IP HIGH COURT OF KOREA

FIFTH-TWO DIVISION

DECISION

Case No.	2022Heo3809 Invalidation of Registration (Patent)
Plaintiff	A Corp. CEO B Counsel for Plaintiff PLUS INTERNATIONAL IP LAW FIRM Patent Attorney in Charge Changhee PARK
Defendant	C Representative D Counsel for Defendant KOREANA PATENT FIRM Patent Attorney in Charge Suyeon SONG and Donghwan KIM
Date of Closing Argument	April 20, 2023
Decision Date	May 25, 2023

ORDER

1. The plaintiff's claims are dismissed.
2. The cost arising from this litigation shall be borne by the plaintiff.

PLAINTIFF'S DEMAND

The IPTAB Decision 2020Dang3238 dated May 20, 2022 shall be revoked.

OPINION

1. Background

A. Plaintiff's Claimed Invention (Plaintiff's Exhibit 2)

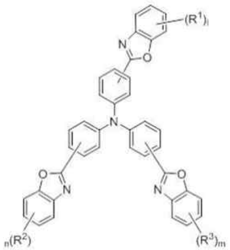
1) Title of invention: Tribenzazole Amine Derivatives and Organic Electroluminescent Device Including the Same

2) Filing date of application / Date of registration / Registration No.: December 12, 2018 / December 26, 2019 / No. 2059550

3) Claims

【Claim 1】 A tribenzazole amine derivative for use as a capping layer in an organic electroluminescent device, represented by the chemical formula below.

[Chemical Formula 1]

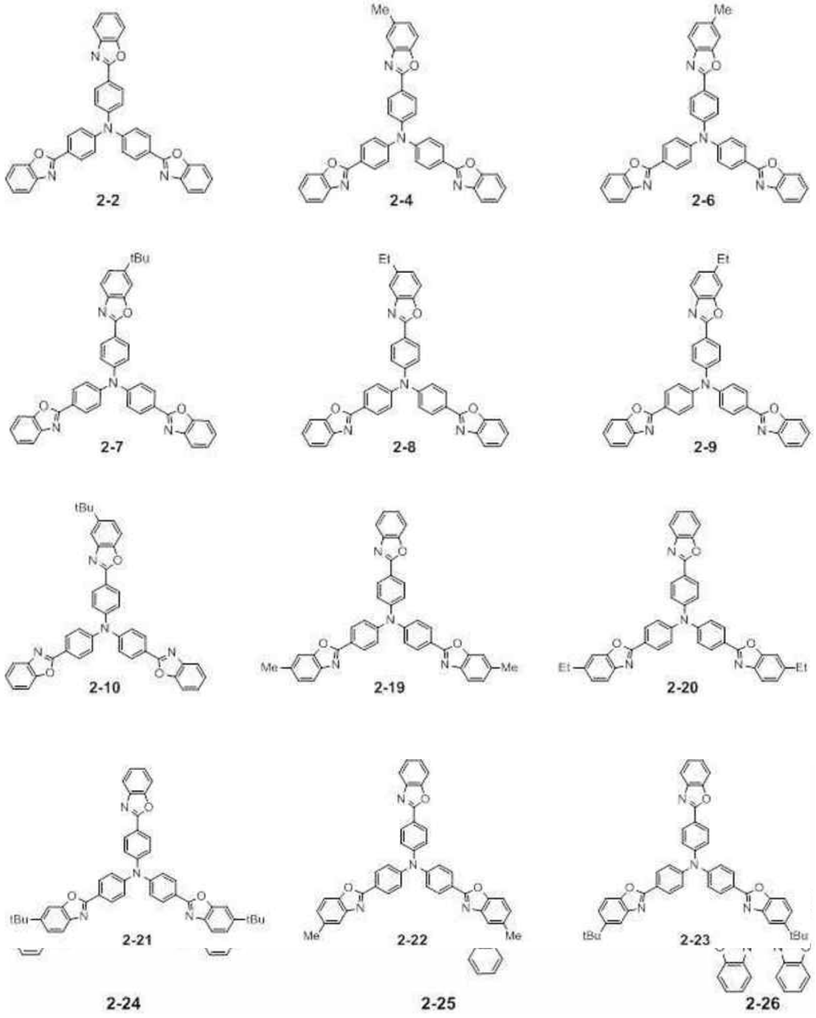


In Chemical Formula 1, R^1, R^2, and R^3 are identical or different, each independently being hydrogen; a C1-C10 alkyl group; and l, n, and m are integers ranging from 0 to 4 (hereinafter, "Claim 1," the same applies to the remaining claims).

【Claim 2】 The tribenzazole amine derivative for use as a capping layer in an organic electroluminescent device according to Claim 1, wherein Chemical

Formula 1 is selected from compounds represented by Chemical Formula 2.

[Chemical Formula 2]



【Claim 3】 An organic light-emitting device comprising: a first electrode; an organic layer comprising a plurality of organic material layers disposed on the first electrode; a second electrode disposed on the organic layer; and a capping layer disposed on the second electrode, wherein the capping layer includes a tribenzazole amine derivative according to any one of Claims 1 or 2.

【Claim 4】 An organic electroluminescent device wherein the organic layer includes a hole injection layer; a hole transport layer; an emitting layer; an electron transport layer; and an electron injection layer, according to Claim 3.

4) Summary of invention

The present invention aims to provide a capping layer material designed to enhance the luminous efficiency, lifespan, and viewing angle characteristics of an organic electroluminescent device (OLED)1), offering a tribenzazole amine derivative with a defined structure. Refer to Appendix 1 for main content of the present invention.

B. Prior Art (Plaintiff's Exhibit 4)2)

This invention, filed on December 21, 2017, and published internationally on June 27, 2019 in Korean Laid-open Patent Publication No. 10-2020-0086747, under the title "Organic Electroluminescence Device and Manufacturing Method Thereof," discloses an amine compound with a benzoazole ring structure suitable for use as a capping layer material in organic electroluminescent devices. Refer to Appendix 2 for main content of the prior art.

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1) Organic Light Emitting Diodes: It is also referred to as an organic electroluminescent device, organic light-emitting device, or organic EL device.

2) The prior art is the Korean translation of WO 2019/124550, which was claimed priority based on Japanese Patent Application No. 2017-244969, filed on December 21, 2017, and was filed internationally on December 21, 2018, and published internationally on June 27, 2019. Since the prior art was applied for registration before the filing date of the subject patent application but published after the date, it qualifies as prior art in relation to the applicability of the expanded first-to-file rule under Article 29(3) of the Patent Act.

C. Procedural History

1) The defendant filed a petition on October 29, 2020, seeking invalidation against the plaintiff in the Korean Intellectual Property Trial and Appeal Board (IPTAB), reasoning, “Claims 1 to 4 at issue all violate the extended first-to-file rule in Article 29(3) of the Patent Act.”

2) The IPTAB reviewed the case under Case No. 2020Dang3238, and issued an administrative decision to invalidate the patented invention, stating “the present invention violates the extended first-to-file rule based on the prior art.”

【Factual Basis】 Undisputed facts, the descriptions on Plaintiff's Exhibits 1 through 6, the purport of the overall arguments

2. Summary of Parties' Arguments and Questions Presented

A. Plaintiff

The subject invention does not violate the extended first-to-file rule due to the following reasons, and the IPTAB decision concluding otherwise is erroneous.

1) Even if the prior art documents disclose the name or chemical structure of a compound, the compound cannot be considered disclosed in the prior art if they fail to provide information on how to prepare the compound, and a person having ordinary skill in the art (hereinafter, a "skilled person") cannot prepare it without undue effort.

2) Although the prior art mentions “compound (27),” corresponding to [Chemical Formula 1] in Claim 1, it does not disclose a method for preparing compound (27), and the preparation method for compound (10), an existing embodiment, cannot be extended to compound (27). Without extensive experimentation, a skilled person would be unable to prepare compound (27). Therefore, compound (27) cannot be considered disclosed in the prior art.

3) Thus, Claim 1 does not violate the extended first-to-file rule based on the prior art. Claims 2 to 4 also do not violate the extended first-to-file rule based on the prior art for the same reasons.

B. Defendant

The subject invention violates the extended first-to-file rule due to the following reasons, and the IPTAB decision is lawful as it is consistent with this conclusion.

1) Whether the prior art discloses a compound shall be determined based on whether a skilled person can directly perceive the presence of the compound from the prior art, based on its specification and the technological common knowledge at the time of the application.

2) The prior art provides a specific description of compound (27), which corresponds to [Chemical Formula 1] in Claim 1. As a result, a skilled person can directly perceive the presence of the compound. Therefore, compound (27) shall be considered disclosed in the prior art.

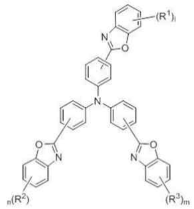
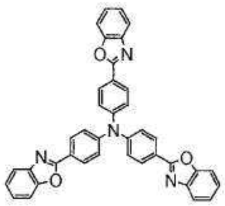
3) Furthermore, regarding the enablement of compound (27), even if the preparation method for compound (27) is not explicitly disclosed in the prior art, a skilled person could readily synthesize and use compound (27) based on the preparation example of compound (10) and the technological common knowledge at the time of the application.

4) Thus, Claim 1 violates the extended first-to-file rule based on the prior art. Claims 2 to 4 also violate the extended first-to-file rule based on the prior art for the same reasons.

3. Whether IPTAB Erred

A. Whether Claim 1 Violates the Extended First-to-File Rule

1) Comparison between Claim 1 and the prior art

Claim 1	Prior Art
A tribenzazole amine derivative for use as a capping layer in an organic electroluminescent device, represented by the chemical formula below.  [Chemical Formula 1] In Chemical Formula 1, R^1, R^2, and R^3 are identical or different, each independently being hydrogen; a C1-C10 alkyl group; and l, n, and m are integers ranging from 0 to 4.	 (27) (Paragraph [0142])

2) Analysis on whether the two inventions are substantially identical

"The compound (27)" described in the prior art in a literal sense is equivalent to the tribenzazole amine derivative in [Chemical Formula 1] where R^1 to R^3 are hydrogen atoms, and there is no dispute between the parties regarding this.

Eventually, regarding whether Claim 1 is equivalent to the invention shown in the specification or the drawing of the prior art and thus violates the extended first-to-file rule in Article 29(3) of the Patent Act, the question is whether the compound (27) in the prior art can be deemed to be an invention disclosed in the prior art. The analysis is provided below.

3) Whether compound (27) is an invention disclosed in the prior art

A) Relevant law

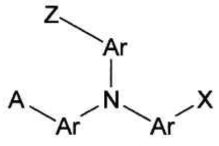
(1) To conclude that the prior art discloses a specific compound, it is insufficient for the compound to be merely encompassed in principle within the chemical formula of a Markush structure or within the range of its substituents. Instead, the compound must be explicitly described in the prior art, or a skilled person in the art must directly recognize the existence of the compound from the prior art, based on its descriptions and the common technical knowledge available at the time of the application (See Supreme Court Decision 2008Hu736,743, dated October 15, 2009).

(2) Meanwhile, even if the specific compound is described in the prior art in a literal sense or a skilled person can directly recognize its existence, in case the compound is inexecutable--such as the person cannot prepare the compound based on the description of the prior art and common technical knowledge at the time of the application--the compound is not deemed to be disclosed in the prior art in determining whether the invention lacks novelty or violates the extended first-to-file rule.

B) Analysis

(1) Detailed description in the compound (27) of the prior art, etc.

(A) The prior art is an invention that aims to provide an organic EL element[3] with good luminous and power efficiency (paragraph [0015]), disclosing amine compounds with capping layer material of organic EL element and a benzoxazole ring structure in General Formula (A-1) (paragraphs [0016] to [0022] and [0097]).

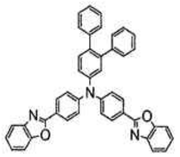

(A−1)

(In General Formula (A-1), A and X may be the same or different, and each represents a monovalent group with one bonding site selected from R_1 to R_6 in General Formula (B-1) below. Z represents a monovalent group with one bonding site selected from R_1 to R_6 in General Formula (B-1), a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, or a substituted or unsubstituted fused polycyclic aromatic group. Ar represents a bivalent group or a single bond, which may be the same or different from one another, selected from substituted or unsubstituted aromatic hydrocarbon groups, substituted or unsubstituted aromatic heterocyclic groups, or substituted or unsubstituted fused polycyclic aromatic groups.)

(B) The prior art discloses 85 compounds as examples of a desirable amine compound in General Formula (A-1), and among them is compound (27).

[0137] Among the compounds represented by General Formula (A-1) or (A-2), which are suitably used in the organic EL device of the present invention, compounds represented by formulas (1) to (85) are presented below as specific examples of desirable compounds.

(The description of the compounds in formulas (1) through (9), (11) through (24), and (28) through (85) is omitted.)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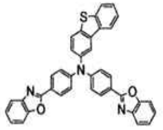
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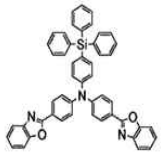
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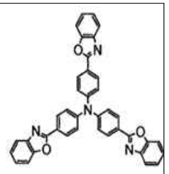
(27)

(C) Therefore, the prior art is deemed to explicitly and thoroughly describe the presence of compound (27). Based on this description, a skilled person could directly recognize compound (27) as a useful material for the capping layer of an organic EL element.

(2) Whether the compound (27) can be executed

(A) Though the prior art does not directly describe a preparation example of compound (27), it shall be deemed that a skilled person could prepare compound (27) from the prior art based on its description and common technical knowledge at the time of the application due to the following reasons.

① According to the description in the specification of the prior art below, the prior art discloses that compounds corresponding to General Formula (A-1), including compound (27), can be prepared using various well-known preparation methods.

o (paragraph [0014]) (non-patent literature 0001) pp. 55-61 of Preview of the 9th Lecture Session of the Korean Physics Society (2001), (non-patent literature 0002) Appl. Phys. Let., 78, 544 (2001), (non-patent literature 0003) Appl. Phys. Let., 82, 466 (2003), (non-patent literature 0004) J. Org. Chem., 71, 1802 (2006), (non-patent literature 0005) J. Org. Ch㎝., 60, 7508 (1995), (non-patent literature 0006) Synth. Co㎜un., 11, 513 (1981), (non-patent literature 0007) Appl. Phys. Lett., 98, 083302 (2011)

o (paragraph [0163]) Amine compounds with a benzoazole ring structure,

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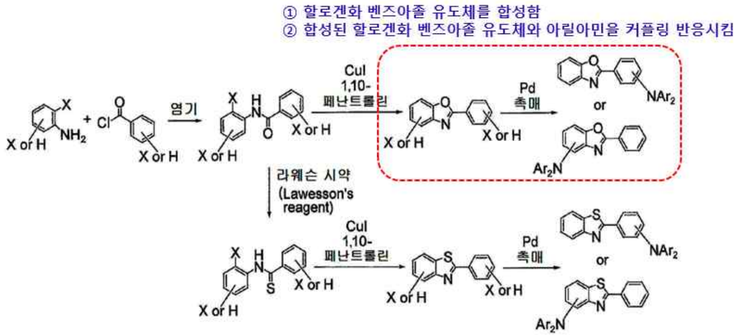
3) It is an abbreviation for organic electroluminescent device.

represented by General Formulas (A-1) and (A-2), can be prepared using the methods described below.

o (paragraph [0164]) First, to form the benzoazole ring structure represented by Formula (B-1) or (B-8), which serve as the main framework of the compounds represented by General Formulas (A-1) and (A-2), a halogenated benzoazole derivative is prepared as the starting material. Halogenated benzoazole derivatives with structures corresponding to the benzoazole ring structure represented by Formula (B-1) or (B-8) can be prepared, for example, using publicly-known methods as described below (See, for instance, non-patent literature 4).

o (paragraph [0165]) Additionally, the synthesized halogenated benzoazole derivative and an arylamine undergo a coupling reaction using a copper catalyst, palladium catalyst, or similar catalysts. Now, the amine compounds with a benzoazole ring structure represented by General Formula (A-1) or (A-2) of the present invention can be synthesized.

o (paragraph [0166]) Also, by subjecting halogenated arylamines and boronic acid derivatives or boronic acid ester derivatives, instead of halogenated benzoazole derivatives, to a coupling reaction, the amine compounds with a benzoazole ring structure represented by General Formulas (A-1) or (A-2) of the present invention can be synthesized (See, for instance, non-patent literature 5 and 6).


① 할로겐화 벤즈아졸 유도체를 합성함
② 합성된 할로겐화 벤즈아졸 유도체와 아릴아민을 커플링 반응시킴

o (paragraph [0167]) [Chemical Formula 16]

② In addition, the prior art describes a specific preparation method for compound (10), one of the desirable compounds represented by General Formula (A-1).

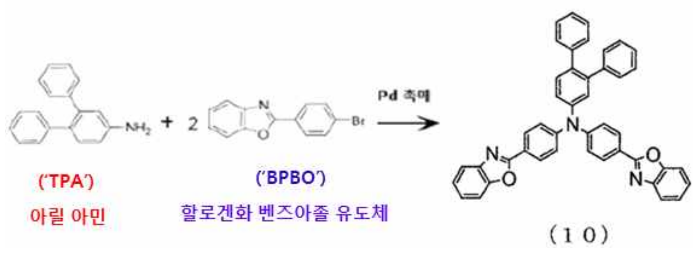
o (paragraph [0224]) 「Embodiment 1」

o (paragraph [0225]) Synthesis of the compound <Bis-{4-(benzoxazol-2-yl)phenyl}-([1,1',2',1'']terphenyl-4'-yl)-amine> represented by Formula (10)

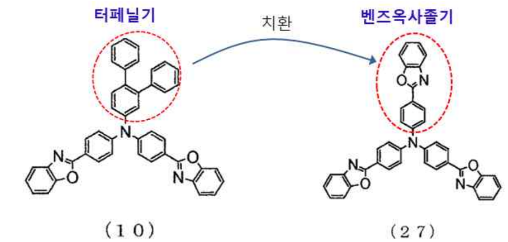
o (paragraph [0226]) A reaction vessel was charged with 5.6 g of ([1,1',2',1'']terphenyl-4'-yl)-amine, 14.4 g of 2-(4-bromophenyl)benzoxazole, 4.4 g of sodium t-butoxide, and 60 mL of toluene, followed by ultrasonic irradiation for 30 minutes under a nitrogen gas purge. Additionally, 0.1 g of palladium acetate and 0.4 mL of a 50% (w/v) toluene solution of tri-(t-butyl)phosphine were added to the vessel, and the mixture was stirred overnight under reflux heating. After allowing the vessel to cool down, methanol was added, and the precipitated solid was collected to obtain a crude product. The crude product was purified through recrystallization using a toluene/acetone mixed solvent, and the precipitated solid was collected to obtain 11.0 g of a yellow powder of bis-{4-(benzoxazol-2-yl)phenyl}-([1,1',2',1'']terphenyl-4'-yl)-amine (the compound represented by formula (10) above) with a yield of 76.4%.

③ The prior art describes the preparation method for halogenated benzoazole derivatives and states that "the synthesized halogenated benzoazole derivative and an arylamine undergo a coupling reaction using a copper catalyst, palladium catalyst, or similar catalysts. Now, the amine compounds with a benzoazole ring structure represented by General Formulas (A-1) or (A-2) of the present invention can be synthesized." Thus, it proposes a coupling reaction between halogenated benzoazole derivatives and arylamines as a process for preparing various amine compounds represented by General Formula (A-1). Furthermore, Embodiment 1 of the prior art demonstrates the synthesis of compound (10) by performing a coupling reaction between a halogenated benzoazole derivative {BPBO (2-(4-bromophenyl)benzoxazole)} and an arylamine {TPA ([1,1’,2’,1“]terphenyl-4’-yl)amine)} under a palladium catalyst (See Chemical Equation 1 below).

Chemical Equation 1

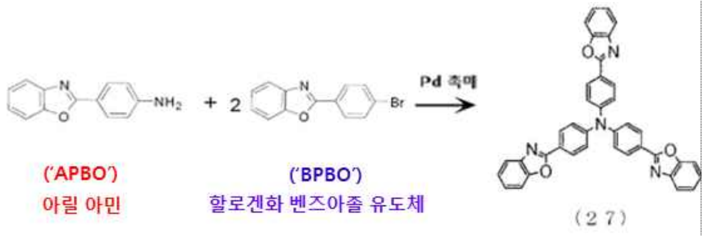


④ However, when comparing compound (27), which is presented as a desirable compound of General Formula (A-1), with compound (10), the only difference lies in the substituent bonded to the central nitrogen atom. In compound (27), the substituent is replaced with a benzoxazole group instead of the terphenyl group in compound (10).



⑤ Therefore, a skilled person would easily understand that, based on the preparation process described in the prior art mentioned above, simply altering the substituents bonded to the central nitrogen atom of the reactant would enable the production of various amine compounds of General Formula (A-1), including compound (27). In other words, it can be easily understood that compound (27) can be prepared through the same reaction process as in Embodiment 1 of the prior art, simply by substituting “TPA,” the reactant in Embodiment 1 of the prior art, with “APBO (2-(4-aminophenyl)benzoxazole),” as shown in Chemical Equation 2 below.

Chemical Equation 2



⑥ Furthermore, the synthesis method for “BPBO” (a halogenated benzoxazole derivative), one of the reactants used in the preparation process of compound (27), is described in the prior art ([Chemical Formula 16] of paragraphs [0164] and [0167]). Also, the structure and various synthesis methods of “APBO,” another reactant, were widely disclosed in multiple references prior to the date of claimed priority of the prior art (December 21, 2017). Therefore, it would not be reasonable to consider that it is difficult to synthesize or acquire “APBO” before the date.

(B) Regarding this, the plaintiff argues that since “APBO,” the starting material in Chemical Equation 2 preparing compound (27), is not commercially common enough to be acquired easily and preparing “APBO” requires finding bibliographic items through meticulous search in paid professional chemical database and finding the source again to check if it is preparable, preparing the material exceeds the level of effort of a skilled person that does not require excessive experiment.

However, considering the level of technological development at the time of the application, that is, the fact that a person can access any time the database about the relevant technical information due to the commercialization of the internet and the development of search engines; can easily look for literature about a specific compound published around the world with the name or the structural formula of the compound without extra time and effort; and can quickly and easily order and receive reagents online from various foreign suppliers, a skilled person does not have to conduct excessive experiment to check if "APBO" can be prepared or acquired based on Defendant's Exhibits 1 to 5 and prepare the material. Therefore, the plaintiff's argument cannot be accepted.

(3) Summary of analysis

In summary, based on the analysis above, compound (27) is deemed to be specifically disclosed as an invention in the prior art.

4) Summary of discussion

Thus, Claim 1 violates the extended first-to-file rule based on the prior art.

B. Whether Claims 2 to 4 Violate the Extended First-to-File Rule

1) Claim 2

Claim 2 is a dependent claim referencing Claim 1, and involves selecting a tribenzazole amine derivative of Chemical Formula 1 from compounds such as compound (2-2), where compound (2-2) is identical to compound (27) disclosed in the prior art above.

2) Claim 3

Claim 3 relates to an organic light-emitting device comprising: a first electrode; an organic layer comprising a plurality of organic material layers disposed on the first electrode; a second electrode disposed on the organic layer; and a capping layer disposed on the second electrode, wherein the capping layer includes a tribenzazole amine derivative according to any one of Claims 1 or 2. However, the prior art discloses "an organic EL device comprising an organic layer including a light-emitting layer between a first and a second electrode, with a capping layer laminated on the surface opposite the organic layer of the first electrode, where the capping layer contains an amine compound with a benzoxazole ring structure represented by General Formula (A-1) or (A-2) [specific example: compound (27)] (paragraphs [0101], [0119], [0122], [0018] through [0093], and [0142]). Accordingly, the structures of the organic electroluminescent devices (organic EL devices) in both inventions are identical.

3) Claim 4

Claim 4 is a dependent claim referencing Claim 3, and it relates to "an organic electroluminescent device wherein the organic layer includes a hole injection layer; a hole transport layer; an emitting layer; an electron transport layer; and an electron injection layer." The prior art also discloses "an organic EL device wherein the organic layer includes a hole injection layer; a hole transport layer; a light-emitting layer; an electron transport layer; and an electron injection layer" (paragraphs [0105] and [0106]). Therefore, the structures of the organic EL devices in both inventions are identical.

4) Summary of discussion

As outlined above, Claims 2 to 4 violate the extended first-to-file rule due to the prior art.

C. Summary of Analysis

In summary, since Claims 1 to 4 violate the extended first-to-file rule due to the prior art, its patent registration should be invalidated. The IPTAB decision consistent with the above shall be upheld, as it is without an error justifying revocation as argued by the plaintiff.

4. Conclusion

Therefore, the plaintiff's petition seeking the revocation of the IPTAB decision is without merit, and accordingly, the decision is rendered as ordered.

Presiding Judge Sungyop WOO
Judge Yongwoo LIM
Judge Kisu KIM

[Appendix 1]

Summary of Invention at Issue

A. Technical Field

[0001] The present invention relates to tribenzazole amine derivatives and organic electroluminescent devices including the same. Specifically, it aims to enable the capping layer of an organic electroluminescent device to simultaneously exhibit high refractive index properties and ultraviolet absorption properties through the use of the tribenzazole amine derivatives above.

B. Background Art

[0010] With the commercialization of organic electroluminescent devices, the demand for properties beyond the devices' intrinsic light-emitting characteristics has emerged. Organic electroluminescent devices are often exposed to external light sources for extended periods, which results in prolonged exposure to ultraviolet light with high energy, having a lasting impact on the organic materials that make up the devices. Applying a capping layer with ultraviolet absorption properties can prevent exposure to such high-energy light sources.

[0011,0012] Additionally, while it is generally known that organic electroluminescent devices have wide viewing angles, the light source spectrum defers significantly depending on the viewing angle. This is due to the discrepancies between the overall refractive index of components such as the glass substrate, organic materials, and electrode materials that constitute the organic electroluminescent device and the appropriate refractive index values corresponding to the emission wavelength of the device. In general, the required refractive index is higher for blue light, and as the wavelength increases, the required refractive index decreases. Accordingly, it is necessary to develop a material for the capping layer that simultaneously has the aforementioned ultraviolet absorption properties and the appropriate refractive index.

C. Problem To Be Solved

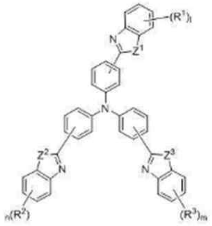
[0016] The present invention aims to provide a capping layer material for organic electroluminescent devices that enhances light-emission efficiency and lifespan while simultaneously contributing to improvements in viewing angle.

[0017] The present invention also aims to provide an organic electroluminescent device with an applied capping layer to improve its characteristics.

D. Means for Solving the Problem

[0018] One embodiment of the present invention comprises: a first electrode; an organic layer disposed on the first electrode; a second electrode disposed on the organic layer; and a capping layer disposed on the second electrode, and the organic layer provides an organic electroluminescent device including a tribenzazole amine derivative represented by Chemical Formula 1.

[0019] [Chemical Formula 1]



[0021 to 0023] In Chemical Formula 1, Z^1, Z^2 and Z^3 are each independently O, S, or NR^4 (where R^4 is phenyl), and R^1, R^2, and R^3 are the same or different and are each independently from hydrogen; deuterium; substituted or unsubstituted alkyl groups; substituted or unsubstituted cycloalkyl groups; substituted or unsubstituted alkoxy groups; substituted or unsubstituted aryloxy groups; substituted or unsubstituted aralkyl groups; substituted or unsubstituted alkylaryl groups; substituted or unsubstituted allyl groups; or substituted or unsubstituted heteroaryl groups. l, n, and m are integers from 0 to 4.

[0024] The organic layer of the organic electroluminescent device above may include a hole transport region, a light-emitting layer disposed on the hole transport region, and an electron transport region disposed on the light-emitting layer. The capping layer may include a tribenzazole amine derivative represented by Chemical Formula 1.

[0025] In addition, the present invention provides tribenzazole amine derivatives represented by Chemical Formula 1 above.

E. Detailed Description for Implementing the Invention

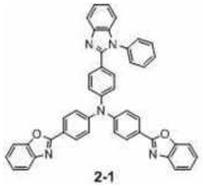
[0041] Referring to Figure 1, the organic electroluminescent device according to one embodiment may include a first electrode (110), a hole injection layer (210), a hole transport layer (215), a light-emitting layer (220), an electron transport layer (230), an electron injection layer (235), a second electrode (120), and a capping layer (300) stacked on a substrate (100) in this order.

[Figure 1] Schematic Diagram of Organic Electroluminescent Device

[0043] Meanwhile, the capping layer (300) presented in the present invention is a functional layer deposited on the second electrode (120) and includes an organic material according to Chemical Formula 1 of the present invention.

[0099] The tribenzazole amine derivative represented by Chemical Formula 1 of the present invention may be any one of the compounds selected from those represented by Chemical Formula 2. However, it is not limited thereto.

[0100] [Chemical Formula 2]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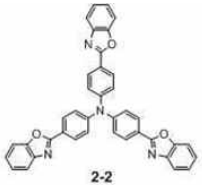
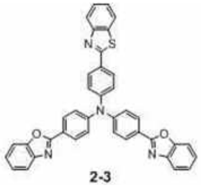
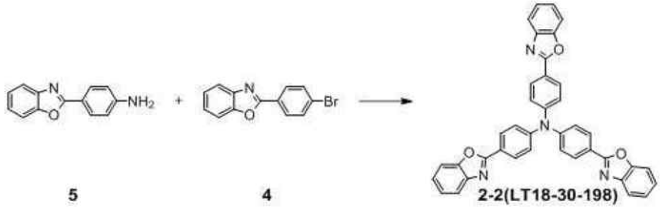

2-2


2-3

(The description of compounds 2-4 to 2-80 is omitted.)

[0244] Embodiment 2: Synthesis of compound 2-2 (LT18-30-198)

[0246] To a 250 mL single-neck flask, 1.8 g (8.56 mmol) of intermediate (5),



5.9 g (21.40 mmol) of intermediate (4), and 85.6 mL of xylene were added and stirred at 50°C. Then, 0.5 g (0.86 mmol) of $Pd(dba)_2$, 4.9 g (51.40 mmol) of sodium tert-butoxide, and 0.69 g (1.71 mmol) of tri-tert-butylphosphine (50 wt% in toluene) were added, and the mixture was stirred for an entire day at 125 to 130°C. After the reaction was complete, the mixture was cooled to room temperature, and the reaction product was passed through a celite pad using $CHCl_3$. Subsequently, the solvent was removed using vacuum distillation. The obtained compound was solidified using hexane to yield a yellow solid, which was dissolved in 800 mL of $CHCl_3$ under heating, and charcoal was added before stirring for 30 minutes. The mixture was passed through a celite and SiO_2 pad using (Hot CHCl3:EA=20:1), and then the solvent was removed by vacuum distillation. The obtained compound was purified using SiO_2 column chromatography (EA:CHCl3:HEX=1:1:5). The compound was slurried with DCM and hexane, yielding 3.0 g of yellow solid compound 2-2 (LT18-30-198) (yield: 59.1%).

[0340,0341] The properties of the optical characteristic samples prepared in the above Comparative Test Example and Test Examples 1 to 23 are shown in Table 1. Table 1 shows the refractive index constants at wavelengths of 420 nm and 620 nm, as well as the absorption coefficient constant at a wavelength of 380 nm.

(The description of Test Examples 4 to 23 is omitted.)

[0356] The electroluminescent properties of the organic electroluminescent devices prepared in the above Comparative Test Example and Test Examples 1 to 23 are shown in Table 2.

Table 1

		(Refractive index constant)	(Absorption coefficient constant)
Classification	Compound	N (450nm, 620nm)	K (380nm)
Comparative example	REF01	2.138, 1.971	0.274
Test Example 1	2-1 (LT18-30-267)	2.227, 1.980	0.634
Test Example 2	2-2 (LT18-30-198)	2.326, 2.081	0.679
Test Example 3	2-3 (LT18-30-238)	2.263, 1.997	0.598

Table 2

Classification	Compound	Driving voltage [V]	Efficiency [cd/A]	Lifespan (%)
Comparative Embodiment 01	REF01	6.60	5.10	88.92
Embodiment 1	2-1 (LT18-30-267)	4.62	6.70	98.87
Embodiment 2	2-2 (LT18-30-198)	3.93	6.07	105.43
Embodiment 3	2-3 (LT18-30-238)	3.81	5.65	98.79

(The description of Embodiments 4 to 23 is omitted.)

F. Effect

[0026] The organic electroluminescent device including the capping layer according to the present invention exhibits ultraviolet absorption properties, minimizing damage to the organic materials within the device caused by external light sources, which helps maintain the inherent efficiency and lifespan of the device as much as possible. Furthermore, the organic electroluminescent device of the present invention, through the use of a capping layer, improves light efficiency, reduces the full width at half maximum of the emission spectrum, and enhances viewing angle. As a result, it meets the diverse performance requirements of currently commercialized organic electroluminescent devices.

[Appendix 2]

Main Content of Prior Art

A. Technical Field

[0001] The present invention relates to an organic electroluminescent device (hereinafter, "organic EL device") and a method for preparing the same.

B. Background Art

[0012] Improving the device characteristics of organic EL devices particularly requires absorbing light with a wavelength of 400 to 410 nm from sunlight while preventing any impact on the materials inside the device. Additionally, to significantly improve light extraction efficiency, materials for the capping layer are required to have a high absorption coefficient, high refractive index, and excellent thin-film stability and durability.

C. Problem To Be Solved

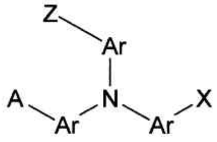
[0015] The present invention has been devised in consideration of the above circumstances, and aims to provide an organic EL device with high brightness, high luminous efficiency, high power efficiency, and long lifespan, along with a method for preparing the same.

D. Means for Solving the Problem

[0018] In other words, the following organic EL device is provided according to the present invention.

[0019] 1) The organic EL device comprising at least an anode electrode, a hole transport layer, a light-emitting layer, an electron transport layer, a cathode electrode, and a capping layer in this order, wherein the capping layer contains an amine compound having a benzoxazole ring structure represented by General Formula (A-1) below.

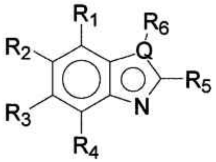
[0020] [Chemical Formula 1]



(A−1)

[0022] (In Formula (A-1), A and X may be the same or different, and each represents a monovalent group with one bonding site selected from R_1 to R_6 in General Formula (B-1) below. Z represents a monovalent group with one bonding site selected from R_1 to R_6 in General Formula (B-1), a substituted or unsubstituted aromatic hydrocarbon group, a substituted or unsubstituted aromatic heterocyclic group, or a substituted or unsubstituted fused polycyclic aromatic group. Ar represents a bivalent group or a single bond, which may be the same or different from one another, selected from substituted or unsubstituted aromatic hydrocarbon groups, substituted or unsubstituted aromatic heterocyclic groups, or substituted or unsubstituted fused polycyclic aromatic groups.)

[0023] [Chemical Formula 2]



(B−1)

[0025] (In Formula (B-1), R_1 to R_6 may be the same or different and represent a linking group as a bonding site, a hydrogen atom, a deuterium atom, a fluorine atom, a chlorine atom, a cyano group, a nitro group, or a straight-chain or branched alkyl group with 1 to 6 carbon atoms that may have substituents, [omitted] and Q represents a nitrogen atom, an oxygen atom, or a sulfur atom. However, when Q is an oxygen atom or a sulfur atom, Q does not have R_6.)

[0049] 19) The organic EL device according to the above 5), wherein A, X, and Z in General Formula (A-1) are the same and the device contains an amine compound having a benzoxazole ring structure.

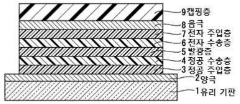
E. Detailed Description for Implementing the Invention

[0097] The present invention is to improve the device characteristics of organic EL devices. It particularly aims to absorb sunlight with wavelengths of 400 nm to 410 nm while preventing any impact on the materials inside the device, and to significantly improves light extraction efficiency. Furthermore, its objective is to provide an organic EL device equipped with a capping layer which is composed of a material that (1) has a high absorption coefficient, (2) has a high refractive index, (3) exhibits good thin-film stability, (4) possesses excellent durability, (5) has superior light resistance, and (6) does not exhibit absorption in the wavelength regions of blue, green, and red light.

[0100] 「Organic EL Device」

[0106] Figure 1 is a schematic cross-sectional view illustrating an example of the organic EL device according to the present embodiment. The organic EL device shown in Figure 1 has a top-emission structure, with an anode (2), a hole injection layer (3), a hole transport layer (4), a light-emitting layer (5), an electron transport layer (6), an electron injection layer (7), a cathode (8), and a capping layer (9) stacked on a glass substrate (1) in this order.

[Figure 1] Example of Organic EL Device

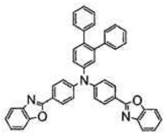


[0109] 「Capping Layer」

[0119] The capping layer of the organic EL device according to the present invention contains an amine compound having a benzoxazole ring structure represented by General Formula (A-1) or (A-2).

[0127] In the organic EL device of the present invention, Ar in General Formulas (A-1) and (A-2) is desirably a divalent or single bond of a substituted or unsubstituted aromatic hydrocarbon group, and more desirably, a divalent group of a substituted or unsubstituted phenyl group.

[0137] Among the compounds represented by General Formula (A-1) or (A-2), which are suitably used in the organic EL device of the present invention, compounds represented by formulas (1) to (85) are presented below as specific examples of desirable compounds.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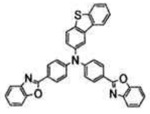
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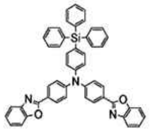
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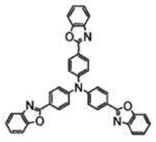

(27)

(The descriptions of the compounds in formulas (1) through (9), (11) through (24), and (28) through (85) are omitted.)

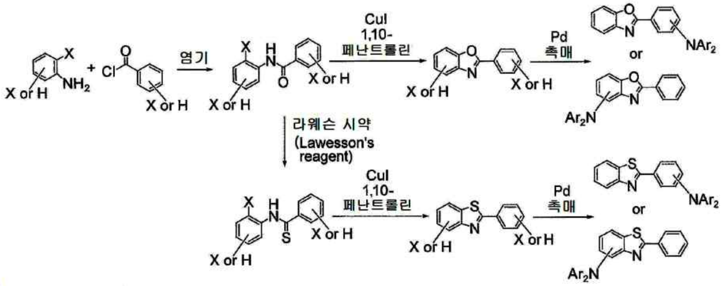
[0162] 「Method for Preparing Compounds Represented by General Formulas (A-1) and (A-2)」

[0163] Amine compounds with a benzoazole ring structure, represented by General Formulas (A-1) and (A-2), can be prepared using the methods described below.

[0164] First, to form the benzoazole ring structure represented by Formula (B-1) or (B-8), which serve as the main framework of the compounds represented by General Formulas (A-1) and (A-2), a halogenated benzoazole derivative is prepared as the starting material. Halogenated benzoazole derivatives with structures corresponding to the benzoazole ring structure represented by Formula (B-1) or (B-8) can be prepared, for example, using publicly-known methods as described below (See, for instance, non-patent literature 4).

[0165] Additionally, the synthesized halogenated benzoazole derivative and an arylamine undergo a coupling reaction using a copper catalyst, palladium catalyst, or similar catalysts. Now, the amine compounds with a benzoazole ring structure represented by General Formula (A-1) or (A-2) of the present invention can be synthesized.

[0167] [Chemical Formula 16]



[0224] 「Embodiment 1」

[0225] Synthesis of the compound <Bis-{4-(benzoxazol-2-yl)phenyl}-([1,1',2',1'']terphenyl-4'-yl)-amine> represented by Formula (10)

[0226] A reaction vessel was charged with 5.6 g of ([1,1',2',1'']terphenyl-4'-yl)-amine, 14.4 g of 2-(4-bromophenyl)benzoxazole, 4.4 g of sodium t-butoxide, and 60 mL of toluene, followed by ultrasonic irradiation for 30 minutes under a nitrogen gas purge. Additionally, 0.1 g of palladium acetate and 0.4 mL of a 50% (w/v) toluene solution of tri-(t-butyl)phosphine were added to the vessel, and the mixture was stirred overnight under reflux heating. After allowing the vessel to cool down, methanol was added, and the precipitated solid was collected to obtain a crude product. The crude product was purified through recrystallization using a toluene/acetone mixed solvent, and the precipitated solid was collected to obtain 11.0 g of a yellow powder of bis-{4-(benzoxazol-2-yl)phenyl}-([1,1',2',1'']terphenyl-4'-yl)-amine (the compound represented by formula (10) above) with a yield of 76.4%.

[0304] (Measurement of peak wavelength, absorbance, and absorption coefficient of the compound)

[Table 1]

	Peak wavelength (λmax)	Absorbance (λ:400nm)	Absorbance (λ:410nm)	Absorption coefficient
Compound 10	387 nm	0.94	0.29	130367
Comparative compound (2-1)	358 nm	0.07	0.02	48856
Alq3	394 nm	0.07	0.06	7518

[0316] (Measurement of refractive index and extinction coefficient of thin films)

[Table 2]

	Refractive Index n (λ:400nm)	Refractive Index n (λ:410nm)	Extinction Coefficient k (λ:400nm)	Extinction Coefficient k (λ:410nm)
Compound 10	2.37	2.47	0.66	0.44
Comparative compound (2-1)	2.13	2.10	0.15	0.06
Alq3	1.86	1.89	0.16	0.14

[0364] (Measurement of device lifespan)

[Table 3]

Capping layer	Voltage [V] (10mA/cm^2)	Luminance [cd/m^2] (10mA/cm^2)	Luminous efficiency [cd/A] (10mA/cm^2)	Power efficiency [lm/W] (10mA/cm^2)	Lifespan of device 95 % degradation
Compound 10	3.65	702	7.02	6.04	157 hours
Comparative compound (2-1)	3.69	668	6.68	5.68	121 hours
Alq3	3.67	647	6.47	5.54	106 hours

E. Effect

[0094] The organic EL device of the present invention exhibits high brightness, high luminous efficiency, high power efficiency, and a long lifespan.