THE OUTCOME OF THE ISOS-3 INTER-LABORATORY COLLABORATION – CHARACTERIZATION OF DEGRADATION PATHWAYS OCCURRING IN A VARIETY OF ORGANIC PHOTOVOLTAIC DEVICES

Birgitta Andreasen,[a] David M. Tanenbaum,[a,b] Martin Hermenau,[c] Eszter Voroshazi,[d] Matthew T. Loyd,[e] Yulia Galagan,[f] Birger Zimmernann,[g] Suleyman Kudret,[h] Wouter Maes,[h] Laurence Lutsen,[i] Dirk Vanderzande,[h] Uli Würfel,[g] Ronn Andriessen,[f] Roland Rösch,[j] Harald Hoppe,[j] Gerardo Teran-Escobar,[k] Monica Lira-Cantu,[k] Agnès Rivaton,[l] Gülşah Y. Uzunoğlu,[m] David Germack,[n] Markus Hösel,[a] Henrik F. Dam,[a] Mikkel Jørgensen,[a] Suren A. Gevorgyan,[a] Morten V. Madsen,[a] Eva Bundgaard,[a] Frederik C. Krebs[a] and Kion Norrman[a]

[a] *Department of Energy Conversion and Storage, Technical University of Denmark, Frederiksborgvej 399, DK-4000, Roskilde, Denmark.*
[b] *Department of Physics and Astronomy, Pomona College, Claremont, CA, 91711, USA.*
[c] *Arbeitsgruppe Organische Solarzellen (OSOL), Institut für Angewandte Photophysik, Technische Universität Dresden, 01062, Dresden, Germany.*
[d] *Imec, Kapeldreef 75, 3000 Leuven, Belgium and Katholieke Universiteit Leuven, ESAT, Kasteelpark Arenberg 10, 3000, Leuven, Belgium.*
[e] *National Renewable Energy Laboratory, Golden, CO, 80401, USA.*
[f] *Holst Centre, High Tech Campus 31, 5656 AE Eindhoven, The Netherlands.*
[g] *Fraunhofer Institute for Solar Energy Systems ISE, Heidenhofstrasse 2, D-79110, Freiburg, Germany.*
[h] *Hasselt University, Campus, Agoralaan 1, Building D, WET/OBPC, B-3590 Diepenbeek, Belgium.*
[i] *IMEC, IMOMEC associated laboratory, Campus University of Hasselt, Wetenscharpspark 1, B-3590, Diepenbeek, Belgium.*
[j] *Institute of Physics, Ilmenau University of Technology, Weimarer Str. 32, 98693, Ilmenau, Germany.*
[k] *Centre d'Investigació en Nanociència i Nanotecnologia (CIN2, CSIC), Laboratory of Nanostructured Materials for Photovoltaic Energy, ETSE, Campus UAB, Edifici Q, 2nd Floor, E-08193, Bellaterra (Barcelona), Spain.*
[l] *Clermont Université, Universit Blaise Pascal, Laboratorie de Photochemie Moléculaire et Macromoléculaire (LPMM), BP10448, F-63000 Clermont-Ferrand, France and CNRS, UMR6505, LPMM, F-63177, Aubiére, France.*
[m] *TÜBITAK National Metrology Institute (UME), Photonic and Electronic Sensors Laboratory, P.O. Box 54, 41470, Gebze, Kocaeli, Turkey.*
[n] *Condensed Matter Physics, Brookhaven National Lab, Building 510B Upton, NY, 11973, USA.*

Introduction

Research within organic photovoltaics (OPVs) has attracted much attention over the past decade, with impressive progress being made with photoelectronic conversion efficiency (PCE) optimization and lifetime and stability [1,2]. For OPV technology to be considered for large scale application it is of outmost importance that acceptable lifetimes are achieved. It is thus necessary to gain a detailed understanding of the multitude of degradation mechanisms that are in play in these complex multilayer systems.

In an effort to further understand the lifetime and stability of OPVs a large inter–laboratory study was initiated as a result of the third International Summits on Organic Photovoltaic Stability (ISOS-3). The study involved seven distinct sets of OPV devices manufactured at six different laboratories (HOLST, NREL, ISE, IMEC, IAPP and DTU Risø), the devices were all degraded under identical conditions at DTU Risø and then subjected to extensive investigation at different analysis laboratories. The ISOS-3 inter–laboratory study has resulted in multiple reports, each with a different area of focus on stability and degradation. The first report included device manufacture methods, degradation procedures and electrical characterization [3]. The second report focused on mapping specific degradation mechanisms by using a suite of imaging techniques including laser-beam induced current (LBIC), photoluminescence imaging (PLI), electroluminescence imaging (ELI) and lock-in thermography (LIT) [4]. Degradation analysis by Incident photon-to-electron conversion efficiency (IPCE) and *in-situ* IPCE were the focus of the third report [5]. A fourth and final report has been submitted in which the main analytical technique was time-of-flight secondary ion mass spectrometry (TOF-SIMS) [6]. This paper will give a short summary of the overall outcome of the ISOS-3 inter–laboratory study.

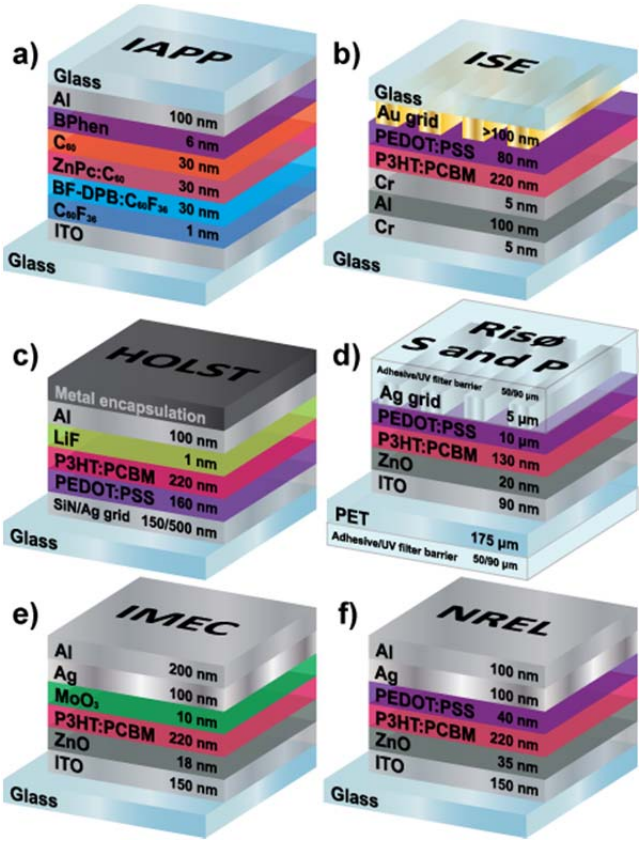

Figure 1. Illustration of the OPV device layer structures used in this ISOS-3 study (materials and thicknesses) (a) IAPP, (b) ISE, (c) HOLST, (d) DTU Risø S and DTU Risø P, (e) IMEC and (f) NREL. The only difference between DTU Risø P and DTU Risø S is the polymer used in the active layer, P3HT and P3HT-co-P3AcET respectively. Further details can be found in **ref. [3]**.

Experimental

The OPV devices that were studied in this collaboration are shown in **Figure 1**, further details are in **ref. [3]**.

The OPVs were degraded under three different conditions: accelerated full sun simulation, low level indoor fluorescent lighting, and dark storage with daily measurements under full sun simulation. The devices were analyzed and characterized at different points of their lifetimes: T100 (pristine device), approximately T80 (when PCE has declined to 80% of its initial value), T50 and T10. Different devices were thus extracted from degradation at specific T-values for destructive testing while selected devices were cycled so that the evolution of spatial defects on the same cell could be monitored.

Results and Discussion

The results from the electrical characterization reveled a dramatically different degradation behavior for the devices exposed to full sun simulation; see **Figure 2,** which presents the best performing device of each type [3].

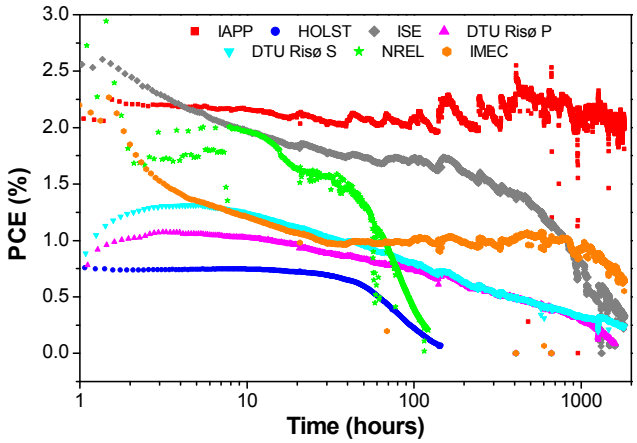

Figure 2. Efficiencies of all the devices degraded under accelerated full sun simulation plotted as a function of time.

Not surprisingly most devices exhibited a better lifetime under low level indoor fluorescent light conditions and during dark storage. For some devices the degradation experiments lasted up to 1830 hours.

The second study utilized LBIC, PLI, ELI and LIT to provide indirect information about the occurring degradation mechanism. Because these techniques are non-destructive it was possible to analyze cycled devises, thus making it possible to follow the spatial evolution of the device degradation [4]. It was found that the main degradation mechanisms were: (i) formation of aluminium oxide (at the aluminium electrode), (ii) formation of blocking contacts caused by either silver oxide formation or dedoping of ZnO, (iii) electro-migration of silver, (iv) ingress of water and oxygen through pinholes and from the edges, and (v) water release from highly conductive PEDOT:PSS. As previously mentioned, the imaging techniques provide indirect information so ideally these techniques should be combined with techniques that provide direct chemical information.

In the third study IPCE and *in-situ* IPCE analyses were conducted in both ambient– and N_2–atmospheres that made it possible to identify materials more susceptible to degradation by molecular oxygen and water [5]. The major observation regarding degradation of the encapsulated devices was that the degradation for some of the devices could have been initiated by the Ag (or Au)/PEDOT:PSS interface by the formation of a chemical bond/interaction between Ag (or Au) and the PEDOT:PSS. For the devices without encapsulation the main degradation pathways observed were: (i) high dependence on atmospheric conditions with a major issue being water uptake, which was attributed to the hygroscopic nature of the PEDOT:PSS and semiconductor oxides, (ii) water uptake was observed to be random and reversible, and (iii) water primarily degraded the electrodes of the cell. The third report also reveled that in devices consisting of multiple cells, the cells did not always degrade in a similar manor.

In the fourth study the mass spectral technique TOF-SIMS was employed, which produces direct chemical information that was used to unravel various degradation mechanisms [6]. One focus of this work was to quantify the oxygen uptake in the active material of the devices, thus revealing the level of photo-oxidation as a function of device performance. No photo-oxidation could be detected in the impermeable encapsulated devices (HOLST and ISE), while there was a linear relationship between the level of oxygen incorporation and performance for the devices with a so-called semi-impermeable encapsulation (DTU Risø S and P) the non-encapsulated devices (IMEC and NREL) showed an initially slow rate of photo-oxidation that accelerated later in the degradation tests.

Conclusions

The outcome of the four reports of the ISOS-3 inter-laboratory collaboration has demonstrated how advantageous it is to combine complementary highly specialized analytical techniques, which has vastly increased the understanding of OPV stability and degradation. It also signifies the need for standardized methods in OPV degradation research along with the need for continuous collaboration across the OPV community, so that ultimately the OPV technology can be commercialized.

References

1. M. Jørgensen, K. Norrman, S.A. Gevorgyan, T. Tromholt, B. Andreasen, F.C. Krebs, Stability of Polymer Solar Cells, Advanced Materials. 24 (2012) 580-612.
2. N. Grossiord, J.M. Kroon, R. Andriessen, P.W.M. Blom, Degradation mechanisms in organic photovoltaic devices, Organic Electronics. 13 (2011) 432-456.
3. D.M. Tanenbaum *et al.*, The ISOS-3 inter-laboratory collaboration focused on the stability of a variety of organic photovoltaic devices, RSC Advances. 2 (2012) 882-893.
4. R. Rösch *et al.*, Investigation of the degradation mechanisms of a variety of organic photovoltaic devices by combination of imaging techniques – the ISOS-3 inter-laboratory collaboration, Energy & Environmental Science. 5 (2012) 6521-6540.
5. G. Teran-Escobar *et al.*, On the stability of a variety of organiv photovoltaic devices by IPCE and in-situ IPCE analyses - the ISOS-3 inter-laboratory collaboration, Physical Chemistry Chemical Physics. (2012). DOI:10.1039/C2CP40821J
6. B. Andreasen *et al.*, TOF-SIMS investigation of degradation pathways occurring in a variety of organic photovoltaic devices – the ISOS-3 inter-laboratory collaboration, (2012). ***Submitted***