Low-Cost Semiconductor Perovskite Radiation Detectors for Medical Imaging and Security Applications

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Purpose: Methylammonium lead tribromide ($CH_3NH_3PbBr_3$, MAPbBr3, or MAPB) is a solution grown perovskite semiconductor that possesses desirable qualities for radiation detection in medical and national security applications. In addition to suitable mobility-lifetime product and bulk resistivity, MAPB has a high effective atomic number as well as an acceptable density (~4 g/cm^3) for efficient gamma and X-ray sensing [1]. As a direct conversion detector that can be grown in large volumes, it has the potential to replace current X-ray imaging detectors such as indirect conversion CsI:Tl scintillators grown on amorphous silicon, or amorphous selenium that has poor transport properties and stopping power for hard X-rays [2]. Moreover, its low cost, up to 50% atomic fraction of hydrogen for fast neutron sensing, ability to incorporate lithium for thermal neutron sensing, and potential for medium resolution gamma spectroscopy make MAPB an attractive alternative to the current gamma/neutron detector arsenal for homeland security. This work presents notable milestones in the development of large area polycrystalline MAPB and MAPB single crystals (SC) at the University of Tennessee that demonstrate its versatility and potential for meeting radiation detection needs in the medical and national security fields.

Methods: The MAPB crystals were grown via inverse temperature crystallization from a precursor solution of MABr and $PbBr_2$ dissolved in dimethylformamide (DMF). $Li_{1-x}MA_xPbBr_{3-y}Cl_y$ single crystals were prepared by stoichiometric substitution of MABr with MACl and LiCl during the precursor solution preparation. The SCs were then mechanically polished with diamond lapping pads, followed by a chemomechanical treatment with DMF to produce a mirror finish (Figure 1a) [3]. Polycrystalline samples were only lightly treated with DMF without mechanical polishing (Figure 1b). Electrical contacts were deposited via plasma sputtering in either a Cr/MAPB/Cr scheme for polycrystalline MAPB or Cr/MAPB/SnO_2/Cr scheme for SCs.

X-ray exposure with a polycrystalline MAPB wafer (2.74 cm^2 area and 0.34 cm thickness) utilized a Moxtek ® MAGPRO 60 kV 12W tube with a tungsten anode and a Keithley 6487 Picoammeter/Voltage source. X-ray imaging was performed using a stepper motor to move a brass key to predefined grid positions (5 mm vertical and 2 mm horizontal steps) between the detector and X-ray source, which were both stationary. The X-ray source, which was operated at 300 µA and 40 kV, was 5 cm away from the MAPB wafer biased at 50V. At each key position, the induced current on the detector was recorded and tabulated to produce an image. X-ray flux was adjusted by adjusting the current applied to the X-ray tube from 300 µA to 30 µA in 30 µA increments while holding a constant tube potential of 40 kV. The dose rate to the MAPB wafer using manufacturer provided specifications was between 0.13 and 1.38 Gy/s while the detector was biased at 50V.

Pulse mode radiation exposure was performed in a custom aluminum enclosure with a CREMAT CR-110 preamplifier, ORTEC 527A amplifier (10 µs shaping time and gain of 100) and ORTEC ASPEC-927 MCA. SCs were exposed to a 0.1 µCi ^{210}Po alpha particle source, separated from the detector by approximately 6 mm of air, three gamma sources and one non-spectroscopic X-ray source (10 µCi ^{133}Ba, 10 µCi ^{137}Cs, 100 µCi ^{57}Co, 0.1 µCi ^{241}Am respectively) separated from the detector by approximately 2 cm of air, and a moderated 2 Ci ^{239}PuBe neutron source.

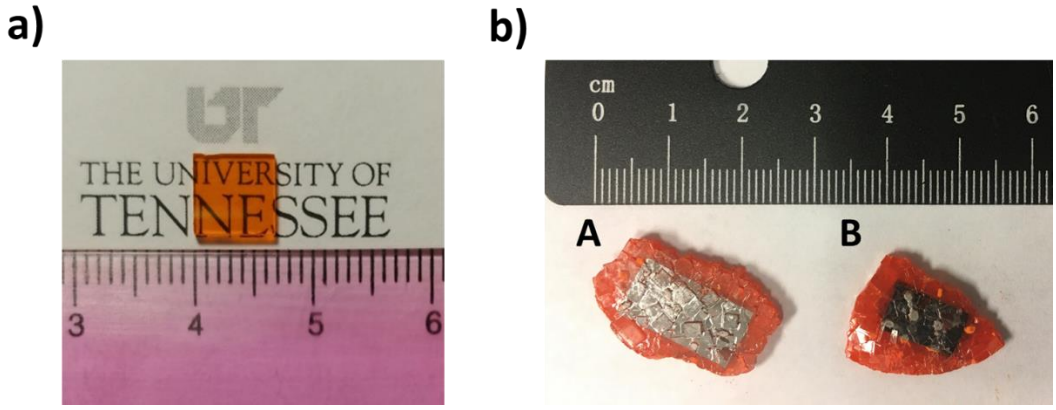

Figure 1. a) MAPB single crystal, b) Fabricated polycrystalline MAPB detectors

Results: An image of a brass key (Figure 2a) was produced from the polycrystalline MAPB wafer with sufficient resolution to resolve the key's general features. Figure 2b also shows that an increase in dark current after an irradiation experiment did not result in a decreased net indued current in the second experiment. Collectively, these results demonstrated that the sensitivity of MAPB was sufficient for producing X-ray images and could be maintained for significant amounts of irradiation time.

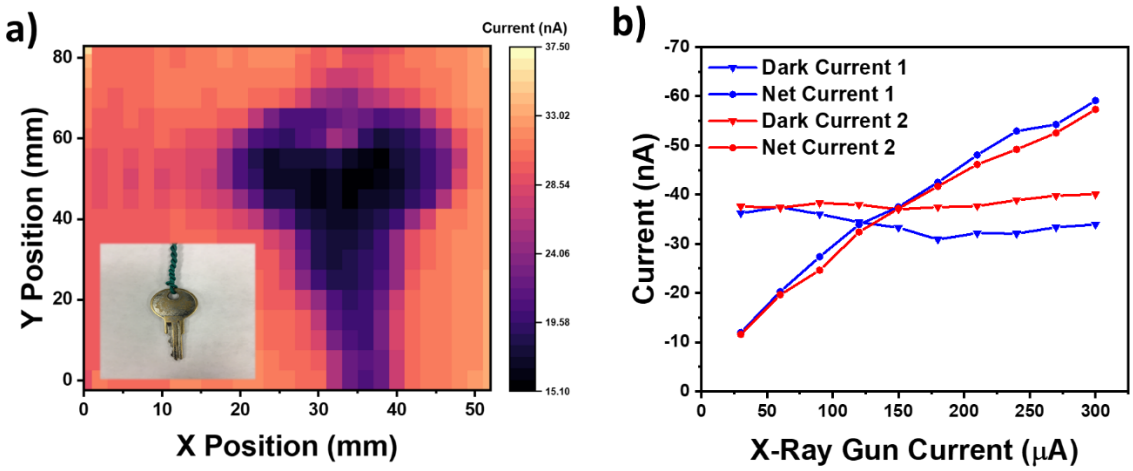

Figure 2. a) X-ray image of a brass key from MAPB, b) Repeated current vs. X-ray flux measurements showing relatively stable sensitivity

$MAPbBr_{2.85}Cl_{0.15}$ SC response to a non-spectroscopic ^{241}Am smoke detector source showed an X-ray photopeak resolution of 15%, which was comparable to the best resolutions achieved in lead halide perovskite sensors [4]. However, variable ballistic deficit due to poor charge transport properties hindered spectroscopic response from highly penetrating gamma rays (Figure 3b). Finally, thermal neutron sensing was made possible with a 2.5% LiCl substitution during growth to produce a sensor with less than 150 ppm ^{6}Li (LiMAPB). Despite this low concentration, exposure to a PuBe source still yielded promising results for thermal neutron sensing, as various shielding configurations were implemented to isolate what was potentially the thermal neutron component (Figure 3d blue shaded region) from ^{113}Cd neutron capture gamma rays (Figure 3d green shaded region) and PuBe gamma rays.

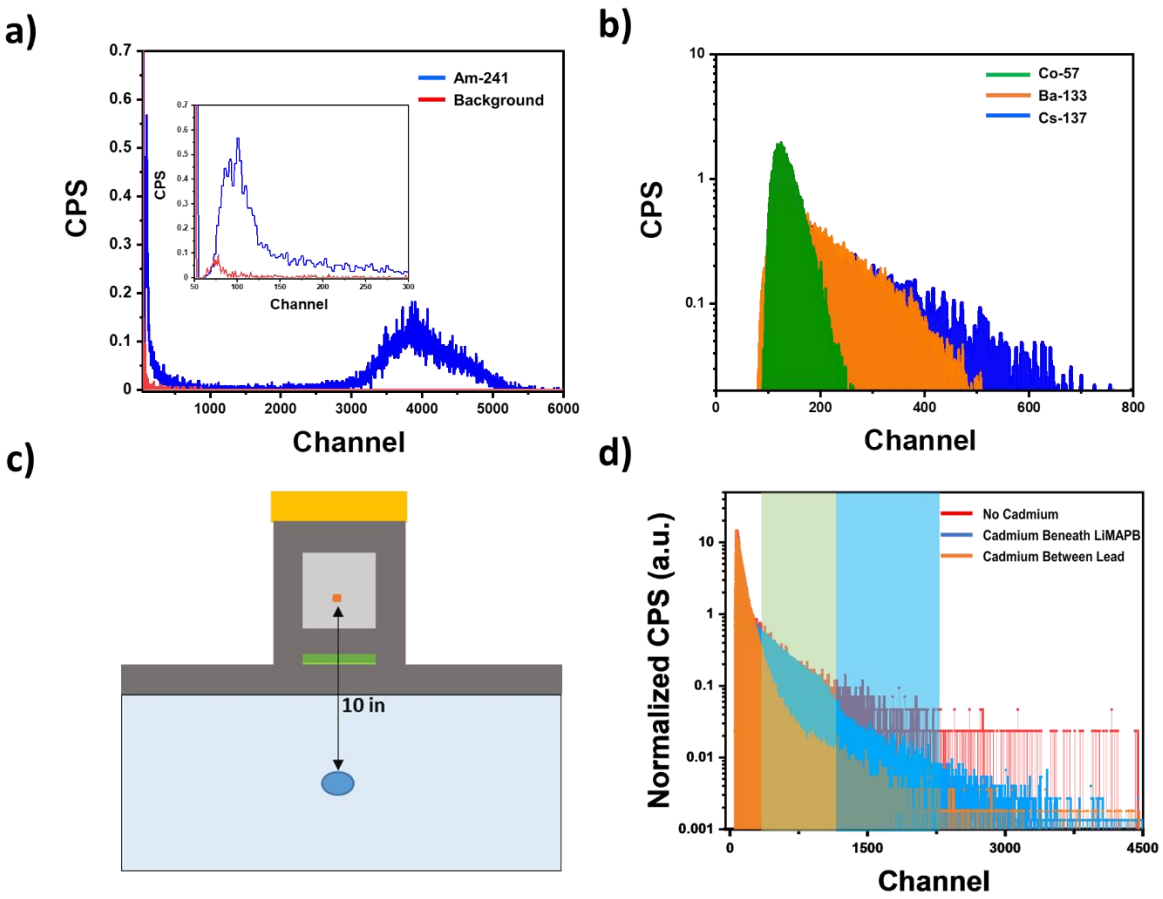

Figure 3. a) Alpha peak and X-ray photopeak (insert) from a non-spectroscopic ^{241}Am smoke detector source observed in $MAPbBr_{2.85}Cl_{0.15}$, b) Background subtracted spectra from various gamma ray sources with LiMAPB, c) Shielding arrangement for LiMAPB thermal neutron sensing with PuBe (dark blue), HDPE (light blue), lead (gray), cadmium (green), and paraffin (yellow), d) LiMAPB response to PuBe with multiple shielding configurations

Conclusions: The development of MAPB at the University of Tennessee has demonstrated its potential for stable X-ray imaging as well as its performance as a radiation detector for national security purposes This justifies continued research to overcome observed deficiencies in charge transport and long-term stability, and current efforts are aimed towards single carrier charge sensing techniques and computationally correcting for charge losses during transport which will lead to improvements in sensitivity and spectroscopic capability.

Relevance to CIRMS: This work is relevant to the CIRMS mission in two ways. First, large area X-ray imaging detectors with high efficiency and sensitivity are critical to minimizing patient dose during imaging operations. Second, lowering the cost of production will allow for detectors to be more widely deployed for national security and public safety operations. The first author aims to bridge the gap between nuclear security science and policy, and currently supports multidisciplinary nonproliferation efforts at the Y-12 National Security Complex.

References:

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