

**UNITED STATES PATENT AND TRADEMARK OFFICE
BEFORE THE PATENT TRIAL AND APPEAL BOARD**

SAMSUNG DISPLAY CO., LTD.
Petitioner,
v.
PICTIVA DISPLAYS INTERNATIONAL LTD.
Patent Owner.

IPR2024-01095

October 24, 2025



pictivadisplays

The Challenges Fail for Multiple Reasons

Claim 1

wherein the organic layer sequence comprises an electron transport layer that is n-doped; and
wherein the organic layer sequence comprises an emission layer comprising an iridium-containing compound.

Claim 2

2. The device of claim 1,
wherein the electron transport layer comprises 8-hydroxy-quinolinolato-lithium.

- Ma not shown as prior art
- Objective evidence shows non-obviousness
 - Only two identified pre-priority references on Liq and only as n-dopant for Alq₃-based fluorescent OLEDs
 - First reference on using Liq-doped ETL in PHOLEDs appeared 3 years after priority, despite “fast moving” OLED community
- Ma’s Liq used a different operating mechanism than Suzuki and Diekmann
- Insufficient evidence of alleged technical benefits

Ma Is Not Shown As Prior Art

ARTICLE INFO

Article history:

Received 11 March 2008

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Keywords:

Organic light-emitting diodes

Doping concentration

Carrier transport

Conductivity

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Synthetic Metals

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Organic light-emitting diodes based on new n-doped electron transport layer

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ARTICLE INFO

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ABSTRACT

Organic light-emitting diodes with 8-hydroxy-quinolino lithium doped 4,7-diphenyl-1, 10-phenanthroline as electron transport layer (ETL), and tetrafluoro-tetracyano-quinodimethane doped 4,4',4''-tris(3-methylphenylphenylamino)triphenylamine as hole transport layer (HTL) are demonstrated. The conductivity of carrier transport layers with different doping concentration is examined by hole-only and electron-only devices. Compared with the referenced device (without doping), the current efficiency and power efficiency of the p-i-n device are enhanced by approximately 51% and 89%, respectively. This improvement is attributed to the improved conductivity of the transport layers and the efficient charge balance in the emission zone.

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1. Introduction

Since the discovery of organic light-emitting devices (OLEDs), there has been a considerable interest in developing OLEDs with high efficiency for display applications [1]. In order to lower driving voltage and enhance power efficiency, it is critical to enhance the carrier injection from the electrode to the transporting layer and to increase the transport conductivity [2,3]. The hole injection can be improved by increasing the work function of ITO with different surface treatments such as O₂ plasma or UV-ozone treatment of ITO surface [4]. It can also be enhanced by introducing hole injection layer (HIL), such as copper phthalocyanine (CuPc) [5], starburst polyamines [6], polymeric PEDT:PSS [7] and 4,4',4''-tris(N-(2-naphthyl)-N-phenylamino)triphenylamine (2-TNATA) between the ITO/hole transport layer (HTL) interface.

Recently, the p-doping of HTL has drawn a lot of attentions because of its ability to enhance hole injection and lower driving voltages in OLEDs. The p-doping HTL is typically made by co-evaporating the hole transporting materials with a strong electron acceptor like tetrafluoro-tetracyano-quinodimethane (F₄-TCNQ) [8], or oxidants like SbCl₅ [9], FeCl₃ [10] and iodine [11]. p-Doping can

also achieve ohmic conductivity to reduce driving voltage of device and the proper control of doping levels can also lead to efficient carrier injection by tunnelling [12].

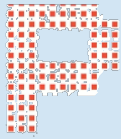
However, organic molecules are known to be poor in electron injection and transport compared with that of hole. Many attempts have been made to improve the electron injection and transport, including using low work function metals [1,13–15] as cathode, or inserting a thin interlayer between cathode and electron transporting layer [16–20] to enhance electron injection. Great efforts have been made to enhance the electron conductivity [21–24]. Kido and Matsumoto [22] reported the improved electron transport by doping Li into tris(8-hydroxyquinoline) aluminum (Alq₃). Fong et al. [21] also demonstrated that the device performance was enhanced significantly by doping 4,7-diphenyl-1, 10-phenanthroline (Bphen) into Alq₃ as a cohost electron transport layer (c-ETL). However, few studies have been reported about using lithium quinolinato complex such as 8-hydroxy-quinolinato lithium (LiQ) as an electron injection layer [25], and no work has yet been reported about using LiQ-doped Bphen as electron transport layer (ETL).

In this paper, we have demonstrated a device with a new n-doping (33 wt.% LiQ:Bphen) layer as ETL and p-type (m-MTDATA:x wt.% F₄-TCNQ) layer as HTL. Here, 33 wt.% LiQ is chosen due to the fact that the device with the doping ratio of 33 wt.% LiQ demonstrates the best current density vs voltage (J–V) curve among all the electron only devices. Using this method, we obtained a sig-

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Unrebutted: Elsevier Had Limited Search Capabilities for Online Only Articles

Dr. Schanze
Expert Witness



pictivadisplays

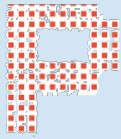
149. Additionally, as I recall, in 2008-2009, Elsevier journals often did not promptly index new articles, especially online-only versions. Thus, it was my recollection that new articles often could not be located unless one knew the authors' names or the title of the article. It is also my recollection that in 2008-2009, Elsevier's search tool⁸ had limited keyword search capability as it often failed to identify articles of interest. Additionally, during the relevant time period (2008-2009), indexing services, such as Clarivate Web of Science did not index online only articles, making them difficult or impossible to locate.

150. For example, as shown by the screen shot below, the Ma article was not indexed by Web of Science until December 2008. I personally captured the screen shot from the Web of Science web site.

EX1046, para. 149-150; Sur-Reply 23-24

Unrebutted: No Indexing of Online Only Articles

Dr. Schanze
Expert Witness



pictivadisplays

149. Additionally, as I recall, in 2008-2009, Elsevier journals often did not promptly index new articles, especially online-only versions. Thus, it was my recollection that new articles often could not be located unless one knew the authors' names or the title of the article. It is also my recollection that in 2008-2009, Elsevier's search tool⁸ had limited keyword search capability as it often failed to identify articles of interest. Additionally, during the relevant time period (2008-2009), indexing services, such as Clarivate Web of Science did not index online only articles, making them difficult or impossible to locate.

150. For example, as shown by the screen shot below, the Ma article was not indexed by Web of Science until December 2008. I personally captured the screen shot from the Web of Science web site.

EX1046, para. 149-150; Sur-Reply 23-24

Munford Declaration Shows Ma as “Article in Press” As of December 2, 2008

Dr. Munford
Expert Witness

SAMSUNG

18. The Articles in Press Listing contains an Elsevier ScienceDirect webpage describing the contents of the Synthetic Metals journal as of December 2, 2008. Appendix 2 at 48/178. “Articles in Press” are described as “peer reviewed accepted articles to be published in this journal.” *Id.* at 53/178. Further, “[t]he ‘Article in Press’ version . . . will appear in the associated published journal issue.” *Id.* When published, “[t]he date [the Article in Press version] was first made available online will be carried over,” and despite not yet being published in paper form, the Article in Press version “can already be cited using the year of online availability . . .” *Id.*

19. An entry for “Organic light-emitting diodes based on new n-doped electron transport layer” (which is the same title as Exhibit 1007) can be found on page 5 of the Articles in Press Listing. Appendix 2 at 51/178. This entry indicates

IPR2024-01094, EX1037, para. 18-19; Sur-Reply 1

12/1/2008 Reflects Month, Not Necessarily Date, Of Indexing

Web of Science

Organic light-emitting diodes based on new n-doped electron transport layer

By [Ma, JW \(Ma, J. W.\) \[1\]](#); [Xu, W \(Xu, Wei\) \[1\]](#); [Jiang, XY \(Jiang, X. Y.\) \[1\]](#); [Zhang, ZL \(Zhang, Z. L.\) \[1\], \[2\]](#)

Are you this author?

Source [SYNTHETIC METALS](#) ▾
Volume: 158 Issue: 21-24 Page: 810-814
DOI: [10.1016/j.synthmet.2008.05.009](https://doi.org/10.1016/j.synthmet.2008.05.009)

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Abstract
Organic light-emitting diodes with 8-hydroxy-quinolinato lithium doped 4'-7-diphenyl-1, 10-phenanthroline as electron transport layer (ETL), and etrafluro-tetracyano-quinodimethane doped 4,4',4''-tris(3-methylphenylphenylamono) triphenylamine as hole transport layer (HTL) are demonstrated. The conductivity of carrier transport layers with different doping concentration is examined by hole-only and electron-only devices. Compared with the referenced device (without doping), the current efficiency and power efficiency of the p-i-n device are enhanced by approximately 51% and 89%, respectively. This improvement is attributed to the improved conductivity of the transport layers and the efficient charge balance in the emission zone. (C) 2008 Elsevier B.V. All rights reserved.

Keywords
Author Keywords: [Organic light-emitting diodes](#); [Doping concentration](#); [Carrier transport](#); [Conductivity](#)
Keywords Plus: [ELECTROLUMINESCENT DEVICES](#); [HOLE-INJECTION](#); [EFFICIENCY](#); [VOLTAGE](#); [PHTHALOCYANINE](#)

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**Indexing Date Predated Publication in Paper Form→
“Indexed” Reflects Month of Indexing**

EX1047

Web-of-Science Indexing “Date” May Reflect Month, and Not Date, Of Indexing

Available online at www.sciencedirect.com

 **ScienceDirect**

Synthetic Metals 157 (2007) 190–197

www.elsevier.com/locate/synmet

Solution-processed polycrystalline copper tetrabenzoporphyrin thin-film transistors

Patrick B. Shea^a, Lisa R. Pattison^b, Manami Kawano^c, Charlene Chen^a, Jihua Chen^d, Pierre Petroff^b, David C. Martin^d, Hiroko Yamada^c, Noboru Ono^c, Jerzy Kanicki^{a,*}

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Solution-processed polycrystalline copper tetrabenzoporphyrin thin-film transistors

[Associated Data](#)

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Indexed
2007-03-01

Published	MAR 2007
Indexed	2007-03-01

EX1042

**Indexing Date Predated Online Posting Date→
“Indexed” Reflects Month of Indexing**

Undisputed: Priority Date for 425 is 12/11/2008

Dr. Holmes
Expert Witness

SAMSUNG

51. I have been asked to assume that the earliest possible priority date for the '425 patent is December 11, 2008, the earliest Foreign Application Priority Date listed on the face of the '425 patent. I have therefore analyzed the prior art as of that date or somewhat before that date.

EX1003, para. 51; Sur-Reply 3; see also EX2032, 220:20-23

Indexing/Publishing Date Not Shown As Before the Priority Date

Web of Science

Organic light-emitting diodes based on new n-doped electron transport layer

By Ma, JW (Ma, J. W.) ^[1]; Xu, W (Xu, Wei) ^[1]; Jiang, XY (Jiang, X. Y.) ^[1]; Zhang, ZL (Zhang, Z. L.) ^[1], [2]

Source SYNTHETIC METALS
Volume: 158 Issue: 21-24 Page: 810-814
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Abstract Organic light-emitting diodes with 8-hydroxy-quinolinato lithium doped 4'7-diphenyl-1, 10-phenanthroline as electron transport layer (ETL), and tetrafluoro-tetracyano-quinodimethane doped 4,4'-tris(3-methylphenylphenylamino) triphenylamine as hole transport layer (HTL) are demonstrated. The conductivity of carrier transport layers with different doping concentration is examined by hole-only and electron-only devices. Compared with the referenced device (without doping), the current efficiency and power efficiency of the p-i-n device are enhanced by approximately 51% and 89%, respectively. This improvement is attributed to the improved conductivity of the transport layers and the efficient charge balance in the emission zone. (C) 2008 Elsevier B.V. All rights reserved.

Keywords Author Keywords: Organic light-emitting diodes; Doping concentration; Carrier transport; Conductivity
Keywords Plus: ELECTROLUMINESCENT DEVICES; HOLE-INJECTION; EFFICIENCY; VOLTAGE; PHTHALOCYANINE

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Categories/ Classification Research Areas: Materials Science; Physics; Polymer Science
Citation Topics: 2 Chemistry > 2.114 Organic Semiconductors > 2.114.321 OLEDs
Sustainable Development Goals: 07 Affordable and Clean Energy

“DEC 2008”: any time between 12/1/2008 and 12/31/2008

12/1/2008

12/11/2008

12/31/2008

EX1047

Availability of Abstract at Unspecified Time Points Does Not Evidence Public Accessibility at the Time of the Invention

Petitioner's Argument

“Ma’s abstract would have been available online ... permitting an interested entity to find the abstract and then, if necessary, gain access to Ma’s full text”

Facts

- Circular argument
- No evidence that a POSITA could locate the abstract of an online only article without first knowing about the article already
- No evidence when the link to abstract was added to the article

No Presumption of Public Accessibility under *VidStream*



- Concerned “a book” authored by Braford et al.
- Book obtained from Library of Congress bearing copyright date
- Says nothing about online only articles
 - Also no evidence that the pre-publication version had any copyright sign or date

VidStream LLC v. Twitter, Inc., 981 F.3d 1060, 1061, 1065 (Fed. Cir. 2020)

Dr. Holmes Had No Information on Public Availability

Dr. Holmes
Expert Witness

SAMSUNG

Q. *And do you know, back in August 29, 2008 when it says it's made available online, was it available only to subscribers or it's available to any general public --*

Q. -- members?

A. Yeah. I don't have knowledge of how the agreements work in terms of how these journals are available. All I could say --

Q. Go ahead.

A. I was just going to add, all I can say is that in August of 2008 and earlier than that, I was pulling journal articles online routinely. And Synthetic Metals is certainly a journal that I read at the time, but *I don't have any broader knowledge of whether it was publicly available, subscription based. I don't know.*

EX2019, 234:14-235:9; Reply 3; Sur-Reply 1

**Testimony Also Does Not Say Whether He Was Able
to Search for and Locate Online Only Articles**

Dr. Holmes Had No Information on Public Availability

Dr. Holmes

Expert Witness

SAMSUNG

- Q. And **were you pulling the articles that has been published for a while or are you pulling the articles that was posted online but before they become published on paper?** Do you know what I mean?
- A. **So I don't know if I can answer that** because I don't know the last time I looked at an article on paper. Certainly I haven't looked at anything on paper since 2008 and I think it probably would have been maybe in very early graduate school would have been the last time I looked up a paper in the library. For my graduate training, everything was done online. And so I don't know the specifics of how a paper publication date would have related to an online date. **When things were available online, that was when we as researchers were reading them.**

EX2019, 235:10-236:4 (objections omitted); Reply 3; Sur-Reply 1

**Testimony Not Specific to Ma;
Testimony Does Not Say When He Considered an Article Became Available Online**

Dr. Holmes Had No Information on Public Availability

Dr. Holmes
Expert Witness
SAMSUNG

- Q. You don't know how long after the journal; published or makes the article available on its website the article actually gets indexed, right?
- A. No. As I've said already, **I don't have any knowledge whatsoever of the indexing process.** I just know that **journals send out table of contents alerts when issues are available** and there's ways to search things online, but I don't know how indexing works and what the timing is of all that.

EX2019, 237:13-22 (objections omitted); Reply 3-4; Sur-Reply 1

**“Table of Contents” and “Issues” Are Terms Used
to Describe Paper Publication, Not Online Articles**

“Issue” Relates to Paper Form, Not Online Posting

Dr. Munford
Expert Witness

SAMSUNG

18. The Articles in Press Listing contains an Elsevier ScienceDirect webpage describing the contents of the Synthetic Metals journal as of December 2, 2008. Appendix 2 at 48/178. “Articles in Press” are described as “peer reviewed accepted articles to be published in this journal.” *Id.* at 53/178. Further, “[t]he ‘Article in Press’ version . . . will appear in the associated published journal issue.” *Id.* When published, “[t]he date [the Article in Press version] was first made available online will be carried over,” and despite not yet being published in paper form, the Article in Press version “can already be cited using the year of online availability . . .” *Id.*

IPR2024-01094, EX1037, para. 18; Sur-Reply 1

Ma Does Not Suggest or Motivate the Use of Liq in Diekmann or Suzuki

- Objective evidence of non-obviousness in using Liq in PHOLEDs
- Different operating principles
- No evidence of non-speculative benefits over Diekmann's and Suzuki's disclosed embodiments

It Took Nine Years to Arrive at the Invention in an Allegedly “Fast Moving” Community

Holmes Reply Declaration

14. Dr. Pattison’s assertions about the motivations and expectations of a POSITA in swapping materials and dopants into different OLED stacks are untethered from the reality of how work was conducted in the OLED community at the time of the ’425 patent. When an interesting new material or device architecture emerges, there is interest from POSITAs to integrate it into their own work. At the time of the ’425 patent, the OLED research community was best described as very high impact and fast moving. Dr. Pattison’s proposed approach of sitting on the sidelines until somebody else figured out the exact physical mechanism by which a new material or architecture functioned was not how POSITAs operated at the time. To the contrary, POSITAs would routinely apply and/or modify materials and architectures whose mechanisms were not fully understood but demonstrated promising device performance. It was by first building OLEDs and running experiments on those devices that POSITAs would discover the underlying mechanisms governing the behavior of OLED materials and architectures. This approach is reflected in much of the literature cited in this proceeding, which are discussed later in this declaration. To illustrate with some examples:

Facts

- **2002:** Kido-Endo paper on Liq-doping with Alq₃ as EML
 - Kido, “an expert in the area of OLEDs” whose work “the community would follow”
- 2008: Ma paper on Liq-doping with Alq₃ as EML
- Dec. 11, 2008: priority date
- **2011:** Kim paper on Liq-doping in PHOLED

Sur-Reply 6-7; EX2032, 28:17-30:5, 217:25-218:25; EX1049; EX1010; EX2003; EX1044, para. 38 (acknowledging Kim as the earliest evidence in the record on Liq in PHOLEDs)

The 9-year Timeline in a “Fast-Moving” Field:

- **No interest/motivation; and/or**
- **No reasonable expectation of success**
- **Not well known**

Dr. Holmes Considers Himself as a Good Literature Researcher

Dr. Holmes
Expert Witness

SAMSUNG

Q. **Do you consider yourself to be a pretty good researcher as far as locating the relevant literature?**

A. Yes, I do.

Q. Okay. So if you want to find a particular paper on a particular topic, you usually find them, right?

A. Well, again, that -- there's some truth to that but you have to remember as well that p- and n-doping was something that was still emerging in the early 2000s, and so, you know, you might have to sometimes look harder than other times to find the paper. Sometimes the terminology is different, sometimes you have to search for particular molecules, so -- but, **yes, I think I'm effective in finding literature.**

EX2032, 220:3-19; Sur-Reply 10

Dr. Holmes Managed to Locate Only Two Liq-n-Dopant Article Before Priority

Dr. Holmes
Expert Witness

SAMSUNG

Q. Do you think that it's relevant to find literature where lithium quinolate is used as a n-dopant in the electron-transfer layer?

A. Yes.

Q. Okay. And the more you can find, **the more it will support your theory, correct?**

A. Yes.

Q. But the fact is, you searched and besides **Ma**, you only located the **Endo** paper, correct? Or at least the -- only cited to the Endo paper?

A. **Those are the two references I'm citing to support the doping of Liq into an ETL.**

Q. **Did you locate other papers?**

A. **None that I'm aware of at the moment.**

EX2032, 222:1-20; Sur-Reply 10

Earliest Work (EX1049) Done by an Expert (Kido)

Dr. Holmes
Expert Witness

SAMSUNG

Q. Let's take a look at Exhibit 1049, which is a Kido paper or Endo paper. Do you [have] it open?

A. I have it open now.

Q. Okay. Are you familiar with Professor Kido's work?

A. **I'm familiar with Professor Kido's work.**

...

Q. He is a person with above-average skill in the art, correct?

A. Again, I don't know if there's, like, a particular -- I'm not familiar with the legal weight of the term "above-average skill in the art." Clearly, he's a professor running a group in this area. So **he would be an expert in the area of OLEDs, I can certainly attest to that.**

Q. Okay. **He's an expert in the field of OLED at the time of the invention in 2008, 2009?**

A. I think **that's a fair comment.**

EX2032, 28:17-30:5 Sur-Reply 6

Others Allegedly Follow Prof. Kido's Work

Dr. Holmes
Expert Witness

SAMSUNG

Q. **Kido is a well-known group at the time of the invention**, right?

A. **Yes.**

Q. People working in the, in that field, generally knows about that group, correct?

A. Yes, that's correct.

Q. And do they follow their work generally?

A. Again, **the community reads Kido's work, certainly.**

Q. So there [sic, they] should be well-aware of Exhibit 1049, for example?

A. Again, I think that **the community would follow Kido's work, certainly.**

Q. They would be aware of Exhibit 1049, for example, of Kido work?

A. Again, it would be -- if it's in the literature if they are following the work, **they would be aware of all of his work.** And so I'm hesitant to single out 1049 versus the dozens of other papers. But I think, **in general, the community is following Kido's work including [EX]1049.**

EX2032, 217:25-218:25 (objections omitted) Sur-Reply 6

6 Years Before Another Group Used Liq-Doped ETL



6-Year Gap in a “Fast-Moving” Field:

- No interest; and/or
- No reasonable expectation of success
- Not well known

Dr. Holmes Conceded the First Reported Use of Liq as n-Dopant in PHOLED Occurred in 2011 (3 Years after Priority)

Dr. Holmes
Expert Witness
SAMSUNG

38. For example, the study from 2011, which is closest to the priority date and thus most likely to be reflective of a POSITA's understanding at the time, shows that the use of an Liq-doped ETL *improved* efficiency and operating voltage, consistent with a POSITA's expectations at the time of the '425 patent based on Ma's teachings:

EX1044, para. 38; Sur-Reply 6-7

- This Timing in a “Fast-Moving” Field:**
- **No interest; and/or**
 - **No reasonable expectation of success**
 - **Not well known**

Ma Does Not Suggest or Motivate the Use of Liq in Diekmann or Suzuki

- Objective evidence of non-obviousness in using Liq in PHOLEDs
- Different operating principles
- No evidence of non-speculative benefits over Diekmann's and Suzuki's disclosed embodiments

Liq in Ma Had a Different Mechanism of Operation Than n-Dopants in Diekmann

Petition (Diekmann)

- h. **Element 1[g]: wherein the organic layer sequence comprises an electron transport layer that is n-doped; and**

Diekmann discloses or at a minimum suggests this element. Ex.1003, ¶¶106-110. Diekmann discloses that its organic layer sequence (organic layer stack 4) “has a doped layer 402 with a dopant 410, which acts as an electron donor...The dopant 410 can be for example cesium, barium or lithium fluoride.” Diekmann, 91.

Pet. 24

Ma

These *J-V* characteristics of electron-only devices suggest that a certain doping ratio of Liq to Bphen can improve the electron transport ability of the co-deposited layer. The advantage of using Bphen:Liq as ETL can be explained by electron hopping exchange alongwith their LUMO. In a single host device, electrons hop along the LUMO in Bphen. Since the LUMO-LUMO difference between Bphen (3.0 eV) [29] and Liq (3.1 eV) [28] is neglectable, subject to their similar LUMO. Transport manifolds alongwith their LUMO are expected to exhibit a certain extent of overlap after a mixing ratio goes beyond 17 wt.%. Therefore, it's likely that a large energetic disorder between Bphen and Liq contributes to electron hopping, implying that electron hopping among Bphen and Liq sites is favorable [29]. Therefore, the high electron conductivity of Bphen:Liq may be originated from short hopping length for electron transport, compared to the pure Bphen HTL. However, the current conduction is reduced slightly as the doping ratio further increases to 50 wt.%. This result may be attributed to the effect of carrier quench and generation of defects. The detailed reasons are possibly the same as that of the heavy doping concentration of F₄-TCNQ into *m*-MTDATA in the hole-only devices.

Ma, 813

Suzuki Also Disclosed Electron-Donor Type n-Dopants

Petition (Suzuki)

h. Element 1[g]: wherein the organic layer sequence comprises an electron transport layer that is n-doped; and

Suzuki discloses or at a minimum suggests this element. Ex.1003, ¶¶194-195. Suzuki's organic layer sequence (organic layer 7) comprises "an electron transport layer," Suzuki, [0274], Suzuki discloses that the "electron transport layer comprises a material having the function of transporting electrons." Suzuki, [0329]. Suzuki specifies that "an electron transport layer with a high n nature doped with impurities can also be used" and lists applications and publications that include examples of this doped electron transport layer. Suzuki, [0333]. A POSITA would understand that this refers to an electron transport layer which has been n-doped, particularly as the publications cited by Suzuki as examples explicitly disclose n-doped electron transport layers. Ex.1003, ¶194; Ex.1019.

EX1019

Al
Bphen:Cs
Bphen
TCTA:Ir(ppy) ₃
Spiro-TAD
MeO-TPD:F4-TCNQ
ITO substrate

p-i-n

EX1019, Fig. 1; EX2101, 1245 ("cesium (Cs) acts as an efficient electron donor in organic films"); EX2018, ¶18-19; POR 2

Ma Had Very Different Configuration than Diekmann/Suzuki

Diekmann/Suzuki

cathode
Doped ETL
Blocking layer
3 emitter-doped emission layers
Hole transport layer
hole injection layer
anode

EX1004 (Diekmann), Fig. 2A, 91; EX1019, Fig. 1; Sur-Reply 12

Ma

cathode
EIL
Doped ETL
Alq3 (neat)
Hole transport layer
P-doped hole injection layer
anode

EX1010 (Ma), 811; Sur-Reply 12; POR 11-12, 29

Dr. Holmes Concedes that Ma Does Not Disclose Increased Electron Density with Liq

Dr. Holmes
Expert Witness

SAMSUNG

Discussing Ma's disclosure on p. 813:

“So again, I'm relying on Ma's interpretation of the results and *Ma does not appear to consider an increase in electron density* due to a charge transfer interaction, but Ma doesn't also entirely exclude it as well”

EX2019:60:13-61:12; POR 1

Dr. Holmes Concedes that Ma Does Not Disclose Increased Electron Density with Liq

Dr. Holmes
Expert Witness
SAMSUNG

- Q. Ma does not say there is an increase in the number of electrons, does it?
- A. **Ma specifically focuses on the change in mobility** and specifically via the improvement in hopping transport in the electron transport material.

EX2019, 47:12-18 (objections omitted); POR 1

Dr. Holmes Concedes that Ma Does Not Disclose Increased Electron Density with Liq

Dr. Holmes
Expert Witness

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“I'll also note that the doping being alluded to here in this patent is a doping by charge transfer. And as we've already discussed, the **conductivity can be improved via charge carrier number or mobility**. And *in Ma*, they're arguing based on their data that *what's happening is an increase in the mobility of electrons* in the electron transport layer.”

EX2019, 53:12-19; POR 1

Dr. Holmes Takes Inconsistent Positions

Initial Deposition

Q. Okay. In your opinion, in your expert opinion, is Lithium Q an electron donor?

A. I think I've already noted that my opinion on Lithium or on Liq is informed by Liu and is informed by Ma in the context of how I'm applying those prior art references with Diekmann and with Suzuki.

And **so in the context of Ma, Liq is mainly serving to improve mobility**. That's what Ma is arguing. And **in the context of Liu, that's really a very different system because it's a neat film of Liq** rather than doped into an electron transport material. So **I don't have any further speculation to offer on what Liq may or may not be doing**.

Reply Declaration

As further explained by Liu et al., Liq dissociates to act like a charge transfer dopant when exposed to the cathode metal. This effect would also be present when Liq is used to n-dope an ETL adjacent the cathode:

- Ex. 1018 (Liu, 2002) at abstract, 212-214: Liu explains that Liq, “covered with aluminum, was used as an electron injection layer.” (abstract). “An Liq electron injecting layer has also been tried in combination with Ca, Mg and Ag cathodes. Our experiments support the assumption that free lithium is released from lithium quinolate, as in the case of lithium fluoride, when Liq is over coated with active metals such as Al.” (abstract). Liu reports “improved electron injection.” (211). Liu explains that “lithium quinolate reacts partially with Ag, to give free lithium that improves electron injection.” (213). Liu further notes that “experimental results for Ca, Mg and Ag cathodes support the assumption that free lithium released from lithium quinolate is responsible for the improved device performance. (214).

Thus, Liq would act as an “electron-donor type n-dopant,” under even Dr. Pattison’s narrow view of what n-doping requires (Ex. 2018, ¶ 17, 23-27), when Liq is used as the ETL n-dopant in either the Diekmann OLED stack or the Suzuki OLED stack.

Liq in Ma Had a Different Mechanism of Operation Than n-Dopants in Diekmann

Petition (Diekmann)

Diekmann's doped layer 402 is situated between cathode 3 and radiation-emitting region 401 and is doped to "increase[] the conductivity of the doped organic layer and...lead[] to better injection of electrons from the cathode into the organic layer stack." Diekmann, 10; *see also id.*, 91. A POSITA would recognize doped layer 402 to be an "electron transport layer" which, as explained by the '425 patent, serves the function of "transport[ing] negative charge carriers—electrons—from the...cathode, to the radiation-emitting region." '425 patent, 24:59-62; Ex.1003, ¶¶32-42, 106-110.

Pet. 24

Ma

These J - V characteristics of electron-only devices suggest that a certain doping ratio of Liq to Bphen can improve the electron transport ability of the co-deposited layer. The advantage of using Bphen:Liq as ETL can be explained by electron hopping exchange alongwith their LUMO. In a single host device, electrons hop along the LUMO in Bphen. Since the LUMO-LUMO difference between Bphen (3.0 eV) [29] and Liq (3.1 eV) [28] is neglectable, subject to their similar LUMO. Transport manifolds alongwith their LUMO are expected to exhibit a certain extent of overlap after a mixing ratio goes beyond 17 wt.%. Therefore, it's likely that a large energetic disorder between Bphen and Liq contributes to electron hopping, implying that electron hopping among Bphen and Liq sites is favorable [29]. Therefore, the high electron conductivity of Bphen:Liq may be originated from short hopping length for electron transport, compared to the pure Bphen HTL. However, the current conduction is reduced slightly as the doping ratio further increases to 50 wt.%. This result may be attributed to the effect of carrier quench and generation of defects. The detailed reasons are possibly the same as that of the heavy doping concentration of F₄-TCNQ into *m*-MTDATA in the hole-only devices.

EX1010 (Ma), 813

With an EIL Layer, Liq in Ma Does Not Reduce Injection Barrier

Dr. Holmes
Expert Witness
SAMSUNG

- Q. So **in Ma, does the addition of Lithium Q to the Bphen layer help reduce the injection barrier?**
- A. That's a good question. **I don't think it does.** And I think that's why Ma intentionally includes a 1 nanometer Lithium fluoride electron injection layer under their cathode. And so I think I mentioned this very early in the day, that that allows all of these different electron transport layers to have good injection from aluminum because Lithium fluoride is there now to provide the assistance for electron injection which allows Ma's data shown in figure 5 to focus on the actual transport of electrons in the doped electron transport material.
- And so **I don't think Ma is looking at a change in injection.** I think Ma is looking at a change in conductivity and transport within the electron transport layer.

EX2019, 291:16-292:11; POR 29

Dr. Holmes Does Not Know How Exactly Ma Would Act if LiF-EIL Were Removed

Dr. Holmes
Expert Witness

SAMSUNG

Q. Do you know how the OLED device in Ma would behave if you removed the Lithium fluoride layer and uses only the Lithium Q and Bphen next to the cathode?

A. That's basically the inversion of the question I was just answering. **I don't know that for a fact.** In that case, if LIF is removed, then now the electronic transport layer will be both improving conduction and potentially also improving injection but **I don't know how that device specifically would look without seeing the data.**

EX2019, 293:14-294:2

Liq-Doped ETL Could Result in Increased Injection Barrier

JOURNAL OF APPLIED PHYSICS 109, 114511 (2011)

Effect of doping of 8-hydroxyquinolinatolithium on electron transport in tris(8-hydroxyquinolinato)aluminum

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Effect of doping of 8-hydroxyquinolinatolithium (Liq) on the electron transport properties of tris(8-hydroxyquinolinato)aluminum (Alq₃) has been investigated as a function of temperature and doping concentration by fabricating electron only devices. It has been observed that current density in the devices increases with the doping of Liq up to a doping concentration of 33 wt. % and then decreases. Current density-voltage (*J*-*V*) characteristics of 0, 15, and 33 wt. % Liq doped Alq₃ devices were found to be bulk limited and analyzed on the basis of trap charge limited conduction model. The *J*-*V* characteristics of 50 and 100 wt. % Liq doped Alq₃ devices were found to be injection limited and were analyzed using the Fowler-Nordheim model. The increase in current density with doping up to 33 wt. % was found to be due to an increase in electron mobility upon doping, whereas the decrease in current density above 33 wt. % was due to the switching of transport mechanism from bulk limited to injection limited type due to an increase in barrier height. Electron mobility and variance of energy distribution have been measured by using transient electroluminescence technique to support our analysis. Electron mobility for pure Alq₃ was found to be 1×10^{-6} cm²/V s, which increased to 3×10^{-5} cm²/V s upon doping with 33 wt. % Liq. The measured values of variance were 95, 87.5, 80, 72, and 65 meV for 0, 15, 33, 50, and 100 wt. % Liq doped Alq₃ respectively. The increase in electron mobility upon doping has been attributed to a decrease in energetic disorder upon doping as evidenced by the decrease in variance. The increase in barrier height for the higher doping concentration was due to the disorder related correction $\sigma^2/2kT$ in the barrier height, which decreases with the increase in doping concentration.

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I. INTRODUCTION

Doping of organic semiconductors was first studied in the 1960s,¹ however they have been extensively studied only after their use in enhancing the carrier injection and transport properties in optoelectronic devices^{2–15} such as organic light-emitting diodes (OLEDs) and organic solar cells (OSCs). p-type doping of organic semiconductors has been studied by using a series of techniques such as ultraviolet photoelectron spectroscopy (UPS), x-ray photoelectron spectroscopy (XPS), and low temperature conductivity measurements.^{16–24} As a result, p-type doping has been explained with a simple picture of doping where holes are generated in the transport level by thermal excitation of electrons to a well defined acceptor level provided by the dopant.²⁵ 2,3,5,6-tetrafluoro-7,7',8,8'-tetracyanoquinodimethane (F₄-TCNQ) has become a vital p-type dopant^{16–25} for hole transport and injecting materials in organic light-emitting diodes.

Despite the extensive studies on p-type doping, n-type doping of organic semiconductors still remains a challenge. To create a free negative carrier in an organic semiconductor, a donor molecule is required whose highest occupied molecular orbital

(HOMO) value should match with the lowest unoccupied molecular orbital (LUMO) of the host molecule. Systematic studies of n-type doping have been performed on materials with low-lying LUMO levels, that is, high electron affinity. Reasonable increases in conductivity have been achieved in naphthalene-tricarboxylic dianhydride (NTCDA) using the dopant bis(ethylenedithio)tetrathiafulvalene (BEDT-TTF).¹ The common electron transporting molecules used in OLEDs, however, have LUMO value around 3 eV which limits the choice of donor molecules for this application. Further, organic molecules with HOMO around 3 eV are highly unstable in air. Alkali metals like Li and Cs and their oxides are also used for n-type doping of organic electron transport material,^{10–12} and were found useful in many instances. But this type of doping has many serious problems, like damage to underlying organic layers due to high evaporation temperatures. Furthermore, they have also been reported as luminescence quenchers,²⁶ which act to reduce the lifetime and stability of devices. Hence, there is a need for molecular dopants for enhancement of electron transporting properties in OLED applications. Recently 8-hydroxyquinolinatolithium (Liq) has been used as a dopant in electron transporting molecules Batho phenanthroline (Bphen) and tris(8-hydroxyquinolinato) aluminum (Alq₃).^{27–29} But the reason for the enhanced electron transport property of these molecules was not well understood, especially the requirement of high doping concentrations (around

IV. CONCLUSION

Effect of doping of Liq on the electron transport in Alq₃ has been studied as a function of doping concentration. Variance of energy distribution has been found to be decreasing with the increase in doping concentration of Liq into Alq₃. Decrease in variance gives rise to the decrease in energetic disorder of Liq doped Alq₃ matrix, thus increasing the electron mobility in Alq₃ with doping of Liq. Furthermore, the energetic disorder related correction $\sigma^2/2kT$ in barrier height decreases with the decrease in variance of energy distribution with doping. This causes the increase in barrier height with doping of Liq, which then resulted in a switching of conduction mechanism from bulk to injection limited type.

EX2004 (Yr. 2011), 114511-7; Sur-Reply 15

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Endo Adopted In Situ Chemical Doping Theory Because of Incorrect Energy Levels

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Organic Electroluminescent Devices Having Metal Complexes as Cathode Interface Layer

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We developed novel cathode systems, composed of metal complexes, for organic electroluminescent (EL) devices. Alkaline metal complexes, such as mono(8-quinolinolato) lithium (Li_q), were used as a cathode interface layer in combination with metallic Al as a cathode material. Similarly, doping the organic electron-transporting layer with alkaline metal complexes was also effective in facilitating the electron injection from the cathode to the organic layer, which results in reducing the drive voltage. In both cases, we assume that alkaline metal ions in the complexes are reduced to free metal by thermally activated Al. Then, the free alkaline metal reacts with electron-transporting materials, forming the radical anions, which facilitate electron injection from the Al cathode. [DOI: 10.1143/JJAP.41.L800]

KEYWORDS: organic, electroluminescence, electron injection, metal complex, thermal reduction

In organic electroluminescent (EL) devices, organic thin layers are sandwiched between the anode and the cathode, and the organic emitting centers are excited by the recombination of holes and electrons injected from the electrodes. It is therefore important to balance the number of holes and electrons injected from the electrodes to the emitter layer to achieve high device performance. A typical material used for the anode is indium-tin oxide (ITO) which is transparent in the visible region and has a high work function of approximately 5.0 eV which is suitable for injecting holes into the typical hole-transporting organic materials with ionization potentials of approximately 5.2–5.4 eV. For the cathode material, Mg:Ag alloy (10:1) was first introduced by Tang and Vanslyke in 1987 and has been widely used.¹⁾ Later, Al:Li alloy was introduced to reduce the drive voltage.²⁾ Double-layer cathodes, such as Li/Ag,³⁾ Li₂O/Al,⁴⁾ and LiF/Al⁵⁾ were also developed and are now widely used. Very recently, we introduced a new type of cathode interface layer composed of metal complexes such as 8-quinolinolato lithium (Li_q) and 8-quinolinolato sodium (Na_q).^{6,7)} The details of the device characteristics using this type of cathode interface layer will be discussed here.

Figure 1 shows organic materials used in this study; a hole-transporting *N,N'*-di(1-naphthyl)-*N,N'*-diphenyl-benzidine (α -NPD), electron-transporting and light-emitting tris(8-quinolinolato) aluminum (III) (Alq₃), electron-transporting bathophenanthroline (Bphen) and several electron-injecting alkaline metal complexes. The ligands for the metal complexes are 8-quinolinolato (q), acetylacetonato (acac) and dipivaloylmethanato (dpm). Liq, for example, was synthesized from lithium hydroxide and 8-quinolinol, and was purified by the train sublimation method. Metal complexes with acac ligands and dpm ligands were purchased from Kojundo Chemical Laboratory Co., Ltd.

Figure 1 also illustrates the two types of device structures, (a) ITO (1100 Å)/ α -NPD (500 Å)/Alq₃ (700 Å)/alkaline metal complex (10 Å)/Al and (b) ITO (1100 Å)/ α -NPD (500 Å)/Alq₃ (x Å)/alkaline metal complex doped layer (700 – x Å)/Al. The organic layers were successively deposited onto a precleaned ITO-coated glass substrate at 1.0 × 10⁻⁵ Torr. Alkaline metal complex-doped organic layers were deposited by coevaporation method and the molar ratio be-

tween the alkaline metal complex and electron-transporting material is fixed to be 1:1. Finally, the Al top electrode was deposited at 7.0 × 10⁻⁶ Torr using a metal shadow mask to define an emitting area of 0.5 × 0.5 cm². EL spectra were taken using an optical multichannel analyzer PMA 11 (Hamamatsu Photonics K.K.). Luminance measurements were carried out with a Topcon BM-8 luminance meter at room temperature.

EL devices having alkaline metal complexes, as a cathode interface layer, between Alq₃ and Al, exhibited high performance compared with a device without a cathode interface layer. Figures 2 and 3 show luminance-voltage (*L-V*) and current density-voltage (*J-V*) characteristics of EL devices having Liq, Naq, Liacac, or Lidpm as a cathode interface layer. The highest luminance values observed for each device were 27000 cd/m² at 12 V for Liq, 31500 cd/m² at 13 V for Naq, 23000 cd/m² at 12 V for Liacac, and 17000 cd/m² at 13 V for Lidpm. Current efficiencies were 7.3 cd/A at 11 V for Liq, 9.0 cd/A at 12 V for Naq, 6.7 cd/A at 11 V for Liacac, and 6.3 cd/A at 12 V for Lidpm. These results clearly demonstrate that the insertion of alkaline metal complexes between electron-transporting layer, Alq₃, and the cathode, Al, is effective in reducing the drive voltages and improving device efficiencies. Devices without the interface layer exhibited low efficiencies which are due to the low electron injection efficiency. Using the alkaline metal complexes as a cathode interface layer improves the electron injection efficiency, resulting in a balanced hole-electron injection and a higher carrier recombination efficiency.

Among the Li complexes, Liq exhibited the highest current efficiency, which may be due to the electron-transporting ability of Liq while other complexes such as Liacac are insulating materials. Comparing central metal ions of the complexes with the same ligand, we found that the current efficiency increases as atomic mass increases. For example, in the case of the devices using quinolinolato ligand, the current efficiency of the device using Naq was higher than that using Liq. Similarly, in the case of dipivaloylmethanato ligand, the device using Kdpm exhibited the highest current efficiency.

The lowest unoccupied molecular orbital (LUMO) level of Liq is –2.5 eV from the vacuum level while that of Alq₃ is –3.0 eV, making the barrier heights for electron injection from Al cathode, having a work function of –4.2 eV, to the organic layers lower for Alq₃. Therefore, the improvement of

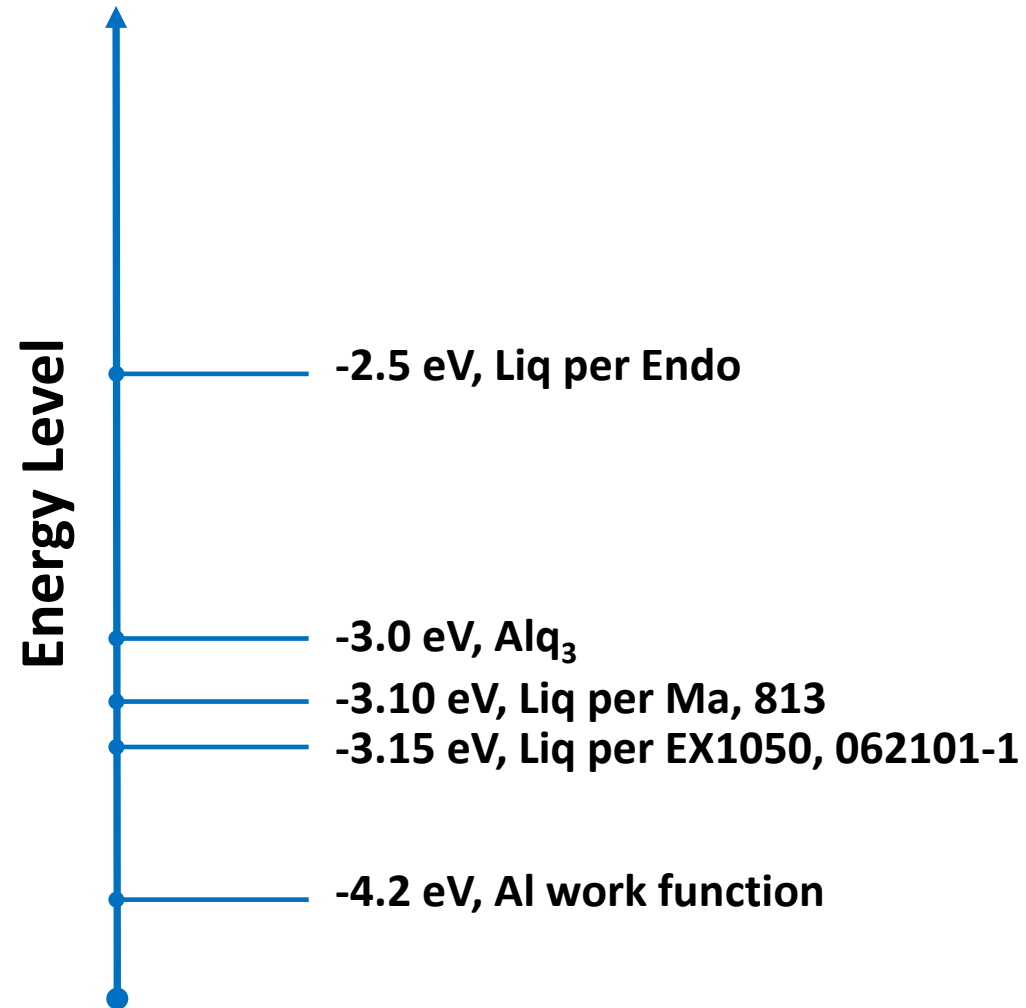
The lowest unoccupied molecular orbital (LUMO) level of Liq is –2.5 eV from the vacuum level while that of Alq₃ is –3.0 eV, making the barrier heights for electron injection from Al cathode, having a work function of –4.2 eV, to the organic layers lower for Alq₃. Therefore, the improvement of electron injection from Al cathode to the organic layer cannot be explained by such simple energy diagrams. We proposed^{6,7)} that the central “metal ions” in these metal complexes are reduced to free metal by the reaction between the complexes and thermally activated aluminum in vacuum. Then the free alkaline metal liberated from the complexes reacts with Alq₃, forming the radical anions, which facilitate electron injection from the Al cathode. We call this an *in situ* chemical doping process. In fact, the reduction of alkaline metal compounds by thermally activated metals such as Al is often used in metallurgy.⁸⁾ In that field, high purity alkaline and alkaline-earth metals are obtained by such reactions. For

EX1049, L800-801; Sur-Reply 15

In-Situ Chemical Doping Theory Relied on in Reply to Argue Liq Acting as Electron Donor

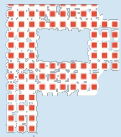
*Corresponding author. E-mail address: kido@yz.yamagata-u.ac.jp

Others Reported Liq LUMO Levels Lower Than That for Alq₃



There Had to Be a Reason for the Swap

Dr. Pattison
Expert Witness



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Q. We look at column 30, back to the '425 patent, and go down to line 25. The patent says that "For all of the layers, of course, other matrix materials, dopants or emitter materials are possible, as are other compositions of the mixed matrix materials. Further materials for emitter materials, transfer materials and dopants are possible and can be exchanged at any time."

Did I read that correctly?

A. Yes.

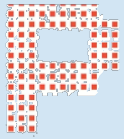
Q. So, I mean, do you agree?

A. Well, I'd say other materials are possible and can be exchanged, but the – the implications -- **you'd need to have some motivation to pursue them and to understand the – the implications of OLED performance.** Right? So this is a -- kind of a vague statement. But, yeah, I -- **you could -- you could try anything. The question would be why.**

EX1043, 135:3-22 (objections omitted); Sur-Reply 23-234

Motivation to Substitute Liq Depended on How Well the Material was Known and Mechanisms of Operation

Dr. Pattison
Expert Witness



pictiva displays

- Q. So independent of the disclosure that we were looking at in the patent, just based on a knowledge that a POSITA at the time of the '425 patent would have, **assuming that we're talking about the layer performing the same function, would that POSITA have any difficulty swapping -- swapping out known materials for that layer in exchange for other known materials for that layer to design a different OLED device stack?**
- A. It depends on the level of mature -- you said "known material." I mean, that --
- Q. Yeah
- A. -- **really would depend on the level of how well the material's known and if the materials that you're talking is actually operating by the same physical mechanism.**

You know, if you change physical mechanisms, there -- there will always be compatibility issues that will need to get resolved and figured out and can't -- you can't be sure that they're not going to be there with a new material.

EX1043, 132:1-134:10 (objections omitted); Sur-Reply 14

Conclusory Expectation of Success

Petition

A POSITA would also have reasonably expected success in implementing Liq as the dopant in the n-doped electron transport layer in Diekmann's OLED . Ex.1003, ¶¶145-146. Doing so would be a routine practice for a POSITA to implement with a high probability of success, particularly as Diekmann already contemplates an n-doped electron transport layer. Ex.1003, ¶145.

Pet. 38

Holmes Declaration (EX1003)

145. Additionally, as I explained above, use of Liq as a dopant in an OLED electron transport layer was well-known in the art at the time, and was specifically taught by Ma. Doing so would be a routine practice for a POSITA to implement with a high probability of success, particularly considering that Diekmann already contemplates an analogous n-doped layer for electron transport. As such, a POSITA could readily have implemented an Liq-doped electron transport layer in Diekmann's OLED.

146. Accordingly, a POSITA would have appreciated that Diekmann in view of Ma discloses this claim.

EX1003, para. 145-146

Conclusory Expectation of Success

Petition

228. A POSITA would have reasonably expected success in using an Liq-doped electron transport layer in Suzuki's OLED for the same reasons discussed above with regards to Diekmann's OLED. Ex.1003, ¶¶225-228. Moreover, Suzuki already contemplates an n-doped electron transport layer, so using an Liq-doped electron transport layer merely involves using a different dopant. This would be a routine matter well within the ability of a POSITA. Ex.1003, ¶228.

Pet. 70

Holmes Declaration

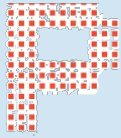
227. Indeed, Suzuki already contemplates an n-doped electron transport layer, so using an Liq-doped electron transport layer merely involves using a different dopant. As I explained above, Liq was well-known in the art at the time to have good electron injection and transport properties. A POSITA thus would have recognized that Liq would be an advantageous dopant for Suzuki's doped electron transport layer.

228. Additionally, as I explained above, use of Liq as a dopant in an OLED's electron transport layer was well-known in the art at the time, and was specifically taught by Ma. Doing so would be a routine practice which would be easy for a POSITA to implement with a high probability of success, particularly considering that Suzuki already contemplates an analogous n-doped layer for electron transport. As such, a POSITA could readily have implemented an Liq-doped electron transport layer in Suzuki's device. Accordingly, a POSITA would have appreciated that Suzuki in view of Ma discloses this claim.

EX1003, para. 227-228 (225-226 re motivation)

No Reasonable Expectation of Success Upon Change of Operating Mechanisms

Dr. Pattison
Expert Witness



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Q. Just as a matter of being able to make the new stack by replacing one type of material for another, I mean, the capability exists for a POSITA to do that, not thinking about, you know, motivations for doing so?

A. **You could make a stack. I'm not sure if you would make an OLED.**

EX1043, 134:17-135:2 (objections omitted); Sur-Reply 14, 23

Low Skill Level

Dr. Holmes
Expert Witness

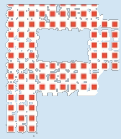
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64. Considering these factors, it is my opinion a person of ordinary skill in the art (POSITA) at the time of the alleged inventions would have had a Bachelor's degree in chemistry, physics, or a comparable field of study, and at least three years of research or industrial experience with organic optoelectronic light-emitting devices. An individual with additional education could have less research or industrial experience and vice versa.

EX1003, para. 64

POSITAs Would Not Have Sufficient Skills to Design Material Sets

Dr. Pattison
Expert Witness



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Q. Would a person of ordinary skill in the art even know how to design the experiments in order to make the appropriate concentration of thickness changes?

A. No.

The way the -- the skill level of the person of skill in the art that we've been talking about is not someone designing experiments with new kind of untested materials.

They'd be more a person operating the machine under the direction of a professor or a more senior researcher, maybe someone with a Ph.D.

Q. Okay. So the people who would design -- strike that.

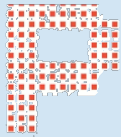
So based on your knowledge and experience, the person who would design the experiments would be a person with high skill levels than that designed for a person of ordinary skill in the art; is that correct?

A. *I agree. That's my experience.*

EX1043, 223:18-224:18 (objections omitted); Sur-Reply 20; Reply 14

Dr. Pattison Has Collaborated with University and Industrial Researchers on Hundreds of DOE OLED Projects

Dr. Pattison
Expert Witness



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- “my job at DoE is, and was, to *oversee the scientific work on OLED materials and devices* and to make sure that it was progressing well and achieving the goals of the contracts”
- “There -- you know, that -- that– *hundreds of projects that I was -- over the years that I’ve managed on behalf of DoE*”
- “the DoE program has internal experts like – like myself, and internal consultants like myself that they -- they hire”
- “when I *started* at DoE, it was *2006*. And that was an early activity of mine, was to – was to recruit new reviewers”
- “There had been hundreds of projects OLED-related with *Universal Display Corporation, Michigan State, I believe Princeton, University of North Texas, OLED Works, Pacific Northwest National Lab. Just – University of Southern California. Private – other private companies. There had just really been hundreds of R&D projects over the years.*”

EX1043, 58:14-62:5 (objections omitted); Sur-Reply 20

Work on Allegedly Analogous LiF (EX1051) Done By Another Expert

Dr. Holmes
Expert Witness
SAMSUNG

Q. So that will be **Exhibit 1051**. (Exhibit 1051 was identified.)

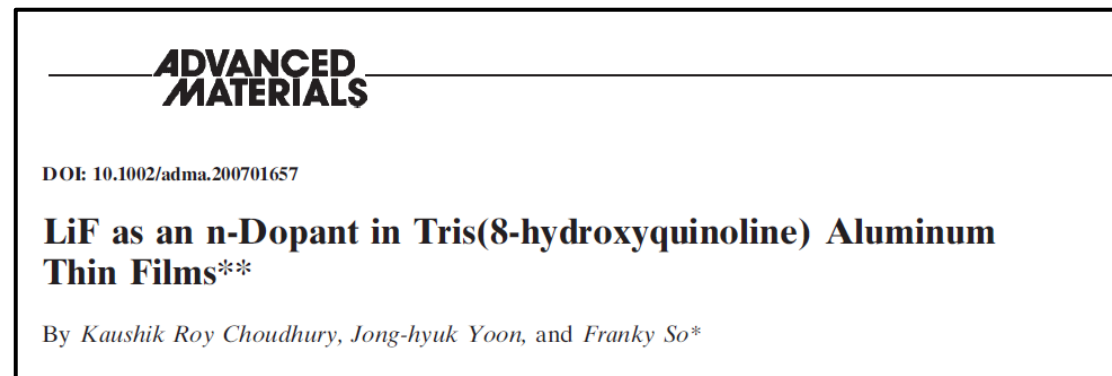
A. I have it. Please repeat your question.

Q. Do you know Professor So, Franky So?

A. Yes, I know Professor So.

Q. Do you know, is he an expert in the field of OLED at the time of the invention?

A. Yes, ***I would categorize Franky So as an expert at the time of the invention.***



EX2032, 61:3-16 (objections omitted); Sur-Reply 20

Newly Cited Work Features a Large Group of Researchers

Japanese Journal of Applied Physics 48 (2009) 062101

REGULAR PAPER

Improved Efficiency and Lifetime of Organic Light-Emitting Diode with Lithium-Quinolate-Doped Electron Transport Layer

Sung Hoon Choi*, Seok Jong Lee, Kwang Yeon Lee, Hee Seok Yang, Kyu-il Han, Kwang Hyun Kim, Sang Dae Kim, Hyo Dae Bae, and Yoon Heung Tak

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In this letter, we analyze the device performance of organic light-emitting diodes (OLEDs) with a lithium-quinolate (Liq)-doped electron transport layer (ETL). When the ETL:Liq doping ratio is 1 : 3, the luminous efficiency of the OLED is 7.1 cd/A; that of an OLED with a nondoped ETL is 5.3 cd/A. The corresponding OLED lifetime is improved by threefold that with a nondoped ETL, owing to the improved hole-electron charge balance and electrical stability of the Liq-doped ETL film. Additionally, when the Liq-doped device was combined with a Mg/Al cathode, the OLED produced had a longer lifetime than the other tested devices. © 2009 The Japan Society of Applied Physics

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EX1050 at 1; Sur-Reply 20

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Diekmann Involved 11 Co-Inventors

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KM, KN, KP, KR, KZ, LA, LC, LK,

[Continued on next page]

Ma Does Not Suggest or Motivate the Use of Liq in Diekmann or Suzuki

- Objective evidence of non-obviousness in using Liq in PHOLEDs
- Different operating principles
- No evidence of non-speculative benefits over Diekmann's and Suzuki's disclosed embodiments

Motivation to Combine Not Shown by the Fact that the Elements were Known or Disclosed as Interchangeable

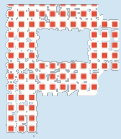


- “the Board erred as a matter of law with regard to the motivation to combine.”
- ***“It does not suffice to meet the motivation to combine requirement to recognize that two alternative arrangements ...were both known in the art”*** and allegedly ***“may be applied to any computer controlled aiming system”***
- Such disclosures “do not provide any reason why a skilled artisan would use [one alternative] instead of [the other]”

Virtek Vision Int’l ULC v. Assembly Guidance Sys., 97 F.4th 882, 886-87 (Fed. Cir. 2024)

There Had to Be a Reason for the Swap

Dr. Pattison
Expert Witness



pictivadisplays

Q. We look at column 30, back to the '425 patent, and go down to line 25. The patent says that "For all of the layers, of course, other matrix materials, dopants or emitter materials are possible, as are other compositions of the mixed matrix materials. Further materials for emitter materials, transfer materials and dopants are possible and can be exchanged at any time."

Did I read that correctly?

A. Yes.

Q. So, I mean, do you agree?

A. Well, I'd say other materials are possible and can be exchanged, but the – the implications -- **you'd need to have some motivation to pursue them and to understand the – thea implications of OLED performance.** Right? So this is a -- kind of a vague statement. But, yeah, I -- you could -- **you could try anything. The question would be why.**

EX1043, 135:3-22 (objections omitted); Sur-Reply 23-234

Comparison Should Be Against an Alternatively n-Doped Device

Alleged Motivation

A POSITA would have been motivated to use an Liq-doped electron transport layer in the organic layer stack of Diekmann due to the known benefits of an Liq-doped electron transport layer. Ex.1003, ¶¶140-143. As Ma explains, “[s]ince the discovery of organic light-emitting devices (OLEDs), there has been a considerable interest in developing OLEDs with high efficiency.” Ma, 810. “In order to lower driving voltage and enhance power efficiency, it is critical to enhance the carrier injection from the electrode to the transporting layer and to increase the transport conductivity.” *Id.* Ma teaches that a “doping concentration of the Liq” in the electron transport layer results in increased current density and reduced driving voltage, compared to the undoped device. *Id.*, 813. Ma further teaches that “a certain doping ratio of Liq to Bphen can improve the electron transport ability of the co-deposited layer.” *Id.*

Pet. 37-38; see also Pet. 69-70

Fact: Comparison Should Be to Doped Devices

Dr. Holmes:

“In Diekmann and Suzuki, the inventor has already decided that they are making a doped device. And ***so the comparison wouldn't be necessarily to an undoped device*** because that decision is already made. The doping's already there. And ***so here the question would be*** is there an expectation of success in ***using Liq as the n-dopant as an alternative to whatever n-dopant is already being used.***”

EX2019, 138:17-139:2; POR 39

No Comparison of Performance Against Suzuki's or Diekmann's Existing Dopants

Dr. Holmes Failed to Make the Proper Comparison

Dr. Holmes
Expert Witness

SAMSUNG

- Q. Do you know what's the relative performance in terms of power efficiency, luminescence efficiency, current efficiency and driving voltages are of a cesium or Lithium doped electron transport layer versus a Lithium Q doped transport layer?
- A. Again, I think I made clear the references that I'm using and **I don't have a direct comparison between Liq doped and alternate doped electron transport materials in terms of performance.**

EX2019, 140:16-141:5; POR 12

Lower Driving Voltage and Higher Efficiency Not Guaranteed Even for BPhen:Liq

Alleged Motivation

A POSITA would have been motivated to use an Liq-doped electron transport layer in the organic layer stack of Diekmann due to the known benefits of an Liq-doped electron transport layer. Ex.1003, ¶¶140-143. As Ma explains, “[s]ince the discovery of organic light-emitting devices (OLEDs), there has been a considerable interest in developing OLEDs with high efficiency.” Ma, 810. “In order to lower driving voltage and enhance power efficiency, it is critical to enhance the carrier injection from the electrode to the transporting layer and to increase the transport conductivity.” *Id.* Ma teaches that a “doping concentration of the Liq” in the electron transport layer results in increased current density and reduced driving voltage, compared to the undoped device. *Id.*, 813. Ma further teaches that “a certain doping ratio of Liq to Bphen can improve the electron transport ability of the co-deposited layer.” *Id.*

Pet. 37-38; *see also* Pet. 69-70

Fact: Alleged Benefits Not Guaranteed

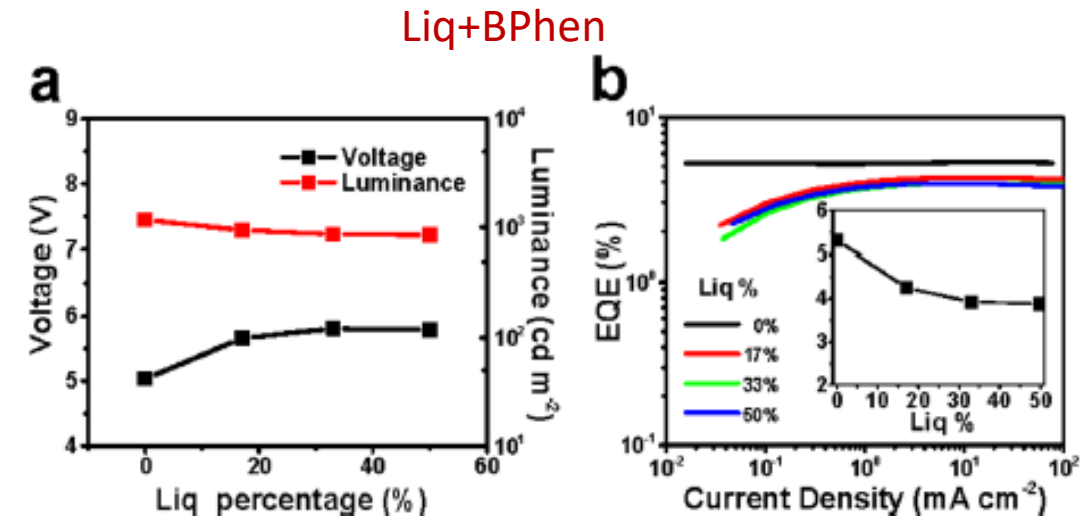


Figure 4. (a) Driving voltages, luminance, and (inset of panel (b)) external quantum efficiency (EQE) of the blue devices upon mixing percentage of Liq in the mETL under a constant current density of 25 mA cm⁻². (b) External quantum efficiency (EQE) versus current density for the devices upon mixing percentage of Liq in the mETL. (c) Relative luminance over the operation time under a constant current density of 25 mA cm⁻² for the devices upon mixing percentage of Liq in the mETL.

EX2104, Figs 4(a), 4(b) inset; POR 33

Liq Doping: Increased Voltage & Reduced EQE

Lower Driving Voltage and Higher Efficiency Not Guaranteed Upon Liq-Doping: Lacking Motivation or Reasonable Expectation of Success

Alleged Motivation

A POSITA would have been motivated to use an Liq-doped electron transport layer in the organic layer stack of Diekmann due to the known benefits of an Liq-doped electron transport layer. Ex.1003, ¶¶140-143. As Ma explains, “[s]ince the discovery of organic light-emitting devices (OLEDs), there has been a considerable interest in developing OLEDs with high efficiency.” Ma, 810. “In order to lower driving voltage and enhance power efficiency, it is critical to enhance the carrier injection from the electrode to the transporting layer and to increase the transport conductivity.” *Id.* Ma teaches that a “doping concentration of the Liq” in the electron transport layer results in increased current density and reduced driving voltage, compared to the undoped device. *Id.*, 813. Ma further teaches that “a certain doping ratio of Liq to Bphen can improve the electron transport ability of the co-deposited layer.” *Id.*

Pet. 37-38; *see also* Pet. 69-70

Fact: Alleged Benefits Not Guaranteed

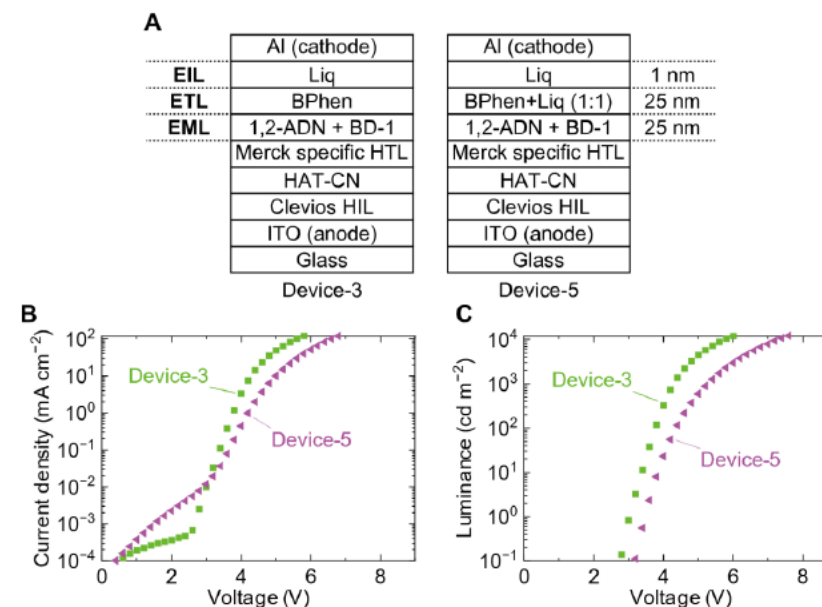


Figure S1. A) Multilayer structure of blue OLEDs employing BPhen and Liq-doped Bphen (50 wt%) as the ETL, and employing Liq as the EIL. B) Current density–voltage characteristics of blue OLEDs. C) Luminance–voltage characteristics of blue OLEDs.

EX2105, S1; POR 33

Liq Doping: Increased Voltage & Reduced EQE

Even Electron Transport May Be Reduced Depending on ETL

Alleged Motivation

A POSITA would have been motivated to use an Liq-doped electron transport layer in the organic layer stack of Diekmann due to the known benefits of an Liq-doped electron transport layer. Ex.1003, ¶¶140-143. As Ma explains, “[s]ince the discovery of organic light-emitting devices (OLEDs), there has been a considerable interest in developing OLEDs with high efficiency.” Ma, 810. “In order to lower driving voltage and enhance power efficiency, it is critical to enhance the carrier injection from the electrode to the transporting layer and to increase the transport conductivity.” *Id.* Ma teaches that a “doping concentration of the Liq” in the electron transport layer results in increased current density and reduced driving voltage, compared to the undoped device. *Id.*, 813. Ma further teaches that “a certain doping ratio of Liq to Bphen can improve the electron transport ability of the co-deposited layer.” *Id.*

Pet. 37-38; see also Pet. 69-70

Fact: Alleged Benefits Not Guaranteed



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Unraveling the complexation effect of electron transporting materials with Liq on the charge transport properties: A theoretical perspective

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A B S T R A C T

The effect of coordinated structures of electron transporting materials (ETMs) with lithium quinolate (Liq) on charge transport characteristics was theoretically investigated. The reorganization energies of holes (λ_h) and electrons (λ_e) of ETM-Liq simultaneously increased compared with those of ETMs, indicating that the charge transporting is retarded. Interestingly, λ_e of ETM-(Liq)_n increased, while λ_h decreased. Regarding transfer integrals for holes (t_h) and electrons (t_e), t_h 's of T2T-Liq and BPyTP2-Liq were significantly reduced with t_e of T2T-Liq decreasing tremendously. Consequently, the hopping rates of holes of Liq-complexes were reduced, while that of electrons of BPyTP2-Liq increased and that of T2T-Liq/TSPO1-Liq was reduced.

EX2006, Abstract; POR 33

Not Obvious if Having to Try Each of Numerous Possible Choices



“to have a reasonable expectation of success, one must be motivated to do more than merely to vary all parameters or try each of numerous possible choices until one possibly arrived at a successful result”

In re Stepan, 868 F.3d 1342, 1347 (Fed. Cir. 2017)

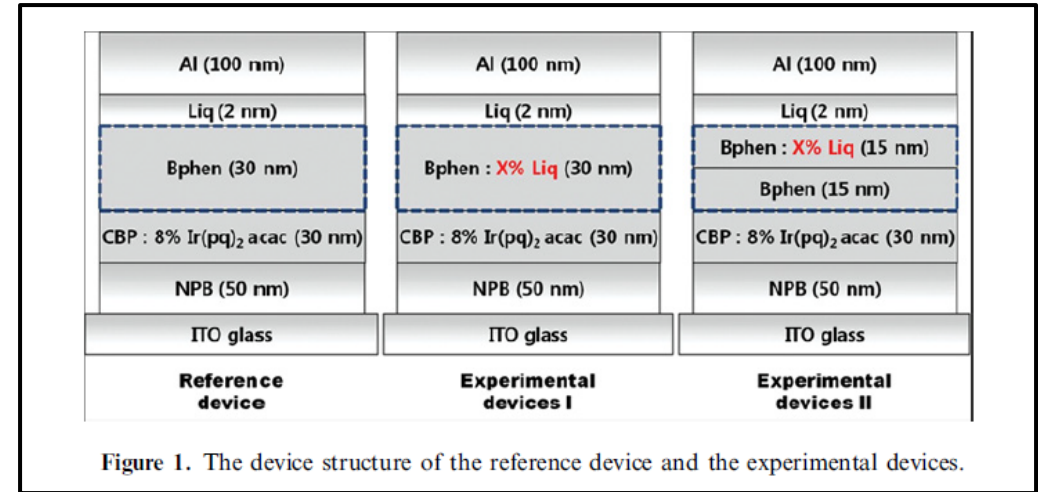
Improved Efficiency Achieved with a Different OLED Device Structure

Alleged Motivation

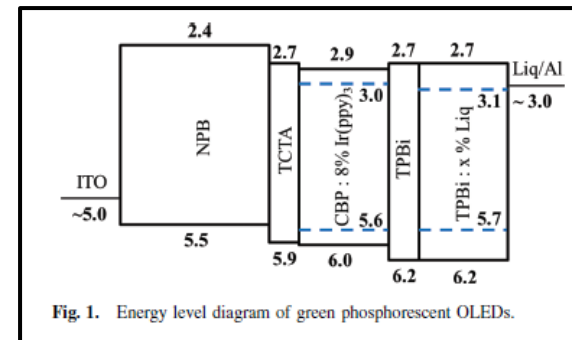
Pictiva cites studies from years after the priority date (POR, 33), which do not reflect the knowledge or expectations of POSITAs at that time. *Bristol-Myers Squibb Co. v. Teva Pharms. USA, Inc.*, 752 F.3d 967, 974 (Fed. Cir. 2014) (post-invention discovery not known to POSITA at time of invention does not negate obviousness); *Ex Parte Jochen G. Salfeld et. al*, APPEAL 2015-007511, 2017 WL 2132356, at *17 (PTAB May 15, 2017). Contrary to Pictiva's claim (POR, 33), these studies do not show reduced performance with Liq-doped ETLs. The studies closest to 2008 show Liq-doped ETL improved efficiency and operating voltage, consistent with POSITAs' expectations based on Ma. Ex.1044, ¶¶37-39; Ex.2024 at 7998; Ex.2003 at 53; Ex.1003, ¶¶140-143; Ma, Fig. 3, 812-813.

Pet. 37-38; see also Pet. 69-70

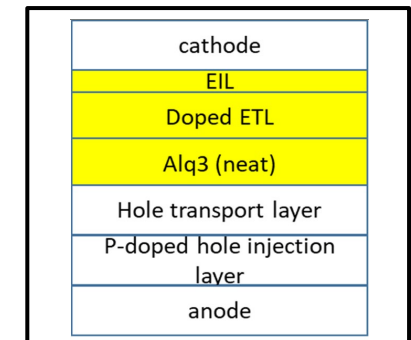
Fact: All Included Separate EIL Layer, Unlike Proposed OLEDs



EX2003 (Yr. 2011)



EX2024 (Yr. 2013); EX2032, 238:4-18 (Dr. Holmes confirming a separate Liq EIL layer between cathode and ETL)



No Demonstration of Electron Emission

Alleged Motivation

Moreover, Diekmann teaches that the n-dopant for its electron transport layer should be “suitable for emitting electrons” and “preferably has a low diffusion constant within the organic layer stack, as is usually the case with large atoms or molecules, for example.” Diekmann, 10. Liq was known to have good electron injection and transport properties and has a larger molecular weight than Diekmann’s preferred n-dopants (cesium, barium, and lithium fluoride). *See* Ex.1003, ¶¶144-145; *see also* Ex.1031, 4495; Ex.1018. A POSITA thus would have recognized that Liq would be an advantageous dopant for Diekmann’s doped layer. Ex.1003, ¶144.

Pet. 37-38; *see also* Pet. 69-70

Fact

- Q. Ma does not say there is an increase in the number of electrons, does it?
- A. ***Ma specifically focuses on the change in mobility*** and specifically via the improvement in hopping transport in the electron transport material.

EX2019, 47:12-18; POR 1

No Evidence that Liq Is Suitable for Emitting Electrons When Doping ETLs

No Demonstration of Reduced Diffusion

Alleged Motivation

Moreover, Diekmann teaches that the n-dopant for its electron transport layer should be “suitable for emitting electrons” and “preferably has a low diffusion constant within the organic layer stack, as is usually the case with large atoms or molecules, for example.” Diekmann, 10. Liq was known to have good electron injection and transport properties and has a larger molecular weight than Diekmann’s preferred n-dopants (cesium, barium, and lithium fluoride). *See* Ex.1003, ¶¶144-145; *see also* Ex.1031, 4495; Ex.1018. A POSITA thus would have recognized that Liq would be an advantageous dopant for Diekmann’s doped layer. Ex.1003, ¶144.

Pet. 37-38; *see also* Pet. 69-70

Fact: Free Li Generated When Liq Abuts Cathode

“So all I can state on that point is referring back to Liu, which is an exhibit I already mentioned earlier. **Liu shows that Liq can release or can be dissociated upon metal deposition.** Whether or not that happens for Liq doped into an ETL of Diekmann, I -- it's hard to say.”

EX2019, 27:4-9; POR 35, 38

Li Has Higher Diffusivity than Cs, Ba & LiF

No Demonstration of Reduced Diffusion

Alleged Motivation

Moreover, Diekmann teaches that the n-dopant for its electron transport layer should be “suitable for emitting electrons” and “preferably has a low diffusion constant within the organic layer stack, as is usually the case with large atoms or molecules, for example.” Diekmann, 10. Liq was known to have good electron injection and transport properties and has a larger molecular weight than Diekmann’s preferred n-dopants (cesium, barium, and lithium fluoride). *See* Ex.1003, ¶¶144-145; *see also* Ex.1031, 4495; Ex.1018. A POSITA thus would have recognized that Liq would be an advantageous dopant for Diekmann’s doped layer. Ex.1003, ¶144.

Pet. 37-38; *see also* Pet. 69-70

Fact: Liq Can Generate Free Li

Pictiva’s argument that Liq does not act as an electron donor “in Ma” (POR, 29) fails to consider Liq’s function when used in Diekmann or Suzuki. In Ma, increased current due to Liq doping could be attributed to increased conductivity in the ETL rather than electron injection because Ma used a separate EIL. Ex.2019, 291:16-293:13; Ex.1044, ¶29. When Liq is used in Diekmann’s or Suzuki’s device, the Liq-doped ETL abuts the cathode. Ex.1044, ¶29; Diekmann, Fig. 2 (layer 402); Suzuki, Fig. 9 (layer 7); Ex.1003, ¶¶41-42. POSITAs would understand, with subsequent cathode deposition, Liq would dissociate, facilitating injection of electrons from the cathode. Ex.1049 at L801-L802 (Liq doping activated by cathode); Ex.1018 at abstract, 212-214 (lithium release from Liq improves electron injection).

Reply 9 (cited on Sur-Reply 16)

Li Has Higher Diffusivity than Cs, Ba & LiF

No Demonstration of Reduced Diffusion

Alleged Motivation

Moreover, Diekmann teaches that the n-dopant for its electron transport layer should be “suitable for emitting electrons” and “preferably has a low diffusion constant within the organic layer stack, as is usually the case with large atoms or molecules, for example.” Diekmann, 10. Liq was known to have good electron injection and transport properties and has a larger molecular weight than Diekmann’s preferred n-dopants (cesium, barium, and lithium fluoride). *See* Ex.1003, ¶¶144-145; *see also* Ex.1031, 4495; Ex.1018. A POSITA thus would have recognized that Liq would be an advantageous dopant for Diekmann’s doped layer. Ex.1003, ¶144.

Pet. 37-38; *see also* Pet. 69-70

Fact: Liq Releases Li More Readily than LiF



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Journal of Industrial and Engineering Chemistry 14 (2008) 676–678

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Efficient electron injection in organic light-emitting diodes
using lithium quinolate/Mg:Ag/Al cathodes

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Received 5 February 2008; accepted 1 April 2008

Liq is useful as an electron injection material as well as electron transport material. Binding energy between Li and quinolate unit is weaker than that of LiF and it can give better performances as an electron injection layer than LiF [10]. LiF has been known to dissociate into Li and F during deposition of Al on top of LiF and Al deposition on Liq can also induce effective dissociation of Liq into Li and quinolate unit. In particular, reactive metals such as Ca and Mg:Ag can activate Liq dissociation due to easy dissociation of Liq [11,12]. It was already reported that Ca activated LiF and Liq dissociation during deposition, resulting in improved electron injection [12]. Therefore, it is expected that Mg:Ag deposition on Liq can also induce efficient electron injection through interfacial energy barrier lowering. In this work, Mg:Ag was used instead of Mg because Mg could not be deposited on organic layer due to low sticking coefficient of Mg.

No Demonstration of Reduced Diffusion

Alleged Motivation

Moreover, Diekmann teaches that the n-dopant for its electron transport layer should be “suitable for emitting electrons” and “preferably has a low diffusion constant within the organic layer stack, as is usually the case with large atoms or molecules, for example.” Diekmann, 10. Liq was known to have good electron injection and transport properties and has a larger molecular weight than Diekmann’s preferred n-dopants (cesium, barium, and lithium fluoride). *See* Ex.1003, ¶¶144-145; *see also* Ex.1031, 4495; Ex.1018. A POSITA thus would have recognized that Liq would be an advantageous dopant for Diekmann’s doped layer. Ex.1003, ¶144.

Pet. 37-38; *see also* Pet. 69-70

Fact: Liq Can Generate Free Li

“So all I can state on that point is referring back to Liu, which is an exhibit I already mentioned earlier. **Liu shows that Liq can release or can be dissociated upon metal deposition.** Whether or not that happens for Liq doped into an ETL of Diekmann, I -- it's hard to say.”

EX2019, 27:4-9; POR 35, 38

Li Has Higher Diffusivity than Cs, Ba & LiF

No Demonstration of Reduced Diffusion

Alleged Motivation

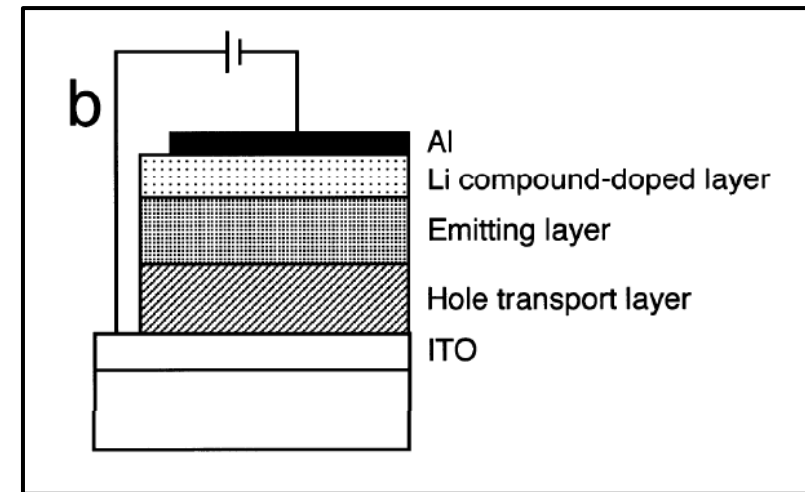
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Pet. 37-38; see also Pet. 69-70

Fact: Free Li Generated When Liq Abuts Cathode

Q. So Endo's theory is *that lithium quinolate is reduced to lithium-free metal*; is that correct?

A. **That's correct.**



EX2032, 49:19-22; Sur-Reply 16

Li Has Higher Diffusivity than Cs, Ba & LiF

Lithium Can Quench Triplet States

Dr. Holmes
Expert Witness

SAMSUNG

“lithium could potentially act as a source for, basically, excitons to transfer their energy to, and that energy is released non-radiatively, so they're quenching sites”

EX2020, 316:16-19, POR 40

Later Studies Confirmed Triplet Quenching Concern with Liq-Doped ETLs

High triplet energy n-type dopants for high efficiency in phosphorescent organic light-emitting diodes

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ABSTRACT

High triplet energy n-type dopants, lithium 2-(oxazol-2-yl)phenolate (LiOx) and lithium 2-(1-methyl-imidazol-2-yl)phenolate (Lilm), were synthesized as n-type doping materials for phosphorescent organic light-emitting diodes and the effect of the n-type doping materials on the electron mobility and device performances of the phosphorescent organic light-emitting diodes was investigated. The LiOx and Lilm n-type dopants were effective to increase the electron mobility of electron transport materials and improve the quantum efficiency of green and blue phosphorescent organic light-emitting diodes.

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voltage [13–16]. The organometallic Liq dopant increased the mobility and conductivity of electron transport materials, resulting in low driving voltage in OLEDs. However, the Liq dopant can act as a luminance quencher in phosphorescent organic light-emitting diodes (PHOLEDs) because of low triplet energy of Liq (2.0 eV) although there has been no report about the triplet exciton quenching effect by Liq. In those works listed above, the Liq dopant was applied as the dopant for electron transport layer of fluorescent OLEDs. Alternatively, exciton blocking layer was inserted between the n-doped layer and emitting layer to block triplet exciton quenching of emitting materials. However, this approach increases the driving voltage of the device due to low electron mobility of the non-doped exciton blocking layer. Therefore, it is strongly required to develop high triplet energy n-type doping materials to suppress the triplet exciton quenching by the n-type doping material.

EX2005 (Yr. 2015) at 1-2; POR 34-35, 39

PHOLEDs Far More Concerned with Quenching of Triplet States than Fluorescent OLEDs

Dr. Holmes

Expert Witness

SAMSUNG

- Q. So when you say lithium diffusion especially -- is problematic especially for phosphorescent OLEDs, why do you say that?
- A. So, again, what I had said was, in devices where you're trying to get very high efficiency, which is what you're doing in a phosphorescent OLED, and so, when people make *phosphorescent OLEDs* in the community, *these are the most efficient devices that we know how to make. And so, as a result, you're trying to move every possible loss in order to get that high efficiency.*

Fluorescent OLEDs frequently are already plagued by low efficiency because they can't emit out of the triplet state and the triplet state's a loss, which means there's already several factors that are preventing high efficiency before you start to consider these finer details.

EX2020, 315:4-316:2 (objections omitted), POR 40

Dr. Holmes Failed to Make the Proper Comparison

Dr. Holmes
Expert Witness

SAMSUNG

Q. What about Lithium fluoride. What is its diffusion coefficient compared to Lithium Q?

A. Again, I haven't looked at that and so I don't know.

EX2019, 182:15-19 (objections omitted), POR 32

Dr. Holmes Failed to Make the Proper Comparison

Dr. Holmes
Expert Witness

SAMSUNG

Q. And do you know what's the relative device life due to Lithium diffusions are for those two systems?

A. So again, not as a comment on stability aside from saying that improved charge balance would be expected to improve stability. But **in terms of the actual measurement of diffusion, that's not included.**

EX2019, 141:7-14; POR 39

Dr. Holmes Does Not Have the Information Needed to Make a Proper Comparison

Dr. Holmes
Expert Witness

SAMSUNG

Q. Okay. You don't know the percentage of disassociation of lithium fluoride?

A. No.

Q. Okay. And you don't know for cesium and barium what is the typical loading, correct?

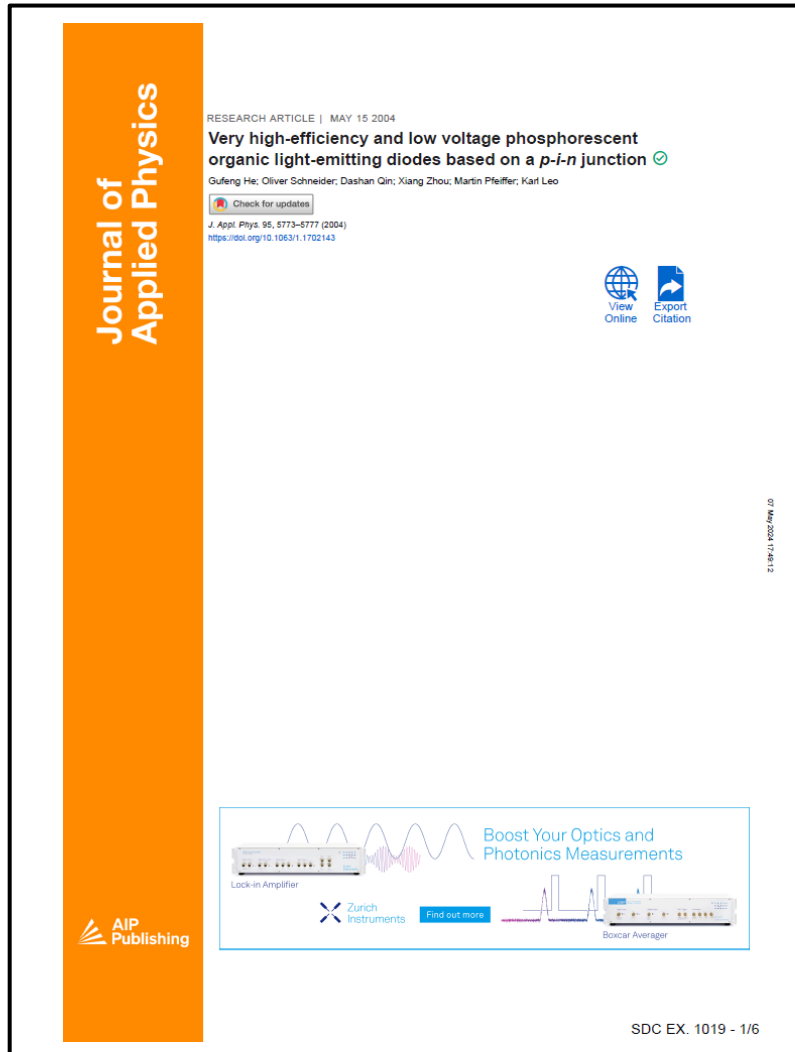
A. No, I don't have a typical loading for cesium or barium.

Q. Do you know any range of loading that might happen?

A. No, if you look in the exhibits I cite, in fact, they don't specify their loading.

EX2032, 184:6-20 (objections omitted)

Blocking Layer Addresses Quenching But Could Lead to Significant Reduction in OLED Device Performance



The image shows the front cover of the Journal of Applied Physics. On the left is an orange vertical bar with the journal title 'Journal of Applied Physics' and the AIP Publishing logo. The main white area contains the article title 'Very high-efficiency and low voltage phosphorescent organic light-emitting diodes based on a *p-i-n* junction', authors 'Gufeng He; Oliver Schneider; Dashan Qin; Xiang Zhou; Martin Pfeiffer; Karl Leo', and publication details 'J. Appl. Phys. 95, 5773-5777 (2004)'. It also includes a 'Check for updates' button, 'View Online' and 'Export Citation' links, and a date stamp '07 May 2004 17:48:12'. At the bottom, there is an advertisement for Zurich Instruments featuring a 'Lock-In Amplifier' and a 'Boxcar Averager' with the text 'Boost Your Optics and Photonics Measurements' and 'Find out more'.

Even as compared to Pfeiffer's previous study on phosphorescent *p-i-n* OLEDs, our latest results show further significant improvement in device performance. Apart from optimized preparation conditions and high material purity, another advantage of our *p-i-n* structure is that we use Cs instead of Li as the dopant for *n* doping. Compared to Li atoms, Cs atoms have larger size, which reduces the probability of Cs diffusing into the emission layer to quench the excitons. Using Li, undoped buffer layers have to be at least 25 nm thick to obtain the full efficiency. Using Cs, however, 10 nm of undoped Bphen between the emission layer and the doped transport layer are enough to avoid any quenching. On the other hand, we find that, if Cs is used, the driving voltage of the OLEDs rapidly increases with the thickness of the undoped buffer layer which is another hint that Cs hardly penetrates into the nominally undoped region.

EX1019 at 5775; Sur-Reply 11

No Demonstration that Ma's OLEDs Would Still Show Improved Performance with a Blocking Layer

No Reason to Use Liq with a Different Mechanism if Diffusion Already a Solved Problem

Alleged Motivation

Moreover, Diekmann teaches that the n-dopant for its electron transport layer should be “suitable for emitting electrons” and “preferably has a low diffusion constant within the organic layer stack, as is usually the case with large atoms or molecules, for example.” Diekmann, 10. Liq was known to have good electron injection and transport properties and has a larger molecular weight than Diekmann’s preferred n-dopants (cesium, barium, and lithium fluoride). See Ex.1003, ¶¶144-145; see also Ex.1031, 4495; Ex.1018. A POSITA thus would have recognized that Liq would be an advantageous dopant for Diekmann’s doped layer. Ex.1003, ¶144.

Pet. 37-38; see also Pet. 69-70

Fact: Diffusion Allegedly Already Addressed with a Blocking Layer

Q. So *Lithium diffusion* is not a concern?

A. So that's not quite the same question. You were asking is Lithium dissociation a concern in phosphorescent OLEDs. Bphen actually does note the potential issue associated with dopant diffusion, which is a general undesirable feature in an OLED with doping that needs to be certainly considered when designing the device. It's something that could be an issue for both fluorescence and phosphorescent OLEDs.

But if you look at the prior art that I cite extensively in my declaration, that *was already a solved issue at the time of the '425[.] work* by He and Pfeiffer and even work at Novaled had shown that the blocking layer that's typically included in a phosphorescent OLED allows the doped layer to be spaced from the emissive layer and reducing the concern related to dopant diffusion.

EX2019, 28:18-29:14 (objections omitted); POR 13, 40

Challenges Against Claim 4 Fail

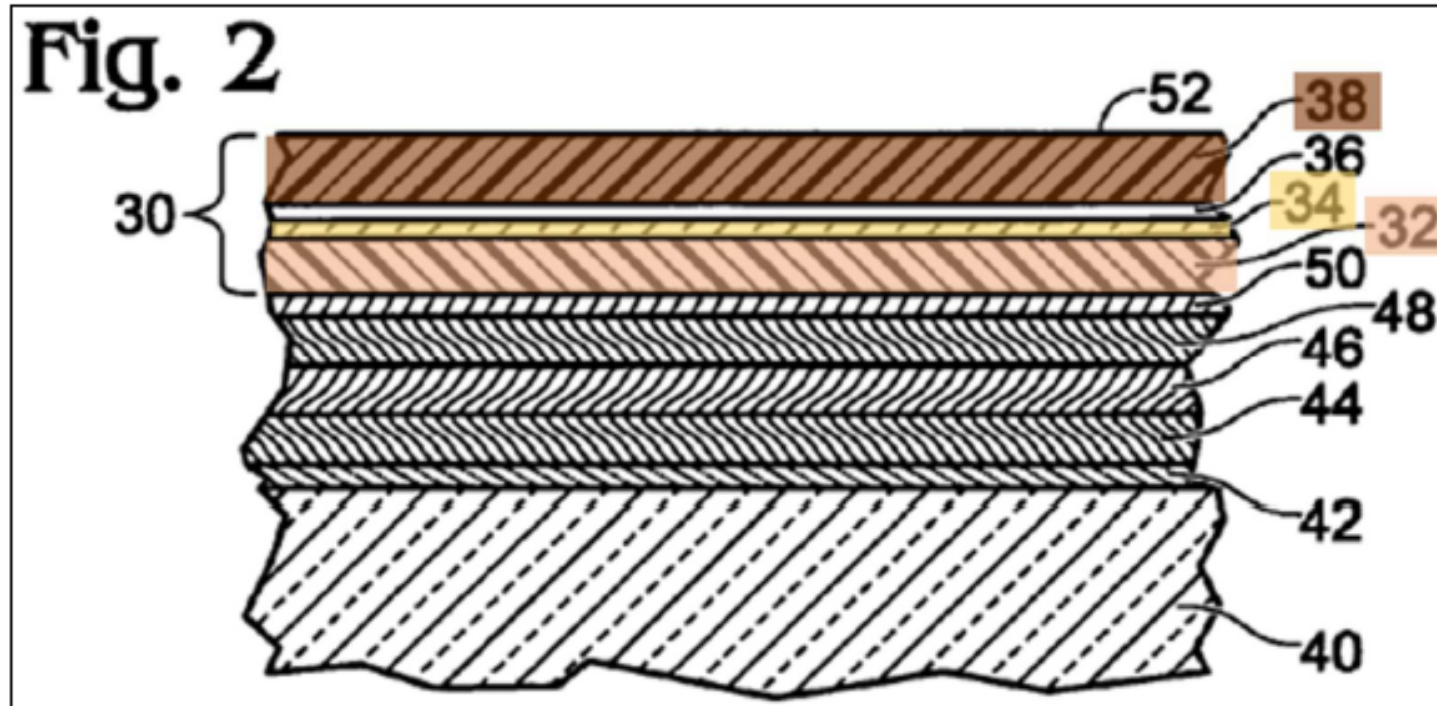
Claim 1

1. A device comprising:
an organic light-emitting diode comprising:
an organic layer sequence,
a radiation exit area, and
an encapsulation,
wherein the organic layer sequence and the encapsulation are disposed on a carrier,
wherein the organic layer sequence comprises at least one radiation-emitting region configured to generate electromagnetic radiation in a spectral range from infrared radiation to UV radiation during operation,
wherein the organic layer sequence comprises an electron transport layer that is n-doped; and
wherein the organic layer sequence comprises an emission layer comprising an iridium-containing compound.

Claim 4

4. The device of claim 1,
wherein the encapsulation comprises a first encapsulation layer and a second encapsulation layer,
the first and the second encapsulation layer each having a respective volume structure,
wherein the volume structure of the second encapsulation layer is independent of the volume structure of the first encapsulation layer, and
the volume structure of the second encapsulation layer has a higher amorphicity than the volume structure of the first encapsulation layer.

Lee Is Not Directed to an n-Doped PHOLED



Pet. 39; EX1008 (Lee), Fig. 2

- 32: 1st semi-crystalline parylene-based polymer layer
- 34: inorganic barrier layer
- 36: reflective layer
- 38: 2nd semi-crystalline parylene-based polymer

- No mentioning of n-doped ETLs
- No mentioning of PHOLEDs
- No demonstration that annealing was compatible with e.g., n-doped PHOLEDs

Lee Is Not Directed to an n-Doped PHOLED

[0033] Some of the organic materials used as electron transport layers, hole transport layers, and/or light emitting layers in OLEDs may not be thermally stable at the desired annealing temperatures. For example, some commonly used OLED organic materials are unstable at temperatures as low as 60° C. To protect these layers during an annealing process, more directed annealing methods than hotplate- or oven-based annealing methods may be used. For example, in some embodiments, laser or focused IR techniques may be used to anneal the second parylene-based polymer film 38. The use of a laser or focused IR may allow energy to be delivered to the semi-crystalline parylene-based polymer layers with a relatively high degree of specificity, thereby avoiding delivering unwanted energy to the organic layers. Reflector 36 may be used to provide further protection to the organic device layers.

EX1008 (Lee), [0033]

No Demonstration that this Was Sufficient for n-doped PHOLEDs

Challenges Against Claim 12 Fail

Claim 1

1. A device comprising:
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a radiation exit area, and
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wherein the organic layer sequence and the encapsulation are disposed on a carrier,
wherein the organic layer sequence comprises at least one radiation-emitting region configured to generate electromagnetic radiation in a spectral range from infrared radiation to UV radiation during operation,
wherein the organic layer sequence comprises an electron transport layer that is n-doped; and
wherein the organic layer sequence comprises an emission layer comprising an iridium-containing compound.

Claim 12

12. The device according to claim 1,
wherein the device is drivable by means of a gesture control.

Dr. Holmes Relies on Solid State White Lighting Device for Element 1[f]

Dr. Holmes
Expert Witness

SAMSUNG

104. Diekmann further explains “[t]he organic layer stack 4 can, for example, comprise the layers of the organic layer stack of Figure 1” Diekmann, 92, which includes as radiation-emitting regions “a SEB-010:SEB020 layer . . . a TMM-004:Ir(ppy)3(15%) layer . . . and a TMM-04:TER012 layer,” Diekmann, 91.

Diekmann discloses that “[t]his organic layer stack is suitable for emitting white light.” Diekmann, 91. Thus, a POSITA would recognize that these emissive layers are configured to generate electromagnetic radiation in a spectral range from infrared radiation to UV radiation during operation.

EX1003, para. 104

- **No Reason to Have Gesture Control for Solid State Light Devices**
- **Secondary Reference Harada “Teaches a Display Device,” (Pet. 77) Not a Lighting Device**

Dr. Holmes Relies on Solid State White Lighting Device for Element 1[f]

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SAMSUNG

“And further *I think supporting the read that I'm offering of Diekmann is the fact that part of the focus of Diekmann* -- and there's obviously many things Diekmann is doing -- but part of the focus in Diekmann *is for solid state lighting, is a white lighting device and a white lighting device would ideally have spectral components throughout the visible*. Otherwise it would have a low color rendering index and wouldn't be able to reproduce all of the colors that are needed to illuminate an internal space.”

EX2019, 192:6-13; POR 59

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Dr. Holmes Is Not Relying on a Display for Element 1[f]

Dr. Holmes
Expert Witness
SAMSUNG

Q. What do you mean by a white light emission device? So *if you have a display that can render color, like a white color, do you consider that to be a white light-emitting device?*

A. *So, no.* What I'm referring to in this case is an OLED that, when energized, emits white light. So a single device that when energized emits white light. And certainly when OLEDs grew in efficiency and performance, there was additional interest in groveling their application and solid state lighting for illumination, general lighting was a significant potential next application for OLEDs.

And so I'm actually referring to a device where, based on Diekmann and based on the claim language, the device itself is emitting -- or sorry, the OLED itself, pardon me, not the device -- the OLED itself is emitting white light.

So, for example, a three emissive layer that are stacked in a single device.

EX2019, 194:7-195:3

Dr. Holmes Relies on Solid State White Lighting Device for Element 1[f]

Dr. Holmes
Expert Witness

SAMSUNG

Q. When you say within a single OLED, what do you mean by that?

A. So element 1F is saying wherein the organic layer sequence, and so when I say a single OLED, I mean within, you know, Diekmann's embodiment in figure 2A, within three layers, red, green, blue, replacing 401, right? So if you look at figure 2A, the emissive layer 401 would now be Diekmann's emissive layer of SEB-10, SEB-20, TMM-4:Ir(ppy) and TMM-4:TER12, right? So, I mean, a single OLED device as shown in Diekmann figure 2A.

EX2019, 195:12-22

Suzuki's White Light Emission Application Relied on for Claim 1

Suzuki explains that its light emission layer “emits light by recombination of electrons and holes injected from the electrode or the electron transport layer and the hole transport layer...” Suzuki, [0299]. Suzuki also discloses using various “light emission hosts” which “may be used in combination” and explains that “by using a plurality of dopant compounds, it is possible to mix different light emissions, thereby obtaining any desired light emission color.” Suzuki, [0316]-[0319]. For example, “[w]hite light emission is possible by adjusting the type and doping amount of phosphorescent compounds, and can be applied to illumination and backlighting.” *Id.*; Ex.1003, ¶¶192-193.

Pet. 56

No Reason Stated for Adding Gesture Control to Illumination & Back Lighting Devices



As explained above §VIII.E.1, Harada teaches a display device which performs “gesture recognition using a hand shape” by recognizing “the shape of a hand 110 touched (or approached) to a display panel.” *Id.*, [0071]; Ex.1003, ¶¶174-178, 249-253.

A POSITA would have been motivated to incorporate this gesture control functionality in Suzuki’s device to allow a user to operate the device “in more natural ways.” Harada, [0073]; Ex.1003, ¶¶174-177. As explained above, §VIII.E, gesture control input for controlling electronic devices, including OLED displays, was well-known by the time of the ’425 patent. Ex.1003, ¶¶177-178. Moreover, Harada teaches that its gesture control functionality can be applied to OLEDs. Harada, [0003].

Challenges Against Claim 9-11 Fail

Claims 9-11: “touch sensor” Claims

9. The device according to claim 1,
wherein the device comprises at least one touch-sensitive
operating element, and
wherein the at least one touch-sensitive operating element
comprises at least one touch sensor.
10. The device according to claim 9,
wherein the at least one touch sensor is integrated into the
organic light-emitting diode.
11. The device according to claim 10,
wherein the touch-sensitive operating element is arranged
at the radiation exit area of the device.

- **Also No Reason to Add Touch Sensors to Lighting Devices**

Additional Slides

Prior Art Provides Motivation to Use Liq as n-dopant

- Increased current density and reduced driving voltage
 - E.g., Ma at 810, 813
- Improved transport conductivity of the ETL
 - E.g., Ma at 813
- Larger molecular weight likely to result in lower diffusion constant
 - E.g., Diekmann at 10
 - E.g., Ex. 1003 (Holmes Declaration) at ¶144
- Good electron injection and transport properties
 - E.g., Ex. 1003 (Holmes Declaration) at ¶¶35-36, 42, 144

Prior Art Provides Motivation to Use Liq as n-dopant

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 - E.g., Ex. 1003 (Holmes Declaration) at ¶144
 - Good electron injection and transport properties
 - E.g., Ex. 1003 (Holmes Declaration) at ¶¶35-36, 42, 144
- Relative to undoped ETL, not ETL doped with other n-dopants
- Diekmann does not mention Liq;
- Dr. Holmes provides no evidence that the effective diffusion of Liq is lower given Liq's in situ formation of Li
- -no evidence that Liq in Ma improves electron injection