ALD of SnO$_2$ as the active component of a Plastic Microchannel-Based Direct Fast Neutron Detector

Philippe de Rouffignac, Neal Sullivan, Anton Tremsin, Dmitry Gorelikov, David Beaulieu - Arradiance

Adam Hock, Jaeyeong Heo, Roy Gordon – Harvard University
Outline

- Arradiance and the Microchannel Plate Amplifier (MCP)
- Motivation I & II
- Theory behind proposed device
- ALD/Film Requirements for Plastic MCP
- SnO₂ ALD Results
- Plastic MCP Beam line Results
  - Efficiency
  - Timing
- From the lab to the field
What is a Micro Channel Amplifier?
Very Fast – Very Low Noise - Charged Particle Amplifier

Single Micro Channel Amplifier

Input Electron

Bias 1000 V

Amplification (SE Cascade)

High Gain up to 1e6
Low noise – Very fast
Pico Second Response

Micro Channel Plate (MCP -Array of pores)

Micro Channel Plate Used In Light Amplification

Finished Night Vision Tube

NV Application

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A 50 year old MEMS Process

Substrate Fabrication

1" Etch-able Core
Lead Glass Rod

Draw Tower

Stacked
Draw Tower
Repeated

Boule
5-100mm Dia

Diced
0.2-0.3 mm thick

Etched
Producing >5M
2-10 um pores

Substrate Functionalize

Furnace H₂ Firing
Both conduction and emission layer produced simultaneously; cannot be optimized independently

TABLE 2
Elemental composition of MCP glass

<table>
<thead>
<tr>
<th>Z</th>
<th>Element</th>
<th>Weight percent</th>
</tr>
</thead>
<tbody>
<tr>
<td>82</td>
<td>Pb</td>
<td>47.8</td>
</tr>
<tr>
<td>8</td>
<td>O</td>
<td>25.8</td>
</tr>
<tr>
<td>14</td>
<td>Si</td>
<td>18.2</td>
</tr>
<tr>
<td>19</td>
<td>K</td>
<td>4.2</td>
</tr>
<tr>
<td>37</td>
<td>Rb</td>
<td>1.8</td>
</tr>
<tr>
<td>56</td>
<td>Ba</td>
<td>1.3</td>
</tr>
<tr>
<td>33</td>
<td>As</td>
<td>0.4</td>
</tr>
<tr>
<td>55</td>
<td>Cs</td>
<td>0.2</td>
</tr>
<tr>
<td>11</td>
<td>Na</td>
<td>0.1</td>
</tr>
</tbody>
</table>

*Density ~ 4.0 g/cm³.

Wiza, Nuclear Inst. & Meth., Vol 162, 1979, 587
Arradiance MCP Technology

◆ Substrate
  ◆ Rigid and electrically insulating

◆ Conductive layer
  ◆ $\sim 10^{13} - 10^{14}$ Ohms/Sq
  ◆ Conformal & uniform up to 200 : 1
    ◆ Thickness and Resistivity
  ◆ Low field effects = Low TCR

◆ Emissive layer
  ◆ Conformal & uniform
  ◆ High secondary yield
    ◆ Contaminants can effect yield

◆ MCP Device
  ◆ High Gain
  ◆ Resistance stability and matching
  ◆ Stable gain following “scrub”
  ◆ Low outgassing
Process: Conductive film
Process: Conductive film - TCR

<table>
<thead>
<tr>
<th></th>
<th>TCR</th>
</tr>
</thead>
<tbody>
<tr>
<td>Film A</td>
<td>0.012</td>
</tr>
<tr>
<td>Film B</td>
<td>0.0098</td>
</tr>
</tbody>
</table>

Thermal coefficient of resistance on par ($B_T < 0.01$) with current state-of-the-art for two Arradiance conductive films.
Process: Secondary electron yield & device gain
Results - Incom 66:1, 20um, 60% OAR
March 2010
Improved Lifetime of Thin Film MCP over Conventional

- Reduced ion feedback
  - Reduce diffusion of mobile ions
- Sustained emission layer response with extracted dose
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Motivation in Two Parts

Scientific Curiosity
- All microchannel plate amplifiers on the market are made from a glass substrate
- Can Arradiance make an MCP out of a seemingly more challenging material like plastic?
- Is there a way to make our high temperature MCP films compatible with plastic?
- What could a functioning plastic MCP be used for?
  - Large area robust MCPs?
  - MCP-PMTs?
  - Detectors?

Revenue Generating Applications
- Detection of Special Nuclear Materials
- Fast neutron counting/spectroscopy
Plastic MCP Applications

Large area MCP (current)

COS detector Hubble telescope

Plastic MCPs are robust and can be potentially be made in large areas for less cost

Market (now): $100k/year

Large Area (>4”) MCP-PMT (Future)

Homeland security X-Ray detection: $100M/year

Medical Imaging: $200M/year


*NURLE TECHNOLOGIES, INC. http://www.burle.com/mcp_pmts.htm
† Philips Healthcare

Nuclear Detection

“Nuclear Car Wash” (Livermore Concept)

Active Scanning

Passive Scanning

Neutron-proton interaction yields detection capabilities

Potential replacement candidate for He-3 detectors

Market: >$1B/year
SNM detection technology overview

- Hydrogen-rich PMMA microchannel structure
- Graded Temperature ALD deposition
  - Active films deposition at 140°C
- Neutron-proton recoil reaction within plastic at better than 1% efficiency
- Proton initiated secondary electron cascade
- Output pulse $10^3 - 10^6$ electrons
- Standard readout electronics
- Technology scalable to large format
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Some polymer candidates and a precursor candidate

<table>
<thead>
<tr>
<th>Material</th>
<th>Tg</th>
<th>MP</th>
<th>CTE</th>
<th>Water Absorption</th>
<th>H Content (mol H/cm³)</th>
<th>Is Substrate Manufacturable?</th>
</tr>
</thead>
<tbody>
<tr>
<td>Radel-R5000 (a polyphenylsulfone)</td>
<td>220°C</td>
<td>360°C</td>
<td>56 µm/m-°C</td>
<td>0.4%</td>
<td>0.018</td>
<td>No</td>
</tr>
<tr>
<td>PMMA</td>
<td>105°C</td>
<td>160°C</td>
<td>75 µm/m-°C</td>
<td>0.3%</td>
<td>0.094</td>
<td>Yes</td>
</tr>
<tr>
<td>HDP Polyethylene</td>
<td>-78°C</td>
<td>130°C</td>
<td>25 µm/m-°C</td>
<td>0.05%</td>
<td>0.073</td>
<td>Yes</td>
</tr>
<tr>
<td>Polypropylene</td>
<td>-10°C</td>
<td>165°C</td>
<td>90 µm/m-°C</td>
<td>0.01%</td>
<td>0.128</td>
<td>Work in progress</td>
</tr>
</tbody>
</table>

- SnO₂ as conductive layer, Al₂O₃ as emission layer
- Tin (II) cyclic stannylene – Gordon group Harvard
  - 30 Torr at 60°C
  - Reacts readily with hydrogen peroxide
  - **ALD window 50-150°C**
- Conductive
- Compatible with TMA
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SnO₂ ALD

(1) Sn precursor saturation

\[
\begin{array}{c}
\text{Deposition Rate (nm/cycle)} \\
0 & 0.04 & 0.08 & 0.12 & 0.16 & 0.20 \\
0 & 1 & 2 & 3 & 4 & \text{Number of Sn Precursor Doses}
\end{array}
\]

(2) AIOX test @120C (TMA+H₂O₂)

\[
\begin{array}{c}
\text{Refractive Index} \\
1.86 & 1.88 & 1.90 & 1.92 & 1.94 & 1.96 & 1.98 \\
\text{Thickness (nm)} \\
5 & 10 & 15 & 20 & 25 & 30 & 35 & 40 & 45 & 50 \\
0 & 100 & 200 & 300 & 400 & \text{Cycles}
\end{array}
\]

\[
\text{data} \\
\text{linear fit} \\
Y = 2.68 + 0.104X \\
G/R: \sim 1 \AA/\text{cycle}
\]

\[
\begin{array}{c}
\text{SnO}_2 \\
\text{rac-} \quad \text{H₂O₂} \\
t-\text{Bu} \\
t-\text{Bu} \\
\text{ALD} \\
\end{array}
\]

For further discussion of ALD characteristics of this precursor system see talks given by Roy Gordon ALD 2010 and Adam Hock ALD 2010

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Properties vs Aspect Ratio

- Nanolaminate structure of SnO₂ and Al₂O₃
- Deposition temp 85°C

Gradient of film thickness for current process
- Likely resistivity gradient as well
- Goal: flatten this curve, then create MCP devices
Plastic substrate MCP (alternative material)

- Reasonable gain for electron amplification, limited by L:D
- Uniform response
- Stable operation
- ALD at higher temperatures (limits plastic choices)
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Detector Hardware Experimental Setup

- 2 & 5 mm polymer MCP, ~50 µm pores, 20 µm walls, 5° bias angle
- Installed above a chevron stack of 50:1 L/D MCPs
- Phosphor screen readout
- Canberra preamp and postamplifier
Neutron detection simulation

\[ P_{\text{detection}} = P_1 \times P_2 \times P_3 \]

- \( P_1 \) – n-p recoil within the MCP substrate
- \( P_2 \) – proton escape into MCP pore
- \( P_3 \) – electron avalanche is formed (MCP \( \sim 1 \))

\[ P_1 \text{ probability} \]

- Pores are 50 \( \mu \text{m} \)
- \( En = 2 \text{ MeV} \)

\[ P_2 \text{ probability} \]

- 2 MeV neutron, 50 \( \mu \text{m} \) pores

\[ P_1 \times P_2 \text{ probability} \]

- Pores are 50 \( \mu \text{m} \)
- \( En = 2 \text{ MeV} \)

\[ P_1 \times P_2 \times P_3 \approx 1\% \text{ for 2MeV neutrons with 20\( \mu \text{m} \) pore walls} \]
Efficiency Results: UNH Beam Line

Isotope sources:
Placed 6” from detector
Stilbene scintillator with a single channel PMT (UNH) for calibration
Cf-252, Am-241/Be \((n, \gamma)\)
Cs-137, Co-60, Am-241 \((\gamma)\)

Cf-252
Neutron Detection Efficiency

Face-on
\[ n \text{ QE} = 0.747\% \]

Edge-on
\[ n \text{ QE} = 2.46\% \]

Measured neutron efficiency matches theoretical (0.8%)
Low dark counts (dark count \(\sim 0.3 \text{ c/cm}^2\text{s}\))

Gamma only source

<table>
<thead>
<tr>
<th>(\gamma)-energy</th>
<th>Face-on</th>
<th>Edge-on</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.122 MeV</td>
<td>0.15%</td>
<td>0.33%</td>
</tr>
<tr>
<td>0.661 MeV</td>
<td>0.6%</td>
<td>1.5%</td>
</tr>
<tr>
<td>(\sim 1.2) MeV</td>
<td>1.3%</td>
<td>2.87%</td>
</tr>
</tbody>
</table>

\[\text{Gamma energy (MeV)}\]

\[\text{QE, face-on} \]
\[\text{QE, edge-on} \]

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Timing and Coincidence

Experiment Summary
- Using 2 detectors offset and some distance apart
- Measure events in Arradiance and commercial detectors
- Gamma or neutron signal detected by Arradiance - starts acquisition window and timer for scintillator
- Time-of-flight is calculated for each event
- Statistics collected on each TOF and analyzed

Coincidence measurements for gamma (35 cm distance)
- Liquid scintillator detector (BC519) (stop signal at TAC)
- Plastic MCP detector (start signal at TAC)
- Gamma and neutrons from Cf$^{252}$
- Recoil neutrons
- Angle $\alpha$

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Coincidence gamma rejection plus timing through TOF

Gamma travel at speed of light – detection in two detectors should happen within ~1 ns

Recoil neutrons arrive with a delay $dT$ to detector2

Temporal Resolution

$\sim 3-4$ ns

Nanosecond resolution = differentiation between incoming gamma and fast neutron radiation
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Combining multiple 50mm plastic MCPs QE goes up

- With 10 planes QE is \(\sim60\%\)
- In coincidence mode \(\sim10\%\)
Coincidence techniques can differentiate Gammas and Neutrons

Combining coincidence with the high efficiency cube yields a state-of-the-art detector

- Provides directionality
- Provides discrimination between neutrons and gammas
- Is sensitive to a large energy range of neutrons and less sensitive to low energy background gammas (not shown)

Compares favorably with liquid scintillator technology

3 x 3 x 3 cube array in an aluminum enclosure
Directionality of source in all spatial dimensions
Acknowledgements

- Dr. James M. Ryan, Professor of Physics, University of New Hampshire
- Mr. Jason S. Legere, Research Project Engineer III Space Science Center, University of New Hampshire
- Dr. Richard Lanza, Senior Research Scientist, MIT Dept. of Nuclear Science and Engineering
- Dr. Gordon Kohse, Ph.D; Principal Research Engineer, MIT Nuclear Reactor Laboratory
- The rest of the Arradiance Team
- DOE LAPPD Collaboration
- NASA SBIR NNX10CD59P
Background
Neutron detection simulation: proton recoil - P1

PMMA \((C_5-O_2-H_8)_n\)

- monomers / cm\(^3\): 7.16x10\(^{21}\)
- H atoms / cm\(^3\): 5.73x10\(^{22}\)
- C atoms / cm\(^3\): 3.58x10\(^{22}\)
- O atoms / cm\(^3\): 1.43x10\(^{22}\)

Cross section of neutron interaction

\[ P = [1- \exp(-N_i \, \sigma_i \, L)](1-A) \]

50 µm circular pores, 20 µm walls, 1.19 g/cm\(^3\)
D-T Source (Thermo 320) Experimental Setup

Filters: Lead (2’’), polyethylene (1”, 2”), borated plastic (1”)

- Polyethylene shielding around the source
- Lead shielding around the detector
- MCPs: 5 mm PMMA MCP, ~50 µm pores, 20 µm walls, 5° bias angle installed above a chevron stack of 50:1 L/D MCPs

Technical Specifications

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Specification</th>
</tr>
</thead>
<tbody>
<tr>
<td>Neutron Yield</td>
<td>1.0E+08 n/s</td>
</tr>
<tr>
<td>Neutron Energy</td>
<td>14 MeV</td>
</tr>
<tr>
<td>Typical Lifetime</td>
<td>1,200 hours @ 1x10^8 n/s</td>
</tr>
<tr>
<td>Pulse Rate</td>
<td>250 Hz to 20 kHz, continuous</td>
</tr>
<tr>
<td>Duty Factor</td>
<td>5% to 100%</td>
</tr>
<tr>
<td>Minimum Pulse Width</td>
<td>5 µsec</td>
</tr>
<tr>
<td>Pulse Rise Time</td>
<td>Less than 1.5 µsec</td>
</tr>
<tr>
<td>Pulse Fall Time</td>
<td>Less than 1.5 µsec</td>
</tr>
<tr>
<td>Maximum Accelerator Voltage</td>
<td>95 kV</td>
</tr>
<tr>
<td>Beam Current</td>
<td>60 uamps</td>
</tr>
</tbody>
</table>