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Semiconducting Polymer X-ray Detectors: Towards Printable, Flexible, and Tissue Equivalent Devices

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Ionizing radiation is the foundation of modern society with rapid growth in medical diagnostics and treatment, space exploration, nuclear energy, and border security. However, increased use in recent decades has correlated to higher radiation exposures, with harmful effects to the health of workers and patients. Active monitoring has now become compulsory in many countries to instantaneously detect, evaluate, and correct for any deviations from the planned exposure. Ideal sensors for active monitoring must be positioned on the body and operated at low voltages to record the total biologically relevant radiation dose absorbed across a wide range of spectra and radiation sources, termed tissue equivalence. These devices should also be relatively transparent to ionizing radiation to minimize impact on protocols [1].

Printed organic semiconductors are to date the only flexible technology capable of active monitoring as a wearable, tissue equivalent sensor [2]. However, they have long been thought of as poor materials to sense ionizing radiation due to low sensitivity and poor tolerance towards high energy radiation. Here we develop a new system to solve these issues, reporting the optoelectronic, x-ray response, and radiation tolerance of an organic photodetector fabricated with solution-based inks prepared with polymer donor P3HT and non-fullerene acceptor o-IDTBR. The optoelectronic properties facilitated a high operating efficiency under x-rays without requiring any external bias when coupled with a plastic scintillator as a safe alternative to high voltage devices [3]. The thin, 500 nm, active layer was demonstrated to be ideal for high spatial resolution reconstruction of 50 μm FWHM radiation beams for novel modalities of Microbeam Radiation Therapy at the Australian Synchrotron, measuring a FWHM of (51 ± 1.4) μm across all 50 microbeams [4]. However, challenges with background luminescence from the polyethylene substrates commonly used for printed electronics were observed. Therefore, an alternative flexible substrate, polyamide (Kapton), was required. P3HT:o-IDTBR performance was found to be both higher and more stable on the Kapton substrates than P3HT:PCBM[5]. The tissue equivalence was determined by measuring the energy dependence of the detector responses across low and high x-ray energies. The response of devices coupled with the plastic scintillator matched the theoretical output of the plastic scintillator, validating the energy independence and tissue equivalence of the materials whilst commercial silicon detectors exhibited a 7-fold over-response compared to human tissue at low energies [6].

The devices also exhibited temporal responses approaching sub-microsecond time scales, the fastest ever observed for organic semiconductors as radiation detectors. High energy x-rays were produced from a Varian Clinac 2liX linear accelerator at 6 MV (<E>=1.2 MeV) with a pulse width of 3.6 μs, with individual pulses able to be resolved with temporal resolution directly comparable to state-of-the-art silicon radiation detectors. This result established organic semiconductors as the first ever ultrafast flexible devices for ionizing radiation with a tissue equivalent response.

The radiolucency was experimentally verified to be 99.8%, enabling operation as a transparent dosimeter with negligible perturbance of radiation beams. Device stability as a function of cumulative ionizing radiation dose past 5 kGy demonstrated P3HT:o-IDTBR was significantly more radiation tolerant than expected. P3HT:PCBM devices exhibited a 98.5% decrease in short circuit current density compared to just an 18.4% for P3HT:o-IDTBR under a radiation exposure equivalent to 10 years in a typical medical clinic. Mechanistic studies employing electrochemical impedance spectroscopy revealed photocurrent degradation occurred primarily in the P3HT polymer via formation of deep trapping sites in the P3HT:PCBM blends however, blends with o-IDTBR were more effective at preventing radiation induced damage. The reported devices provided a successful demonstration of the first fully organic ionizing radiation detectors, with further optimization of the scintillator required to enhance the sensitivity and tissue equivalence.

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