

## DESCRIPTION

TITLE OF THE INVENTION: PRESSURE-SENSITIVE ADHESIVE LAYER FOR FLEXIBLE IMAGE DISPLAY DEVICES, LAMINATE FOR FLEXIBLE IMAGE DISPLAY DEVICES, AND FLEXIBLE IMAGE DISPLAY DEVICE

TECHNICAL FIELD

[0001]

The present invention relates to a pressure-sensitive adhesive layer for a flexible image display device. The present invention further relates to a laminate for a flexible image display device to which, for example, an optical film including at least a polarizer is applied together with the pressure-sensitive adhesive layer for a flexible image display device. The present invention furthermore relates to a flexible image display device in which the laminate for a flexible image display device is disposed.

[0002]

As an organic EL display device integrated with a touch sensor is shown in Fig. 1, an optical laminate 20 is provided on the viewing side of an organic EL display panel 10, and a touch panel 30 is provided on the viewing side of the optical laminate 20. The optical laminate 20 includes a polarizer 1 having protective films 2-1 and 2-2 bonded on both sides thereof and a retardation film 3, and the

polarizer 1 is provided on the viewing side of the retardation film 3. Further, in the touch panel 30, transparent conductive films 4-1 and 4-2 having a structure in which base material films 5-1 and 5-2 and transparent conductive layers 6-1 and 6-2 are laminated are disposed with a spacer 7 interposed therebetween (see, for example, Patent Document 1).

[0003]

It is also expected to realize a foldable organic EL display device which is more excellent in portability. For example, for a pressure-sensitive adhesive layer applied to an organic EL display device and the like, it has been proposed to improve flexibility and the like by controlling a storage elastic modulus  $G'$  (see, for example, Patent Documents 2 to 4).

#### PRIOR ART DOCUMENTS

#### PATENT DOCUMENTS

[0004]

Patent Document 1: JP-A-2014-157745

Patent Document 2: JP-A-2016-108555

Patent Document 3: JP-A-2017-095657

Patent Document 4: JP-A-2017-095659

#### SUMMARY OF THE INVENTION

## PROBLEMS TO BE SOLVED BY THE INVENTION

[0005]

The conventional organic EL display device as disclosed in Patent Document 1 is not designed with bending in mind. When a plastic film is used for a base material of an organic EL display panel, bendability can be imparted to the organic EL display panel. In addition, even when the plastic film is used for a touch panel and incorporated in the organic EL display panel, bendability can be imparted to the organic EL display panel. However, a problem of hindering the bendability of the organic EL display device occurs due to an optical laminate formed by laminating a conventional polarizer, or the likes on the organic EL display panel.

[0006]

In conventional organic EL display devices, there are problems that when the organic EL display device is repeatedly bent at normal temperature, a minute distortion occurs between layers such as an optical film and a pressure-sensitive adhesive layer constituting the organic EL display device and, in each layer, and peeling or cracking (breaking) occurs. Furthermore, in addition to the problem at normal temperature, when bend is performed in a high temperature environment, the pressure-sensitive adhesive layer may be coagulated and broken, and peeling

tends to be remarkable.

[0007]

In Patent Document 2, from the viewpoint of bending resistance and heat resistance, it has been proposed to use a pressure-sensitive adhesive layer having a storage elastic modulus  $G'$  at  $-20^{\circ}\text{C}$  of  $1 \times 10^5$  Pa or less and a storage elastic modulus  $G'$  at  $85^{\circ}\text{C}$  of  $1 \times 10^4$  Pa or more. In Patent Document 3, from the viewpoint of adhesive strength and bending resistance at low temperature, it has been proposed to use a pressure-sensitive adhesive layer having a storage elastic modulus  $G'$  at  $23^{\circ}\text{C}$  of  $3.0 \times 10^5$  Pa or less. In Patent Document 4, from the viewpoint of bending resistance at low temperature, it has been proposed to use a pressure-sensitive adhesive layer having a storage elastic modulus  $G'$  at  $-20^{\circ}\text{C}$  of  $1.3 \times 10^5$  Pa or more,  $1 \times 10^6$  Pa or less, and  $3.0 \times 10^5$  Pa or less. However, even with the pressure-sensitive adhesive layer satisfying the storage elastic modulus  $G'$  in the above range, none of the pressure-sensitive adhesive layers can satisfy the bending resistance from low temperatures to high temperatures.

[0008]

Therefore, an object of the present invention is to provide a pressure-sensitive adhesive layer for a flexible image display device that can satisfy bending resistance from low temperatures to high temperatures.

[0009]

The present invention further provides a laminate for a flexible image display device to which an optical film including at least a polarizing film is applied together with the pressure-sensitive adhesive layer for a flexible image display device, and furthermore provides a flexible image display device in which the laminate for a flexible image display device is disposed.

#### MEANS FOR SOLVING THE PROBLEM

[0010]

As a result of active investigations for solving the problems, the inventors have found the pressure-sensitive adhesive layer for a flexible image display device and the like described below, so that the invention has been completed.

[0011]

The present invention relates to a pressure-sensitive adhesive layer for a flexible image display device, having a storage elastic modulus  $G'$  at  $-20^{\circ}\text{C}$  of  $3.5 \times 10^4$  to  $1.7 \times 10^5$  Pa and a storage elastic modulus  $G'$  at  $23^{\circ}\text{C}$  of  $1.0 \times 10^4$  to  $5.0 \times 10^4$  Pa, and a difference between the storage elastic modulus  $G'$  at  $23^{\circ}\text{C}$  and the storage elastic modulus  $G'$  at  $85^{\circ}\text{C}$  being  $5.2 \times 10^3$  Pa or more.

[0012]

In the pressure-sensitive adhesive layer for a flexible image display device, an average value of the storage elastic modulus  $G'$  at  $-20^{\circ}\text{C}$  and the storage elastic modulus  $G'$  at  $23^{\circ}\text{C}$  is preferably  $4.5 \times 10^4$  to  $1.5 \times 10^5$  Pa.

[0013]

In the pressure-sensitive adhesive layer for a flexible image display device, a gel fraction is preferably 70% by weight or more.

[0014]

The pressure-sensitive adhesive layer for a flexible image display device is able to formed of a pressure-sensitive adhesive composition including a (meth)acrylic polymer containing an alkyl (meth)acrylate as a monomer unit.

[0015]

In the pressure-sensitive adhesive layer for a flexible image display device, the alkyl (meth)acrylate preferably includes an alkyl (meth)acrylate having an alkyl group having 10 or more carbon atoms.

[0016]

In the pressure-sensitive adhesive layer for a flexible image display device, the (meth)acrylic polymer preferably includes, as a monomer unit, an N-vinyl group-containing lactam monomer in addition to the alkyl (meth)acrylate.

[0017]

The present invention also relates to a laminate for a flexible image display device including:

the pressure-sensitive adhesive layer described above; and

an optical film containing at least a polarizer.

[0018]

The present invention also relates to a flexible image display device including:

the laminate for a flexible image display device; and

an organic EL display panel,

wherein the laminate for a flexible image display device is disposed on a viewing side with respect to the organic EL display panel.

[0019]

In the flexible image display device, a window is preferably disposed on the viewing side with respect to the laminate for a flexible image display device.

#### EFFECT OF THE INVENTION

[0020]

The pressure-sensitive adhesive layer laminated on a base material requires a stress that follows minute deformation of the adjacent base material. In particular, in bending in a high temperature environment, a cohesive

force of the pressure-sensitive adhesive layer decreases at high temperature, and it is conceivable that when bending is performed repeatedly, a distortion is repeatedly applied to the pressure-sensitive adhesive layer, causing cohesive fracture of the pressure-sensitive adhesive layer and causing peeling. Since the pressure-sensitive adhesive layer of the present invention has a predetermined storage elastic modulus at a predetermined temperature, distortion-stress can be dispersed in the pressure-sensitive adhesive layer even when exposed to a low temperature, normal temperature, or high temperature environment, and a distortion applied to the base material can be reduced. That is, the pressure-sensitive adhesive layer is easily deformed by a minute distortion, and a distortion applied to the other layers (each layer) can be reduced. As a result, in the pressure-sensitive adhesive layer of the present invention, the base material does not crack, peel or break even when bending is performed repeatedly, and the bending resistance can be satisfied. In addition, it is possible to prevent occurrence of peeling between the base material and the pressure-sensitive adhesive layer, and the pressure-sensitive adhesive layer can be suitably applied to applications of flexible image display devices. In particular, when the optical laminate is bent, a minute distortion is likely to occur in each layer, and there is a

high possibility that the base material cracks or peeling occurs between the base material and the pressure-sensitive adhesive layer. Therefore, the pressure-sensitive adhesive layer of the present invention is suitably applied.

#### BRIEF DESCRIPTION OF THE DRAWINGS

[0021]

Fig. 1 is a cross-sectional view showing a conventional organic EL display device.

Fig. 2 is a cross-sectional view showing a flexible image display device according to an embodiment of the present invention.

Fig. 3 is a cross-sectional view showing a flexible image display device according to another embodiment of the present invention.

Fig. 4 is a cross-sectional view showing a flexible image display device according to another embodiment of the present invention.

Fig. 5 is a view showing a bending test ((A) bending angle  $0^{\circ}$ , (B) bending angle  $180^{\circ}$ ).

Fig. 6 is a cross-sectional view showing a sample for evaluation used in examples.

Fig. 7 is a view showing a method of producing a retardation used in examples.

## MODE FOR CARRYING OUT THE INVENTION

[0022]

A pressure-sensitive adhesive layer for a flexible image display device of the present invention has a storage elastic modulus  $G'$  at  $-20^{\circ}\text{C}$  of  $3.5 \times 10^4$  to  $1.7 \times 10^5$  Pa and a storage elastic modulus  $G'$  at  $23^{\circ}\text{C}$  of  $1.0 \times 10^4$  to  $5.0 \times 10^4$  Pa, and a difference between the storage elastic modulus  $G'$  at  $23^{\circ}\text{C}$  and the storage elastic modulus  $G'$  at  $85^{\circ}\text{C}$  is  $5.2 \times 10^3$  Pa or more.

[0023]

As described above, the pressure-sensitive adhesive layer of the present invention satisfies the bending resistance in a low temperature environment by controlling the storage elastic modulus  $G'$  at  $-20^{\circ}\text{C}$  in the range of  $3.5 \times 10^4$  to  $1.7 \times 10^5$  Pa. The storage elastic modulus  $G'$  at  $-20^{\circ}\text{C}$  is preferably  $5 \times 10^4$  to  $1.6 \times 10^5$  Pa and more preferably  $7.0 \times 10^4$  to  $1.5 \times 10^5$  Pa, from the viewpoint of the bending resistance in a low temperature environment.

[0024]

Furthermore, in the pressure-sensitive adhesive layer of the present invention, the storage elastic modulus  $G'$  at  $23^{\circ}\text{C}$  is controlled in the range of  $1.0 \times 10^4$  to  $5.0 \times 10^4$  Pa, and the difference between the storage elastic modulus  $G'$  at  $23^{\circ}\text{C}$  and the storage elastic modulus  $G'$  at  $85^{\circ}\text{C}$  is controlled to be  $5.2 \times 10^3$  Pa or more, so that the bending

resistance in normal temperature and high temperature environments is satisfied. The range of the storage elastic modulus  $G'$  at 23°C is preferably designed to be lower than the storage elastic modulus  $G'$  at -20°C, and this is preferable in order to satisfy the bending resistance in a normal temperature environment. In addition, by designing the storage elastic modulus  $G'$  at 85°C to a range lower than the storage elastic modulus  $G'$  at 23°C by a predetermined amount, the bending resistance in a high temperature environment is satisfied. That is, in addition to controlling the storage elastic modulus  $G'$  at -20°C, it is possible to satisfy the bending resistance in a wide temperature range from low temperatures to high temperatures, which requires reliability assurance.

[0025]

The storage elastic modulus  $G'$  at 23°C is preferably  $1.3 \times 10^4$  to  $4.0 \times 10^4$  Pa and more preferably  $1.5 \times 10^4$  to  $3.5 \times 10^4$  Pa, from the viewpoint of the bending resistance in a normal temperature environment. The difference between the storage elastic modulus  $G'$  at 23°C and the storage elastic modulus  $G'$  at 85°C is preferably controlled to be  $5.3 \times 10^3$  Pa or more, and further  $5.5 \times 10^3$  Pa or more, from the viewpoint of the bending resistance in a high temperature environment. If the difference in storage elastic modulus  $G'$  is too large, fluidity of a pressure-

sensitive adhesive becomes high, and therefore, from the viewpoint of processing, the difference in storage elastic modulus  $G'$  is preferably controlled to be  $8.0 \times 10^3$  Pa or less, and further  $7.0 \times 10^3$  Pa or less.

[0026]

The storage elastic modulus  $G'$  at  $85^\circ\text{C}$  is preferably  $8.0 \times 10^3$  to  $3.4 \times 10^4$  Pa and more preferably  $1.0 \times 10^4$  to  $3.0 \times 10^4$  Pa, from the viewpoint of the bending resistance in a high temperature environment.

[0027]

In the pressure-sensitive adhesive layer of the present invention, from the viewpoint of satisfying the bending resistance in both low temperature and normal temperature environments, an average value of the storage elastic modulus  $G'$  at  $-20^\circ\text{C}$  and the storage elastic modulus  $G'$  at  $23^\circ\text{C}$  is preferably  $4.0 \times 10^4$  to  $1.5 \times 10^5$  Pa. The average value is more preferably  $4.5 \times 10^4$  to  $1.0 \times 10^5$  Pa.

[0028]

In addition, in the pressure-sensitive adhesive layer of the present invention, from the viewpoint of satisfying the bending resistance in any of low temperature, normal temperature, and high temperature environments, an average value of the storage elastic modulus  $G'$  at  $-20^\circ\text{C}$ , the storage elastic modulus  $G'$  at  $23^\circ\text{C}$ , and the storage elastic modulus  $G'$  at  $85^\circ\text{C}$  is preferably  $5.0 \times 10^4$  to  $4.0 \times 10^5$  Pa.

The average value is more preferably  $8.0 \times 10^4$  to  $3.0 \times 10^5$  Pa.

[0029]

By controlling the average value of the storage elastic modulus  $G'$  within the above range, a difference in elastic modulus for each temperature condition becomes small, which is preferable for exhibiting bending resistance in a wide temperature range.

[0030]

A gel fraction of the pressure-sensitive adhesive layer of the present invention is preferably 70% by weight or more, more preferably 70 to 95% by weight, still more preferably 80 to 90% by weight, even more preferably 82 to 90% by weight, and further more preferably 85 to 90% by weight. When the gel fraction of the pressure-sensitive adhesive layer is within the above range, the cohesive force of the pressure-sensitive adhesive layer can be increased, and appearance (glue dents, etc.), processability, durability, and flexibility are improved. In particular, the flexibility in normal temperature and high temperature environments can be easily simultaneously achieved, which is a preferred embodiment.

[0031]

Although a glass transition temperature ( $T_g$ ) of the pressure-sensitive adhesive layer of the present invention

is not particularly limited, the upper limit is preferably 5°C or lower. Considering the flexibility in a low temperature environment or in a high-speed region, the glass transition temperature is more preferably -20°C or less, and still more preferably -25°C or less. If T<sub>g</sub> of the pressure-sensitive adhesive layer is within such a range, it is difficult for the pressure-sensitive adhesive layer to become hard even at the time of bending in a low temperature environment and in such a high-speed region where a bending rate exceeds 1 second/time, and it is possible to achieve a laminate for a flexible image display device which is excellent in stress relaxation property and is bendable or foldable, and a flexible image display device in which the laminate for a flexible image display device is disposed. The glass transition temperature (T<sub>g</sub>) is a theoretical value derived from Fox's equation.

[0032]

The total light transmittance (according to JIS K7136) in the visible light wavelength region of the pressure-sensitive adhesive layer used for the laminate for a flexible image display device of the present invention is preferably 85% or more and more preferably 90% or more.

[0033]

Hereinafter, a pressure-sensitive adhesive forming the pressure-sensitive adhesive layer of the present

invention, its composition, and the like will be described.

[0034]

Examples of the pressure-sensitive adhesive forming the pressure-sensitive adhesive layer of the present invention include an acrylic pressure-sensitive adhesive, a rubber-based pressure-sensitive adhesive, a vinyl alkyl ether-based pressure-sensitive adhesive, a silicone-based pressure-sensitive adhesive, a polyester-based pressure-sensitive adhesive, a polyamide-based pressure-sensitive adhesive, a urethane-based pressure-sensitive adhesive, a fluorine-based pressure-sensitive adhesive, an epoxy-based pressure-sensitive adhesive, and a polyether-based pressure-sensitive adhesive. The pressure-sensitive adhesives constituting the pressure-sensitive adhesive layer is used alone or in combination of two or more. However, from the viewpoints of transparency, processability, durability, adhesiveness, bending resistance, etc., it is preferable to use an acrylic pressure-sensitive adhesive (composition) containing a (meth)acrylic polymer alone.

[0035]

<(Meth)acrylic Polymer>

The acrylic pressure-sensitive adhesive, which is used as the pressure-sensitive adhesive composition, preferably includes a (meth)acrylic polymer preferably

containing a (meth)acrylic monomer having a linear or branched alkyl group of 1 to 30 carbon atoms as a monomer unit. By using the (meth)acrylic monomer having a linear or branched alkyl group of 1 to 30 carbon atoms, a pressure-sensitive adhesive layer excellent in bendability can be obtained. In the present invention, the term “(meth)acrylic polymer” refers to an acrylic polymer and/or a methacrylic polymer, and the term “(meth)acrylate” refers to an acrylate and/or a methacrylate.

[0036]

Specific examples of the (meth)acrylic monomer having a linear or branched alkyl group of 1 to 30 carbon atoms forming the main skeleton of the (meth)acrylic polymer include methyl (meth)acrylate, ethyl (meth)acrylate, n-butyl (meth)acrylate, s-butyl (meth)acrylate, t-butyl (meth)acrylate, isobutyl (meth)acrylate, n-pentyl (meth)acrylate, isopentyl (meth)acrylate, n-hexyl (meth)acrylate, isohexyl (meth)acrylate, isoheptyl (meth)acrylate, 2-ethylhexyl (meth)acrylate, n-octyl (meth)acrylate, isooctyl (meth)acrylate, n-nonyl (meth)acrylate, isononyl (meth)acrylate, n-decyl (meth)acrylate, isodecyl (meth)acrylate, n-dodecyl (meth)acrylate (lauryl ), n-tridecyl (meth)acrylate, n-tetradecyl (meth)acrylate, etc.

[0037]

Among the alkyl (meth)acrylates, an alkyl (meth)acrylate having a linear or branched alkyl group having 6 to 30 carbon atoms (hereinafter may be referred to as "alkyl (meth)acrylate having a long-chain alkyl group") is preferable from the viewpoint of flexibility due to reduction of microdeformation stress. As the alkyl (meth)acrylate having a long-chain alkyl group, it is preferable to contain an alkyl (meth)acrylate having an alkyl group having 10 or more carbon atoms. In particular, as the alkyl (meth)acrylate having a long-chain alkyl group, n-dodecyl (meth)acrylate (lauryl (meth)acrylate) is more preferable. By using the alkyl (meth)acrylate having a long-chain alkyl group, entanglement of polymers is reduced, deformation is likely to occur with respect to minute distortions, and this is a preferable embodiment for flexibility. From the viewpoint of flexibility and adhesiveness at low temperature, an alkyl (meth)acrylate having a homopolymer glass transition temperature ( $T_g$ ) of -70 to  $-20^{\circ}\text{C}$  is preferably used, and an alkyl (meth)acrylate having an alkyl group having 6 or more and 9 or less carbon atoms, particularly 2-ethylhexyl acrylate is more preferably used. As the alkyl (meth)acrylates, one or two or more acrylates can be used. As the long-chain alkyl (meth)acrylate, it is preferable to use a mixture of an alkyl (meth)acrylate having an alkyl group having 10 or

more and 30 or less carbon atoms and an alkyl (meth)acrylate having an alkyl group having 6 or more and 9 or less carbon atoms. In the mixture, the mixed weight ratio is preferably (alkyl (meth)acrylate having an alkyl group having 10 or more and 30 or less carbon atoms) : (alkyl (meth)acrylate having an alkyl group having 6 or more and 9 or less carbon atoms) = 40 : 60 to 90 : 10. In the mixture of the alkyl (meth)acrylate having an alkyl group having 10 or more and 30 or less carbon atoms and the alkyl (meth)acrylate having an alkyl group having 6 or more and 9 or less carbon atoms, n-dodecyl (meth)acrylate and 2-ethylhexyl acrylate are preferably used in combination.

[0038]

The (meth)acrylic monomer having a linear or branched alkyl group of 1 to 24 carbon atoms is a main component in all the monomers forming the (meth)acrylic polymer. Here, as the main component, the total amount of (meth)acrylic monomer having a linear or branched alkyl group of 1 to 30 carbon atoms in all the monomers constituting the (meth)acrylic polymer is preferably 50 to 100% by weight, more preferably from 80 to 100% by weight, even more preferably from 90 to 99.9% by weight, particularly preferably from 94 to 99.9% by weight.

[0039]

The monomer component constituting the (meth)acrylic

polymer may contain a monomer which is copolymerizable (copolymerizable monomer) in addition to the alkyl (meth)acrylate having a linear or branched alkyl group having 1 to 30 carbon atoms. The copolymerizable monomer may be used alone or in combination of two or more.

[0040]

The copolymerizable monomer is not particularly limited, and a copolymerizable monomer having a reactive functional group of a polymerizable unsaturated double bond is preferable. As the copolymerizable monomer having the reactive functional group, a hydroxyl group-containing monomer is preferable. By using the hydroxyl group-containing monomer, a pressure-sensitive adhesive layer having excellent adhesiveness and flexibility can be obtained. The hydroxyl group-containing monomer contains a hydroxyl group in its structure and is a compound containing a polymerizable unsaturated double bond such as a (meth)acryloyl group and a vinyl group.

[0041]

Specific examples of the hydroxyl group-containing monomer include hydroxyalkyl (meth)acrylates such as 2-hydroxyethyl (meth)acrylate, 3-hydroxypropyl (meth)acrylate, 4-hydroxybutyl (meth)acrylate, 6-hydroxyhexyl (meth)acrylate, 8-hydroxyoctyl (meth)acrylate, 10-hydroxydecyl (meth)acrylate, 12-hydroxylauryl

(meth)acrylate, etc. and (4-hydroxymethylcyclohexyl)-methyl acrylate. Among the hydroxyl group-containing monomers, 2-hydroxyethyl (meth)acrylate and 4-hydroxybutyl (meth)acrylate are preferable from the viewpoints of adhesiveness and durability. As the hydroxyl group-containing monomers, one or two or more kinds of the monomers can be used.

[0042]

Examples of the copolymerizable monomer having a reactive functional group include a carboxyl group-containing monomer, an amino group-containing monomer, and an amide group-containing monomer, which having a reactive functional group. These monomers are preferable from the viewpoint of adhesiveness in a humidification environment or a high temperature environment.

[0043]

By using the carboxyl group-containing monomer, a pressure-sensitive adhesive layer having excellent adhesiveness in a humidification environment or a high temperature environment can be obtained. The carboxyl group-containing monomer contains a carboxyl group in its structure and is a compound containing a polymerizable unsaturated double bond such as a (meth)acryloyl group and a vinyl group.

[0044]

Specific examples of the carboxyl group-containing monomer include (meth)acrylic acid, carboxyethyl (meth)acrylate, carboxypentyl (meth)acrylate, itaconic acid, maleic acid, fumaric acid, crotonic acid, isocrotonic acid, and the like.

[0045]

By using the amino group-containing monomer, a pressure-sensitive adhesive layer having excellent adhesiveness in a humidification environment or a high temperature environment can be obtained. The amino group-containing monomer contains an amino group in its structure and is a compound containing a polymerizable unsaturated double bond such as a (meth)acryloyl group and a vinyl group.

[0046]

Specific examples of the amino group-containing monomer include aminoethyl (meth)acrylate, N,N-dimethylaminoethyl (meth)acrylate, N,N-dimethylaminopropyl (meth)acrylate, and the like.

[0047]

By using the amide group-containing monomer, a pressure-sensitive adhesive layer having excellent adhesiveness can be obtained. The amide group-containing monomer contains an amide group in its structure and is a compound containing a polymerizable unsaturated double bond

such as a (meth)acryloyl group and a vinyl group.

[0048]

Specific examples of the amide group-containing monomer include acrylamide-based monomers such as (meth)acrylamide, N,N-dimethyl (meth)acrylamide, N,N-diethyl (meth)acrylamide, N-isopropylacrylamide, N-methyl (meth)acrylamide, N-butyl (meth)acrylamide, N-hexyl (meth)acrylamide, N-methylol (meth)acrylamide, N-methylol-N-propane (meth)acrylamide, aminomethyl (meth)acrylamide, aminoethyl (meth)acrylamide, mercaptomethyl (meth)acrylamide, and mercaptoethyl (meth)acrylamide; N-acryloyl heterocyclic monomers such as N-(meth)acryloyl morpholine, N-(meth)acryloyl piperidine, and N-(meth)acryloyl pyrrolidine; N-vinyl group-containing lactam monomers such as N-vinylpyrrolidone and N-vinyl- $\epsilon$ -caprolactam; and the like.

[0049]

Among the copolymerizable monomers such as the carboxyl group-containing monomer, the amino group-containing monomer, and the amide group-containing monomer, the amide group-containing monomer is preferable, and an N-vinyl group-containing lactam monomer is particularly preferable. The N-vinyl group-containing lactam monomer is preferable from the viewpoint of improving the adhesiveness to an adherend, particularly the adhesiveness during

heating.

[0050]

As the copolymerizable monomer having a reactive functional group, it is preferable to use a hydroxyl group-containing monomer and an amide group-containing monomer (particularly, an N-vinyl group-containing lactam monomer) in combination.

[0051]

In a monomer unit constituting the (meth)acrylic polymer, a blending ratio of the above-exemplified copolymerizable monomer having a reactive functional group is preferably 20% by weight or less, more preferably 15% by weight or less, and still more preferably 10% by weight or less, in all monomers constituting the (meth)acrylic polymer.

[0052]

When the hydroxyl group-containing monomer is contained, the blending ratio is preferably 0.01 to 8% by weight, more preferably 0.01 to 5% by weight, and still more preferably 0.05 to 3% by weight.

[0053]

When a copolymerizable monomer such as the carboxyl group-containing monomer, the amino group-containing monomer, or the amide group-containing monomer is contained, the blending ratio is preferably 0.01 to 15% by

weight, more preferably 0.1 to 12% by weight, and still more preferably 0.1 to 10% by weight. In particular, the N-vinyl group-containing lactam monomer is preferably used in an amount of 3 to 12% by weight, and more preferably 4 to 10% by weight. If a proportion of N-vinyl group-containing lactam monomer becomes too large, the glass transition temperature rises, and the storage elastic modulus  $G'$  rises at normal temperature (23°C), which may cause peeling and cracking in the bending test. Therefore, it is preferable to use the N-vinyl group-containing lactam monomer in the above range.

[0054]

As the copolymerizable monomer, in addition to the above-exemplified copolymerizable monomer having a reactive functional group, other copolymerizable monomers can be used as long as the effects of the present invention are not impaired.

[0055]

Examples of the other copolymerizable monomer include alkoxyalkyl ester (meth)acrylates [e.g., 2-methoxyethyl (meth)acrylate, 2-ethoxyethyl (meth)acrylate, methoxytriethylene glycol (meth)acrylate, 3-methoxypropyl (meth)acrylate, 3-ethoxypropyl (meth)acrylate, 4-methoxybutyl (meth)acrylate, 4-ethoxybutyl (meth)acrylate]; epoxy group-containing monomers [e.g., glycidyl

(meth)acrylate and methyl glycidyl (meth)acrylate]; sulfonate group-containing monomers [such as sodium vinyl sulfonate]; phosphate group-containing monomers; (meth)acrylates having an alicyclic hydrocarbon group [e.g., cyclopentyl(meth)acrylate, cyclohexyl(meth)acrylate, isobornyl(meth)acrylate]; (meth)acrylates having an aromatic hydrocarbon group [e.g., phenyl(meth)acrylate, phenoxyethyl(meth)acrylate, benzyl(meth)acrylate]; vinyl esters [e.g., vinyl acetate, vinyl propionate]; aromatic vinyl compounds [e.g., styrene, vinyl toluene]; olefins or dienes [e.g., ethylene, propylene, butadiene, isoprene, isobutylene]; vinyl ethers [e.g., vinyl alkyl ether]; and vinyl chlorides.

[0056]

The blending ratio of the other copolymerizable monomers is not particularly limited, and is preferably 30% by weight or less, more preferably 10% by weight or less, and still more preferably zero, in all monomers constituting the (meth)acrylic polymer. When the blending ratio of the other copolymerizable monomers exceeds 30% by weight, in particular when a material other than the alkyl (meth)acrylate is used, the reaction point between the pressure-sensitive adhesive layer and the other layer (film, base material) tends to be small, and the adhesiveness tends to decrease.

[0057]

In the monomer component constituting the (meth)acrylic polymer, in addition to a monofunctional monomer having one reactive functional group of a polymerizable unsaturated double bond exemplified above, a polyfunctional monomer having a plurality of the reactive functional groups can be used.

[0058]

When the polyfunctional monomer is contained, a crosslinking effect can be obtained by polymerization, and the gel fraction can be easily adjusted and the cohesive force can be easily improved. Thus, cutting becomes easy, and processability is easily improved. In addition, at the time of bending (particularly in a high temperature environment), peeling due to cohesive fracture of the pressure-sensitive adhesive layer can be prevented. Examples of the polyfunctional monomer include, but are not particularly limited to, polyfunctional acrylates such as hexanediol di(meth)acrylate (1,6-hexanediol di(meth)acrylate), butanediol di(meth)acrylate, (poly)ethylene glycol di(meth)acrylate, (poly) propylene glycol di(meth)acrylate, neopentyl glycol di(meth)acrylate, pentaerythritol di(meth)acrylate, pentaerythritol tri(meth)acrylate, dipentaerythritol hexa(meth)acrylate, trimethylolpropane tri(meth)acrylate, tetramethylolmethane

tri(meth)acrylate, allyl (meth)acrylate, vinyl (meth)acrylate, epoxy acrylate, polyester acrylate, and urethane acrylate, and divinylbenzene. Among them, as the polyfunctional acrylate, 1,6-hexanediol diacrylate and dipentaerythritol hexa(meth)acrylate are preferable. The polyfunctional monomer may be used alone or in combination of two or more.

[0059]

The blending ratio of the polyfunctional monomer is preferably 10 parts by weight or less, more preferably 5 parts by weight or less, and still more preferably 3 parts by weight or less with respect to 100 parts by weight of the total amount of the monofunctional monomer constituting (meth)acrylic polymer. As the blending ratio of the polyfunctional monomer increases, the number of crosslinking points increases, and the flexibility of the pressure-sensitive adhesive (pressure-sensitive adhesive layer) is lost, so that the stress relaxation property tends to be poor.

[0060]

The pressure-sensitive adhesive layer is formed of a pressure-sensitive adhesive composition, and the pressure-sensitive adhesive composition may be a pressure-sensitive adhesive composition in any form, and examples thereof include emulsion-type, solvent-type (solution-type), active

energy ray-curable type, heat-fusing type (hot melt-type) and others. Among them, as the pressure-sensitive adhesive composition, solvent-type pressure-sensitive adhesive compositions and active energy ray-curable pressure-sensitive adhesive compositions are preferable.

[0061]

As the solvent-type pressure-sensitive adhesive composition, a pressure-sensitive adhesive composition containing the (meth)acrylic polymer as an essential component is preferable. Examples of the active energy ray-curable pressure-sensitive adhesive composition preferably include a mixture of the monomer components (monomer mixture) constituting the (meth)acrylic polymer, or the pressure-sensitive adhesive composition including as an essential component a partially polymerized substance of the mixture. The "partially polymerized substance" means a composition in which one or two or more components of the monomer components included in the monomer mixture are partially polymerized. The "monomer mixture" includes a case where there is only one type of monomer component.

[0062]

In particular, from the viewpoints of productivity, effect on the environment and easiness in obtaining a pressure-sensitive adhesive layer having a certain thickness, the pressure-sensitive adhesive composition is

preferably an active energy ray curable-type pressure-sensitive adhesive composition including, as an essential component, a mixture (monomer mixture) of the monomer components constituting the (meth)acrylic polymer or a partially polymerized substance of the mixture.

[0063]

The (meth)acrylic polymer is obtained by polymerizing the monomer component. More specifically, the (meth)acrylic polymer is obtained by polymerizing, with heretofore known conventional methods, the monomer components, the monomer mixture or the partially polymerized substance thereof. Examples of the polymerization method include solution polymerization, emulsion polymerization, bulk polymerization, and polymerization based on heat or active energy ray irradiation (thermal polymerization and active energy ray polymerization). Among these methods, from the viewpoint of transparency, water resistance, cost and others, the solution polymerization method and the active energy ray polymerization method are preferable. For the purpose of suppressing the inhibition of polymerization by oxygen, the polymerization is preferably performed by avoiding the contact with oxygen. For example, the polymerization is preferably performed in a nitrogen atmosphere, or by blocking oxygen with a release film (separator). The

(meth)acrylic polymer to be obtained may be any of a random copolymer, a block copolymer, a graft copolymer and the like.

[0064]

Examples of the active energy ray used for irradiation in the active energy ray polymerization (photopolymerization) include ionizing radiation such as  $\alpha$ -ray,  $\beta$ -ray,  $\gamma$ -ray, neutron ray and electron beam, and ultraviolet ray, in particular, ultraviolet ray is preferable. The irradiation energy, irradiation time, irradiation method and the like of the active energy ray are not particularly limited as long as the active energy ray can activate the photopolymerization initiator and allows the reaction of the monomer components to occur.

[0065]

In the solution polymerization, various common solvents can be used. Examples of such solvents include the following organic solvents: esters such as ethyl acetate and n-butyl acetate; aromatic hydrocarbons such as toluene and benzene; aliphatic hydrocarbons such as n-hexane and n-heptane; alicyclic hydrocarbons such as cyclohexane and methyl cyclohexane; and ketones such as methyl ethyl ketone and methyl isobutyl ketone. The solvent may be used alone or in combination of two or more.

[0066]

In the polymerization, according to the type of the polymerization reaction, a polymerization initiator such as a photopolymerization initiator (photoinitiator) or a thermal polymerization initiator may be used. The polymerization initiator may be used alone or in combination of two or more.

[0067]

The photopolymerization initiator is not particularly limited; however, examples of the photopolymerization initiator include benzoin ether-based photopolymerization initiators, acetophenone-based photopolymerization initiators,  $\alpha$ -ketol-based photopolymerization initiators, aromatic sulfonyl chloride-based photopolymerization initiators, optically active oxime-based photopolymerization initiators, benzoin-based photopolymerization initiators, benzil-based photopolymerization initiators, benzophenone-based photopolymerization initiators, ketal-based photopolymerization initiators and thioxanthone-based photopolymerization initiators.

[0068]

Examples of the benzoin ether-based photopolymerization initiators include benzoin methyl ether, benzoin ethyl ether, benzoin propyl ether, benzoin isopropyl ether, benzoin isobutyl ether, 2,2-dimethoxy-1,2-

diphenylethan-1-one and anisole methyl ether. Examples of the acetophenone-based photopolymerization initiators include 2,2-diethoxyacetophenone, 2,2-dimethoxy-2-phenylacetophenone, 1-hydroxycyclohexyl phenyl ketone, 4-phenoxydichloroacetophenone and 4-(t-butyl)-dichloroacetophenone. Examples of the  $\alpha$ -ketol-based photopolymerization initiators include 2-methyl-2-hydroxypropiophenone and 1-[4-(2-hydroxyethyl)-phenyl]-2-hydroxy-2-methylpropan-1-one. Examples of the aromatic sulfonyl chloride photopolymerization initiator include 2-naphthalenesulfonyl chloride. Examples of the photoactive oxime type photopolymerization initiator include 1-phenyl-1,1-propanedione-2-(o-ethoxycarbonyl)-oxime. Examples of the benzoin-based photopolymerization initiator include benzoin. Examples of the benzyl-based photopolymerization initiator include benzyl. Examples of the benzophenone type photopolymerization initiator include benzophenone, benzoylbenzoic acid, 3,3'-dimethyl-4-methoxybenzophenone, polyvinylbenzophenone, and  $\alpha$ -hydroxycyclohexyl phenyl ketone. Examples of the ketal-based photopolymerization initiator include benzyldimethyl ketal. Examples of the thioxanthone-based photopolymerization initiator include thioxanthone, 2-chlorothioxanthone, 2-methylthioxanthone, 2,4-dimethylthioxanthone, isopropylthioxanthone, 2,4-diisopropylthioxanthone, and dodecylthioxanthone.

[0069]

The used amount of the photopolymerization initiator is not particularly restricted, and is preferably 0.01 to 1 part by weight and more preferably 0.05 to 0.5 parts by weight with respect to 100 parts by weight of all the monomer components.

[0070]

Examples of the polymerization initiator used in the solution polymerization include azo polymerization initiators, peroxide polymerization initiators (such as dibenzoyl peroxide and tert-butyl permaleate) and redox polymerization initiators. Among these initiators, azo polymerization initiators disclosed in JP-A-2002-69411 are preferable. Examples of the azo polymerization initiator include 2,2'-azobisisobutyronitrile (AIBN), 2,2'-azobis-2-methylbutyronitrile, dimethyl 2,2'-azobis(2-methylpropionate) and 4,4'-azobis-4-cyanovalerianic acid.

[0071]

The used amount of the azo polymerization initiator is not particularly limited, and is preferably 0.05 to 0.5 parts by weight and more preferably 0.1 to 0.3 parts by weight with respect to 100 parts by weight of all the monomer components.

[0072]

The polyfunctional monomer (polyfunctional acrylate)

used as the copolymerizable monomer can also be used in a solvent-type or active energy ray-curable pressure-sensitive adhesive composition. For example, when the polyfunctional monomer (polyfunctional acrylate) and the photopolymerization initiator are mixed with the solvent-type pressure-sensitive adhesive composition and used, active energy ray curing is performed after heat drying. [0073]

In the present invention, as the (meth)acrylic polymer used in the solvent-type pressure-sensitive adhesive composition, such polymer usually having a weight average molecular weight (Mw) in the range of 1,000,000 to 2,500,000 is used. In consideration of durability, particularly heat resistance and flexibility, the weight average molecular weight is preferably from 1,200,000 to 2,000,000 and more preferably from 1,400,000 to 1,800,000. When the weight average molecular weight is smaller than 1,000,000, at the time of crosslinking the polymer chains with each other in order to ensure durability, the number of crosslinking points is increased to lose the flexibility of the pressure-sensitive adhesive (pressure-sensitive adhesive layer), compared with those having a weight average molecular weight of 1,000,000 or more, and as a result, the distortion of the outer bend side (convex side) and the inner bend side (concave side) occurring between

the layers (films) at the time of bending cannot be alleviated, and the layers tend to break easily. In addition, when the weight average molecular weight exceeds 2,500,000, a large amount of a diluting solvent is required for adjusting the viscosity for coating, which undesirably leads to an increase in cost, and since the entanglement of the polymer chains of the resulting (meth)acrylic polymer becomes complicated, the flexibility is inferior, and breakage of the layers (films) is likely to occur at the time of bending. The weight average molecular weight (Mw) is a value calculated in terms of polystyrene as measured by GPC (gel permeation chromatography).

[0074]

<(Meth)acrylic oligomer>

The pressure-sensitive adhesive composition may contain a (meth)acrylic oligomer. As the (meth)acrylic oligomer, it is preferable to use a polymer having a weight average molecular weight (Mw) smaller than that of the (meth)acrylic polymer. By using such a (meth)acrylic oligomer, the (meth)acrylic oligomer is interposed between the (meth)acrylic polymers to reduce entanglement of the (meth)acrylic polymers, which makes it easier to deform with respect to minute distortions, which is a preferable embodiment for flexibility.

[0075]

Examples of monomers constituting the (meth)acrylic oligomer include: an alkyl (meth)acrylate such as methyl (meth)acrylate, ethyl (meth)acrylate, propyl (meth)acrylate, isopropyl (meth)acrylate, butyl (meth)acrylate, isobutyl (meth)acrylate, sec-butyl (meth)acrylate, tert-butyl (meth)acrylate, pentyl (meth)acrylate, isopentyl (meth)acrylate, hexyl (meth)acrylate, 2-ethylhexyl (meth)acrylate, heptyl (meth)acrylate, octyl (meth)acrylate, isooctyl (meth)acrylate, nonyl (meth)acrylate, isononyl (meth)acrylate, decyl (meth)acrylate, isodecyl (meth)acrylate, undecyl (meth)acrylate, and dodecyl (meth)acrylate; an ester of (meth)acrylic acid and an alicyclic alcohol, such as cyclohexyl (meth)acrylate, isobornyl (meth)acrylate and dicyclopentanyl (meth)acrylate; an aryl (meth)acrylate such as phenyl (meth)acrylate and benzyl (meth)acrylate; and a (meth)acrylate derived from a terpene compound derivative alcohol. Such (meth)acrylates can be used alone or in combination of two or more.

[0076]

The (meth)acrylic oligomer preferably contains, as a monomer unit, an acrylic monomer having a relatively bulky structure, typified by an alkyl (meth)acrylate whose alkyl group has a branched structure, such as isobutyl

(meth)acrylate or tert-butyl (meth)acrylate; an ester of (meth)acrylic acid and an alicyclic alcohol, such as cyclohexyl (meth)acrylate, isobornyl (meth)acrylate or dicyclopentanyl (meth)acrylate; or an aryl (meth)acrylate such as phenyl (meth)acrylate or benzyl (meth)acrylate, or any other cyclic structure-containing (meth)acrylate. The use of a (meth)acrylic oligomer with such a bulky structure can further improve the tackiness of the pressure-sensitive adhesive layer. In terms of bulkiness, cyclic structure-containing oligomers are highly effective, and oligomers having two or more rings are more effective. When ultraviolet light is used in the process of synthesizing the (meth)acrylic oligomer or forming the pressure-sensitive adhesive layer, an oligomer having a saturated bond is preferred because such an oligomer is less likely to inhibit polymerization, and an alkyl (meth)acrylate whose alkyl group has a branched structure or an ester of an alicyclic alcohol and (meth)acrylic acid can be suitably used as a monomer constituting the (meth)acrylic oligomer.

[0077]

From these points of view, preferred examples of the (meth)acrylic oligomer include a copolymer of butyl acrylate (BA), methyl acrylate (MA) and acrylic acid (AA), a copolymer of cyclohexyl methacrylate (CHMA) and isobutyl methacrylate (IBMA), a copolymer of cyclohexyl methacrylate

(CHMA) and isobornyl methacrylate (IBXMA), a copolymer of cyclohexyl methacrylate (CHMA) and acryloyl morpholine (ACMO), a copolymer of cyclohexyl methacrylate (CHMA) and diethylacrylamide (DEAA), a copolymer of 1-adamantyl acrylate (ADA) and methyl methacrylate (MMA), a copolymer of dicyclopentanyl methacrylate (DCPMA) and isobornyl methacrylate (IBXMA), a copolymer of dicyclopentanyl methacrylate (DCPMA), cyclohexyl methacrylate (CHMA), isobornyl methacrylate (IBXMA), isobornyl acrylate (IBXA), cyclopentanyl methacrylate (DCPMA) and methyl methacrylate (MMA), and a homopolymer of each of dicyclopentanyl acrylate (DCPA), 1-adamantyl methacrylate (ADMA), and 1-adamantyl acrylate (ADA).

[0078]

As the polymerization method for the (meth)acrylic oligomer, similarly to the (meth)acrylic polymer, examples thereof include solution polymerization, emulsion polymerization, bulk polymerization, emulsion polymerization, and polymerization based on heat or active energy ray irradiation (thermal polymerization and active energy ray polymerization). Among these methods, from the viewpoint of transparency, water resistance, cost and others, the solution polymerization method and the active energy ray polymerization method are preferable. The (meth)acrylic oligomer to be obtained may be any of a

random copolymer, a block copolymer, a graft copolymer and the like.

[0079]

The (meth)acrylic oligomer can be used in the solvent-type pressure-sensitive adhesive composition and the active energy ray-curable pressure-sensitive adhesive composition, similarly to the (meth)acrylic polymer. For example, as the active energy ray-curable pressure-sensitive adhesive composition, the (meth)acrylic oligomer can be further mixed with a mixture of the monomer components (monomer mixture) constituting the (meth)acrylic polymer, or the partially polymerized substance thereof and can be used. When the (meth)acrylic oligomer is dissolved in a solvent, the pressure-sensitive adhesive composition is heat-dried to remove the solvent, and then active energy ray curing can be completed to obtain a pressure-sensitive adhesive layer.

[0080]

The weight average molecular weight ( $M_w$ ) of the (meth)acrylic oligomer used in the solvent-type pressure-sensitive adhesive composition is preferably 1,000 or more, more preferably 2,000 or more, still more preferably 3,000 or more, and particularly preferably 4,000 or more. The weight average molecular weight ( $M_w$ ) of the (meth)acrylic oligomer is preferably 30,000 or less, more preferably

15,000 or less, still more preferably 10,000 or less, and particularly preferably 7,000 or less. By adjusting the weight average molecular weight (Mw) of the (meth)acrylic oligomer within the above range, for example, when the (meth)acrylic oligomer is used in combination with the (meth)acrylic polymer, the (meth)acrylic oligomer is interposed between the (meth)acrylic polymers, and entanglement of the (meth)acrylic polymers is reduced, so that the pressure-sensitive adhesive layer is likely to deform with respect to minute distortions. As a result, a distortion applied to the other layers can be reduced, and cracking of each layer, peeling between the pressure-sensitive adhesive layer and the other layer, and other damage can be suppressed, which is a preferable embodiment. The weight average molecular weight (Mw) of the (meth)acrylic oligomer is a value calculated in terms of polystyrene as measured by GPC (gel permeation chromatography), similarly to the (meth)acrylic polymer.

[0081]

When the (meth)acrylic oligomer is used in the pressure-sensitive adhesive composition, the blending amount thereof is not particularly limited, and is preferably 70 parts by weight or less, more preferably 1 to 70 parts by weight, still more preferably 2 to 50 parts by weight, and even more preferably 3 to 40 parts by weight

with respect to 100 parts by weight of the (meth)acrylic polymer. By adjusting the blending amount of the (meth)acrylic oligomer within the above range, the (meth)acrylic oligomer is properly interposed between the (meth)acrylic polymers, and entanglement of the (meth)acrylic polymers is reduced, so that the pressure-sensitive adhesive layer is likely to deform with respect to minute distortions. As a result, a distortion applied to the other layers can be reduced, and cracking of each layer, peeling between the pressure-sensitive adhesive layer and the other layer, and other damage can be suppressed, which is a preferable embodiment.

[0082]

<Measurement of Weight Average Molecular Weight (Mw) of (Meth)acrylic Polymer and acrylic oligomer>

The weight average molecular weight (Mw) of the obtained (meth)acrylic polymer and the acrylic oligomer were measured by GPC (gel permeation chromatography).

- Analyzer: HLC-8120 GPC, manufactured by Tosoh Corporation
- Column: G7000H<sub>XL</sub> + GMH<sub>XL</sub> + GMH<sub>XL</sub>, manufactured by Tosoh Corporation
- Column size: each 7.8 mm $\phi$  × 30 cm, 90 cm in total
- Column temperature: 40°C
- Flow rate: 0.8 ml/min
- Injection volume: 100  $\mu$ l

- Eluent: Tetrahydrofuran
- Detector: Differential refractometer (RI)
- Standard sample: Polystyrene

[0083]

<Crosslinking Agent>

The pressure-sensitive adhesive composition of the present invention may contain a crosslinking agent. An organic crosslinking agent or a polyfunctional metal chelate may be used as the crosslinking agent in the case of solvent-type or the active energy ray-curable pressure-sensitive adhesive composition. Examples of the organic crosslinking agent include an isocyanate-based crosslinking agent, a peroxide-based crosslinking agent, an epoxy-based crosslinking agent, an imine-based crosslinking agent, and the like. The polyfunctional metal chelate may include those in which a polyvalent metal is covalently or coordinately bonded to an organic compound. Examples of the polyvalent metal atom include Al, Cr, Zr, Co, Cu, Fe, Ni, V, Zn, In, Ca, Mg, Mn, Y, Ce, Sr, Ba, Mo, La, Sn, and Ti. Examples of the atom in the organic compound that is covalently or coordinately bonded include an oxygen atom and the like. Examples of the organic compound include alkyl esters, alcohol compounds, carboxylic acid compounds, ether compounds, ketone compounds, and the like. In particular, in the case of the solvent-type pressure-

sensitive adhesive composition, a peroxide-based crosslinking agent or an isocyanate crosslinking agent is preferable, and among them, the peroxide-based crosslinking agent is preferably used. The peroxide-based crosslinking agent, for example, generates a radical due to abstraction of hydrogen from the side chain of the (meth)acrylic polymer to promote crosslinking between the side chains of the (meth)acrylic polymer. Therefore, compared with the case of crosslinking using an isocyanate-based crosslinking agent (for example, a polyfunctional isocyanate-based crosslinking agent), a crosslinked state is relatively loose, and it is possible to increase the cohesive force while maintaining the ease of deformation with respect to minute distortions, which is a preferable embodiment in terms of achieving flexibility (suppression of cracking and peeling) in a normal temperature environment and a high temperature environment. Also, an isocyanate-based crosslinking agent (particularly, a trifunctional isocyanate-based crosslinking agent) is preferable from the viewpoint of durability. In addition, a peroxide-based crosslinking agent and an isocyanate-based crosslinking agent (particularly, a bifunctional isocyanate-based crosslinking agent) are preferable from the viewpoint of bendability. Both the peroxide-based crosslinking agent and the bifunctional isocyanate-based crosslinking agent

form a flexible two-dimensional crosslinking, whereas the trifunctional isocyanate-based crosslinking agent forms a stronger three-dimensional crosslinking. When bending, two-dimensional crosslinking, which is a more flexible crosslinking, is advantageous. However, since only two-dimensional crosslinking is poor in durability and peeling is likely to occur, hybrid crosslinking between two-dimensional crosslinking and three-dimensional crosslinking is favorable, so that a trifunctional isocyanate-based crosslinking agent and a peroxide-based crosslinking agent or a bifunctional isocyanate-based crosslinking agent are preferably used in combination. As the active energy ray-curable pressure-sensitive adhesive composition, it is preferable to obtain the crosslinking effect by polymerization using the polyfunctional monomer from the viewpoint of productivity and thick film coating; however, it is also possible to use the above crosslinking agent or use the polyfunctional monomer in combination. For example, a crosslinking agent is mixed with a mixture of the monomer components (monomer mixture) constituting the (meth)acrylic polymer, or the partially polymerized substance thereof, and before and after active energy ray curing of the pressure-sensitive adhesive composition, the reaction of the crosslinking agent can be completed by heat drying.

[0084]

The amount of the crosslinking agent to be used is preferably 0.1 to 10 parts by weight, more preferably 0.2 to 5 parts by weight, even more preferably less than 0.3 to 3 part by weight, per 100 parts by weight of the (meth)acrylic polymer. Within the above range, a preferred embodiment excellent in bending resistance is obtained. In order to increase the cohesive force without changing the microdeformation stress, it is preferable to increase the amount of the crosslinking agent.

[0085]

When the peroxide-based crosslinking agent is used alone, the amount of the crosslinking agent is preferably 0.5 to 5 parts by weight and more preferably 1 to 3 parts by weight with respect to 100 parts by weight of the (meth)acrylic polymer. If the amount of the crosslinking agent is within the above range, the cohesive force can be sufficiently increased while maintaining the ease of deformation with respect to minute distortions, and the durability and bending resistance can be improved, which is a preferable embodiment.

[0086]

When a peroxide-based crosslinking agent and an isocyanate-based crosslinking agent are used in combination, the lower limit of the weight ratio of the

peroxide-based crosslinking agent to the isocyanate-based crosslinking agent (peroxide-based crosslinking agent/isocyanate-based crosslinking agent) is preferably 1.2 or more, more preferably 1.5 or more, and still more preferably 3 or more. The upper limit of the weight ratio is preferably 500 or less, more preferably 300 or less, and still more preferably 200 or less. If the weight ratio is within the above range, the cohesive force can be sufficiently increased while maintaining the ease of deformation with respect to minute distortions, which is a preferable embodiment.

[0087]

<Other Additives>

Further, the pressure-sensitive adhesive composition of the present invention may contain any other known additives, including, for example, various silane coupling agents, polyether compounds such as polyalkylene glycol (e.g. polypropylene glycol etc.), powder such as coloring agents and pigments, dyes, surfactants, plasticizers, tackifiers, surface lubricants, leveling agents, softeners, antioxidants, anti-ageing agents, light stabilizers, ultraviolet absorbers, polymerization inhibitors, antistatic agents (alkali metal salt or ionic liquid which are an ionic compound, etc.), inorganic or organic fillers, metal powder, particle- or foil-shaped materials, and the

like, and such additives can be appropriately added depending on the intended use. In addition, a redox system including a reducing agent to be added may also be used in the controllable range.

[0088]

Although the preparation method of the pressure-sensitive adhesive composition is not particularly limited, known methods can be used, and, for example, as described above, a solvent-type acrylic pressure-sensitive adhesive composition is prepared by mixing a (meth)acrylic polymer and components (such as the (meth)acrylic oligomer, crosslinking agent, silane coupling agent, solvent and additive) added if necessary. As described above, the active energy ray curable-type acrylic pressure-sensitive adhesive composition is prepared by mixing a monomer mixture or the partially polymerized substance thereof and components (such as the photopolymerization initiator, polyfunctional monomer, the (meth)acrylic oligomer, crosslinking agent, silane coupling agent, solvent and additive) added if necessary.

[0089]

The pressure-sensitive adhesive composition preferably has a viscosity suitable for handling and coating. Thus, the active energy ray curable-type acrylic pressure-sensitive adhesive composition preferably includes

the partially polymerized substance of a monomer mixture. The polymerization percentage of the partially polymerized substance is not particularly limited, and is preferably 5 to 20% by weight and more preferably 5 to 15% by weight.

[0090]

The polymerization percentage of the partially polymerized substance is derived as follows.

A sample is prepared by sampling a fraction of the partially polymerized substance. The sample is precisely weighed to obtain the weight thereof, and the resulting weight is defined as "the weight of the partially polymerized substance before drying." Next, the sample is dried at 130°C for 2 hours, and the sample after drying is precisely weighed to obtain the weight thereof, and the resulting weight is defined as "the weight of the partially polymerized substance after drying." From "the weight of the partially polymerized substance before drying" and "the weight of the partially polymerized substance after drying," the weight decrement of the sample due to the drying at 130°C for 2 hours is derived, and the resulting weight decrement is defined as "the weight decrement" (the weights of the volatilized component and the unreacted monomers).

From the resulting "weight of the partially polymerized substance before drying" and the resulting

"weight decrement," the polymerization percentage (% by weight) of the partially polymerized substance of the monomer components is derived on the basis of the following formula.

polymerization percentage (% by weight) of the partially polymerized substance of the monomer components =  $[1 - (\text{weight decrement}) / (\text{weight of partially polymerized substance before drying})] \times 100$

[0091]

<Formation of pressure-sensitive adhesive layer>

Examples of a method of forming the pressure-sensitive adhesive layer include a method including applying the solvent-type pressure-sensitive adhesive composition to a release-treated separator (release film) or the like and removing a polymerization solvent and the like by drying to form a pressure-sensitive adhesive layer, a method including applying the solvent-type pressure-sensitive adhesive composition to a polarizing film or the like, and removing the polymerization solvent and the like by drying to form a pressure-sensitive adhesive layer on the polarizing film or the like, and a method including applying the active energy ray-curable pressure-sensitive adhesive composition to a release-treated separator or the like and applying an active energy ray to form a pressure-sensitive adhesive layer. If necessary, heat drying may be

performed in addition to irradiation with an active energy ray. In applying the pressure-sensitive adhesive composition, one or more kinds of solvents other than the polymerization solvent may be newly added as needed.

[0092]

A silicone release liner is preferably used as the release-treated separator. When the pressure-sensitive adhesive composition of the present invention is applied to such a liner and dried to form a pressure-sensitive adhesive layer, any appropriate drying method may be suitably adopted depending on the purpose. A method of drying under heating is preferably used. The heat drying temperature, in a case using an acrylic pressure-sensitive adhesive containing a (meth)acrylic polymer, is preferably from 40°C to 200°C, more preferably from 50°C to 180°C, particularly preferably from 70°C to 170°C. When the heating temperature is set in the above range, a pressure-sensitive adhesive layer having good adhesive property can be obtained.

[0093]

Any suitable drying time may be used as appropriate. The drying time, in a case using an acrylic pressure-sensitive adhesive containing a (meth)acrylic polymer, is preferably from 5 seconds to 20 minutes, more preferably from 5 seconds to 10 minutes, particularly preferably from

10 seconds to 5 minutes.

[0094]

As a coating method of the pressure-sensitive adhesive composition, various methods may be used. Specific examples of such methods include a roll coating method, a kiss roll coating method, a gravure coating method, a reverse coating method, a roll brush coating method, a spray coating method, a dip roll coating method, a bar coating method, a knife coating method, an air knife coating method, a curtain coating method, a lip coating method, and an extrusion coating method with a die coater or the like.

[0095]

The thickness of the present invention is preferably 1 to 200  $\mu\text{m}$ , more preferably 5 to 150  $\mu\text{m}$ , even more preferably 15 to 100  $\mu\text{m}$ . The pressure-sensitive adhesive layer may be a single layer or may have a laminated structure. A preferred embodiment is within the above range in terms of not inhibiting the bending and also in terms of adhesiveness (retention resistance). Further, in the case of having a plurality of pressure-sensitive adhesive layers, all the pressure-sensitive adhesive layers are preferably within the above-mentioned range.

[0096]

Hereinafter, embodiments of an optical film, a

laminate for a flexible image display device, and a flexible image display device according to the present invention will be described in detail below with reference to the drawings and the like.

[0097]

The present invention is hereunder described in detail, but it should not be construed that the present invention is limited to the following embodiments, and the present invention may be arbitrarily modified and carried out within the range where the gist of the present invention is not deviated.

[0098]

[Laminate for flexible image display device]

The laminate for a flexible image display device of the present invention is characterized by including a pressure-sensitive adhesive layer and an optical film.

[0099]

[Optical film]

The laminate for a flexible image display device of the present invention is characterized by including an optical film including at least a polarizer, and the optical film refers to one including, for example, a film such as a protective film or a retardation film formed of a transparent resin material in addition to the polarizer. In the present invention, as the optical film, a

configuration including the polarizer, a protective film of a transparent resin material on a first surface of the polarizing film, and a retardation film on a second surface of the polarizer different from the first surface is referred to as an optical laminate. The optical film does not include a pressure-sensitive adhesive layer such as the first pressure-sensitive adhesive layer described later.

[0100]

The thickness of the optical film is preferably 92  $\mu\text{m}$  or less, more preferably 60  $\mu\text{m}$  or less, even more preferably 10 to 50  $\mu\text{m}$ . Within the above range, a preferred embodiment is obtained without hindering the bending.

[0101]

As long as the properties of the present invention are not impaired, a protective film may be bonded to at least one side of the polarizer with an adhesive (layer) (not shown in the drawing). An adhesive can be used for the adhesion treatment of the polarizer and the protective film. Examples of the adhesive include isocyanate-based adhesives, polyvinyl alcohol-based adhesives, gelatin-based adhesives, vinyl-based latex, aqueous-based polyester and the like. The adhesive is usually used as an adhesive made of an aqueous solution, and usually contains 0.5 to 60% by weight of a solid content. Besides the above, as an

adhesive between the polarizer and the protective film, an ultraviolet curable adhesive, an electron beam-curable adhesive and the like can be mentioned. The adhesive for electron beam-curable type polarizer shows suitable adhesion property to the various protective films mentioned above. The adhesive used in the present invention may contain a metal compound filler. In the present invention, those obtained by laminating a polarizer and a protective film with an adhesive (layer) may be sometimes referred to as a polarizing film (polarizing plate).

[0102]

<Polarizer>

In the polarizer used in the optical film of the present invention, a polyvinyl alcohol (PVA)-based resin which is stretched by a stretching step such as an in-air stretching (dry stretching) or a stretching step in an aqueous boric acid and in which iodine is aligned can be used.

[0103]

Typically, as a method for producing the polarizer, there is a production method including a step of dyeing a single layer body of a PVA-based resin and a step of stretching such a single layer body as described in JP-A-2004-341515 (a monolayer stretching method). In addition, as described in JP-A-51-069644, JP-A-2000-338329, JP-A-

2001-343521, WO 2010/100917, JP-A-2012-073563, and JP-A-2011-2816, there is exemplified a production method including a step of stretching a PVA-based resin layer and a stretching resin base material in the state of a laminate and a step of dyeing the laminate. According to this production method, even when the PVA-based resin layer is thin, such resin layer can be stretched without inconveniences such as breakage due to stretching because the resin layer is supported by the stretching resin base material.

[0104]

As the production method including a step of stretching in the state of a laminate and a step of dyeing the laminate, an air stretching (dry stretching) method as described in JP-A-51-069644, JP-A-2000-338329, or JP-A-2001-343521 is exemplified. From the viewpoint of being able to stretching to a high drawing ratio and improve the polarization performance, a production method including a step of stretching in an aqueous boric acid solution as described in WO 2010/100917 A and JP-A-2012-073563 is preferable, and a production method (two-step stretching method) including a step of performing an auxiliary in-air stretching before stretching in an aqueous boric acid solution as described in JP-A-2012-073563 is particularly preferable. In addition, as described in JP-A-2011-2816, a

method of stretching a PVA-based resin layer and a stretching resin base material in a laminate state, excessively dyeing the PVA-based resin layer, and then decoloring the dyed resin layer (excess dyeing decolorization method) is also preferable. The polarizer used in the optical film of the present invention is made of the polyvinyl alcohol-based resin in which iodine is aligned as described above and can be formed by laminating the polyvinyl alcohol-based resin stretched by a two-step stretching method including an auxiliary in-air stretching and a stretching in an aqueous boric acid solution. The polarizer is made of the polyvinyl alcohol-based resin in which iodine is aligned as described above and can be prepared by excessively dyeing a laminate of a stretched PVA-based resin layer and a resin base material for stretching, followed by decoloring.

[0105]

The thickness of the polarizer is 20  $\mu\text{m}$  or less, preferably 12  $\mu\text{m}$  or less, more preferably 9  $\mu\text{m}$  or less, even more preferably 1 to 8  $\mu\text{m}$ , particularly preferably 3 to 6  $\mu\text{m}$ . Within the above range of the thickness of the polarizer, a preferred embodiment is obtained without hindering the bending.

[0106]

<Retardation film>

As the retardation film that can be used in the optical film in the present invention, one obtained by stretching a polymer film or one obtained by aligning and fixing a liquid crystal material can be used. In this specification, the retardation film means a film having birefringence in the plane and/or thickness direction.

[0107]

Examples of the retardation film may include an anti-reflection retardation film (see paragraphs [0221], [0222], and [0228] in JP-A-2012-133303), a viewing-angle compensating retardation film (see paragraphs [0225] and [0226] in JP-A-2012-133303), and a viewing-angle compensating obliquely-aligned retardation film (see paragraph [0227] in JP-A-2012-133303).

[0108]

Any known retardation film substantially having any of the functions described above can be used irrespective of, for example, the retardation value, the arrangement angle, the three-dimensional birefringence index, whether or not a single layer or a multilayer, and other factors.

[0109]

The thickness of the retardation film is preferably 20  $\mu\text{m}$  or less, more preferably 10  $\mu\text{m}$  or less, even more preferably 1 to 9  $\mu\text{m}$ , particularly preferably 3 to 8  $\mu\text{m}$ . Within the above range, a preferred embodiment is obtained

without hindering the bending.

[0110]

<Protective film>

As the protective film, formed of the transparent resin material, that can be used in the optical film in the present invention, the protective film (also referred to as a transparent protective film) includes a cycloolefin-based resin such as a norbornene resin, an olefin-based resin such as polyethylene and polypropylene, a polyester-based resin, a (meth)acrylic resin or the like.

[0111]

The thickness of the protective film ranges of preferably 5 to 60  $\mu\text{m}$ , more preferably 10 to 40  $\mu\text{m}$ , even more preferably 10 to 30  $\mu\text{m}$ , and a surface treatment layer, such as an anti-glare layer or an antireflection layer, may be provided as appropriate. Within the above range, a preferred embodiment is obtained without hindering the bending.

[0112]

[Pressure-sensitive adhesive layer]

The laminate for a flexible image display device of the present invention is a laminate for a flexible image display device including a pressure-sensitive adhesive layer, and the pressure-sensitive adhesive layer has the predetermined storage elastic modulus.

[0113]

Although the pressure-sensitive adhesive layer may be one layer, two or more pressure-sensitive adhesive layers may be provided for laminating a transparent conductive film, an organic EL display panel, a window, a decorative printing film, a retardation layer, a protective film, etc. in addition to the optical film (for example, as a case where a plurality of pressure-sensitive adhesive layers are provided in the laminate for a flexible image display device such as a first pressure-sensitive adhesive layer and a second pressure-sensitive adhesive layer, for example, see Fig. 2, etc.). When a plurality of pressure-sensitive adhesive layers are provided, it is preferable to provide two or more and five or less layers. If the number of layers is more than five, the thickness of the entire laminate becomes thick, so that a distortion difference between an outermost layer and an innermost layer at a bent portion of the laminate becomes large, and peeling or breakage is likely to occur, which is not preferable.

[0114]

[First pressure-sensitive adhesive layer]

Among the pressure-sensitive adhesive layers used in the laminate for a flexible image display device of the present invention, the first pressure-sensitive adhesive layer is preferably disposed on a side opposite to a

surface in contact with the polarizer with respect to the protective film (see Fig. 2).

[0115]

[Other pressure-sensitive adhesive layers]

Among the pressure-sensitive adhesive layers used in the laminate for a flexible image display device of the present invention, the second pressure-sensitive adhesive layer can be disposed on the side opposite to the surface in contact with the polarizer with respect to the retardation film (see Fig. 2).

[0116]

Among the pressure-sensitive adhesive layers used in the laminate for a flexible image display device of the present invention, a third pressure-sensitive adhesive layer can be disposed on a side opposite to a surface in contact with the second pressure-sensitive adhesive layer with respect to a transparent conductive layer constituting the touch sensor (see Fig. 2).

[0117]

Among the pressure-sensitive adhesive layers used in the laminate for a flexible image display device of the present invention, the third pressure-sensitive adhesive layer can be disposed on a side opposite to a surface in contact with the first pressure-sensitive adhesive layer with respect to the transparent conductive layer

constituting the touch sensor (see Fig. 3).

[0118]

In the case of using the second pressure-sensitive adhesive layer and further other pressure-sensitive adhesive layer (for example, and the third pressure-sensitive adhesive layer or the like) in addition to the first pressure-sensitive adhesive layer, these pressure-sensitive adhesive layers may have the same composition (same pressure-sensitive adhesive composition), may have the same characteristics, or may have different characteristics, but are not particularly limited. From the viewpoints of workability, economy, and flexibility, it is preferable that all the pressure-sensitive adhesive layers are each a pressure-sensitive adhesive layer having substantially the same composition and the same characteristics.

[0119]

[Transparent Conductive Layer]

A member having a transparent conductive layer is not particularly limited and known materials can be used as such a member. The member includes those having a transparent conductive layer on a transparent base material such as a transparent film or the like and a member having a transparent conductive layer and a liquid crystal cell can be mentioned.

[0120]

The transparent base material may be of any type having transparency, and examples thereof include a base material (for example, a sheet-like, film-like, or plate-like base material) made of a resin film or the like. The thickness of the transparent base material is not particularly limited, but is preferably about 10 to 200  $\mu\text{m}$ , more preferably about 15 to 150  $\mu\text{m}$ .

[0121]

The resin film may be made of any material, such as any of various plastic materials having transparency. Examples of such materials include polyester-based resins such as polyethylene terephthalate and polyethylene naphthalate, acetate-based resins, polyethersulfone-based resins, polycarbonate-based resins, polyamide-based resins, polyimide-based resins, polyolefin-based resins, (meth)acrylic resins, polyvinyl chloride-based resins, polyvinylidene chloride-based resins, polystyrene-based resins, polyvinyl alcohol-based resins, polyarylate-based resins, and polyphenylene sulfide-based resins. Among them, polyester-based resins, polyimide-based resins, and polyethersulfone-based resins are particularly preferred.

[0122]

The surface of the transparent base material may be previously subjected to sputtering, corona discharge

treatment, flame treatment, ultraviolet irradiation, electron beam irradiation, chemical treatment, etching treatment such as oxidation, or undercoating treatment so that the transparent base material can have improved adhesiveness to the transparent conductive layer formed thereon. Before the transparent conductive layer is formed, if necessary, the transparent base material may be subjected to solvent washing or ultrasonic washing for removal of dust and cleaning.

[0123]

Examples of the material used to form the transparent conductive layer include, but not limited to, metal oxides of at least a metal selected from the group consisting of indium, tin, zinc, gallium, antimony, titanium, silicon, zirconium, magnesium, aluminum, gold, silver, copper, palladium, tungsten and molybdenum, and an organic conductive polymer such as polythiophene. If necessary, the metal oxides may be doped with any metal from the group shown above. For example, tin oxide-doped indium oxide (ITO) and antimony-doped tin oxide are preferably used, and in particular, ITO is preferably used. ITO preferably includes 80 to 99% by weight of indium oxide and 1 to 20% by weight of tin oxide.

[0124]

The ITO may be crystalline or amorphous. The

crystalline ITO can be obtained by high-temperature sputtering or further heating an amorphous ITO.

[0125]

The thickness of the transparent conductive layer of the present invention is preferably 0.005 to 10  $\mu\text{m}$ , more preferably 0.01 to 3  $\mu\text{m}$ , even more preferably 0.01 to 1  $\mu\text{m}$ . When the thickness of the transparent conductive layer is less than 0.005  $\mu\text{m}$ , the transparent conductive layer tends to be more variable in electric resistance. On the other hand, the transparent conductive layer with a thickness of more than 10  $\mu\text{m}$  may be produced with lower productivity at higher cost and tend to have a lower level of optical properties.

[0126]

The total light transmittance of the transparent conductive layer of the present invention is preferably 80% or more, more preferably 85% or more, even more preferably 90% or more.

[0127]

The density of the transparent conductive layer of the present invention is preferably 1.0 to 10.5  $\text{g}/\text{cm}^3$ , more preferably 1.3 to 3.0  $\text{g}/\text{cm}^3$ .

[0128]

The surface resistance value of the transparent conductive layer of the present invention is preferably 0.1

to 1000  $\Omega/\square$ , more preferably 0.5 to 500  $\Omega/\square$ , even more preferably 1 to 250  $\Omega/\square$ .

[0129]

The method for forming the transparent conductive layer is not particularly limited, and conventionally known methods can be adopted. Specifically, for example, a vacuum deposition method, a sputtering method, and an ion plating method can be exemplified. In addition, an appropriate method can be adopted according to the required film thickness.

[0130]

In addition, an undercoat layer, an oligomer prevention layer, and the like can be provided between the transparent conductive layer and the transparent base material, if necessary.

[0131]

The transparent conductive layer forms a touch sensor and is required to be configured to be bendable.

[0132]

The transparent conductive layer forming a touch sensor used in the laminate for a flexible image display device of the present invention may be disposed on the side opposite to the surface in contact with the retardation film with respect to the second pressure-sensitive adhesive layer (see Fig.2).

[0133]

The transparent conductive layer forming a touch sensor used in the laminate for a flexible image display device of the present invention may be disposed on the side opposite to the surface in contact with the protective film with respect to the first pressure-sensitive adhesive layer (see Fig.3).

[0134]

The transparent conductive layer forming a touch sensor used for the laminate for a flexible image display device of the present invention may be disposed between the protective film and a window film (OCA) (see Fig.3).

[0135]

The transparent conductive layer can be suitably applied to a liquid crystal display device incorporating a touch sensor such as an in-cell type or an on-cell type as a case of being used for a flexible image display device, and in particular, a touch sensor may be built in (even incorporated) in an organic EL display panel.

[0136]

[Conductive Layer (Antistatic Layer)]

Further, the laminate for a flexible image display device of the present invention may have a layer having conductivity (a conductive layer, an antistatic layer). Since the laminate for a flexible image display device has

a bending function and has a very thin thickness structure, such a laminate is highly responsive to feeble static electricity generated in a manufacturing process or the like and is easily damaged, but by providing a conductive layer in the laminate, the load due to static electricity in the manufacturing process and the like is largely reduced, which is a preferable embodiment.

[0137]

In addition, one of the major features of the flexible image display device including the laminate is to have a bending function, but in the case of continuous bending, static electricity may be generated due to shrinkage between the films (base material) at the bent portion. Therefore, when conductivity is imparted to the laminate, generated static electricity can be promptly removed and damage caused by static electricity of the image display device can be reduced, which is a preferable embodiment.

[0138]

Further, the conductive layer may be an undercoat layer having a conductive function, a pressure-sensitive adhesive containing a conductive component, or a surface treatment layer containing a conductive component. For example, a method of forming a conductive layer between a polarizing film and a pressure-sensitive adhesive layer by

using an antistatic composition containing a binder and a conductive polymer such as polythiophene can be employed. Further, a pressure-sensitive adhesive containing an ionic compound which is an antistatic agent can also be used. The conductive layer preferably has one or more layers and may contain two or more layers.

[0139]

[Flexible Image Display Device]

The flexible image display device of the present invention includes the laminate for a flexible image display device and an organic EL display panel configured to be foldable, wherein the laminate for a flexible image display device is disposed on a viewing side and configured to be foldable with respect to the organic EL display panel. Although arbitrary, a window can be disposed on the viewing side with respect to the laminate for a flexible image display device (see Figs.2 to 4).

[0140]

Fig. 2 is a cross-sectional view showing one embodiment of a flexible image display device according to the present invention. A flexible image display device 100 includes a laminate 11 for the flexible image display device and an organic EL display panel 10 configured to be foldable. The laminate 11 for the flexible image display device is disposed on the viewing side with respect to the

organic EL display panel 10, and the flexible image display device 100 is configured to be foldable. Further, although optional, a transparent window 40 can be disposed on the viewing side of the laminate for the flexible image display device with an interposed first pressure-sensitive adhesive layer 12-1 with respect to the laminate 11 for the flexible image display device.

[0141]

The laminate 11 for the flexible image display device includes the optical laminate 20 and the pressure-sensitive adhesive layer forming a second pressure-sensitive adhesive layer 12-2 and a third pressure-sensitive adhesive layer 12-3.

[0142]

The optical laminate 20 includes a polarizer 1, a protective film 2 made of a transparent resin material, and a retardation film 3. The protective film 2 made of a transparent resin material is bonded to a first surface on the viewing side of the polarizer 1. The retardation film 3 is bonded to a second surface different from the first surface of the polarizer 1. For example, the polarizer 1 and the retardation film 3 generate circularly polarized light in order to prevent light incident inside from the viewing side of the polarizer 1 from being internally reflected and emitted to the viewing side, or to compensate

a viewing angle.

[0143]

In the present embodiment, a protective film is provided on one side of a polarizer only, whereas a protective film is conventionally provided on both sides of a polarizer, and the thickness of the optical laminate 20 can be reduced by using a polarizer having a very thin thickness (for example, 20  $\mu\text{m}$  or less) as compared with the polarizer used in the conventional organic EL display device. In addition, since the polarizer 1 is much thinner than the polarizer used in the conventional organic EL display device, stress due to expansion and contraction occurring under temperature or humidity conditions becomes extremely smaller. Therefore, the possibility that the stress caused by the shrinkage of the polarizer causes deformation such as warping in the adjacent organic EL display panel 10 is greatly reduced, and the deterioration of the display quality due to deformation and breakage of the panel sealing material can be greatly suppressed. In addition, by using a thin polarizer, bending is not hindered, which is a preferable embodiment.

[0144]

In the case of bending the optical laminate 20 with the protective film 2 side as the inside, the thickness (for example, 92  $\mu\text{m}$  or less) of the optical laminate 20 is

thinned and the first pressure-sensitive adhesive layer 12-1 having the properties of moduli at 100% and 500% as described above is disposed on the side opposite to the retardation film 3 with respect to the protective film 2 to make it possible to reduce the stress applied to the optical laminate 20, whereby the optical laminate 20 can be folded. Cracking of each layer and peeling of the pressure-sensitive adhesive layer can be suppressed at the bent portion, and finally the laminate 11 for a flexible image display device can be bent. Therefore, an appropriate range of the moduli at 100% and 500% may be set according to the environmental temperature in which the flexible image display device is used.

[0145]

Optionally, a foldable transparent conductive layer 6 forming a touch sensor may further be disposed on the side opposite to the protective film 2 with respect to the retardation film 3. The transparent conductive layer 6 is configured to be directly bonded to the retardation film 3 by a manufacturing method as disclosed in, for example, JP-A-2014-219667, whereby the thickness of the optical laminate 20 is reduced and the stress applied to the optical laminate 20 when the optical laminate 20 is folded can be further reduced.

[0146]

Optionally, a pressure-sensitive adhesive layer forming a third pressure-sensitive adhesive layer 12-3 can be further disposed on the side opposite to the retardation film 3 with respect to the transparent conductive layer 6. In the present embodiment, the second pressure-sensitive adhesive layer 12-2 is directly bonded to the transparent conductive layer 6. By providing the second pressure-sensitive adhesive layer 12-2, it is possible to further reduce the stress applied to the optical laminate 20 when folded.

[0147]

The flexible image display device shown in Fig. 3 is substantially the same as that shown in Fig. 2. In the flexible image display device of Fig. 2, a foldable transparent conductive layer 6 forming a touch sensor is disposed on the side opposite to the protective film 2 with respect to the retardation film 3, whereas in the flexible image display device of Fig. 3, a foldable transparent conductive layer 6 forming a touch sensor is disposed on the side opposite to the protective film 2 with respect to the first pressure-sensitive adhesive layer 12-1. This is a different point. Further, there is a different point in that in the flexible image display device of Fig. 2, the third pressure-sensitive adhesive layer 12-3 is disposed on the side opposite to the retardation film 3 with respect to

the transparent conductive layer 2, whereas in the flexible image display device of Fig. 3, the second pressure-sensitive adhesive layer 12-2 is disposed on the side opposite to the protective film 2 with respect to the retardation film 3.

[0148]

In addition, although optional, the third pressure-sensitive adhesive layer 12-3 can be disposed when the window 40 is disposed on the viewing side with respect to the laminate 11 for the flexible image display device.

[0149]

The flexible image display device of the present invention can be suitably used as a flexible liquid crystal display device, an organic EL (electroluminescence) display device, a PDP (plasma display panel), and an electronic paper. Further, such a flexible image display device can be used irrespective of a type of a touch panel or the like such as a resistive film type or a capacitive type.

[0150]

In addition, as shown in Fig. 4, the flexible image display device of the present invention may also be used as an in-cell type flexible image display device in which the transparent conductive layer 6 forming a touch sensor is incorporated in the organic EL display panel 10.

## EXAMPLES

[0151]

Several examples relating to the present invention will be described below, but the present invention is not intended to be limited to such specific examples.

[0152]

### Example 1

#### <Preparation of prepolymer>

In a four-neck flask, a monomer mixture containing 59 parts by weight of lauryl acrylate (LA), 40 parts by weight of 2-ethylhexyl acrylate (2EHA), and 1 part by weight of 4-hydroxybutyl acrylate (4HBA), 0.05 parts by weight of 2,2-dimethoxy-1,2-diphenylethan-1-one (trade name: "Irgacure 651," manufactured by BASF Japan Ltd) which was a photopolymerization initiator, and 0.05 parts by weight of 1-hydroxy-cyclohexyl-phenyl-ketone (trade name: "Irgacure 184," manufactured by BASF Japan Ltd) which was a photopolymerization initiator were placed; the resulting mixture was irradiated with ultraviolet light to be photopolymerized in a nitrogen atmosphere, until the viscosity of the mixture reached approximately 15 Pa·s (BH viscometer, No. 5 rotor, 10 rpm, temperature: 30°C), and thus a partially polymerized monomer syrup (a partially polymerized substance of the monomer components) was obtained.

[0153]

<Preparation of acrylic pressure-sensitive adhesive composition>

With 100 parts by weight of the obtained partially polymerized monomer syrup, as additional monomers, 5 parts by weight of N-vinylpyrrolidone, 0.15 parts by weight of 1,6-hexanediol diacrylate (trade name: "A-HD-N," manufactured by Shin-Nakamura Chemical Co, Ltd., HDDA, polyfunctional monomer), 0.1 parts by weight of 2,2-dimethoxy-1,2-diphenylethan-1-one (trade name: "Irgacure 651," manufactured by BASF Japan Ltd, additional initiator) which was a photopolymerization initiator, and 0.3 parts by weight of silane coupling agent (trade name: "KBM-403", manufactured by Shin-Etsu Chemical Co., Ltd.) were uniformly mixed to yield an acrylic pressure-sensitive adhesive composition.

[0154]

<Formation of pressure-sensitive adhesive layer>

The acrylic pressure-sensitive adhesive composition was applied to the release-treated surface of a release film (trade name: "MRF#38," manufactured by Mitsubishi Plastics, Inc.) so as for the thickness after the formation of the pressure-sensitive adhesive layer to be 70  $\mu\text{m}$ , and thus a pressure-sensitive adhesive composition layer was formed. Then, a release film (trade name: "MRN#38,"

manufactured by Mitsubishi Plastics, Inc.) was laminated on the surface of the pressure-sensitive adhesive composition layer. Then, the pressure-sensitive adhesive composition layer was irradiated with ultraviolet ray under the conditions of the illuminance: 4 mW/cm<sup>2</sup> and the light intensity: 1200 mJ/cm<sup>2</sup>, and thus the pressure-sensitive adhesive composition layer was photocured to form a pressure-sensitive adhesive layer. Then, a pressure-sensitive adhesive layer in which both sides of the pressure-sensitive adhesive layer were each protected with a release film was obtained.

[0155]

Examples 2 to 3, Comparative Examples 1 to 5

A prepolymer and an acrylic pressure-sensitive adhesive composition were prepared in the same manner as in Example 1 except that in the preparation of the prepolymer of Example 1, the monomer mixture (type, composition) of the prepolymer was changed as shown in Table 2, and in the preparation of the acrylic pressure-sensitive adhesive composition, additional monomers (type, blending amount) were changed as shown in Table 2, and then a pressure-sensitive adhesive layer was formed.

In Comparative Examples 2 and 3, as shown in Table 2, with the change in the blending amount of 1,6-hexanediol diacrylate to 0.3 parts by weight, the blending amount of

2,2-dimethoxy-1,2-diphenylethan-1-one (trade name: "Irgacure 651," manufactured by BASF Japan Ltd, additional initiator) which was a photopolymerization initiator was changed to 0.6 parts by weight.

[0156]

After an optical laminate was produced using each member produced by the following method, an optical laminate with a pressure-sensitive adhesive layer was produced using the pressure-sensitive adhesive layers obtained in the above Examples and Comparative Examples.

[0157]

[Polarizer]

An amorphous polyethylene terephthalate (hereinafter referred to as "PET") (IPA-copolymerized PET) film (thickness: 100  $\mu\text{m}$ ) with 7 mol% of isophthalic acid unit was used as a thermoplastic resin base material, and a surface of the film was subjected to a corona treatment (58 W/m<sup>2</sup>/min). Further, a PVA (polymerization degree: 4200, saponification degree: 99.2%) added with 1 wt% of acetoacetyl-modified PVA (trade name: Gohsefimer Z200 (average polymerization degree: 1200, saponification degree: 98.5 mol%, acetoacetyl-modification degree: 5 mol%), manufactured by Nippon Synthetic Chemical Industry Co., Ltd.) was used to preliminarily prepare a coating solution consisting of an aqueous PVA solution containing

5.5 wt% of PVA-based resin. Then, the coating solution was applied onto a base material to allow a film thickness after drying to become 12  $\mu\text{m}$  and subjected to hot-air drying under an atmosphere at 60°C for 10 minutes to prepare a laminate in which a layer of the PVA-based resin is provided on the base material.

[0158]

Then, this laminate was first subjected to free-end stretching in air (auxiliary in-air stretching) at 130°C at a stretching ratio of 1.8 times to form a stretched laminate.

Then, the stretched laminate was immersed in a boric acid insolubilizing aqueous solution having a temperature of 30°C for 30 seconds to perform a step of insolubilizing a PVA layer in which PVA molecules are aligned and which is contained in the stretched laminate. The boric acid insolubilizing aqueous solution in this step was prepared to allow a boric acid to be contained in an amount of 3 weight parts with respect to 100 weight parts of water. The stretched laminate was subjected to dyeing to form a dyed laminate. The dyed laminate was prepared by immersing the stretched laminate in a dyeing solution containing iodine and potassium iodide and having a temperature of 30°C for an arbitrary time, in such a manner that a single layer transmittance of a PVA layer making up a polarizer to

be finally obtained falls with the range of 40 to 44%, thereby causing the PVA layer included in the stretched laminate to be dyed with iodine. In this step, the dyeing solution was prepared using water as a solvent to allow an iodine concentration and a potassium iodide concentration to fall with the range of 0.1 to 0.4% by weight, and the range of 0.7 to 2.8% by weight, respectively. A concentration ratio of iodine to potassium iodide was 1: 7. Then a step of immersing the dyed laminate in a boric acid crosslinking aqueous solution at 30°C for 60 seconds so as to subject PVA molecules in the PVA layer having iodine adsorbed therein to a cross-linking treatment was performed. The boric acid crosslinking aqueous solution in this step was set to contain boric acid in an amount of 3 weight parts with respect to 100 parts by weight of water and contain potassium iodide in an amount of 3 parts by weight with respect to 100 parts by weight of water.

[0159]

Further, an obtained dyed laminate was stretched in an aqueous boric acid solution (stretching in an aqueous boric acid solution) at a stretching temperature of 70°C, at a stretching ratio of 3.05 times in the same direction as that during the previous in-air stretching to obtain an optical film laminate stretched at a final (total) stretching ratio of 5.50 times. The optical film laminate

was taken out of the aqueous boric acid solution, and a boric acid attaching on a surface of the PVA layer was washed with an aqueous solution containing 4 parts by weight of potassium iodide with respect to 100 parts by weight of water. The washed optical film laminate was dried through a drying step using hot air at 60°C. The polarizer including the obtained optical film laminate had a thickness of 5  $\mu\text{m}$ .

[0160]

[Protective Film]

A protective film obtained by extruding a methacrylic resin pellet having a glutarimide ring unit to form a film shape and then stretching the film was used. This protective film had a thickness of 20  $\mu\text{m}$  and was an acrylic film having a moisture permeability of 160 g/m<sup>2</sup>.

[0161]

Next, the polarizer and the protective film were bonded using an adhesive shown below to obtain a polarizing film.

[0162]

As the adhesive (active energy ray-curable adhesive), each component was mixed according to the formulation table shown in Table 1 and stirred at 50°C for 1 hour to prepare an adhesive (active energy ray-curable adhesive A). Numerical values in the table indicate weight% when the

total amount of the composition is taken as 100% by weight.

Each component used is as follows.

HEAA: Hydroxyethylacrylamide

M-220: ARONIX M-220, tripropylene glycol diacrylate)  
manufactured by Toagosei Co., Ltd.

ACMO: Acryloyl morpholine

AAEM: 2-Acetoacetoxyethyl methacrylate, manufactured  
by The Nippon Synthetic Chemical Industry Co., Ltd.

UP-1190: ARUFON UP-1190, manufactured by Toagosei  
Co., Ltd.

IRG 907: IRGACURE 907, 2-methyl-1-(4-  
methylthiophenyl)-2-morpholinopropan-1-one, manufactured by  
BASF

DETX-S: KAYACURE DETX-S, diethylthioxanthone,  
manufactured by Nippon Kayaku Co., Ltd.

[0163]

[Table 1]

| (wt%)   | Adhesive<br>composition |
|---------|-------------------------|
| HEAA    | 11.4                    |
| M-220   | 57.1                    |
| ACMO    | 11.4                    |
| AAEM    | 4.6                     |
| UP-1190 | 11.4                    |

|         |     |
|---------|-----|
| IRG 907 | 2.8 |
| DETX-S  | 1.3 |

[0164]

It should be noted that after the protective film and the polarizer were laminated with the interposed adhesive, the adhesive was cured by irradiation with ultraviolet light to form an adhesive layer. For irradiation with ultraviolet rays, a gallium-encapsulated metal halide lamp (trade name "Light HAMMER 10" manufactured by Fusion UV Systems, Inc., bulb: V bulb, peak illuminance: 1600 mW/cm<sup>2</sup>, integrated irradiation amount: 1000/mJ/cm<sup>2</sup> (wavelength 380 to 440 nm)) was used.

[0165]

[Retardation film]

The retardation film (a quarter wavelength retardation plate) of this example was a retardation film composed of two layers of a retardation layer for a quarter wavelength plate and a retardation layer for a half wavelength plate, in which a liquid crystal material is aligned and fixed. Specifically, such retardation film was manufactured as follows.

[0166]

(Liquid Crystal Material)

A polymerizable liquid crystal material (trade name:

Paliocolor LC242, manufactured by BASF) showing a nematic liquid crystal phase was used as a material for forming a retardation layer for a half wavelength plate and a retardation layer for a quarter wavelength plate. A photopolymerization initiator (trade name Irgacure 907, manufactured by BASF) for the polymerizable liquid crystal material was dissolved in toluene. Further, for the purpose of improving the coating property, a MEGAFACE series manufactured by DIC was added in an amount of about 0.1 to 0.5% according to the liquid crystal thickness to prepare a liquid crystal coating solution. The liquid crystal coating solution was applied on an alignment base material with a bar coater, dried by heating at 90°C for 2 minutes, and subjected to alignment fixation by ultraviolet curing under a nitrogen atmosphere. As the base material, for example, one capable of transferring the liquid crystal coating layer later, such as PET, was used. Further, for the purpose of improving coatability, a fluorine-based polymer which is a MEGAFACE series made by DIC Corporation was added in an amount of about 0.1% to 0.5% depending on the thickness of the liquid crystal layer, and MIBK (methyl isobutyl ketone), cyclohexanone, or a mixed solvent of MIBK and cyclohexanone was used to dissolve the polymer to a solid content concentration of 25%, thereby to prepare a coating solution. This coating solution was applied on a

base material with a wire bar, dried at 65°C for 3 minutes, and subjected to alignment fixation by ultraviolet curing under a nitrogen atmosphere to perform the preparation. As the base material, for example, one capable of transferring the liquid crystal coating layer later, such as PET, was used.

[0167]

(Manufacturing Process)

The manufacturing process of the present embodiment will be described with reference to Fig. 7. The numbers in Fig. 7 are different from the numbers in other drawings. In this manufacturing process 20, a base material 14 was provided by a roll, and this base material 14 was supplied from a supply reel 21. In the manufacturing process 20, a coating solution of an ultraviolet curable resin 10 was applied to the base material 14 by a die 22. In the manufacturing process 20, a roll plate 30 was a cylindrical shaping mold in which a concavo-convex shape relating to an alignment film for a quarter wavelength plate of a quarter wavelength retardation plate was formed on the peripheral side surface. In the manufacturing process 20, the base material 14 coated with the ultraviolet curable resin is pressed against the circumferential side surface of the roll plate 30 by a pressure roller 24, and the ultraviolet curable resin was irradiated with ultraviolet light by an

ultraviolet irradiation device 25 composed of a high-pressure mercury lamp and then cured. As a result, in the manufacturing process 20, the concavo-convex shape formed on the peripheral side surface of the roll plate 30 was transferred to the base material 14 so as to be at 75° with respect to the MD direction. Thereafter, the base material 14 integrally with the cured ultraviolet curable resin 10 was peeled from the roll plate 30 by a peeling roller 26, and the liquid crystal material was applied by a die 29. After that, the liquid crystal material was cured by irradiation with ultraviolet rays by an ultraviolet irradiation device 27, whereby a configuration relating to the retardation layer for a quarter wavelength plate was formed.

Subsequently, in this step 20, the base material 14 is conveyed to a die 32 by a conveying roller 31, and the coating solution of an ultraviolet curing resin 12 is applied onto the retardation layer for a quarter wavelength plate of the base material 14 by the die 32. In this manufacturing process 20, a roll plate 40 was a cylindrical shaping mold in which a concavo-convex shape relating to the alignment film for a half wavelength plate of the quarter wavelength retardation plate was formed on the circumferential side surface. In the manufacturing process 20, the base material 14 coated with the ultraviolet curing

resin was pressed against the peripheral side surface of the roll plate 40 by a pressure roller 34, and the ultraviolet curable resin was irradiated with ultraviolet rays by an ultraviolet irradiation device 35 composed of a high-pressure mercury lamp, and then cured. As a result, in the manufacturing process 20, the concavo-convex shape formed on the circumferential side surface of the roll plate 40 was transferred onto the base material 14 so as to be at 15° with respect to the MD direction. Thereafter, the base material 14 integrally with the cured ultraviolet curable resin 12 was peeled from the roll plate 40 by a peeling roller 36, and the liquid crystal material was applied thereon by a die 39. After that, the liquid crystal material was cured by irradiation with ultraviolet rays by an ultraviolet irradiation device 37, whereby a configuration relating to the retardation layer for a half wavelength plate was obtained. Thus, a retardation film having a thickness of 7  $\mu\text{m}$  and composed of two layers of a retardation layer for a quarter wavelength plate and a retardation layer for a half wavelength plate was obtained.

[0168]

[Optical Film (Optical Laminate)]

The retardation film obtained as described above and the polarizing film obtained as described above were continuously laminated by the roll-to-roll method using the

adhesive to prepare a laminated film (optical laminate) so that an axis angle became 45° between the slow axis and the absorption axis.

[0169]

<Production of optical laminate with pressure-sensitive adhesive layer>

A separator in which the pressure-sensitive adhesive layer (second) obtained in the above Examples and Comparative Examples was formed was transferred to the protective film side (corona-treated) of the obtained optical laminate, and an optical laminate with a pressure-sensitive adhesive layer was produced.

[0170]

[First pressure-sensitive adhesive layer]

The same pressure-sensitive adhesive layer (first) as that of respective examples was formed except that the thickness of the pressure-sensitive adhesive layer used in each Example was changed to 50  $\mu\text{m}$ . A separator having the pressure-sensitive adhesive layer (first) formed therein was transferred to a surface (corona-treated) of a 75  $\mu\text{m}$  thick polyimide film (PI film, manufactured by Du Pont-Toray Co., Ltd., KAPTON 300V, base material), and a PI film with a pressure-sensitive adhesive layer was formed.

[0171]

[Third pressure-sensitive adhesive layer]

The same pressure-sensitive adhesive layer (third) as that of each Example was formed except that the thickness of the pressure-sensitive adhesive layer used in each Example was changed to 50  $\mu\text{m}$ . A separator having the pressure-sensitive adhesive layer (third) formed therein was transferred to a surface (corona-treated) of a 125  $\mu\text{m}$  thick PET film (transparent base material, manufactured by Mitsubishi Plastics Inc., trade name: Diafoil), and a PET film with a pressure-sensitive adhesive layer was formed.

[0172]

<Production of laminate for flexible image display device>

As shown in FIG. 6, for the first to third pressure-sensitive adhesive layers (along with each transparent base material) obtained as described above, a second pressure-sensitive adhesive layer 12-2 was laminated on a PET film having a thickness of 25  $\mu\text{m}$  to serve as a transparent base material 8-1, a third pressure-sensitive adhesive layer 12-3 was laminated on a retardation film 3, and, in addition, a first pressure-sensitive adhesive layer 12-1 was laminated on the transparent base material 8-1 (PET film) having the second pressure-sensitive adhesive layer 12-2 thereon, whereby the laminate 11 for a flexible image display device was produced.

[0173]

[Evaluation]

The following evaluations were performed on the pressure-sensitive adhesive layers and the laminate bodies for a flexible image display device obtained in Examples and Comparative Examples. The results are shown in Table 2.

[0174]

<Storage elastic modulus  $G'$  of pressure-sensitive adhesive layer>

The separator was peeled off from the pressure-sensitive adhesive layer obtained in each Example and Comparative Example, and a plurality of pressure-sensitive adhesive layers were laminated to prepare a test sample having a thickness of about 2 mm. The test sample was stamped into a disk having a diameter of 7.9 mm and sandwiched between parallel plates, and the dynamic viscoelasticity was measured under the following conditions using "Advanced Rheometric Expansion System (ARES)" manufactured by Rheometric Scientific, Inc. The storage elastic moduli  $G'$  of the pressure-sensitive adhesive layer at  $-20^{\circ}\text{C}$ ,  $23^{\circ}\text{C}$ , and  $85^{\circ}\text{C}$  were read from the measurement results.

(Measurement condition)

Deformation mode: twisting

Measurement temperature:  $-70^{\circ}\text{C}$  to  $150^{\circ}\text{C}$

Rate of temperature increase: 5°C/min

[0175]

<Measurement of gel fraction of pressure-sensitive adhesive layer>

About 0.2 g was scraped from the pressure-sensitive adhesive layer formed on a release-treated surface of the separator film after 1 week of production and used as a sample 1. The sample 1 was wrapped with a Teflon (registered trademark) film (trade name "NTF1122", manufactured by Nitto Denko Corporation) having a diameter of 0.2  $\mu\text{m}$ , then strapped with a kite string, and used as a sample 2. The weight of the sample 2 before being subjected to the following test was measured, which was taken as a weight A. The weight A is a total weight of the sample 1 (pressure-sensitive adhesive layer), the Teflon (registered trademark) film, and the kite string. A total weight of the Teflon (registered trademark) film and the kite string was defined as a weight B. Then, the sample 2 was placed in a 50 ml container filled with ethyl acetate and left standing at 23°C for 1 week. Thereafter, the sample 2 was taken out from the container, and dried in a dryer at 130°C for 2 hours to remove ethyl acetate, and then the weight of the sample 2 was measured. The weight of the sample 2 after being subjected to the above test was measured, which was taken as a weight C. Then, the gel

fraction (% by weight) was calculated from the following formula.

$$\text{Gel fraction (wt\%)} = (C-B)/(A-B) \times 100$$

[0176]

The gel fraction of the pressure-sensitive adhesive layer is preferably 55 to 90% by weight, more preferably 57 to 90% by weight, still more preferably 60 to 88% by weight, even more preferably 62 to 88% by weight, particularly preferably 65 to 86% by weight, and most preferably 70 to 86% by weight. When the gel fraction of the pressure-sensitive adhesive layer is within the above range, the appearance (glue dents, etc.), processability, durability, and flexibility are improved. In particular, the flexibility in normal temperature and high temperature environments can be easily simultaneously achieved, which is a preferred embodiment.

[0177]

(Measurement of Thickness)

The thickness of each of the polarizer, the retardation film, the protective film, the optical laminate, and the pressure-sensitive adhesive layer was calculated together with measurement using a dial gauge (manufactured by Mitutoyo Corporation).

[0178]

<Folding endurance (continuous bending) test method>

Figs. 5(A) and 5(B) each show a schematic diagram of a bending test based on a U-shape folding tester (Yuasa System Co., Ltd.).

The tester has a mechanism in which a planar body workpiece is repeatedly bent at 180° in a U shape with no load in a constant temperature bath, and a bending radius can be changed by adjusting a distance between surfaces bent into a U shape.

In the test, a 2.5 cm × 10 cm laminate for a flexible image display device obtained in each Example and Comparative Example was set in the tester so as to be bent in the long side direction, and evaluation was performed at -20°C, 25°C × 50% RH, and 85°C under conditions of a bending angle of 180°, a bending radius of 3 mm, and a bending rate of 1 second/time.

As a sample for measurement (evaluation), the configuration shown in Fig. 6 was adopted, a transparent base material 8-2 (PET film) was on the concave side (inside), and a base material 9 (PI film) was on the convex side (outside). The evaluation was performed by bending the sample in the long side direction near the center. The folding endurance was evaluated by the number of times until cracking or peeling between layers occurred at the bent portion of the laminate for a flexible image display device. Here, when the number of bends reached 200,000,

the test was terminated.

<Presence of peeling/cracking>

⊙: No defect occurs when the number of bends is 200,000 times or more (no problem in practical use)

○: Defect occurs when the number of bends is 100,000 to less than 200,000 times (no problem in practical use)

△: Defect occurs when the number of bends is 50,000 to less than 100,000 times (no problem in practical use)

×: Defect occurs when the number of bends is less than 50,000 times (there is a problem in practical use)

[0179]

<Measurement of modulus stress>

Using the same pressure-sensitive adhesive composition as those in Examples and Comparative Examples, a pressure-sensitive adhesive layer with a thickness of 50 μm having release films (trade name: "MRF#38," manufactured by Mitsubishi Plastics, Inc.) on both sides was prepared. After the pressure-sensitive adhesive layer was cut into a width of 30 mm and a length of 100 mm, the release films on both sides were peeled off, and the pressure-sensitive adhesive layer was rolled up to prevent entrance of bubbles and formed into a columnar shape as a measurement sample. This sample was measured using a Tensilon type tensile tester under conditions of a distance between chucks of 10 mm and a tensile speed of 300 mm/min, and from the obtained

load-elongation curve, stresses (N/mm<sup>2</sup>) at moduli at 100%, 500% and 700% elongation were determined. Here, 100% elongation refers to a state in which the distance between chucks is 20 mm, 500% elongation refers to a state in which the distance between chucks is 60 mm, and 700% elongation refers to a state in which the distance between chucks is 80 mm.

[0180]

<Adhesiveness>

The PI film, optical laminate, and PET film used above were adhesively fixed onto a glass plate via a general-purpose double-sided tape (manufactured by Nitto Denko Corporation, No. 500) and prepared as an adherend.

Using the same pressure-sensitive adhesive composition as those in Examples and Comparative Examples, a pressure-sensitive adhesive layer with a thickness of 50  $\mu$ m having release films (trade name: "MRF#38," manufactured by Mitsubishi Plastics, Inc.) on both sides was prepared. The pressure-sensitive adhesive layer was cut into a size of 25 mm in width and 100 mm in length, one of the release films was peeled off and laminated on a PET film (manufactured by Toray Industries, Inc., Lumirror S10) having a thickness of 25  $\mu$ m, and the other separator was then peeled off to expose a pressure-sensitive adhesive layer surface. This was used as a sample, and the sample

was laminated on the adherend produced earlier. Pressure-bonding was performed at linear pressures of 78.5 N/cm and 0.3 m/min, respectively. After this was left for 30 minutes in an environment of 23°C and 50% RH, the sample was peeled off from the adherend under conditions of a peeling speed of 0.3 m/min and a peeling angle of 180° using a universal tensile tester in the same environment. The peeling force at this time was taken as the adhesion (N/25 mm), and evaluation was performed.

[0181]

[Table 2]

|                       | PRESSURE-SENSITIVE ADHESIVE LAYER |  |                    |                                     |                                 |          |          |            |                          |                             | EVALUATION                             |   |                                     |            |                   |        |        |        |     |                  |     |                 |             |        |
|-----------------------|-----------------------------------|--|--------------------|-------------------------------------|---------------------------------|----------|----------|------------|--------------------------|-----------------------------|----------------------------------------|---|-------------------------------------|------------|-------------------|--------|--------|--------|-----|------------------|-----|-----------------|-------------|--------|
|                       | ACRYLIC POLYMER                   |  |                    |                                     | STORAGE ELASTIC MODULUS G' (Pa) |          |          |            |                          |                             | BENDING TEST (LAMINATION CONSTITUTION) |   | MODULAR STRESS (N/cm <sup>2</sup> ) |            | ADHESION (N/25mm) |        |        |        |     |                  |     |                 |             |        |
|                       | MONOMER MIXTURE OF PREPOLYMER     |  | ADDITIONAL MONOMER |                                     | -20°C                           | 23°C     | 85°C     | DIFFERENCE | AVERAGE VALUE            |                             | GEL FRACTION                           |   | -20°C                               | 23°C/50RH% | 85°C              | 100%   | 500%   | 700%   | PI  | POLARIZING PLATE | PET |                 |             |        |
|                       |                                   |  | TYPE               | BLENDING RATIO (PART (S) BY WEIGHT) |                                 |          |          |            | NVP (PART (S) BY WEIGHT) | A-HD-N (PART (S) BY WEIGHT) |                                        |   |                                     |            |                   |        |        |        |     |                  |     | (-20, 23, 85°C) | (-20, 23°C) | (wt %) |
|                       |                                   |  |                    |                                     |                                 |          |          |            |                          |                             |                                        |   |                                     |            |                   |        |        |        |     |                  |     |                 |             |        |
| Example 1             | LA/2EHA/4HBA                      |  | 59/40/1            | 5                                   | 0.15                            | 7.4.E+04 | 1.7.E+04 | 1.1.E+04   | 5.7.E+03                 | 4.5.E+04                    | 84.6                                   | ⊙ | ⊙                                   | ⊙          | ⊙                 | 0.0363 | 0.1118 | 0.1675 | 3.9 | 3.3              | 3.9 |                 |             |        |
| EXAMPLE 2             | LA/2EHA/4HBA                      |  | 59/40/1            | 10                                  | 0.15                            | 1.7.E+05 | 3.5.E+04 | 2.8.E+04   | 6.8.E+03                 | 1.0.E+05                    | 88.8                                   | ⊙ | ⊙                                   | ⊙          | ⊙                 | 0.0408 | 0.1343 | 0.1836 | 3.4 | 4.3              | 5.1 |                 |             |        |
| EXAMPLE 3             | LA/2EHA/NVP/4HBA                  |  | 50/40/9/1          | 0                                   | 0.15                            | 1.5.E+05 | 2.9.E+04 | 2.2.E+04   | 6.5.E+03                 | 9.1.E+04                    | 89.3                                   | ⊙ | ⊙                                   | ⊙          | ⊙                 | 0.0396 | 0.1403 | 0.2121 | 7.1 | 7.3              | 7.1 |                 |             |        |
| COMPARATIVE EXAMPLE 1 | LA/2EHA/4HBA                      |  | 59/40/1            | 0                                   | 0.15                            | 3.5.E+04 | 1.3.E+04 | 1.3.E+04   | -9.9.E+01                | 2.4.E+04                    | 84.1                                   | ⊙ | ⊙                                   | ×          | ⊙                 | 0.0198 | 0.0619 | 0.1046 | 1.8 | 1.8              | 1.5 |                 |             |        |
| COMPARATIVE EXAMPLE 2 | 2EHA/4HBA                         |  | 99/1               | 0                                   | 0.15                            | 9.4.E+04 | 3.0.E+04 | 2.5.E+04   | 5.1.E+03                 | 6.2.E+04                    | 81.1                                   | ⊙ | ⊙                                   | ×          | ⊙                 | 0.0318 | 0.1381 | 0.1640 | 1.2 | 1.2              | 1.3 |                 |             |        |
| COMPARATIVE EXAMPLE 3 | LA/4HBA                           |  | 99/1               | 0                                   | 0.3                             | 1.1.E+08 | 1.4.E+04 | 1.5.E+04   | -2.7.E+02                | 5.6.E+07                    | 77                                     | × | ⊙                                   | ×          | ⊙                 | 0.0215 | 0.0857 | 0.1040 | 1.0 | 1.0              | 1.6 |                 |             |        |
| COMPARATIVE EXAMPLE 4 | LA/2EHA/4HBA                      |  | 79/20/1            | 0                                   | 0.3                             | 7.2.E+06 | 1.3.E+04 | 1.3.E+04   | -7.1.E+01                | 3.6.E+06                    | 73.4                                   | × | ⊙                                   | ×          | ⊙                 | 0.0221 | 0.0709 | 0.0869 | 1.1 | 1                | 1.3 |                 |             |        |
| COMPARATIVE EXAMPLE 5 | LA/2EHA/4HBA                      |  | 59/40/1            | 15                                  | 0.15                            | 3.3.E+05 | 4.0.E+04 | 3.4.E+04   | 5.9.E+03                 | 1.9.E+05                    | 92.1                                   | × | ○                                   | ⊙          | ⊙                 | 0.0562 | 0.1892 | 0.2232 | 3.9 | 5.1              | 5.7 |                 |             |        |

[0182]

Abbreviations in Table 2 are as follows.

LA: Lauryl acrylate

2EHA: 2-Ethylhexyl acrylate

4HBA: 4-Hydroxybutyl acrylate

NVP: N-Vinylpyrrolidone

A-HD-N: 1,6-hexanediol diacrylate (manufactured by Shin-Nakamura Chemical Co, Ltd., trade name: A-HD-N)

[0183]

From the evaluation results in Table 2, in all the examples, the storage elastic modulus  $G'$  at  $-20^{\circ}\text{C}$  of the pressure-sensitive adhesive layer was  $3.5 \times 10^4$  to  $1.7 \times 10^5$  Pa, the storage elastic modulus  $G'$  at  $23^{\circ}\text{C}$  was  $1.0 \times 10^4$  to  $5.0 \times 10^4$  Pa, and the difference between the storage elastic modulus  $G'$  at  $23^{\circ}\text{C}$  and the storage elastic modulus  $G'$  at  $85^{\circ}\text{C}$  was  $5.2 \times 10^3$  Pa or more. According to the folding endurance (continuous bending) test, even after exposure at  $-20^{\circ}\text{C}$ ,  $25^{\circ}\text{C} \times 50\% \text{ RH}$ , and  $85^{\circ}\text{C}$  (in a low temperature, normal temperature, or high temperature environment), it could be confirmed that cracking (breaking) and peeling were not at a practically problematic level. That is, in the laminate for a flexible image display device of each Example, by using the pressure-sensitive adhesive layer having a storage elastic modulus  $G'$  in a predetermined range, it could be confirmed

that cracking (breaking) or peeling did not occur with respect to repeated bends and a laminate for a flexible image display device having excellent bending resistance and adhesiveness could be obtained.

[0184]

On the other hand, in Comparative Examples 1 to 4, the storage elastic modulus  $G'$  was not within the predetermined range, and therefore, according to the folding endurance (continuous bending) test, after exposure at  $-20^{\circ}\text{C}$ ,  $25^{\circ}\text{C} \times 50\% \text{ RH}$ , and  $85^{\circ}\text{C}$  (in a low temperature, normal temperature, or high temperature environment), it could be confirmed that cracking (breaking) and peeling were at a practically problematic level, and the bending resistance or the adhesiveness were poor.

#### DESCRIPTION OF REFERENCE SIGNS

[0185]

- 1 Polarizer
- 2 Protective film
- 2-1 Protective film
- 2-2 Protective film
- 3 Retardation layer
- 4-1 Transparent conductive film
- 4-2 Transparent conductive film
- 5-1 Base material film

- 5-2 Base material film
- 6 Transparent conductive layer
- 6-1 Transparent conductive layer
- 6-2 Transparent conductive layer
- 7 Spacer
- 8 Transparent base material
- 8-1 Transparent base material (PET film)
- 8-2 Transparent base material (PET film)
- 9 Base material (PI film)
- 10 Organic EL display panel
- 10-1 Organic EL display panel (With touch sensor)
- 11 Laminate for flexible image display device
- (laminate for organic EL display device)
- 12 Pressure-sensitive adhesive layer
- 12-1 First pressure-sensitive adhesive layer
- 12-2 Second pressure-sensitive adhesive layer
- 12-3 Third pressure-sensitive adhesive layer
- 13 Decorative printing film
- 14 Double-sided pressure-sensitive adhesive tape
- 20 Optical laminate
- 30 Touch panel
- 40 Window
- 100 Flexible image display device (organic EL display device)

## CLAIMS

1. A pressure-sensitive adhesive layer for a flexible image display device, having a storage elastic modulus  $G'$  at  $-20^{\circ}\text{C}$  of  $3.5 \times 10^4$  to  $1.7 \times 10^5$  Pa and a storage elastic modulus  $G'$  at  $23^{\circ}\text{C}$  of  $1.0 \times 10^4$  to  $5.0 \times 10^4$  Pa, and a difference between the storage elastic modulus  $G'$  at  $23^{\circ}\text{C}$  and the storage elastic modulus  $G'$  at  $85^{\circ}\text{C}$  being  $5.2 \times 10^3$  Pa or more.

2. The pressure-sensitive adhesive layer for a flexible image display device according to claim 1, wherein an average value of the storage elastic modulus  $G'$  at  $-20^{\circ}\text{C}$  and the storage elastic modulus  $G'$  at  $23^{\circ}\text{C}$  is  $4.5 \times 10^4$  to  $1.5 \times 10^5$  Pa.

3. The pressure-sensitive adhesive layer for a flexible image display device according to claim 1 or 2, wherein a gel fraction is 70% by weight or more.

4. The pressure-sensitive adhesive layer for a flexible image display device according to any one of claims 1 to 3, which is formed of a pressure-sensitive adhesive composition comprising a (meth)acrylic polymer containing an alkyl (meth)acrylate as a monomer unit.

5. The pressure-sensitive adhesive layer for a flexible image display device according to claim 4, wherein the alkyl (meth)acrylate comprises an alkyl (meth)acrylate having an alkyl group having 10 or more carbon atoms.

6. The pressure-sensitive adhesive layer for a flexible image display device according to claim 4 or 5, wherein the (meth)acrylic polymer comprises, as a monomer unit, an N-vinyl group-containing lactam monomer in addition to the alkyl (meth)acrylate.

7. A laminate for a flexible image display device comprising:

the pressure-sensitive adhesive layer according to any one of claims 1 to 6; and

an optical film containing at least a polarizer.

8. A flexible image display device comprising:

the laminate for a flexible image display device according to claim 7; and

an organic EL display panel,

wherein the laminate for a flexible image display device is disposed on a viewing side with respect to the organic EL display panel.

9. The flexible image display device according to claim 8, wherein a window is disposed on the viewing side with respect to the laminate for a flexible image display device.