SRNL™
SAVANNAH RIVER NATIONAL LABORATORY
Operated by Savannah River Nuclear Solutions, LLC

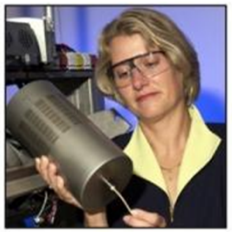
We Put Science To Work

Radiation Detection with Nanomaterial-Based Gas Filled Detectors

Lindsay T. Sexton[1]

Steven M. Serkiz[1], Joeseph Cordaro[1], Tim Devol[2], Apparao Rao[2]

ACS Spring 2011 Meeting, March 27, 2011



[1]Savannah River National Laboratory

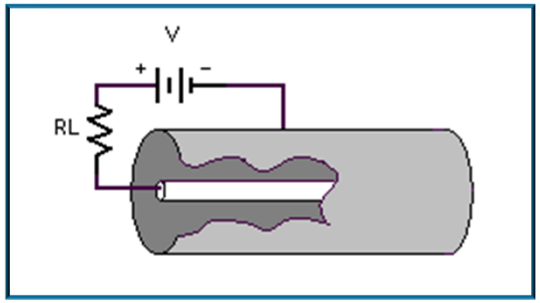
[2]Clemson University

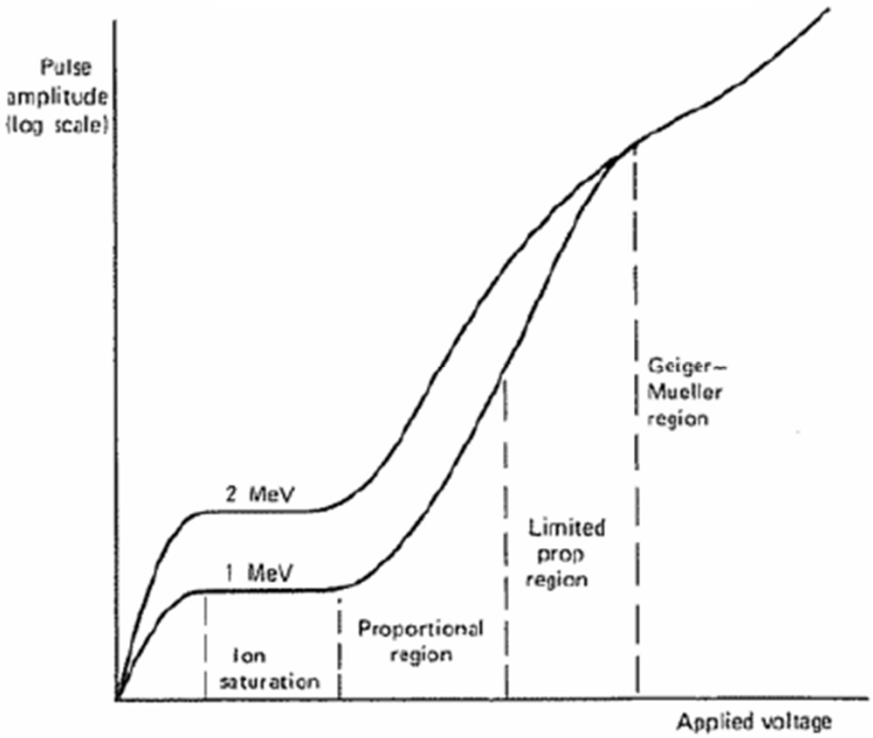
SRNL-L7100-2012-00040

Outline

- **Background**
  - Gas-Filled Radiation Detectors – Proportional Counters
  - Scale Effects
  - Carbon Nanotube Anodes
- **Fabrication of Nanostructured Arrays**
- **Electrical Properties of Arrays – HV Breakdown Studies**
- **Preliminary Results – Radiation Detection with Arrays**
- **Summary and Path Forward**

Background: Gas-Filled Ionizing Radiation Detection





Knoll, 2000

- **Depending on the applied potential of the instrument:**

  - ionization chambers, proportional counters (PCs), or Geiger-Muller detectors

- **Of these detectors, PCs are capable of detecting a range of radiation types and energies.**

  - gas multiplication to amplify the charge

  - pulse generated proportional to energy of the radiation

  - electrical potential required: 1-2 KV

SRNL

Background: Proportional Counter - Scale Effects

The electric field strength for a typical cylindrical PC at radius r is:

$$E = \frac{V}{r \ln(b/a)}$$

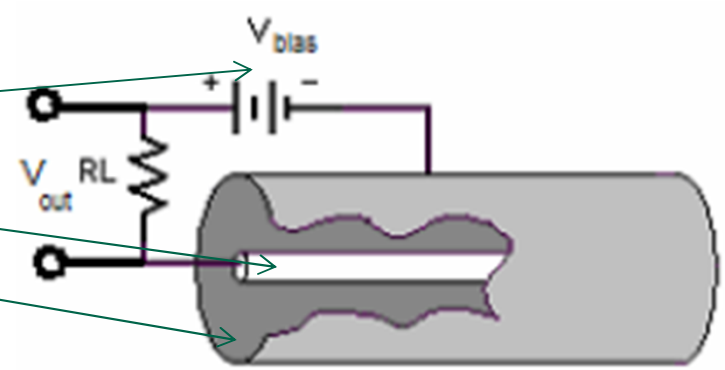
where

V = voltage applied between the anode and cathode

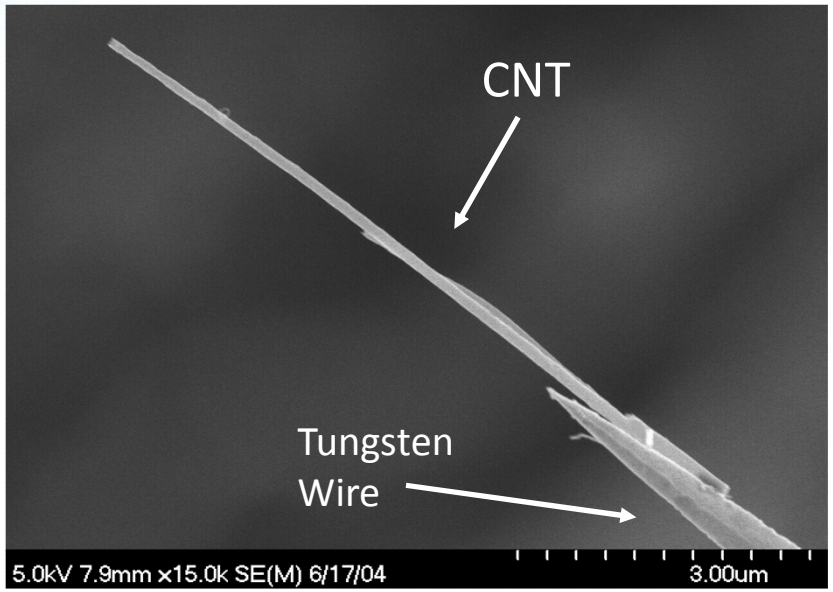
a = anode wire radius

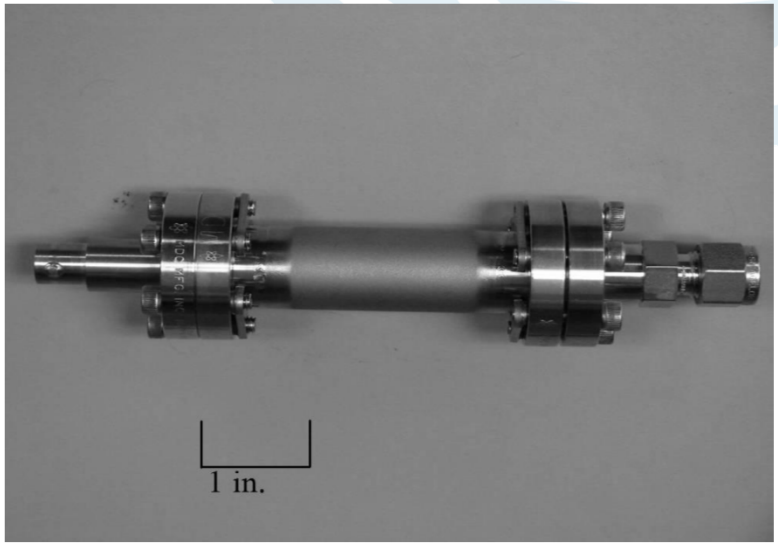
b = cathode inner radius

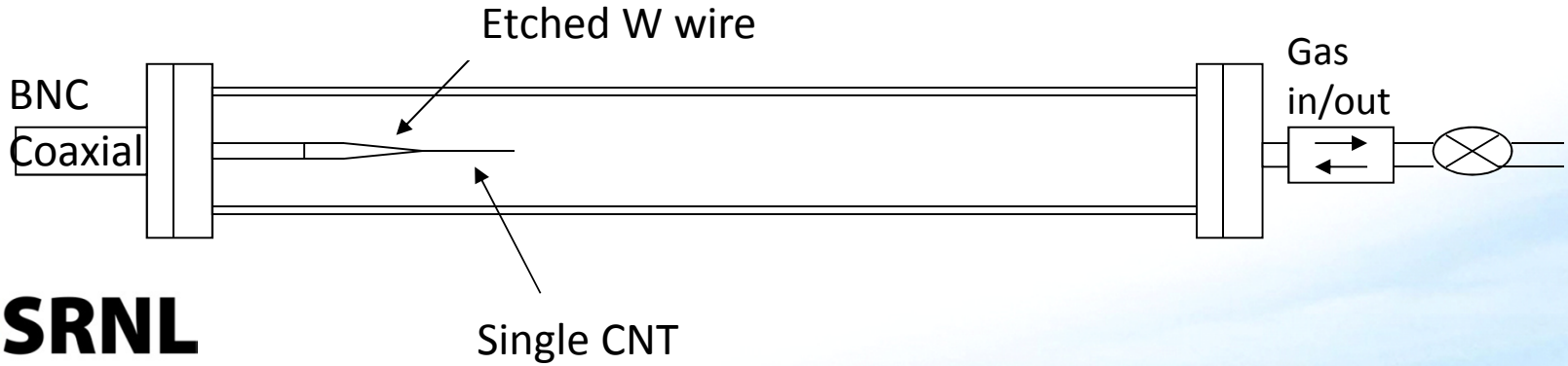
Field strength for the ***n*-PC will have electric field strength 570 times the micro scale**. Or the ***n*-PC could operate at 3.5 volts** and have the same electric field strength at the surface of the anode.



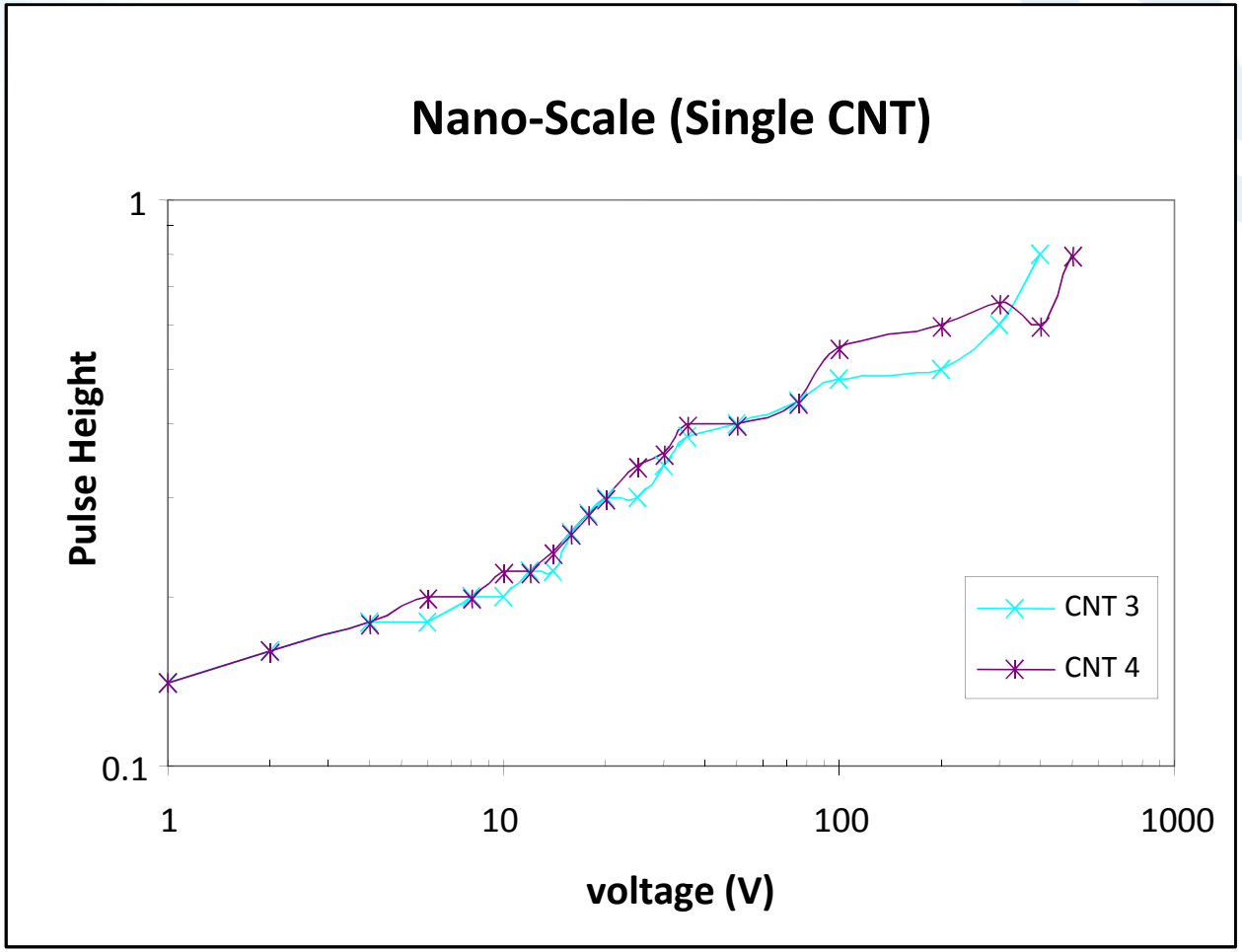
Scale Effects: Single Carbon Nanotube (CNT) PCs







Scale Effects: Single Carbon Nanotube (CNT) PCs



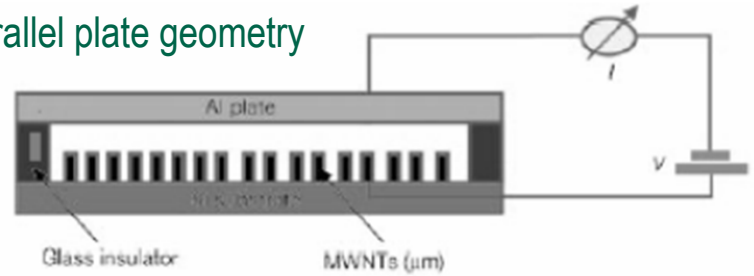
Experimental conditions:
- coaxial geometry
- Sr-90 source (activity of 20 mCi in Nov. 1980)

Single CNT – low detector volume/slow response

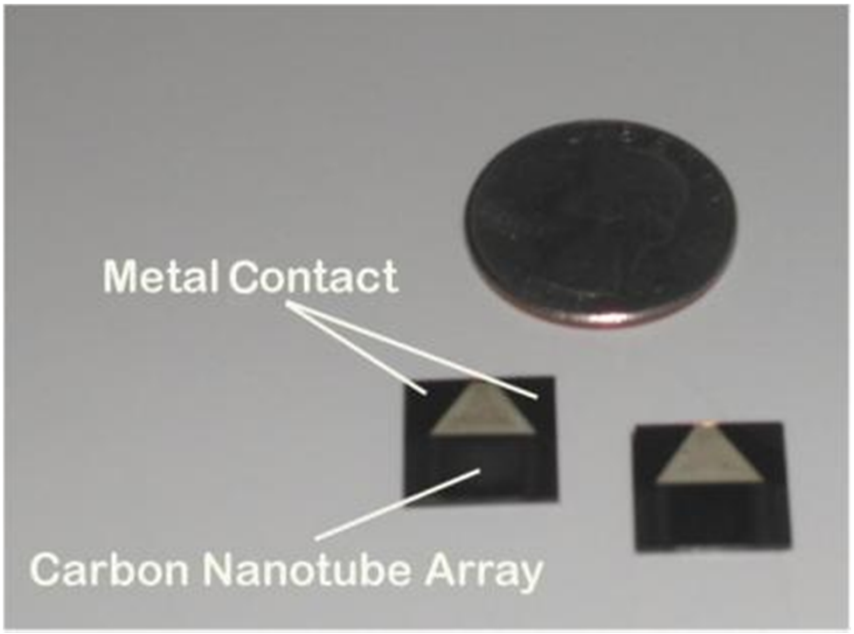
SRNL

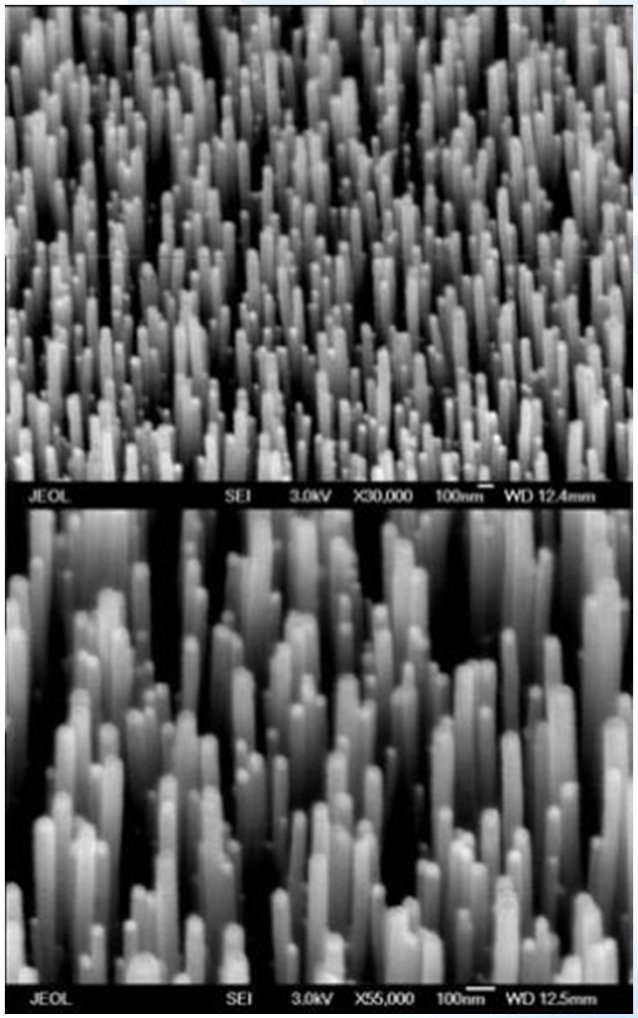
Carbon Nanotube (CNT) Arrays

parallel plate geometry



Modi, et. al. *Nature,* **2001,** 171.

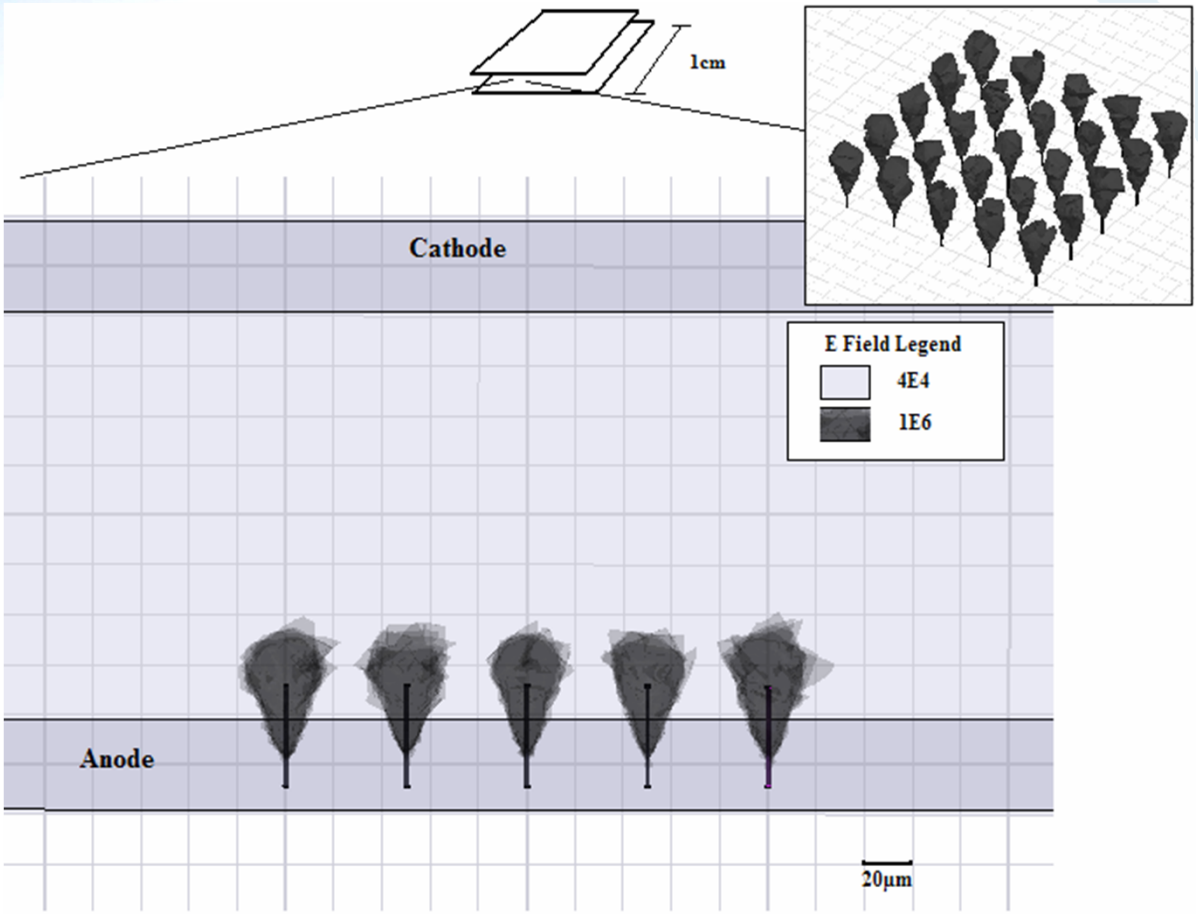




CNT array – acts as macro electrode

SRNL

Electric Field Simulations: CNT Arrays with Controlled Pitch



CNTs of 100nm diameter, 40µm length, with a pitch of 50µm,

plates 200µm apart,

biased at 10V

CNTs need to be spaced at ~2 times their height

Outline

- Background
  - Gas-Filled Radiation Detectors – Proportional Counters
  - Scale Effects
  - Carbon Nanotube Anodes
- **Fabrication of Nanostructured Arrays**
- **Electrical Properties of Arrays – HV Breakdown Studies**
- **Preliminary Results – Radiation Detection with Arrays**
- **Summary and Path Forward**

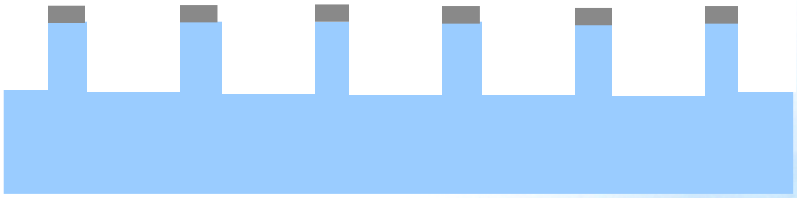
Fabrication of Nanoscale Arrays with Controlled Pitch

PE-CVD – growth of vertically aligned carbon nanofiber arrays

E-beam lithography – define array pattern

Substrate: B-doped 4” Si wafer
Array pattern: 100-200 nm diameter dots of 10-50 nm thick Ni (VACNFs) or Cr (Si posts) with pitch of either 1, 2, 3, 5, or 10 um

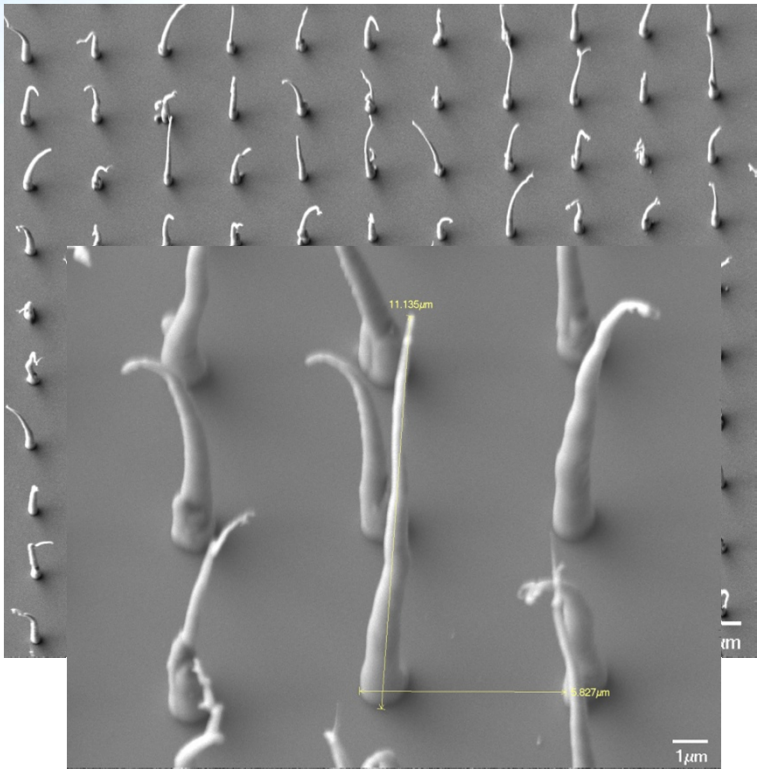
RIE – etching of nanostructured Si post arrays



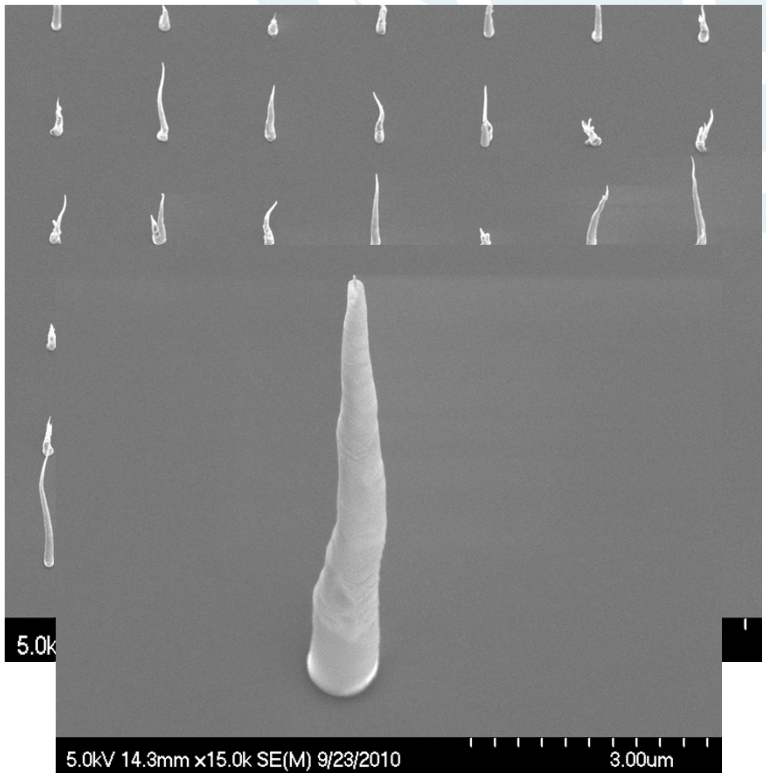
SRNL

Growth of VACNFs via E-beam Lithography and PECVD

Control of Array Pitch



5 micron pitch, ~30 micron height



10 micron pitch, ~12-15 micron height

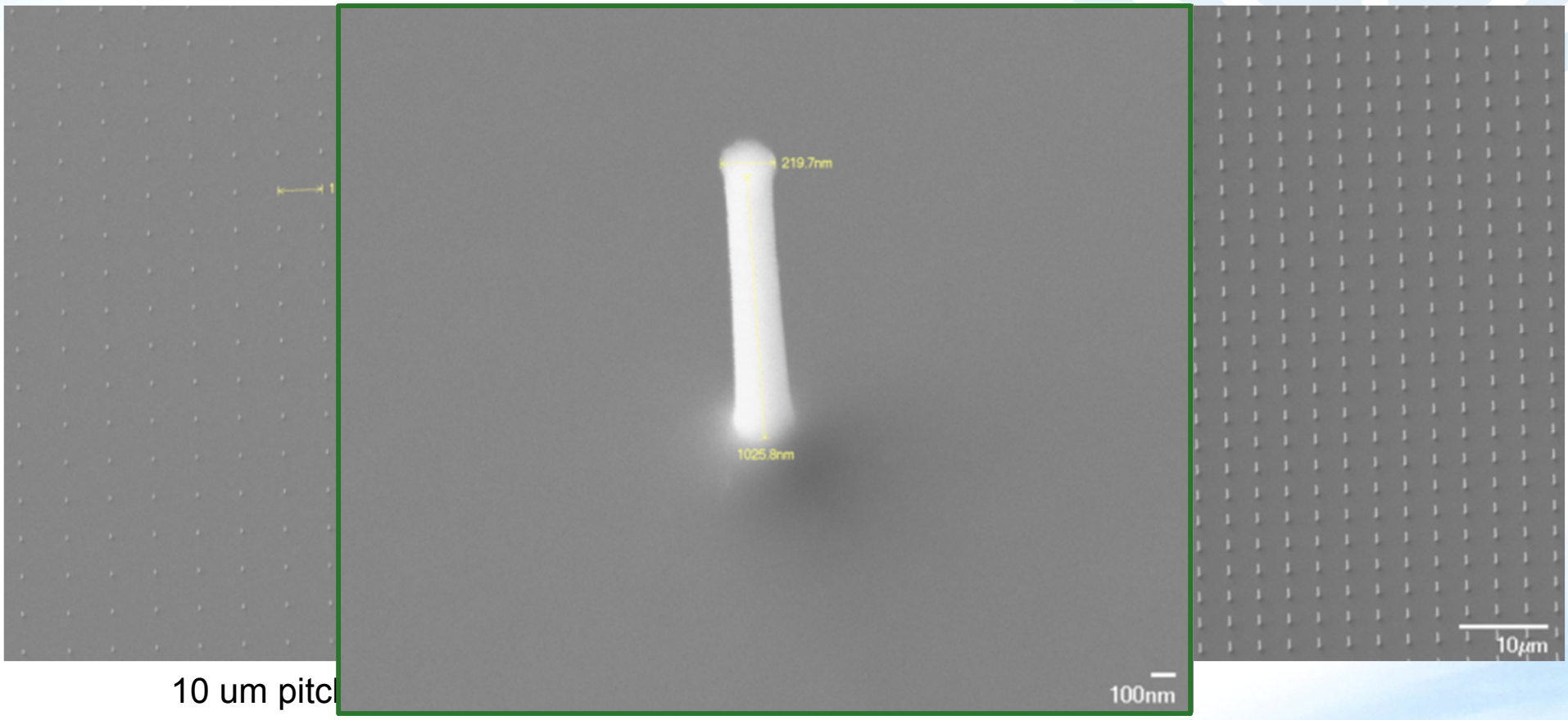
PECVD conditions: 665-710 °C, 100 sccm NH_3, 60 sccm C_2H_2, current 1.5-3A in 15 sec, 5 min growth time

PECVD conditions: 650-700 °C, 100 sccm NH_3, 58 sccm C_2H_2, current 3A, 4 min growth time

SRNL

Growth of Si Posts via E-beam Lithography and Reactive Ion Etching

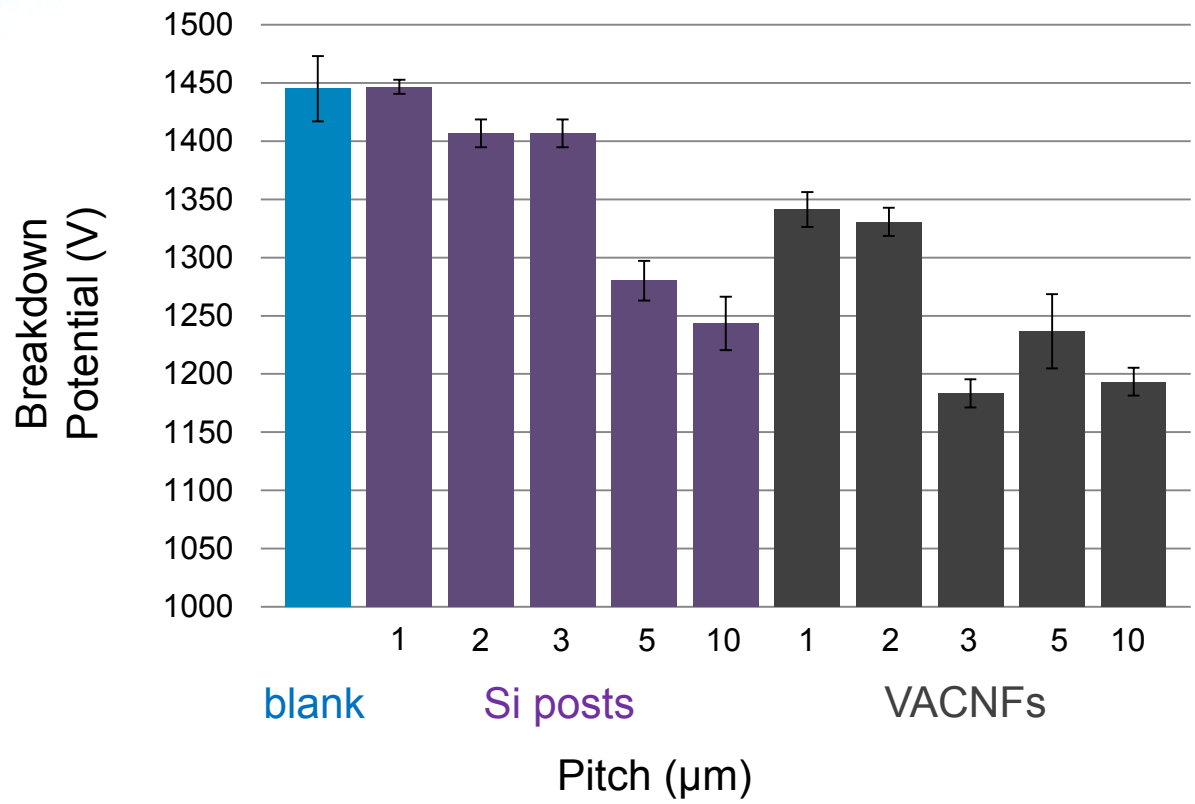
*diameter of spot ~200 nm for all pitches
*mask is 10 nm Cr



10 um pitc

RIE conditions: -110 °C, 80 sccm SF_6, 14 sccm O_2, 1 min etch time

Electrical Properties: High Voltage Breakdown Studies



High voltage electrical breakdown studies to:

1) obtain a qualitative measure of the electric field

2) test electrical stability of nanostructures

- HV breakdown in He
- 1 atm
- 0.5 cm spacing between parallel plates
- Error bars represent 3 measurements with the same array

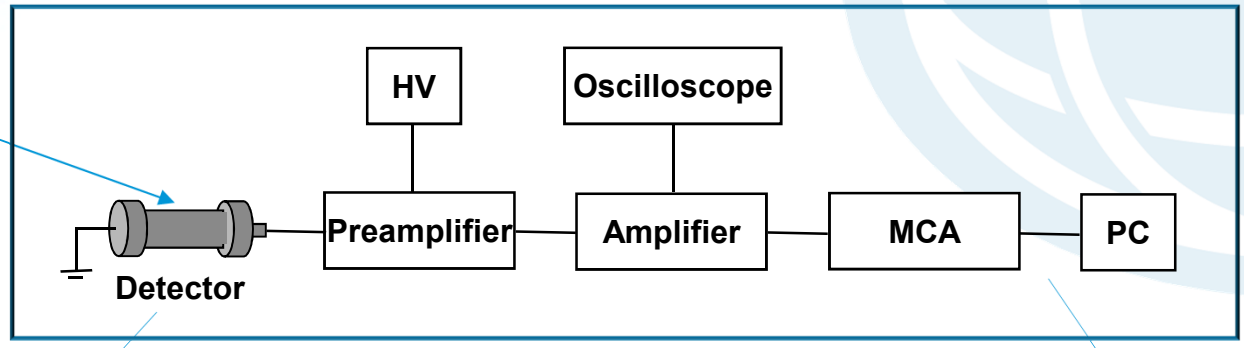
SRNL

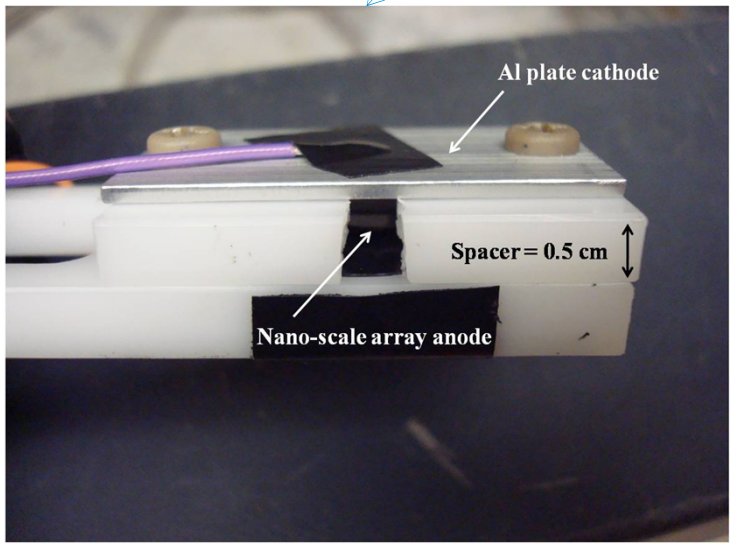
* Lower breakdown voltage corresponds to an increased electric field

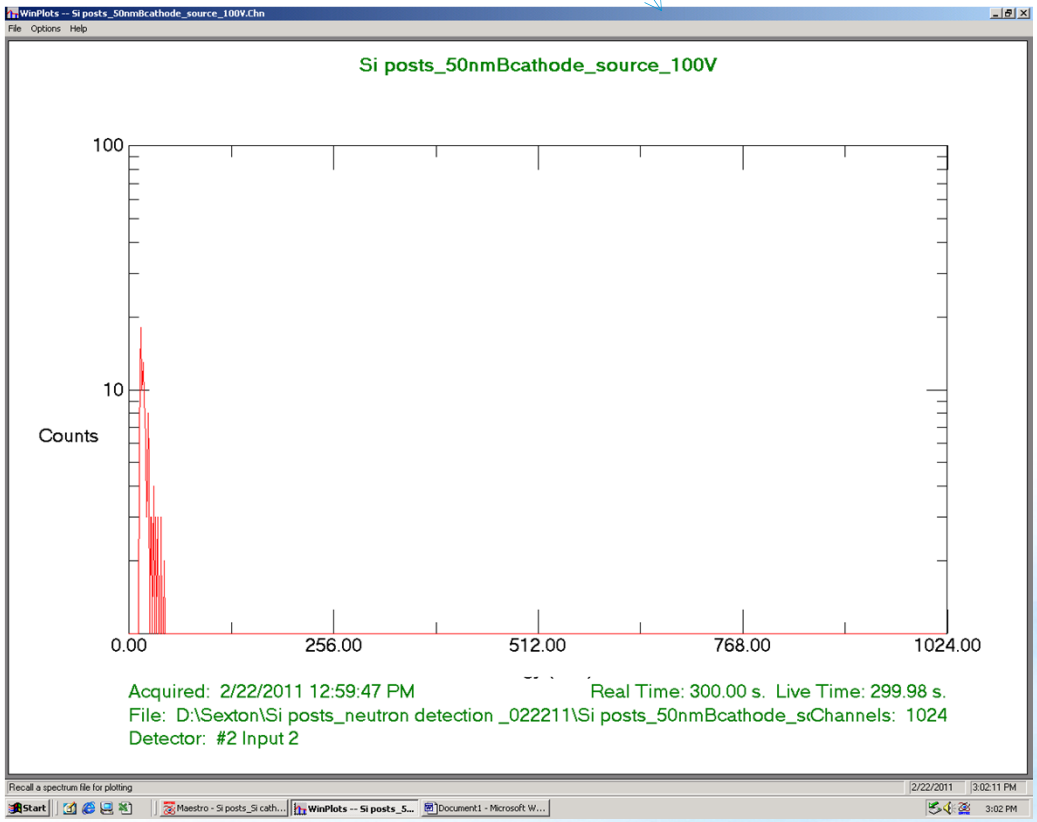
Experimental Setup for Proportional Counter

10 mCi Am-241 source placed inside detector tube

P10 fill gas at 1 atm

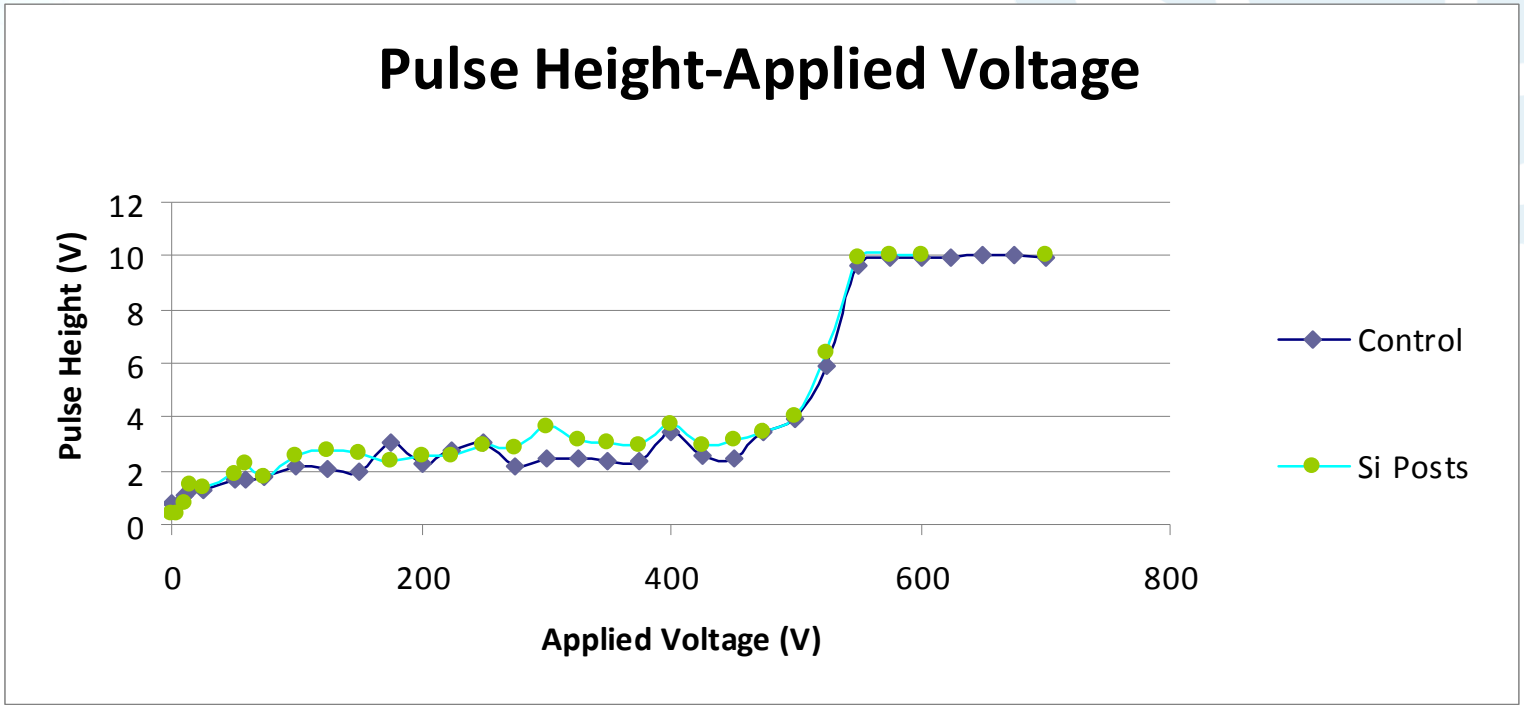






SRNL

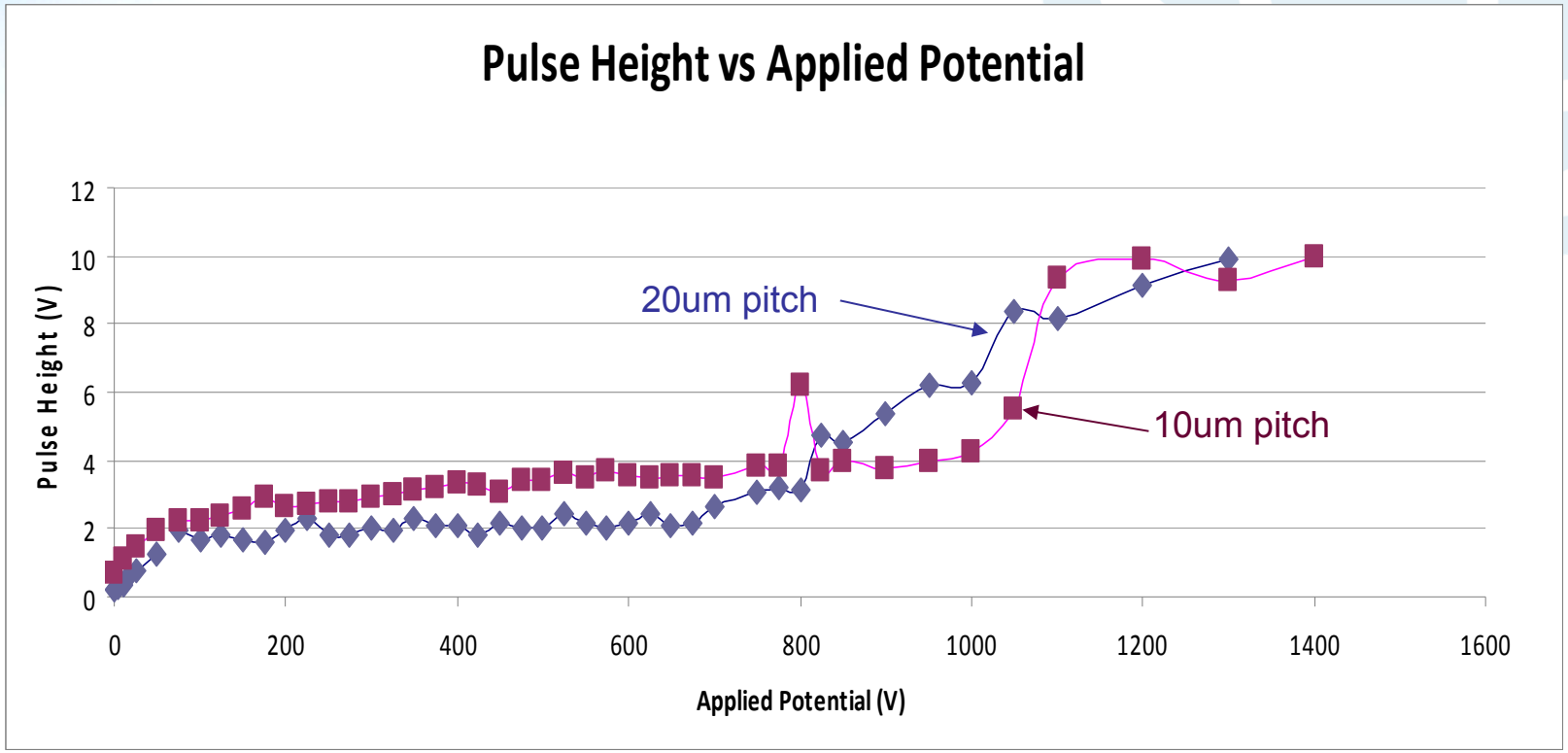
Preliminary Results: Proportional Response of Nanoscale Arrays



Si post arrays and VACNF arrays (data not shown) as prepared (100nm diameter x 10um pitch x 2um height with 1 mm spacer) showed no difference in proportional response compared to controls

SRNL

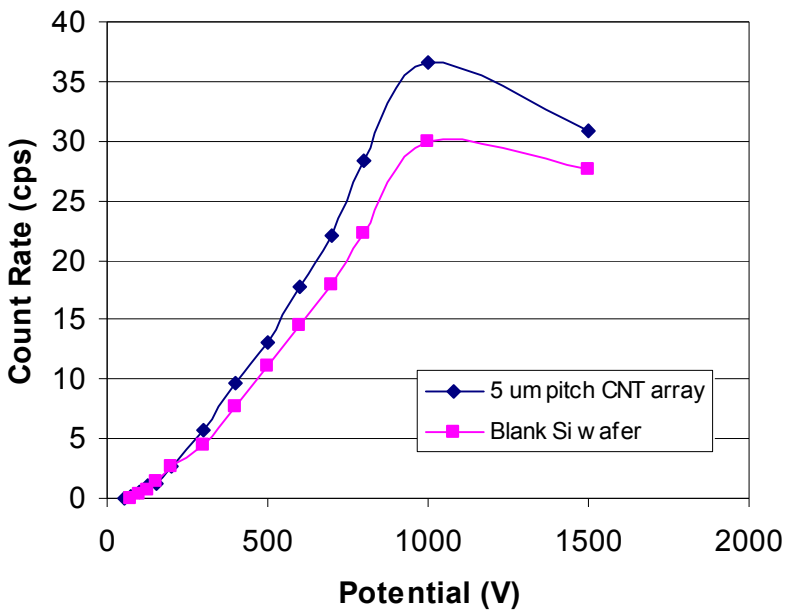
Preliminary Results: Proportional Response of Nanoscale Arrays

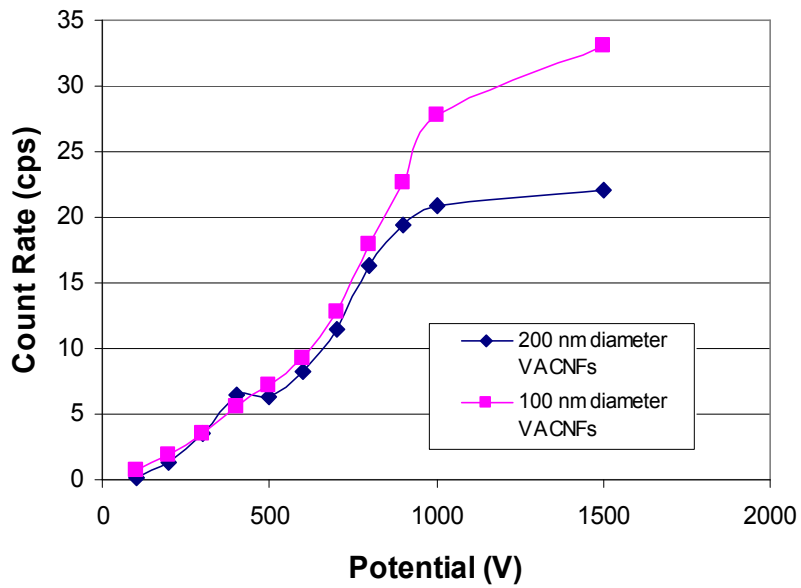


Si post arrays (100nm diameter, 2um height, 2mm spacer) with 20um pitch and 10um pitch showed little difference in proportional response

SRNL

Preliminary Results: Count Rate of Nanoscale Arrays



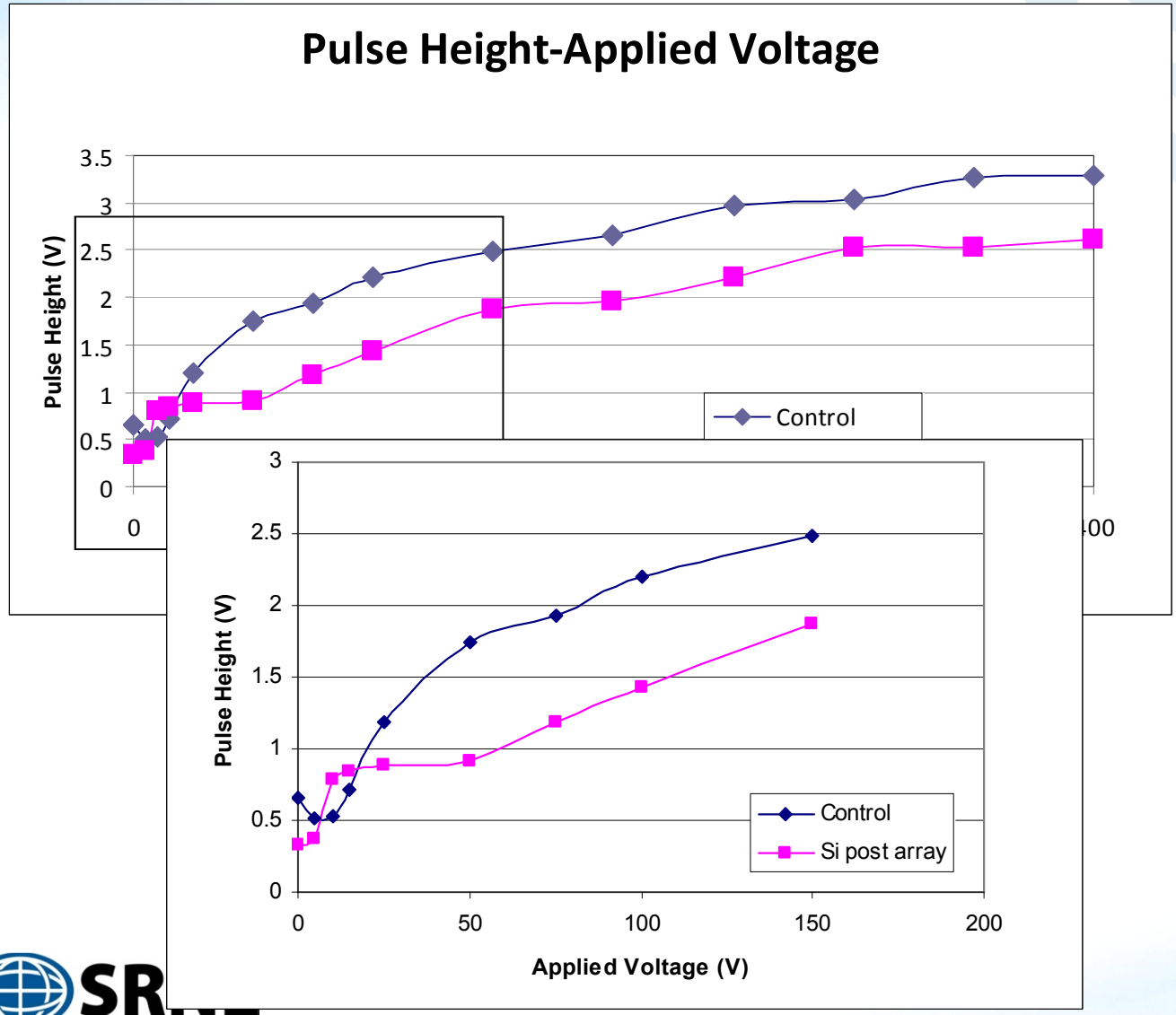


(10 um pitch for VACNF arrays above)

Effect on count rate is observed when nanoscale arrays are present and when diameter of nanostructures is decreased

SRNL

Preliminary Results: Metal Coated Si Post Arrays



Source: 10 mCi Am-241

Cathode: Al plate

Anode: B-doped Si wafer with Si post arrays (100 nm diameter x 10 um pitch) **coated with an additional 50 nm B**

Control anode: B-doped Si wafer coated with an additional 50 nm B

Summary & Future Work

Proof-of-concept for nano-scale shown for single carbon nanotube in gamma sealed mode

Testing of electrical properties of nanoscale arrays – high voltage breakdown studies

Proof-of-concept testing with gamma sources – *ongoing*

- array pitch and nanostructure diameter found to effect count rate
- conductive metal coatings allow for an observable nanoscale response

Future work:

Proof-of-concept testing of arrays with neutron sealed sources

Scale-up of array fabrication

Potential advantages include:

- Increased counting sensitivity by achieving higher electrical field strengths
- Lower operating voltages due to optimized electrode geometry and lower cost electronics costs.
- Reduced platform size and through micro- and nano-fabrication techniques as well as electronic requirements.
- Improved ruggedness from the radiation-resistant and material-strength properties of carbon nanotubes.

SRNL

Acknowledgements

SRNL:

Matt Howard
Matt Seigfried
Kevin Kane

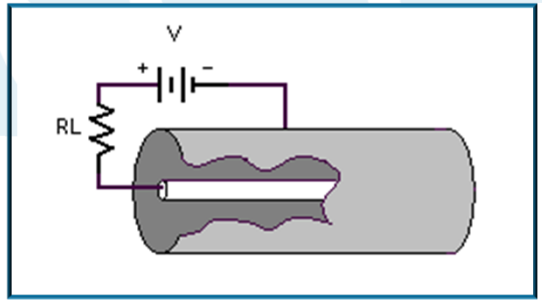
Center for Nanophase Materials Science at ORNL:

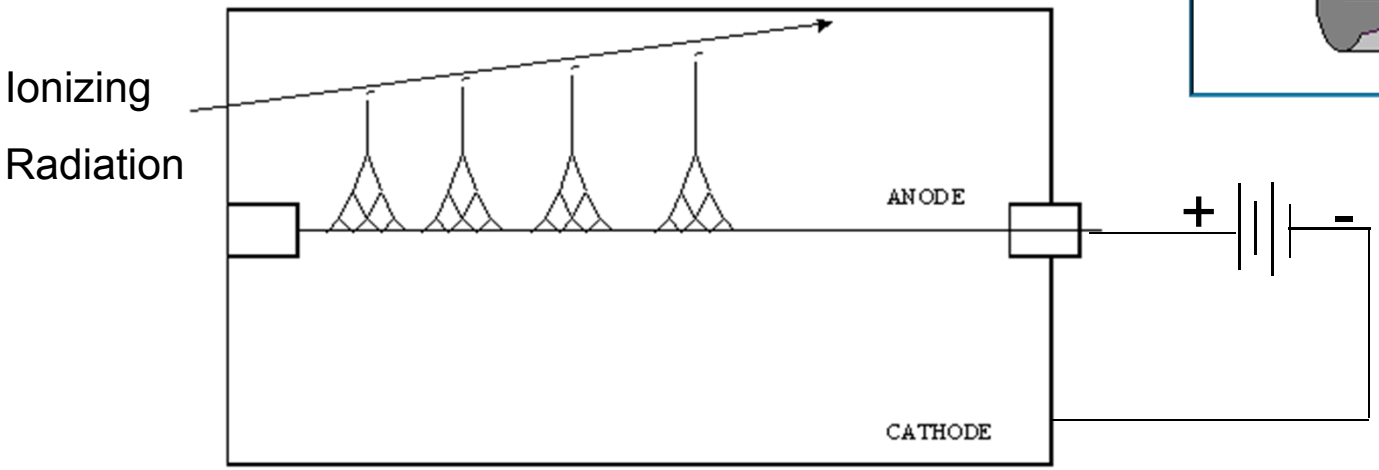
Scott Retterer
Dale Hensley

Funding:

Next Generation Safeguards Initiative Fellowship
SRNL LDRD FY2010
SRNL PDRD

Background: Proportional Counter (PC)





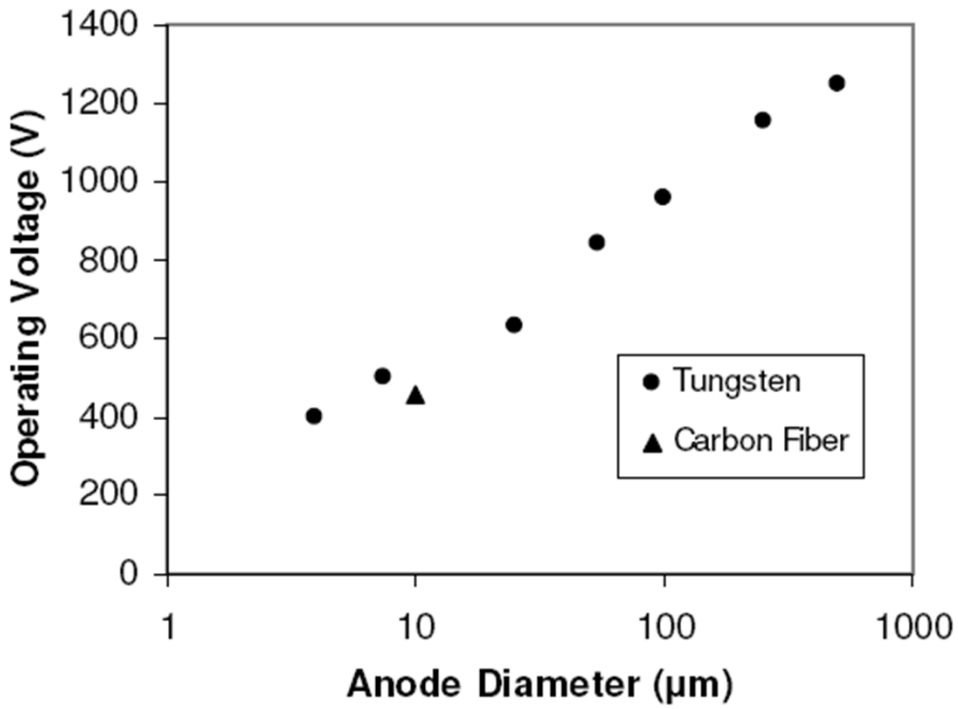
Gas multiplication to amplify the charge from the ion pair created by ionizing radiation in the gas phase created by large electric field

Detection : Pulse generated proportional to the energy of the radiation allowing for radiation spectroscopy

Electrical potential in 1-2 KV range

SRNL

Scale Effects: Tungsten Wires and Carbon Fiber



- Operating voltage decreases with anode diameter
- Material effects observed with carbon fiber

Experimental conditions:
- coaxial geometry
- Cs-137 source (activity of ~18 µC)

SRNL

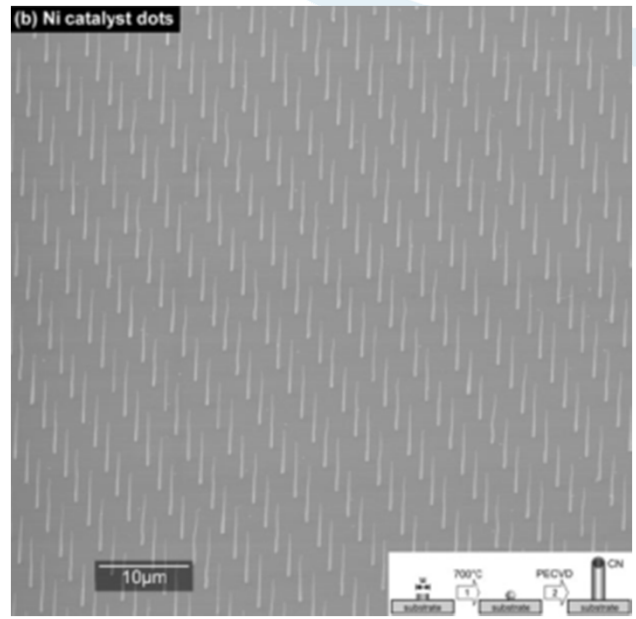
Fabrication of CNT Arrays with Controlled Pitch – E-beam Lithography + PECVD

Substrate Preparation:
Spin coat e-beam resist: 100-300 nm

Electron Beam Lithography:
Define array pattern

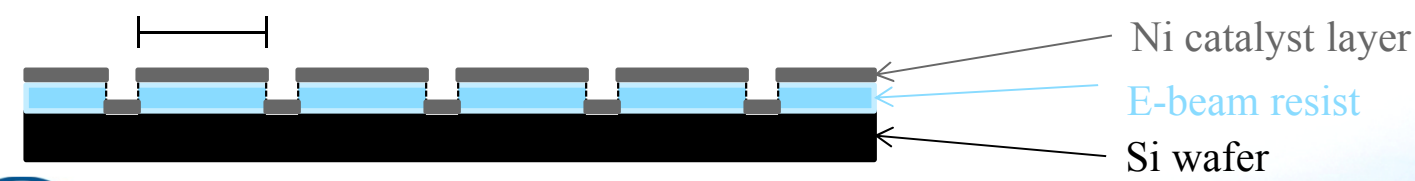
Patterning of Ni Catalyst:
Develop resist
Sputter Ni catalyst layer: 5-10 nm
Lift-off of resist

Grow Carbon Nanotubes:
Plasma Enhanced CVD



Teo, KBK, *et. al. Nanotechnology*, 14, **2003,** 204-211

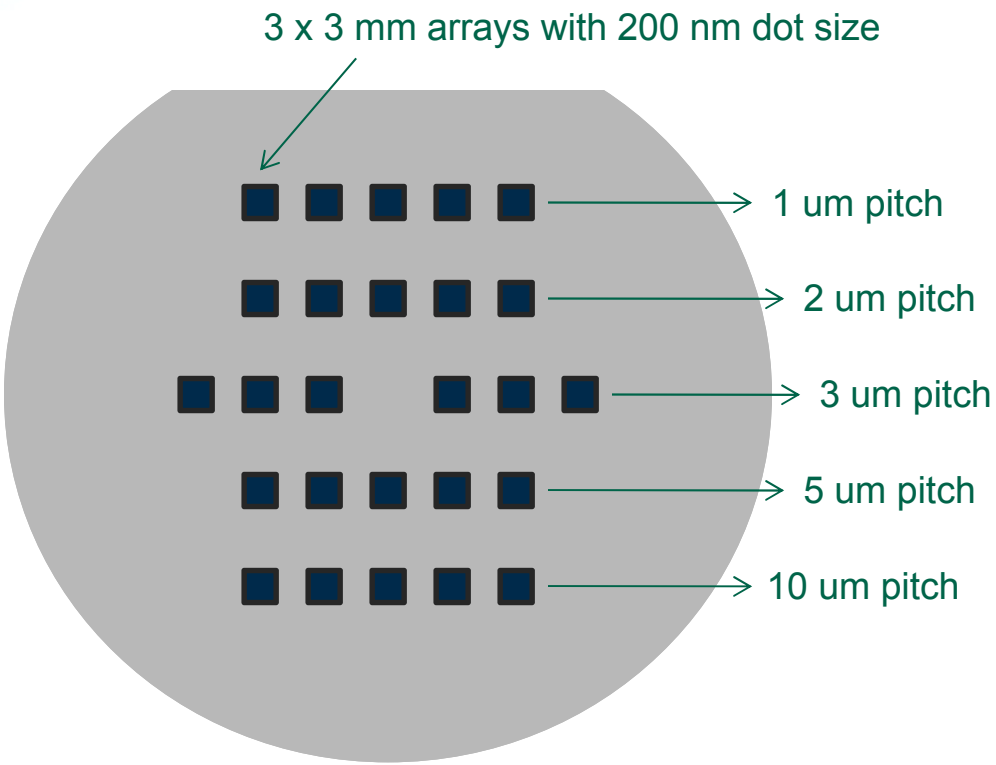
Controlled pitch defined by e-beam



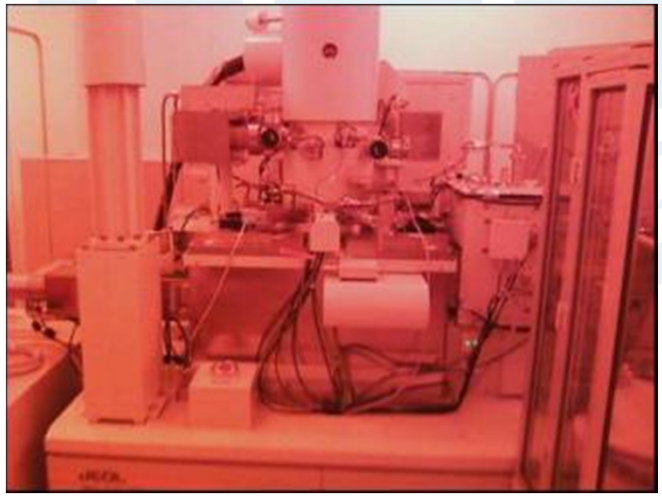
SRNL

Si Wafer Array Patterning

Schematic of Si Wafer and Arrays



Conductive 4 inch Si wafer



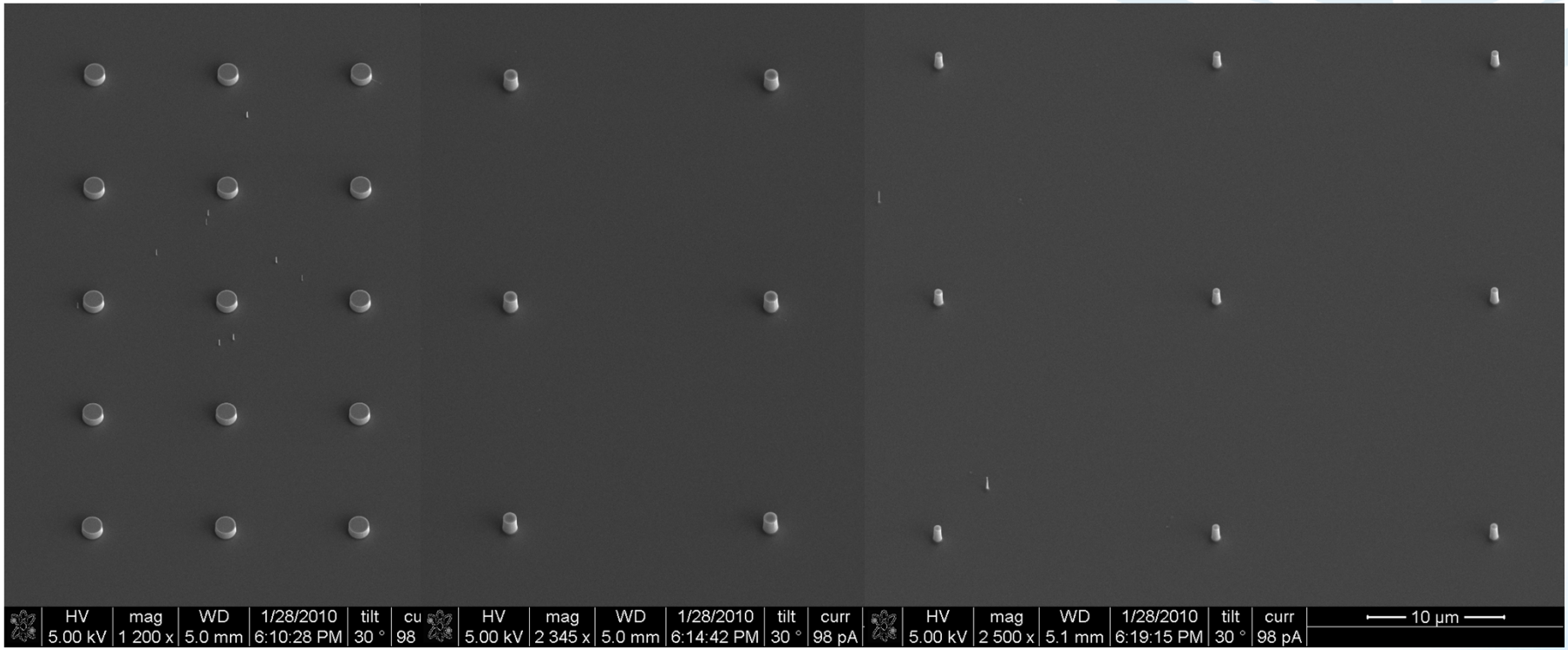
E-beam lithography system

PE-CVD

SRNL

Growth of Si Posts via E-beam Lithography and Reactive Ion Etching

Control of Si Post Diameter

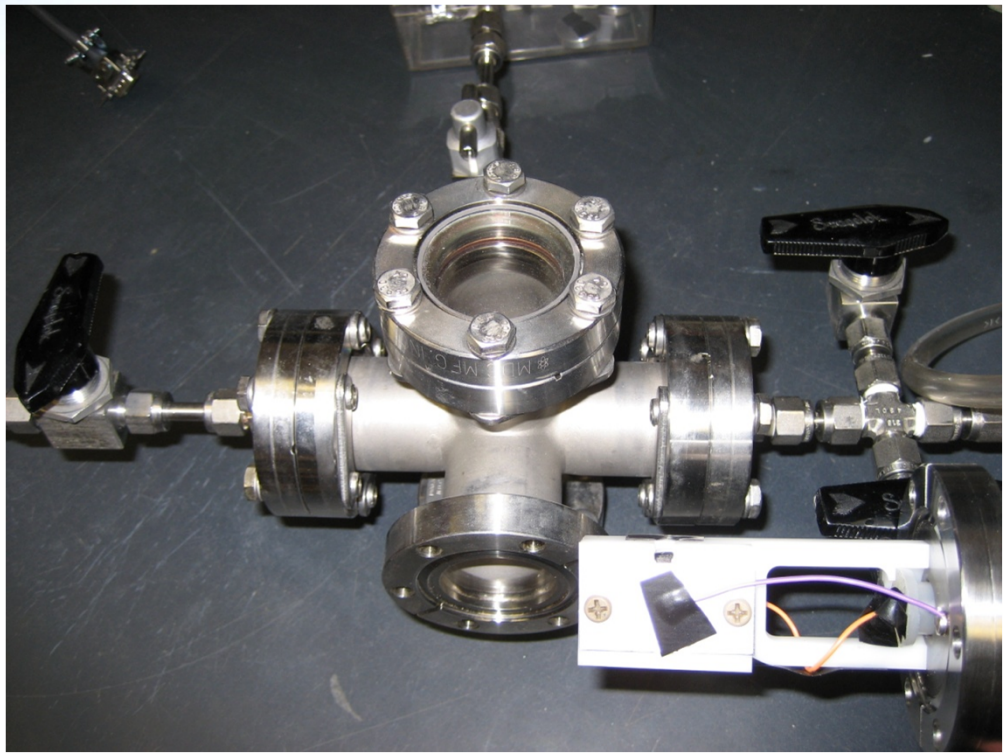


3 um diameter 1 um diameter 0.5 um diameter

*pitch kept constant at 20 um
*mask is 10 nm Cr

SRNL

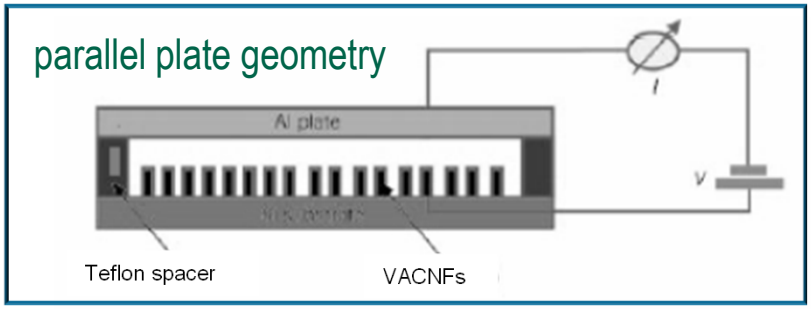
Testing Electrical Properties of Nanostructured Arrays



High voltage electrical breakdown studies to:

1) obtain a qualitative measure of the electric field

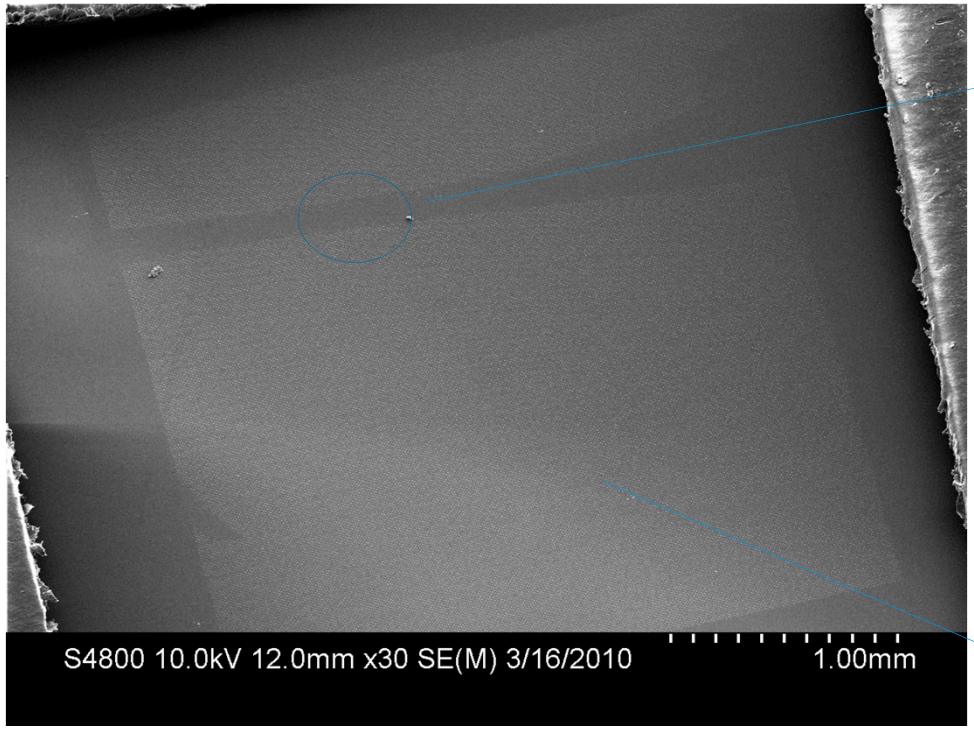
2) test electrical stability of nanostructures

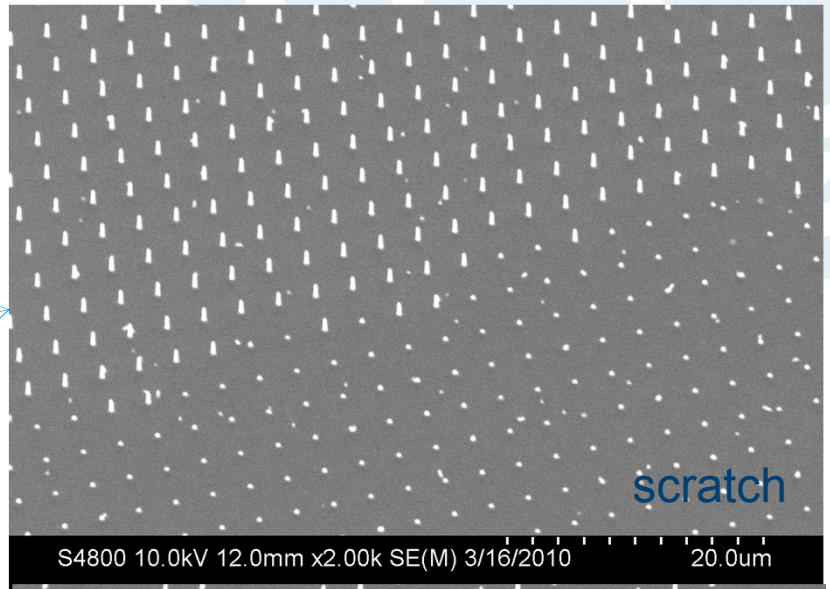


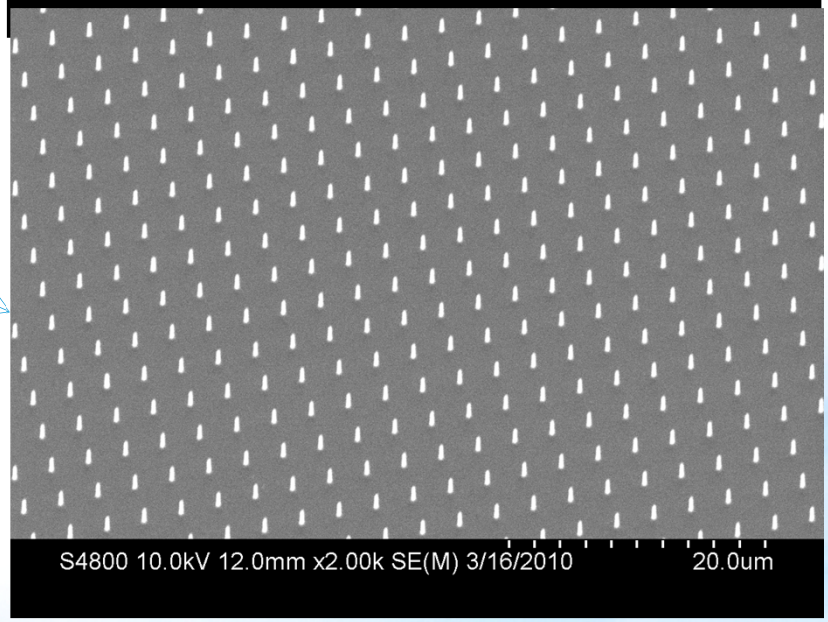
Modi, et. al. *Nature,* **2001,** 171.

Effects of HV Breakdown

Si posts - 3 µm pitch array
Breakdown in **He** with 0.5 cm gap
@ ~1.4 KV



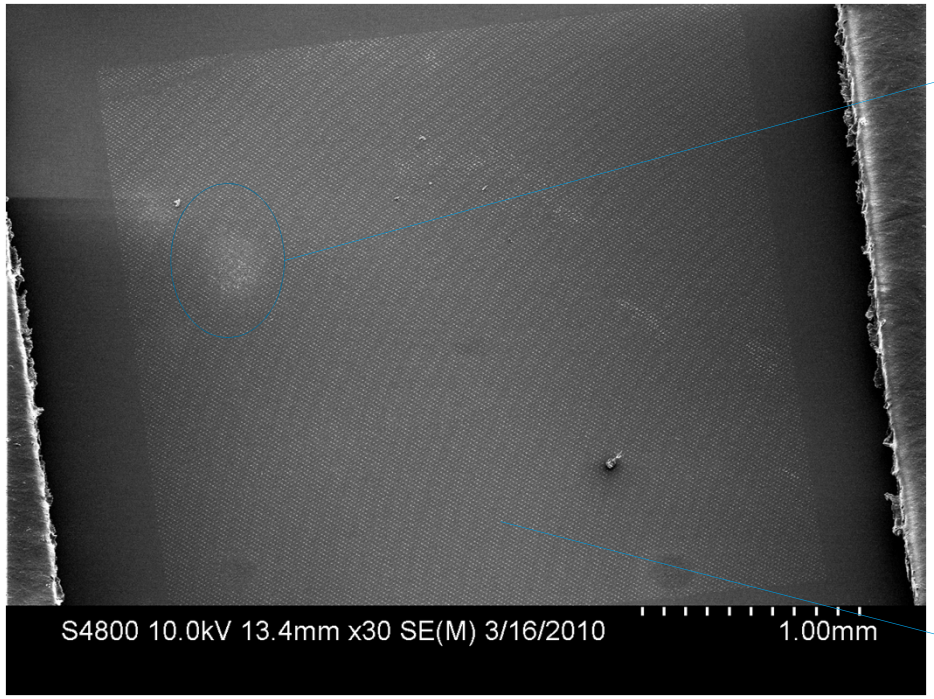


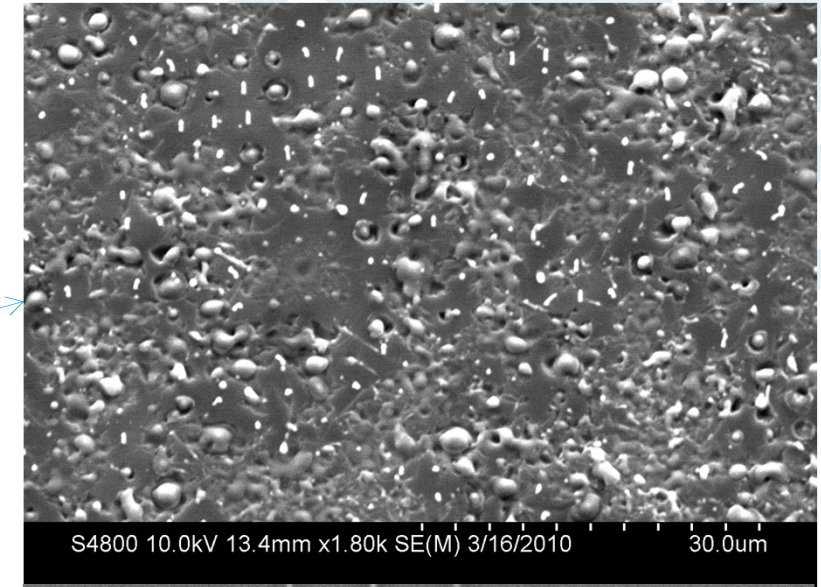


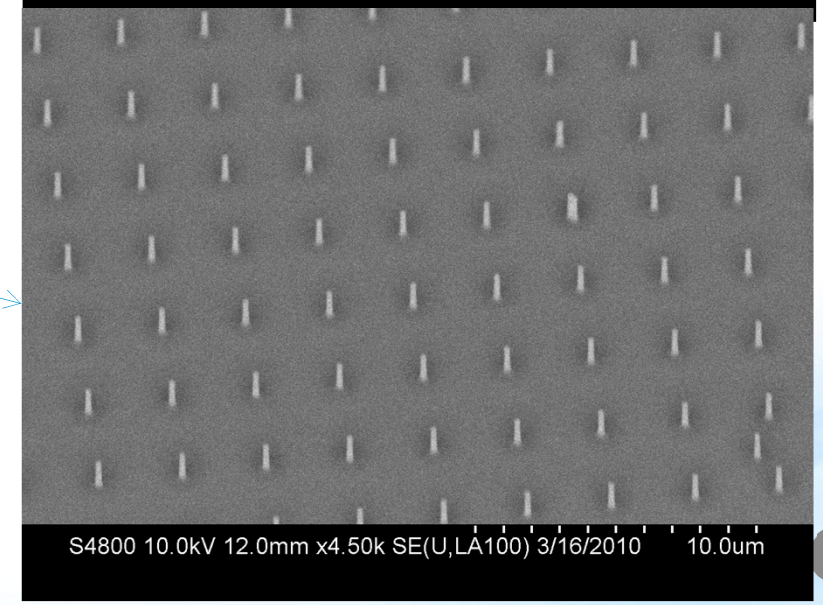
SRNL

Effects of HV Breakdown

Si posts - 3 µm pitch array
Breakdown in **Ar** with 0.5 cm gap
@ ~3.5 KV

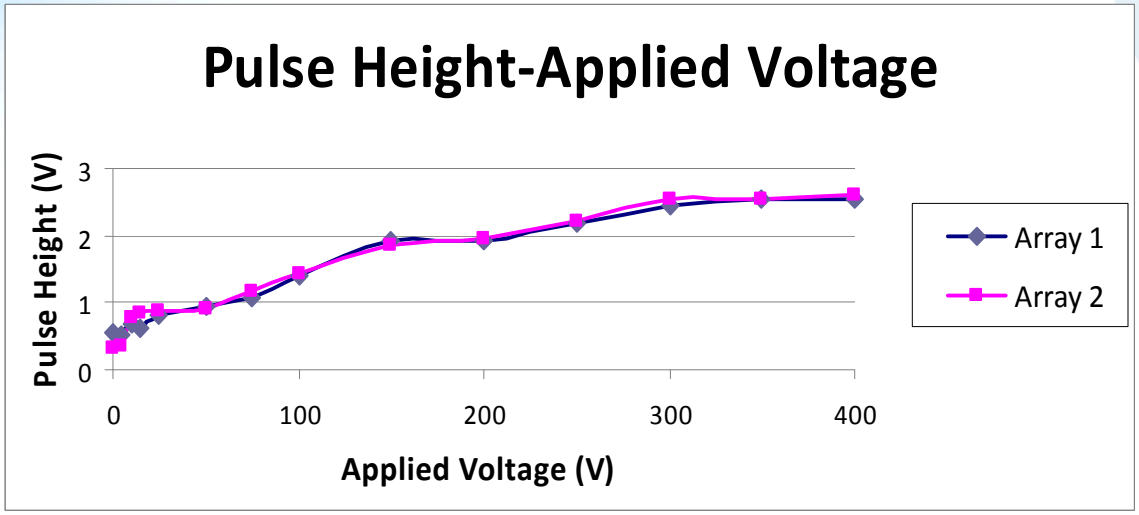


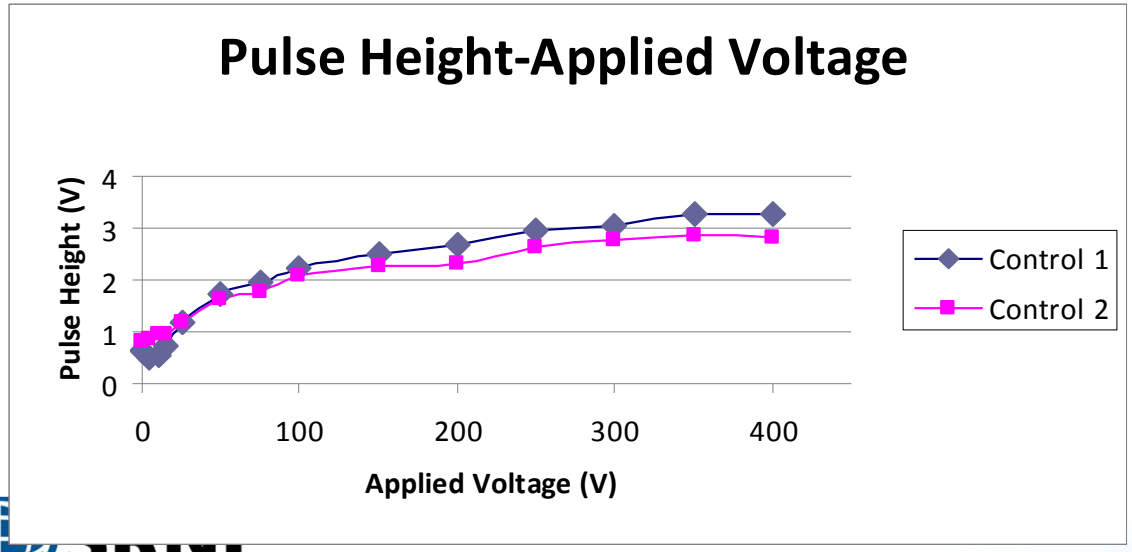




SRNL

Preliminary Results: Metal Coated Si Post Arrays





Source: 10 mCi Am-241

Cathode: Al plate

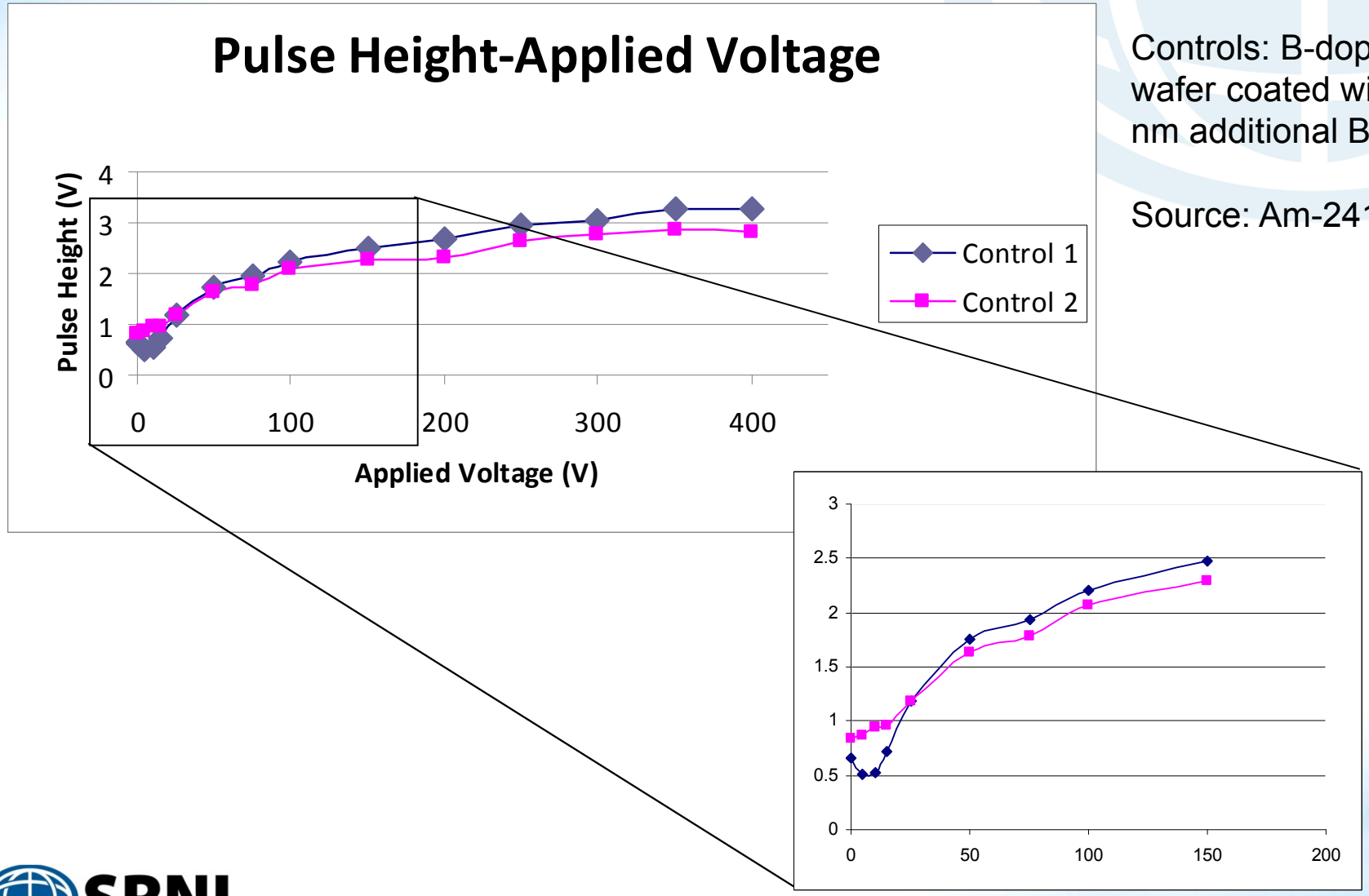
Anode: B-doped Si wafer with Si post arrays (100 nm diameter x 10 um pitch) **coated with an additional 50 nm B**

Array 1: posts etched for ~12 min.

Array 2: posts etched for ~20 min.

Control anode: B-doped Si wafer coated with an additional 50 nm B

Preliminary Results: Control Experiments

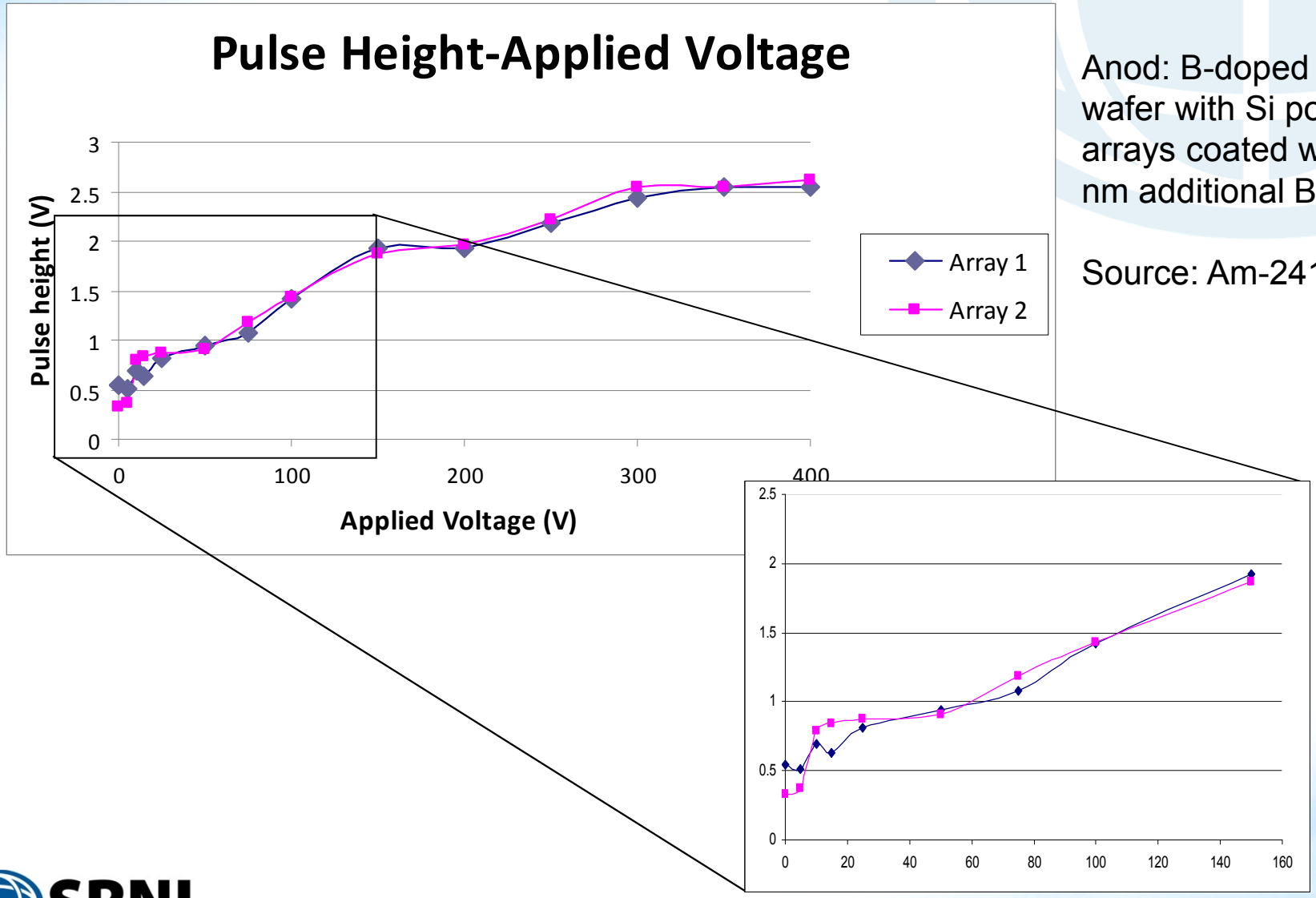


Controls: B-doped Si wafer coated with 50 nm additional B

Source: Am-241

SRNL

Preliminary Results: Nanoscale Arrays

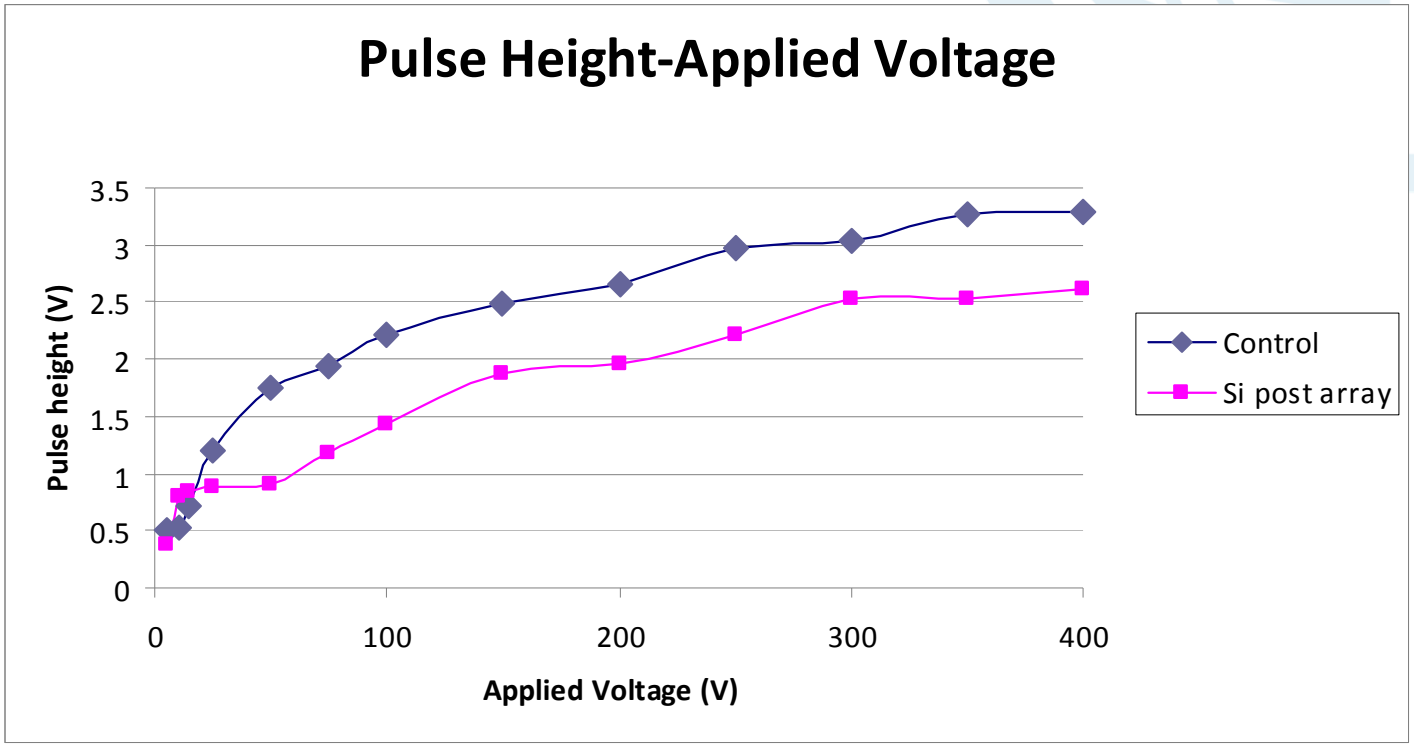


Anod: B-doped Si wafer with Si post arrays coated with 50 nm additional B

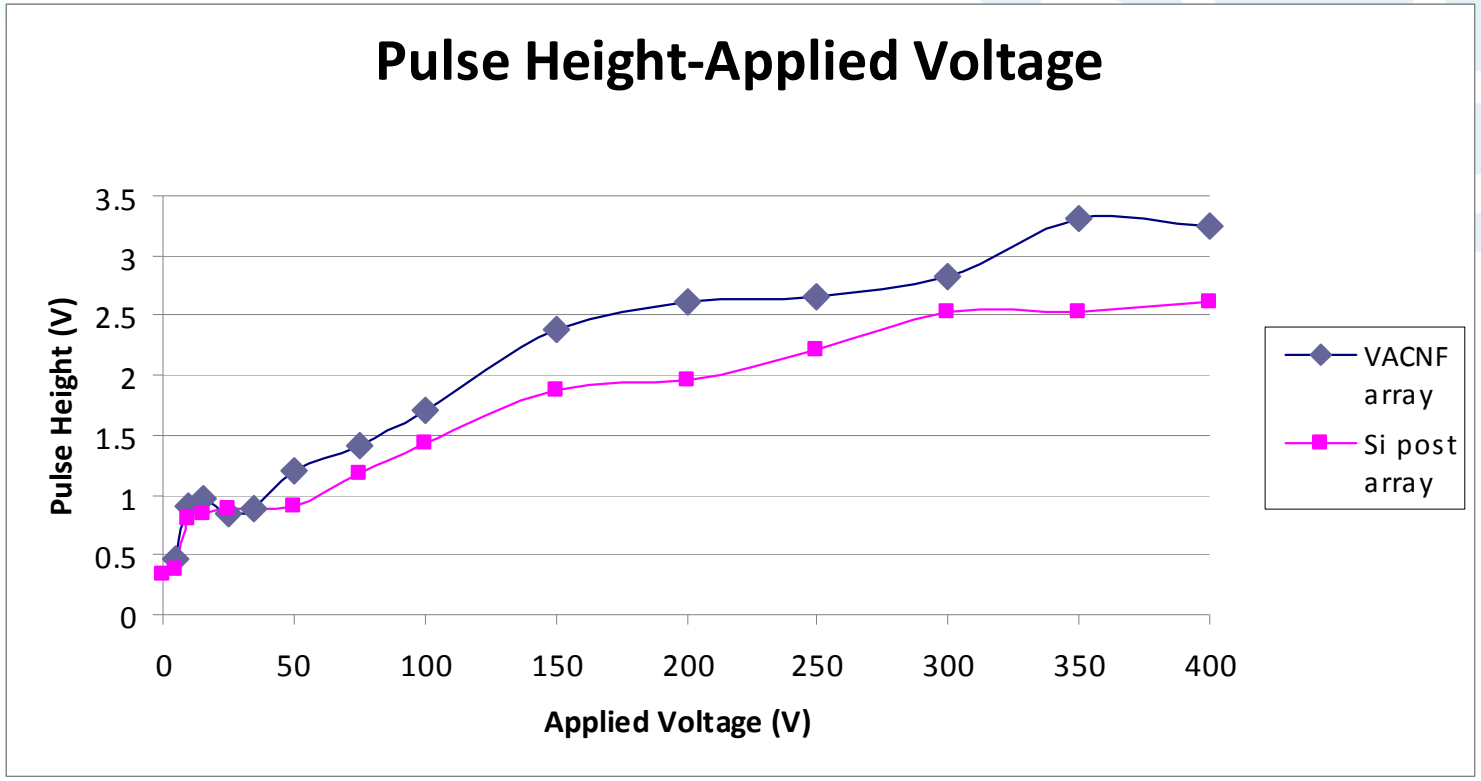
Source: Am-241

SRNL

Controls vs. Arrays



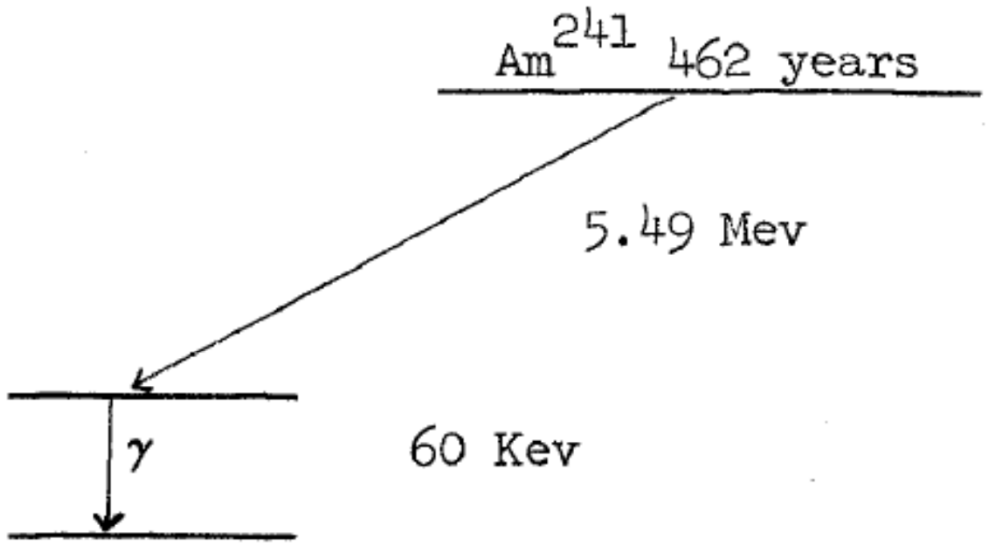
VACNF Array vs. Si Post Array



100 nm diameter, 10 um pitch, coated with an additional 50 nm B for both VACNF and Si post arrays

SRNL

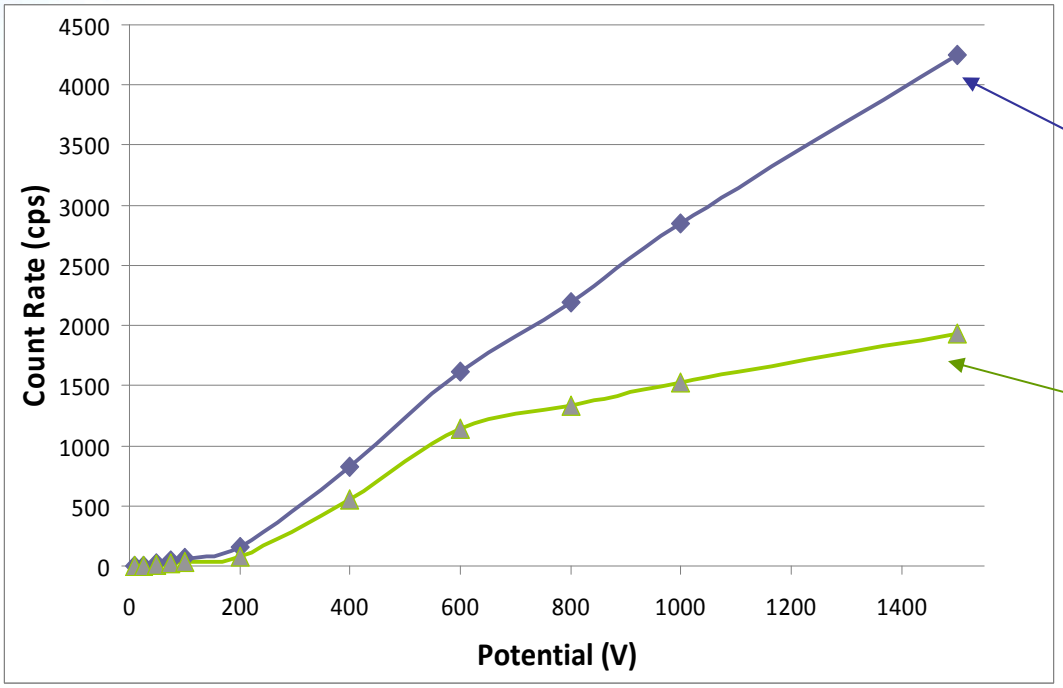
Am-241 Decay Scheme



Simplified Decay Scheme

Strain, et. al. *Preparation, properties, and uses of Am-241, alpha-, gamma-, and neutron sources,* ORNL-3335, 1962.

Preliminary Results: Count Rate Data



3 mm plate spacing

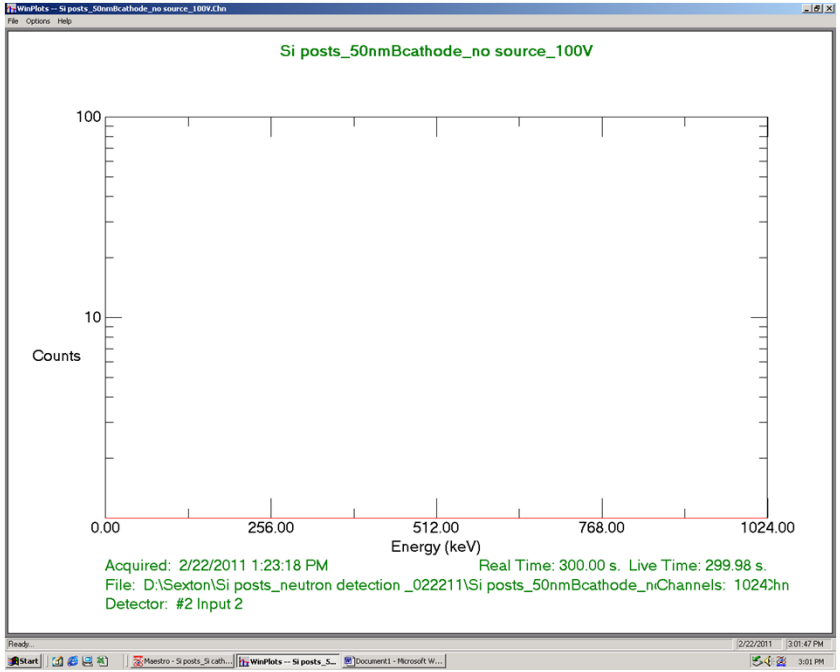
Nanoarray anode: 3x3 mm VACNF array with 100 nm diameter x 5 um pitch nanostructures

Control anode: B-doped Si wafer

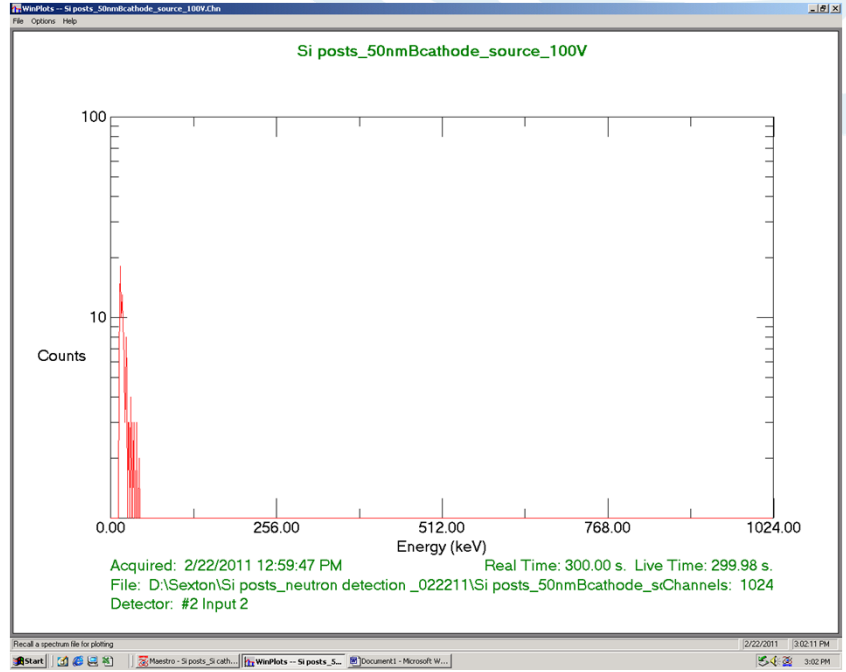
Cathode: Al plate

Preliminary Results: Neutron Detection

No source, 100 V applied potential



0.1 uCi Cf-252 source, 100 V applied potential



Anode: Si post array, 100 nm diameter posts, 250 um pitch

Cathode: Si wafer coated with 50 nm of B

Plate spacing: 2 mm