Detector and Conventional Facilities Materials Assay Results: Introduction, Steel & Foam Results, and Outlook

Juergen Reichenbacher

South Dakota School of Mines & Technology (SDSMT)

DUNE DEEP UNDERGROUND NEUTRINO EXPERIMENT

DUNE Backgrounds Mitigation Strategies Workshop

July 22, 2020

2019/20 Assays Activities

SDSMT assayed shotcrete & concrete materials from ALL potential bidders in Black Hills area for informed decision making of LBNF for Conventional Facilities at SURF:
https://indico.fnal.gov/event/20144/session/19/contribution/262/material/slides/0.pdf

- GCC Cement Plant (Rapid City): Portland cement with rock from local RC quarry, fly ash (coal firing plant in NB)
- Pete Lien (Rapid City): GCC cement, fly ash (coal firing plant in NB), gravel (RC quarry), sand (Oral/SD, near Uranium-Mine)
- TCC (Rapid City): GCC cement, fly ash (coal firing plant in NB), gravel (South America), sand (packaged)
- Croell (Spearfish): GCC cement, fly ash (coal firing plant in NB), gravel (Sundance/WY), sand (Nisland/SD)
- Industrial and sump water from SURF (Davis 4850 ft)
- ProtoDUNE Cu-getter & molecular sieve (zeolite) -> Stephen Pordes, David Montanari
- ProtoDUNE Steel (I-beams) and SS cryo-wall -> Filippo (Luxembourg corp.)

Gamma-, Alpha- and Beta-Ray Spectrometers at SDSMT for Radiological Assays

RABBIT:

Large, Fast Loading and Versatile Gamma-Ray Spectrometer (BEGe)

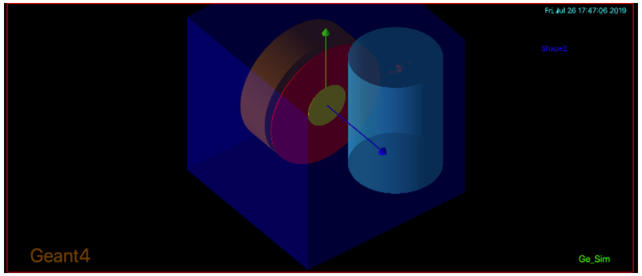
Large Alpha-, Beta-Ray Spectrometers (big AlphaBACH and portable AlphaBACH)

Uniquely large volume AND meeting world's best sensitivity!

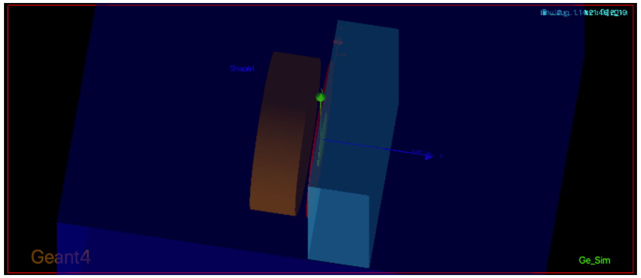
Measuring Samples for Gamma-Ray Assays with RABBIT

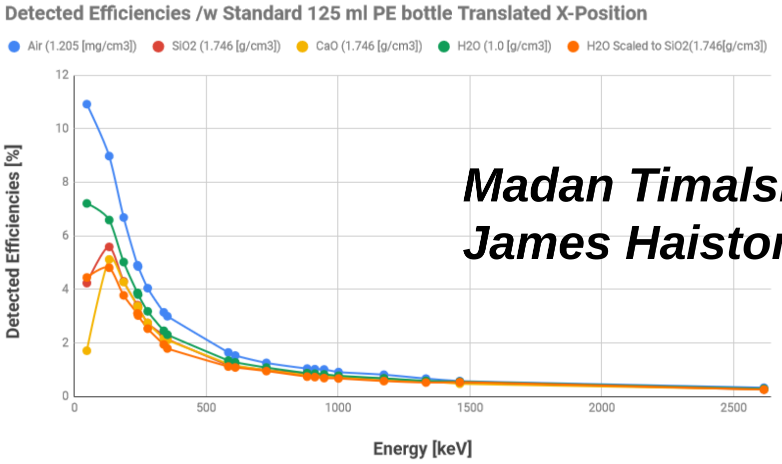
125 ml bottles of cement, fly ash, sand, gravel, water, Cu-getter, molecular sieve

Steel samples

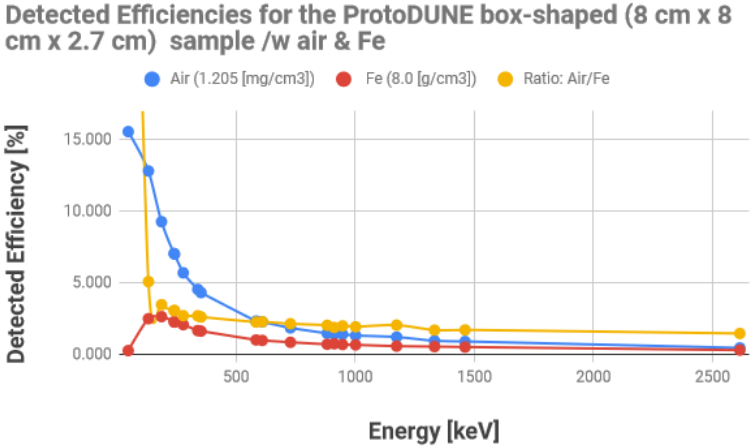


Geant4 simulations of each sample with Ge-detector geometry





Madan Timalsina (LZ), James Haiston & Juergen



Geant4 Simulated Detection Efficiencies (e.g. Steel Samples)

Madan Timalsina (LZ),
James Haiston & Juergen

Isotope	Energy [keV]	Detected Efficiencies for the ProtoDUNE Box-shaped sample /w air & Fe									
		1mm /w 1mylar&1C (10k Events)	76 mm /w 1mylar&1C (10m Events)	76 mm /w 1mylar&1C (250m Events)	Box: 8 cm x 8 cm x 2.7 cm			Box: 8 cm x 8 cm x 0.95 cm			
					Air (1.205 [mg/cm3])	Fe (8.0 [g/cm3])	Ratio: Air/Fe	Air (1.205 [mg/cm3])	Fe (8.0 [g/cm3])	Ratio: Air/Fe	
Pb-210	46.539	37.899	3.84371		15.570	0.285	54.632	20.244	0.875	23.136	
Pa-234	131.3	33.034	3.09356		12.825	2.515	5.099	16.577	6.528	2.539	
Ra-226	186.211	25.216	2.20493		9.282	2.657	3.493	12.317	6.309	1.952	=> Rn-222
Pb-212	238.632	19.487	1.63666		7.068	2.297	3.077	9.329	5.072	1.839	
Ra-224	240.986	19.229	1.61381		7.020	2.278	3.082	9.249	5.032	1.838	
Tl-208	277.351	15.908	1.31439		5.715	2.099	2.723	7.637	4.344	1.758	
Ac-228	338.32	12.575	1.01519		4.564	1.682	2.713	5.978	3.679	1.625	=> Th-232
Pb-214	351.932	11.904	0.96011		4.335	1.646	2.634	5.665	3.493	1.622	
Tl-208	583.191	6.653	0.50911		2.346	1.030	2.278	3.045	2.043	1.490	
Bi-214	609.312	6.306	0.48592		2.292	0.996	2.301	2.974	1.976	1.505	
Bi-212	727.33	5.337	0.39969		1.870	0.866	2.159	2.470	1.675	1.475	
Pa-234	881.826	4.425	0.32423		1.493	0.728	2.051	2.071	1.411	1.468	
Ac-228	911.204	4.173	0.31264		1.444	0.759	1.903	1.987	1.436	1.384	=> Th-232
Pa-234	946.183	4.114	0.30000		1.426	0.710	2.008	1.948	1.321	1.475	
Pa-234m	1001.026	3.844	0.28043		1.3350	0.6858	1.9466	1.7914	1.2982	1.3799	=> U-238
Co-60	1173.237	3.217	0.23504		1.231	0.597	2.062	1.479	1.113	1.329	
Co-60	1332.501	2.794	0.20796		0.968	0.564	1.716	1.312	0.984	1.333	
K-40	1460.83	2.579	0.19113		0.923	0.534	1.728	1.183	0.928	1.275	
Tl-208	2614.533	1.431	0.09861	0.0984096	0.473	0.319	1.483	0.662	0.549	1.206	

Existing 'Steel' Assays on DB on Radiopurity.org

Grouping	Name	Isotope	Amount	Isotope	Amount
ILIAS UKDM	Steel	Th-232	0.4 ppb	U-238	1 ppb
ILIAS UKDM	Steel, sheet	Th-232	150 ppm	U-238	160 ppb
ILIAS Edelweiss	Stainless steel				
ILIAS ROSEBUD	Stainless steel				
Rau (2000)	Stainless steel				
ILIAS UKDM	Steel, mild	Th-232	100 ppm	U-238	60 ppb
ILIAS ANAIS	Stainless steel				
ILIAS Edelweiss	Stainless steel				
BOREXINO (2002)	Stainless steel, piping	Th-232	2.5E-9 g/g	U-238	1.1E-9 g/g
BOREXINO (2002)	Stainless steel for flanges	Th-232	1.6E-9 g/g	U-238	5.0E-10 g/g
ILIAS UKDM	Steel, scaffold	Th-232	150 ppm	U-238	160 ppb
ILIAS UKDM	Steel, stainless, frame, USA	Th-232	1.6 ppb	U-238	0.37 ppb
ILIAS UKDM	Steel, stainless, USA	Th-232	0.6 ppb	U-238	0.9 ppb
ILIAS Edelweiss	Stainless steel, 304L				
BOREXINO (2002)	Stainless steel for storage vessel	Th-232	9.3E-10 g/g	U-238	1.4E-9 g/g
ILIAS Edelweiss	Stainless steel, Jacquet SAS				
ILIAS UKDM	Steel, stainless, cryogenic	Th-232	2 ppb	U-238	3 ppb
ILIAS UKDM	Steel, stainless, rod, USA	Th-232	0.24 ppb	U-238	0.24 ppb
ILIAS Edelweiss	Stainless steel, Jacquet SAS				
ILIAS UKDM	Steel, stainless 304	Th-232	3.6 ppb	U-238	3.2 ppb
ILIAS UKDM	Steel, stainless, USA				
ILIAS Edelweiss	Stainless steel, 304L				
EDELWEISS (2011)	Mild steel, support	Th-232	0.01 ppb	U-238	0.01 ppb
ILIAS UKDM	Steel, stainless, USA	Th-232	4.0 ppb	U-238	3.4 ppb
ILIAS Edelweiss	Stainless steel, 304L				
ILIAS UKDM	Steel, stainless 316L	Th-232	1.7 ppb	U-238	1.3 ppb

= 12 mBq/kg

= 2 Bq/kg

= 3/4 Bq/kg

= 2 Bq/kg

1 Bq ^{238}U/kg = 81 ppb U (81 10^{-9} gU/g)

1 Bq ^{232}Th/kg = 246 ppb Th (246 10^{-9} gTh/g)

1 Bq ^{40}K/kg = 32.3 ppm K (32.3 10^{-6} gK/g)

Intrinsic Neutron Rate from Detector Materials and Cryostat (Requirements)

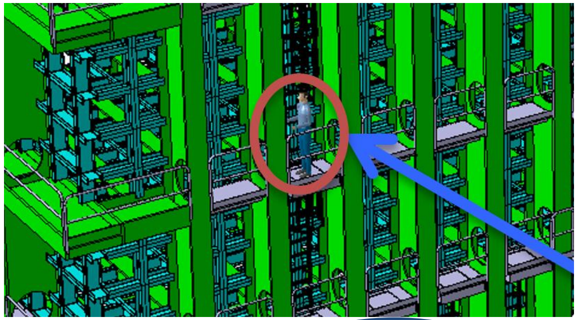
U-238 w/ SF factor of 5.45e-7

Mode	Branching (%)
a	100
SF	5.45E-5 7

Mass I-Beam max: 9,500 kg x 40 x 4 sides = 1.52 ktons

⇒ **0.83 Hz Neutron Rate with 1 Bq/kg of U-238 (and just 1 neutron per fission enters of the <2.1> neutrons / fission emitted)**

⇒ **Requirement (<1 Hz): < 1 Bq/kg < 81 ppb**



External moderator only partially useful for this

Mass APA steel frame max: 0.5 ton x 150 APAs = 75 tons

⇒ **0.08 Hz Neutron Rate with 1 Bq/kg of U-238 (and 2 neutrons per fission contained)**

⇒ **Requirement (<1 Hz): < 12 Bq/kg [no issue expected]**

Mass CPA (3 mm thick FR4): 0.3 cm x 1.85 g/cm^3 x 12 m x 60 m x 2 = 8 tons

⇒ **0.009 Hz Neutron Rate with 1 Bq/kg of U-238 (and 2 neutrons per fission contained)**

⇒ **Requirement (<1 Hz): < 110 Bq/kg [no issue expected]**

Assay Results of ProtoDUNE Steel Samples

Steel I-beams: ***(2.6 +/- 0.8) Bq/kg U-238***
= (211 +/- 65) ppb U-238

(Ra-226 <1.1 Bq/kg @ 90%CL and
Th-232 <0.1 Bq/kg @ 90%CL)

SS of cryostat wall: ***(3.3 +/- 2.3) Bq/kg U-238***

(Ra-226 <1.7 Bq/kg @ 90%CL and
Th-232 <0.1 Bq/kg @ 90%CL)

Steel of I-Beams (Warm Structure of Cryostat): U-238 SF

Mass of I-beams: 1.52 ktons, *U-238 w/ SF factor of 5.45e-7*

~2 Hz Neutron Rate with 2.6 Bq/kg of U-238

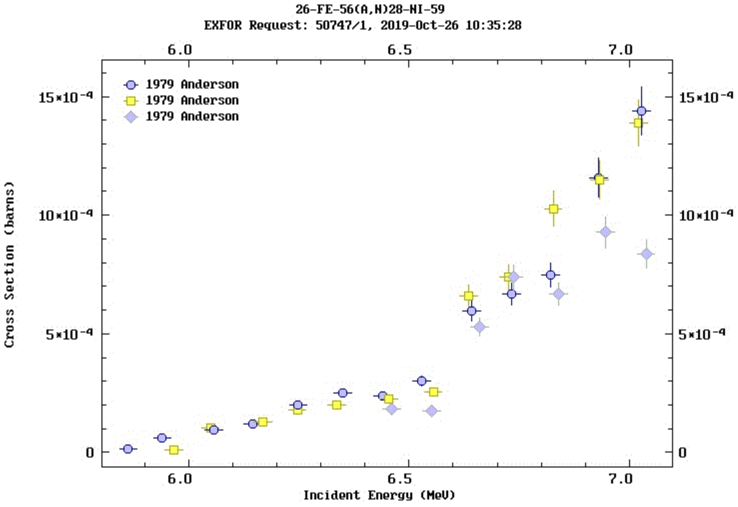
(if just 1 neutron per fission enters of the <2.1> neutrons / fission emitted)

[Simulations with 15 keV, Cf-252 and AmBe neutron energy spectra started inside our LAr cryostat show that only about half of neutrons capture in active LAr)

Steel of I-Beams (Warm Structure of Cryostat): U-238 SF + (a,n)

Mass of I-beams: 1.52 ktons, *U-238 w/ SF factor of 5.45e-7*

~2 Hz Neutron Rate with 2.6 Bq/kg of U-238 (and just 1 neutron per fission enters)



<u>Neutron production from 56Fe(alpha, neutron):</u>

*1 Bq/kg * 8 = ~10 Bq/kg alpha activity*
*=> 10 Bq/kg * 1,520,000 kg = 15,200,000 alpha/sec*

[8.6e22/cm^3 (56 u in 8 g/cm^3)]
If I assume a cross section for 56Fe(alpha, neutron) of 1 mbarn
*expect 1e-3 * 10^-24 cm^2 * 1.52e7 * 8.6e22/cm^3 * 0.002 cm / 2 = 1.3 neutron / sec*

-> 2.8 neutrons/sec from (a,n) could enter per 1 Bq/kg U-238 (daughters in sec. equil.)

[54Fe(alpha,n) about equivalent because ~20 mbarn with just 5.8% abundance]

Most Important Concerning Results from Steel Assays:

- *Steel of I-beams is higher than expected and in principle could even be higher and we still had to accept the I-beams from exclusive manufacturer in Luxembourg:*

  (2.6 +/- 0.8) Bq/kg in U-238 could translate into many Hz of neutron rate!

  But signs of stratification during manufacturing process
  (Ra-226 out of equilibrium)

  => sample steel coupons from different heights in smelt (-> Dimitar, Filippo)

  => Try to establish simple acceptance criterion that does not increase costs

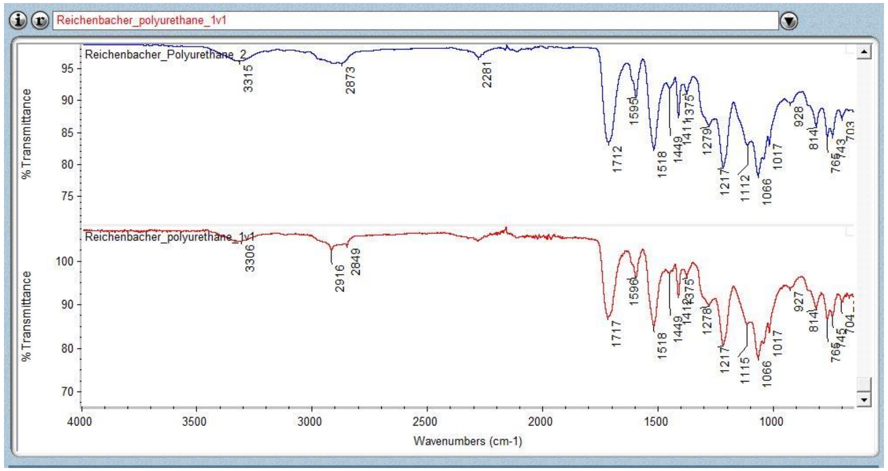
R-PUF ProtoDUNE Foam Samples:

H-Content (Neutron Moderation)

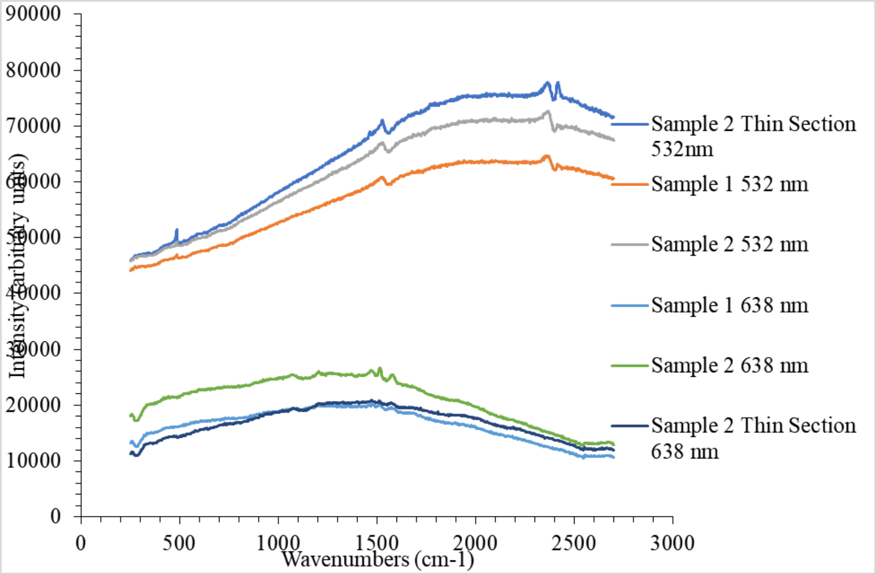
Specific Activity

FT-IR and Laser Raman Spectroscopy on 2 R-PUF ProtoDUNE Foam Samples

FT-IR



Raman



FT-IR indicates polyESTER-urethane (but one table assigns polyETHER-urethane

Raman result hints at polyESTER-urethane, but is even less conclusive

CHN Analysis at UIowa

<64.41> mass % C
<5.95> mass % H
<5.5> mass % N
<24.14> mass % O (100% - CHN%)

With a factor of 10 the relative quantity unit for N becomes approx. 4.
And including oxygen (accounting for the remainder), we get then a stoichiometric formula of our molecule of

$C54\ H60\ N4\ O15$ **-> this measured value is now in GDML file for full 10 kton cryostat geometry**
or
$C27\ H30\ N2\ O7.5$
or
$C13.5\ H15\ N1\ O3.75$

compared to literature polyESTER urethane:
$C17\ H15\ N2\ O4$

This gives us a good match for polyESTER urethane (instead of polyETHER urethane)!

And compared to our existing computer simulation (GDML material definition "Layer2Molecule"):
~~$C17\ H16\ N2\ O4$~~ **-> has been substituted with measured value now**
it is not that far off.

But it is largely different from literature polyETHER urethane R-PUF w/ HFC 245 "our best case for neutron moderation" (most H abundance):
$C16\ H32\ N2\ O4$ **-> would have been better, but GTT company is apparently not using it ☹**

Material budget and expected source rates

- From radioactive isotopes in cryostat and other materials
- Radiopurity very dependent on the material/production process
- Order of magnitude for steel >1mBq/kg
- **Assumed here 10mBq/kg in all materials, 50% U, 50% Th , except foam ==2Bq/kg**
- Decay photons included

		Mass [t]	Disint/year 10mBq/kg	Fission [n/year]	Alpha,n [n/year] 10mBq/kg
External SS	SS460ML	250	7.9e10	71 000	53 490
Foam	polyurethane	34	~~1.1e10~~ 7.5E12 ?	~~9 600~~ 6.7e6 ?	~~22 516~~
Internal SS	SS304	4	0.13e10	3 400	1 168
Wood	plywood	12	0.4e10	1 200	3 257

EMES RESEARCH LABORATORY ~
ENGINEERING and MINING EXPERIMENT STATION

Home Facilities Personnel

ICP-MS Facility Primary Contact
Dr. Scott Beeler
Research Scientist II - Analytical Chemistry
Email: Scott.Beeler@sdsmt.edu
Office: Mineral Industries 212B
ph. (605) 394-2380

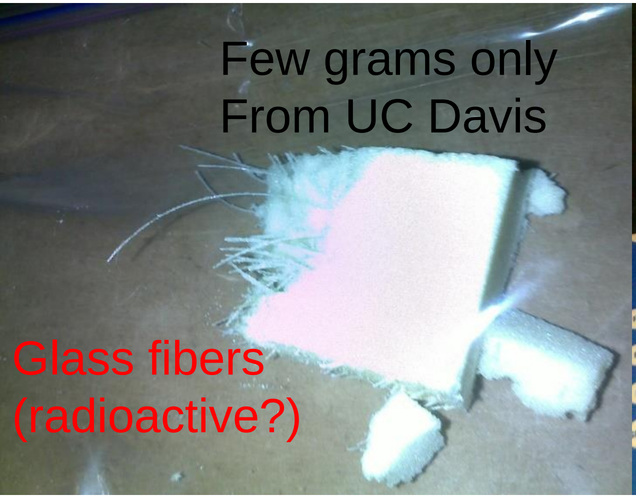
EMES
Providing analytical services to public and private sectors since 1903.

Inductively Coupled Plasma Mass Spectrometry (ICP/MS)

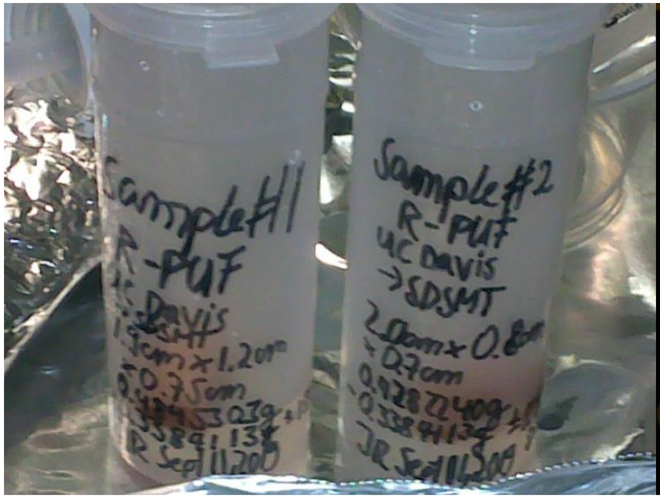
Facility Overview

Inductively coupled plasma mass spectrometry (ICP-MS) can be utilized for both quantitative and semi-quantitative analysis of most elements in the periodic table and is commonly employed in a wide range of geologic, environmental, biological, and industrial applications. The primary strengths of ICP-MS lie in its ability to provide rapid, sensitive, and precise multi elemental analyses across a high dynamic range enabling measurement of elemental concentrations ranging from sub parts per billion to hundreds of parts per million in a single run. The EMES is equipped with an Agilent 7900 quadrupole ICP-MS for determination of trace, minor, and major elements in aqueous solutions and acid digestions.

Foam Radioactivity Assay with ICP-MS



(=> Need 10 cm x 10 cm x 10 cm
sample for gamma assay)



Failed because foam
does not digest in HNO3

-> we need HF
-> BHSU has HF kit
-> BHSU has Laser Ablation

(BHSU has no tech,
but we do at MI-Dept.)

UCL @ Boulby
Or PNNL as backup