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Organic thin films as flexible, large area X-ray and proton detectors for medical therapy

Beatrice Fraboni
Department of Physics and Astronomy, University of Bologna, Bologna, Italy
beatrice.fraboni@unibo.it

1. Introduction

High-energy radiation detectors based on organic semiconductor materials have already demonstrated to be promising candidates fulfilling the quest for large-area, flexibility, low power operation low-cost scalable fabrication [1-4]. All these requirements are essential in several application fields, from nuclear waste management to space mision monitoring, but the more relevant one is medical diagnostics and radio/hadrontherapy, thanks to the unique human-tissue equivalence of organic materials. Such property allows organic-based detectors to provide in real-time, without further calibration and data correction processes, the required information on irradiation dosimetry and spatial distribution.

Here we present our recent results on bis-(triisopropylsilylethynyl)-pentacene thin-films deposited from solution onto flexible substrates, as direct X-ray and proton detectors. We obtained record sensitivity for organic-based direct X-ray detectors of $1.3 \cdot 10^4$ μC $Gy^{-1} \cdot cm^{-2}$ and a very low minimum detectable dose rate (35 μGy s^{-1}) assessing their great potential for medical applications We clarified and modeled the photoconversion processes responsible for their excellent performance, also when compared with other emerging solution deposited materials, such as perovskites [4]. Figure 1 reports a comparison of recently reported organic/hybrid thin film detectors performance, including stae-of-the-art a-Se and ppolycrystalline CZT ones (not human tissue equivalent).

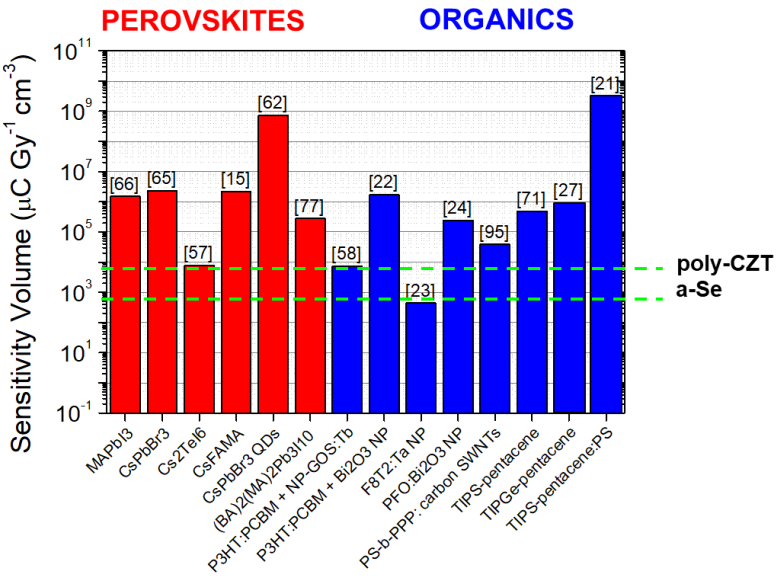

Figure 1: Comparison of top sensitivity values for organic and perovskite-based thin film high-energy radiation detectors: Histograms of the top sensitivity (a) and LoD (b) of different materials reported for OSCs- (blue) and PSCs- (red) based X-ray detectors.

2. Ionizing radiation detection processes

Organic thin film devices are characterized by an unexpected high X-ray sensitivity that we justified by interpreting the detection mechanism as a photo-modulation of the semiconductor conductivity due to charge accumulation during X-ray exposure (photoconductive gain). We proposed that, during X-ray irradiation, additional free carriers

are generated and accumulate in the organic thin film and to derive a model for such accumulation's impact on photocurrent we considered the differences in hole and electron carrier transport in organic materials [2]. In fact, the additional electrons and holes generated by the interaction with the X-ray follow a different fate: holes drift along the electric field until they reach the collecting electrode while electrons remain trapped. To guarantee charge neutrality, mobile holes that are collected at the collecting electrode are continuously re-injected from the injecting electrode, i.e. for each electron-hole pair created, more than one hole contributes to the photocurrent, leading ultimately to a photoconductive gain. Crucial for the amplification in this mechanism is the slow recombination dynamics of X-ray generated carriers, resulting here from the presence of deep trap levels which remove free electron carriers from the recombination process. We further investigated the origin of the physical processes and parameters controlling the minority carrier traps that assist the photoconductive gain effect and demonstrated that by reducing the grain size and increasing the number of grain boundaries, we can increase the density of electron trap states within the material, enhancing the photoconductive gain for the X-ray induced photocurrent. Further, by adding polystyrene to the semiconductor solution, we can reduce the interface hole trap density and consequently the charge carrier mobility is enhanced, as well as the device sensitivity [3].

3. Direct, real-time proton detection

We demonstrate the direct detection of 5 MeV protons by organic thin film devices that act as a solid-state detector in which the energy released by the protons within the active layer of the sensor is converted into an electrical current. such sensors demonstrate a stable and reproducible response to proton beams in a range of fluences between 10^9 -10^{11} H^+cm^{-2} and maintain a linear response up to a total dose of 28.5 kGy. By exploiting the structure of this sensor, two different operation modes can be effectively used: i) real-time mode sensing, where the amount of charges generated and collected at the electrodes is proportional to the released dose; ii) integration-mode sensing, where the energy released in the plastic substrate by the impinging protons generates static long lifetime charges that accumulate in the polymeric substrate and induce an increase of conductivity in the semiconducting layer (Figure 2). The measured sensitivity S = (5.15 ± 0.13) pC Gy^{-1} and limit of detection LOD = (30 ± 6) cGy s^{-1}, of the here proposed detectors assess their efficacy and their potential as proton dosimeters in medical proton-therapy.

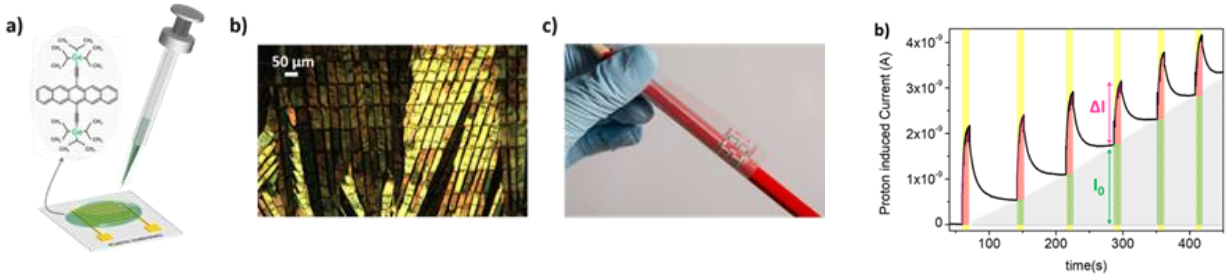

Figure 2. Organic Thin film-based device: architecture and morphology. a) Co-planar structure composed by two interdigitated electrodes (Au) deposited onto a plastic substrate (125 µm thick) followed by drop casting TIPGe-Pn film; b) Optical image of microcrystalline thin film. c) Flexibility of the here presented TIPGe-Pn – based detectors. c) Dynamic response curve (black line) to different fluences of incident protons ((4.5 · 10-11 – 6.6 · 10-11) H+cm-2). Two different contributions can be distinguished: ΔI (pink shadow) is the real-time response proportional to the dose (already described in Fig. 3), while I0 (green shadow) represents the baseline shift due to the fixed charges trapped in the plastic substrate

4. References

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