

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.

Case No. IPR2024-01095
Patent No. 11,828,425

**SUPPLEMENTAL DECLARATION OF
DR. P. MORGAN PATTISON, PhD.**

TABLE OF CONTENTS

	<u>Page</u>
I. INTRODUCTION	1
II. A POSITA WOULD NOT HAVE USED LIQ AS AN ETL DOPANT IN DIEKMANN OR SUZUKI.....	2
A. The Role Of Liq in Ma Is Not That of An Electron Donor	4
B. There is No Showing that Liq-Doping Would Have Led to Lower Diffusion of Impurities or Higher Efficiency	11
C. A Skilled Artisan Would Not Have Used Ma's Fluorescent OLED Dopant in the Claimed Ir-based PHOLED Due to Significant Quenching Problems and Traps.....	24
D. A Skilled Artisan Would Not Have Reasonably Expected Success in Doping an Unidentified Host ETL with Liq in a System Without a Specified PHOLED Emitter Matrix	29
III. DIEKMANN AND SUZUKI HAVE NO NEED FOR LEE'S ENCAPSULATION (CLAIM 4).....	33
A. Diekmann and Suzuki Have No Further Need of Encapsulation	36
B. Neither Diekmann or Suzuki Use Polyacrylate or Amorphous Parylene Barrier Layers.....	37
C. Samsung's Expectations of Success are Conclusory	40

1. I, P. Morgan Pattison, PhD, declare as follows:

I. INTRODUCTION

2. I have been retained as an independent expert on behalf of Pictiva Displays International Ltd. (Patent Owner) to provide technical background and opinions related to U.S. Patent No. 11,828,425 (EX1001) (the '425 Patent) at issue in in IPR2024-01095.

3. I previously provided a declaration regarding Petitioner Samsung Display Co., Ltd.'s position regarding Grounds II and VII (Pet. 36-38, 69-70) involving claims 2 and 17 and other matters addressed herein. EX2002.

4. I provide this supplemental declaration to address additional opinions in this case and also incorporate EX2002 herein.

5. For this service, I am compensated for my time at my normal consulting rate. My compensation does not depend on the outcome of any matter or on any of the opinions provided below. I have no financial interest in this matter.

6. My testimony herein is based on my technical background, my years of experience in the field of organic light emitting diodes (OLED) and my review of the materials cited herein.

7. My qualifications and experience as well as my curriculum vitae (CV) are detailed in my prior declaration. EX2002, § II, Ex. A.

8. I also apply the same legal standards as previously described in my prior declaration. EX2002, § III.A.

9. In forming my opinions expressed in this declaration, I have again relied on my own knowledge, experience, and expertise, as well as the knowledge of a POSITA in the relevant timeframe. In addition, I have reviewed and relied upon the materials cited in this declaration and my prior declaration. *See* EX2002.

II. A POSITA WOULD NOT HAVE USED LIQ AS AN ETL DOPANT IN DIEKMANN OR SUZUKI

10. Given the early stages of this research into Liq at the time of filing the '425 Patent, a skilled artisan armed with only a bachelor's degree in physics or chemistry and three years of experience would not have been motivated to and could not reasonably expect to just swap materials or dopants into an entirely different stack with different host and emissive materials with a reasonable expectation of success, as I previously explained. EX2002, ¶¶ 86, 114, 116, 122; *see id.*, ¶¶ 69, 71, 78, 85. Such efforts would require significant experimentation and a large team of cross-disciplinary scientists with far more advanced experience to even begin to accomplish. *Id.*

11. There are significant difficulties in using unproven materials in different OLED stacks. The choice of which layers to use, the specific materials for each layer, the dopant and its concentration, if any, for each layer, the size of each layer, the layer order, and the choice of manufacturing processes (deposition, co-

evaporation, temperature, and so forth) must be carefully selected to create an operational, efficient, and reliable OLED. EX2002, ¶¶ 123, 138; EX2021 (Yang 2024) at 1 (“optimizing excitons generation and minimizing their loss are essential” to OLED design and “necessitates precise control of polarons—the source of excitons—to ensure their efficient and balanced transport, injection, and confinement within the EML while mitigating their involvement in excitons loss mechanisms”). Otherwise, there would be no “light emitting device” to qualify as the claimed OLED device that emits EM radiation in the claimed range. EX2002, ¶¶ 123, 138; *see id.*, ¶¶ 35-36.

12. In my prior declaration, I explained why a POSITA would not have used 8-hydroxyquinolinolato-lithium (“Liq” or lithium quinolate) as a dopant in either Diekmann’s or Suzuki’s electron transport layers based on Ma (EX1010) at the time of filing the ’425 Patent. EX2002, § VI.

13. In that prior discussion, I explained why:

- Samsung and Dr. Holmes provided no reason for why a POSITA would use Liq in Diekmann’s or Suzuki’s ETL with a reasonable expectation of success. EX2002, § VI.A.
- Ma does not provide a motivation to dope every ETL with Liq. EX2002, § VI.B.

- The use of Liq as an ETL dopant was experimental for limited use in stable, fluorescent OLED devices with Bphen at the time. EX2002, § VI.C.
- Samsung and Dr. Holmes do not account for the differences between Ma's Alq₃-based fluorescent OLED system and the claimed iridium-based phosphorescent OLED. EX2002, § VI.D.
- Samsung and Dr. Holmes do not address the delicate energy-level balance required for the different organic layers in an OLED stack. EX2002, § VI.E.
- Samsung and Dr. Holmes offer no evidence that Diekmann and Suzuki would have gained any benefit from using an Liq-doped ETL. EX2002, § VI.F

14. I provide the following additional opinions related to that testimony.

A. The Role Of Liq In Ma Is Not That Of An Electron Donor

15. For element 1[g], which recites “wherein the organic layer sequence comprises an electron transport layer that is n-doped,” both Samsung and Dr. Holmes point to Diekmann’s disclosure that it “has a doped layer 402 with a dopant 410, which acts as an electron donor.” Pet. 24-25; EX1003, ¶¶ 106, 109; Diekmann, 91. The preferred dopants include “cesium, barium, or lithium fluoride.” *Id.*

16. They further point to Diekmann's disclosure that layer 402 "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.'" Pet. 24; EX1003, ¶ 106 (citing Diekmann 10 that provides "doping advantageously increases the conductivity of the doped organic layer and, in the case of an n-doped layer adjacent to the cathode, leads to better injection of electrons from the cathode into the organic layer stack"); Diekmann, 91 (disclosing that doping with an electron donor "advantageously increases the injection of electrons from the cathode into the organic layer").

17. Diekmann's description that n-dopants in organic semiconductors would be understood as electron donors comports with a skilled artisan's understanding of n-type doping at the time of the invention. For example, Walzer and co-authors described in a 2007 Chemical Reviews article:

The basic principles of doping in organic semiconductors are similar to those in inorganic materials: Mobile carriers are generated by electron donors or acceptors. In organics, one has to add *constituents*, which either *donate electrons to the lowest unoccupied orbital (LUMO) states (n-type doping)* or remove electrons from the highest occupied orbital (HOMO) states to generate holes (p-type doping) (Figure 4).

EX2101 [2007 Chem Review Walzer], 1236, 1238.

18. Likewise, when Suzuki mentions “an electron transport layer with a high n nature doped with impurities” and gives as an example of an OLED described in “J. Appl. Phys., 95 5773(2004)” (that is the He paper in EX1019),¹ a skilled artisan would understand that, to the extent Suzuki is referencing an n-doped electron transport layer, that n-dopant—such as cesium in He—serves as an electron donor. *See* EX2101 at 1245 (“Similar to lithium, cesium (Cs) acts as an efficient electron donor in organic films.”). Like Li, when Cs is used to dope an organic material such as BPhen, it may help to lower the electron injection barrier. *Cf.*, EX2101, Fig. 5 (showing that upon doping of another alkali metal, Li, the Fermi level in BCP shifts upward, which narrows the gap between the Fermi-level and the LUMO of BCP, making electron injection into the LUMO level more efficient and hence a higher electron density).

¹ Institution Decision at 19-20 (citing EX1019, Abstract) (“... 4,7-diphenyl-1,10-phenanthroline *and cesium* are co-evaporated *as a n-doped electron transport layer...*”).

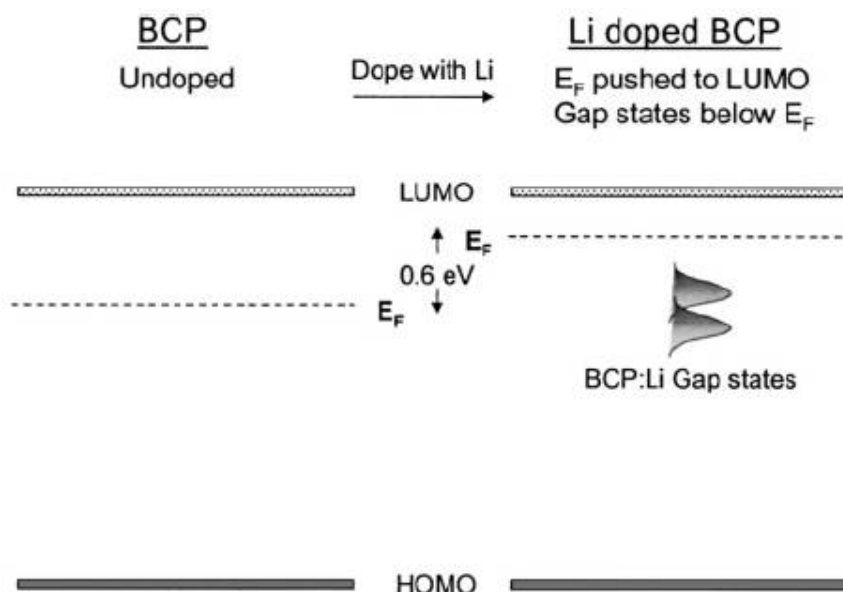


Figure 5. Proposed energy level scheme of undoped (left) and Li-doped (right) BCP thin films. The lowest unoccupied and highest occupied molecular orbitals are labeled LUMO and HOMO, respectively. Reused with permission from G. Parthasarathy, C. Shen, A. Kahn, and S. R. Forrest, *Journal of Applied Physics*, 89, 4986 (2001). Copyright 2001, American Institute of Physics.

EX2101 at 1236, 1238, Fig. 5.

19. Thus, in both Diekmann and Suzuki, n-dopants, if any, are electron donors. In contrast, there is no indication that Liq in Ma serves acts as an electron donor.

20. When Ma describes the implication of J-V characteristics of the electron-only devices, it does not describe the observed trend in terms of increased electron densities, only in terms of “short hopping length for electron transport, compared to the pure BPhen [E]TL.” Ma at 813.

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.

21. Consequently, a skilled artisan would understand that in Ma, Liq helps improve conductivity by modulating the mixed layer's electron mobility, but not by affecting its electron density. They would therefore not view Liq as described in Ma as serving an “electron donor” that is explicitly called out in Diekmann or implicitly in He reference by Suzuki. In contrast, Ma describes its p-doping as involving a charge transfer from the HOMO of the matrix material to the LUMO of the p-dopant. Ma at 812-813. Ma's description of its p-doping mechanism indicates that Ma is aware of the charge-transfer mechanism of doping, and that it is not an accident that Ma describes Liq's role only in terms of improving electron mobility, but not in terms of charge transfer or increased electron density.

22. I am asked to comment on Dr. Holmes' characterization of Liq as a molecular dopant. The use of Liq by Ma is as an alloy to improve the mobility of the electron transport layer but not to increase the concentration of charge carriers. This differs from n-doping by molecular dopants having high-lying HOMOs, such as the "proprietary n-dopant released by Novaled AG." *See* EX2101 [2007 Chem. Review], 1245-1246 (listing "proprietary n-dopant released by Novaled AG" under the heading "n-Type Doping by Organic Materials with High-Lying HOMO"); EX2100 [Novaled US7,911,129], 10:56-63 (specifying that the electron affinity of a matrix material to be n-doped should be greater in value than the ionization potential of a molecular n-dopant minus 0.5 eV).

23. I am also asked to comment on Dr. Holmes' view that "[a]n n-type dopant is an atomic or molecular species that when added to the ETL increases the conductivity for electrons in the layer." EX1003, ¶ 40. Dr. Holmes' definition encompasses dopants that affect both carrier concentration and electron mobility. While conductivity of a layer is the product of carrier concentration and mobility, Dr. Holmes definition does not seem to comport with the understanding and context of n-doping at the time of the invention. For example, as I note above, Walzer describes n-type doping as involving electron donation to the LUMO of the matrix material. EX2101 at 1236 ("The basic principles of doping in organic semiconductors are similar to those in inorganic materials: Mobile carriers are

generated by electron donors or acceptors. In organics, one has to add constituents, which either donate electrons to the lowest unoccupied orbital (LUMO) states (n-type doping) or remove electrons from the highest occupied orbital (HOMO) states to generate holes (p-type doping) (Figure 4).”).

24. But even if n-doping encompasses any dopants that increase electron conductivity of a doped layer compared to an undoped layer, Diekmann and Suzuki used the electron-donor type n-dopants, which also helped increase the injection of electrons from the cathode into the organic layer stack. These are the characteristics and operating principles of n-dopants used by Diekmann and Suzuki. Ma’s Liq, as explained above, has neither characteristics.

25. Hence, to the extent that a skilled artisan would have explored other n-dopants in Diekmann or Suzuki, they would not have chosen Liq in light of Ma because Ma’s Liq does not operate in the same way as the electron-donor type n-dopants in Diekmann and Suzuki. Rather, the use of Liq as described in Ma would alter Diekmann and Suzuki’s operating principle, which is to modulate the electron conductivity through the use of an electron donor, which may, for example, reduce the electron injection barrier. Diekmann, 91. Liq as described in Ma does not appear to have those properties.

26. That a skilled artisan would not have used Liq in place of other preferred n-dopants in Diekmann and Suzuki is reinforced by Samsung’s assertion

that “[a] POSITA would have been motivated to include an n-doped electron transport layer *to promote injection of electrons from the cathode and* improve transportation of electrons to the light-emitting region.” Pet. 25. Nothing indicates that in Ma’s mixed BPhen:Liq ETL layer, Liq helped reduce injection barrier to the ETL layer or otherwise promoted electron injection into the ETL layer.

27. Thus, it is my opinion that a skilled artisan would not have abandoned the proven n-doping mechanism of Diekmann and Suzuki—electron donor n-dopants at significantly lower concentrations (e.g., cesium, barium, and LiF)—for a heavy Liq dopant that appears to affect conductivity through a different mechanism (that is, affecting hopping length and electron mobility). *See* Ma 813.

28. As I will explain further below, this lack of motivation is further reinforced by other significant issues with Liq at causing carrier imbalance, charge carrier traps, and quenching in PHOLEDs discussed below and in my prior declaration.

B. There is No Showing that Liq-Doping Would Have Led to Lower Diffusion of Impurities or Higher Efficiency

29. At his deposition, Dr. Holmes stated two reasons for using Liq: (1) “implementation of Liq into their device so that they could take advantage of the reduced diffusion that would come with the molecular n-dopant,” and (2) “the device operates with potentially higher efficiency and lower voltage.”

EX2019_2025-04-24_Holmes, 238:10-11 (discussing EX1003, ¶ 145), 240:1-

241:4. But Samsung and Dr. Holmes did not present any evidence on the relative performance of a PHOLED with a Liq-doped ETL versus one with an ETL doped with Cs, Ba or LiF. For example, I have not seen any evidence on (1) how the diffusion in a Liq-doped PHOLED compares to PHOLED devices with cesium, barium, or LiF-doped ETLs (especially after both having been optimized); (2) how their efficiency and driving voltages compare.

30. At his deposition, Dr. Holmes admitted that he did not investigate these relevant issues despite prior reports of potential Liq dissociation:

Q. Have you done any testing to see whether Liq offers reduced diffusion?

A. I have not --

Q. Compared to cesium or Lithium, for that matter.

A. I have not --

MR. WEIDENFELLER: Objection.

THE WITNESS: I have not done any testing myself. But Diekmann acknowledges the challenge and acknowledges molecular dopants as an alternative to elemental metals, and a person skilled in the art would recognize Liq as a molecular dopant that would fit that bill.

Q. But we've also established that at least based on Liu, when aluminum is deposited on Lithium Q a neat film, there is some Lithium dissociation or Lithium generation happening, correct?

A. That is what Liu teaches for a neat film of Liq under a metal cathode. But even in the case where some Lithium were generated, as a person of skill in the art -- a person skilled in the art would look and expect that there would be potentially less Lithium generated than strictly Lithium metal going in as an n-dopant. And so I think even still there'd be an expectation for success.

Q. But with the reduced amount of Lithium -- let's take one step back.

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?

MR. WEIDENFELLER: Objection.

THE WITNESS: 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.

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

MR. WEIDENFELLER: Objection.

THE WITNESS: Again, I haven't looked at that and so I don't know.

Q. Do you know whether it's higher or lower?

A. No, I don't. As I've already said, I don't know what the diffusion coefficient is for Liq and I separately don't know what the diffusion coefficient is for Lithium fluoride, so I'm not able to answer that question

EX2019 [2025-04-24 Holmes Tr.] at 139:10-141:5, 182:15-183:4.

31. As I noted above, should Liq operate as a molecular n-dopant, given its HOMO energy level, a skilled artisan would not have viewed it as a suitable one for OLEDs. But should Liq somehow dissociate,² it would be important to investigate the resulting diffusion and performance compared to, not ETLs doped with lithium metal, but to ETLs doped with preferred dopants Cs, Ba and LiF. Dr. Holmes did not conduct the relevant analysis and therefore has not shown that Liq-doping would have led to lower diffusion or higher efficiency as he speculates.

32. In that regard, I also notes that Dr. Holmes testified that dopant diffusion “was already a solved issue at the time of the ‘425 work by He [EX1019] and Pfeiffer [EX106] and even work at Novaled [EX1029] had shown that the blocking layer that’s typically included in a phosphorescent OLED allows the

² EX2023 [Lee 2008, J. Indus. & Eng’g Chem. 14 (2008) 676-678] at 677, § 3 (“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”).

doped layer to be spaced from the emissive layer and reducing the concern related to dopant diffusion.” EX2019 [2025-04-24 Holmes Tr.] at 29:7-14. If dopant diffusion ‘was already a solved issue’ at the time of the invention, that would beg the question of why a skilled artisan would have attempted to substitute preferred n-dopants (Cs, Ba and LiF) with Liq which operated using a different mechanism and whose behavior and performance were less understood.

33. Additionally, to the extent that Dr. Holmes relies on an OLED device with a blocking layer (such as the one between layers 401 and 402 in Diekmann’s Figure 2A), there is no evidence of the relative performance or diffusion when layer 402 is a Liq-doped layer versus when it is a Cs-, Ba- or LiF doped layer. EX2019 [2025-04-24 Holmes Tr.] at 141:1-5 (“I think I made 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.”). Without such comparison, however, a skilled artisan would not be motivated to trade the unknown Liq for the well-studied and preferred n-dopants such as cesium, barium or LiF.

34. I further note that Liq does not necessarily lead to improvement in performance. For example, in 2022, Sasaki and co-workers examined the relative performance of Device-3 and Device-5, differing only in that Device-5 has a Liq-

doped ETL layer and Device-3 has an undoped ETL layer. *See* EX2105, Figure S1.

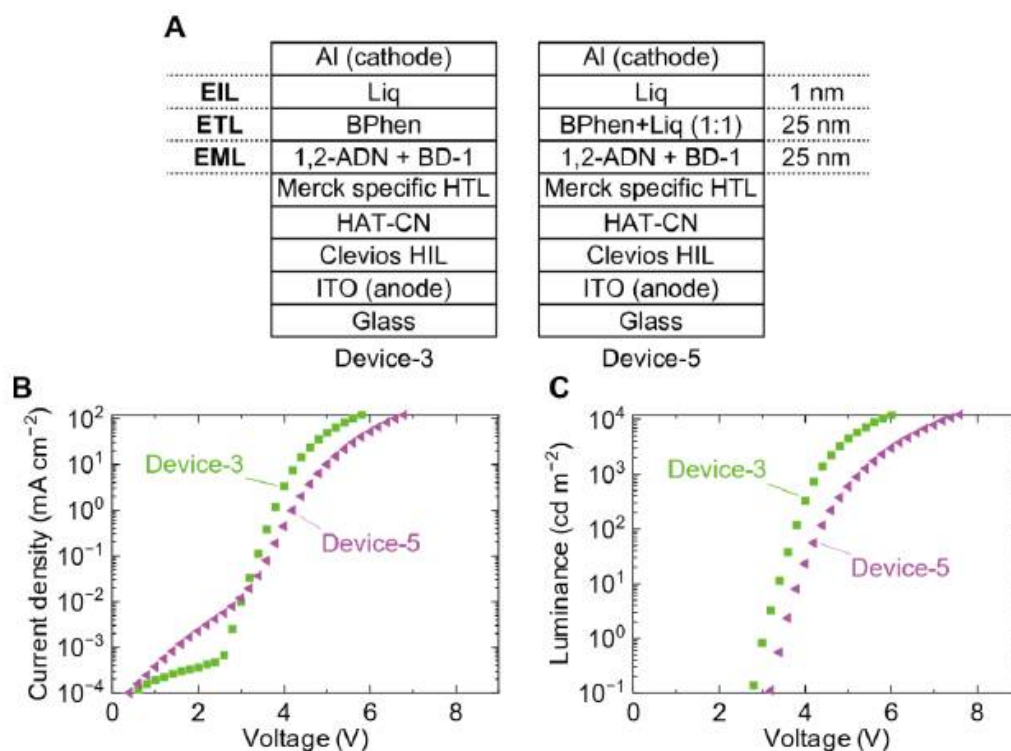


Figure S1. A) Multilayer structure of blue OLEDs employing BPhen and Liq-doped BPhen (50 wt%) as the ETL, and employing Liq as the EIL. B) Current density–voltage characteristics of blue OLEDs. C) Luminance–voltage characteristics of blue OLEDs.

35. Sasaki stated that “[a]lthough we fabricated a blue OLED [Device-5] with Liq used as the EIL and BPhen doped with Liq used as the ETL, its operating voltage was *much higher than that of Device-3* (Figure S1, Supporting Information).”). EX2007, 2201925-2. The authors further noted, 14 years after Ma’s report, “OLEDs fabricated using BPhen doped with Liq as the ETL have rarely been reported.” *Id.*

36. In another example recent example by Angel and co-workers, they also examined the behavior of OLED devices with BPhen:Liq doped ETL. EX2103. In particular, they examined a series of devices with the same HTL (NPB), EML (Alq₃), and ETL (BPhen:Liq) materials as Ma. But as shown below, the addition of Liq resulted in slightly decreased external quantum efficiencies (EQE) and slightly increased driving voltages (V), especially when there is not a separate Liq-injection layer. EX2103, Fig. 2 (Figure A on the left is without a Liq injection layer, and Figure B on the right has a Liq injection layer). In fact, the results below would suggest that it would be more advantageous to use Liq as an injection layer than a dopant in ETL: with Liq serving as an injection layer, the driving voltage dropped from 8V to 6V, and the EQE increased from 7% to 8%.

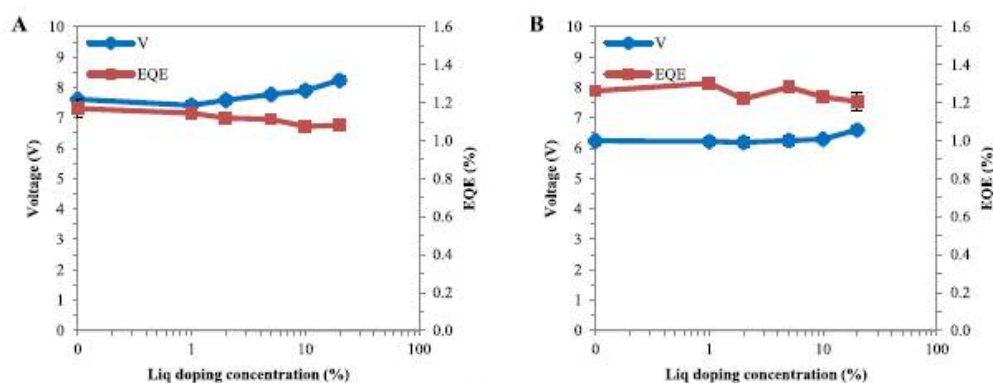


Fig. 2. Drive voltage and EQE dependence on Liq doping concentration for devices: (C) ITO|MoO_x (20 Å)|NPB(600 Å)|Alq₃ (400 Å)|Bphen:Liq(100 Å:x%)|Ag, and (D) ITO|MoO_x (20 Å)|NPB(600 Å)|Alq₃ (400 Å)|Bphen:Liq(100 Å:x%)|Liq (20 Å)|Ag. The voltage and EQE were measured at 20 mA/cm².

37. In a third example, Ki and co-authors also observed that with addition of Liq to Bphen, the driving voltage increased somewhat and the luminance as well as EQE (Figure B inset) decreased. EX2104, Figures 4(a)-(b). In the figures

below, the lifetime did show significant improvement with Liq, which allegedly was due to increased glass transition temperatures of the mixed ETL compared to pure BPhen films. *Id.* at 1681 (“the improved thermal and morphological stabilities of mETL resulted in the better thermal stability and the prolonged operational lifetime of blue devices.”).

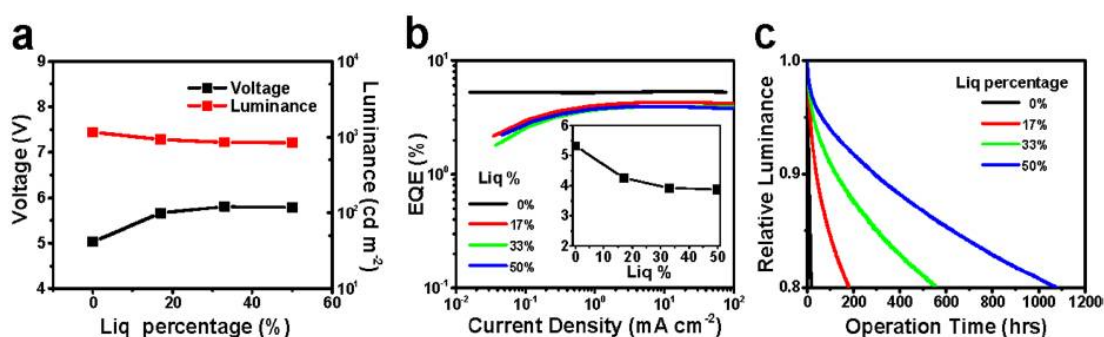


Figure 4. (a) Driving voltages, luminance, and (inset of panel (b)) external quantum efficiency (EQE) of the blue devices upon mixing percentage of Liq in the mETL under a constant current density of 25 mA cm⁻². (b) External quantum efficiency (EQE) versus current density for the devices upon mixing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

38. Ma, however, never mentioned this aspect of the improvement, and there is no evidence that a skilled artisan would have known about this benefit. For example, in 2011, Kumar and co-workers attempted to study the reason for the enhanced electron transport properties in Liq-doped BPhen and Alq₃. EX2004 at 114511-1 to 114511-2. Kumar selected Liq-doped Alq₃ for further study because “Bphen has very low glass transition temperature ..., which causes issues related to device lifetimes.” *Id.*, 114511-2. Kumar’s reason for selecting Alq₃ over BPhen suggests that even as of 2011, skilled artisans were not aware that Liq-doping

would have induced morphological change in BPhen to increase its glass transition temperature and improve the resulting devices' life time.

39. In addition to increased driving voltages that led to reduced power efficiency,³ Liq also causes “low efficiency” through “obstruct[ing] ... electron movement” due to charge carrier traps and “reduc[ing] carrier transport” because of its insulating properties with “a high dielectric constant.” EX2024 (Kim 2013) at 7999-8000; EX2025 (Le 2024) at 69636 (describing degradation due to unstable molecules, annihilation, instability of cations and anions, and traps as well as avoiding quenching); EX2003 (Kim 2011) at 54 (“low efficiency” of Liq); EX2006

³ Dr. Holmes justified n-doping by arguing that n-doping would have led to reduced driving voltages and improved power efficiency. EX1003, ¶¶ 110 (“A POSITA would have known that n-doping the electron transport layer would have clear benefits, including promoting injection of electrons from the cathode and improving transportation of electrons to the light-emitting region. Diekmann, 10. A POSITA would have understood that this would also lead to improved device characteristics, such as lower driving voltage and better power efficiency.”), 142 (“Ma explicitly teaches that an the current and power efficiency of an OLED can be increased at a low driving voltage by utilizing an ETL which is doped with Liq.”).

(Kang 2022), Abstract, 6 (Liq doping could “retard[]” and “weaken” charge transport); EX2103, Abstract (“Liq is a relatively poor electron-transport material”).

40. Additionally, a skilled artisan would understand that Ma involved a fluorescent OLED with a single EML layer consisting of a single material, Alq3. In contrast, under Dr. Holmes’s theory and mapping, the Diekmann and Suzuki OLEDs each include three emissive layers, each layer with a guest-host structure and at least one of the emissive layers includes a phosphorescent iridium-based emitter. EX1003, ¶¶ 104, 194.

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.

transport layer and the hole transport layer...” Suzuki, [0299]. Suzuki further discloses using various different “light emission hosts” which Suzuki teaches “may be used in combination.” Suzuki, [0316]–[0319]. Suzuki further 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, Suzuki teaches that “[w]hite light emission is possible by adjusting the type and doping amount of phosphorescent compounds, and can be applied to illumination and backlighting.” Suzuki, [0316]–[0319]. A POSITA would recognize that emitting of lights of various colors, as disclosed by Suzuki, entails generating electromagnetic radiation in a spectral range from infrared radiation to UV radiation.

See EX2019 [2025-04-24 Holmes Tr.], 200:6-9 (“Q: You are inferring that there are multiple emissive layers because it is white light-emitting; is that right? A: That’s correct.”).

41. Ma emphasizes the importance of charge balance in the emissive layer. *See* Ma, Figure 6 (attempting to find p-dopant loading and n-dopant loading that would lead to similar hole conductivity and electron conductivity). Charge balances would differ for different emission layers, because of the differences in electron and hole transport capabilities of the host materials. There is no showing by Dr. Holmes or Samsung that charge balances could be achieved in all three emissive layers with an Liq-doped ETL. For example, experimental device 1 of

Kim 2011 [EX2003], which like Ma does not have a separate blocking layer, achieved a best efficiency at 5 wt% Liq loading. EX2003 at 58. In contrast, Ma's best efficiency was at 33 wt%. Ma, Fig. 3. And with a blocking layer in Kim 2011, the best efficiency was achieved at 65% Liq; but in Kim 2013, the performance at 5 wt% Liq was better than at 10 wt % Liq. EX2003 at 59; EX2024, 7999 (Device A is without Liq in ETL, and Devices B-D have respectively 3, 5 and 10% Liq in ETL), 8001, Table 1 (Device C with 5% Liq showed the highest luminance efficiency, power efficiency, EQE and lifetime). Thus, depending on the host materials for the emissive layers and the thicknesses of any blocking layer, the optimal loading for Liq would differ. It's not clear how far apart the suitable ranges of doping levels are for the three layers. There is also no information on whether, if compromises were possible to choose a loading level that would provide suitable performance to all three emissive layers, the Liq-doped OLED would have provided better performance or lower diffusion than an OLED with an ETL doped with preferred dopants such as Cs, Ba or LiF.

42. Additionally, BPhen is not a preferred ETL material in Diekmann. For PHOLEDs, Diekmann's Alq₃ and BAlq may not be suitable because their triplet state energies may be too low to confine the triplet excitons of the phosphorescent emitter. *See* EX2026 (Tanaka 2008) at 695 (the lowest triplet energy states of Alq₃ and BAlq is about 0.4 and 0.2 eV lower than the T1 state of

Ir(ppy)₃). There is no evidence at the time of the invention of what would be a suitable Liq loading level for TPBi, or that Liq-doped TPBi could provide suitable electron transport to all three emissive layers.

43. The only example of a “prior art” white OLED with a doped ETL in the record involved a molecular dopant that allegedly did not generate “alkali-metal.” EX1029, 7. A skilled artisan would understand that this molecular dopant would not be Liq because Liq was known for potential dissociation. EX2023 [Lee 2008], 677 (binding energy in Liq “between Li and quinolate unit is weaker than that of LiF”). Hence, there is no teaching or suggestion of using Liq for a white OLED that includes an emissive layer having a PHOLED emitter.

44. In fact, a comparison of the J-V curve of Novaled’s devices with those of Ma shows that the Novaled’s material set provided far better driving voltages (4-5 V versus 6V and higher at 100 cd/m²) and power efficiency (over 80 lm/W at 100 cd/m² versus no more than 3-3.5 lm/W in Ma at 100 cd/m²). EX1029 at 17, EX1010, Figs 2-3. Noavled’s material set also allegedly provided “Record Lifetimes of PIN-OLEDs.” EX1029 at 19. A skilled artisan, if concerned with diffusion and OLED device performance, would have started with Novaled’s material set that used a non-Liq-doped ETL, instead of experimenting with Liq.

45. This is not surprising because PHOLEDs are especially sensitive to triplet exciton quenching by lithium. This is because PHOLEDs have theoretical

internal efficiencies of up to 100%, thus a skilled artisan would remove all possible sources that could quench the excitons. On the other hand, traditional fluorescent materials do not approach such high efficiency. Thus, the efficiencies of the fluorescent systems would be low to begin with and a skilled artisans would be much less concerned with lithium quenching triplet excitons that do not emit light at any rate.

C. A Skilled Artisan Would Not Have Used Ma's Fluorescent OLED Dopant in the Claimed Ir-based PHOLED Due to Significant Quenching Problems and Traps

46. I previously noted that the use of Liq as a dopant remained almost exclusively on fluorescent OLEDs until well after the filing date of the '425 Patent. *See* EX2002, ¶ 119 (citing EX2005 (Oh 2015) at 34). To date, the earliest reported use of Liq as an ETL dopant in PHOLED that I am aware of was 2011. *See* EX2003 (Kim 2011), Fig. 1 (using Liq as a dopant in PHOLED devices I and II). This is because Liq has significant problems as a dopant, particularly with PHOLED devices. I detailed many of them in my prior declaration. *E.g.*, EX2002, §§ VI.C.-VI.F. I provide additional explanations and support below.

47. As I previously explained, unlike fluorescent with OLEDs singlet excitons, Liq was known to quench the triplet excitons in phosphorescent OLED devices. EX2002, ¶¶ 106-107; *see id.*, ¶¶ 44, 95, 103, 115, 121, 127, 136; EX2005 (Oh 2015) at 34; EX2002, ¶ 38-39, 44, 106-107, 121(Liq describing as “act[ing] as

a luminance quencher in phosphorescent organic light-emitting diodes (PHOLEDs).”).

48. Post-425 solutions required a blocking layer to address the quenching problem. EX2024 (Kim 2013) at 1 (proposing using a triplet excitons blocking layer when using an ETL of TPBi doped with Liq in an phosphorescent system). Neither Samsung nor Dr. Holmes have presented any data to show that Liq would have provided improved performance and reduced diffusion over cesium, barium or LiF when a blocking layer is used in conjunction with a doped ETL.

49. Specifically, Liq can generate free lithium when cathode is deposited on it. EX2019_2025-04-24_Holmes, 27:4-9. A skilled artisan would have understood that small Li^+ cation in Liq would dissociate from the hydroquinolate anion and diffuse through the organic layers to wreak havoc on the device. *See* EX2002, ¶ 82. A 2008 paper by Lee expressly stated that the “[b]inding energy between Li and quinolate unit is weaker than that of LiF.” EX2023 (Lee 2008) at 676-678 (citing EX2027 (Schmitz 2000)). Liq can exist in multiple forms, including as $[\text{Liq}_2]^-\text{[Li]}^+$, from which the lithium cation is more easily disassociated and diffused. EX2027 (Schmitz 2000) at 3014; EX2023 [Lee 2008] at 677-78. Petitioner’s expert admits he did not analyze the disassociation of Liq or diffusion of Liq or Li^+ or make any comparison to more common dopants like LiF, barium, or cesium. EX2019_2025-04-24_Holmes, 138:17-141:22, 65:1-5.

50. Liq, with disassociated lithium cation or atom, can interact with the triplet excitons, causing them to lose energy through non-radiative pathways instead of emitting light. OLED emission is then quenched by lithium in phosphorescent OLED devices. EX2002, ¶ 121; EX2005 (Oh 2015) at 34.

51. Phosphorescent OLEDs—including Iridium-based Ir(ppy)₃ OLEDs—were known to be particularly susceptible to quenching problems. *See* EX2028 (Reineke 2007) at Abstract, 1-3, 7-8 (In contrast to fluorescent OLEDs, “phosphorescent OLED’s suffer from a decrease of efficiency when the current density is increased,” investigating triplet-polaron quenching in PHOLED devices, including Ir(ppy)₃-based OLEDs); *see* EX2021 (Yang 2024) at Abstract, 1 (discussing interfacial exciton-polaron quenching at the interface between emission and transport layers as significantly reducing OLED efficiency).

52. Triplet quenching is a concern in PHOLEDs but not in fluorescent OLEDs because triplets in a fluorescent OLED decays non-radiatively but should decay radiatively in PHOLEDs to achieve the 100% theoretical internal quantum efficiency.

53. As Dr. Holmes explained:

“[W]hen 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 [sic, remove] 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.

EX2020-2025-04-18_Holmes, 315:4-316:2; *see also* EX2002, ¶¶ 44, 106-107, 115, 121; EX3006 (Oh 2015) at 34-35. Thus, given the known weak binding energy of Liq, a skilled artisan would not have chosen Liq to dope an ETL in a PHOLED, especially when non-alkali-metal-containing dopants were known at the time of the invention. EX1029 at 7.

54. In addition, Liq introduces traps for charge carriers (electrons or holes). In phosphorescent OLEDs, where charge balance is critical for efficient exciton formation and actual operation, trapping exacerbates charge imbalances. EX2024 (Kim 2013) at 7999-8000 (recognizing in PHOLED that a “thick Liq layer” that “reduced carrier transport because it has an insulating property with a high dielectric constant” and that Liq has “low electron transport[] properties” and “obstruct[s] ... electrons movement by the existence of *many shallow traps* in the Liq molecules”); EX2025 (Le 2024) at 69636 (describing degradation due to unstable molecules, annihilation, instability of cations and anions, and traps as well as avoiding quenching).

55. As a result Liq doping could “retard[]” and “weaken” charge transport. EX2006 (Kang 2022), Abstract, 6 (theorizing the formation of an ETL-Liq

complex). The resulting charge imbalance due to an electron surplus and undesirable traps would have led to “dark spots, luminance decay[,] and subsequent low efficiency.” EX2003 (Kim 2011) at 54.

56. As I have noted before, significant work continued well after the filing date in trying to determine the correct energy band levels to dope materials with Liq for an operable system even when just based on Bphen as in Ma. EX2003 (Kim 2011), Abstract, 59 (testing multiple configurations of an ETL with doping concentrations of 5-65% of Liq in Bphen, reporting efficiency points as low as 5% and as high as 65% depending on configuration); EX2002, ¶ 131.

57. Dr. Holmes and Samsung do not explain how Liq would even be reasonably expected to work with the material sets in Diekmann or Suzuki, which would have different injection barriers and charge carrier mobilities than the Ma system. As further noted above, Diekmann and Suzuki include multiple emissive layers, which could have required different doping concentrations for charge balances. *E.g.*, EX2003, 59 (65% optimal range for red-emitting Ir(pq)₂acac with BPhen:Liq and a blocking BPhen layer); EX2024, Fig. 1 and Table 1 (optimal range between 3-10 % Liq for green-emitting Ir(ppy)₃ in TPBi and TPBi blocking layer); EX2007 & EX2105 (*compare* Devices-2, 3 and 5). Dr. Holmes and Samsung did not present any evidence that the delicate charge balance emphasized

by Ma would be maintained if Liq-doped ETL is used in Diekmann's or Suzuki's multi-EML OLEDs. EX2002, ¶¶ 123-131.

D. A Skilled Artisan Would Not Have Reasonably Expected Success in Doping an Unidentified Host ETL with Liq in a System Without a Specified PHOLED Emitter Matrix

58. The delicate and complicated nature of the relationship between the stacks (emitter matrix and materials, host materials, charge balance, hole/electron surplus, and quenching concerns) and problems with Liq and PHOLEDs, means that a skilled artisan would not have reasonably expected success in combining Ma's Liq-doped Bphen in an Alq₃ fluorescent system with Diekmann's or Suzuki's Iridium-based emitter in a phosphorescent system with different stacks. The complexity is further increased because Diekmman and Suzuki involved multiple emission layers; and it is unclear whether there could be a suitable Liq doping range and ETL/blocking layer thickness for these white OLEDs. A POSITA with a bachelors in physics or chemistry and three years of experience could not even attempt using Ma's novel Liq-doping scheme in Diekmann's or Suzuki's PHOLED devices in an unnamed ETL material with an expectation of success.

59. I understand that to have a reasonable expectation of success, one must be motivated to do more than merely to vary all parameters ... until one possibly arrived at a successful result. I also understand that no reasonable expectation of success is shown when defendants throw "metaphysical darts at a

board filled with combinatorial prior art possibilities.” A reasonable expectation of success requires “[s]ufficient guidance” from the asserted references.

60. I have reviewed Diekmann and Suzuki, neither provided any guidance on how to vary the parameters to accommodate Liq as opposed to Cs, Ba or LiF.

61. Doping of organic semiconductors was and still is unpredictable, especially when it comes to doping with Liq. EX2002, ¶ 103; *see id.*, ¶¶ 71, 113, 124. Years after the filing of the '425 Patent, “the reason for the enhanced electron transport property” of Liq in Bphen and Alq₃ and why such high doping (around 30%) concentrations were needed of Liq still “was not well understood.” EX2004 (Kumar 2011) at 1-2 (“[N]-type doping of organic semiconductors still remain[ed] a challenge”); EX2002, ¶ 119.

62. As I previously mentioned, as recently as 2022, researchers were still trying to figure out just how Liq works. EX2006 (Kang 2022), Abstract, 1 (investigating the “effect of coordinated structures of electron transporting materials (ETMs) with lithium quinolate (Liq) on charge transport characteristics.”); EX2002, ¶ 119. Doping of organic semiconductors was and still is unpredictable, especially when it comes to doping with Liq, as I previously explained. EX2002, ¶ 103; *see id.*, ¶¶ 71, 113, 124. Given this deep lack of understanding, Petitioner has not shown a reasonable expectation of success from a

bachelors in chemistry or physics with only three years of experience at the time of filing of the '425 Patent.

63. To illustrate, Petitioner identifies no particular host material for either the ETL in Diekmann or Suzuki. This makes it impossible to determine if the doping method is even compatible with the ETL host. The choice of dopant, the amount of dopant, and the method of doping must also be tailed to the choice of ETL host. And that host and dopant combination also needs to be chosen so at to be compatible with the overall system, in this case a phosphorescent, iridium-based OLED device. This leads to significant experimentation in finding compatible materials that maintain the correct energy level balance, avoid quenching and polaron-exciton annihilation, and do not trap charge carriers, among other problems. *See* Sections B-C, *supra*.

64. The host-dopant combination also needs to be chosen so at to be compatible with the overall system, in this case a phosphorescent, iridium-based OLED devices. Samsung and Dr. Holmes have not identified what ETL host material from Diekmann or Suzuki is being doped, its dimensions, or even the amount of Liq dopant to use. For instance, optimizing the thickness of the host and doping concentration of Liq in phosphorescent OLEDs is extremely challenging. The amount of dopant and the method of doping must be tailored to the choice of ETL host. Without knowing a particular host material for either the ETL in

Diekmann or Suzuki, it is impossible to determine if the doping method is even compatible with the ETL host. This also requires finding the right balance between electron injection and minimizing triplet exciton quenching while avoiding Liq's documented issues with acting as an insulator and charge carrier trapping. This would require careful experimentation and device engineering and is beyond a bachelors with just three years of experience.

65. In addition, Petitioner has not explained how it would even begin to deal with the quenching and charge imbalance problems caused by Liq. While other references attempted to quell cesium diffusion using an additional undoped Bphen layer (*See* EX1019 (He), Fig. 1), Petitioner has neither proposed such a modification nor provided any explanation of how thick of an undoped layer would be sufficient, how the increased thickness would affect the optical performance, what the appropriate material would, or how that material would alter the charge balance and driving voltage of the overall system. Ma did not propose any such blocking layer. The previously discussed weak binding energy in Liq and multiple forms of Liq would include lithium cations (or even lithium atoms) that can more easily diffuse into the emission layer than the heavier Cs atom in He. Indeed, even as of 2011, skilled artisans gravitated away from Bphen because of BPhen's known low T_g "which causes issues related to device lifetimes." EX2004, 114511-2.

66. No skilled artisan with only a bachelor's degree in physic or chemistry and three years of experience would have reasonably expected Ma's extreme doping to work in Diekmann or Suzuki's phosphorescent system.

III. DIEKMANN AND SUZUKI HAVE NO NEED FOR LEE'S ENCAPSULATION (CLAIM 4)

67. Claim 4 of the '425 Patent claims:

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.

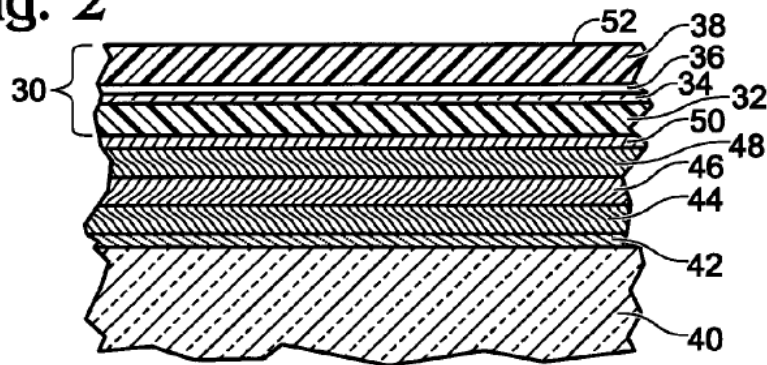
68. Lee describes a multi-layered encapsulation using parylene-based polymer layers 32 (a "semi-crystalline parylene-based polymer") and 38 (an annealed "highly crystalline parylene-based polymer") sandwiching an inorganic barrier layer 34. Lee, [0002], [0005], [0020]-[0024], Fig. 2. Lee reports an

“improved lifetime” only in contrast to “prior OLEDs having polyacrylate or amorphous parylene barrier layers.” *Id.*, [0020].

69. The relevant portions of Lee are reproduced below:

[0002] The present disclosure relates to an organic light emitting device, and more particularly to an organic light emitting device having a barrier for protection against environmental degradation.

Fig. 2



[0020] The semi-crystalline parylene-based polymer films of the embodiments described herein are deposited under such conditions that the films has a relatively high initial crystallinity, for example, equal to or above 10%. A semi-crystalline, and especially a highly crystalline parylene-based polymer film has a greatly improved density and decreased free volume (which is defined as open volume in the amorphous area excluding pinholes and defects) compared to amorphous parylene films and other organic polymer films, such as polyacrylate films. The highly crystalline parylene-based polymer layers have low-enough WVTR and OTR values that they not only “decouple” adjacent inorganic layers to prevent the propagation of defects between inorganic layers, but also themselves help block water vapor and oxygen permeation into the OLED. In this sense, the semi-crystalline and highly crystalline parylene-based polymer films actually act as barrier layers. Therefore, an OLED having barrier constructed with the semi-crystalline and highly crystalline parylene-based polymer barrier layers as described herein may have a significantly improved lifetime compared to prior OLEDs having polyacrylate or amorphous parylene barrier layers. Each of these embodiments is described in more detail below.

[0021] FIG. 1 illustrates, generally at 10, one exemplary embodiment of a method for forming a barrier layer in an OLED, and FIG. 2 shows, generally at 30, a greatly magnified, sectional view of an OLED having barrier formed via the method of FIG. 1. Method 10 includes forming (at 12) a first semi-crystalline parylene-based polymer layer over an underlying layer, forming (at 14) an inorganic barrier layer over the first semi-crystalline parylene-based polymer layer, optionally forming (at 16) a reflective layer over the inorganic barrier layer, forming (at 18) a second semi-crystalline parylene-based polymer layer over the reflective layer, and annealing (at 20) the second semi-crystalline parylene-based polymer layer to form a highly crystalline parylene-based polymer layer.

[0022] The first semi-crystalline parylene-based polymer layer is shown in FIG. 2 at 32, the inorganic barrier layer is shown at 34, an optional anti-reflective layer is shown at 36, and the second semi-crystalline parylene-based polymer layer is shown at 38. The OLED device layers are formed on

a substrate **40**, and include a transparent anode **42**, an organic hole transport layer **44**, an organic light emitting layer **46**, an organic electron transport layer **48**, and a cathode **50** formed from a low work function metal. It will be appreciated that the depicted OLED architecture is merely exemplary, and that any other suitable architecture may be used. The specific materials used for the device layers are well known in the art, and are therefore not described in further detail herein.

70. Samsung proposes using Lee’s encapsulation in Diekmann and Suzuki to meet claim 4. Pet. 38-41, 71-72. Samsung and Dr. Holmes, however, do not explain why a POSITA would have been motivated to use Lee’s encapsulation method over Diekmann and Suzuki’s existing encapsulation.

A. Diekmann and Suzuki Have No Further Need of Encapsulation

71. Samsung cites Lee’s statements about protecting cathodes from air and the “multi-layer” encapsulation in Lee as decoupling the adjacent “inorganic layers” to prevent “defects” and “block water and oxygen permeation into the OLED.” Pet. 40 (citing Lee, ¶¶ [0005], [0020]). For Suzuki, Samsung ostensibly points back to protection from “outside materials ... for the same reasons discussed above in §VIII.C.1.” Pet. 71.⁴

⁴ Petitioner refers broadly to the entire eight-page of analysis for claim 4 in the Diekmann ground. This too is an improper incorporation by reference and woefully nonspecific. 37 C.F.R. §§ 42.6(a)(3), 42.104(b)(5); CTPG at 35-36.

72. Diekmann and Suzuki already encapsulate their OLED devices for these same reasons and thus have no need for further encapsulation. *See* Diekmann at 30-31, 94, 109, 120, Figs. 2A (thin film encapsulation 6), 3, 17A; Suzuki, ¶¶ [0004] (describing problem of permeation), [0067] (addressing that problem with “barrier film ... has a high density and low gas and moisture vapor permeability”), [0115] (“silicon oxide is particularly preferable from the perspective of moisture permeability, light permeability”). The statements about protection from outside materials and blocking water and oxygen permeation are merely, again, describing what encapsulation generally does and are not tailored to Lee’s specific form of encapsulation or why it would be better than Diekmann’s or Suzuki’s existing encapsulation or what would actually motivate a skilled artisan to use Lee’s complicated annealing method over Diekmann’s or Suzuki’s encapsulation.

B. Neither Diekmann or Suzuki Use Polyacrylate or Amorphous Parylene Barrier Layers

73. Samsung alleges encapsulation generally was “well-known.” Pet. 40-41, 71-72. Whether encapsulation generally is well known does not say why a skilled artisan would use Lee’s specific semi-crystalline encapsulation approach. Samsung’s blanket citation to the entirety of Exhibits 1020 and 1021 are not explained.

74. As for Lee’s claim of “improved lifetime” (Pet. 40, 71) from annealing its semi-crystalline, parylene-based polymer barrier, Lee is clear that this

improved lifetime was “compared to prior OLEDs having polyacrylate or amorphous parylene *barrier* layers,” as Samsung admits. Pet. 40 (citing Lee, [0020]) (emphasis added). Neither Diekmann nor Suzuki use polyacrylate or amorphous parylene *barrier* layers.

75. Thin film encapsulation, such as in Diekmann or Suzuki, sometimes uses a polymer layer as a stress relief between inorganic barrier layers (to prevent cracks propagating through the barrier layers). The inorganic barrier layers—commonly silicon oxide or silicon nitride—are what blocks the percolation of water and oxygen. Even though Lee’s parylene-based polymer might be more crystalline, a skilled artisan would not expect it to improve or even act equally as well as a traditional silicon nitride barrier in terms of its barrier properties. Lee’s process would not have been expected to improve Diekmann or Suzuki in any way.

76. To illustrate, Samsung points to Diekmann’s “intermediate polymer layers,” labelling this “the precise prior art structure that Lee’s encapsulation is intended to improve upon.” Pet. 40 (citing Diekmann, 15; Lee, ¶ [0008]). Samsung points to the wrong layer. Diekmann’s intermediate layer is a single *buffer* layer, sandwiched between the Diekmann’s two barrier layers as part of “thin film encapsulation 6.” Diekmann, 15, 94, Fig. 3. Diekmann’s two barrier layers are not made of polyacrylate or parylene, but are silicon oxide and silicon nitride layers, as Samsung admits and relies on elsewhere. Pet. 21, 26-27 (citing Diekmann, 94, Fig.

3). Neither Samsung nor Dr. Holmes provides any explanation for how Lee's annealing of parylene-based polymer barriers would improve the lifetime of Diekmann's device with its non-parylene-based barrier layers of silicon oxide and silicon nitride.

77. Samsung also never identifies a specific barrier layer in Suzuki to implement Lee or explains how Lee would improve Suzuki's lifetime. *See* Pet. 71. Samsung refers back to its Diekmann analysis with little explanation. *Id.* (alleging motivation "to protect the OLED from outside materials and improve the lifetime of the device for the same reasons discussed above in §VIII.C.1 with regards to Diekmann."). Samsung makes no allegation that Suzuki uses a polyacrylate or an amorphous parylene barrier layer. Nor could it.

78. Samsung relied on Suzuki's "gas barrier thin film laminate 9" comprising "buffer film 3," "a barrier film 4," and another "buffer film 3" (Pet 54 (citing Suzuki, Fig. 9, ¶¶ [0273]-[0274])). Samsung agrees that barrier film 4 is a "dense intervening layer" with "low water vapor transmission" comprising "a ceramic composition with low or no carbon content and having at least Si and O atoms." Pet. 59 (citing Suzuki, ¶ [0117]); *see* Pet. 59-60 (citing Suzuki, ¶¶ [0068]-[0069]) (alleging barrier and buffer films are made of silicon oxide, silicon oxide nitride, or silicon nitride). But this barrier is not polyacrylate or parylene. Samsung

has not identified any motivation to use Lee's annealing of parylene-based polymer barriers in Suzuki.

79. Samsung has not suggested replacing either of the silicon oxide or silicon oxide nitride, or silicon nitride barrier layers from Diekmann or Suzuki with the parylene barrier layers. Thus, it remains unclear exactly what the resulting barrier or overall encapsulation stack would be. Nor has Samsung explained why a skilled artisan would do so or that the resulting structure would have good interlayer adhesions and compatibility to provide the required reliability and protection from water and air. Nor has Samsung explained whether in the resulting structure, the annealed parylene would have a different amorphicity than silicon oxide or silicon nitride to satisfy the claimed requirement that "the volume structure of the second encapsulation layer has a higher amorphicity than the volume structure of the first encapsulation layer" in claim 4.

C. Samsung's Expectations of Success are Conclusory

80. First, Samsung asserts without evidence that reasonable expectation of success is to be expected because Diekmann and Suzuki was "already contemplating" encapsulation and the proposal was merely replacing one known element for another. Pet. 41, 71-72 (no cited support beyond Samsung's expert parroting identical language in EX1003, ¶¶ 151, 233). Mere replacement is not a reason for why a skilled artisan would reasonably expect success using Lee's

encapsulation. Samsung ignores the tremendous considerations needed in OLED systems: is the replacement material going to be compatible with the rest of the materials to provide good adhesions and the right mechanical support to provide a desired reliability?

81. For example, Lee does not mention a doped OLED. But the annealing process would necessarily subject the device to heat. Although Lee teaches ways to mitigate the effect of temperature, the OLED stack can still be raised to a sufficient temperature to promote diffusion. Dr. Holmes never examined the temperature profile during annealing and that was not surprising because he did not even know what parylene materials would be suitable for Diekmann or Suzuki, without which, one would not know the temperature profile. EX2019_2025-04-24_Holmes, 118:6-11, 120:10-125:10. Increased diffusion rate at high temperatures would be a real concern and would be a reason to avoid Lee's system.

82. Samsung states that encapsulation using alternating organic and inorganic layers was "well-known." Pet. 41, 71 (citing EX1003, ¶¶ 43-44, 151, 231-232; EX1020, EX1021). That says nothing about expected success using the specific encapsulation in Lee of a semi-crystalline, parylene-based polymer barrier and an annealed, highly crystalline, parylene-based polymer barrier. Increasing the crystallinity of one of the encapsulation layers will logically only make the encapsulation even more brittle.

83. Samsung and Dr. Holmes state that this modification was within the ability of a skilled artisan. Pet. 41, 72; EX1003, ¶¶ 151, 233. I disagree that a POSITA with only a bachelor's in chemistry or physics and three years experience would have been able to or reasonably expected success in attempting to implement Lee's complicated encapsulation techniques in Diekmann or Suzuki.

84. In fact, Lee gives no guidance on the appropriate parylene system, the annealing temperature, or temperature profile across the encapsulation layer and OLED during annealing for Diekmann or Suzuki's system. Lee does not even give a starting point for its annealed, parylene barrier system. I understand that varying numerous parameters to arrive at a combination is not obviousness.

I declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code.

Executed on May 5, 2025, in Ringgold, GA, _____.

A handwritten signature in black ink, appearing to read 'P.M. Pattison', is written over a horizontal line.

P. Morgan Pattison, Ph.D.

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.

Case No. IPR2024-01095
Patent No. 11,828,425

DECLARATION OF DR. P. MORGAN PATTISON, PhD.

TABLE OF CONTENTS

	<u>Page</u>
I. INTRODUCTION	1
II. QUALIFICATION AND EXPERIENCE	1
III. LEGAL STANDARDS	5
A. Claim Construction	5
B. Obviousness.....	6
C. Level of Ordinary Skill in the Art	9
D. Materials Relied on in Forming My Opinions	11
IV. TECHNOLOGICAL BACKGROUND	11
V. OVERVIEW OF THE ALLEGED PRIOR ART.....	16
A. Diekmann (EX1004 & EX1005).....	16
B. Suzuki (EX1006 & EX1007)	20
C. Ma (EX1010).....	22
VI. A POSITA WOULD NOT HAVE USED LIQ AS A ETL DOPANT IN DIEKMANN OR SUZUKI.....	24
A. Samsung and Dr. Holmes Provide No Reason for Why a POSITA Would Use Liq in Diekman’s or Suzuki’s ETL With a Reasonable Expectation of Success	25
B. Ma Does Not Provide a Motivation to Dope Every ETL with Liq.....	33
C. The Use of Liq as an ETL Dopant Was Experimental for Limited Use in Stable, Fluorescent OLED Devices with Bphen at the Time	39

D.	Samsung and Dr. Holmes Do Not Account for the Differences Between Ma’s Alq ₃ -based Fluorescent Emission System Versus the Claimed Iridium-based Phosphorescent Emission Systems.....	48
E.	Samsung and Dr. Holmes Do Not Address the Delicate Energy-Level Balance Required for the Different Organic Layers in an OLED Stack.....	52
F.	Samsung and Dr. Holmes Offer No Evidence that Diekmann and Suzuki Would Have Gained Any Benefit from Using a Liq-doped ETL	56

1. I, P. Morgan Pattison, PhD, declare as follows:

I. INTRODUCTION

2. I have been retained as an independent expert on behalf of Pictiva Displays International Ltd. (Patent Owner) to provide technical background and opinions related to U.S. Patent No. 11,828,425 (EX1001) (the '425 Patent) at issue in in IPR2024-01095, including to Petitioner Samsung Display Co., Ltd.'s position regarding Grounds II and VII (Pet. 36-38, 69-70) involving Claims 2 and 17 and other matters addressed herein.

3. For this service, I am compensated for my time at my normal consulting rate. My compensation does not depend on the outcome of any matter or on any of the opinions provided below. I have no financial interest in this matter.

4. My testimony herein is based on my technical background, my years of experience in the field of organic light emitting diodes (OLED) and my review of the materials cited herein.

II. QUALIFICATION AND EXPERIENCE

5. My curriculum vitae (CV) is attached hereto as Exhibit A and provides an accurate identification of my background and experience.

6. I have over 25 years of experience in the areas of light emitting diodes (LEDs), organic light emitting diodes (OLEDs), lighting, solid state lighting (SSL), displays, lasers, and optoelectronic materials and devices.

7. I attended the University of California, Santa Barbara, receiving my M.S. in Electrical Engineering, Electronics and Photonics in 2003 and my Ph.D. Materials Science, Electronic and Photonic Materials in 2006. I received my B.S. in Electrical Engineering, Electronic Devices and Materials from the University of California, San Diego, in 1996.

8. This coursework included significant classwork in both chemistry and electronic devices as well as classwork and research on the fabrication and evaluation of optoelectronic devices, such as light emitting diodes, which require understanding of both materials chemical properties as well as electronic properties. This coursework through my Ph.D. exceeds those offered to a bachelors in chemistry and physics relevant to OLED technology at the time of the '425 Patent.

9. Presently, I am the President of Solid State Lighting Services, Inc. In that capacity, I am responsible for, among other things, providing technical, strategic, and market expertise in the areas of LEDs, OLEDs, lighting, SSL, displays, and optoelectronic materials and devices. I am also involved in connecting optical device technical performance with application requirements for

displays, general illumination, color discrimination applications (medical, retail), and various novel lighting applications enabled by SSL technology. Solid State Lighting Services also provides services related to LED and lighting product failure and reliability evaluation. I founded Solid State Lighting Services in 2007, and I have worked at Solid State Lighting Services, Inc. for almost 17 years.

10. From January 2006 through November 2007, I worked as a Project Manager at the U.S. Department of Energy (DOE). In that capacity, I was responsible for managing DOE funded research and development projects in SSL, including efficient LED and OLED-based lighting technologies. My duties included managing the solicitation and selection of R&D projects with the objective of saving energy. As part of the solicitation and selection process, I evaluated and identified priority areas of research necessary for advancement of SSL in terms of efficiency, lighting performance, and cost. In this capacity I identified priority R&D areas to achieve the maximum energy savings, I managed R&D funding solicitations, and I managed DOE funded R&D projects covering all aspects of LED and OLED lighting technology including, light emitting materials and devices (LEDs and OLEDs), power supplies, optics, integrated fixtures, lighting controls, and lighting applications.

11. From September 2000 through January 2006, I was a Graduate Student Researcher at University of California Santa Barbara (UCSB). I performed

my Doctoral thesis research under Professor Shuji Nakamura (Nobel Prize in Physics 2014) on gallium-nitride (GaN) based micro-cavity light emitting diodes. The research included: epitaxial structure design and growth by MOCVD, semiconductor process development, materials characterization, and electro-optical device characterization. While at UCSB, I participated in additional areas of research, including research and development of UV LEDs, vertical cavity surface emitting lasers (VCSEL), and edge emitting laser diodes.

12. From November 1996 through May 2000, I worked as a Laser Test Engineer at Cymer Laser. I was responsible for testing and developing test methodology for production and prototype KrF and ArF excimer lasers used in deep UV photolithography for manufacturing of advanced semiconductor products. Testing included optical characterization and full system troubleshooting. I also developed new electrical and optical test methodologies to improve test cycle times, automated test procedures, and developed test documentation.

13. I am listed as an inventor on at least 11 issued patents, including multiple patents in the field of LED materials and device design, LED lighting product design, and home automation and controls for lighting.

14. I have authored numerous peer reviewed journal articles and publications on the topics of SSL technology and applications.

15. Since 2008, I have also authored annually updated R&D and technology and manufacturing roadmaps used by the DOE to guide federal lighting R&D efforts. I frequently give presentations and talks on the topic of LED and OLED technology, SSL status and R&D opportunities, and application advancements enabled by LED technology.

III. LEGAL STANDARDS

16. I am not an attorney. I have relied on instructions from counsel as to the applicable legal standards to use in arriving at my opinions in this Declaration. I am relying only on instructions from Patent Owner's attorneys for these legal standards.

17. I understand that Samsung, as the Petitioner, has the burden of proving that the challenged claims of the '425 Patent are unpatentable by a preponderance of the evidence.

A. Claim Construction

18. I understand that claim terms in an IPR are given their ordinary and customary meaning as understood by one of ordinary skill in the art (POSITA) in view of the entire patent and the prosecution history pertaining to the patent. I understand claim terms are to be given the meaning they would have to a person having ordinary skill in the art at the time of the invention, taking into consideration the patent, its file history, and, secondarily, any applicable extrinsic

evidence (*e.g.*, dictionary definitions). I understand that applicants sometimes provide special definitions for claim terms that differ from the meaning the term might otherwise possess, also known as lexicography. I have applied these standards in this declaration.

19. I have no opinion on any particular claim construction at this time.

B. Obviousness

20. I understand that a patent claim is invalid if it would have been obvious in view of the prior art. I further understand that obviousness is determined from the perspective of a POSITA at the time the invention was made.

21. Whether or not explicitly stated below, my analysis of the prior art is made as of the time the invention was made. For purposes of this *inter partes* review, I have assumed that the priority date for the '425 Patent is December 11, 2008. My analysis, however, does not change if that date was December 9, 2009.

22. I understand that a claim may be unpatentable as obvious if the differences between the subject matter in the prior art and the subject matter in the claim would have been obvious to a POSITA at the time of the invention. I understand that obviousness can be based on a single prior art reference or a combination of prior art references.

23. I understand that a prior art reference must be viewed as a whole. I further understand that the following factors must be evaluated to determine

whether the claimed subject matter is obvious: (1) the scope and content of the prior art; (2) the difference or differences, if any, between each claim of the patent and the prior art; (3) the level of ordinary skill in the art at the time the patent was filed; (4) objective indicia of non-obviousness.

24. I understand certain objective indicia can be important evidence regarding whether a patent is obvious or nonobvious. Such indicia include: commercial success of products covered by the patent claims; a long-felt need for the invention; failed attempts by others to make the invention; copying of the invention by others in the field; unexpected results achieved by the invention as compared to the closest prior art; praise of the invention by the infringer or others in the field; the taking of licenses under the patent by others; expressions of surprise by experts and those skilled in the art at the making of the invention; and the patentee proceeding contrary to the accepted wisdom of the prior art.

25. I have been informed and understand that the teachings of two or more references may be combined if such a combination would have been obvious to one having ordinary skill in the art. It is also my understanding that where there is a reason to modify or combine the prior art to achieve the claimed invention, there must also be a reasonable expectation of success in so doing to render the claimed invention obvious. I also understand that the references themselves need not provide a specific hint or suggestion of the alteration needed to arrive at the

claimed invention; the analysis may include recourse to logic, judgment, and common sense available to a POSITA.

26. I understand that when the prior art “teaches away” from combining prior art references or certain known elements, discovery of a successful means of combining them is less likely to be obvious. A prior art reference may be said to “teach away” from a patent when a person of ordinary skill, upon reading the reference, would be discouraged from following the path set out in the patent or would be led in a direction divergent from the path that was taken by the patent.

27. I have been informed and understand that one of ordinary skill in the art has ordinary creativity, and is not an automaton. I have been informed and understand that in considering obviousness, it is important not to determine obviousness using the benefit of hindsight derived from the patent being considered.

28. Finally, I understand that Patent Examiners at the U.S. Patent and Trademark Office (“USPTO”) rely upon certain exemplary rationales in review patent applications to understand whether the subject matter of the claims is obvious. I understand that the following is a list of exemplary rationales relied upon by Patent Examiners at the USPTO:

A. Combining prior art elements according to known methods to yield predictable results;

- B. Simple substitution of one known element for another to obtain predictable results;
- C. Use of a known technique to improve similar devices (methods, or products) in the same way;
- D. Applying a known technique to a known device (method, or product) ready for improvement to yield predictable results;
- E. “Obvious to try”- Choosing from a finite number of identified, predictable solutions, with a reasonable expectation of success;
- F. Known work in one field of endeavor may prompt variations of it for use in either the same field or a different one based on design incentives or other market forces if the variations are predictable to one of ordinary skill in the art; and
- G. Some teaching, suggestion, or motivation in the prior art that would have led one of ordinary skill to modify the prior art reference or to combine prior art reference teachings to arrive at the claimed invention.

C. Level of Ordinary Skill in the Art

29. I understand that interpreting patent claims and validity must be undertaken from the perspective of a hypothetical POSITA. I understand that in determining the characteristics of a hypothetical person having ordinary skill in the art of the patent at the time of the claimed invention, I must consider several

things, including the type of problems encountered in the art, the solutions to those problems, the rapidity with which innovations are made, the sophistication of the technology, and the education level and experience of people working in the field.

30. I understand that Samsung and Dr. Holmes defines a POSITA at the time of the invention as someone with “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.” Pet. 10 (citing EX1003, ¶ 63). It is not clear what Samsung means by “a comparable field of study.” For the purposes of this Declaration, I have been instructed to apply Samsung’s proposed level of skill in the art.

31. I express no opinion on whether this level of ordinary skill is correct. Yet this level of ordinary skill in the art is quite low. At the time of the invention, OLED design was a unique, cross-disciplinary field, requiring expertise in chemistry, materials science, electronic devices, and optics. Large teams of scientists and engineers with advanced degrees, usually doctorate level, were required to develop materials, designs, and processes for advanced OLED devices.

32. Samsung’s and Dr. Holmes’s low level of ordinary skill would mean that a POSITA would not be motivated to or reasonable able to easily deviate from the express teachings in the prior art to extrapolate broader uses with a reasonable expectation of success. The theoretical basis and experimentation necessary to do

deviate from express teachings in the prior art would not be routine for one of this skill level.

33. Given my education and experience at the time, I was at least a person of ordinary skill in the art, if not one with above average skills, at the time of the invention.

D. Materials Relied on in Forming My Opinions

34. In forming my opinions expressed in this declaration, I have relied on my own knowledge, experience, and expertise, as well as the knowledge of a POSITA in the relevant timeframe. In addition, I have reviewed and relied upon the materials cited in this declaration.

IV. TECHNOLOGICAL BACKGROUND

35. A light emitting diode (LED) uses two semiconductor layers—one rich in electrons (n-type) and one rich in “holes” (*i.e.*, the absence of electrons) (p-type)—to form a junction where electrons and holes meet, recombine, and emit light when electricity is applied. This LED is a “diode” because electricity can only flow in one direction. And an LED is “light emitting” because electrons and holes enter this diode junction and recombine and their energy is released in the form of light.

36. Effective LEDs require that there are not energy barriers or energy traps in the path of the electrons or holes. So, the energy levels of any materials in

the along the paths of electrons and holes from the electrodes to the LED junction must be selected or engineered for suitable energy levels with respect to the LED junction. In addition, the injection of electrons and holes must be balanced such that the electrons and holes meet with similar quantities within the junction region of the LED. Excess holes or electrons will be unable to recombine to emit light and will find energy release pathways that damage the LED materials reducing the efficiency and lifetime of the LED. Electrons and holes that meet away from the junction may combine non-radiatively and also reduce efficiency and lifetime of the LED device.

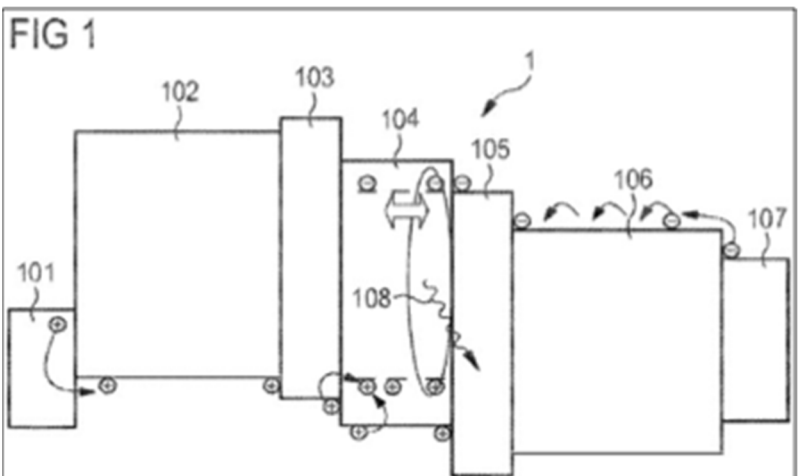
37. An QLED is an LED with light emitted by organic (*i.e.*, carbon-containing) molecules. Generally, OLEDs are made of a series of thin, carbon-based semiconductor layers sandwiched between a cathode (the negative electrode) and an anode (the positive electrode). All of these layers are deposited on a substrate.

38. Applying a voltage across the anode and cathode, the electrons and holes travel into an emissive layer (where light is produced) and recombine. When a hole meets an electron, they form an “exciton” (an electron-hole pair), releasing energy in the form of light as the exciton decays. Excitons are either “triplet” or “singlet” excitons, formed in a ratio of 3 triplets for every 1 singlet (3:1).

39. Emitters can be fluorescent or phosphorescent. Fluorescent emitters primarily use the singlet excitons for emission as triplet excitons do not decay rapidly enough in fluorescent materials to emit light. A fluorescent emitter is only 25% efficient as a result as only 1 in 4 excitons decay to emit light. A phosphorescent emitter is significantly more efficient than a fluorescent emitter because the light emission is also caused by the triplet excitons in addition to the singlet excitons. In contrast to a fluorescent emitter, a phosphorescent emitter is theoretically 100% efficient.

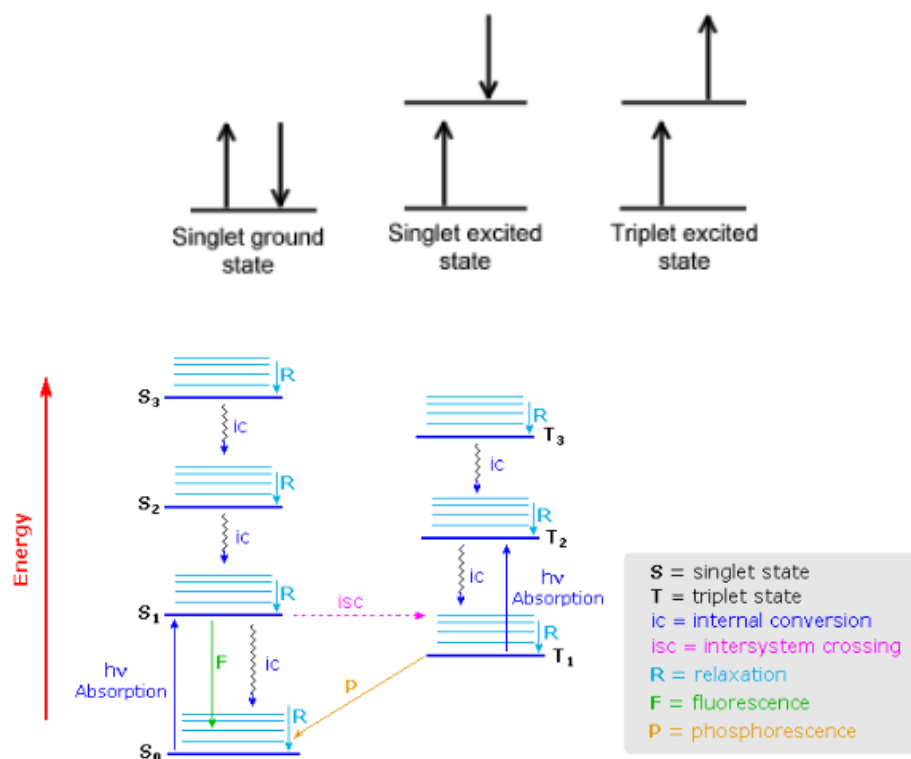
40. There are variations of this basic layered OLED structure. Layers can be added to align energy levels, to improve conduction of the charge carriers (electrons or holes), or to create energy barriers for travel of electrons or holes beyond the junction or emission region.

41. The '425 Patent illustrates one arrangement in Figure 1 with: (1) hole and electron transport layers to efficiently “transport[] the positive charge carriers”—holes—from the anode and “negative charge carriers—electrons—from the cathode” to the emission layer; and (2) electron and hole blocking layers to “prevent[] the penetration of electrons” from the emission layer into the hole transport layer and to “prevent[] the penetration of positive charge carriers into the adjoining electron transport layer,” respectively. '425 patent at 24:33–62, Fig. 1.

anode (first electrode 101)	
hole transport layer 102	
electron barrier layer 103	
radiation-emitting region 104	
hole barrier layer 105	
electron transport layer 106	
cathode (second electrode 107)	

42. Additional “injection” layers can be added to the cathode or anode to improve electron and hole injection into the transport layers. *E.g.*, ’425 Patent at 28:16-18 (“In addition, the second electrode **107** can have, on a side facing the radiation-emitting region **104**, a layer comprising LiF, which has good electron injection properties.”).

43. The ’425 Patent claims are directed to an OLED device with an “iridium-containing compound.” ’425 Patent at 88:7-9. Iridium-based compounds are phosphors, where light emission involves a change in electron spin from the higher energy, excited state to a lower energy, ground state. This results in a lifetime from seconds to minutes, significantly longer than that for molecules undergoing fluorescence, which does not change in electron spin.

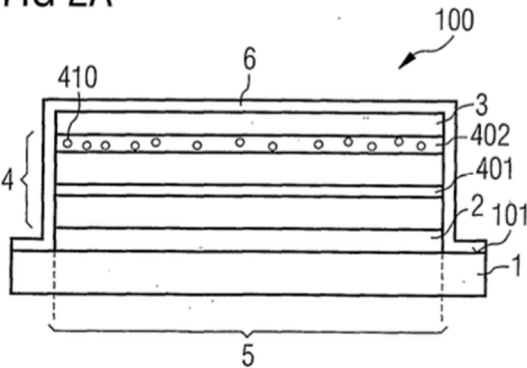


44. Material selections—especially for the emission layer and the layers immediately adjacent to the emission layer—are considerably different for a phosphorescent OLED than a fluorescent OLED. For the latter, the chosen materials must be carefully chosen not to quench the triplet excited state, which could then make the entire system unsuitable for OLED application.

V. OVERVIEW OF THE ALLEGED PRIOR ART

A. Diekmann (EX1004 & EX1005)

45. Diekmann describes an “an organic lighting device and lighting equipment with such a lighting device.” Diekmann, 1. Figure 2A illustrates a series of OLED layers in Diekmannn:

	Thin-film encapsulation 6
	Second electrode 3 (cathode)
	Doped layer 402 with dopant 410
	Unlabeled Layer
	Layer 401 suitable for generating light
	Unlabeled layer
	First electrode 2 (anode)
	Substrate 1 with main surface 101

Diekmann, 91-92, Fig. 2A.

46. Diekmann’s organic layer stack includes separate electron injection and transport layers. *See id.* at 4 (“the organic layer stack may comprise further organic layers, such as a hole-injecting layer, a hole-conducting layer, an electron-injecting layer and an electron-conducting layer”), 10 (“the doped organic layer forms an outermost layer of the organic layer stack, which particularly preferably forms a common interface with the respective electrode facing it”; “n-doped layer adjacent to the cathode, leads to better injection of electrons from the cathode into the organic layer stack.”).

47. Diekmann lists its “electron-conducting layer” materials as “Alq₃,” “BALq,” “TPBi.” *Id.* at 5.

48. Diekmann identifies its “electron injection layer” as containing at least one of an organic material NET-5 with NDN-1 and inorganic materials cesium carbonate (“Cs₂CO₃,”), lithium or sodium fluoride (“LiF,” “NaF”), and barium (“Ba”). Diekmann, at 5.

49. NET-5 with NDN-1 is referring to a doped injection layer. *See* EX1029 at 7 (listing NDN-1 as an n-dopant).

50. Diekmann further discloses:

[T]he organic layer stack comprises a doped organic layer with a dopant, which is arranged between the at least one organic layer suitable for generating light and one of the electrodes. Particularly preferably, the doped organic layer forms an outermost layer of the organic layer stack, which particularly preferably forms a common interface with the respective electrode facing it. electrode facing it.

...

[D]oping advantageously increases the conductivity of the doped organic layer and, in the case of an n-doped layer adjacent to the cathode, leads to better injection of electrons from the cathode into the organic layer stack ... Preferred n-dopants are cesium, barium and lithium fluoride.

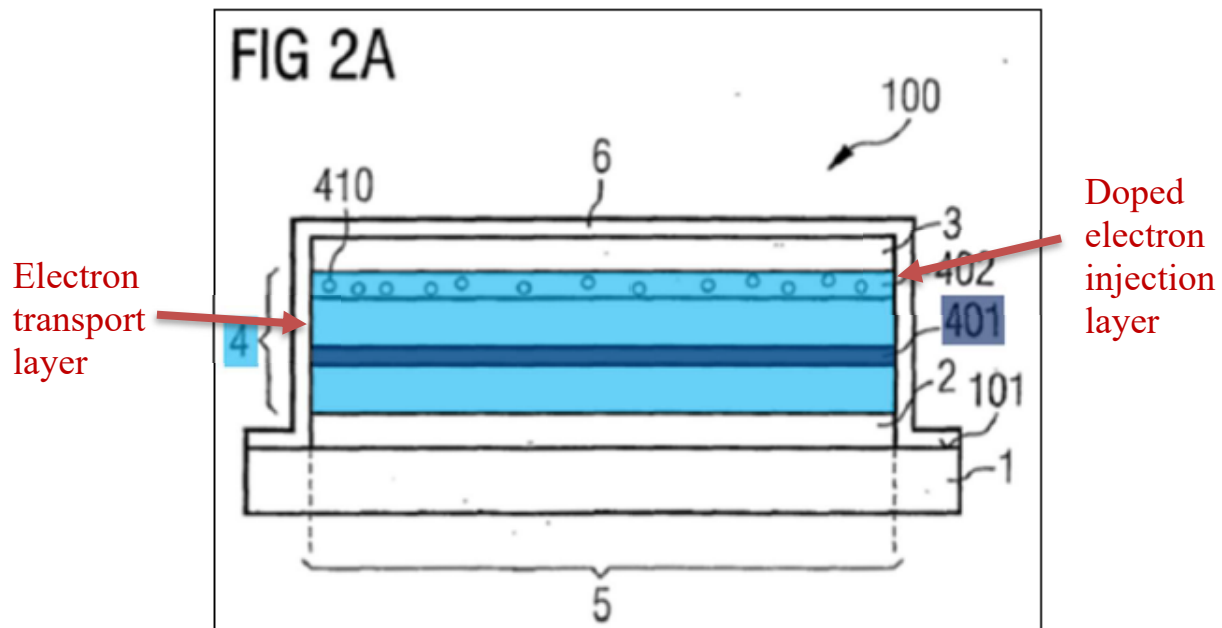
...

On its outer side facing the cathode, the organic layer stack 4 has a doped layer 402 with a dopant 410, which acts as an electron donor. This advantageously increases the injection of electrons from the cathode into the organic layer stack. The dopant 410 can be for example cesium, barium or lithium fluoride.

Id. at 10, 91.

51. Doped layer 402, lying next to the cathode, is expressly designed to improve electron **injection**. Diekmann, 91 (“dopant 410, which acts as an electron donor” ... “advantageously increases the **injection** of electrons from the cathode into the organic layer stack.”); *see id.*, 10 (“in the case of an n-doped layer adjacent to the cathode,” doping “leads to better **injection** of electrons from the cathode into the organic layer stack.”).

52. The location of doped layer 402 in Figure 2A shows that doped layer 402 abuts the electrode 3 and there is another layer between doped layer 402 and the light generating layer 401 that would have been understood to be the conducting layer:

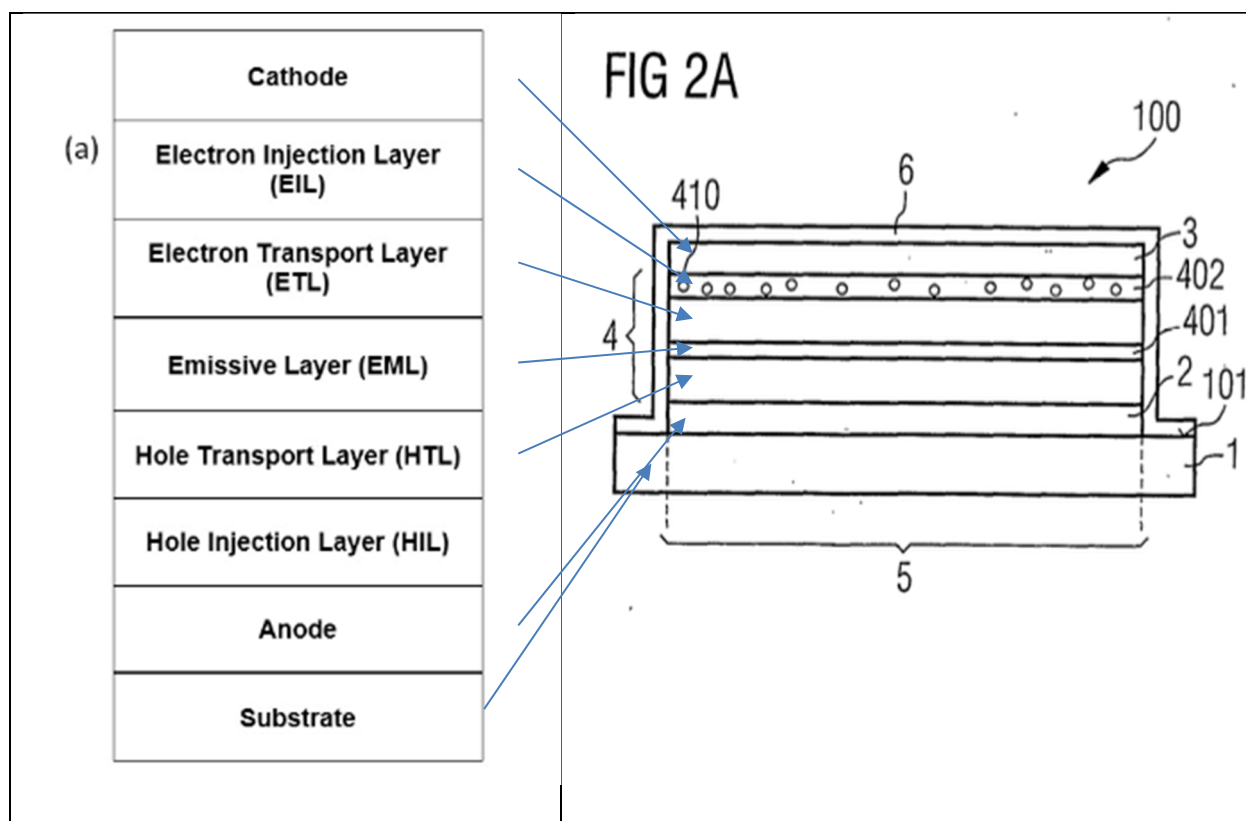


Diekmann, Fig. 2A (red annotations added; *see* Pet. 23 (adding blue annotations)).

53. Relative to the cathode, a POSITA would have understood that the electron injection layer abuts the cathode and, next, the electron transport layer abuts the emissive layer. *See* EX1003, Fig. 3(a) (illustrating this arrangement, discussed below).

54. Aside from an electron injection layer and an electron conducting layer, Diekmann does not describe any other possible layers between the electrode and the emissive layer (*e.g.*, a hole barrier layer 105 as in the '425 Patent, Fig. 1, 24:55–62).

55. This arrangement is shown in the figure provided by Dr. Holmes when compared to Diekmann Figure 2A:



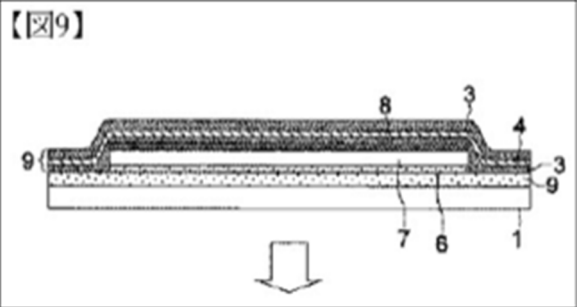
EX 1003, Fig. 3(a), ¶ 34; Diekmann, Fig. 2A (annotations added).

56. A POSITA would understand from these disclosures and the fact that another layer separates layer 402 and the emission layer 401 that layer 402 is an electron injection layer while the unlabeled layer between layers 401 and 402 are an electron-transport layer.

57. A skilled artisan would understand that doped layer 402 is thus describing an electron injection layer, not an electron transport layer given its location and express purpose of increasing injection.

B. Suzuki (EX1006 & EX1007)

58. Suzuki describes bottom emitting “organic electroluminescent device” with a “gas barrier resin substrate material.” Suzuki, ¶ [0001]. Samsung relies primarily on Figure 9 and the accompanying description in paragraph 274:

	Thin film laminate 9	Buffer film 3
		Barrier film 4
		Buffer film 3
	Cathode 8	
	Organic layer 7	“comprising a hole transport layer, a light emission layer, an electron transport layer, and the like”
	Anode 6	
	Thin film laminate 9	
	Resin substrate 1	

Id., Fig. 9, ¶ [0274]. Suzuki also discloses using electron injection layers next to the cathode, such as a layer of LiF. *Id.*, ¶¶ [0295], [0338].

59. Suzuki then describes that “an electron transport layer with a high n nature doped with impurities.” *Id.*, ¶ [0333]. Suzuki does not say that the ETL has been *n-doped*. A POSITA would have understood that this meant a material with an already “high n nature” is then “doped with impurities.” Because an ETL is for transporting negative charge carriers (electrons), ETL materials tend to have a high n (or negative charge) characteristic. That is because electron transport requires both a high number of electrons in the ETL and good electron mobility. Thus, if there are no electrons in the material, there would be no electron transport. This material with n characteristic may be then doped.

60. Here, a “high n nature” simply means that it is a material with a high electron (negative charge carrier) transport capability. But that does not necessarily mean the high n nature is derived from the doped impurities. A POSITA would have been well-aware of impurity doping for other reasons than n-type doping, such as colorant dopants, hole trapping, or hole blocking doping. *E.g.*, EX1013 (Baldo 1998) at 151 (doping a phosphorescent dye (PtOEP) into an Alq3 “electron transport material” for red coloring); EX2008 (Adamovich 2003), Abstract (doping a combined “hole blocking and electron transport layer”), 82 (doping FIrrpic into wide gap matrix to block holes). Hole blocking or trapping is

particularly important, as it can prevent hole leakage from the EML into the adjacent layers, which may then upset hole-electron balance, damage the device, and significantly degrade device performance.

C. Ma (EX1010)

61. Ma describes an Alq₃-based fluorescent OLED stack using both “8-hydroxy-quinolinato lithium doped 4,7-diphenyl-1,10-phenanthroline as [the] electron transport layer (ETL)” (Liq-doped Bphen) and “[t]etrafluoro-tetracyano-quinodimethane doped 4,4',4''-tris(3-methylphenylphenylam[i]no)triphenylamine as hole transport layer (HTL)” (F₄-TCNQ doped *m*-MTDATA). Ma, Abstract.

62. Ma admits that “no work” had previously been done on Liq as a dopant for an ETL based on Bphen. *Id.* at 810.

63. Ma compares “[t]he conductivity of [those] carrier transport layers with different doping concentration[s].” *Id.*, Abstract. Ma’s results are then “[c]ompared with the referenced device (without doping)” to show a “current efficiency and power efficiency.” *Id.*

64. The specific devices fabricated in Ma are compared by varying the doping ratios “x” and “y” within the specific layered arrangements (Ma, 811):

	Complete Device
Anode	ITO
HTL	<i>m</i> -MTDATA: x wt.% F ₄ -TCNQ (40nm)
Interlayer	NPB (10nm)

Emission layer	Alq ₃ (20 nm)
ETL	BPhen:y wt.% Liq (50 nm)
EIL	LiF (1 nm)
Cathode	Al (130nm)

65. Ma repeatedly emphasizes the need for maintaining “efficient charge balance in the emission zone” for its reportedly improved efficiency. *Id.*, Abstract, 812 (“[t]he high carrier balance is reached in the emission zone, which leads to the enhanced current and power efficiency at low driving voltage”), 813 (“current efficiency” depends “on the carrier balance”), 814 (discussing “good balance” between electrons and holes with “efficient charge balance” in the emission layer). This “good carrier balance” is “one of the most important factors in getting excellent performance in OLEDs.” *Id.* at 814. In Ma, that balance is tailored to a fluorescent emission zone material using Alq₃ and the specific layer materials for the injection and transport layers surrounding that zone.

66. Alq₃ emission material was used in the seminal OLED reference, cited by Ma as reference 1 from 1987. As evidenced by Diekmann and Suzuki this material was no longer being considered for commercial grade OLED devices, having been surpassed in performance by phosphorescent emitters, but was rather a research grade material that was easy to work with and somewhat useful for researching basic OLED materials and device physics within the context of Alq₃-based devices. The charge balance, balance of energy levels, and conduction

requirements would be very different for a basic fluorescent monochromatic Alq₃ device compared to a state of the art, multi-emitter and phosphorescent device structure at the time of filing of the '425 Patent. In addition, the operating voltages described by Ma, which are a function of the device structure and charge injection and transport layers, are too high to be relevant to any practical OLED device application since they represent a significant efficiency loss.

67. Ma's results focused on a "33 wt.% ratio" of Liq to BPhen for the electron transport layer with a "0wt.%, 0.3wt.%, 2wt.% and 4wt.%" ratio of F4-TCNQ to *m*-MTDATA for the hole transport layer. *Id.* at 810-811, 814. Ma makes no general statement about doping any other materials besides BPhen with Liq or with different layers besides fluorescent Alq₃ as the emission zone materials.

VI. A POSITA WOULD NOT HAVE USED LIQ AS A ETL DOPANT IN DIEKMANN OR SUZUKI

68. I have been asked to comment on Samsung's position that a POSITA would "use 8-hydroxyquinolinolato-lithium as [a] dopant" in either Diekmann's or Suzuki's electron transport layers based on Ma (EX1018) at the time of filing the '425 Patent. Pet. 36-38, 69-70.

69. To summarize, I disagree that a POSITA—as defined by Samsung and Dr. Holmes—would have been motivated to combine either Diekmann or Suzuki with Ma and reasonably expected success in doing so at the time of filing the '425 Patent.

70. I express no opinion on whether Ma qualifies as prior art to the '425 Patent. For the purposes of this analysis, I merely assume Ma is prior art to the '425 Patent. Prior to this proceeding, I have not seen Ma (EX1018), despite being active in the OLED industry at the time of its alleged publication.

71. At a high level: (1) Samsung's stated reasoning does not explain why a POSITA would use Liq with either Diekman's or Suzuki's ETL with a reasonable expectation of success; (2) Ma does not provide a general motivation to dope every ETL with Liq at the time of filing the '425 Patent; (3) the use of Liq as a dopant was experimental and only shown to operate in a BPhen matrix with an Alq₃ emissive layer—a known stack for a fluorescent OLED device; and (4) there was unpredictability and technical and compatibility issues with attempting to use an unproven dopant. Therefore, a POSITA, particularly as described by Samsung and Holmes, would neither have doped Diekmann's or Suzuki's phosphorescent OLED ETL layers with Liq nor reasonably expected success in doing so.

A. Samsung and Dr. Holmes Provide No Reason for Why a POSITA Would Use Liq in Diekman's or Suzuki's ETL With a Reasonable Expectation of Success

72. For ease of reference, I copy Samsung's entire analysis of Ma:

Ma teaches an OLED with an electron transport layer doped with 8-hydroxy-quinolinatolithium ("Liq"). Ma, Abstract, 810-811.

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.*

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.

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.

73. For Suzuki, Samsung similarly contends:

It would have been obvious to a POSITA to use 8-hydroxyquinolinolato-lithium (“Liq”) as the dopant in Suzuki’s n-doped electron transport layer which, as discussed above in §VIII.B.1, is taught by Ma. Ma, Abstract, 810-811; Ex.1003, ¶224.

A POSITA would have been motivated to use an Liq-doped electron transport layer of Ma in the organic layer stack of Suzuki due to the known benefits of an Liq- doped electron transport layer, discussed above in §VIII.B.1. Ex.1003, ¶¶32-42, 225-228. 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.

74. Samsung's only evidence that there were "known" benefits for a Liq-doped electron transport layer is its expert, Dr. Holmes. Pet. 37-38, 70. Dr. Holmes, however, merely repeats Samsung's argument without support or further explanation:

The use of Liq-doped electron transport layers in OLEDs was a well-known practice in the art at the time, and had well-known benefits. See §V.A.4 above.

See EX1003, ¶¶140, 145, 225. I note that Dr. Holmes in these sections does not explain what the purported "well-known benefits" were in doping an ETL with Liq in this section beyond a vague reference to "improve the performance" and, in his later discussion, "achieving ... efficiency." EX1003, ¶¶ 42, 140, 225.

75. In the cited background discussion from Dr. Holmes in Section V.A.4, the only mention of using Liq as a potential dopant in any ETL was from Ma's doping Liq into Bphen:

42 ... In a 2008 paper, Ma et al. similarly used doped transport layers to improve the performance of an OLED, by using a p-doped hole transport layer (doped with p- dopant F4-TCNQ) and an n-doped electron transport layer (doped with n-dopant Liq). Ex. 1010 (Ma). Ma et al.'s work shows that the community was also actively working

on molecular dopants containing lithium (for example, Liq) to increase the conductivity of electron transporting layers in OLEDs.

76. But citing only to Ma does not show that Ma's supposed teachings of were "well known," "community" work before the filing of the '425 Patent, as Dr. Holmes states. On the contrary, Ma acknowledges in its introduction that "[n]o work has been reported about using Liq-doped BPhen as electron transport layer." Ma at 810.

77. For Diekmann, Samsung and Dr. Holmes cites Liu (EX1018) as evidence of Liq's "well-known ... electron injection and transport properties." Pet. 37; EX1003, ¶144. There is no pin-cite or explanation of this citation. I disagree that Liq had well-known electron injection and transport properties as a dopant at the time of filing and see no evidence to the contrary in the record. To illustrate, Liu was discussing using Liq as an electron injection layer, not a ETL dopant:

Lithium quinolate (Liq), covered with aluminium, ***was used as an electron injection layer*** in a double layer organic light emitting diode consisting of NPD as the hole transport layer and Alq as the emitting layer resulting in lower turn on voltage and increased power efficiency. The driving voltage required to achieve a luminance of 100 cd/m decreased from 5.8 V for the Ca/Al to 4.2 V when Liq/Al was used, improving device power efficiency from 2.3 to 4.1 lm/W. The performance tolerance to layer thickness of Liq devices is also better than that of the devices with lithium fluoride (LiF). Due to the highly insulating nature of LiF, it can only be used when deposited as an ultra-thin layer, while the Liq can be deposited into layers as thick as 5 nm without significantly affecting the EL properties. 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.

EX1018, Abstract. Like Ma (Ma, 810), Liu also notes that Liq “has not yet been widely investigated.” EX1018 at 211.

78. And his conclusion that a “POSITA thus would have recognized that Liq would be an advantageous dopant for Diekmann’s doped layer” lacks explanation for why a POSITA would have specifically used Liq. EX1003, ¶ 144. As I explain below, a POSITA would not have used Liq as an ETL dopant in Diekmann or Suzuki.

79. While Dr. Holmes states that “[a]t the time, it was well understood that dopant metal ions could readily diffuse in the OLED during film growth and operation, compromising performance” and that “[a] larger, molecular dopant would be less able to diffuse in the transport layer, reducing the severity of these issues,” he cites no support for this statement. EX1003, ¶ 144.

80. While Diekmann states “the dopant preferably has a low diffusion constant within the organic layer stack, as is usually the case with large atoms or molecules, for example,” (EX1003, ¶ 144 (quoting Diekmann, 10)), Diekmann’s preferred dopants are cesium, barium, and LiF. Each of these materials has a low diffusion and sufficient size to be preferred in Diekmann:

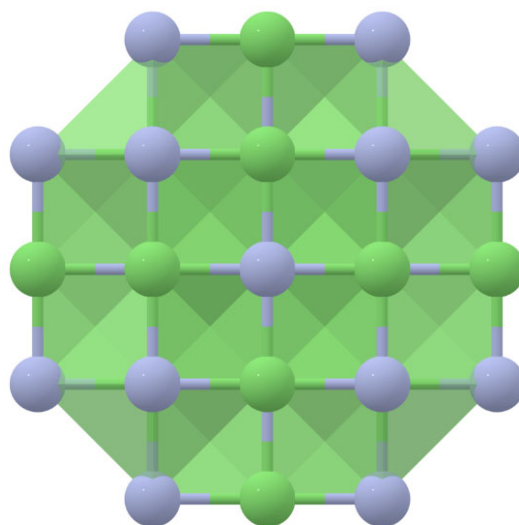
Diekmann 10: Preferably, the dopant of the doped layer is as large as possible atoms or molecules, which in the case of an n-dopant are suitable for emitting electrons ... Furthermore, the dopant preferably has a *low diffusion constant* within the organic layer stack, as is usually the case with *large atoms or molecules*, for example. ... *Preferred n-dopants are cesium, barium and lithium fluoride.*

Diekmann 34: ... The outermost organic layer is doped with a dopant. Preferably, the dopant of the doped layer - as described above - is as *large as possible atoms or molecules*, which are suitable for emitting electrons in the case of an n-dopant and for emitting holes in the case of a p-dopant. Furthermore, the dopant preferably has a *low diffusion constant* within the organic layer stack, as is usually the case with large atoms or molecules, for example. *Cesium, for example, is a particularly suitable dopant*

Diekmann 119: The outermost organic layer is doped with a dopant 410. Preferably, the dopant 410 of the doped layer comprises – as explained above - atoms or molecules which are as large as possible and which are suitable for emitting electrons - n-dopant - or holes - p-dopant. Furthermore, the dopant has a low diffusion constant within the organic layer stack 4. For this purpose, the dopant is formed from the largest possible atoms or molecules. Cesium, for example, proves to be a suitable dopant.

Diekmann, 10, 34, 119; *see id.*, 91 (“The dopant 410 can be for example cesium, barium or lithium fluoride.”).

81. Cesium and barium are large row 6 main group metals. And LiF, although composed of the smallest metal cation Li^+ and smallest anion F^- , has a cubic crystal structure in which Li^+ is bonded by six equivalent F^- to form a mixture of corner and edge sharing LiF_6 octahedra as shown below. This reduces the diffusion rate of Li^+ and F^- .



82. While Liq might have a larger molecular weight, that does not mean that Liq would have been recognized by a POSITA at the time as suitable as a dopant with low diffusion rate in Diekmann's OLED device over cesium, barium, and LiF. For instance, there is no explanation why the small Li^+ cation in Liq would not dissociate from the hydroquinolate anion and diffuse through the organic layers to wreak havoc on the device. Neither Samsung nor Dr. Holmes provide any explanation for why a POSITA would use Liq in lieu of these already low-diffusion constant, large atom/molecule dopants already in Diekmann.

83. In addition, Diekmann specifies three materials for its electron conducting layer:

For example, the electron-conducting layer contains or consists of at least one of the following materials:

- Alq_3 = Tris(8-hydroxyquinolin)aluminium

- BALq = Bis-(2-methyl-8-chinolinolate)-4-(phenylphenolato)aluminium

- TPBi = 1,3,5-Tris-(1-phenyl-1H-benzimidazol-2-yl)-benzol.

Diekmann, 5. Diekmann neither suggests doping Alq₃, Balq or TPBi nor suggests replacing these ETL materials with BPhen. I have seen no reason offered by Samsung or Dr. Holmes to substitute Bphen in Diekmann's system.

84. Finally, for an expectation of success, Samsung and Dr. Holmes conclude that using Liq as a dopant in an OLED electron transport layer was a "routine" with a "high probability of success" given Diekmann and Suzuki contemplated an "n-doped electron transport layer." Pet. 38, 70 (citing EX1003, 145-146, 225-228). I disagree. There was nothing routine about using a novel, untested dopant in an entirely different system, as I explain below. And Samsung does not explain why a POSITA, with their low level of skill, would expect success in using *Liq* specifically as a dopant in either Diekmann or Suzuki. Dr. Holmes similarly provides no explanation for how attempting to use a new, specific Liq dopant in a different host in a different device structure than Ma is "routine" with a "high probability of success" for his definition of a POSITA. EX1003, ¶¶ 145, 228

85. As explained below, using a novel and unproven Liq dopant on a different OLED stack for a different emissive material would have hardly been

routine for such an ordinary skilled artisan and Samsung and Dr. Holmes provide no evidence on why a POSITA would have replaced the listed dopants in Diekmann with Liq or why Liq would be used in Suzuki as an impurity dopant in Suzuki's already high-n nature ETL.

B. Ma Does Not Provide a Motivation to Dope Every ETL with Liq

86. In my opinion, no POSITA would have understood Ma to teach doping every ETL in every OLED stack with Liq for “performance,” “efficiency,” or any other reason as Samsung offers.

87. While Samsung and Dr. Holmes both cite Ma's background statement that there was an “interest in developing OLEDs with high efficiency.” Pet. 37 (quoting Ma at 810); EX1003, 140 (same), 225 (same). This interest could only be taken within the context of improving the very low efficiency of Alq₃ emitter-based devices, which have low internal quantum efficiency and high voltages which result in undesirable monochromatic device operation that is much less efficient than state of the art OLED structures. In addition, this says nothing about using *Liq* as a general ETL dopant. Nor does this show that using Liq as an ETL dopant in Diekmann's or Suzuki's OLED stack would have been reasonably expected to result in high efficiency in an entirely different system.

88. To a POSITA, Ma narrowly describes an improvement to a known, stable, and undoped fluorescent OLED stack:

Compared to the referenced device (without doping)....

1. Introduction

...

In this paper, we have demonstrated a device with an new n-doping (33wt.% 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 significant improvement in the device performance *as compared with the referenced device*.

2. Experiments

The structure of the complete devices fabricated in this work is as follows:

ITO/*m*-MTDATA:*x* wt.% F₄-TCNQ (40nm)/NPB (10nm)/Alq₃ (20 nm)/ BPhen:*y* wt.% Liq (50 nm)/LiF (1 nm)/ Al (130 nm). *Device A (referenced device)*: *x* = 0, *y* = 0; *Device B*: *x* = 0, *y* = 33; *Device C*: *x*=0.3, *y*=33; *Device D*: *x*=2, *y*=33; *Device E*: *x*=4, *y*=33.

In the devices, 4,4',4"-tris(3-methylphenylphenylamono) triphenylamine (*m*-MTDATA):*x* wt.% etrafluro-tetracyano-quino-dimethane (F₄-TCNQ)and Bphen:*y*wt.% Liq are used as p-doping HTL and n-doping (ETL), respectively. *N,N*-diphenyl-*N,N*-bis(1,1'-biphenyl)-4,4'-diamine (NPB) and Alq₃ are used as interlayer and emission layer (EML), respectively, while LiF and Al are used as electron injection layer and cathode, respectively.

Ma, Abstract, 810–811.

89. No POSITA would have understood Ma to suggest Liq is a panacea dopant for all OLED systems. Instead, a POSITA would have understood that Ma's improved performance is specific to the BPhen host and specific to this

specific OLED stack and only in comparison to a device with an undoped version of its OLED stack, as explained further below.

90. As Ma explains:

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).*

Ma, 810 (emphasis added).

91. From this passage, Ma is clear that “no work” had been previously reported on Liq doping an ETL in any form prior to Ma. Ma, 810. Ma admits it builds off prior efforts to “improve electron injection and transport” of doping an emissive layer, Alq₃, with lithium (Li) or Bphen. *Id.* Yet Ma admits that there were only a “few studies” about Liq even as a separate electron injection layer. *Id.* Samsung and Dr. Holmes ignore this key context.

92. A POSITA would understand only that Ma is describing doping Bphen with Liq alongside doping *m*-MTDATA with F₄-TCNQ surrounding its Alq₃ emission layer:

Anode	ITO
HTL	m-MTDATA (40nm)
Interlayer	NPB (10nm)
Emission layer	Alq3 (20 nm)
ETL	BPhen (50 nm)
EIL	LiF (1 nm)
Cathode	Al (130nm)

Ma, Abstract, 810-811. Ma specifically proposes heavily doping Bphen with Liq in this stack. *Id.*

93. Ma seeks to find the optimal dopant levels for correcting a known charge imbalance in this specific stack, testing power efficiency by comparing this prior undoped system to its dual doped variations.

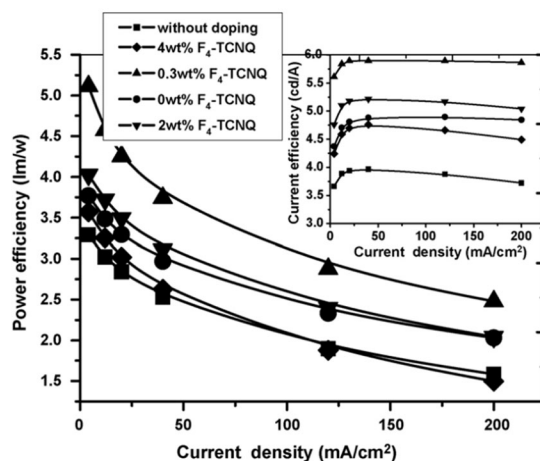


Fig. 3. Power efficiency vs current density characteristics of the complete devices.
Inset: Current efficiency – current density characteristics of the complete devices.

Ma, Fig. 3.

94. Ma's ultimate conclusion about performance enhancement is expressly tied, though, to the specific layer materials, widths, and doping

concentrations of its *m*-MTDATA and Bphen layers as used in this simplified, fluorescent Alq₃-based stack. To illustrate, Ma expressly concludes, based on this testing, that:

This significant enhancement in the device performance is attributed to the improved transport conductivity of both p-doped HTL and n-doped ETL at a particular doping concentration of F4-TCNQ into *m*-MTDATA and Liq into Bphen.

Ma, 812.

95. Ma tied the performance improvement to a specific synergy in the Liq and Bphen combination. Specifically, the “certain doping ratio of Liq to Bphen can improve the electron transport ability of the co-deposited layer” in Ma’s Alq₃-based stack:

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 along with 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 along with their LUMO are expected to exhibit a certain extent of overlap after a mixing ratio goes beyond 17wt.%. 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.

Ma, 813.

96. A skilled artisan would have understood that the performance enhancement is thus tied to the specific layers with the specific dopants in an Alq₃-based fluorescent stack. A POSITA with merely a bachelor's degree in physics or chemistry and 3 years of industry experience would not read Ma any broader.

97. Contrary to Dr. Holmes, Ma does not describe that “current and power efficiency of an OLED can be increased at a low driving voltage by utilizing an ETL which is doped with Liq” or that doping with Liq in the ETL “results in increased current density and reduced driving voltage, compared to the undoped OLED.” EX1003, ¶¶ 142-143, 225-226 (citing Ma, 812-813). I do not see such general propositions for doping any ETL with Liq anywhere in Ma.

98. Ma also states that its doping was to correct a hole surplus in its known, undoped device:

For the stability of the devices, we have known that in un-doping devices, such as *Device A* [(the reference device)], the number of holes is much more than electrons. So, a surplus of holes at the NPB/Alq₃ interface increases the probability that Alq₃ cations are formed, which are known to be unstable and lead to rapid device degradation.

Ma, 814. This is why Ma proposed doping its Bphen layer with Liq and its *m*-MTDATA layer with F₄-TCNQ—to achieve a “good balance” between holes and electrons and “decrease the surplus of holes”:

[I]n this paper, we introduce a p-i-n devices with a novel n-doping (33wt.% Liq:BPhen) layer as ETL and a p-doped (0wt.%, 0.3wt.%, 2 wt.% and 4 wt.% F₄-TCNQ:m-MTDATA) layer as HTL, in which

the holes and electrons have a good balance seen from Fig. 6, leading to the decrease in surplus of holes.

Id. I have seen no evidence that Diekmann or Suzuki has that hole imbalance to begin with.

99. To illustrate, unlike Ma, there is no NPB/Alq₃ interface in Diekmann. For instance, in Diekmann, the hole conducting layer (such as aNPB, *see* Diekmann at 4) is separated from the electron transport layer (such as Alq₃, *see* Diekmann at 5) by at least the emissive layer:

The organic layer, which is suitable for generating light, is preferably arranged between the hole-conducting layer and the hole-injecting layer on one side and the electron-conducting layer and the electron-injecting layer on the other side. As a rule, the organic materials are translucent, in particular for light emitted by the organic layer stack.

Diekmann, 4; *see generally* EX1003, Fig. 3(a), ¶ 34 (Dr. Holmes illustrating and discussing OLED layers). A skilled artisan would understand that Ma's observation about preventing a hole surplus at the NPB/Alq₃ interface cannot be readily applied to Diekmann as there is no NPB/Alq₃ interface and BPhen was not even mentioned as a potential ETL matrix material. *See* Diekmann at 5 (listing Alq₃, BAlq and TPBi as the ETL material).

C. The Use of Liq as an ETL Dopant Was Experimental for Limited Use in Stable, Fluorescent OLED Devices with Bphen at the Time

100. Neither Diekmann nor Suzuki mention using Liq as an ETL n-dopant or otherwise. This is because Liq as an ETL dopant was experimental at the time.

101. As Ma's limited use shows, Liq was only starting to be tested in single, very specific and known fluorescent Alq₃-based OLED stack as a dopant for Bphen. Given the early stages of this research into Liq, a POSITA armed with only a bachelors in physics or chemistry and three years of experience would not and could not just swap materials or dopants into an entirely different stack with a reasonable expectation of success. That skilled artisan simply lacks the experience and expertise to do so and such efforts would require significant experimentation and a large team of scientists with far more advanced experience to even being to accomplish.

102. Ma admits that "no work" had previously been done on Liq as a dopant for a Bphen ETL. Ma, 810. This is because, at the time of filing the '425 Patent, the research into using Liq in OLED devices was nascent and focused primarily on Liq as an injection layer material in legacy fluorescent OLED stacks, not as an ETL dopant in phosphorescent OLED devices:

However, few studies have been reported about using lithium quinolinato complex such as 8-hydroxy-quinolinato lithium (Liq) as an electron injection layer, and no work has yet been reported about using Liq-doped BPhen as electron transport layer (ETL).

Ma, 810

103. In 2011, Kumar recognized that "the reason for the enhanced electron transport property" of Liq in Bphen and Alq₃ "was not well understood" or why such high doping (around 30%) concentrations were needed of Liq in the prior

research. EX2004 (Kumar) at 1-2. Indeed, Kumar recognized that Liq had only

“recently” been used as a dopant of Bphen and Alq₃ electron transport materials.

Id. at 1. Thus, Kumar states, “n-type doping of organic semiconductors still

remains a challenge.” *Id.* In other words, n-type doping of organic semiconductors

was and remains unpredictable. As Kumar states:

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 materials 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, 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). But the reason for the enhanced electron transport property of these molecules was not well understood, especially the requirement of high doping concentrations (around 30%) in these molecular systems.***

EX2004 (Kumar) at 1-2. That challenge is exactly why, years before Kumar, no

POSITA with only bachelor’s degree and three years of experience would have

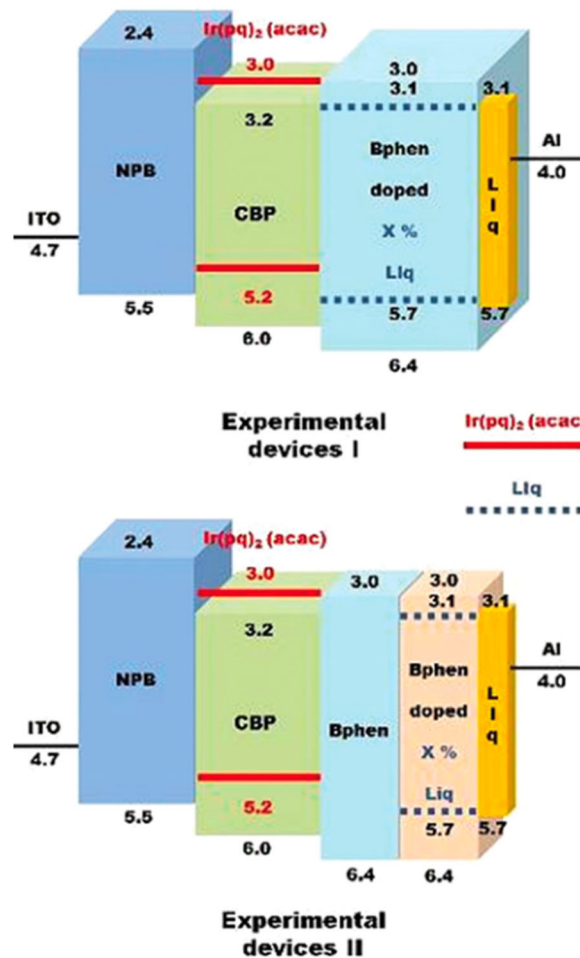
been motivated to or reasonably expected success in doping the ETL layers of

Diekmann and Suzuki.

104. Also in 2011, Kim experimented with a wide range of doping (5% to 65%) Bphen and multiple configurations of the ETL. EX2003 (Kim 2011),

Abstract, 59. While Kim achieved some positive results, Kim also reported concerns of Liq having “poor electron transportation properties” as doping increased passed 15% (at 57), that “heavy doping of Liq (15%~50%) causes unbalanced holes and electrons in EML due to excess or insufficient electrons and decreases the device efficiency” (at 58).

105. For reference, Kim created two experimental devices:



EX2003 (Kim 2011), Fig. 3. As Kim explains:

The device configuration was ITO=NPB (50 nm)=4,4'-N-N'-dicarbazole-biphenyl (CBP) doped with 8% bis(2-phenylquinoline)iridium(III) (acetylacetonate) [Ir(pq)₂(acac)] (30nm)=Bphen doped with X% Liq (30nm)=Liq (2nm)=Al (100nm), where the doping concentration of Liq in ETL was varied as 5%~65%.

...

The doping ratio of the Liq was controlled to range from 5%~[5]0% in experimental devices I. The current density was increased according to the increase of the doping ratio of Liq, ***but it was decreased by more than 15% because of the poor electron transportation properties of the Liq.*** However, the current density of all experimental devices I is higher than that of the reference device. It proves that the Liq doped in the ETL improves the driving voltage of device. It is assumed to be due to good interfacial contact between the cathode and the ETL when the Liq was doped in the ETL.

...

Although the electron injection into the device is the most efficient in the device with the 15% Liq, the maximum efficiency value was obtained in the device with a doping ratio 5% Liq because the recombination of holes and electrons is well balanced in the 5% Liq doped device. Moreover, ***heavy doping of Liq (15%~50%) causes unbalanced holes and electrons in EML due to excess or insufficient electrons and decreases the device efficiency compare with 5% Liq doped device.***

EX2003 (Kim 2011), Abstract, 57-58.

106. As Oh observed in 2015, the prior documented doping of an ETL with Liq was conducted primarily on ***fluorescent*** OLEDs. EX2005 () at 34 (“In those works listed above, the Liq dopant was applied as the dopant for electron transport layer of fluorescent OLEDs.”). Yet, as Oh reports, in a phosphorescent OLED, an Liq dopant was expected to act as a “luminance quencher” because of the low

triplet energy of Liq (2.0 eV). *Id.* at 34 (“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.”). Work arounds in 2015 required an exciton blocking layer between the n-doped layer and the emitting layer so as to attempt to block triplet exciton quenching. *Id.* at 34-35 (“Alternatively, exciton blocking layer was inserted between the n-doped layer and emitting layer to block triplet exciton quenching of emitting materials.”). But that would increase the driving voltage due to the low electron mobility of the non-doped exciton blocking layer. *Id.* at 35 (“However, this approach increases the driving voltage of the device due to low electron mobility of the non-doped exciton blocking layer.”).

107. The full passage from Oh is copied below:

Cs₂CO₃, CsN₃, and LiF were reported as n-type doping materials and increased the conductivity of an electron transport layer, decreasing the driving voltage of OLEDs. However, the ionic compounds quenched the excitons in the emitting layer and had negative effect on the quantum efficiency of OLEDs. Therefore, organometallic complexes such as 8-hydroxyquinolinatolithium (Liq) were utilized as a molecular dopant to reduce the driving voltage. 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 (Oh 2015) at 34-35. Neither Samsung nor Dr. Holmes address quenching or propose using any such exciton blocking layer to prevent quenching in Diekmann or Suzuki.

108. The industry is still trying to figure out just how Liq works as a dopant. In a 2022 paper by Kang (EX2006) investigated the “effect of coordinated structures of electron transporting materials (ETMs) with lithium quinolate (Liq) on charge transport characteristics.” EX2006 (Kang 2022), Abstract, 1 (“[T]he purpose of our theoretical investigation [is] to understand the effect of ETL-Liq complexation on the charge transport property.”). Again, while Kang reported some positive results, Kang also concluded that “charge transport” was actually “retarded” and that “co-deposition of Liq in an ETL solid matrix generally weakens the characteristics of hole/electron carrier transport via forming an ETL-Liq complex”:

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/TSP01-Liq was reduced.

...

Based on the comprehensive consideration of these results, it can be concluded that the co-deposition of Liq in an ETL solid matrix generally ***weakens the characteristics of hole/electron carrier transport via forming an ETL-Liq complex.***

Id., Abstract, 6.

109. And, even in 2023, reliance on Ma's approach of doping an ETL Bphen with Liq was still rare, as reported by Sasaki:

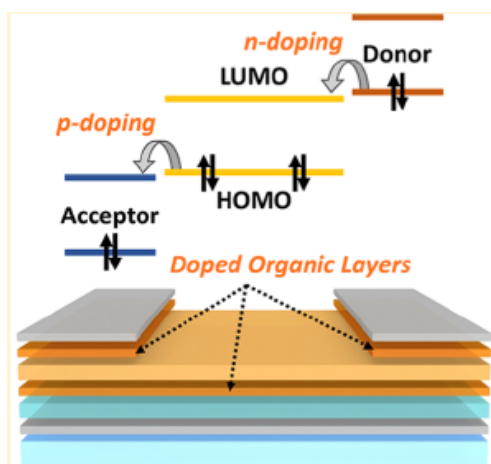
Although we fabricated a blue OLED with Liq used as the EIL and BPhen doped with Liq used as the ETL, its operating voltage was much higher than that of Device-3 (Figure S1, Supporting Information). Actually, ***OLEDs fabricated using BPhen doped with Liq as the ETL have rarely been reported.***

EX2007 (Sasaki 2023) at 1.

110. Moreover, Ma describes increased conductivity of the Bphen transport layer due to "electron hopping among BPhen and Liq sites." Ma, 813. This "hopping" mechanism differs from the n-doping mechanism described in the '425 patent of an "electron donor," which improves the conductivity of the electron transport layer by increasing the number of electrons. '425 patent at 26:61-62. Diekmann's dopant of its injection layer is as an "electron donor" (Diekmann, 91)

and similarly focused on materials “suitable for emitting electrons” (*Id.*, 10, 34, 119), not electron hopping. This is likely why Ma required such heavy Liq doping. No POSITA would turn to a heavy dopant using an unproven transport mechanism (hopping) as in Ma over a simpler electron donor dopant that operates at significantly lower concentrations.

111. Indeed, a POSITA would be skeptical as to whether Liq is even an n-dopant as in the '425 Patent as Liq is described by Ma. Generally, a molecular dopant n-dopes an ETL by transferring electrons from the dopant's HOMO to the ETL's LUMO, as illustrated below:



EX2009, Abstract.

112. The HOMO of Liq is 5.8eV (<https://www.ossila.com/products/8-quinolinolato-lithium>) while the LUMO of BPhen—according to Ma—is about 3.0eV. A POSITA would have regarded the 2.8eV energy gap as too much for any electron transfer from n-doping to occur from Liq to BPhen. Thus, although Ma described BPhen as n-doped, Samsung and Dr. Holmes have not explained why

shortening the molecule distance between electron carrier sites to improve “hopping” is n-doping as opposed to modulating the electron mobility.

113. In short, the use of Liq as an ETL dopant at the time of filing the ’425 Patent was experimental and unpredictable and only demonstrated in Bphen in a fluorescent Alq₃ stack alongside an F₄-TCNQ-doped m-MTDATA layer. A POSITA would not have read Ma any broader.

D. Samsung and Dr. Holmes Do Not Account for the Differences Between Ma’s Alq₃-based Fluorescent Emission System Versus the Claimed Iridium-based Phosphorescent Emission Systems

114. A skilled artisan would understand that Iridium-based compounds are phosphorescent emitters whose emission involves a triplet excitation state. This differs from a fluorescent emitter like Alq₃ used by Ma, which involves only a singlet excitation state.

115. As previously explained, Liq, with its relatively low triplet energy potentially “act[s] as a luminance quencher in phosphorescent organic light-emitting diodes (PHOLEDs).” EX2005 (Oh 2015) at 34. This is because PHOLEDs (unlike fluorescent OLEDs) use triplet excitation for emission. Neither Samsung nor Dr. Holmes account for this difference and indeed ignores the difference between Iridium-based phosphorescent and Alq₃-based fluorescent emitters entirely.

116. Given the early stages of this research into Liq, a skilled artisan armed with only a bachelor's degree in physics and chemistry and three years of experience would not and could not just swap materials or dopants into an entirely different stack with a reasonable expectation of success. That level of POSITA simply lacks the experience and expertise to do so. Such efforts would require a large team of cross-disciplinary scientists with far more advanced experience to even being to accomplish.

117. At the time of filing the '425 Patent, the research into using Liq in OLED devices was nascent and focused primarily on Liq as an *injection* layer material in *fluorescent* OLED stacks, not as an ETL dopant in phosphorescent OLED devices. As Ma's limited use shows, Liq was only starting to be tested in single, very specific and known fluorescent Alq₃-based OLED stack as a dopant for BPhen. Ma, 810.

118. Ma admits that "no work" had previously been done on Liq as an ETL dopant in BPhen:

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).

Ma, 810. Indeed, in 2023, Sasaki reported that "using Bphen doping with Liq as the ETL" has "rarely been reported":

Although we fabricated a blue OLED with Liq used as the EIL and BPhen doped with Liq used as the ETL, its operating voltage was much higher than that of Device-3 (Figure S1, Supporting Information). ***Actually, OLEDs fabricated using BPhen doped with Liq as the ETL have rarely been reported.***

EX2007 (Sasaki 2023) at 1. Ma thus describes itself as the first use of Liq as a heavy dopant in its Bphen ETL in its fluorescent (Alq₃ emitter) OLED.

119. The use of Liq as a dopant remained almost exclusively on fluorescent OLEDs through at least 2015. EX2005 (Oh 2015) at 34. Yet, in that time frame and still years after the filing of the '425 Patent, “the reason for the enhanced electron transport property” of Liq in Bphen and Alq₃ and why such high doping (around 30%) concentrations were needed of Liq still “was not well understood.” EX2004 (Kumar 2011) at 1-2. “[N]-type doping of organic semiconductors still remain[ed] a challenge” even then. *Id.* And, even as recently as 2022, researchers were still trying to figure out just how Liq works. EX2006 (Kang), Abstract, 1 (investigating the “effect of coordinated structures of electron transporting materials (ETMs) with lithium quinolate (Liq) on charge transport characteristics.”).

120. Yet the embodiments relied on by Samsung in Suzuki and Diekmann are ***phosphorescent*** OLEDs, not fluorescent OLEDs. Pet. 26 (relying on Diekmann’s emissive layer of “TMM-004:Ir(ppy)₃(15%) layer (Irppy = fac-tris(2-phenyl- pyridyl)iridium complex)), 58 (Suzuki’s alleged Iridium complex is a

PHOLED); *see* EX1003, ¶¶ 31 (Iridium as a phosphorescent material), 38, 112 (“Ir(ppy)₃, which is an iridium-containing compound which functions as a green light-emitting phosphorescent dopant”), 196 (“Suzuki teaches that in the case of phosphorescent dopants, “iridium compounds are the most preferable.”); *see* EX1014 (Baldo 1999), 4. There is no iridium-containing emission layer in Ma’s fluorescent system. Instead, Ma uses a single layer of Alq₃—the oldest and most studied fluorescent emissive layer material in OLED. Samsung and Dr. Holmes make no effort to explain how or why a Liq dopant tailored for Alq₃ and Bphen with the device-specific charge balance challenges would work with either Iridium-based emitters or their other emissive materials in the stack with their different HOMO and LUMO levels as well as other properties.

121. OLED emission is known to be quenched by Li⁺ ions from using a Liq layer or a Liq bulk dopant in phosphorescent OLED devices. Whatever the benefits to a fluorescent Alq₃ OLED, in a phosphorescent OLED, a Liq dopant acts as a “luminance quencher” because of the low triplet energy of Liq (2.0 eV). EX2005 (Oh 2015) at 34 (Liq as a dopant “act[s] as a luminance quencher in phosphorescent organic light-emitting diodes (PHOLEDs).”). As Oh reports, there was still a need, long after the filing date, of a high triplet energy n-type doping material that suppress triplet exciton quenching in phosphorescent OLEDs. EX2005 (Oh 2015) at 35 (“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.”). Samsung ignores the quenching problem. But, more fundamentally, Samsung never explains why a POSITA would have used Liq as a dopant in Diekmann’s or Suzuki’s phosphorescent system with a reasonable expectation of success given these unsolved quenching concerns.

122. No POSITA at the time of filing the ’425 Patent would have been motivated to use this experimental and unexplored Liq dopant from a fluorescent Alq₃ stack in Diekmann’s or Suzuki’s entirely different phosphorescent OLED given its limited and experimental use with a reasonable expectation of success.

E. Samsung and Dr. Holmes Do Not Address the Delicate Energy-Level Balance Required for the Different Organic Layers in an OLED Stack

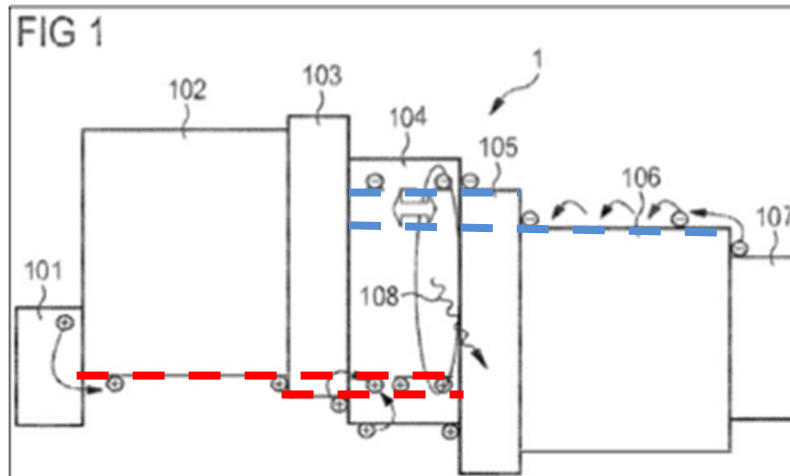
123. OLED stack layers and dopants are not modular, swappable components. There are significant difficulties in using unproven materials in a different OLED stack. To illustrate, the choice of which layers to use, the specific materials for each layer, the dopant and its concentration, if any, for each layer, the size of each layer, the layer order, and the choice of manufacturing processes (deposition, co-evaporation, temperature, and so forth) must be carefully chosen to maintain that balance that still create an operable OLED. Otherwise, there would be no “light emitting device” to qualify as the claimed OLED device that emits EM radiation in the claimed range.

124. For instance, maintaining the proper energy level balance between the layers is extremely difficult and necessary to solve before potentially expecting to build an operable OLED system that emits any electromagnetic radiation. Using the wrong materials, dopants, amount of doping, layer thickness, manufacturing processes, among many other variables could end up preventing an operable OLED entirely. At its core, OLED device engineering remains somewhat of an art form subject to complex and unpredictable results.

125. For an operational OLED, the LUMO and HOMO levels of the OLED layers must be carefully engineered. As Dr. Holmes admits, efficient injection of electrons and holes requires “relative energy level alignment between adjacent layers of the organic stack as well as the electrical conductivity of each layer.” EX1003, ¶ 32; *see* EX2004 (Kumar 2011) at 1 (matching the “HOMO” value “of the donor molecule” to the “LUMO” value “of the host molecule.”).

126. This is accomplished by matching the LUMO and HOMO levels of the various OLED layers so that the electrons and holes can traverse from the cathode or anode, respectively, to the emissive layer. The consequence of misaligning the charge levels is that the OLED might be inoperable.

127. Figure 1 of the '425 Patent is an energy level diagram, showing the LUMO (blue) and HOMO (red) balance in the '425 Patent's stack:

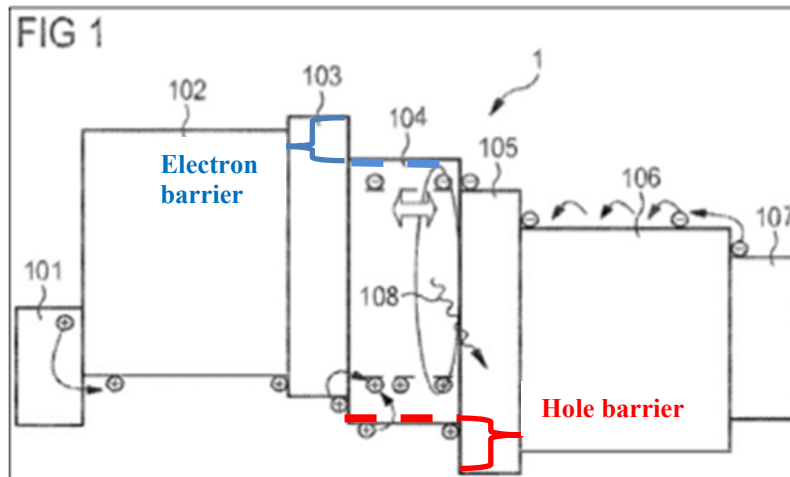


'425 Patent, Fig. 1 (annotated). The gap between neighboring levels must be sufficiently close to permit the injected electrons and holes to move (transport) from the electrode toward the emissive layer. Yet, at the same time, a charge balance must also be maintained to prevent holes from crossing into the ETL and to prevent electrons from passing into the HTL to avoid quenching and degradation of the OLED.

128. The HOMO and LUMO levels on the other side of the emissive layer must be sufficiently lower than the hole (p) or electron (n) side to avoid the holes entering the electron side or the electrons entering the hole-side. *E.g.*, EX2003 (Kim 2011) at 54 (higher mobility of holes results in “non-emissive cationic species” in the emission layer, obstructing “hole-electron recombination” and forming “dark spots, luminance decay[,] and subsequent low efficiency”), 56 (depicting charge levels for Liq doping), 57 (Liq has “poor electron transportation properties”), 58 (“heavy doping of Liq (15%~50%) causes unbalanced holes and

electrons in EML due to excess or insufficient electrons and decreases the device efficiency.”); EX1029 at 6.

129. The ’425 Patent’s embodiment below illustrates this by the large band gaps on the electron blocking layer (103) and hole barrier layer (105):



’425 Patent, Fig. 1 (annotated). The result is that there is only a narrow range of permissible LUMO and HOMO levels for each layer and dopant to create an operable OLED. Adding a Liq as dopant—especially as a heavy 30%+ dopant as in Ma—would greatly throw off this delicate charge balance in Diekmann and Suzuki.

130. Ma repeatedly emphasizes this need for maintaining “efficient charge balance in the emission zone.” Ma, Abstract, 810 (“[t]he high carrier balance is reached in the emission zone, which leads to the enhanced current and power efficiency at low driving voltage”), 813 (“current efficiency” depends “on the carrier balance”), 814 (discussing “good balance” between electrons and holes with “efficient charge balance” in the emission layer). This “good carrier balance” is

“one of the most important factors in getting excellent performance in OLEDs.” *Id.* at 814.

131. Significant work continued well after the filing date in determining the correct energy band levels to dope materials with Liq for an operable system even when just based on Bphen as in Ma. EX2003 (Kim 2011). Abstract, 59 (testing multiple configurations of an ETL with doping concentrations of 5-65% of Liq in Bphen, reporting efficiency points as low as 5% and as high as 65% depending on configuration). Samsung makes no effort to explain how this would even be reasonably expected to work within the LUMO and HOMO level constraints of the different neighboring layers in Diekmann or Suzuki.

F. Samsung and Dr. Holmes Offer No Evidence that Diekmann and Suzuki Would Have Gained Any Benefit from Using a Liq-doped ETL

132. Ma’s performance enhancement due to Liq was to correct a known hole surplus.

For the stability of the devices, we have known that in un-doping devices, such as *Device A* [(the reference device)], the number of holes is much more than electrons. So, a surplus of holes at the NPB/Alq3 interface increases the probability that Alq3 cations are formed, which are known to be unstable and lead to rapid device degradation. However, in this paper, we introduce a p-i-n devices with a novel n-doping (33wt.% Liq:BPhen) layer as ETL and a p-doped (0wt.%, 0.3wt.%, 2 wt.% and 4 wt.% F4 -TCNQ:m-MTDATA) layer as HTL, in which the holes and electrons have a good balance seen from Fig. 6, leading to the decrease in surplus of holes.”

Ma, 814. Samsung and Dr. Holmes do not contend Diekmann or Suzuki have such a surplus of holes to correct.

133. Without that hole surplus, Liq would have been expected to create an *electron surplus*, breaking charge carrier balance for generating excitons and degrading the emitter molecules. A skilled artisan would have expected that doping Liq into Diekmann and Suzuki as proposed by Samsung would thus only create charge balance problems. *See* EX2003 (Kim 2011) at 54 (charge imbalance leads to “dark spots, luminance decay[,] and subsequent low efficiency.”). This is contrary to Ma and the extremely important need to maintain the charge balance, as discussed in the prior section.

134. Again, Ma repeatedly emphasizes that need for maintaining “efficient charge balance in the emission zone” for its reportedly improved efficiency. Ma, Abstract, 810 (“[t]he high carrier balance is reached in the emission zone, which leads to the enhanced current and power efficiency at low driving voltage”), 813 (“current efficiency” depends “on the carrier balance”), 814 (discussing “good balance” between electrons and holes with “efficient charge balance” in the emission layer). This “good carrier balance” is “one of the most important factors in getting excellent performance in OLEDs.” *Id.* at 814. A POSITA, reading Ma, would not risk throwing off the carrier balance by creating an electron surplus. In

my opinion, Ma teaches away from Liq doping an ETL in an OLED device without a known hole surplus to correct.

135. Further, Samsung does not specify how much to dope the ETL layers in Diekmann or Suzuki and thus cannot show a reasonable expectation of success. This is not a scenario where more is better.

136. Ma illustrates this difficulty. Ma proposed *heavily* doping its BPhen layer with 33% Liq. Ma, Abstract. But Ma reported reduced conduction as the ratio increases to 50% Liq “attributed to the effect of carrier quench and generation of defects.” Ma, 813. Such heavy doping of Liq turns Liq into more of a host material given the significant amount used, leading to the effects of Liq as a bulk material to become more present. For instance, even 5 nm of Liq (as an electron injection layer), risked “significantly affecting the EL properties” similar to LiF’s instability and emission blocking. EX1018 (Liu), Abstract, 214 (“Liq layers should be under 5 nm to avoid “significantly affecting the EL properties”); *see generally id.* at 213 (“When the LiF thickness is increased to 1.0 nm, the EL efficiency deteriorates. When the thickness of LiF is 5 nm, the device is very unstable, and when it reaches 10 nm, the device does not give emission at all.”).

137. In 2011, the uncertainty in the even the amount of doping remained even for BPhen. *See* EX2003 (Kim 2011). Abstract, 59 (testing multiple configurations of an ETL with doping concentrations from 5-65% of Liq in Bphen,

reporting efficiency points as low as 5% and as high as 65% depending on layer configuration and thickness). While Kim achieved some positive results, Kim also reported concerns of Liq having “poor electron transportation properties” as doping increased passed 15% (at 57) and that “heavy doping of Liq (15%~50%) causes unbalanced holes and electrons in EML due to excess or insufficient electrons and decreases the device efficiency” (at 58). Samsung and Dr. Holmes do not mention the Liq doping concentration at all.

138. Samsung also ignores the significant technical barriers to phosphorescent OLED fabrication in even attempting to utilize a new, unproven dopant from a different OLED layer stack that are well-beyond the limited level of ordinary skill proposed by Samsung. Liq doping could “retard[]” and “weaken” charge transport, for example. EX2006 (Kang 2022), Abstract, 6 (theorizing the formation of an ETL-Liq complex). EX2003 (Kim 2011) at 54 (charge imbalance leads to “dark spots, luminance decay[,] and subsequent low efficiency”). Samsung does not explain how doping could be accomplished with Liq in Diekmann or Suzuki in a manner that would have been reasonable expected to work in Diekmann or Suzuki’s system without these significant problems.

139. I declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code.

140. Executed on October 28, 2024, in Ringgold, Georgia, USA.

A handwritten signature in black ink, appearing to read "P. Morgan Pattison", written over a horizontal line.

P. Morgan Pattison, Ph.D.

EXHIBIT A

P. Morgan Pattison
morgan@sslsinc.com
(805)217-3878

Experience

Solid State Lighting Services, Inc.

November 2007 - Present

Founder/President- Provide technical, strategic, and market expertise in the areas of LEDs, OLEDs, Lighting, Solid State Lighting (SSL), displays, and optoelectronic materials and devices. LED device, lighting, and materials product development. Horticultural lighting research and strategy. Light pollution measurement, evaluation, and mitigation. LED and lighting product failure and reliability evaluation. Manage research on human and animal physiological responses to light. Scout and evaluate new technologies for investment.

U.S. Department of Energy

January 2006 – November 2007

National Energy Technology Laboratory, Morgantown WV

Project Manager- Managed DOE funded R&D projects in SSL including LED and OLED lighting technologies. Managed solicitation and selection of new funded R&D projects. Identified priority areas of research necessary for advancement of SSL efficiency, performance, and price.

University of California, Santa Barbara

September 2000 – January 2006

Graduate Student Researcher- Doctoral thesis research under Professor Shuji Nakamura (Nobel Prize in Physics 2014) on GaN based micro-cavity light emitting diodes. Research included: epitaxial structure design and growth by MOCVD, semiconductor process development, materials characterization, and electro-optical device characterization. Additional areas of research included UV LEDs, vertical cavity surface emitting laser (VCSEL), and edge emitting laser diodes.

Cymer Laser, San Diego

November 1996 – May 2000

Laser Test Engineer- Tested and developed test methodology for production and prototype KrF and ArF excimer lasers used in deep UV photolithography for manufacturing of advanced semiconductor products. Testing included optical characterization and full system troubleshooting. Test development of new electrical and optical test methodologies to improve test cycle, Labview automation of test procedures, and development of test documentation.

Education

Ph.D. Materials Science, Electronic and Photonic Materials

University of California, Santa Barbara, 2006

Thesis: *Fabrication and Characterization of Gallium Nitride Based Micro-Cavity LEDs*

M.S. Electrical Engineering, Electronics and Photonics

University of California, Santa Barbara, 2003

B.S. Electrical Engineering, Electronic Devices and Materials

University of California, San Diego, 1996

Expertise

Optoelectronic materials and devices, lighting, LED, OLED, UV-LEDs, Germicidal UV application, R&D and technology evaluation, μ LED-LED-OLED-Quantum Dot display technology, medical lighting, SSL product evaluation and selection, LED lighting product development, R&D Management, technology roadmapping, technical meeting moderation, technical writing, IP evaluation, trial expert witness, lighting color evaluation and selection, advanced lighting performance evaluation, Energy Star LED lighting testing, photometric characterization and testing, R&D Management, Federal R&D funding processes, R&D Proposal review and evaluation, ESCO contracts and performance measurement & verification (M&V), horticultural lighting, wildlife responses to light, human physiological responses to light

Accreditations, Affiliations, and Professional Societies

Fellow - Illuminating Engineering Society (IES)

Chair – Outdoor Nighttime Environments (ONE) Committee

Voting Member – IES Horticultural Lighting Committee

Oregon State University, Civil and Construction Engineering, Courtesy Faculty

Lighting Certified (LC) by National Council on Qualifications for Lighting Professionals

Member: International Society of Horticultural Scientists (ISHS), International Dark-Sky Association (IDA), International Society for Optics and Photonics (SPIE), American Society of Agricultural and Biological Engineers (ASABE), American Physical Society (APS)

Project Management Professional (PMP) *Lapsed*, Project Management Institute (2006)

U.S. Government Contracting Officer's Representative (COR) (2006)

Leadership

Senior Technical and Strategic Advisor to DOE SSL R&D Program

Lead writer of DOE SSL R&D Opportunities, R&D Plan, and Multi-Year Program Plan

Technical content supervisor of annual DOE SSL R&D Workshops

Manage organization of DOE SSL topical R&D meetings on: LED devices and materials, OLEDs, luminaire integration, roadway lighting, germicidal ultraviolet (UV) LEDs, human physiological responses to light, domesticated and wild animal responses to light, roadway and area lighting, and horticultural lighting

New business creation/development –*Action Spectrum*, *Ultra-Yield Solutions*, *Guidance LED*, *Zuli*, *Canvas*, *Intellilight/Next*, *SSLs, Inc.*, *Diel Lighting Science*, *Blue Laser Fusion*

Scientific Advisory Board member to *GLASE (Greenhouse Lighting and System Engineering)*, NYSERDA funded controlled environment agriculture R&D program

Technical Advisory Committee of Resource Innovation Institute

Advisory Board Member of *Phosphor Global Summit* annual meeting

Advisor to *Cyclotron Road* – Clean Technology Incubator at Lawrence Berkeley National Lab

Project sponsor for Senior Design Course at UCSB (2011-2012)

1. Design and development of spectro-goniophotometer
2. Design and development of smartphone light controller (co-founder and original board member of resulting company renamed *Zuli*)

Designed, fabricated, and characterized world's first gallium nitride microcavity LED

Fabricated and characterized world's first gallium nitride semipolar LED

Publications

Minimizing Ecological Impacts of Marine Energy Lighting. Reilly, C. E., Larson, J., Amerson, A. M., Staines, G. J., Haxel, J. H., & Pattison, P. M. (2022). *Journal of Marine Science and Engineering*, 10(3), 354.

Department of Energy SSL Manufacturing Status and Opportunities (2022)

Photon Efficacy in Horticulture. Kusuma, P, Pattison, PM, Bugbee, B. (2021) Chapter 7 in *Plant Factory: Basics, Applications and Advances*

A Review of Human Physiological Responses to Light: Implications for the Development of Integrative Lighting Solutions. Vetter, C, Pattison, PM, Houser, K, Herf, M, Phillips, AJK, Wright, KP, Skene, DJ, Brainard, GC, Boivin, DB, Glickman, G. (2021) *Leukos*

DOE 2020 LED Manufacturing Supply Chain. Lee, K, Nubbe, V, Rego, B, Hansen, M, Pattison, M. *U.S. DOE Lighting R&D Program*. July 2021.

Recommended Practice: Horticultural Lighting, An American National Standard. ANSI/IES RP-45-21. *Illuminating Engineering Society*. June 2021.

Energy Savings Potential of SSL in Agricultural Applications. Lee, K, Elliott, C, and Pattison, PM. *U.S. DOE Lighting R&D Program*. June 2020.

From physics to fixtures to food: current and potential LED efficacy. Kusuma, P, Pattison, PM, and Bugbee, B. (2020) *Horticultural Research*. 7, 56.

What's next for LEDs. Pattison, PM (2019) *Electroindustry*. March/April 2019.

Building the evidence outside the lab. Pattison, PM (2018) *Lighting Design & Application*. December

LEDs for photons, physiology and food. Pattison, PM, Tsao, JY, Brainard, GC, Bugbee, B. (2018) *Nature*. 563, 493-499.

LEDs for Horticulture: More than Meets the Eye. Pattison, PM. (2018) *Lighting Design & Application*. August.

LED lighting efficacy: Status and directions. Pattison, PM, Hansen, M, Tsao, JY. (2018) *Comptes Rendus Physique*. 19, 134-145.

Energy Savings Potential of SSL in Horticultural Applications. Stober, K, Lee, K, Yamada, M, and Pattison, PM. *U.S. DOE Lighting R&D Program*. December 2017.

Department of Energy Solid State Lighting R&D Opportunities/Plan, lead author (2022, 2019, 2018, 2017, 2016, 2015)

Light-emitting diode technology status and directions: opportunities for horticultural lighting. Pattison, PM, Tsao, JY, Krames, MR. (2016) *Acta Horticulturae*. 1134, 413-426.

Status of SSL Product Development and Future Trends for General Illumination. Katona TM, Pattison PM, Paolini S. (2016) *Annual Review of Chemical and Biomolecular Engineering* 7 (1).

The Blue LED Nobel Prize: Historical context, current scientific understanding, human benefit. Tsao, J, Han, J, Haitz, R, Pattison, PM. (2015) *Annalen der Physik* 527, No. 5–6, A53–A61.

Department of Energy SSL R&D Multi-Year Program Plan (2008-2014)

Department of Energy SSL Manufacturing Roadmap (2009-2014)

Gallium nitride based microcavity light emitting diodes with 2 lambda effective cavity thickness. Pattison, P. M., David, A., Sharma, R., Weisbuch, C., DenBaars, S., & Nakamura, S. (2007). *Appl. Phys. Lett.*, 90, 031111.

First-moment analysis of polarized light emission from InGaN/GaN light-emitting diodes prepared on semipolar planes. Masui, H., Baker, T. J., Sharma, R., Pattison, P. M., Iza, M., Zhong, H., Nakamura, S., & DenBaars, S. P. (2006). *Japanese journal of applied physics*, 45(9L), L904.

Gallium nitride based micro-cavity light emitting diodes emitting at 498 nm. Pattison, P. M., Sharma, R., David, A., Waki, I., Weisbuch, C., & Nakamura, S. (2006). *Physica Status Solidi (a)*, 203(7), 1783-1786.

Demonstration of a semipolar (10 (1) over-bar (3) over-bar) InGaN/GaN green light emitting diode. Sharma, R., Pattison, P. M., Masui, H., Farrell, R. M., Baker, T. J., Haskell, B. A., Wu, F., DenBaars, S.P., Speck, J.S., & Nakamura, S. (2005). *Applied Physics Letters*, 87(23).

Micro cavity effect in GaN-based light-emitting diodes formed by laser lift-off and etch-back technique. Fujii, T., David, A., Schwach, C., Pattison, P. M., Sharma, R., Fujito, K., Margalith, T., DenBaars, S.P., Weisbuch, C., & Nakamura, S. (2004). *Japanese journal of applied physics*, 43(3B), L411.

292 nm AlGaIn single-quantum well light emitting diodes grown on transparent AlN base. Hanlon, A., Pattison, P. M., Kaeding, J. F., Sharma, R., Fini, P., & Nakamura, S. (2003). *Japanese journal of applied physics*, 42(6B), L628.

Higher efficiency InGaIn laser diodes with an improved quantum well capping configuration. Hansen, M., Piprek, J., Pattison, P. M., Speck, J. S., Nakamura, S., & DenBaars, S. P. (2002). *Applied physics letters*, 81(22), 4275-4277.

Presentations

2024 Joint CEA Workshop, U.S. Fish & Wildlife Service Light Pollution & Bird Collisions expert panel, Workshop on Disorder in Semiconductor Physics and Devices 2024, University of Cincinnati IRIS 2023 Working Group, IES Raleigh Section “Horticultural Lighting – A Growing Opportunity”, IES Street and Area Lighting Conference 2022, National Park Service Science and Technology – Skyglow Measurements at Channel Islands National Park 2022, UCSB Nitride Seminar 2022, International SSL Alliance 2021 General Assembly, The Limits of Food Production – Vertical Farming 2021, Horticonn 2020, U.C. Davis Energy and Efficiency Institute,

California Energy Administration, 2020 DOE Lighting R&D Workshop, Charleston Catholic High School STEM Seminar, East Tennessee State University Physics and Astronomy Seminar, IES Rocky Mountain Section – CU Boulder, 2019 DOE SSL R&D Workshop, Harvard ES-173 Guest Lecture, U.S. DOE Building Technologies Office Brown Bag Seminar, U.C. Santa Barbara Nitrides Seminar 2018, Phosphor Global Summit 2018, West Virginia State University – Biology Seminar, East Tennessee State University Entrepreneur Club 2018, IES Aviation Lighting Committee Meeting 2017 (Best Technical Paper), ICCEA Panama 2017, NCERA 101 Controlled Environment Agriculture meeting, DOE SSL R&D Workshop 2017, The College of New Jersey Physics Colloquium, DOE SSL R&D Workshop 2017, NREL 2016, Harvard University Engineering Seminar, LightSym 2016, DOE SSL R&D Workshop 2016, DOE SSL Technology development Workshop 2015, DOE SSL R&D Workshop 2015, IDTechX Internet of Things Applications, Phosphor Global Summit 2014, International Conference on Nitride Semiconductors 2013, IES Illuminate Philadelphia 2012, Strategies in Light 2012, Energy Efficiency Expo 2011, Tennessee Association of Physical Plant Administrators 2011, East Tennessee State University Faculty Development Sustainability Seminar 2011, LED Discovery 2010, Think Tomorrow Today 2008, International Conference on Nitride Semiconductors 2005, International Symposium on Blue Lasers and Light Emitting Diodes 2004, Fall Materials Research Society 2001.

Patents

Technique for growth and fabrication of semipolar (Ga, Al, In, B) N thin films, heterostructures, and devices. #9793435, #9231376, #8686466, #7846757, #10529892

Lighting unit having lighting strips with lighting elements and a remote luminescent material. #8491165

Lighting unit with heat dissipating chimney #8360607

Lighting unit with indirect light source #8684566 B2

System for monitor and control of equipment. #9432210

Semiconductor light emitting device #8227820, #8643036

Etching technique for the fabrication of thin (Al, In, Ga) N layers. #7795146

Semiconductor micro-cavity light emitting diode. Application # 11/510,240 (abandoned), WO2007025122