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Electrical and thermal analysis of frequency dependent filamentary switching in printed rectifying diodes

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ABSTRACT

Filamentary conduction and switching properties are studied in printed rectifying diodes with a poly(triaryl amine) (PTAA) semiconductor layer sandwiched between Cu and Ag electrodes. Formation of conductive filaments caused defective operation of the rectifier at low frequencies. In contrast, the normal operation was restored at high frequencies. Reversible switching was observed between the low and high frequency states. Therefore, it is clear that the operational frequency has a significant effect on the filament formation and switching characteristics. The filamentary conduction was confirmed by lock-in IR thermography and physical defect analysis. The results reveal the existence of filamentary operation in p-type rectifying diodes and clearly demonstrate the effect of the device operation frequency on the switching properties. This has far-reaching implications on the switching properties in similar devices in literature.

1. Introduction

The quality and performance of organic electronic devices has improved significantly during the past decades. At the same time the required performance level of the devices is increasing. The performance requirements as well as new application markets of vertical organic devices have driven the research and development toward a trend of thinner active material layers. Extremely thin layers are beneficial, for example, in organic light emitting diodes (OLED) [1,2], capacitors, transistor gate dielectrics [3] and high frequency rectifying diodes [4] for application as charge-pump circuits [5]. A thinner active layer, in general, leads to enhanced device operations and additional freedom in design and integration. However, the decrease of the active layer thickness also increases the risk of layer defects which influence or prevent the overall device functionality. Following the market requirements, these factors can influence both reliability and lifetime of the device and need, therefore, careful and intense defect analysis.

In organic electronic devices, the operation voltages are conventionally high due to the performance limitations of the organic materials [2,6,7]. Therefore, the electric field in the active materials of the devices easily reaches 10 MV/m level or even higher. The high operation voltage and electric field may cause various phenomena, including dielectric breakdown, electrode redox reactions (dendrite formation) [8] and conductive filament formation [9–11]. The conductive filament formation is a known critical and undesired defect phenomena which can lead into local ohmic short circuiting. However, in recent years, the filament formation, knowingly or unknowingly, has been utilized also for organic resistive memory applications

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[8,12–15], where bi-stable resistance states of the switching are utilized to fabricate simple and small dimension memory circuits. In most cases the origin of switching was filamentary conduction. The main reasons proposed for the filamentary conduction are carbon-rich filament formation by local degradation of the material and metallic filament formation resulting from migration of ionized metal through the film [9,10]. Due to the thin and fragile structure of the filaments, the existence is hard to verify or locate [16] and requires non-destructive analysis methods. It is also to be noted that most of the filamentary switching studies have been performed using only DC characterization or low frequency signals.

In this paper, the effect of filament formation on organic rectifying diodes is studied by the built up of defined test structures with two different active layer thicknesses and an analysis method for the phenomena is proposed. The electrical characteristics of the diodes are measured to study the switching properties of the diodes. To confirm the filamentary nature of the conduction, local heat sources are investigated and recorded in detail using high sensitive IR thermography imaging. In addition, physical failure analysis (PFA), using focused ion beam (FIB) sample preparation and scanning electron microscopy (SEM) imaging, is utilized to further analyze the defective areas of the devices.

2. Materials and methods

The diodes under investigation were fabricated on a PET foil (Melinex ST506, Dupont Teijin Films) using a sheet-fed gravure printing press (Labratester Automatic from Norbert Schläfli Maschinen). First, a 100 nm copper layer was evaporated onto the substrate and patterned using a reel-to-reel wet etching process, where the etch resist was printed using a rotary screen printer. Before printing of the poly(triaryl amine) (PTAA) semiconductor (SC), the samples were cleaned using deionized water and 2-propanol. The PTAA semiconductor was then gravure printed onto the patterned Cu structures and cured for 5 min at 115 °C. Two different dilutions of the PTAA were used to fabricate devices with different semiconductor layer thicknesses. After semiconductor curing, the top Ag electrodes were gravure printed using silver flake ink (PM460A from Acheson Industries Ltd.). The silver top electrodes were cured at 115 °C for 5 min. All the fabrication steps were performed in ambient air and in a dust free environment. As a result, a "tree-shaped" sample structure was fabricated with five different device areas (Fig. 1). The advantage of the sample structure is an easy contacting of each individual sample using micro-manipulators. The electrical contact to the anode can hereby be retained. This decreases the risk of mechanical sample damage. For systematic investigation, two different sample sets were fabricated with different semiconductor thicknesses.

In order to investigate the fabricated samples, electrical and thermal as well as non-destructive and invasive methods were applied. To ensure a high success rate, a work flow which allows a long existence of the overall device functionality is proposed:

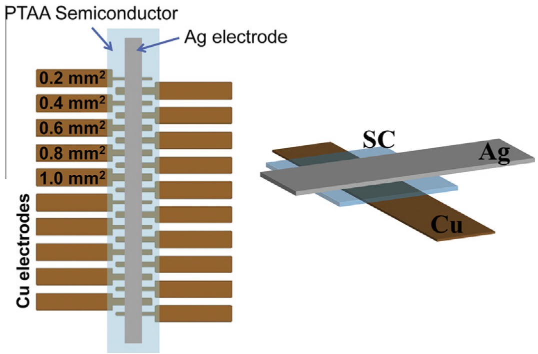

Fig. 1. Schematic illustration of the sample and diode structures. A stack of copper–PTAA–silver is printed on a flexible PET. The structure is printed as a "tree shaped structure" which enables an easy electrical contacting. Two different PTAA layer thicknesses were fabricated for the analysis.

1. Electrical testing of the device and determination of the *I–V* and device specific characteristics.
2. Thermal testing of the device for precise localization of electrical conduction pathways.
3. Target preparation in the hot spot area using focused ion beam technique (FIB) and SEM imaging for root cause determination.

This analysis flow was applied on both sample thicknesses, where the sample with thicker semiconductor layer is regarded as a reference.

2.1. Electrical characterization

As a first investigation step, the diode DC current–voltage (*I–V*) characteristics were measured using a Keithley 236 source measure unit. In addition, a Zennium workstation (ZAHNER-Elektrik GmbH & CoKG) was used to measure the *I–V* hysteresis. The half-wave rectification measurements were performed using a 10 $V_{zero\text{-}to\text{-}peak}$ sinusoidal input signal, 47 nF load capacitor and 1 MΩ load. The high-frequency half-wave rectification measurements are described in detail in [5,17]. The semiconductor layer thicknesses in the printed diodes were calculated based on the diode geometric capacitance (HP Network analyzer 8752A) and diode active area using a relative permittivity of 3 for the PTAA semiconductor.

2.2. Thermal inspection and short localization using Lock-in thermography

For thermal investigation and as a pre-step to further physical failure analysis, Lock-in thermography (LIT) was utilized following the electrical characterization. Hereby, the sample under investigation is stimulated electrically using a rectangle-shaped stimulation signal. This simulates a periodical ON/OFF switching of the device with a certain (lock-in) frequency. The used lock-in-frequency is chosen by the operator and lies in the range of a Hertz to several tens of Hertz. By stimulating the device electrically, heat is dissipated internally and the resulting temperature