resin composition such as those used for producing color filters for liquid crystal display devices (see, for example, Patent document 3). In general, however, an acrylic resin is used as the negative type photosensitive resin composition, leading to poor heat resistance and resulting in a light emitting element with a decreased reliability. To provide a photosensitive resin composition with an increasing the sensitivity, it is known that the use of a negative type photosensitive is essentially more advantageous than the use of a positive type photosensitive from the viewpoint of the photosensitivity development mechanism. Accordingly, a negative type photosensitive resin composition is generally used to increase the sensitivity. However, since the surface of the film is photo-cured first according to the negative type photosensitivity mechanism, a rectangular or a backward tapered pattern shape tends to be formed while those regions where the exposure is small tend to fail to be photo-cured sufficiently, frequently resulting in the absence of patterns being left after the alkaline development step. Therefore, it is difficult for a negative type photosensitive resin composition to form a step-shaped pixel dividing layer having a sufficient thickness difference between the thick parts and the thin parts even when a halftone photomask is used. A known method for producing a negative photosensitive resin composition with an increased sensitivity is to increase the content of a compound having a plurality of ethylenically unsaturated double bond groups in the molecule, but this can lead to a pattern shape having a rectangular or backward tapered cross section. Thus, there has been a call for a highly sensitive negative type photosensitive resin composition that has excellent characteristics helpful to form a step-shaped pattern in a batch process using a halftone photomask (hereinafter referred to as halftone characteristics) and serves to produce a small-tapered pattern shape.

PRIOR ART DOCUMENTS

PATENT DOCUMENTS

[0008]

[Patent document 1] Japanese Unexamined Patent Publication (Kokai) No. 2002-091343
[Patent document 2] Japanese Unexamined Patent Publication (Kokai) No. 2005-322564
[Patent document 3] Japanese Unexamined Patent Publication (Kokai) No. 2015-093986

SUMMARY

PROBLEMS TO BE SOLVED

[0009]

However, none of the conventionally known photosensitive resin compositions have required characteristics to serve as a material for forming a step-shaped pixel dividing layer of an organic EL display device. Specifically, they are inferior in terms of sensitivity, halftone characteristics, or pattern shape.

[0010]
Thus, an object of the embodiments disclosed herein is to provide a negative type photosensitive resin composition having high sensitivity, excellent halftone characteristics, capability to form a small-tapered pattern shape, and alkali-developability.
[0011]
As another problem, it is difficult for the conventionally known negative type photosensitive resin compositions to form a step-shaped pixel dividing layer and accordingly a contact between the vapor deposition mask and the pixel dividing layer can result in a light emitting element with a poor reliability.
[0012]
Thus, another object of the embodiments disclosed herein is to provide an organic EL display device containing a step-shaped pixel dividing layer with a sufficiently large thickness difference between the thick parts and the thin parts and enabling the production of a light emitting element with high reliability.
[0013]
As still another problem, a complicated process is necessary in some cases to produce a step-shaped pixel dividing layer from a negative type photosensitive resin composition.
[0014]
Thus, still another object of the embodiments disclosed herein is to provide a method to form a step-shaped pattern in a batch process using a halftone photomask and a process using it to produce an organic EL display device.

MEANS OF SOLVING THE PROBLEMS
[0015]
The negative type photosensitive resin composition according to the present disclosure includes, as an alkali-soluble resin (A), at least a weakly acidic group-containing resin (A1) and an unsaturated group-containing resin (A2), the weakly acidic group-containing resin (A1) containing an acidic group having an acid dissociation constant of 13.0 to 23.0 in dimethyl sulfoxide, and the unsaturated group-containing resin (A2) containing an ethylenically unsaturated double bond group.
[0015a]
In another aspect, there is provided a negative type photosensitive resin composition comprising, as an alkali-soluble resin (A), at least a weakly acidic group-containing resin (A1), an unsaturated group-containing resin (A2), a radical-polymerizable compound (B) and a photosensitive agent (C),
the weakly acidic group-containing resin (A1) containing an acidic group having an acid dissociation constant in the range of 13.0 to 23.0 in dimethyl sulfoxide, and
the unsaturated group-containing resin (A2) having an ethylenically unsaturated double

bond group;
wherein the weakly acidic group-containing resin (A1) includes one or more selected from the group consisting of polyimide (A1a-1), polyimide precursor (A1a-2), carboxylic acid-modified polysiloxane (A1a-3), polybenzoxazole (A1b-1), polybenzoxazole precursor (A1b-2), and polysiloxane (A1b-3).

[0015b]

In antoher aspect, there is provided a cured film formed by curing a negative type photosensitive resin composition as defined above.

[0015c]

In another aspect, there is provided an organic EL display device comprising a cured film as defiend above, wherein the cured film serves as at least one selected from pixel dividing layers, color filters, black matrices of color filters, and planarization films for TFTs.

[0015d]

In another aspect, there is provided an organic EL production method for display device comprising:

(1) a step for forming a coating film of a negative type photosensitive resin composition as defined above on a substrate,

(2) a step for applying active actinic ray to the resin composition through a photomask,

(3) a step for developing it with an alkaline solution to form a step-shaped pattern of the resin composition, and,

(4) a step for heating the pattern to produce a step-shaped cured pattern of the resin composition,

the photomask being halftone photomask that has a pattern containing transparent portions and light blocking portions and that also has translucent portions lower in transmittance than the transparent portions, higher in transmittance than the light blocking portions, and located between the transparent portions and the light blocking portions.

ADVANTAGEOUS EFFECT

[0016]

The present disclosure can provide a negative type photosensitive resin composition having high sensitivity, excellent halftone characteristics, capability to form a small-tapered pattern shape, and alkali-developability. The use of this negative type photosensitive resin composition serves to form a step-shaped cured film with a sufficiently large thickness difference between the thick parts and the thin parts and accordingly serves to produce a

CLAIMS

[Claim 1]

A negative type photosensitive resin composition comprising, as an alkali-soluble resin (A), at least a weakly acidic group-containing resin (A1), an unsaturated group-containing resin (A2), a radical-polymerizable compound (B) and a photosensitive agent (C),

the weakly acidic group-containing resin (A1) containing an acidic group having an acid dissociation constant in the range of 13.0 to 23.0 in dimethyl sulfoxide, and

the unsaturated group-containing resin (A2) having an ethylenically unsaturated double bond group;

wherein the weakly acidic group-containing resin (A1) includes one or more selected from the group consisting of polyimide (A1a-1), polyimide precursor (A1a-2), carboxylic acid-modified polysiloxane (A1a-3), polybenzoxazole (A1b-1), polybenzoxazole precursor (A1b-2), and polysiloxane (A1b-3).

[Claim 2]

A negative type photosensitive resin composition as set forth in claim 1, wherein the weakly acidic group-containing resin (A1) includes a double acidic group-containing resin (A1a) and a weakly acidic group-containing second resin (A1b), and

the double acidic group-containing resin (A1a) includes one or more selected from the group consisting of the following as described later: polyimide (A1a-1), polyimide precursor (A1a-2), carboxylic acid-modified polysiloxane (A1a-3), carboxylic acid-modified novolac resin (A1a-4), and carboxylic acid-modified polyhydroxystyrene (A1a-5), and

the weakly acidic group-containing second resin (A1b) includes one or more selected from the group consisting of the following as described later: polybenzoxazole (A1b-1), polybenzoxazole precursor (A1b-2), polysiloxane (A1b-3), novolac resin (A1b-4), and polyhydroxystyrene (A1b-5).

[Claim 3]

A negative type photosensitive resin composition as set forth in either claim 1 or 2, wherein the weakly acidic group-containing resin (A1) having one or more acidic groups selected from the group consisting of phenolic hydroxyl group, silanol group, hydroxyimide group, and hydroxyamide group.

[Claim 4]

A negative type photosensitive resin composition as set forth in any one of claims 1 to 3 further comprising a coloring agent (D).

[Claim 5]

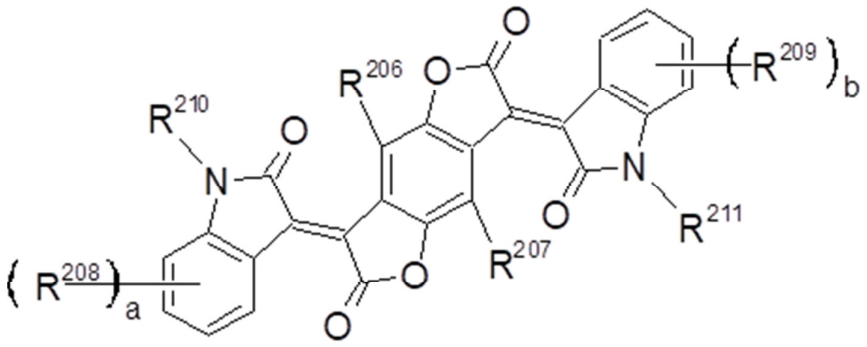
A negative type photosensitive resin composition as set forth in claim 4, wherein the coloring agent (D) contains a pigment (D1), and the pigment (D1) contains a black pigment (D1a).

[Claim 6]

A negative type photosensitive resin composition as set forth in claim 5, wherein the black pigment (D1a) contains a benzofuranone based black pigment (D1a-1a) and/or a perylene based black pigment (D1a-1b) as a black organic pigment (D1a-1), and

the benzofuranone based black pigment (D1a-1a) includes a benzofuranone compound as represented by any of general formulae (63) to (68), a geometrical isomer thereof, a salt thereof, or a geometrical isomer salt thereof, and

the perylene based black pigment (D1a-1b) includes a perylene compound as represented by any of general formulae (69) to (71), a geometrical isomer thereof, a salt thereof, or a geometrical isomer salt thereof;



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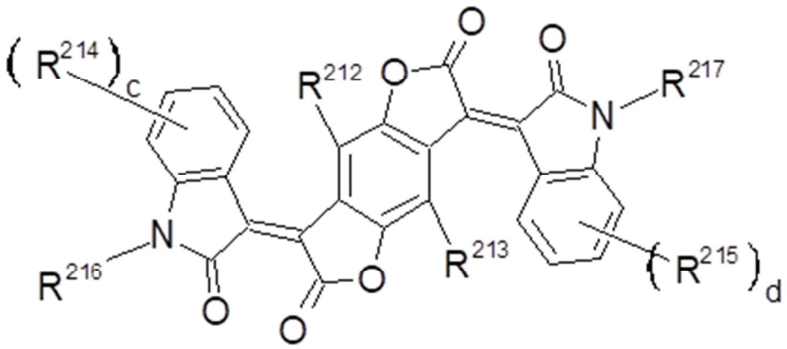

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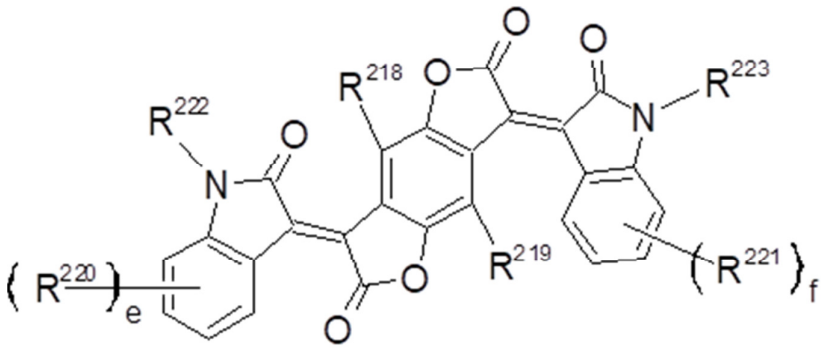
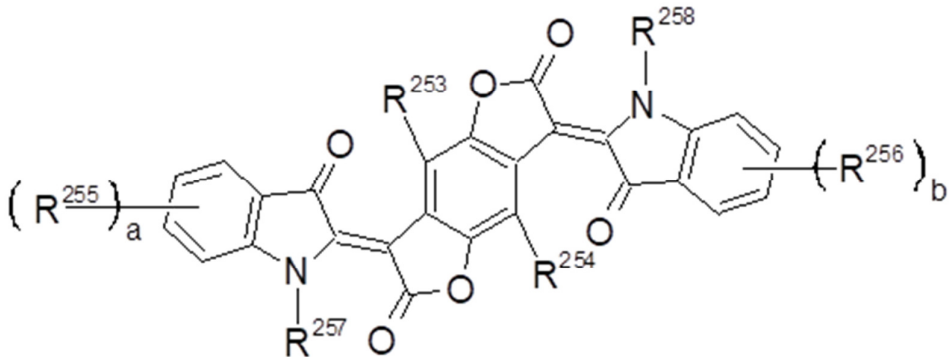


(65)

wherein in general formulae (63) to (65), R^{206}, R^{207}, R^{212}, R^{213}, R^{218}, and R^{219} are each independently a hydrogen atom, a halogen atom, an alkyl group containing 1 to 10 carbon

atoms, or an alkyl group containing 1 to 10 carbon atoms and 1 to 20 fluorine atoms; R^{208}, R^{209}, R^{214}, R^{215}, R^{220}, and R^{221} are each independently a hydrogen atom, a halogen atom, R^{251}, COOH, $COOR^{251}$, COO-, $CONH_2$, $CONHR^{251}$, $CONR^{251}R^{252}$, CN, OH, OR^{251}, $OCOR^{251}$, $OCONH_2$, $OCONHR^{251}$, $OCONR^{251}R^{252}$, NO_2, NH_2, NHR^{251}, $NR^{251}R^{252}$, $NHCOR^{251}$, $NR^{251}COR^{252}$, $N=CH_2$, $N=CHR^{251}$, $N=CR^{251}R^{252}$, SH, SR^{251}, SOR^{251}, SO_2R^{251}, SO_3R^{251}, SO_3H, SO_3^-, SO_2NH_2, SO_2NHR^{251}, or $SO_2NR^{251}R^{252}$, and R^{251} and R^{252} are each independently an alkyl group containing 1 to 10 carbon atoms, a cycloalkyl group containing 4 to 10 carbon atoms, an alkenyl group containing 2 to 10 carbon atoms, a cycloalkenyl group containing 4 to 10 carbon atoms, or alkynyl group containing 2 to 10 carbon atoms; a plurality selected from R^{208}, R^{209}, R^{214}, R^{215}, R^{220}, and R^{221} may be connected by a direct bond, oxygen atom bridge, sulfur atom bridge, NH bridge, or NR^{251} bridge to form a ring; R^{210}, R^{211}, R^{216}, R^{217}, R^{222}, and R^{223} are each independently a hydrogen atom, an alkyl group containing 1 to 10 carbon atoms, or an aryl group containing 6 to 15 carbon atoms; and furthermore, a, b, c, d, e, and f are each independently an integer of 0 to 4;



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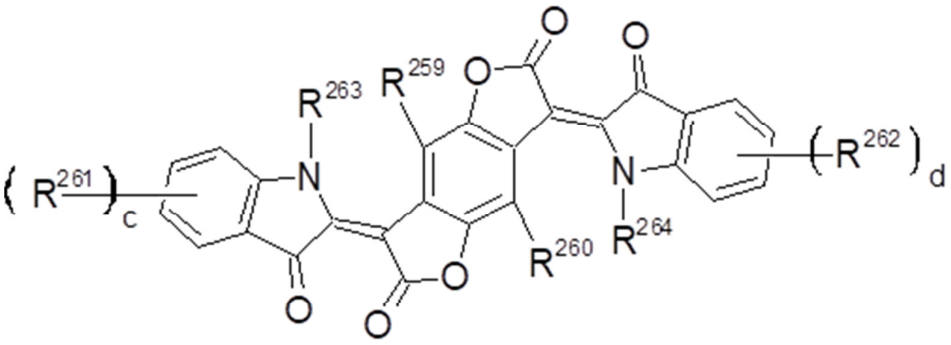

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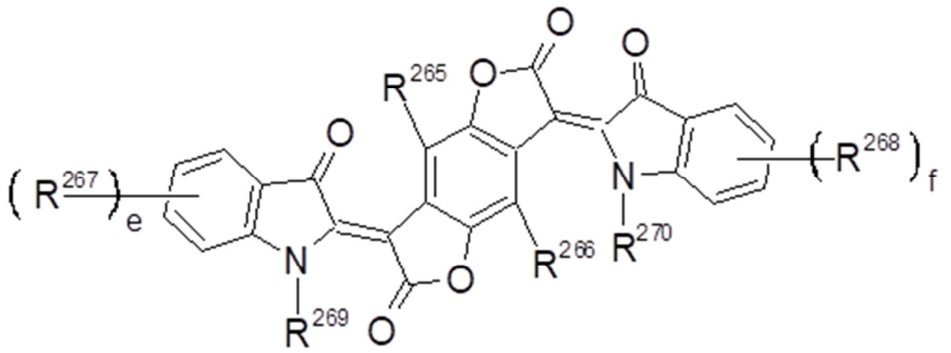
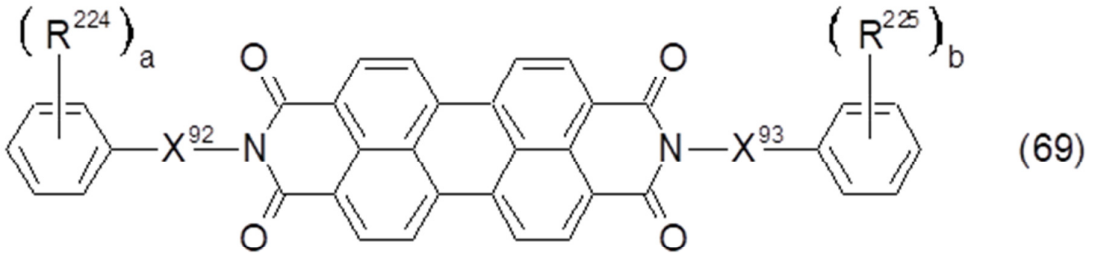


(68)

wherein in general formulae (66) to (68), R^{253}, R^{254}, R^{259}, R^{260}, R^{265}, and R^{266} are each independently a hydrogen atom, a halogen atom, an alkyl group containing 1 to 10 carbon atoms, or an alkyl group containing 1 to 10 carbon atoms and 1 to 20 fluorine atoms; R^{255}, R^{256}, R^{261}, R^{262}, R^{267}, and R^{268} are each independently a hydrogen atom, a halogen atom, R^{271}, COOH, $COOR^{271}$, COO-, $CONH_2$, $CONHR^{271}$, $CONR^{271}R^{272}$, CN, OH, OR^{271}, $OCOR^{271}$, $OCONH_2$, $OCONHR^{271}$, $OCONR^{271}R^{272}$, NO_2, NH_2, NHR^{271}, $NR^{271}R^{272}$, $NHCOR^{271}$, $NR^{271}COR^{272}$, $N=CH_2$, $N=CHR^{271}$, $N=CR^{271}R^{272}$, SH, SR^{271}, SOR^{271}, SO_2R^{271}, SO_3R^{271}, SO_3H, SO_3^-, SO_2NH_2, SO_2NHR^{271}, or $SO_2NR^{271}R^{272}$, and R^{271} and R^{272} are each independently an alkyl group containing 1 to 10 carbon atoms, a cycloalkyl group containing 4 to 10 carbon atoms, an alkenyl group containing 2 to 10 carbon atoms, a cycloalkenyl group containing 4 to 10 carbon atoms, or alkynyl group containing 2 to 10 carbon atoms; a plurality selected from R^{255}, R^{256}, R^{261}, R^{262}, R^{267}, and R^{268} may be connected by a direct

bond, oxygen atom bridge, sulfur atom bridge, NH bridge, or NR^{271} bridge to form a ring; R^{257}, R^{258}, R^{263}, R^{264}, R^{269}, and R^{270} are each independently a hydrogen atom, an alkyl group containing 1 to 10 carbon atoms, or an aryl group containing 6 to 15 carbon atoms; and furthermore, a, b, c, d, e, and f are each independently an integer of 0 to 4;



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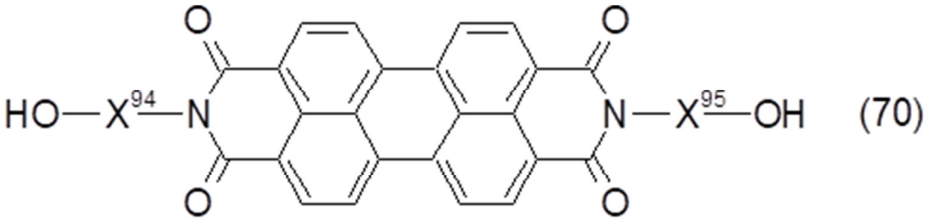

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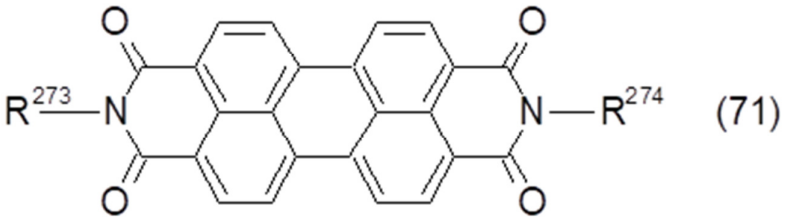

(71)

wherein in general formulae (69) to (71), X^{92}, X^{93}, X^{94}, and X^{95} are each independently an alkylene chain containing 1 to 10 carbon atoms; R^{224} and R^{225} are each independently a hydrogen atom, a hydroxyl group, an alkoxy group containing 1 to 6 carbon atoms, or an acyl group containing 2 to 6 carbon atoms; R^{273} and R^{274} are each independently a hydrogen atom or an alkyl group containing 1 to 10 carbon atoms; and furthermore, a and b are each an integer of 0 to 5.

[Claim 7]

A negative type photosensitive resin composition as set forth in claim 6, wherein the black organic pigment (D1a-1) contains the benzofuranone based black pigment (D1a-1a).

[Claim 8]

A negative type photosensitive resin composition as set forth in any one of claims 1 to 7, wherein the unsaturated group-containing resin (A2) includes one or more selected from the group consisting of unsaturated group-containing polyimide (A2a-1), unsaturated group-containing polyimide precursor (A2a-2), carboxylic acid-modified unsaturated group-containing polysiloxane (A2a-3), unsaturated group-containing polybenzoxazole (A2b-1),

unsaturated group-containing polybenzoxazole precursor (A2b-2), unsaturated group-containing polysiloxane (A2b-3), polycyclic side chain-containing aromatic resin (A2c-1), acrylic resin (A2c-2), and carboxylic acid-modified epoxy resin (A2c-3).

[Claim 9]

A negative type photosensitive resin composition as set forth in claim 8, wherein the unsaturated group-containing resin (A2) includes one or more selected from the group consisting of unsaturated group-containing polyimide (A2a-1), unsaturated group-containing polyimide precursor (A2a-2), carboxylic acid-modified unsaturated group-containing polysiloxane (A2a-3), unsaturated group-containing polybenzoxazole (A2b-1), unsaturated group-containing polybenzoxazole precursor (A2b-2), and unsaturated group-containing polysiloxane (A2b-3).

[Claim 10]

A negative type photosensitive resin composition as set forth in any one of claims 1 to 9, wherein the weakly acidic group-containing resin (A1) having no ethylenically unsaturated double bond group, and

the weakly acidic group-containing resin (A1) accounting for 30 to 99 mass% of the alkali-soluble resin (A).

[Claim 11]

A negative type photosensitive resin composition as set forth in claim 10, wherein the weakly acidic group-containing resin (A1) accounts for 70 to 97 mass% of the alkali-soluble resin (A).

[Claim 12]

A negative type photosensitive resin composition as set forth in any one of claims 1 to 11, wherein a step-shaped cured pattern of the resin composition obtained by a step for forming a coating film of a negative type photosensitive resin composition, a step for applying active actinic ray to the resin composition through halftone photomasks, a step for developing it with an alkaline solution to form a step-shaped pattern of the resin composition, and, a step for heating the step-shaped pattern, and

the film thickness difference (ΔT_{FT-HT}) μm between (T_{FT}) and (T_{HT}) wherein (T_{FT}) μm denotes the film thickness of the thick parts of the transparent portions of the halftone photomasks and (T_{HT}) μm denotes the film thickness of the thin parts of the translucent portions of the halftone photomasks in the stepped shape of the cured pattern is in the range of 1.0 to 10.0 μm.

[Claim 13]

A cured film formed by curing a negative type photosensitive resin composition as set forth in any one of claims 1 to 12.

[Claim 14]

A cured film as set forth in claim 13, wherein the optical density per micrometer of film

thickness of the cured film is in the range of 0.3 to 5.0, and the cured film includes the step-shaped cured pattern.

[Claim 15]

A cured film as set forth in claim 14, wherein the film thickness difference (ΔT_{FT-HT}) μm between (T_{FT}) and (T_{HT}) wherein (T_{FT}) μm denotes the film thickness of the thick parts and (T_{HT}) μm denotes the film thickness of the thin parts in the stepped shape of the cured pattern is in the range of 1.0 to 10.0 μm.

[Claim 16]

A cured film as set forth in claim 15, wherein (T_{FT}) and (T_{HT}) further meet the relations represented by general formulae (δ) to (ζ):

$3.0 \leq (T_{FT}) \leq 9.0$ (δ)

$0.20 \leq (T_{HT}) \leq 7.0$ (ε)

$0.20 \times (T_{FT}) \leq (T_{HT}) \leq 0.70 \times (T_{FT})$ (ζ)

[Claim 17]

A cured film as set forth in any one of claims 14 to 16, wherein an inclined slope in the cross section of a cured pattern having the step-shaped cured pattern has a taper angle in a range of 1° to 60°.

[Claim 18]

An organic EL display device comprising a cured film as set forth in any one of claims 13 to 17, wherein the cured film serves as at least one selected from pixel dividing layers, color filters, black matrices of color filters, and planarization films for TFTs.

[Claim 19]

An organic EL display device as set forth in claim 18 comprising a curved display part having a curvature radius in the range of 0.1 to 10 mm.

[Claim 20]

An organic EL production method for display device comprising:

(1) a step for forming a coating film of a negative type photosensitive resin composition as set forth in any one of claims 1 to 10 on a substrate,

(2) a step for applying active actinic ray to the resin composition through a photomask,

(3) a step for developing it with an alkaline solution to form a step-shaped pattern of the resin composition, and,

(4) a step for heating the pattern to produce a step-shaped cured pattern of the resin composition,

the photomask being halftone photomask that has a pattern containing transparent portions and light blocking portions and that also has translucent portions lower in transmittance than the transparent portions, higher in transmittance than the light blocking portions, and located between the transparent portions and the light blocking portions.