Exploration of the challenges with radon-generated Po-210 surface contamination

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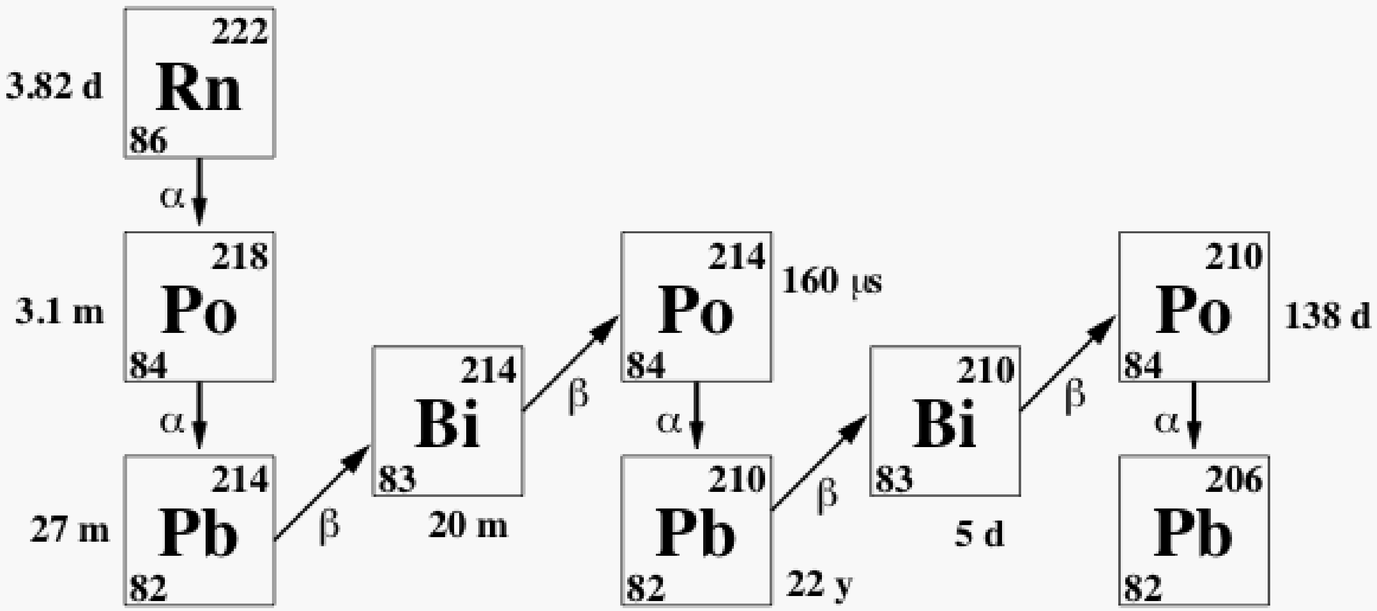
Motivation



Exposure to radon gas leaves behind progeny on surfaces with the short-lived portion of the radon decay chain deposited on a surface

Radon gas can diffuse into the material during exposure - ^{210}Po is an alpha emitter

Being of the most concern

Depending on the material, progeny are deposited deeper into the subsurface

Challenge:

Findings from studies evaluating cleaning and surface removal techniques give varying results

Pb and Bi generally remove easily using a variety of standard methods

Po shows to be more difficult to remove – more involved techniques needed

Next generation experiments will have more parts with greater surface contamination control requirements

Example Po Removal Techniques

E.W. Hoppe et. al [NIM A579 (2007) 486]

Cu **etching** using HNO_3

Effective at removing all surface contaminants, but not always the most practical

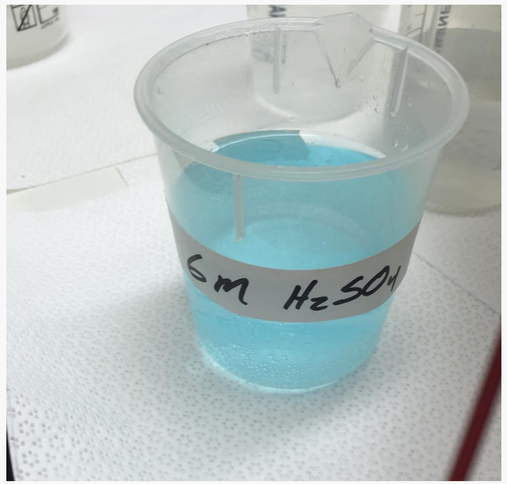
Limited Po removal

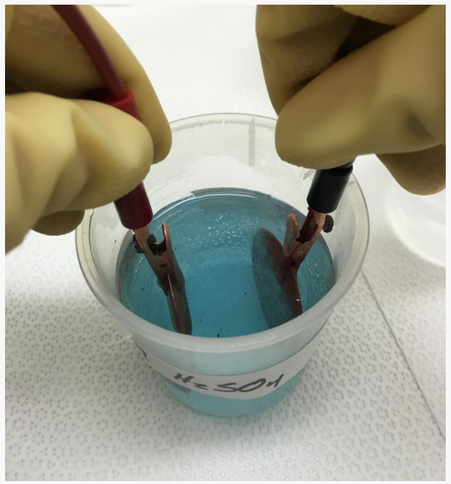
Nitric most effective

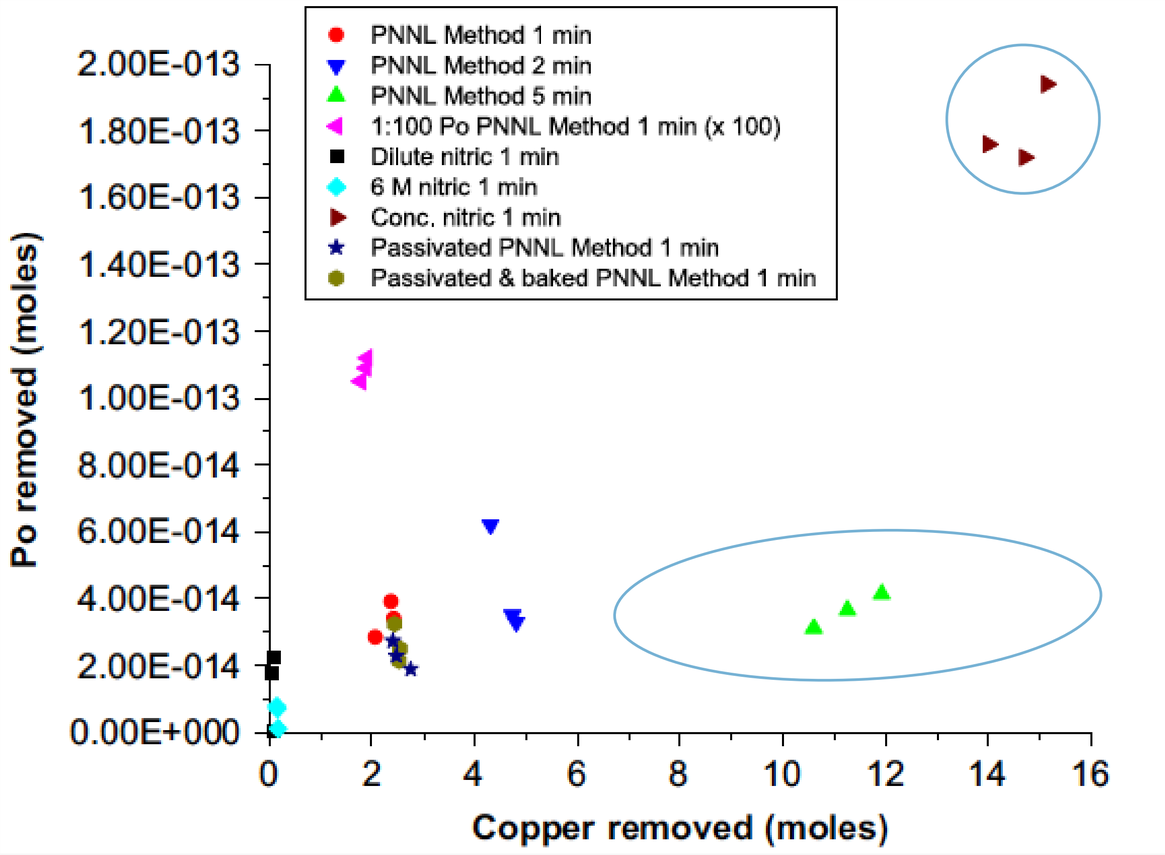
PNNL method: Dilute: H_2SO_4 / H_2O_2

Showed promise but limited ^{210}Po removal

Method works well for removing surface U/Th







Po Chemical Behavior

Previous findings show mixed results for Po removal

Po^0 can exist in solution over the entire pH range

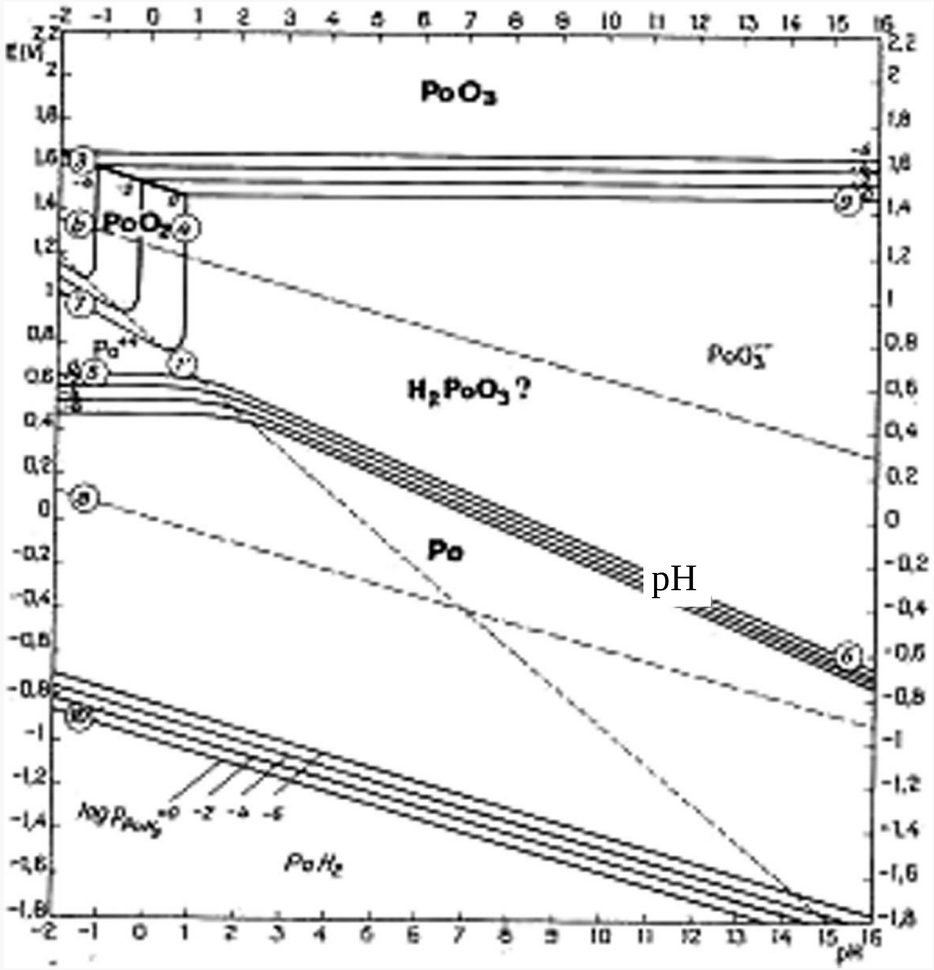
All techniques use an acidic solution

Force Po to be soluble through oxidation; Po^{4+} is the most favorable ion state unlike Th/U

Po^{4+} favors staying in solution; forced through applying an oxidation potential or an oxidizing agent

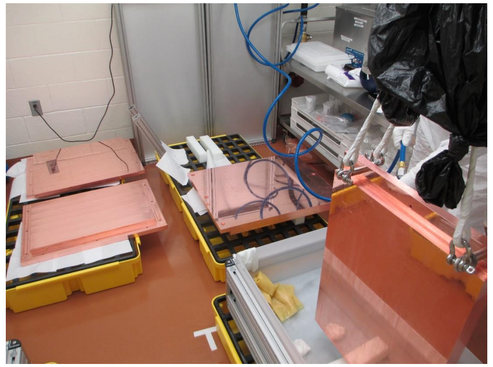
Sufficient exposure to the oxidizing agent may be the determining factor for effective Po removal

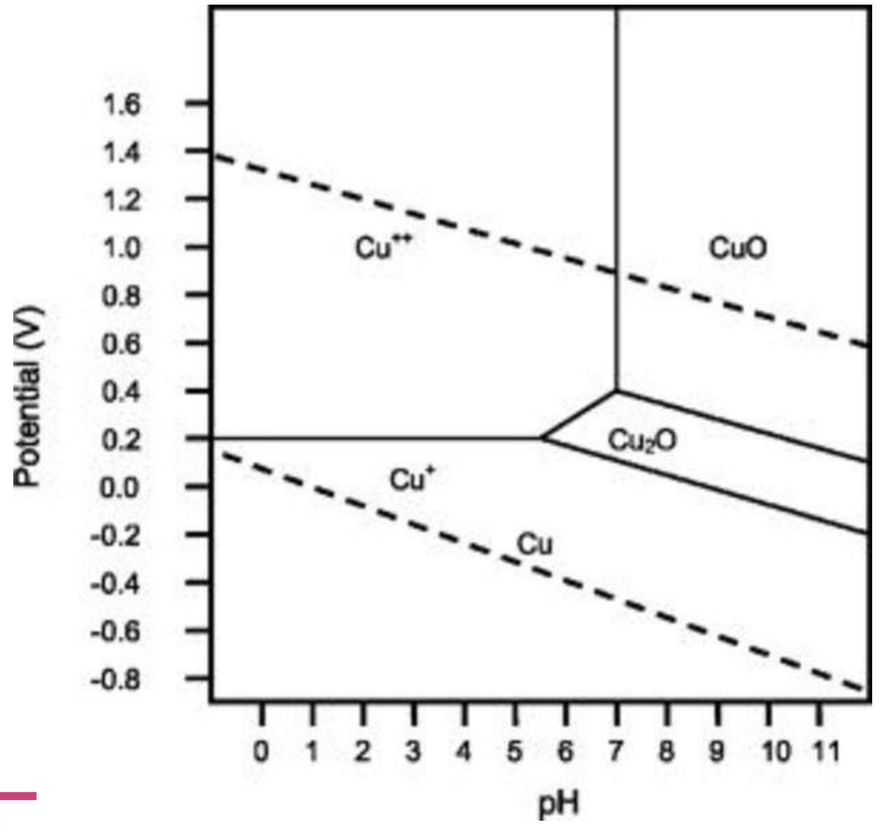
- Established Po removal from Cu
  - increased H_2O_2 concentration with agitation [V. Guiseppe, LRT 2017]
  - dynamic (multi-stage) etching [K. Pelczar, LRT 2017]



Po Pourbaix Diagram

Oxidation Method of Cu





Cu Pourbaix Diagram

Half reactions using hydrogen peroxide to **etch**

~~$H_2O_2 + 2e \longrightarrow 2OH^-$ (in alkaline conditions)~~

$H_2O_2 + 2H^+ + 2e^- \longrightarrow 2H_2O$ (in acidic conditions)

$H_2O_2 \longrightarrow 2H^+ + O_2 + 2e^-$ (in acidic conditions)

PTFE (polytetrafluoroethylene) Properties

Key properties

- Excellent chemical resistance (C-F bonding)
- Anti-stick properties
- Excellent thermal working properties
- Greatest resistance to fatigue
- Low co-efficient of friction
- UV-resistant (does not age)
- Not hygroscopic (water absorption < 0,01%)
- Very high dielectric insulation properties but that contributes to low static dissipation factor (*unless treated)*

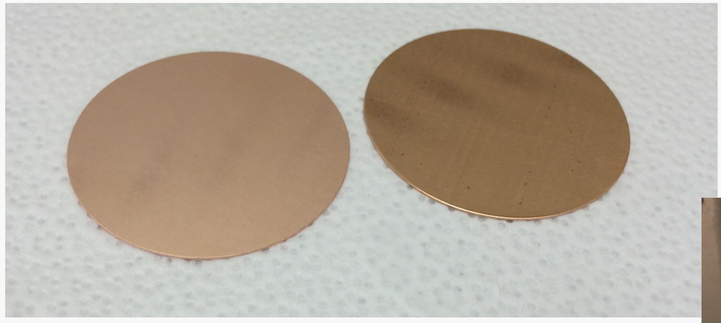
Easily **leached** for cleaning with high concentration nitric solution

$$2H^{+}(aq) + 2HNO_3(aq) + M(s) + 2NO_3^{-}(aq) \rightarrow 2NO_2(g) + 2H_2O(l) + M(NO_3)_2(aq)$$

Po Removal Study

Focus on removal of ^{210}Po from Cu and PTFE

Two common low background materials



Samples: untreated chemically

50-mm diameter, 0.5 mm-thick Cu foil disks

50-mm diameter, 5 mm-thick PTFE disks

Exposed to a 100 kBq radon source for ~ 1 month

Achieved an alpha count rate of ~300 counts/day

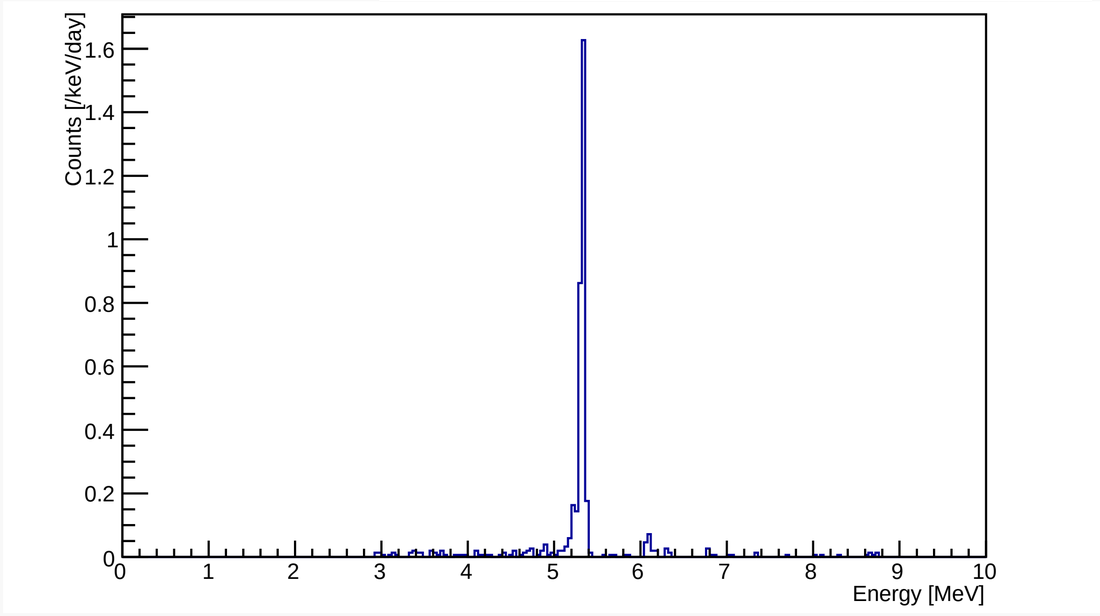
Alpha Detector

Alpha spectrometer with an ion-implanted silicon detector.

Samples counted before and after treatment

Background of ~6 counts/day

Example of the measured ^{210}Po α-peak on PTFE sample



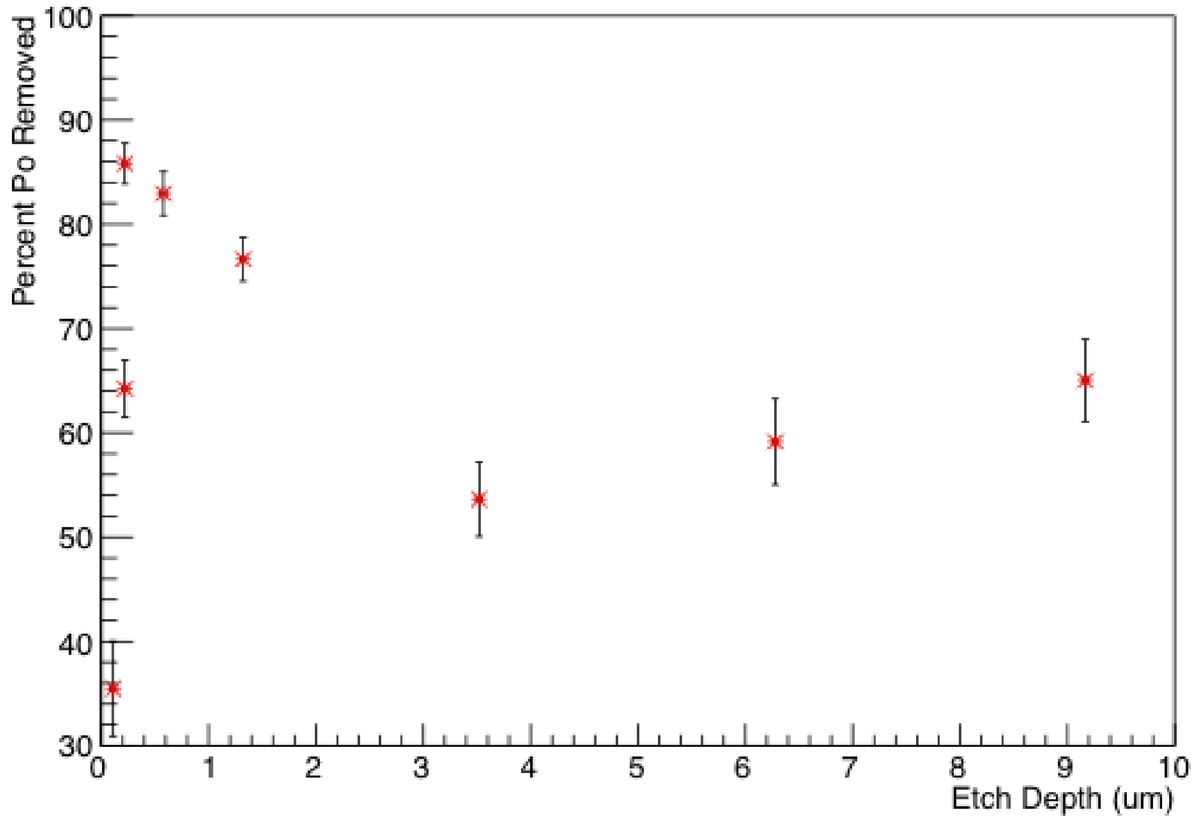
Po removal from Cu

Method:

- Etch using 3% H_2O_2/1% H_2SO_4 solution

Effective for removing of Bi, Pb, U, Th but why not Po?

Po removed as a function of etch depth



100 ml of 1% H_2SO_4, 3% H_2O_2

Po removal from Cu

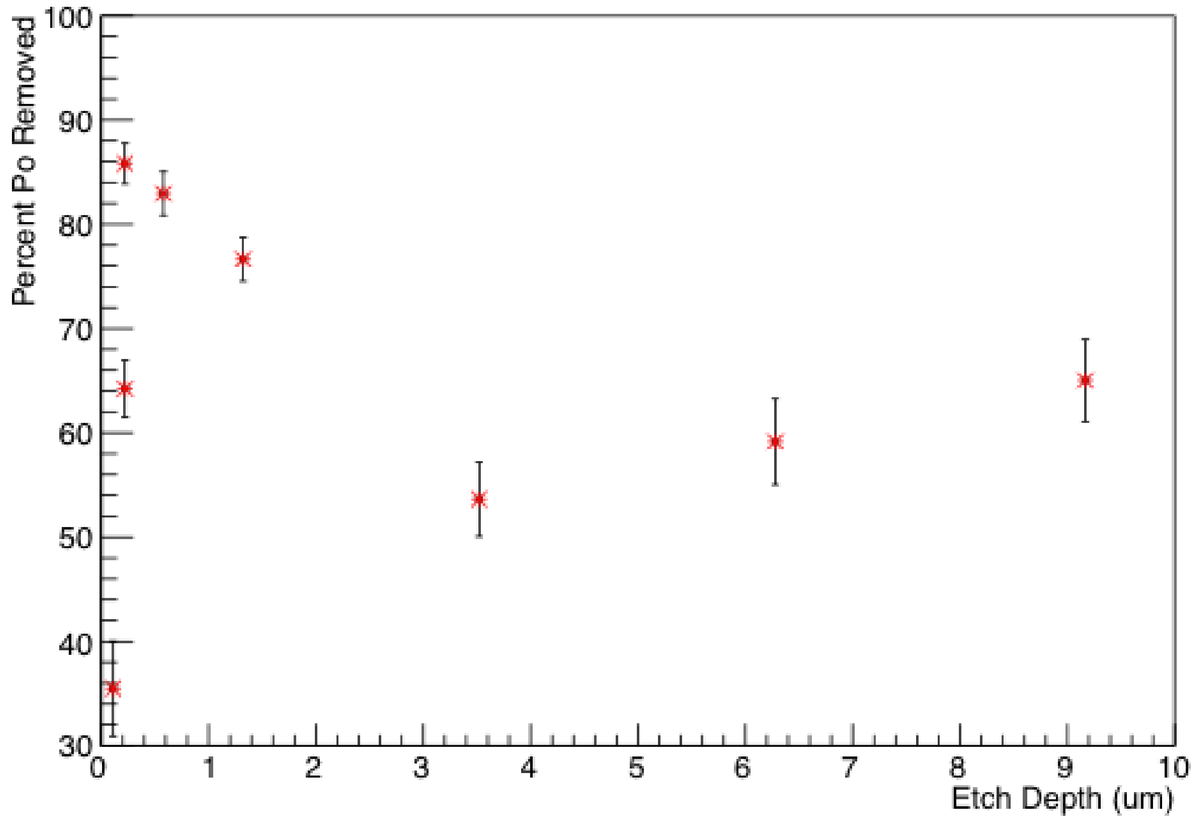
Method:

- Etch using 3% H_2O_2/1% H_2SO_4 solution

Evaluation of this shows initial Po removal, but subsequent re-deposition after removal of first 0.05 microns

Po redeposits as the etch depth of Cu continues showing there is a competitive half reaction within the hydrogen peroxide

Po removed as a function of Cu etch depth

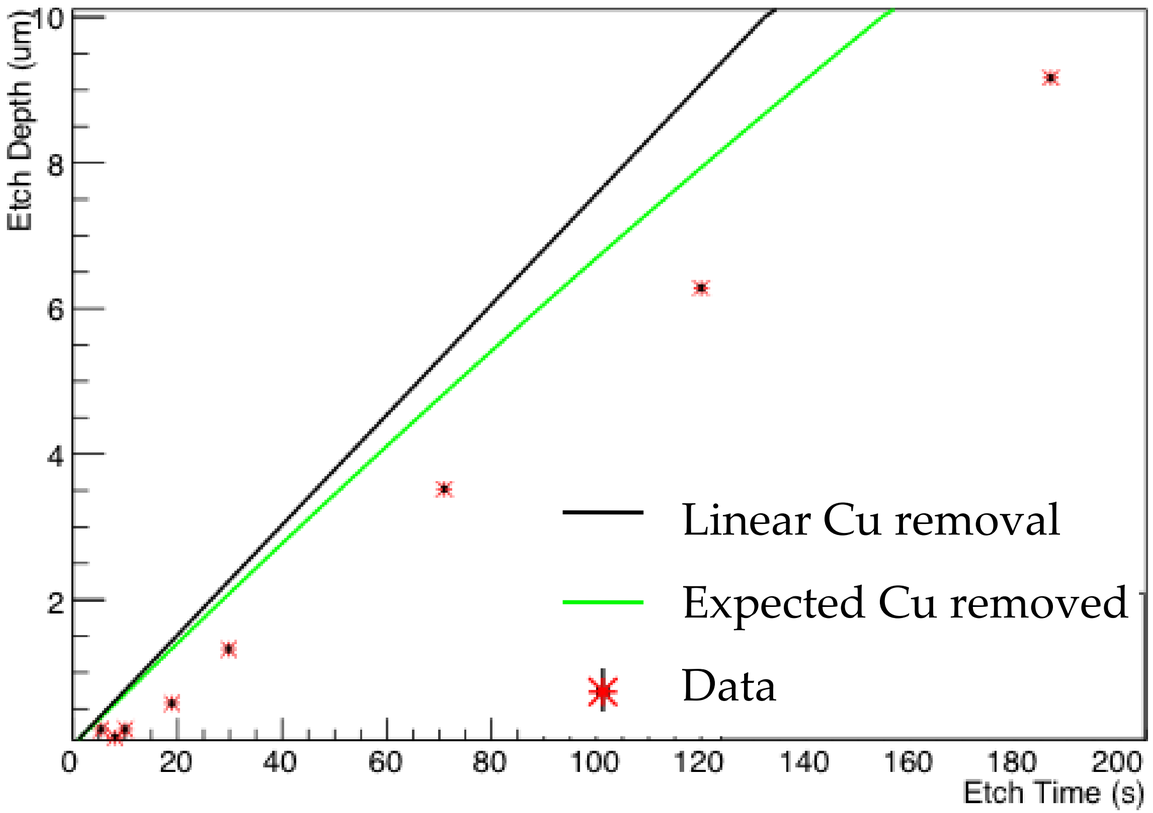


100 ml of 1% H_2SO_4, 3% H_2O_2

with agitation

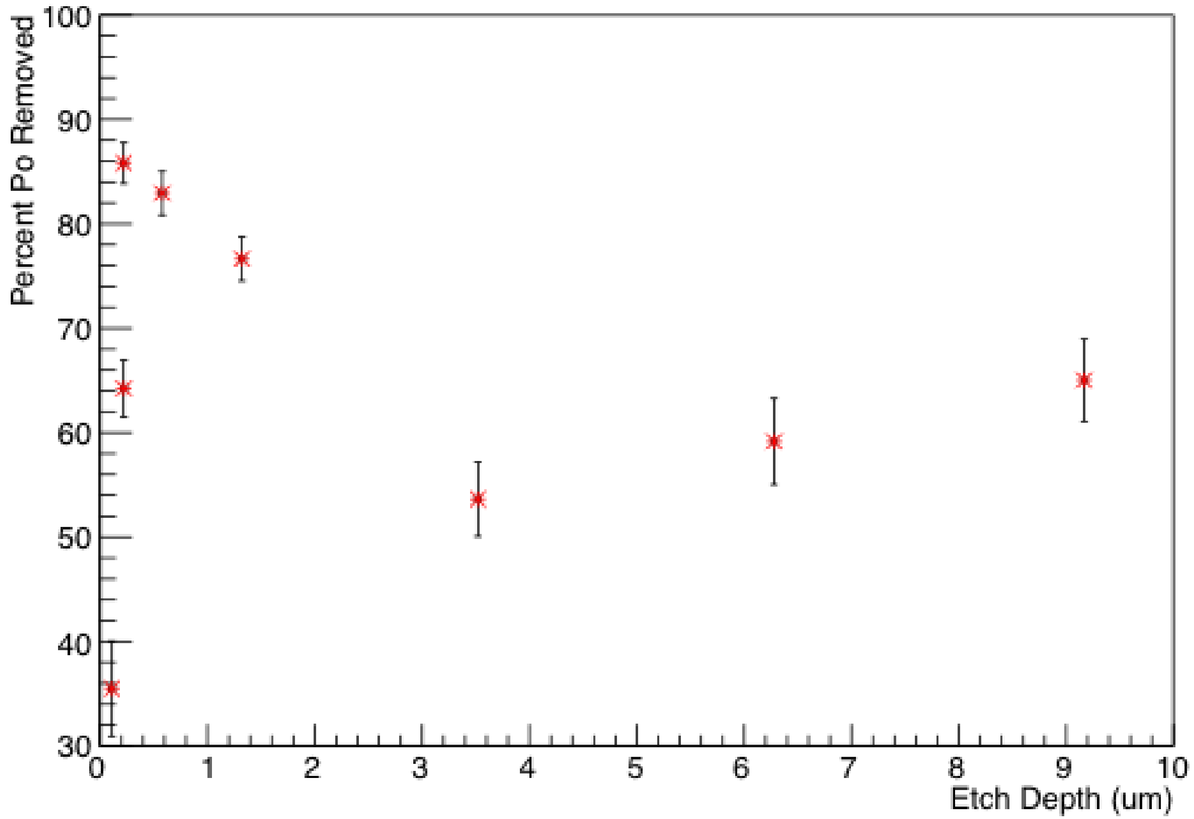
Po Removal from Cu

Cu removed vs. time for Po-spiked Cu samples



Expected Cu removal based on measured rate of Cu removal due to consumption of H_2O_2

Po removed as a function of Cu etch depth



Po removal but then subsequent reduction within the 1% H_2SO_4, 3% H_2O_2 solution during oxidation of Cu and consumption of H_2O_2

Po Removal from PTFE

Exploration of the standard plastic leaching process of a 72 hour period in high concentration HNO_3 solution to remove Po deposited from Rn exposure

Sample 1:

Nitric leaching does remove a fraction of Po, but only 40% here

Sample 2:

Repeat etches, following a stronger leach but does not change the amount of Po that remains on a surface

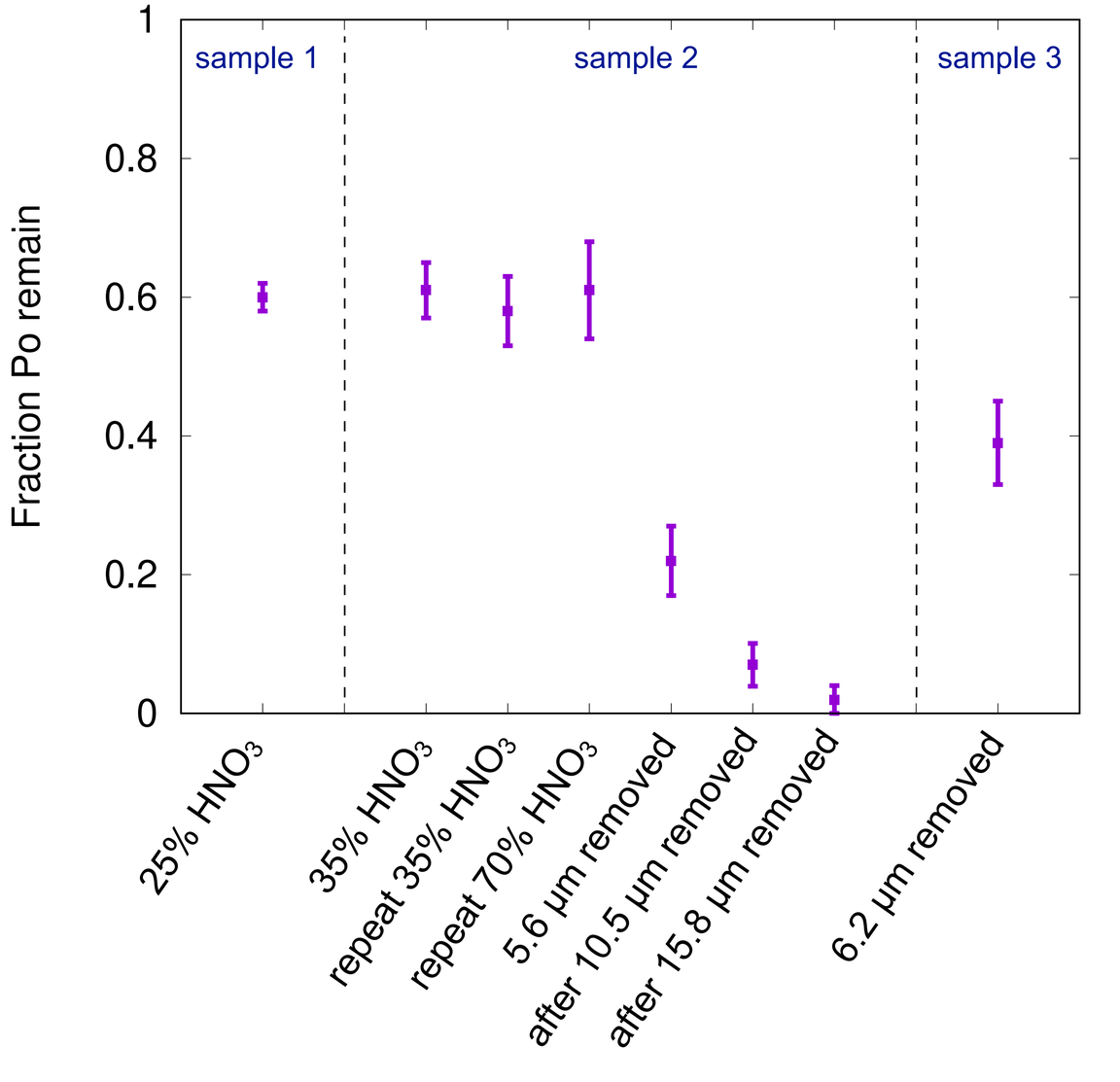
Physical surface removal was implemented after 3 attempts showing removal of the deposited Po

This is not a surprise nor new finding:

For example: DEAP-3600 resurfacing their acrylic vessel to remove Rn progeny [P. Giampa, LRT 2017]

Sample 3:

Only sanded, showing a decrease in Po yet must account for no removal of other substances due to lack of nitric leaching



Error bars are from counting statistics

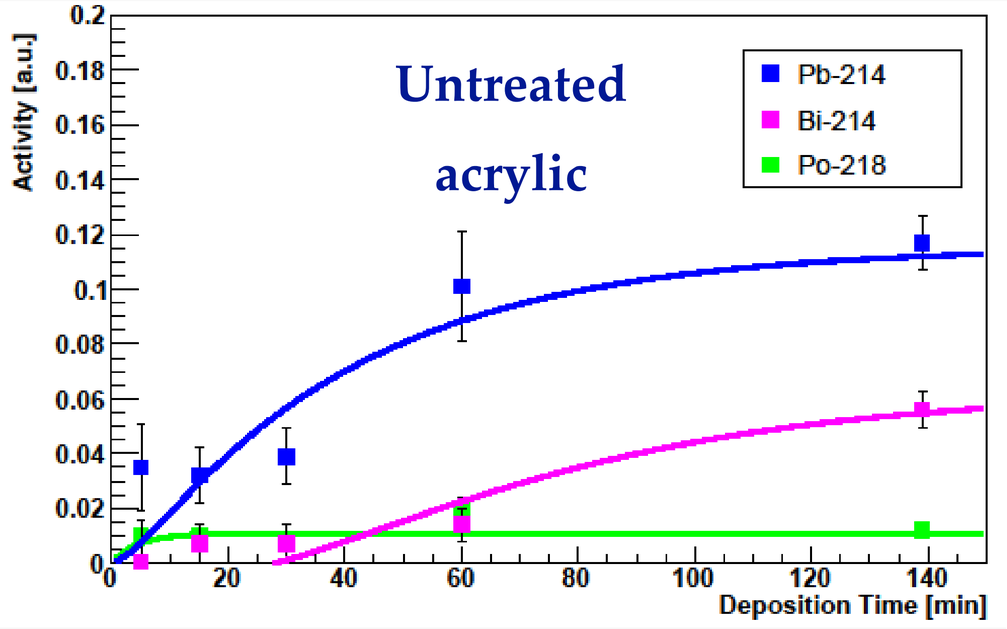
Rn progeny deposition on PTFE

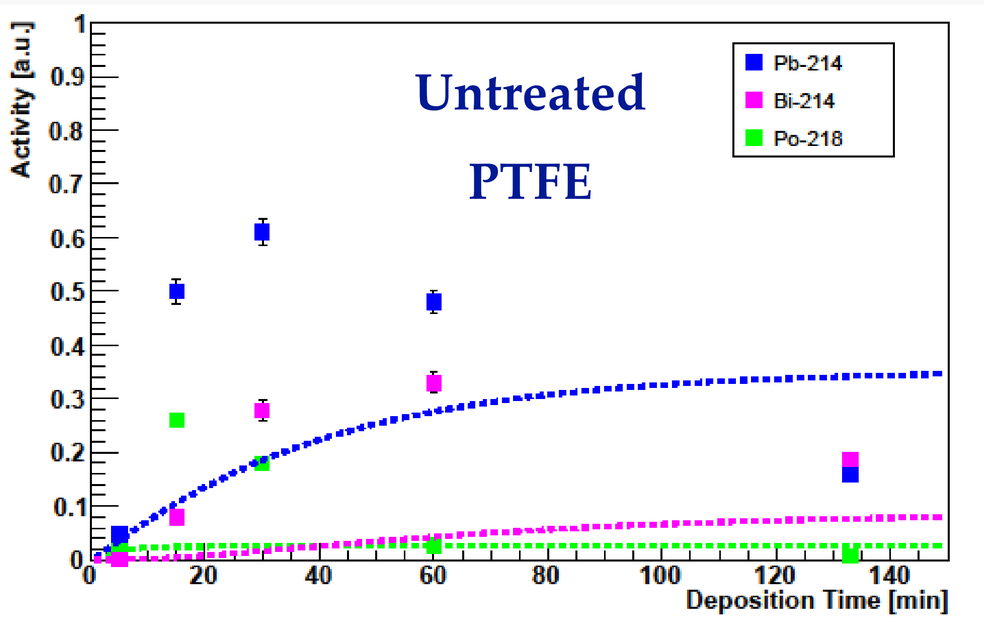
The Rn progeny deposition on **acrylic** follows a linear deposition model

The activity of each Rn progeny **approaches** a steady-state activity (concentration) signaling constant rate of deposition

For PTFE the activity of each Rn progeny **spikes** before dropping to a steady-state activity (concentration) signaling a non-linear rate of deposition

Rn progeny deposition as a function of Rn exposure time





Dashed lines represent acrylic linear deposition activities

Rn progeny deposition on PTFE

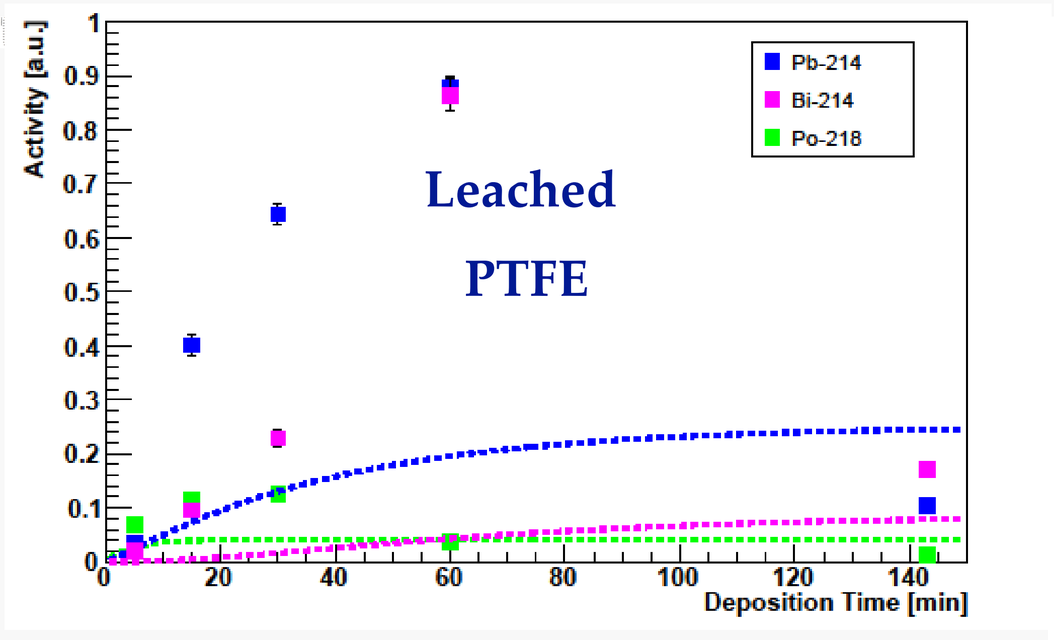
PTFE appears to be attract Rn progeny at a greater rate than other plastics

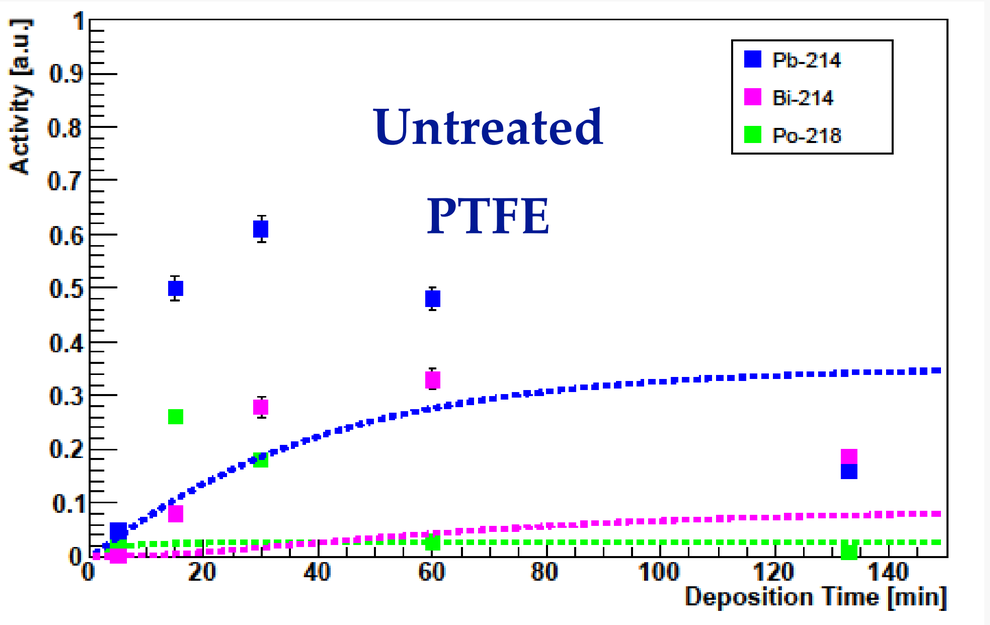
Nitric Leaching PTFE further strengthens the electrostatic attraction

For leached PTFE, an even greater Rn progeny activity (concentration) **spike** occurs before dropping to a steady-state activity

For PTFE the activity of each Rn progeny **spikes** before dropping to steady-state activity (concentration) signaling a non-linear rate of deposition

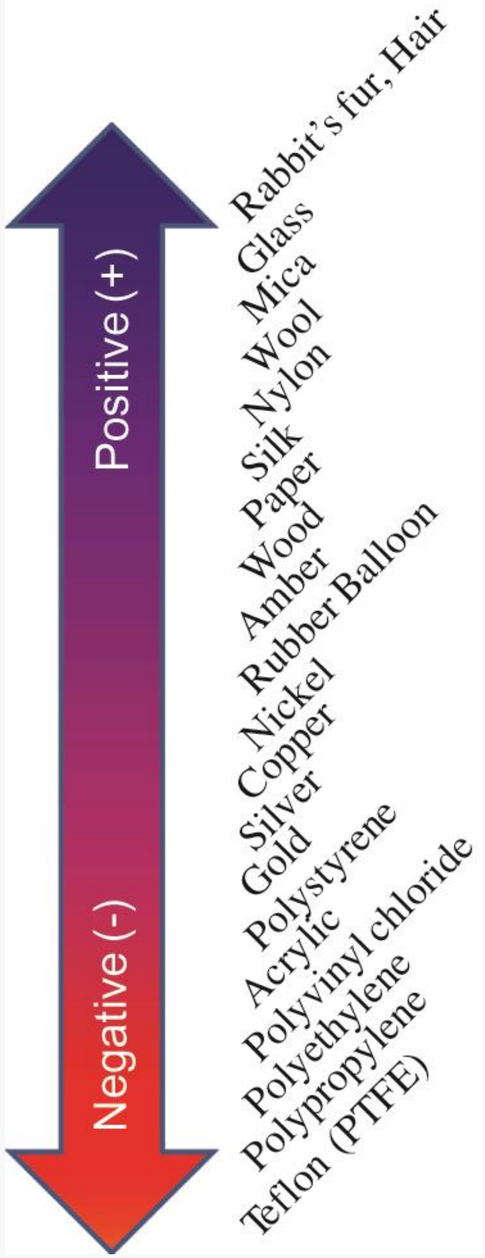
Rn progeny deposition as a function of Rn exposure time





Dashed lines represent acrylic linear deposition activities

PTFE Behavior



PTFE seems to attract Rn progeny at a greater rate than other plastics and other metals (Cu)

Based on **Triboelectric Series** you can see that PTFE has the greatest negative charge build up capability of static electricity, therefore strongly favoring Rn progeny

Leached PTFE may show a higher favorability for this state due to reduced contamination from other substrates and low humidity environments

Data shows the initial time after leaching attracts the most Rn progeny

Summary

Etching:

Based on the results of Cu etching, further development of the procedure must be done using hydrogen peroxide which allows for surface removal control that takes into account rate kinetics of the redox reactions by validation of technique

Leaching:

Surface contamination is still an issue and leaching should continue to handle the Th/U but unlike etching it does not penetrate deep enough to remove Rn progeny

Additional steps and handling must be accounted for after leaching to prevent against amplification of Rn progeny attraction

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Thank you

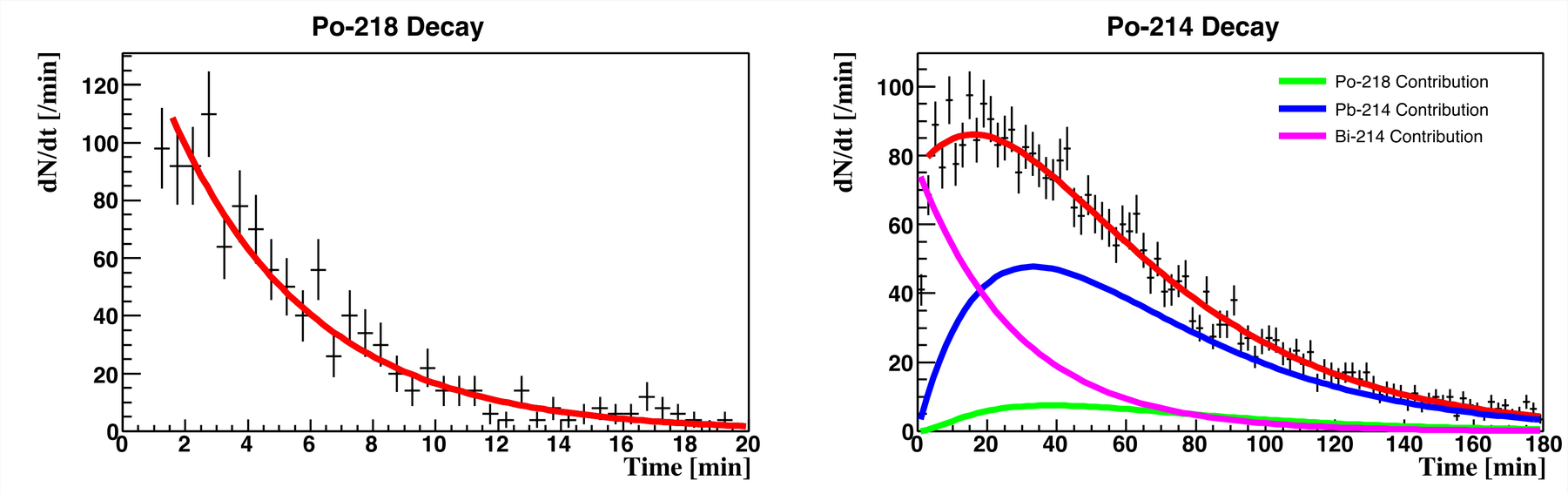
Questions?

Rn progeny deposition on PTFE

It is well known that Rn progeny are attracted to plastics (many examples cited just at this meeting)
It is known that electrostatics plays a strong role in the attraction of Rn progeny

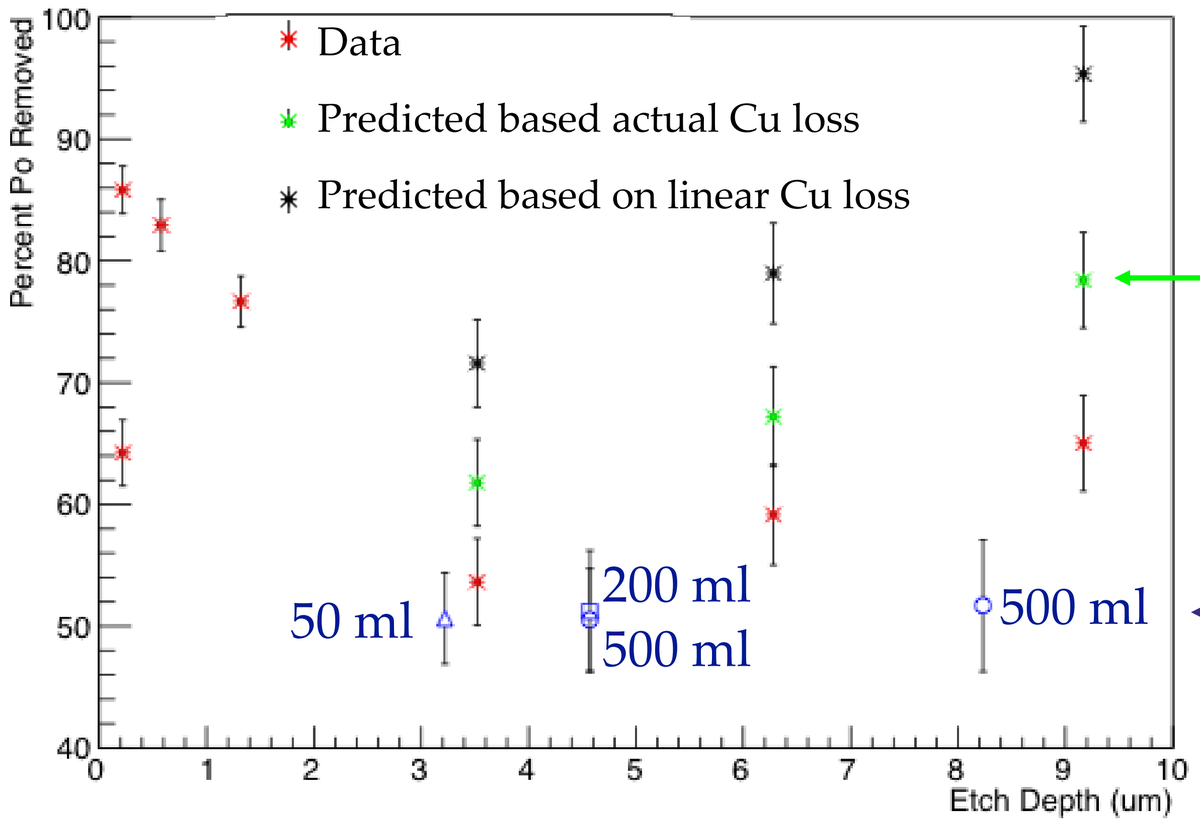
PTFE samples exposed to Rn to measure the Rn progeny deposition rate

The concentration/activity of the immediate progeny can be calculated from the Po-218 and Po-214 alpha decay rates after Rn exposure



Po Removal from Cu

Po removed as a function of the Cu etch depth



Predict a modest improvement if corrected for actual Cu removed

No improvement at greater volume

Even with the larger etch volume, the Po is not being oxidized

While more Cu is removed , the oxidation of Po is inhibited

Normalizing to the H_2O_2 lost, the predicted Po removal modestly improves

Competition between Po and Cu remains