curable
compositions I and II are the same or different.

In one preferred embodiment, the epoxy resin compounds in the UV-curable compositions I and II are
one or more selected from aromatic epoxy resins or epoxidized polyolefins.

In one preferred embodiment, the UV-curable composition I comprises 20-30 wt. % of the epoxy
resin compound based on the total weight of the UV-curable composition I; and the UV-curable
composition II comprises 30-50 wt. % of the epoxy resin compound based on the total weight of
the UV-curable composition II.

In one preferred embodiment, the epoxy equivalent weights of the epoxy resin compounds in the
UV-curable compositions I and II are within a range of 150-600 grams.

In one preferred embodiment, the epoxy resin compounds in the UV-curable compositions I and
II are the same or different in type and/or epoxy equivalent weight.

In one preferred embodiment, the hydroxyl-containing compounds in the UV-curable
compositions I and II are polyols, such as polyester polyols or polyether polyols.

In one preferred embodiment, the photoinitiators in the UV-curable compositions I and II are
cationic photoinitiators.

In one preferred embodiment, the cationic photoinitiators are one or more selected from diazo salts,
iodonium salts, sulfonium salts, antimony onium salts and iron aromatic hydrocarbon salts.

In one preferred embodiment, the UV-curable compositions I and II independently further comprise
one or more selected from an electric conducting agent, a heat conducting agent, a flame retardant,
and a filler.
In one preferred embodiment, the electric conducting agent is electric conducting particles or fibers, the heat conducting agent is heat conducting particles or fibers, and the filler is silicon dioxide.

In one preferred embodiment, the UV-curable compositions I and II do not contain solvents.

In one preferred embodiment, the thickness of the intermediate adhesive film layer is 300-800 μm.

In one preferred embodiment, the thickness of the two surface adhesive film layers is independently 50-100 μm.

In one preferred embodiment, the intermediate adhesive film layer and two surface adhesive film layers are formed through hot melt extrusion.

In one preferred embodiment, the multilayer UV-curable adhesive film further comprises a release paper or a release film disposed on one or two of the two surface adhesive film layers.

The advantages of the present invention include, but are not limited to, the following aspects:

A multilayer (usually three-layer) UV-curable adhesive film structure is designed in the present invention by using UV-curable compositions with specific components and contents, wherein the intermediate adhesive film layer and two surface adhesive film layers on two sides of the intermediate adhesive film layer use ethylene-vinyl acetate copolymers with rubber elastomer properties as base materials, and use epoxy resin compounds as curable materials. Moreover, the content of the ethylene-vinyl acetate copolymers in the intermediate layers is not lower than and is preferably higher than that in the surface layers on the two sides; and the content of epoxy resin in the intermediate layers is not higher than and is preferably lower than that in the surface layers. Higher epoxy content in the surface layers can provide a stronger interfacial effect, thus achieving higher initial tack and bonding strength after curing. Lower epoxy content in the intermediate layers imparts good energy absorption and cushioning effects to the adhesive film. As a result, the obtained multilayer adhesive film structure has good initial tack as well as impact resistance. The obtained multilayer adhesive film structure further has excellent structural strength after curing, and would not cause problems of adhesive overflow.

In addition, the multilayer UV-curable adhesive film structure obtained by the present invention can be stored for a long time because of the specific content of antioxidants that is contained in the UV-curable compositions I or II of the adhesive film. For example, the multilayer UV-curable adhesive film provided by the present invention can be stored at room temperature for a long time, and can be stored for more than 24 months if stored in a dark place. Compared with the cryopreservation requirement for most of the existing adhesive films, the transportation and storage costs for the UV-curable adhesive film of the present invention are greatly reduced.
In addition, the compositions I and II which are used to form the multilayer UV-curable adhesive film structure provided by the present invention adopt EVA copolymers with large molecular weight. The use of organic solvents is thus avoided in the preparation of the multilayer adhesive film, imparting the feature of low odor or odorlessness to the multilayer UV-curable adhesive film obtained by the present invention.

In addition, the UV-curable compositions of the adhesive film provided by the present invention can be used to form an intermediate adhesive film layer and surface adhesive film layers through, for example, hot melt extrusion method. The required multilayer adhesive film structure can thereby be easily and conveniently prepared. Preferably, the intermediate layers prepared through hot melt extrusion are thicker than the surface layers on the two sides, thus ensuring that the prepared adhesive film structure can be fully cured through ultraviolet irradiation.

In addition, the multilayer UV-curable adhesive film provided by the present invention can be conveniently prepared at room temperature through a simple process; large scale production can be easily achieved.

In addition, the multilayer UV-curable adhesive film provided by the present invention possesses features similar with the common pressure-sensitive adhesive tape at the initial stage of bonding. That is, the adhesive film of the present invention has initial tack, can create a bond between objects when a small amount of pressure is applied, and can be molded through die cutting. After being induced by UV, the multilayer adhesive film can be cured into an adhesive tape with semi-structural strength to structural strength at room temperature, and thus it can be used to bond non-heat-resistant plastics such as PC, PMMA, and ABS. Moreover, the adhesive film provided by the present invention has no odor or low odor, making it especially suitable for the bonding between internal parts of household appliances and automobiles, between plastic parts, or between plastic parts and metal parts in electronic products such as hand-held mobile devices.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a structural diagram of a multilayer UV-curable adhesive film according to one embodiment of the present invention.

DETAILED DESCRIPTION

After in-depth studies, the inventor of the invention discovered the following: by using UV-curable compositions with specific components and contents to form the intermediate layer and two side surface layers respectively of the multilayer UV-curable adhesive film, a multilayer (usually three-layer) UV-curable adhesive film structure formed therefrom has excellent and balanced performance. Herein, the terms “adhesive film” and: “adhesive tape” are interchangeable.
In the multilayer UV-curable adhesive film structure provided by the present invention, the UV-
induced curable compositions of ethylene-vinyl acetate copolymer/epoxy resin hybrid systems with
specific components are used to form an intermediate adhesive film layer and two surface adhesive
film layers on two sides (or upper and lower surfaces) thereof respectively.

As used in the description about the adhesive film and composition I and/or II herein, the term “curable”
means that the film or composition referred to can be cured by chemical reaction of epoxy resin
components in the adhesive film or composition initiated by a photoinitiator therein under the induction
by UV light. In the present invention, after irradiation or induction by UV light, even after the UV source
is removed, the reaction of epoxy groups can continue to be initiated at room temperature, thus
completing the curing process (so-called active polymerization process). In addition, each layer of the
multilayer adhesive film is produced by hot melting method. The use of solvents is avoided to achieve
the effect of low odor. The intermediate layers of the multilayer adhesive film have a low modulus, which
imparts good impact resistance to the cured adhesive film.

The low-odor multilayer UV-curable adhesive film provided by the present invention exhibits features
similar to the common pressure-sensitive adhesive tape at the initial stage of bonding (i.e., having initial
tack), and it can bond the bonded objects by applying small pressure and can be molded through die
cutting; after being induced by UV, the adhesive film can be cured into a tape with semi-structural
strength to structural strength at room temperature. As a result, the adhesive film of the present invention
can bond some plastic products such as PC, PMMA and ABS which are not resistant to high temperature.

At the same time, since the UV-curable composition I or II in the present invention does not contain
solvents, and the adhesive film is produced by solvent-free hot melt extrusion, the present invention meets
the requirements of low odor or odorlessness. The invention is especially suitable for the bonding
between internal parts of household electric appliances and automobiles, and between plastic parts and
plastic parts, or between plastic parts and metal parts in electronic products such as hand-held mobile
 terminals. In addition, the UV-induced curable high-temperature pressure-sensitive adhesive tape
provided by the present invention has a room-temperature storage period of more than 24 months in a
dark place.

As used herein, the term “structural adhesive” refers to an adhesive with a shear strength greater
than 1000 psi between the adhesive tape and the bonded parts (1 MPa is about 145 psi); the term
“structural strength” refers to the shear strength of the bonded member formed by the adhesive tape
and the bonded parts which is greater than 1000 psi; and the term “semi-structural strength” refers
to the shear strength of the bonded member formed by the adhesive tape and the bonded parts which
is greater than 100 psi but smaller than 1000 psi.
Unless otherwise specified, all numbers used in this description and the claims representing the characteristic sizes and quantities and physical properties should be understood as being modified by the term “approximately” under any and all circumstances. Therefore, unless stated on the contrary, parameters in numerical values listed in the above description and in the attached claims are all approximate values, and one skilled in the art is capable of seeking to obtain desired properties by taking advantage of contents of the teachings disclosed herein, and changing these approximate values appropriately. The use of a numeric value range represented by endpoints includes all numbers within such range and any range within such range. For example, the range “1 to 5” intends to include 1, 1.1, 1.3, 1.5, 2, 2.75, 3, 3.80, 4, 5, etc.

In the embodiment of the present invention, the present invention provides a multilayer UV-curable adhesive film, comprising an intermediate adhesive film layer and two surface adhesive film layers respectively disposed on two sides of the intermediate adhesive film layer, the intermediate adhesive film layer being formed by a UV-curable composition I, and the surface adhesive film layers being formed by a UV-curable composition II, wherein

the UV-curable composition I comprises the following components based on a total weight of the UV-curable composition I: a) 70-85 wt. % of an ethylene-vinyl acetate copolymer; b) 15-30 wt. % of an epoxy resin compound; c) 0.3-8 wt. % of hydroxyl-containing compound; d) 0.5-5 wt. % of a photoinitiator; and e) 0.5-2 wt. % of an antioxidant,

and wherein the UV-curable composition II comprises the following components based on a total weight of the UV-curable composition II: a) 30-70 wt. % of an ethylene-vinyl acetate copolymer; b) 30-70 wt. % of an epoxy resin compound; c) 0.3-8 wt. % of hydroxyl-containing compound; d) 0.5-5 wt. % of a photoinitiator; and e) 0.5-2 wt. % of an antioxidant.

a) Ethylene-vinyl acetate copolymer

In the multilayer UV-curable adhesive film structure provided by the present invention, the ethylene-vinyl acetate copolymers with rubber elastomer properties are used as the base materials in the intermediate adhesive film layer and two surface adhesive film layers on two sides, or upper and lower surfaces. The ethylene-vinyl acetate copolymer (abbreviated as “EVA”) used in the present invention may be a non-cross-linked or non-cross-linked linear copolymer or a pre-cross-linked ethylene-vinyl acetate copolymer with a certain degree of cross-linking. In one preferred embodiment, the ethylene-vinyl acetate copolymers in the UV-curable compositions I and II used in the present invention may be the same or different, and the ethylene-vinyl acetate copolymers in the UV-curable compositions I and II are not pre-cross-linked polymers at the same time.
In some preferred embodiments, the content of vinyl acetate units in the ethylene-vinyl acetate copolymers is within a range of 60-90 wt. %, preferably 70-80 wt. %. This is because the higher the content of vinyl acetate, the higher the Tg of the copolymer, and the higher the modulus of the cured adhesive film. Thereby, the obtained adhesive film or tape can be enabled to have higher shear strength.

The ethylene-vinyl acetate copolymers that can be used for the present invention may be commercially obtained, such as LEVAPREN 600, LEVAPREN 700, LEVAPREN 800, LEVAPREN 900, LEVAPREN 700 XL, LEVAPREN 800 XL and the like from LANXESS.

In order for the adhesive film structure obtained by the present invention to have good balanced performance in initial tack, impact resistance, structural strength after curing, and especially good impact resistance, it is required that the amount of the ethylene-vinyl acetate copolymer contained in the UV-curable composition I used to form the intermediate adhesive film layer is not lower than (preferably higher than) that in the UV-curable composition II used to form the surface adhesive film layers on the two sides.

b) Epoxy resin compound

In the UV-curable compositions in the two-component adhesive film of the present invention, in addition to the ethylene-vinyl acetate copolymers as the base materials, the epoxy resin compounds are further used as base materials and as curing components at the same time.

The epoxy resin compound that can be used for the present invention may be epoxy resin compounds known in the field. For example, in some embodiments, each molecule of the used epoxy resin compounds may contain one or more epoxy groups, and the epoxy equivalent weight thereof is preferably within a range of 150-600 grams. Preferably, the present invention may use aromatic epoxy resins and their derivatives such as glycidyl ethers or esters obtained by reaction of polyphenols such as bisphenol A, bisphenol F, bisphenol S, hexahydrobisphenol A, tetramethyl bisphenol A, diaryl bisphenol A and tetramethyl bisphenol F with epichlorohydrin. In addition, epoxidized polyolefins are known epoxy resins that can be used.

The epoxy resin compounds that can be used for the present invention may be commercially obtained, and such as YD 128 (with an epoxy equivalent weight of about 187 grams) and KD212 (with an epoxy equivalent weight of 535 grams) from Kudko Chemical Co., Ltd. (Korea).

In order for the adhesive film structure obtained by the present invention to have good balanced performance in initial tack, impact resistance, structural strength after curing, and especially good initial tack, it is required that the amount of the epoxy resin compound contained in the UV-curable composition I used to form the intermediate adhesive film layer is not higher than (preferably lower than) that in the UV-curable composition II used to form the surface adhesive film layers.
on the two sides. More specifically, the content of the epoxy resin compound in the UV-curable composition II used to form the surface adhesive film layers on the two sides is generally within a range of 30-70%, more preferably within 35-60%, and further preferably within 40-50%. Higher epoxy content in the surface layers can provide strong interfacial effect, which can ensure the initial tack of the formed adhesive film and the sufficient strength of the cured adhesive tape, while the content of the epoxy resin compound in the UV-curable composition I used to form the intermediate adhesive film layer is generally within a range of 15-30%, and more preferably within 20-30%. This not only ensures that the intermediate adhesive film layer have a certain structural strength, but also ensures that the curing speed of the adhesive film meets the actual requirements.

Unbound by specific theories, it is believed that, as mentioned above, the relatively low content of the ethylene-vinyl acetate copolymer and relatively high content of epoxy in the surface adhesive film layers can provide a strong interfacial effect, thus achieving higher initial adhesiveness and bonding strength after curing. At the meantime, the relatively low content of epoxy and relatively high content of the ethylene-vinyl acetate copolymer in the intermediate adhesive film layer enable the adhesive film to have good energy absorption and cushioning effects, thus forming a structure having three layers with different modulus, i.e., surface adhesive film layer/intermediate adhesive film layer/surface adhesive film layer, which can impart good impact resistance to the cured adhesive film.

c) Hydroxyx-containing compound

The UV-curable compositions used to form the multilayer UV-curable adhesive film in the present invention comprise hydroxyl-containing compounds, which act as chain transfer agents when the epoxy groups in the epoxy resin compounds react according to a cationic mechanism.

The hydroxyl-containing compounds that can be used for the present invention include ether or ester derivatives of such hydroxyl-containing compounds. In some preferred embodiments, the hydroxyl-containing compounds may be polyol compounds. Examples of the polyols that can be used include but are not limited to: polyether polyols such as polyether diols; polyester polyols such as polyester diols; bisphenol A polyols, etc. One or mixture of more of the aforementioned polyols may be used.

The hydroxyl-containing compounds that can be used for the present invention may be commercially obtained, and such as TONE 0230 Polyl, VORANOL 230-238 and VORANOL 2070 from the Dow Chemical Company (USA); Dianol 285 from J4T Seppic Company (France), etc. In some embodiments, VORANOL 2070 from the Dow Chemical Company is used, which is a polyether diol with a molecular weight of 700 grams/mol.
In the present invention, the content range of the hydroxyl-containing compounds in the UV-curable compositions I and II is 0.3-8.0 wt. %, more preferably 1.0-6.0 wt. %, and further preferably 2.0-4.0 wt. % (respectively based on the total weight of the UV-curable composition I or II). The inventor of the present invention has found that, only when the content of the hydroxyl-containing compounds respectively existing in the UV-curable compositions I and II of the present invention is within the above range, can the obtained adhesive film or tape have the above balanced performance. Moreover, if the content of the polyol compound is too low, the curing speed of the UV-induced pressure-sensitive adhesive tape is slow after induction by UV and the cured adhesive tape is brittle. On the other hand, if the content of the polyol is too high, the cured adhesive tape is too soft, which affects the shear strength.

d) Photoinitiator

The UV-curable compositions used to form the multilayer UV-curable adhesive film in the present invention comprise photoinitiators. Although the amount of the photoinitiators in the UV-curable compositions of the adhesive film is small, it has a great influence on the curing speed and storage stability of the UV-curable compositions of the adhesive film.

The photoinitiators that can be used for the present invention may be at least one selected from cationic photoinitiators. Cationic photoinitiators that can be used include, but are not limited to: diazo salts, iodonium salts, sulfonium salts, antimony onium salts and iron aromatic hydrocarbon salts. Specific examples thereof include diaryl iodonium salts, diaryl sulfonium salts, alkyl sulfonium salts, iron aromatic hydrocarbon salts, sulfonyloxanone and diaryl siloxane. In some embodiments, diaryl sulfonium hexafluorophosphate or hexafluoroantimonate is used. Such photoinitiators may be commercially obtained, such as DOUBLECURE 1176 from Taiwan Double Bond Chemical Ind., Co., Ltd.

In the present invention, the content range of the photoinitiators such as cationic photoinitiators in the above UV-curable compositions I and II is respectively within a range of 0.5-5%, and preferably within 0.75-2.5% (respectively based on the total weight of UV-curable composition I or II). Generally speaking, as the content of the photoinitiators such as cationic photoinitiators increases, the curing speed of the UV-curable compositions increases, but if the content is too high, the curing speed is too fast and it can be curved even under sunlight or fluorescent lamp light (containing a small amount of UV), which will cause some performance of the cured adhesive film or tape to be impaired, such as poor storage stability at room temperature. When the amount of the used cationic photoinitiators is too small, the requirement on ultraviolet radiation energy is high during and the curing speed is slow. At the same time, some performance of the cured adhesive film or tape will be impaired.
e) Antioxidant

The UV-curable compositions I and/or II used to form the multilayer UV-curable adhesive film comprise antioxidants. The used antioxidants can prevent the polymer from being oxidized in the process of hot melt extrusion of the UV-curable compositions I and/or II of the adhesive film in the present invention.

The antioxidants that can be used for the present invention include, but are not limited to: phenolic antioxidants such as alkyl monophenols, alkyl polyphenols and thiopolyphenols. Such antioxidants are known in the field, may be commercially obtained, and such as antioxidant 264, IRGANOX 1076, IRGANOX 2246 and IRGANOX 1010 from BASF Corporation.

In the present invention, the content range of the antioxidants in the above UV-curable compositions I and II is respectively 0.5-2% (respectively based on the total weight of the UV-curable compositions I or II). If the amount of the antioxidant is too small, such as smaller than 0.5%, it will not have a good anti-oxidizing effect; and if the amount of the antioxidant is too great, such as greater than 2%, it will impair other performance.

Other components

As known by one skilled in the art, the UV-curable compositions I and II in the adhesive film provided by the present invention may contain other components or additives known in the field according to actual application requirements. There is no special limitation to the types and content of these other components, as long as the required performance of the UV-curable compositions in the adhesive film provided by the present invention is not affected. In one preferred embodiment, the UV-curable compositions I and II in the present invention may independently further comprise one or more selected from an electric conducting agent, a heat conducting agent, a flame retardant, and a filler. In one further preferred embodiment, the electric conducting agent may be, for example, electric conducting particles or fibers (e.g. 2-45 wt. %), the heat conducting agent is heat conducting particles or fibers (e.g. 2-45 wt. %), the flame retardant may be, for example, zinc borate (e.g. 2-30 wt. %), and the filler may be silicon dioxide (e.g. 0.5-8 wt. %) such as amorphous silica, wherein the content of each component is based on the total weight of the corresponding UV-curable composition I or II.

In one preferred embodiment, the UV-curable compositions I and II used in the present invention do not contain solvents, especially organic solvents. In this case, the UV-curable compositions I and II in the present invention may be in the form of powder or particle mixture, for example. Such UV-curable compositions I and II may be prepared by, for example, simply mixing components in a mixing container or machine.
Adhesive film/tape

In the present invention, an adhesive film/tape may be formed by using the UV-curable compositions I and II of the present invention. Preferably, an adhesive film/tape may be formed through hot extrusion or melt extrusion, the adhesive film/tape comprising an intermediate adhesive film layer and surface adhesive film layers respectively formed by the UV-curable composition I and the UV-curable composition II. More specifically, the UV-curable composition I may be extruded onto a flexible and non-flexible substrate (including a release film or release paper) by hot extrusion or melt extrusion to form the intermediate adhesive film layer, and then the UV-curable composition II may be extruded onto both sides of the intermediate adhesive film layer by hot extrusion or melt extrusion to form two surface adhesive film layers, thereby forming the required adhesive film in the present invention.

As one example, FIG. 1 is a double-sided adhesive tape structural diagram of a multilayer UV-curable adhesive film/tape (excluding a substrate) obtained according to one embodiment of the present invention. As illustrated in FIG. 1, the adhesive film/tape comprises an intermediate layer or an intermediate adhesive film layer 1 formed by the UV-curable composition I, upper and lower surface adhesive film layers 2 and 3 on two sides of the intermediate layer or the intermediate adhesive film layer 1, and release films or release paper 4 and 5 respectively disposed on the surface adhesive film layers 2 and 3, wherein the two surface layers or surface adhesive film layers 2 and 3 on the two sides of the intermediate adhesive film layer 1 may be fully the same or different in composition (such as component content), and the release films or release paper 4 and 5 on the sides of the surface adhesive film layers may be the same or different in type or other aspects. The adhesive film/tape structure provided by the present invention is not limited to the form illustrated in the drawing. For example, a substrate may be provided on one surface adhesive film layer, or release paper or a release film may be provided only on one surface adhesive film layer of the adhesive film. As the release films or release paper, release films or release paper known in the prior art, such as PET release films, glass paper, laminated paper, and polypropylene thin films, may be used.

In addition to the double-sided adhesive tape, the present invention may also provide a single-sided adhesive tape by replacing the release paper or release film on one side of the adhesive film with a bonding substrate. Such a substrate is already known in this field. Examples include but are not limited to: a polymer film, a woven or non-woven fabric, a metal foil, foam and a combination thereof.

In one preferred embodiment, the thickness of the intermediate adhesive film layer formed by the UV-curable composition I may be 300-800 μm, and the thickness of the two surface adhesive film layers on the two sides of the intermediate adhesive film layer may independently be 50-100 μm.
The adhesive film structure with the thicker intermediate layer and the thinner two surface layers can ensure the curing speed and impact resistance of the formed adhesive tape.

The multilayer UV-curable adhesive film provided by the present invention exhibits the features of the common pressure-sensitive adhesive tape at the initial stage of bonding. That is, the adhesive film of the present invention has initial tack, can bond the bonded objects by applying small pressure, and can be molded through die cutting; and the adhesive film can be cured into an adhesive tape with semi-structural strength to structural strength at room temperature after being induced by UV, and thus it can bond some plastic products such as PC, PMMA and ABS which are not resistant to high temperature. Moreover, the adhesive film provided by the present invention has no odor or low odor, so that it is especially suitable for the bonding between internal parts of household electric appliances and automobiles, and between plastic parts and plastic parts, or between plastic parts and metal parts in electronic products such as hand-held mobile terminals.

EXAMPLES

The present invention will be further described below in detail with reference to the examples and the comparative examples. It should be understood that the present invention is not limited to the following examples.

In the following examples and comparative examples, unless otherwise specified, “parts” refers to “parts by weight”, “%” refers to “wt. %”, and “g” refers to weight unit “gram”. In the present invention, unless otherwise indicated, the used reagents are all commercially available and are used directly without being further purified.

Table 1 List of raw materials

<table>
<thead>
<tr>
<th>Product name</th>
<th>Description</th>
<th>Supplier</th>
</tr>
</thead>
<tbody>
<tr>
<td>LEVAPREN 600</td>
<td>Non-cross-linked ethylene-vinyl acetate copolymer, content of vinyl acetate units: 60 wt.%</td>
<td>LANXESS</td>
</tr>
<tr>
<td>LEVAPREN 700</td>
<td>Non-cross-linked ethylene-vinyl acetate copolymer, content of vinyl acetate units: 70 wt.%</td>
<td>LANXESS</td>
</tr>
<tr>
<td>LEVAPREN 800</td>
<td>Non-cross-linked ethylene-vinyl acetate copolymer, content of vinyl acetate units: 80 wt.%</td>
<td>LANXESS</td>
</tr>
<tr>
<td>LEVAPREN 900</td>
<td>Non-cross-linked ethylene-vinyl acetate copolymer, content of vinyl acetate units: 90 wt.%</td>
<td>LANXESS</td>
</tr>
<tr>
<td>LEVAPREN 700 XL</td>
<td>Pre-cross-linked ethylene-vinyl acetate copolymer, content of vinyl acetate units: 70 wt. %</td>
<td>LANXESS</td>
</tr>
<tr>
<td>LEVAPREN 800 XL</td>
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<td>Polyether polyol</td>
<td>Dow Chemical Company</td>
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<tr>
<td>DOUBLECURE 1176</td>
<td>Photoinitiator</td>
<td>Taiwan Double Bond Chemical Ind., Co., Ltd.</td>
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</tbody>
</table>

**Test methods**

The 180-degree peeling force, cohesive force (dynamic shear strength) and failure mode of various multilayer UV-curable adhesive films prepared in the examples and comparative examples were tested according to the specific methods listed below.

**Odor detection**

The odors of the obtained adhesive films/tapes were assessed by artificial olfaction and were considered as qualified when there was no any irritating odor (i.e., no odor or low odor).

**180° peeling force**

The adhesive film samples not irradiated by UV and obtained in the examples or the comparative examples were cut into adhesive strips of 12.7 mm, the adhesive strips were attached to a stainless steel plate, which was rolled back and forth once by using a 2 Kg roller (machine: Rolldown, purchased from Cheminstruments, USA). After the preparation, the stainless steel plate to which the adhesive samples were attached was allowed to stand at room temperature for 20 minutes; and then the peeling force was tested at a peeling speed of 305 mm/min (machine: Instron 3300, purchased from Instron, USA). The measurements of 5 peeling force tests were recorded and the average value was taken as 180° peeling force (unit: N/mm), i.e., initial tack.

**Dynamic shear strength**

(1) UV triggered curing test

The multilayer UV-curable adhesive films/tapes obtained in the examples or comparative examples were cut into adhesive strips of 25.4 mm, the release paper was removed, one adhesive
surface was attached to a standard test steel plate, the adhesive surface on the test steel plate was placed upward, a UV-LED ultraviolet lamp (Model KT403) of AVENTK was used to control the UV irradiation amount at 1J/cm² (8 min), the standard test steel plate was laminated with another standard test steel plate in 5 min, which was rolled back and forth once by using a 2 kg roller (machine: Rolldown, purchased from Cheminstruments, USA), and it was stored for 48 h at constant temperature of 23°C and constant humidity of 50% before testing. The specific procedure is performed in accordance with ASTM D3330.

(2) Dynamic shear strength test

According to the method described in FINAT FTM 2 (FINAT Technical Manual Test Method, 8th Edition) (FTM 2 is equivalent to the second test method), the above stored samples were tested and the shear strength was obtained. The used instrument was Instron 3300 of Instron, USA. The measurements of 5 shear strength tests were recorded and the average value was taken as dynamic shear strength (unit: MPa).

Failure modes

The failure modes of the adhesive film are defined as follows. Interfacial failure after the dynamic shear test: after the test, the adhesive film completely fell off from the bonding surface. Cohesive failure: after the test, the adhesive film was damaged from the intermediate layer. Adhesive film failure: after the test, the surface layers and the intermediate layer were peeled off. Cohesive failure and interfacial failure indicate that the effect is good, and the adhesive film failure mode indicates that the effect is not good.

Storage stability

The prepared adhesive film was sealed and packed in a black plastic bag, stored for one year at room temperature, and then cured under the induction by UV. Then the shear strength of the adhesive film/tape with respect to the standard test steel plate was tested.

Preparation of surface adhesive film layers and intermediate adhesive film layer

The intermediate adhesive film layer and the two surface adhesive film layers on the two sides (hereinafter referred to as the first surface adhesive film layer and the second surface adhesive film layer respectively for the sake of clarity) were prepared through hot melt extrusion, wherein, according to the designated components for the surface layers on the two sides, the components such as ethylene-vinyl acetate copolymer, epoxy resin compound, hydroxyl-containing compound, photoinitiator and antioxidant were fully mixed at 155°C in a CPM-40 double-screw extruder produced by CPM and then were extruded onto the release film, and wherein, according to the designated components for the intermediate layer, the components such as ethylene-vinyl
acetate copolymer, epoxy resin compound, hydroxyl-containing compound, photoinitiator and antioxidant were fully mixed at 155°C in a CPM-40 double-screw extruder produced by CPM, then were extruded and then attached to the surface layers on the two sides.
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<th>Layer 3</th>
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Table 3 Formula of surface (adhesive film) layers

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<td>Example 4</td>
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<td>Inter-facial failure</td>
<td>Inter-facial failure</td>
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</table>
Firstly, for the three-layer adhesive film/tape structures prepared through the hot melt extrusion method in the above examples and comparative example, all of them are odorless or low-odor qualified products according to odor detection, which means that these adhesive film/tape products can be applied to the bonding between internal parts of household electric appliances and automobiles, and between plastic parts and plastic parts, or between plastic parts and metal parts in electronic products such as hand-held mobile terminals.

Secondly, the following results regarding the adhesive films/tapes in examples 1-8 are shown in Tables 1-5 above: the intermediate layer and two surface layers, which are formed respectively by using the UV-curable compositions I and II comprising the specific components with specific content required by the present invention, achieve a good balance among initial tack, peeling force after curing and storage stability. In contrast, the adhesive films/tapes formed in comparative examples 1-14 by using the UV-curable compositions not comprising the specific components required by the present invention, cannot achieve a good balance in different areas.

More specifically, in example 1, an ethylene-vinyl acetate copolymer with 60% of vinyl acetate units is used. Because the content of the vinyl acetate units is low, the glass transition temperature of the copolymer is relatively low, which makes the cured adhesive film have a relatively low modulus, resulting that the adhesive film failure mode is cohesive failure.

In examples 2 and 3, the content of the vinyl acetate units in the used ethylene-vinyl acetate copolymer is increased (i.e. relatively high), and the modulus of the cured adhesive film is increased gradually, resulting that the dynamic shear strength is increased accordingly and the failure mode is interfacial failure. At the same time, since the ethylene-vinyl acetate copolymer used in examples 1, 2 and 3 are non-cross-linked, the adhesive films before curing have 180° peeling force (initial tack) greater than 0.48N/mm.

In example 4, the content of the vinyl acetate units in the used ethylene-vinyl acetate copolymer is 90% (i.e. the maximum content), resulting that the glass transition temperature of the EVA polymer is higher, the modulus of the cured adhesive film is higher, and the adhesive film after curing has very high shear strength.

Pre-cross-linked ethylene-vinyl acetate copolymers are used in examples 5 and 6 because the pre-cross-linked ethylene-vinyl acetate copolymers have higher modulus. Compared with examples 2 and 3, the pre-cross-linked ethylene-vinyl acetate copolymers help to improve the shear strength after curing.
In example 7, the adhesive film obtained by using the ethylene-vinyl acetate copolymer with 70% of vinyl acetate units for forming the surface layers and using the ethylene-vinyl acetate copolymer with 80% of vinyl acetate units for forming the intermediate layer also has better shear strength after curing.

In example 8, the surface layers formed by using the ethylene-vinyl acetate copolymer with 80% of vinyl acetate units and the intermediate layer formed by using the ethylene-vinyl acetate copolymer with 80% of vinyl acetate units can obtain dynamic shear strength greater than 9.0 MPa.

In comparative examples 1 and 2, because of the use of the surface layers with too low or too high content of epoxy resin compound, very good dynamic shear strength cannot be obtained.

In comparative examples 3 and 4, because of the use of the intermediate layer with too low or too high content of epoxy resin compound, very good dynamic shear strength cannot be obtained.

In comparative examples 5, 6, 7 and 8, the intermediate layer and surface layers with too low or too high content of initiator are used. Too low content of initiator makes the adhesive film very difficult to cure; and too high content of initiator causes the adhesive film to lose adhesiveness after irradiation by light. In addition, because the content of vinyl acetate units in the ethylene-vinyl acetate copolymers used in comparative examples 5, 6, 7 and 8 is 80%, the glass transition temperature is lower than that when the content of the vinyl acetate units is 90%.

In comparative examples 9, 10, 11 and 12, too low or too high content of hydroxyl-containing compounds are used. Too low content of hydroxyl-containing compounds make the curing speed slower and the shear strength of the adhesive film cannot reach a good level within a certain period of time; and too high content of hydroxyl-containing compounds causes the decrease of the overall modulus of the adhesive film, and the shear modulus of the adhesive film will also decrease.

In comparative example 13, only the intermediate layer and one surface layer are used to form the adhesive film sample. Because the content of epoxy in the intermediate layer is low, it cannot provide good bonding performance, resulting low dynamic shear strength.

Comparative example 14 is an adhesive film with only two surface layers. Although the adhesive film has shear strength greater than 10 MPa, there is no intermediate layer with a lower modulus. The adhesive film failure mode during dynamic shearing is adhesive film failure, resulting that the adhesive film does not have good energy absorption performance.

After being sealed and packed in a black plastic bag and stored for one year at room temperature, the adhesive film samples in examples 1-8 can still maintain very good dynamic shear strength. This result indicates that
the UV-curable adhesive film has good storage stability. On the contrary, the storage stability cannot be measured (i.e. "NA") because adhesive film failure occurs in comparative examples 4, 8-10 and 14. However, the adhesive film samples in comparative examples 1-3, 5-7 and 11-13, after being sealed and packed in a black plastic bag, do not have good storage stability, the dynamic shear strength is very poor. Therefore, “NA” is also used to express the storage stability test result.

The embodiment embodiments and examples of the present invention described above are just exemplary descriptions of the present invention, and are not intended to limit the concept and scope of the present invention. Various modifications and improvements may be made to the technical solution of the present invention by one skilled in the art without departing from the design concept of the present invention, which shall be all included in the protection scope of the present invention.
CLAIMS

1. A multilayer UV-curable adhesive film, comprising an intermediate adhesive film layer and two surface adhesive film layers respectively disposed on two sides of the intermediate adhesive film layer, the intermediate adhesive film layer being formed by a UV-curable composition I, and the surface adhesive film layers being formed by a UV-curable composition II, wherein based on a total weight of the UV-curable composition I, the UV-curable composition I comprises the following components:
   a) 70-85 wt. % of an ethylene-vinyl acetate copolymer;
   b) 15-30 wt. % of an epoxy resin compound;
   c) 0.3-8 wt. % of a hydroxyl-containing compound;
   d) 0.5-5 wt. % of a photoinitiator; and
   e) 0.5-2 wt. % of an antioxidant,
   and wherein based on a total weight of the UV-curable composition II, the UV-curable composition II comprises the following components:
   a) 30-70 wt. % of ethylene-vinyl acetate copolymer;
   b) 30-70 wt. % of epoxy resin compound;
   c) 0.3-8 wt. % of a hydroxyl-containing compound;
   d) 0.5-5 wt. % of a photoinitiator; and
   e) 0.5-2 wt. % of antioxidant.

2. The multilayer UV-curable adhesive film according to claim 1, wherein in the ethylene-vinyl acetate copolymers of the UV-curable compositions I and II, the content of vinyl acetate units is 60-90 wt. %.

3. The multilayer UV-curable adhesive film according to claim 1 or 2, wherein the ethylene-vinyl acetate copolymer is a pre-cross-linked polymer or a non-cross-linked linear polymer, and the ethylene-vinyl acetate copolymers in the UV-curable compositions I and II are the same or different.

4. The multilayer UV-curable adhesive film according to claim 1, wherein the epoxy resin compounds in the UV-curable compositions I and II are one or more selected from aromatic epoxy resins or epoxidized polyolefins.
5. The multilayer UV-curable adhesive film according to claim 1, wherein the UV-curable composition I comprises 20-30 wt. % of the epoxy resin compound based on the total weight of the UV-curable composition I, and the UV-curable composition II comprises 30-50 wt. % of the epoxy resin compound based on the total weight of the UV-curable composition II.

6. The multilayer UV-curable adhesive film according to claim 1 or 5, wherein the epoxy equivalent weights of the epoxy resin compounds in the UV-curable compositions I and II are within a range of 150-600 grams.

7. The multilayer UV-curable adhesive film according to claim 1 or 5, wherein the epoxy resin compounds in the UV-curable compositions I and II are the same or different in type and/or epoxy equivalent weight.

8. The multilayer UV-curable adhesive film according to claim 1, wherein the hydroxyl-containing compounds in the UV-curable compositions I and II are polyester polyols or polyether polyols.

9. The multilayer UV-curable adhesive film according to claim 1, wherein the photoinitiators in the UV-curable compositions I and II are cationic photoinitiators.

10. The multilayer UV-curable adhesive film according to claim 9, wherein the cationic photoinitiators are one or more selected from diazo salts, iodonium salts, sulfonium salts, antimonyonium salts and iron aromatic hydrocarbon salts.

11. The multilayer UV-curable adhesive film according to claim 1, wherein the UV-curable compositions I and II independently further comprise one or more selected from an electric conducting agent, a heat conducting agent, a flame retardant, and a filler.

12. The multilayer UV-curable adhesive film according to claim 11, wherein the electric conducting agent is an electric conducting particle or fiber, the heat conducting agent is heat conducting particles or fibers, and the filler is silicon dioxide.
13. The multilayer UV-curable adhesive film according to claim 2 or 5, wherein the UV-curable compositions I and II do not contain solvents.

14. The multilayer UV-curable adhesive film according to claim 1, wherein a thickness of the intermediate adhesive film layer is 300-800 μm.

15. The multilayer UV-curable adhesive film according to claim 1, wherein a thickness of the two surface adhesive film layers is independently 50-100 μm.

16. The multilayer UV-curable adhesive film according to claim 1, wherein the intermediate adhesive film layer and the two surface adhesive film layers are formed through hot melt extrusion.

17. The multilayer UV-curable adhesive film according to claim 1, wherein the multilayer UV-curable adhesive film further comprises a release paper or a release film disposed on one or two of the two surface adhesive film layers.
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*Fig. 1*
**INTERNATIONAL SEARCH REPORT**

### A. CLASSIFICATION OF SUBJECT MATTER

INV. C09J7/30  C09J7/20  C09J7/38

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

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)

EP0-Internal

### C. DOCUMENTS CONSIDERED TO BE RELEVANT

<table>
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<th>Citation of document, with indication, where appropriate, of the relevant passages</th>
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<td>paragraphs [0012] - [0022], [0033], [0037] - [0040], [0042], [0043], [0065]; claims 1-8; figure 3; example 4</td>
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- Further documents are listed in the continuation of Box C.
- X See patent family annex.

**Date of the actual completion of the international search**

25 June 2020

**Date of mailing of the international search report**

10/07/2020

**Name and mailing address of the ISA/ European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-3040, Fax: (+31-70) 340-3016**

Sperry, Pascal
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<tr>
<td></td>
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<td>DE 69901827 T2</td>
<td>06-02-2003</td>
</tr>
<tr>
<td></td>
<td></td>
<td>EP 1073697 A1</td>
<td>07-02-2001</td>
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<tr>
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<td></td>
<td>JP 2002513841 A</td>
<td>14-05-2002</td>
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<td></td>
<td>US 6057382 A</td>
<td>02-05-2000</td>
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<tr>
<td></td>
<td></td>
<td>US 6136384 A</td>
<td>24-10-2000</td>
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<tr>
<td></td>
<td></td>
<td>US 6153302 A</td>
<td>28-11-2000</td>
</tr>
<tr>
<td></td>
<td></td>
<td>WO 9957216 A1</td>
<td>11-11-1999</td>
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