

Organic Light Emitting Devices for Lighting

by Joseph J. Shiang

Lighting is one of the oldest electronics industries, and has been characterized throughout its history by its adoption and extension of new technologies. Most recently, solid-state lighting (SSL) sources have become available and have the potential to offer better energy efficiency than any other lighting technology. These potential energy efficiency gains are predicated on the assumption that solid-state devices will have efficient electron to photon conversion (> 0.9), operate at low voltage ($< 3.5\text{V}$), exhibit high light extraction efficiencies (> 0.7), and can be produced at a low enough cost to satisfy the general lighting market.¹

There are two different approaches for SSL: semiconductor light emitting diodes (LEDs) and organic light emitting devices (OLEDs). Both of these approaches have their origins in the 1960s and 1970s with the first OLED lighting device patented in 1976² (Fig. 1). This article will focus on some of the progress made in developing OLED technology for lighting applications. The performance of white light OLEDs has greatly increased, from ~ 4 lumens per watt (LPW) in 2001 to 100 LPW.³

Furthermore, it appears that the first two requirements necessary to achieve high efficiency lighting—efficient electron-to-photon conversion⁴ and low voltage operation—have been largely achieved in OLEDs. Thus, this article will not be a review of these topics but will discuss some technical developments relating to the development of practical OLED light sources that have high light extraction and can be manufactured at low cost.

Current Status of OLED-based Lighting

Many of the fundamental characteristics of OLEDs can be understood through an examination of the device in Fig. 1.⁶ The substrate is glass or plastic that has been pre-coated with a metal oxide or in some cases, a highly conductive organic film. In many OLED lighting devices, indium tin oxide (ITO) is utilized as the transparent anode electrode. The next set of layers is a series of organic layers that transport charge and emit light; they are applied via either a solution

or a physical vapor deposition process. In the solution coated device in Fig. 1, there are three separate regions in the organic layer: an oxidized region near the anode, a region that is neither oxidized nor reduced but contains dopant molecules that emit light and transport charge, and a region near the cathode that is highly reduced. The top, electron injecting electrode is applied on top of the organic layers. The top electrode is commonly applied by thermal evaporation of a low work function metal (such as an alkali or alkaline earth) or alloy, or a combination of an alkali-halide salt and a metal.⁷ Other high work function metals or solution based cathode deposition processes are used less often. Due to the reactive nature of the oxidized or reduced organic layers and the low work function cathode, the OLED is sealed to prevent ingress of water or oxygen.⁸

The operation of a typical OLED is illustrated in Fig. 1b. When voltage is applied between the two electrodes, positive charge carriers are injected into the organic layers from the anode and negative charge carriers are injected from the cathode. These

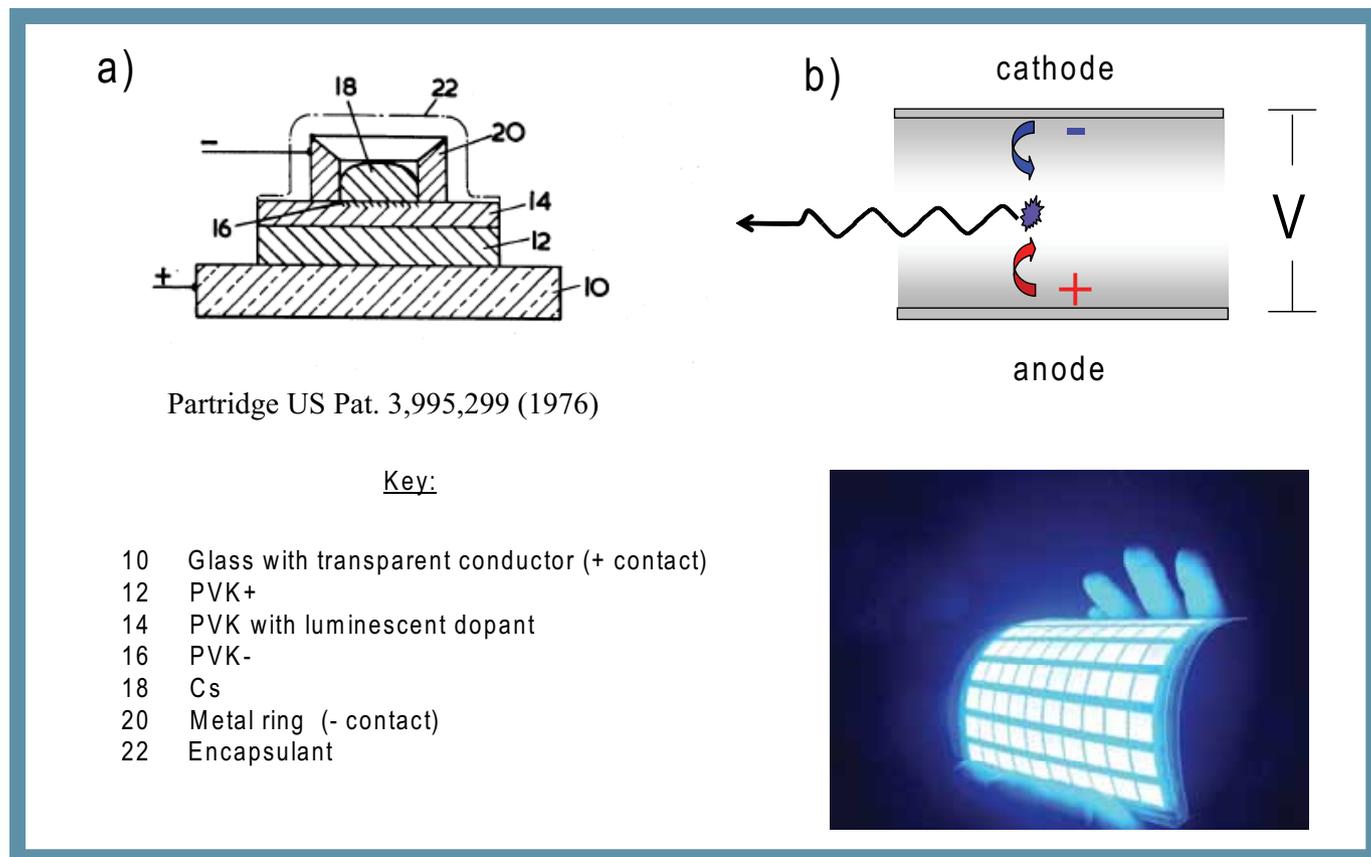


Fig. 1. (a) Schematic illustration of a solution processed OLED made using polyvinyl carbazole (PVK) as the primary electroactive component. (Figure taken from U.S. patent 3,995,299); (b) Simplified schematic of the physical processes involved in electroluminescence from organic materials. Lower portion shows an operating OLED made using state-of-the-art polymer materials on a flexible substrate.

Shiang

(continued from previous page)

oppositely charged carriers migrate under the applied electric field and recombine in the organic layer. Charge recombination either occurs in the host or at a luminescent dopant molecule. Following light generation, light either travels through the structure to the ambient or is lost through waveguiding or absorption in the OLED structure.

Since the original development of the OLED light source, the basic architecture shown in Fig. 1a has been extensively improved. Due to the broad flexibility afforded by synthetic organic chemistry there is a myriad of possible materials and layer structures, and improved material sets continue to appear in both the scientific literature⁹ and commercial marketplace.¹⁰ For example, the charge recombination events can be efficiently directed toward luminescent emission through the use of heavy metal organo-metallic dopants.^{11,12} Since different materials have different relative charge carrier mobilities and different energy levels of the electron (LUMO) and hole (HOMO), the location of the charge recombination event can be tightly controlled through the selection of materials in the organic layers. Since the binding between organic materials is van der Waals and not covalent (as in inorganic semiconductors) many different material combinations are possible. In addition, since these organic layers are amorphous, there are no epitaxial constraints that limit the selection of different organic layers. Consequently, device design has also been improved through the incorporation of additional layers that each serve a specific function; some

efficient designs have between four¹³ and six discrete organic layers between a single pair of electrodes.¹⁴ In addition, OLEDs can also be stacked vertically, so that the cathode of one device is the anode of the next device above it; these devices have even more layers.¹⁵

A common feature of organic materials is their low charge carrier mobility relative to inorganic semiconductors. For example, the OLEDs that were developed in the 1970s were quite thick by today's standards (ranging from 0.5 μm to several μm in thickness) and required high voltage for operation. Reducing the operating voltage requires a considerable reduction in the overall thickness of the device to $\sim 0.1 \mu\text{m}$.¹⁶ The requirement of very thin films for OLEDs has several other implications that impact device design. The proximity of the electrodes to the location of the emission zone makes the details of the physical optics very important for determining the optical loss mechanisms in the device. An additional consequence of the thin film nature of the device is that each molecule or polymer chain within the device must be able to repeat the charge injection, charge transport, and radiative emission cycle many times, since the total amount of material is limited. While OLEDs lifetimes at low brightness (150 cd/m^2) can be quite long ($> 200,000$ hours),¹⁷ the operating lifetime of devices decreases rapidly as the current density is increased beyond a few mA/cm^2 .¹⁸ High overall currents are required for lighting applications. For example, at a brightness 1000 cd/m^2 , $\sim 1 \text{ m}^2$ of emissive area is required to match the total lumen output of a standard 30W T8 linear fluorescent lamp; for an OLED design with an efficacy of 100 cd/A , the required current density

is 1 mA/cm^2 . For a planar device, the total emissive area is the product of the total area times the fill factor. Designs that have low fill factors require more total material (substrate, conductors, encapsulation) to be processed, which increases to the overall cost. Thus fill factors that are greater than 0.5 are desirable. The combination of fill-factor requirements (necessary for life) and device thickness (necessary for low voltage) places limits on the types of structures that can be used to promote high light extraction efficiencies.

As a result of these geometric limitations, the best OLEDs still have external quantum efficiencies (EQE) of 0.3-0.4, limited by the extraction efficiency of transporting light from the organic layers into the ambient. This present EQE would limit OLED efficiencies to slightly below that of current mercury fluorescent lighting technology. Solving this light extraction problem has been the focus of much theoretical and experimental effort. The light extraction problem can be partitioned into two different terms. One is the efficiency of coupling the light from the thin organic layers (refractive indices ranging from 1.5-2.0) into the optically thick substrate. The other is efficiency of coupling the light from the substrate that typically has an optical index of 1.5 (glass) or 1.6-1.7 (high index polymers) into air. Modeling the coupling of light from the electro-active organic layers into the substrate requires a detailed physical model of the near-field, or microcavity, properties of dipole emission in the presence of dielectric interfaces. For planar geometries, these microcavity models are extensions of Sommerfeld's original theory of radio wave propagation near the

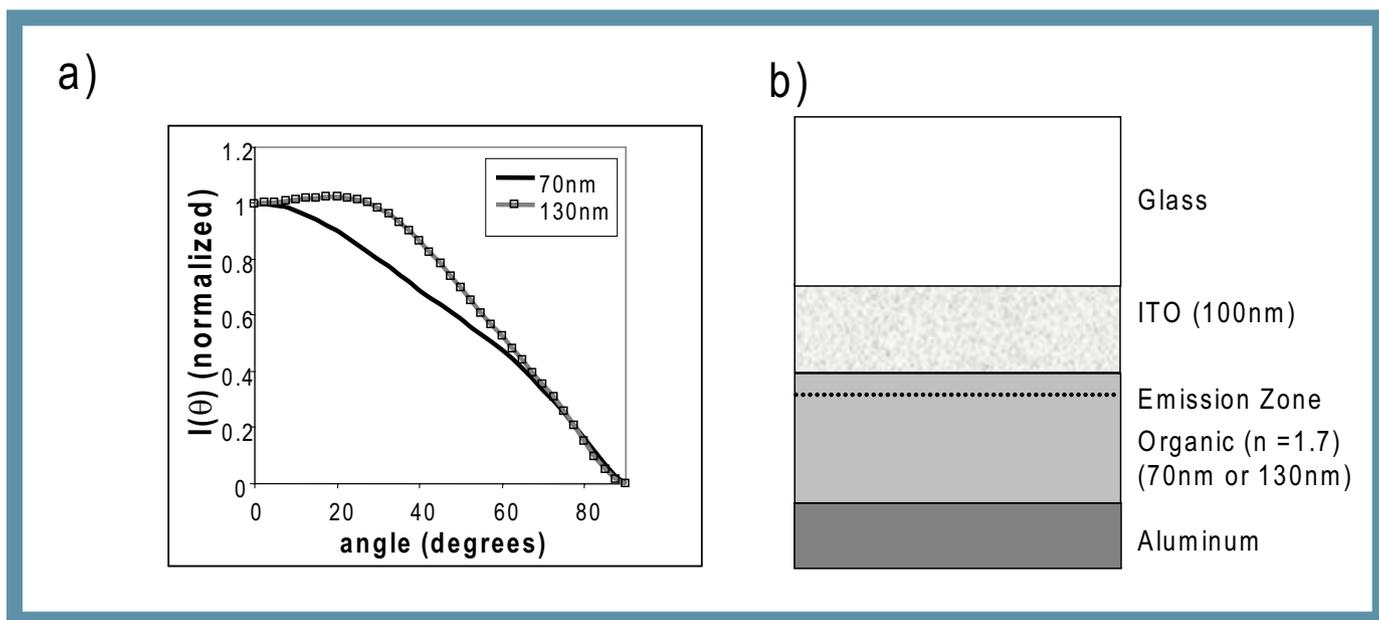


Fig. 2. (a) The calculated emission intensity $I(\theta)$ versus angle (in degrees) in the glass substrate. For both curves, $I(\theta)$ is normalized to be equal to 1 at 0 degrees. (b) The model OLED layer structure. The glass has an optical index of 1.518, the ITO has a complex index of refraction, with $n = 1.80$ and $k = 0.024$. The organic index is set to $n = 1.7$ and the metal cathode has complex index of refraction, with $n = 0.62$ and $k = 5.25$. The emission wavelength is 614 nm.

earth's surface.^{19,20} These models are quite rich in their implications and have interesting properties as different device parameters are varied. These models indicate that the details of layer design and emission zone position are critical for optimizing the angular emission pattern and total amount of light coupled from the electro-active organic layers of the device.²¹

The impact of these physical optics phenomena on OLED efficiency has led to many results in OLED modeling and device design.^{22,23} These effects are illustrated using a simplified OLED model consisting of a 100 nm thick ITO film on glass, an organic layer of either 130 nm or 70 nm total thickness, and an optically thick metal cathode. In both cases, the emission wavelength is 614 nm and the emission location is fixed at 20 nm away from the ITO interface. The assumed optical index of each layer is given in the figure caption. Using a microcavity analysis, the emission intensity versus angle in the glass substrate were calculated (Fig. 2). These results indicate that the 70 nm OLED will have a more forward directed emission. Based on only these data, it could be supposed that setting to organic layer thickness equal to 70 nm would lead to an optimal design. However, a detailed account for all the loss mechanisms, such as plasmons and absorption loss in the metal, indicates that the structure with a 130 nm organic film has a total light coupling into the substrate that is about 1.3 times greater than the structure with 70 nm film. In an actual design, these optical considerations would have to be balanced against the increase in

operating voltage due to the increased organic layer thickness.

Using inputs from a microcavity model, the coupling of light from the substrate to air can then be modeled in detail via radiative transport methods or through ray tracing simulations. In bulk media, the efficacy of coupling light from a high index medium into air is limited to between 0.2 and 0.4.²⁴ In bulk emissive systems, the fraction of light emitted into the ambient can be increased via appropriate shaping or by the introduction of light scattering materials. In OLEDs, a similar effect is achieved through substrate modification. For example, one solution, most applicable to point sources, is to use a shaped substrate, and it has been demonstrated that certain lens-like shapes will result in almost complete light extraction.²⁵ This method has been applied to patterned OLED devices as well, where the active area of the OLED was placed at the center of a large hemisphere or at the bottom of an etched well.²⁶ However, fill factor considerations limit the applicability of this approach to lighting applications. In addition, light scattering within the substrate or device may be used to increase the OLED light extraction efficiency.^{27,28} Calculations and experiments indicate that the overall extraction efficiency can be quite high with values for light extraction from the substrate into ambient >75%.²⁷ However, at the present time, the development of a fully satisfactory out-coupling solution from planar OLED emitters remains a topic of active research.

The preceding example is also helpful in understanding the importance of system level considerations in OLED design. At the device level, there are multiple factors with significant interactions that must be addressed for the best net system performance. These constraints and trade-off analysis increase in complexity when taking account the stringent color requirements for general illumination lighting. In addition, OLED-based lighting will also require new luminaire system designs (including electrical contacts and power supply) that maximize the potential benefits of these light sources. An additional challenge is that these design constraints are coupled with potential processing concerns. For example, electrical defects such as pinholes result in shorting of the anode and cathode, leading to device failure. Thus an OLED architecture must not only meet the design requirements, it also must be robust to manufacture. Given all these constraints, it would seem to be nearly impossible to create a functional lighting source of any significant size. In reality, the OLED fabrication process is relatively robust. Large area devices have been demonstrated through the development of defect tolerant architectures²⁹ and through careful process control; some results of these efforts at GE are shown Fig. 3. All of these devices were prepared using solution processing to deposit the organic layers and have high white light color quality (correlated color temperature between 3200K and 4100K with color rendering indices ≥ 85). In addition, these devices could be operated for sustained periods of time at brightness levels suitable for illumination.



Fig. 3. White light OLED based luminaires produced over a several year period at GE.

Shiang

(continued from previous page)

To transition these demonstration prototypes into lighting products, low cost, large area, OLED manufacturing systems need to be developed. The best method to transition OLED devices to manufacturing is also an active area of discussion with several possible approaches under consideration³⁰: (1.) applied research into multilayer solution coating processes; (2.) making physical vapor deposition methods more efficient with higher throughput; and (3.) novel methods of OLED device fabrication.³¹

At GE, we have also demonstrated the feasibility of large area manufacturing through the output of our roll-to-roll OLED manufacturing test facility.³² The device in Fig. 4 is an example of the potential of multilayer solution coating processes. The OLED layers in these devices were deposited using solution web coating methods adapted from high speed printing technology. While the devices were produced using this manufacturing process are not yet suitable as general illumination quality lighting sources, the ability to develop the tooling and machinery to coat OLEDs over large areas is a critical proof of concept for low cost manufacturing. An example of novel methods for device fabrication that could significantly reduce manufacturing costs are devices where a multilayer OLED is formed via the lamination of two separate components. The feasibility of this concept was demonstrated in a relatively simple polymeric test structure,³⁰ and still needs to be applied to highly efficient device architectures. This lamination process used to make this device would completely remove all vacuum processing steps from the actual manufacture of OLED devices and enable a considerable reduction in OLED manufacturing complexity and cost.

Conclusions and Future Outlook

The successful transition of OLED technology would have many benefits with many new applications within the lighting industry. Apart from new form factors and design considerations, OLED technology offers a means of reducing the overall amount of energy used for lighting. It will be necessary to have a truly interdisciplinary effort that integrates OLED technology from molecular design all the way through the ultimate product manufacturing process. For example, OLED light extraction efficiencies could be increased substantially by developing materials and structures that relax the present constraints on OLED design. Similarly, many of the limitations of the current manufacturing process for OLEDs are dictated by the fundamental

properties of the materials, such as their ability to form solution coated films, and their air stability both during and after OLED manufacture. These challenges toward transitioning OLED to the lighting marketplace are being taken up by a variety of groups around the world, so there is good reason to be optimistic about the impact that OLED technology will have on the future of lighting.

Acknowledgments

The author wishes to acknowledge the many technical conversations and collaborations he has had with the members of the Electronic Materials System Advanced Technology Program at GE. He also wishes to thank Anant Setlur for a critical reading of the manuscript.

Some of the results presented here were from work partially supported by the U.S. Department of Energy through contract #s DE-FC26-07NT43226, DE-FC26-05NT42343, DE-FC-00NT40989; and the National Institute of Standards and Technology through contract # 70NANB3H303. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of author expressed

herein do not necessarily state or reflect those of the United States Government or any agency thereof.

About the Author

JOSEPH SHIANG is a chemist at the GE Global Research Center in Niskayuna New York. He may be reached via e-mail at shiang@ge.com.

References

1. A. R. Duggal, in *Organic Electroluminescence*, Z. H. Kafafi, Editor, p. 437, Taylor & Francis Group, Boca Raton (2005).
2. R. H. Partridge, U.S. Patent 3,995,299; R. H. Partridge, *Polymer*, **24**, 733, (1983); R. H. Partridge, *Polymer*, **24**, 739, (1983); R. H. Partridge, *Polymer*, **24**, 748, (1983); R. H. Partridge, *Polymer*, **24**, 755, (1983).
3. U.S. DOE Solid State Lighting MYPP09, available from: http://apps1.eere.energy.gov/buildings/publications/pdfs/ssl/ssl_mypp2009_web.pdf.
4. C. Adachi, M. A. Baldo, M. E. Thompson, S. R. Forrest, *J. Appl. Phys.*, **90**, 5048, (2001).
5. K. Walzer, B. Maennig, M. Pfeiffer, K. Leo, *Chem. Rev.*, **107**, 1233, (2007).
6. G. Parthasarathy, J. Liu, A. R. Duggal, *Electrochem. Soc. Interface*, **12(2)**, 42, (2003).
7. L. S. Hung, C. W. Tang, M. G. Mason, *Appl. Phys. Lett.*, **70**, 152 (1997).
8. M. Yan, T. W. Kim, A. G. Erlat, M. Pellow, D. F. Foust, J. Liu, M. Schaeckens, C. M. Heller, P. A. McConnelee, T. P. Feist, and A. R. Duggal, *Proc. of IEEE*, **93**, 1468 (2005).
9. G. Malliaras and R. Friend, *Physics Today*, **58**, 53, (2005).

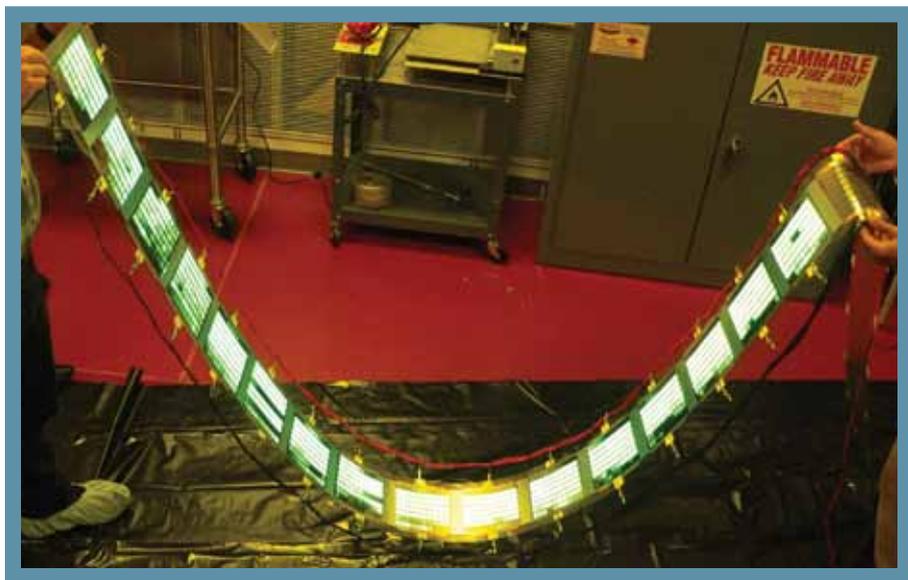


Fig. 4. One of the first examples of a roll-to-roll OLED manufactured at the test facility at GE.

10. W. Wu, M. Inbasekaran, M. Hudack, D. Welsh, W. Yu, Y. Cheng, C. Wang, S. Kram, M. Tacey, M. Bernius, R. Fletcher, K. Kiszka, S. Munger, J. O'Brien, *Microelectronics Journal*, **35**, 343 (2004).
11. M. A. Baldo, S. Lamansky, P. E. Burrows, M. E. Thompson, and S. R. Forrest, *Appl. Phys. Lett.*, **75**, 4 (1999).
12. S. Lamansky, P. Djurovich, D. Murphy, F. Abdel-Razzaq, R. Kwong, I. Tsyba, M. Bortz, B. Mui, R. Bau, and M. E. Thompson, *Inorg. Chem.*, **40**, 1704 (2001).
13. D. Tanaka, H. Sasabe, Y.-J. Li, S.-J. Su, T. Takeda, and J. Kido, *Jpn J. Appl. Phys.*, **46**, L10 (2007).
14. G. He, M. Pfeiffer, K. Leo, M. Hofmann, J. Birnstock, R. Pudzich, and J. Salbeck, *Appl. Phys. Lett.*, **85**, 3911 (2004).
15. G. Gu, G. Parthasarathy, P. Tian, P. E. Burrows, and S. R. Forrest, *J. Appl. Phys.*, **86**, 4076 (1999).
16. C. W. Tang, S. A. VanSlyke, and C. H. Chen, *J. Appl. Phys.*, **65**, 3610 (1989).
17. P. Wellmann, M. Hofmann, O. Zeika, A. Werner, J. Birnstock, R. Meerheim, G. He, K. Walzer, M. Pfeiffer, and K. Leo, *Journal of the SID*, **13**, 393 (2005).
18. Z. Popovic and H. Aziz, *IEEE J. Quantum Electronics*, **8**, 362 (2002).
19. A. Sommerfeld, *Partial Differential Equations in Physics*, Academic, New York (1949).
20. R. Chance, A. Prock, and R. Silbey, *Adv. Chem. Phys.*, **37**, 1 (1978).
21. V. Bulovic, V. B. Khalfin, G. Gu, P. E. Burrows, D. Z. Garbuzov, and S. R. Forrest, *Phys Rev. B*, **58**, 3730 (1998).
22. L. H. Smith, J. A. E. Wasey, I. D. W. Samuel, and W. L. Barnes, *Adv. Funct. Mater.*, **15**, 1839 (2005).
23. J. M. Ziebarth and M. D. McGehee, *J. Appl. Phys.*, **97**, 064502 (2005).
24. J. J. Shiang and A. Duggal, *J. Appl. Phys.*, **95**, 2880 (2004).
25. W. N. Carr, *Infrared Physics*, **6**, 1 (1966).
26. G. Gu, D. Z. Garbuzov, P. E. Burrows, S. Venkatesh, S. R. Forrest, and M. E. Thompson, *Optics Letters*, **22**, 396 (1997).
27. J. J. Shiang, T. J. Faircloth, and A. R. Duggal, *J. Appl. Phys.*, **95**, 2889 (2004).
28. Y. Sun and S. R. Forrest, *Nature Photonics*, **2**, 483 (2008).
29. A. R. Duggal, D. F. Foust, W. F. Nealon, and C. M. Heller, *Appl. Phys. Lett.*, **82**, 2580 (2003).
30. F. So, J. Kido, and P. Burrows, *MRS Bulletin*, **33**, 663 (2008).
31. J. Liu, L. N. Lewis, T. J. Faircloth, and A. R. Duggal, *Appl. Phys. Lett.*, **88**, 223509 (2006).
32. "From Edison's Desk," GE Global Research Blog, March 11, 2008: <http://www.grcblog.com/?p=247>.

ECS Monograph Series

Electrochemical Impedance Spectroscopy

by Mark E. Orazem & Bernard Tribollet



MARK E. ORAZEM, University of Florida and BERNARD TRIBOLLET, Centre National de la Recherche Scientifique (CNRS) and Pierre and Marie Curie University; 524 pages, ISBN 978-0-470-04140-6

This book provides the background and training suitable for application of impedance spectroscopy to varied applications, such as corrosion, biomedical devices, semiconductors and solid-state devices, sensors, batteries, fuel cells, electrochemical capacitors, dielectric measurements, coatings, electrochromic materials, analytical chemistry, and imaging. The emphasis is on generally applicable fundamentals rather than on detailed treatment of applications. With numerous illustrative examples showing how these principles are applied to common impedance problems, *Electrochemical Impedance Spectroscopy* is ideal either for course study or for independent self-study, covering:

Special discount for ECS members!

Order your copy from ECS

E-mail: customerservice@electrochem.org

Tel: 609.737.1902 / Fax: 609.737.2743



Published by John Wiley & Sons, Inc.