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REPORT OF DOCTORAL THESIS

Adaptation of Inkjet Printing Technique and Slot-Die Coating Technique for OLED fabrication,
Doctoral Thesis of Mr. Amruth C, Lodz University of Technology

The PhD Thesis, "*Adaptation of Inkjet Printing Technique and Slot-Die Coating Technique for OLED fabrication*" focus a particular critical aspect of Organic Light Emitting Diode technology development related with the "technological triangle" simple structure / efficiency / large area. In fact, regardless the huge developments of OLEDs fabrication and applications, the technological issues associated with the main fabrication technique (thermal evaporation) and the consequent required device complexity to achieve suitable final devices, impacting in the more advance applications, particularly in lighting. In that way, new techniques, specifically based in wet deposition process, compatible with large area fabrication, becomes (after a first idea in the beginnings of OLEDs displays production) of a great of interest and being an important area of research in main scientific and technological groups working in devices field. Notably, the physical assessments required for efficient, profitable and reliable OLEDs can easier ne obtained in wet deposition process; the reverse of such interesting framework, is that high degrees of freedom for parameters are available with a noteworthy complexity, that imposes in the end a very complex framework not easy to overcome. In spite of that, developing scientific and technological work trying to improve these methods are nor easy but simultaneously, a great challenge towards the everyday more important concept of "printing electronics". Thus, the present thesis is indeed important, and the topic focused is without doubt, current and of total interest. Based on the concept of "paper publications", overall, the thesis is well conceptualized and easy to read and understand the results, discussions and conclusions. Apart from minor absence of connecting ideas among the published papers, that are not important in the thesis concept, the document contains no major flaws that could compromise presentation and discussion. Moreover, from my personal opinion, presents very interesting results that are not only important now, but sure, with impact in the future of this field research.

The introductory chapter is clear to explain the thesis scope. Follow a simple line of presentation, focusing, in the natural work evolution, the main achievements and (although not in an explicit form) try to show the options made during the work development. In that point, is

perfect, with a good state of the art (further complete in the publications themselves) and clear of the work developed.

The first work published (*Slot-Die Coating of Double Polymer Layers for the Fabrication of Organic Light Emitting Diodes*) is, in my opinion, one of the most important parts of this thesis. Besides good results, although not exceptional, the most important question is a clear explanation of the main issues related with the slot-die (although not the case is the base of widely spread Roll-to-Roll continuous fabrication line concept). From my personal experience, this technique is hard to improve, particularly regarding the optimization of organic layers. The data presented, (for instance in sections 3.1.1, 3.1.2 and 3.1.3) are crucial for all the process. Achieving optimal conditions is the fundamental question. This paper shows without doubt the ideal process to do that, opening the real future possibilities to a more idealistic development. The results are interesting for a very simple device structure. Regardless the low efficiency (and I do not understand how a simple estimation of external quantum efficiency was not provided, although for the case study is not of high interest) it can be prove a clear possibility for the use of slot-die process for OLEDs fabrication. Notwithstanding do not have a precise "fence" for applicable rules limiting the use of slot-die technique, the results opens a very large possibility for future. As for the scientific discussions and conclusions, both are well framed and reflects in a correct way, the experimental results obtained. Perhaps, this paper is the most interesting in technological point of view.

The second paper "*Inkjet Printing of Super Yellow: Ink Formulation, Film Optimization, OLEDs Fabrication, and Transient Electroluminescence*" denotes a clear more deep scientific work, besides the main idea of printed devices. The inkjet printing technique has been used from many years to produce organic based devices. The concept appears to be simple, but as very well known, is very complex. This paper shows that but the main achievement is a critical and very well driven idea to establish the basic relationships in order to reveals the main factors for efficient printed OLEDs. Using an well-known super-efficient emitter ("super yellow") that, in principle assure a useful emission, the main interesting results, in my opinion, are two: the determination of effective mobility and the dependence of the results (particularly in the current efficiency) on the active layer thickness. This last, is critically depend on the exciton (and obviously on the electrons / holes) profile inside the active layer that in turn, depends on the mobility (and this is the first main interesting result). This less explained idea was the only less positive unexplored concept in the paper but, of course, it does not detract from the extraordinary importance of the overall results that have been obtained. AFM, by turn, was excellent to prove the influence of parameters adjustment for the final results. Based on a large set of experimental data, very well grounded and explained, the results prove without doubt, that useful and suitable printed OLEDs in simplified and structures can be possible. The original idea exposed in the paper introduction (and in the

main goal of this thesis) is completely achieved in this second paper, that, joined with the first one, shows that slot-die and inkjet deposition / printing techniques can surprisingly compete in an remarkable way with the basic spin-coting, achieving device figures of merit absolutely compatibles.

The third paper, "*Inkjet printing of thermally activated delayed fluorescence (TADF) dendrimer for OLED application*" introduces a new idea in the work timeline. As very well know, the basic organic emitters suffers from the low internal efficiency due to the spin-forbidden T-S radiative emission. Triplet harvesting is, therefore, the most studied process in order to increase the internal efficiency of organic emitters and consequentially to increase the external efficiency. TADF materials are currently, the best possibilities (in the balance cost / results). Is therefore natural, that, for all the previous discussed ideas, attempts to print TADF based OLEDs takes place. The work described in this paper, have, in my opinion, the main importance in the optimization conditions for efficient print of multilayer device. The huge advance compared with the both previous papers, is the increase of devices complexity (although keeping them in an affordable structure) with simultaneously final good success as the main results proves. Several factors influences the printing techniques: fluid viscosity, temperature, concentration, surface tension, etc. In this paper, almost of them are detailed explored (drop formation, contact angle, for instance). Noticeable, is the result that shows the ability to pattering the emission area that can obviously be further applied in complex shapes. Again, the determination of carriers mobility are of maximum importance to a complete understand of the physical process that, in final, should condition the results. Again, the results, in same magnitude of that obtained by spin-coating technique, are clear to definitely proof the viability of the idea to produce OLEDs by inkjet.

Finally, the last paper published under this thesis, "*Inkjet Printing Technique and Its Application in Organic Light Emitting Diode*" is an overall presentation of the different issues / solution / process, etc., related with the device printing. Well done, focusing the main important questions is noticeable as state of the art and opens in a clear way, what we can expect for the future. In my opinion, should be considered simultaneously as a very good state of the art assessment and the base of the present thesis formulation and work plan. In that concept, it is very good to show that the followed work plan was well establish and, with the results found, perfectly done.

As a general idea, this work is clearly important and introduces new approaches for use more interesting printing techniques than simple spin-coating process to made OLEDs, in a research field every day as more important in both scientific and technological point of view. In my opinion, we are in the presence of an excellent work where, the implications for future will be crucial. It is relevant to mention once again that the future of several widely used devices,

including electroluminescence ones, will be the printing techniques towards large area / low cost in simplified device structures. The present work, adds relevant news in such idea.

Less well done, I would point some absence of a more deep physical discussion related with the electrical transport phenomena and its correlation with the results found. All the data was there and would be very interesting to discuss that. In any case, and as I pointed before, this was not a fundamental question in the work plan and I believe that, with the excellent results now obtained, a very interesting development will be done.

Finally, the excellent publications made based in this work, is obviously one of the most important consequences for the validity and importance for the research field, adding suitable new and valuable approaches, that the scientific and technological community has recognized the value.

Thus, considering the overall thesis document, including the organization, writing and concept, the novelty of the work and the related publications**, I recommend that the thesis "*Adaptation of Inkjet Printing Technique and Slot-Die Coating Technique for OLED fabrication*" should be accept as it is, ready for exam.**

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