Evaluating the suitability of Organic Semiconductor Detectors for Nuclear Security

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Introduction

- He-3 used in neutron detectors becoming scarce therefore expensive
- Neutron detector usages:
    1. Security (detecting fissile materials shielded from gamma radiation)
    2. Scientific research (dark matter, radiation monitoring)
- Organic materials:
    - Tunable, **scalable**, low-cost
    - Can be used for detecting alpha, beta, gamma and x-ray radiation
    - Alpha detection possible, can adjust to neutrons

Organic semiconductor devices: How I build them

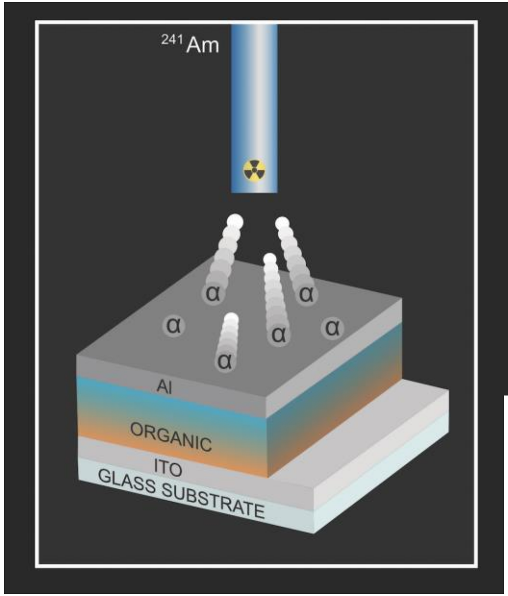
1. Photolithography: Pattern ITO on glass substrate into desired “fingers” shape
2. Drop-casting: Using a pipette, drop polymer/solvent mixture onto substrate (left to set)
3. Evaporation: Evaporate aluminium top contact through shadow mask
4. Uncovering: Dissolve/scrape away polymer to give access to ITO contacts

Device: each area of overlapping top/bottom contacts (typically 4 mm^2)

Figure 1: Step-by-step animation of device fabrication.

Organic semiconductor detectors: Alpha particles

- Use organic polymers to detect alpha particles
- Many different polymers and polymer blends used successfully internally (P3HT, P3HT:PC$_{60}$BM, PNDI(2HD)2T, etc.)
- Alpha particles come in, electron-hole pairs form (depending on HOMO/LUMO levels, medium's thickness), make a current when under a voltage bias
- Tested by opening and closing a shutter: significant increase in current when shutter is opened → radiation is being detected



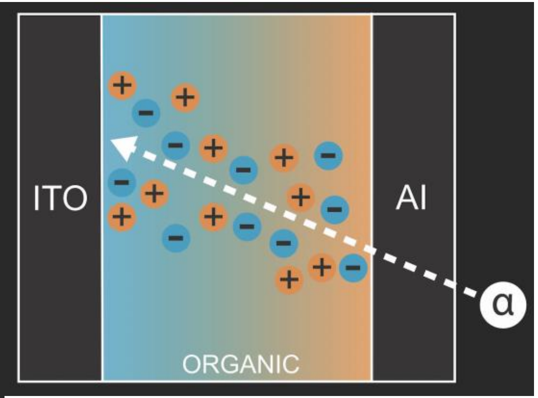


Figure 2: Left: Composition of a simple "diode-style" device. Above: Formation of electron-hole pairs in organic medium of devices irradiated by alpha radiation.

Organic semiconductor detectors: Neutrons

- Neutrons do not interact much with matter, need a different way to detect
- Thermal neutrons have a high cross section with ^{10}boron, making detectable alpha particles (and lithium)
- Solution: stir in some boron nanoparticles (NPs), i.e., boron carbide, boron nitride, etc.
- Issue: big problem with clumping and causing conductive paths between electrodes
- Solution in [1] to add a surfactant to prevent agglomeration
- Currently only alpha sources easily accessible without travelling to external neutron source (NPL, ISIS, AWE)

$$^{10}B + n \rightarrow \begin{cases} ^{7}\mathrm{Li}(1015)\mathrm{keV}) + \alpha(1777\ \mathrm{keV}) & 6\% \\ ^{7}\mathrm{Li}(840\ \mathrm{keV}) + \alpha(1470\ \mathrm{keV}) + \gamma(480\ \mathrm{keV}) & 94\% \end{cases}$$

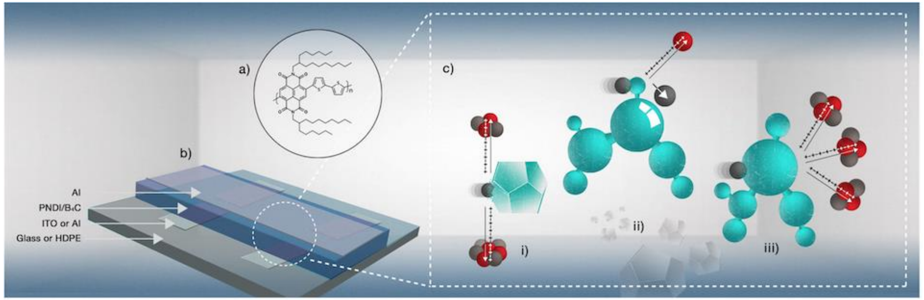


Figure 3: a) Chemical structure of PNDI(2HD)2T organic material. b) Schematic of four diode devices fabricated on a single substrate. c) Schematic cross section through a single device. Three schematic neutron interactions are indicated as i) thermal neutron ^{10}B capture at a B_4C microparticle, ii) elastic neutron-proton scattering at a PNDI H site, iii) ^{12}C decay into 3 α particles at a PNDI C site.

[1] Chatzispyroglou, P.; Keddie, J. L.; Sellin, P. J., *Boron-Loaded Polymeric Sensor for the Direct Detection of Thermal Neutrons*. ACS Appl. Mater. Interfaces **2020**, 12 (29), 33050-33057.

Organic material: PNDI(2HD)2T

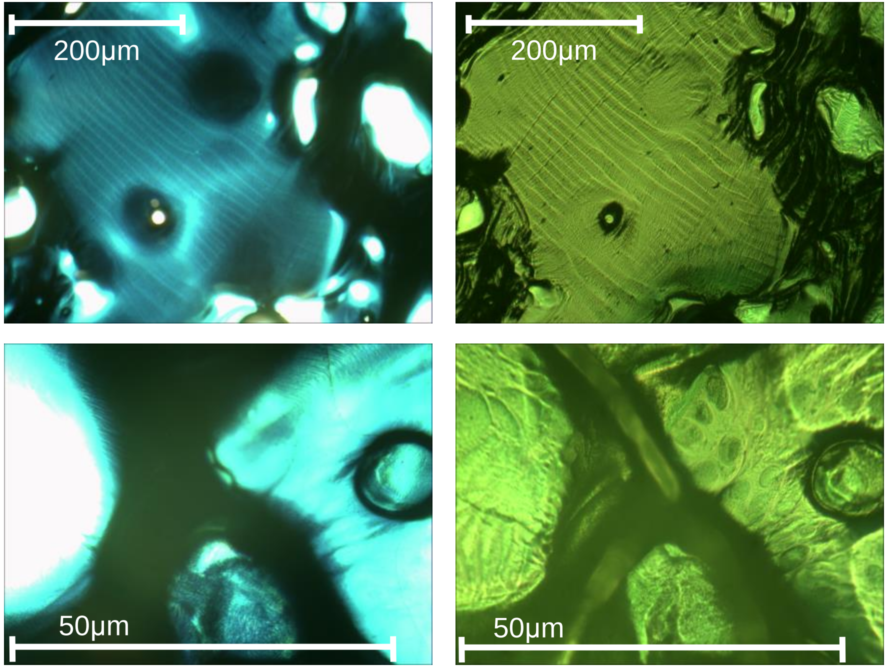


Figure 4: Pictures of a PNDI(2HD)2T film under a microscope. Left: Transverse images at x5 (top) and x20 (bottom) zoom. Right: Reflected images at x5 (top) and x20 (bottom) zoom.

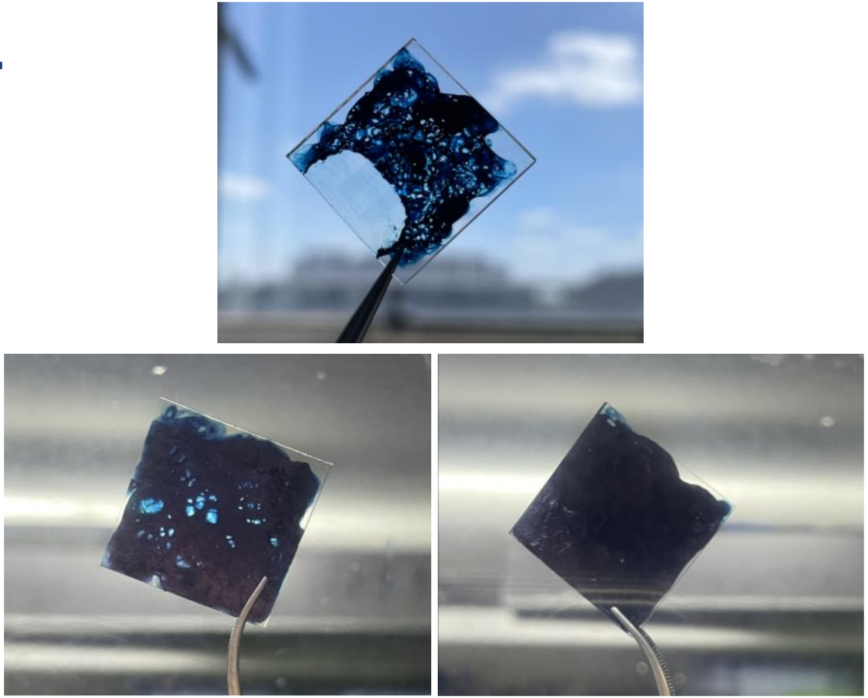
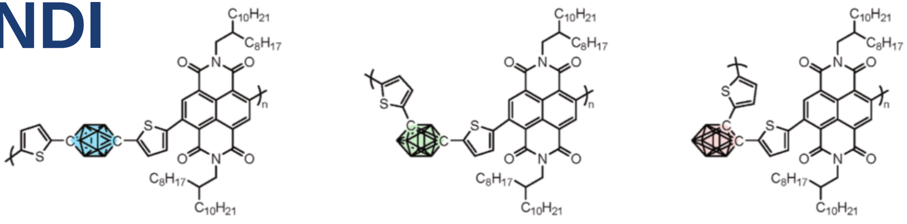


Figure 5: Top: Picture of PNDI(2HD)2T film made using typical method but lower concentration (film pictured under microscope in Fig. 4). Bottom Left: PNDI(2HD)2T film with same concentration but lower drop-casting temperature. Bottom Right: PNDI(2HD)2T film with same concentration but double the proportions and lower drop-casting temperature.

Organic material: Carborane-NDI

- Carborane: a polyhedral cluster of carbon, boron and hydrogen [2]
- Carborane derivatives highly relevant for medical applications, particularly for potential cancer treatments via boron neutron capture therapy
- Carborane-NDI: a variant of PNDI(2OD)2T with carborane directly incorporated in the molecular backbone
- If carborane-NDI can detect alpha particles, boron inside should allow it to detect neutrons like the nanoparticles
- Given a small supply of *meta*-, *ortho*-, and *para*-carborane-NDI by authors of [2]



pCbT_2-NDI *m*CbT_2-NDI *o*CbT_2-NDI

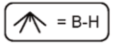


Figure 6: Top: Chemical compositions of the three isomer forms of carborane-NDI polymers (taken from [2]). Bottom: Picture of carborane-NDI films before top contacts were added via evaporation. Left: *o*CbT_2-NDI. Right: *p*CbT_2-NDI. A noticeable difference in colour may be seen in this picture between the two isomer forms.

[2] Aniés, F. *et al, N-type polymer semiconductors incorporating para, meta, and ortho-carborane in the conjugated backbone*, Polymer **2022** 240 124481.

*Some timings modified to allow overlapping regions of interest. No current response values have been modified.

Organic material: *Ortho*-carborane-NDI

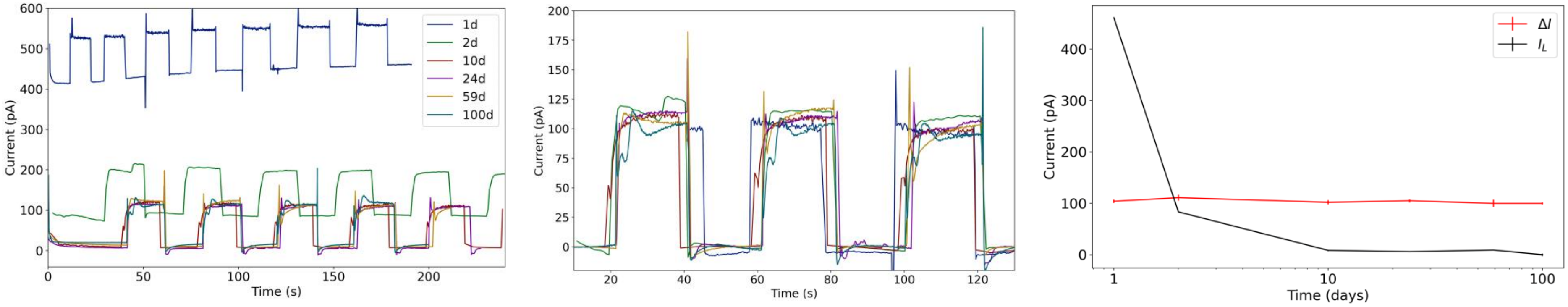


Figure 7: Left: Plot showing response of *o*CbT$_2$-NDI device under 10V bias being exposed to 370 kBq Americium-241 alpha source after various days' exposure to air over time. Middle: Same data*, first three periods of irradiation with leakage current (I_L) subtracted away, showing consistency of current enhancement (***ΔI***) being around 100 pA. Right: Variation of current enhancement and leakage current over the course of 100 days.

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DETECTOR DEVELOPMENT GROUP

Organic material: *Ortho*-carborane-NDI

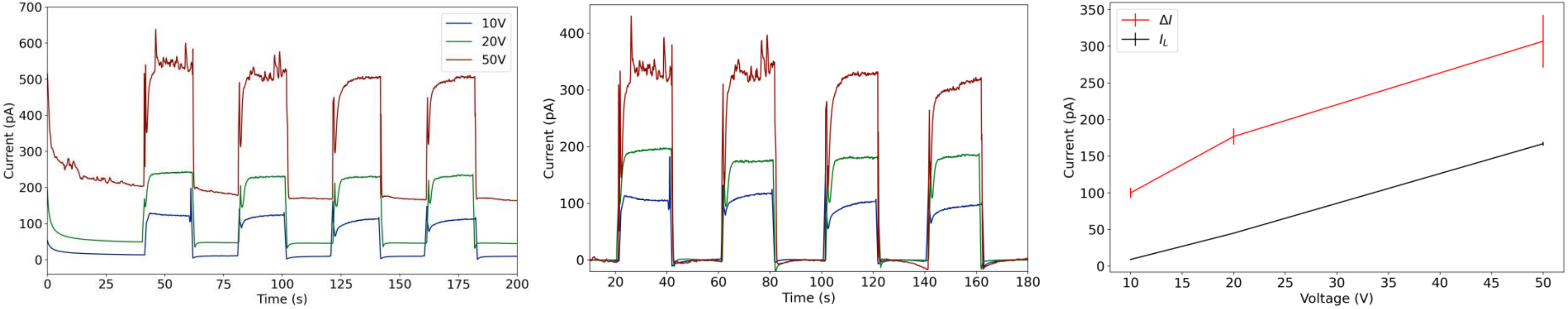

Figure 8: Left: Plot showing current response of *o*CbT_2-NDI device under different voltage biases being exposed to alpha source after 59 days' exposure to air. Middle: Same data, with leakage current subtracted away, highlighting changes in current enhancement. Right: Variation of current enhancement and leakage current with voltage bias.

Leakage current and current enhancement both increased with voltage bias, however enhancement appears to saturate whilst leakage continues to steadily rise

Large-area OSC detector

1. Scaled up detector
   - Simply take current method, make it larger
   - Less resolution
2. Array of detectors
   - Increase effective area
   - Triangulate location of source
   - Use a data acquisition device with multiple channels to simultaneously get results such as DAQ6510 with 7700 multiplexer

Figure 9: Keithley DAQ6510 with 7700 multiplexer.

Detector array: Current circuit design

- Capacitor bank to integrate charge over time, increase signal-to-noise ratio, higher standoff distance:

$$CV_0 = I\Delta t$$

includes four unknowns to be appropriately obtained. *Ortho*-carborane-NDI results in I ~ 100 pA under a voltage bias V_0 ~ 10V and time to walk past a device Δt ~1s lead to choosing a C = 10 pF capacitor.

- A closed → A opened, B closed → B opened, C closed → repeat
- Battery charges capacitor to V_0, which discharges through diode, resulting in lower voltage across capacitor when read out after 1s

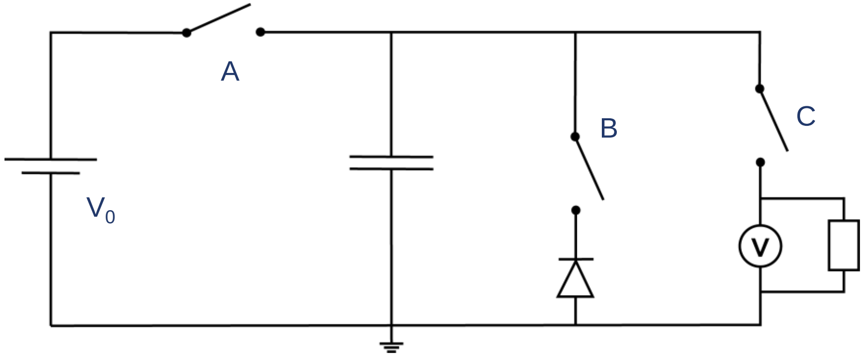

Figure 11: Circuit diagram of single detector-capacitor system. Switches and voltage readout done by multiple channels of a multiplexer. This diagram may be scaled to include multiple capacitors, detectors, and readouts, all working together.

Conclusion

- Successfully manufactured an alpha radiation detector using a new type of organic polymer
- Successful alpha detection implies neutron detection should be possible with carborane-containing polymers; will be tested in visit to NPL neutron source next month
- Standard-sized *meta*-, *ortho*-, and *para*-carborane-NDI devices have been manufactured in the lab awaiting characterisation
- Can make many comparisons with standard PNDI(2HD)2T, explored more in-depth during NuSec poster session
- PNDI(2OD)2T should be better to compare with carborane-NDI, being the basis of these polymers
- The use of an array of detectors attached to a capacitor bank should allow for removal of noise increasing step-off distance and allow triangulation of radiation sources

Thank you

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Latest group publication:

Ref 1:

Ref 2:

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