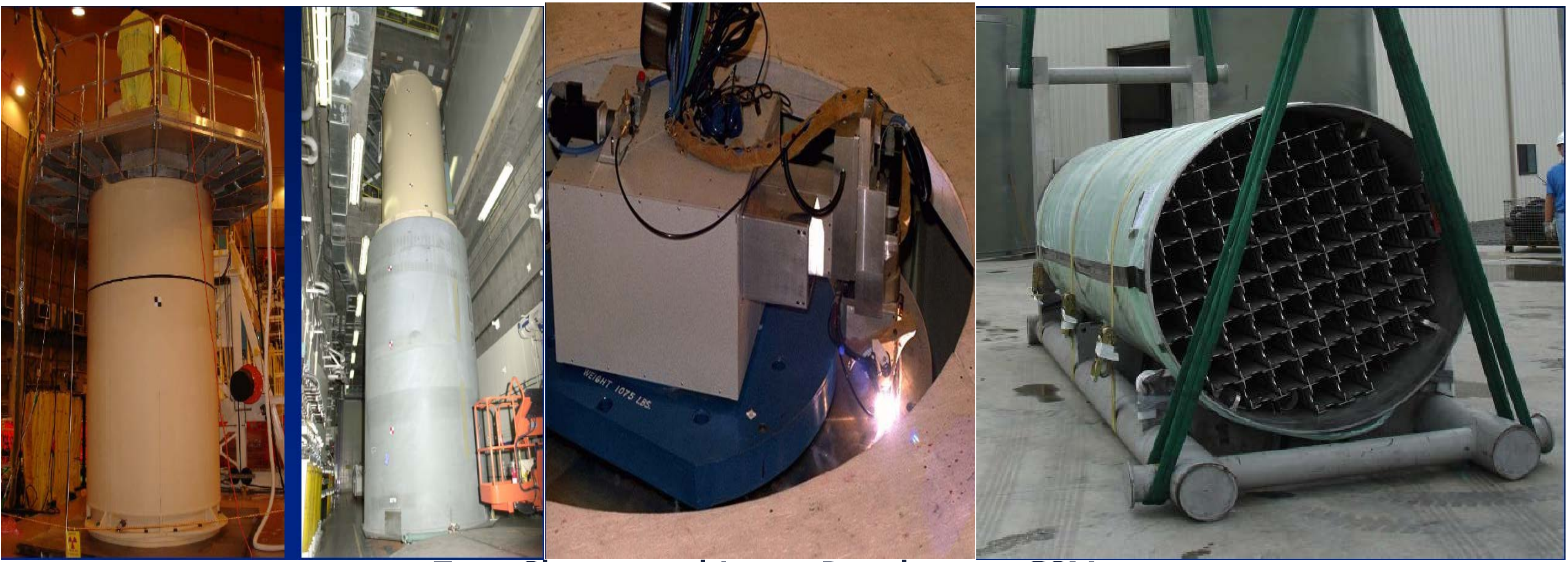
Innovative Approach to SCC Inspection and Evaluation of Canister in Dry Storage
NEUP\IRP-15-9318

Chloride Detection and Life Prediction of Dry Storage Casks Using PGAA and NAA Techniques



Zeev Shayer and Jason Brookman– CSM

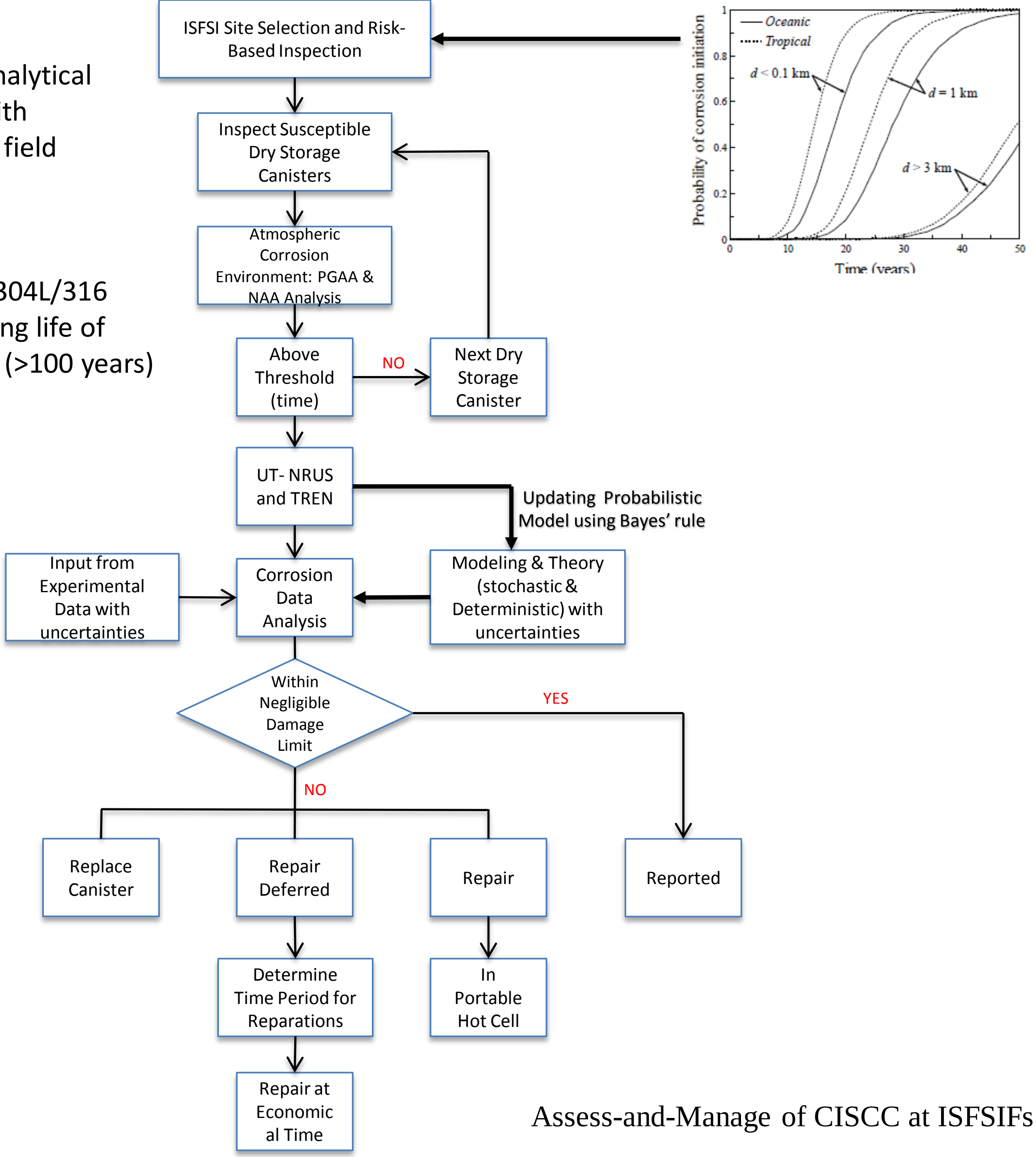
2016 UFD WG Meeting, UNLV, June 7-9

Outline

- PPGAA and NAA Methodology – Application to UNF Dry Storage Canister Susceptible to CISCC
- Simulation Methodology – Monte Carlo Code (MCNP6)
- Neutron Sources and Materials Data
- Background, Signal Processing and Probability of Detection
- Summary– Preliminary Results, Conclusions and Future Work.

Predict the service life through analytical rate expressions and modeling with uncertainties, and data from the field measurements

Lead to Better Validation of 304/304L/316 stainless steels survivability to long life of Canister at UNF Dry Storage Cask (>100 years)



ISFSI Site Selection and Risk-Based Inspection

Inspect Susceptible Dry Storage Canisters

Atmospheric Corrosion Environment: PGAA & NAA Analysis

Above Threshold (time)

NO

Next Dry Storage Canister

UT- NRUS and TREN

Updating Probabilistic Model using Bayes' rule

Input from Experimental Data with uncertainties

Corrosion Data Analysis

Modeling & Theory (stochastic & Deterministic) with uncertainties

Within Negligible Damage Limit

YES

NO

Replace Canister

Repair Deferred

Repair

Reported

Determine Time Period for Reparations

In Portable Hot Cell

Repair at Economical Time

Assess-and-Manage of CISCC at ISFSIFs

PGAA&NAA Methodology

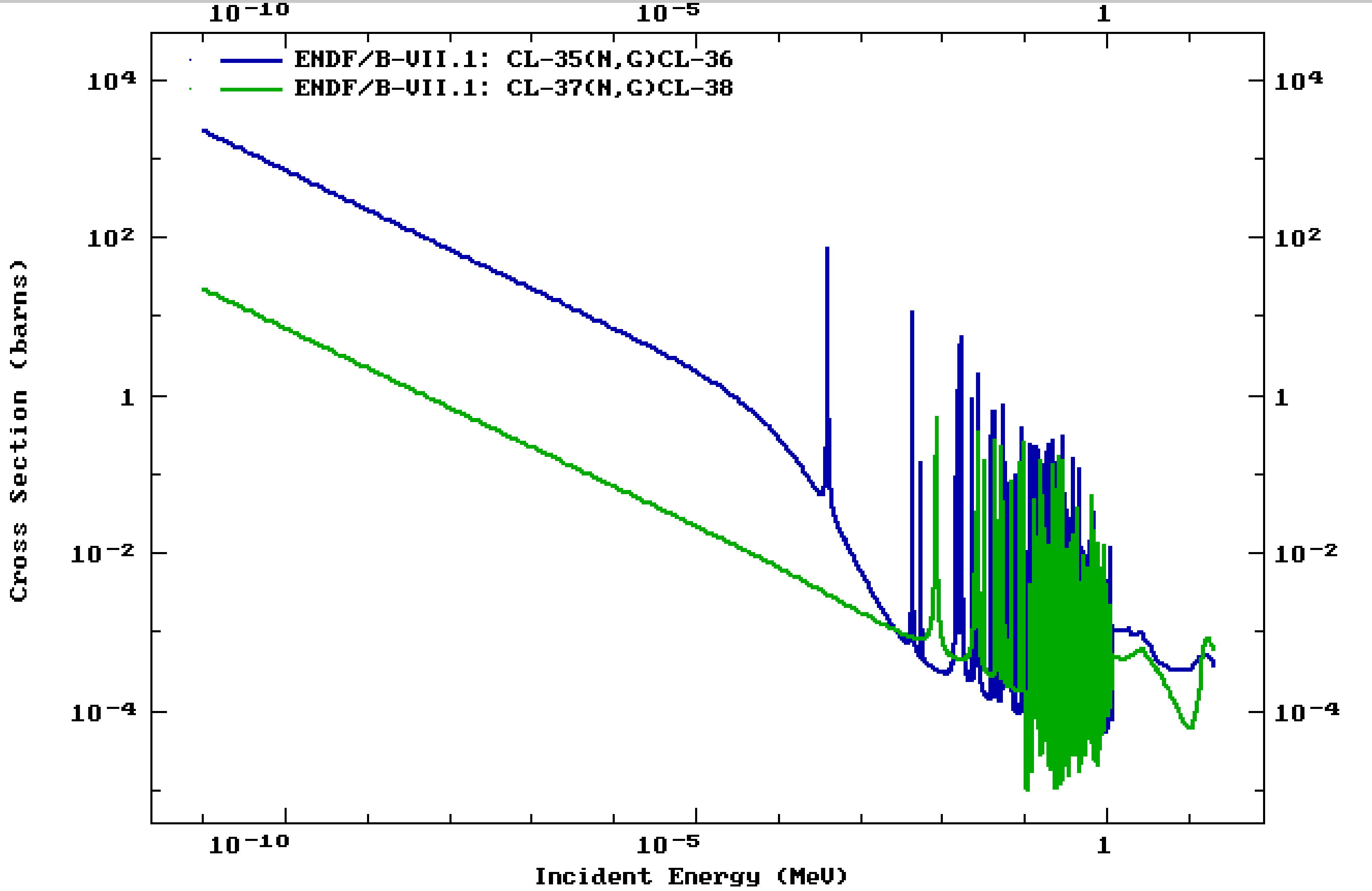
Neutron Activation Analysis for Salt Deposition

Natural chlorine consists of two isotopes ^{35}Cl (75.5%) and ^{37}Cl (24.5%).

^{35}Cl(n,γ)^{36}Cl, where ^{36}Cl is a radioactive isotope (half-life of 3.0×10^5 years) that upon decay, emits gamma ray energies of 517, 786, 1165, 1951 KeV and **6.1 and 7.4 MeV**. The second is ^{37}Cl(n,γ)^{38}Cl; ^{38}Cl is an unstable isotope with a half-life of 37.3 min, that decays by emission of a β particle and a γ ray with an energy of **1.64 or 2.17 MeV**, and forms a stable argon isotope, ^{38}Ar.

PGAA and NAA Simulated Using Monte Carlo (MCNP6) to estimate minimum detectable concentration of chlorine.

Absorption Cross-Section Data of Chlorine
ENDF/B-VII

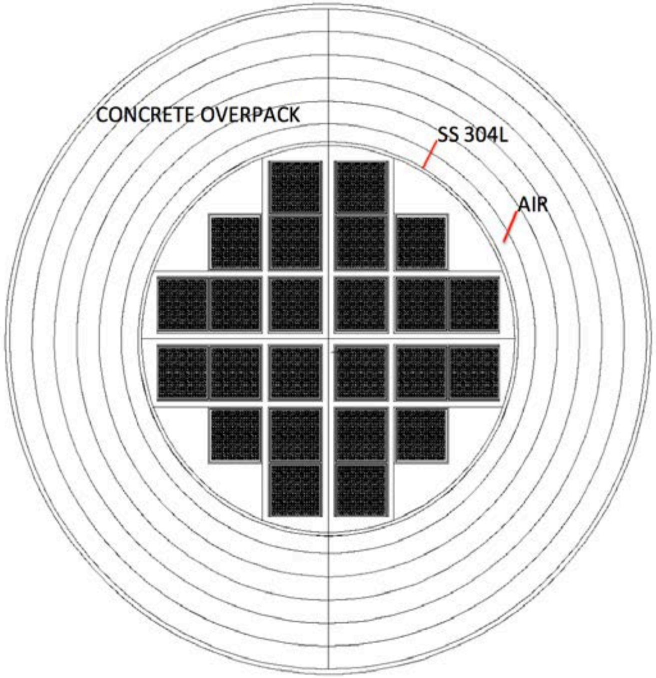


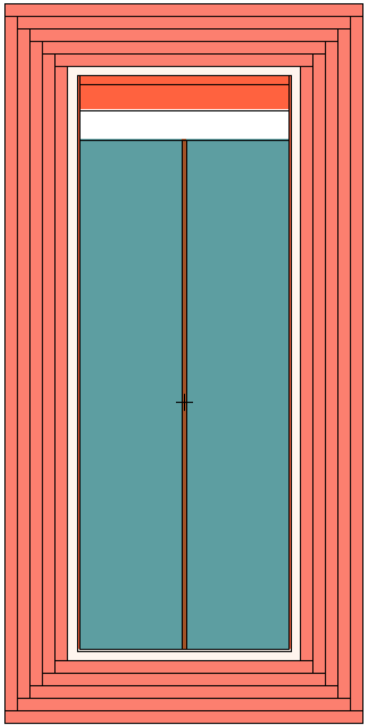
Summary of Cross-section Data of Chlorine

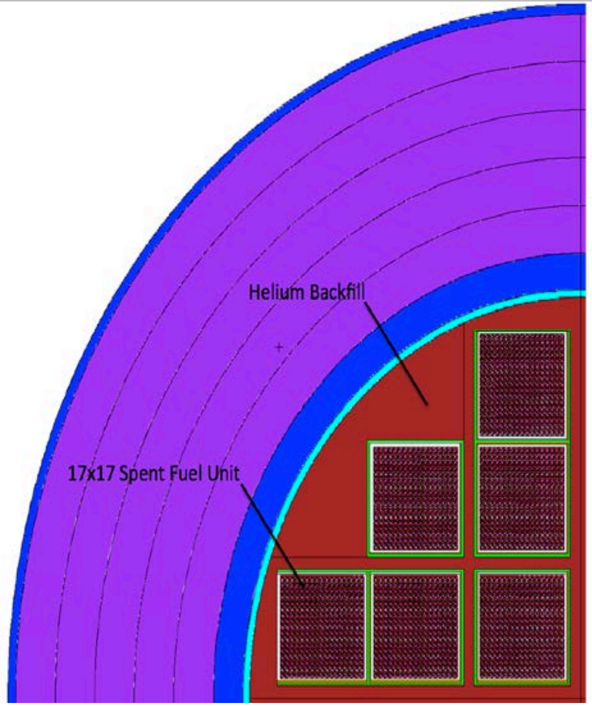
Element	Reactions	Neutron Energy	Reaction Type	Energy (MeV)
H	^{2}H (n, γ) ^{2}H	Thermal	Prompt	2.23
N	^{14}N (n, γ) ^{15}N	Thermal	Prompt	2.31
Cl (75.5% ^{35}Cl+24.5% ^{37}Cl)	^{35}Cl (n, γ) ^{36}Cl	Thermal	Prompt	1.96 ; 6.1; 7.41
	^{35}Cl (n, n $'\gamma$) ^{36}Cl	Fast	Prompt	0.6
	^{37}Cl (n, γ) ^{38}Cl	Thermal	delayed	1.64; 2.16
		$T_{1/2}$		
^{36}Cl	3.0x10^{5} years	^{36}Ar		
^{38}Cl	37.3 min	^{38}Ar		

Geometrical Simulation – MCNP6

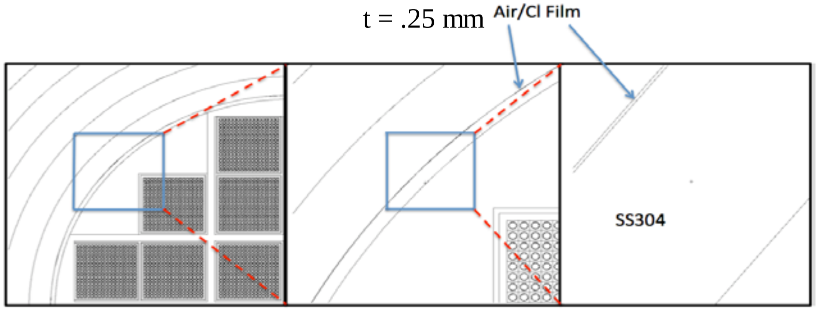
Holtec, International and Sierra Nuclear Corporation VSC-24 storage system, 10 Years Cooling (typical PWR storage arrangement)







- Helium back-filled
- 17x17 spent fuel storage units (24 count)
- SS304 canister thickness = 5/8"
- Air gap between outer wall of canister and inner wall of concrete overpack = 3"
- A total concrete thickness of 50 cm
- Concrete shelled with surfaces at 10 cm intervals for tally analysis



Marine Atmospheric & Materials Data – MCNP6

Dry Air Composition	
Element	Wt %
N	0.75527
O	0.23178
C	0.00012
Ar	0.01283

Sea Salt Composition	
Element	Wt %
Na	0.308
Cl	0.555
Mg	0.037
Ca	0.012
K	0.011
S	0.026
O	0.051

CONCRETE [LOS ALAMOS (MCNP) Mix] - Density = 2.25 g/cm^3

Nuclide	Weight Fraction	Atom Density
H	0.00453	0.006094
O	0.5126	0.043421
Si	0.36036	0.01739
Al	0.03555	0.001786
Na	0.01527	0.0009
Ca	0.05791	0.001958
Fe	0.01378	0.000334

Most vendors use about 35 cm of concrete

Nuclide	Weight Fraction	Atom Density
Fe	0.705280	0.678055
C	0.000216	0.000966
Mn	0.018325	0.017909
Si	0.002510	0.004798
Cr	0.183257	0.189225
Ni	0.081143	0.074226
P	0.000325	0.000563
S	0.000010	0.000017
N	0.008933	0.034241

Neutron Sources at Dry Storage Casks

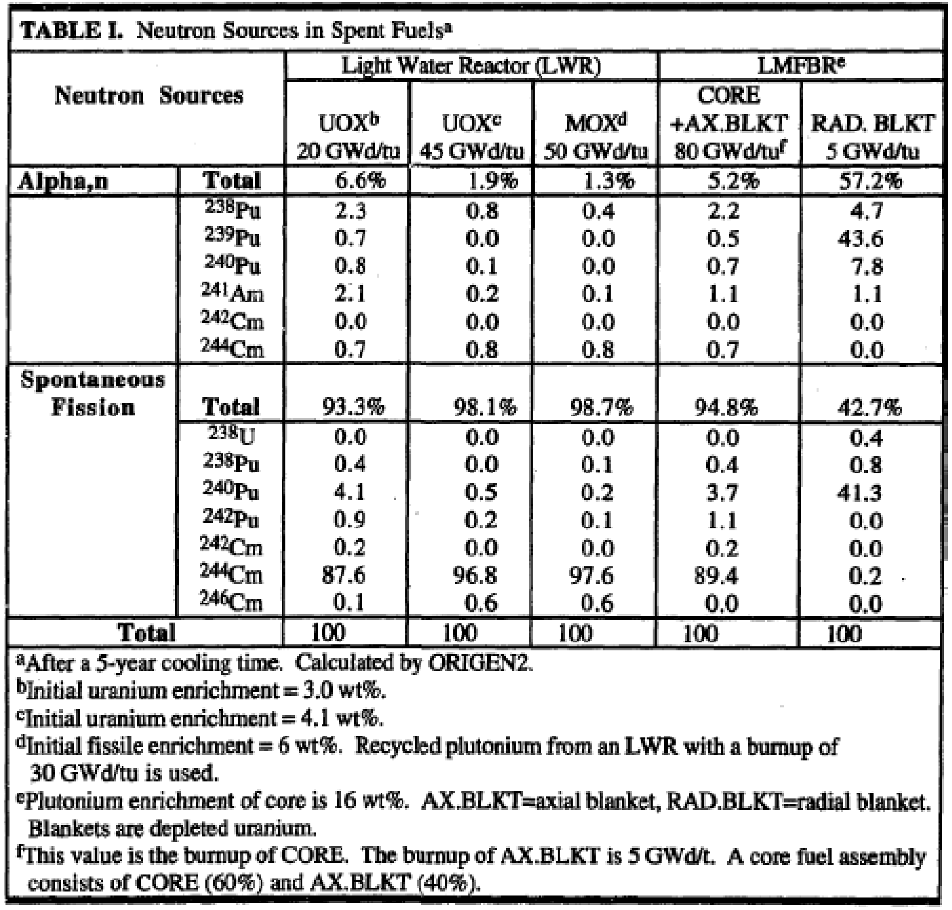


TABLE I. Neutron Sources in Spent Fuels[a]

Neutron Sources		Light Water Reactor (LWR)			LMFBR[e]	
		UOX[b] 20 GWd/tu	UOX[c] 45 GWd/tu	MOX[d] 50 GWd/tu	CORE +AX.BLKT 80 GWd/tu[f]	RAD. BLKT 5 GWd/tu
Alpha,n	**Total**	6.6%	1.9%	1.3%	5.2%	57.2%
	^{238}Pu	2.3	0.8	0.4	2.2	4.7
	^{239}Pu	0.7	0.0	0.0	0.5	43.6
	^{240}Pu	0.8	0.1	0.0	0.7	7.8
	^{241}Am	2.1	0.2	0.1	1.1	1.1
	^{242}Cm	0.0	0.0	0.0	0.0	0.0
	^{244}Cm	0.7	0.8	0.8	0.7	0.0
Spontaneous Fission	**Total**	93.3%	98.1%	98.7%	94.8%	42.7%
	^{238}U	0.0	0.0	0.0	0.0	0.4
	^{238}Pu	0.4	0.0	0.1	0.4	0.8
	^{240}Pu	4.1	0.5	0.2	3.7	41.3
	^{242}Pu	0.9	0.2	0.1	1.1	0.0
	^{242}Cm	0.2	0.0	0.0	0.2	0.0
	^{244}Cm	87.6	96.8	97.6	89.4	0.2
	^{246}Cm	0.1	0.6	0.6	0.0	0.0
Total		100	100	100	100	100

[a] After a 5-year cooling time. Calculated by ORIGEN2.

[b] Initial uranium enrichment = 3.0 wt%.

[c] Initial uranium enrichment = 4.1 wt%.

[d] Initial fissile enrichment = 6 wt%. Recycled plutonium from an LWR with a burnup of 30 GWd/tu is used.

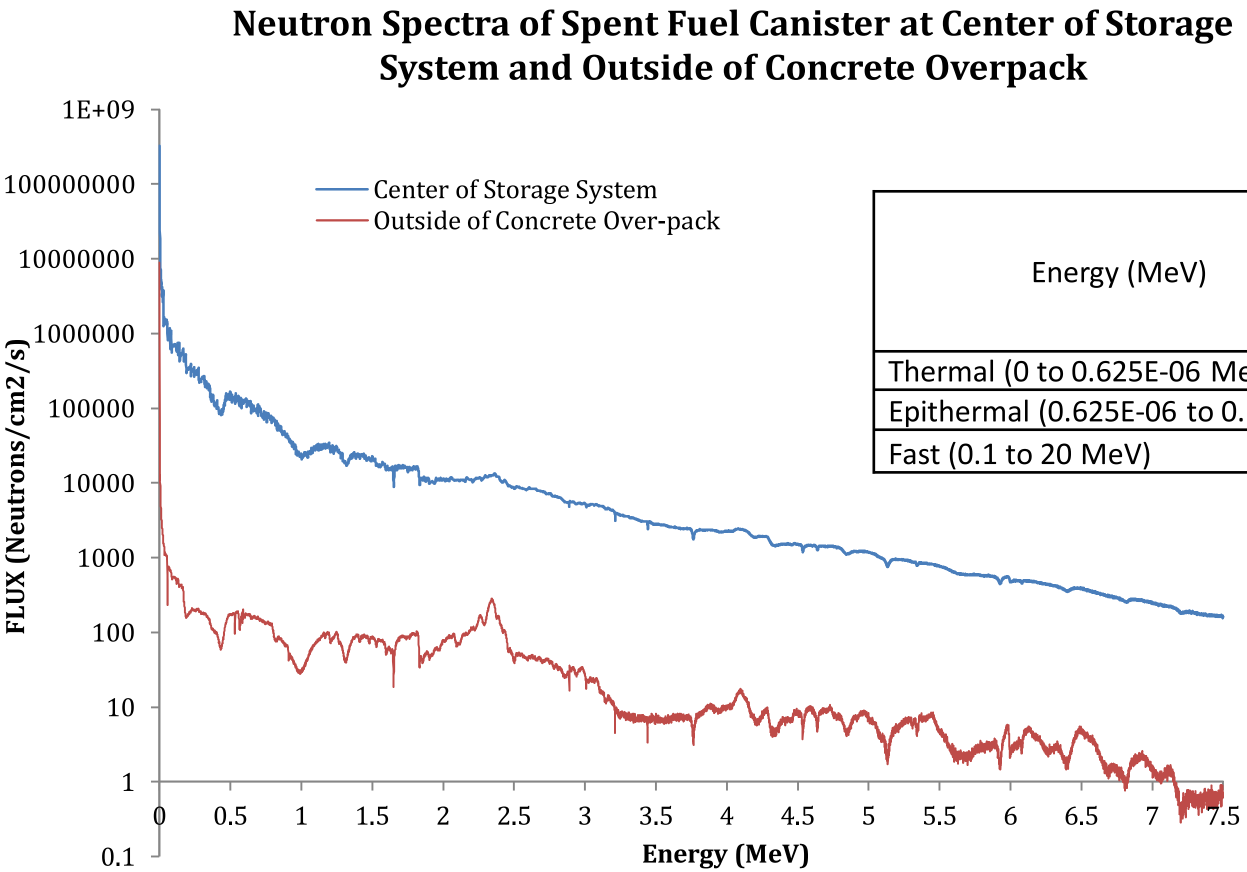
[e] Plutonium enrichment of core is 16 wt%. AX.BLKT=axial blanket, RAD.BLKT=radial blanket. Blankets are depleted uranium.

[f] This value is the burnup of CORE. The burnup of AX.BLKT is 5 GWd/t. A core fuel assembly consists of CORE (60%) and AX.BLKT (40%).

Figure 9: Excerpt for LANL note LA-12774-MS
(http://inis.iaea.org/search/search.aspx?orig_q=RN:25070652)

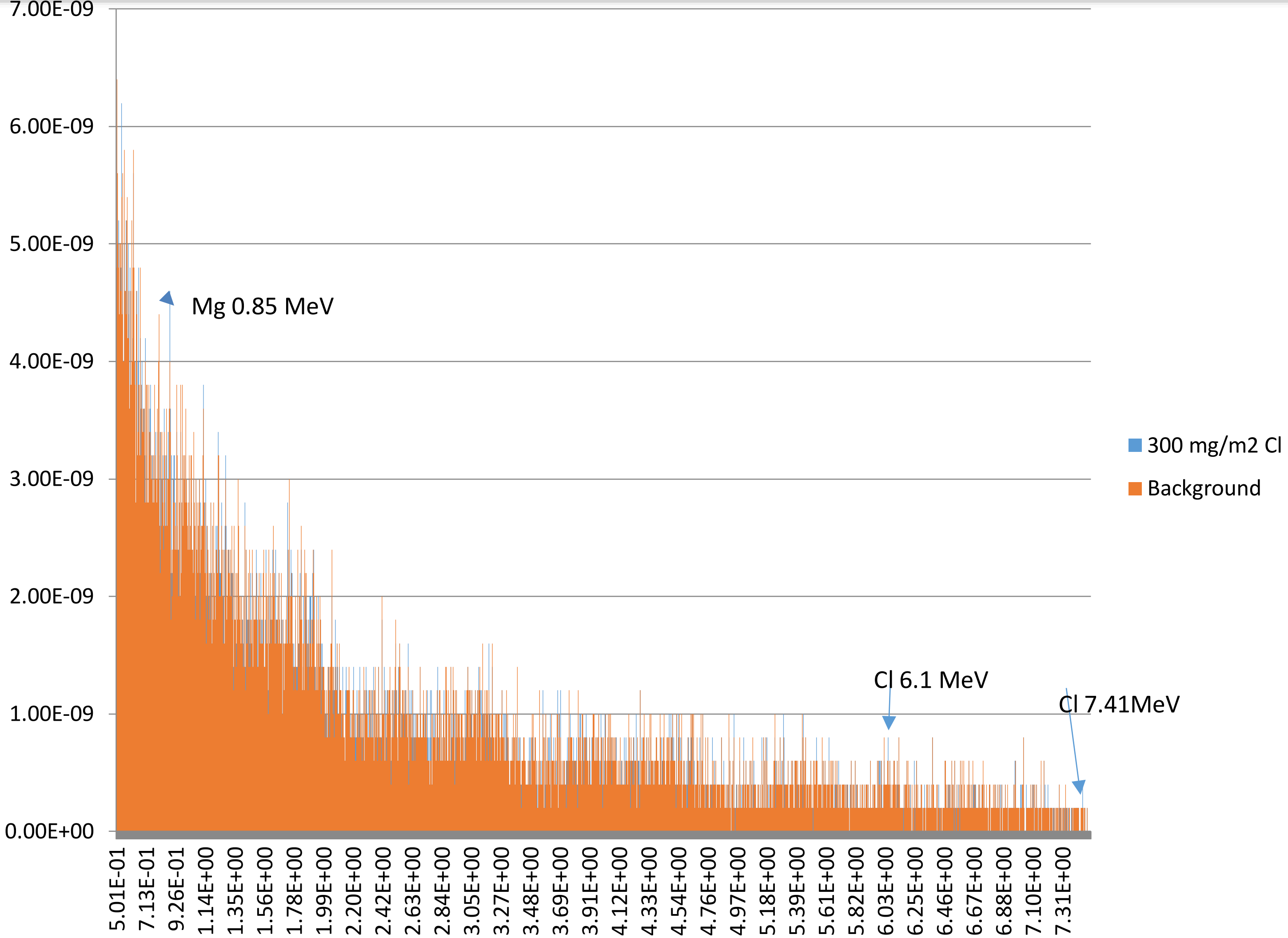
MCNP6 results for a SNC VSC-24 Storage System	
Criticality Eigenvalue	0.377
Q Value (MeV/fission)	198
ν (neutrons/fission)	2.702

Neutron Source Spectrum – MCNP6



Energy (MeV)	Neutrons at Center of Storage System (%)	Neutrons at Outside of Overpack (%)
Thermal (0 to 0.625E-06 MeV)	0.13	0.80
Epithermal (0.625E-06 to 0.1 MeV)	0.58	0.15
Fast (0.1 to 20 MeV)	0.30	0.05

Gamma Spectra at Concrete outer surface of a Typical Westinghouse MC-10 Spent Fuel Cask (SNC VSC-24)



Spent Nuclear Fuel Measurements

PNNL-23561

JE Fast, JW Chenault, BD Glasgow, DC Rodriguez, BA VanDevender, LS Wood

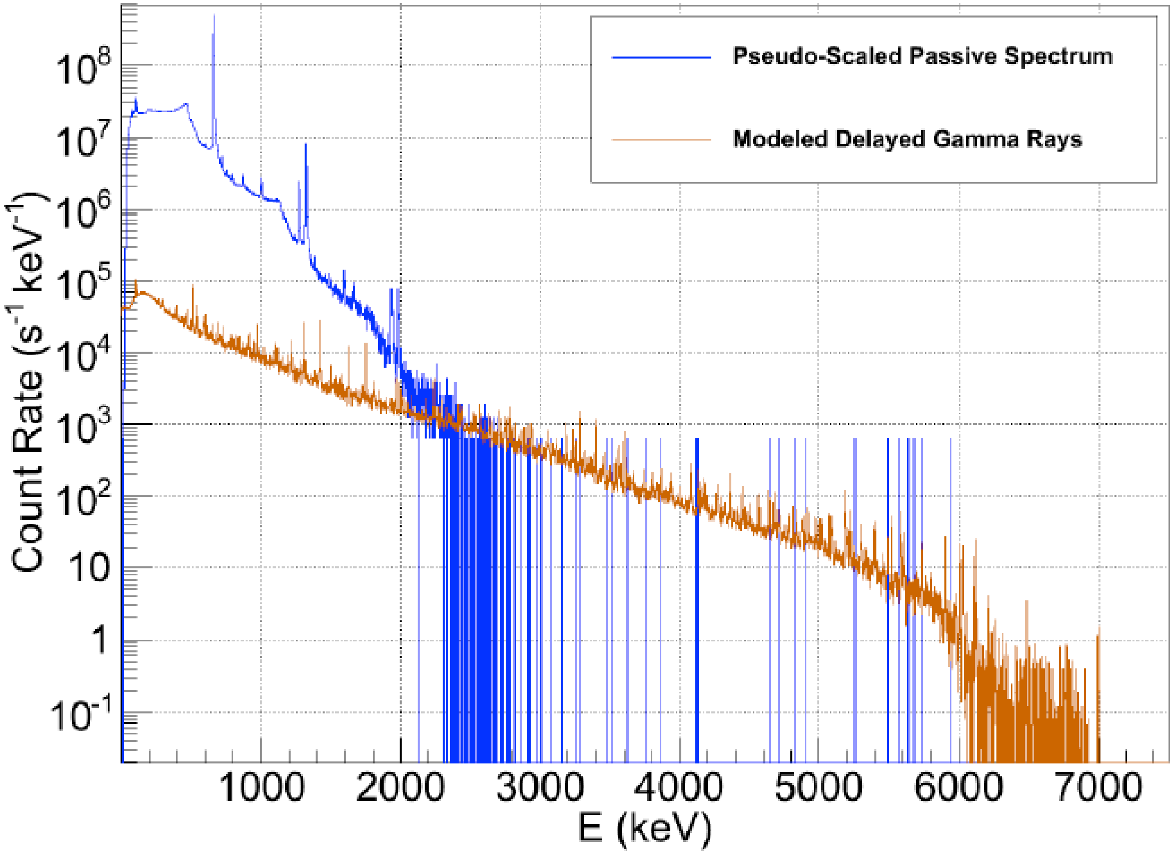
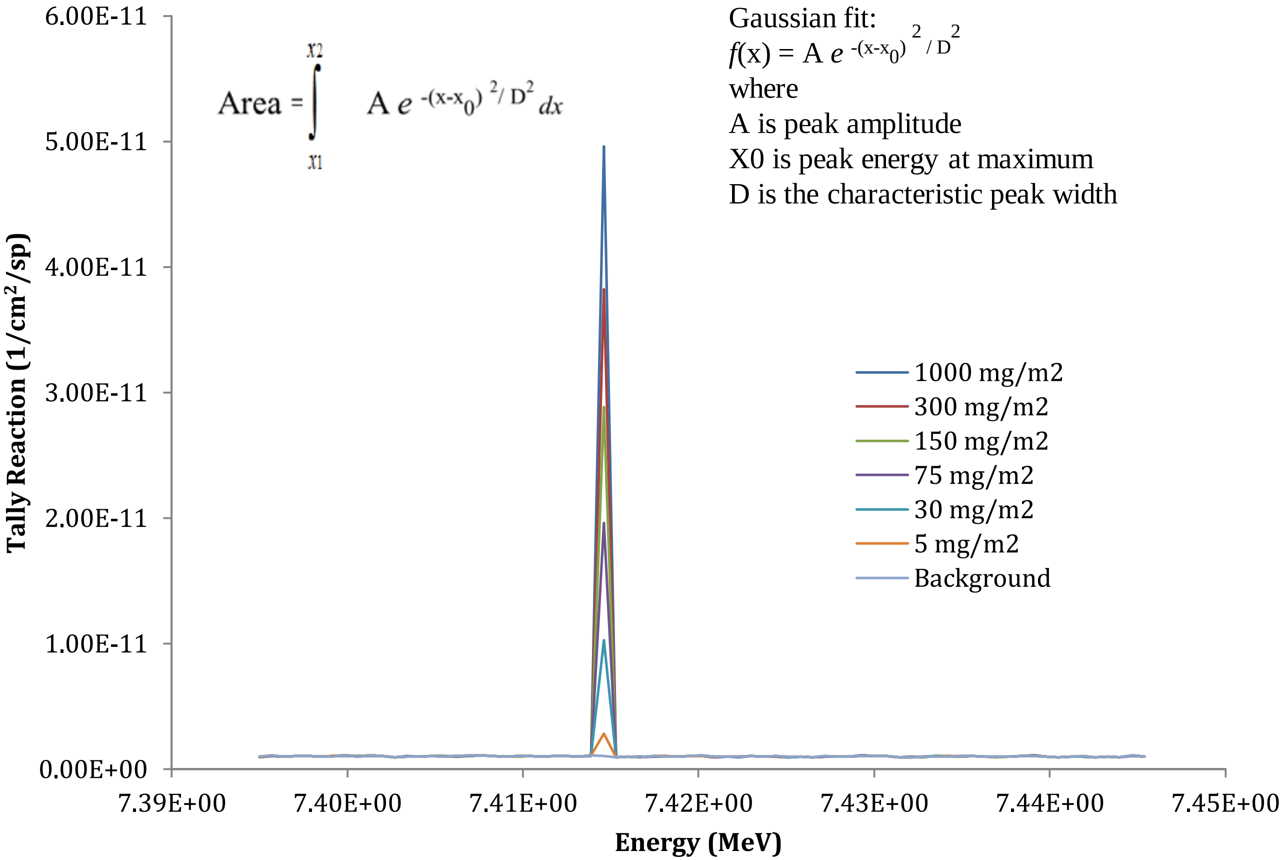


Figure 1: Measured passive background of ~65 GWd/tU burnup, 22.5 year cool down BWR fuel scaled to match equivalent exposure time for simulated delayed gamma assay scenario with 10^{10} n/s (thermal) neutron irradiation with 10 second irradiation/10 sec measurement cycles.

Chloride Detection and Life Prediction of Dry Storage Casks Using PGAA and NAA Techniques Signal from MCNP6 Simulation


7.41 MeV Cl-36 Signal Alongside the Background Signal at Outside Surface of Concrete Overpack

$$\text{Area} = \int_{x_1}^{x_2} A\, e^{-(x-x_0)^2 / D^2}\, dx$$

Gaussian fit:

$f(x) = A\, e^{-(x-x_0)^2 / D^2}$

where

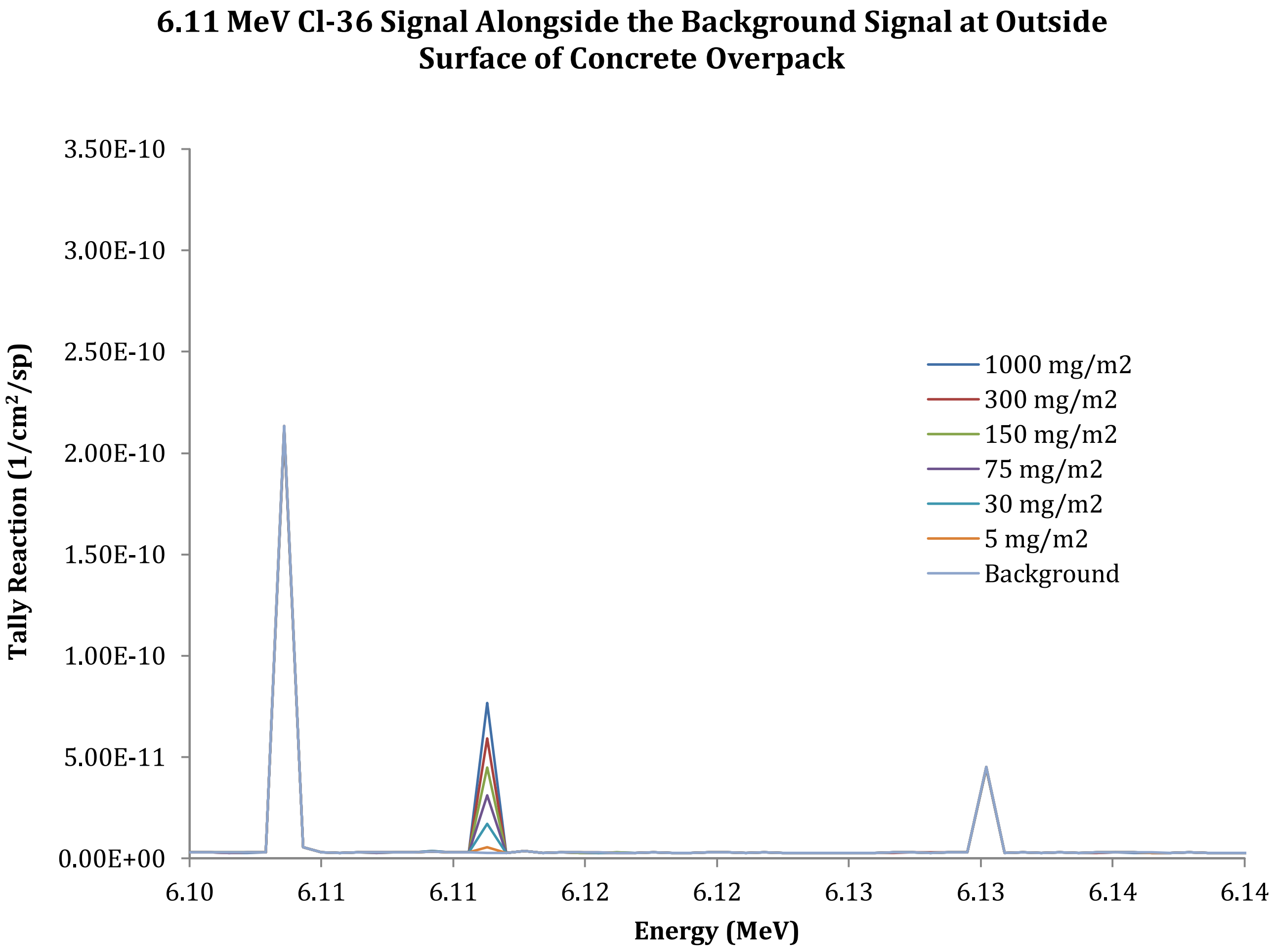
A is peak amplitude

X0 is peak energy at maximum

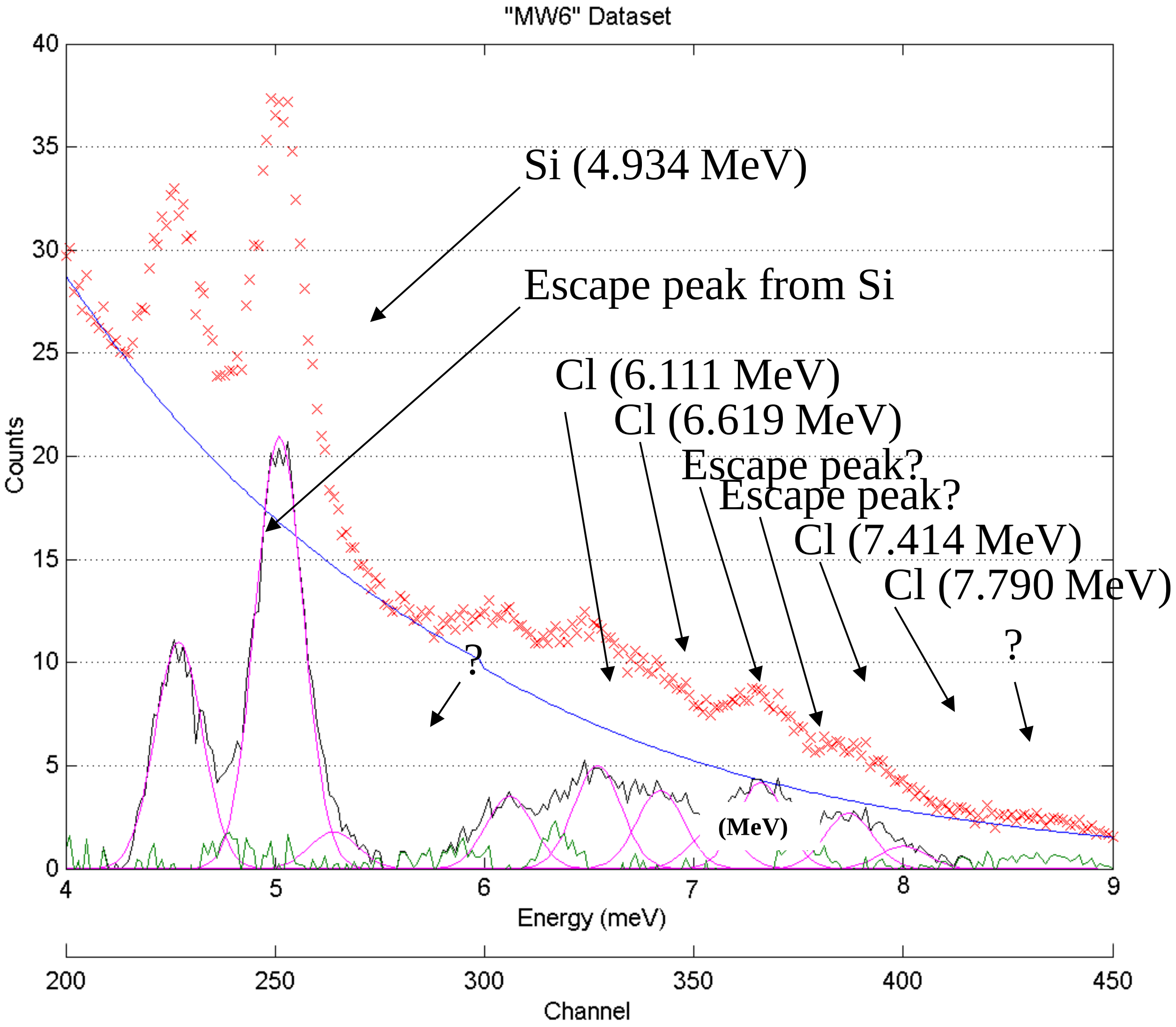
D is the characteristic peak width

Chloride Detection and Life Prediction of Dry Storage Casks Using PGAA and NAA Techniques

Signal from MCNP6 Simulation

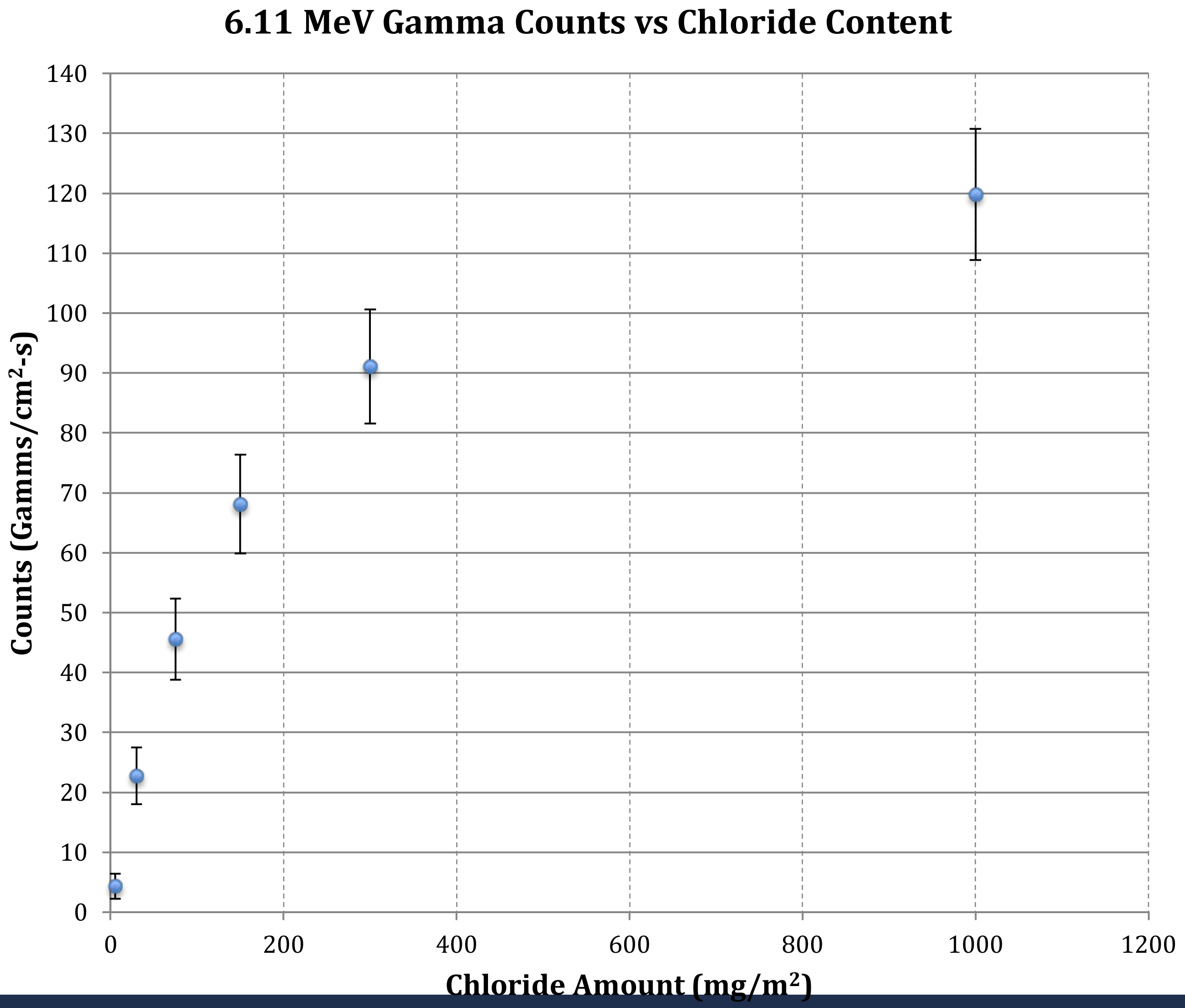


Chlorine Detection in Soil–^{252}Cf Neutron Source Literature Review

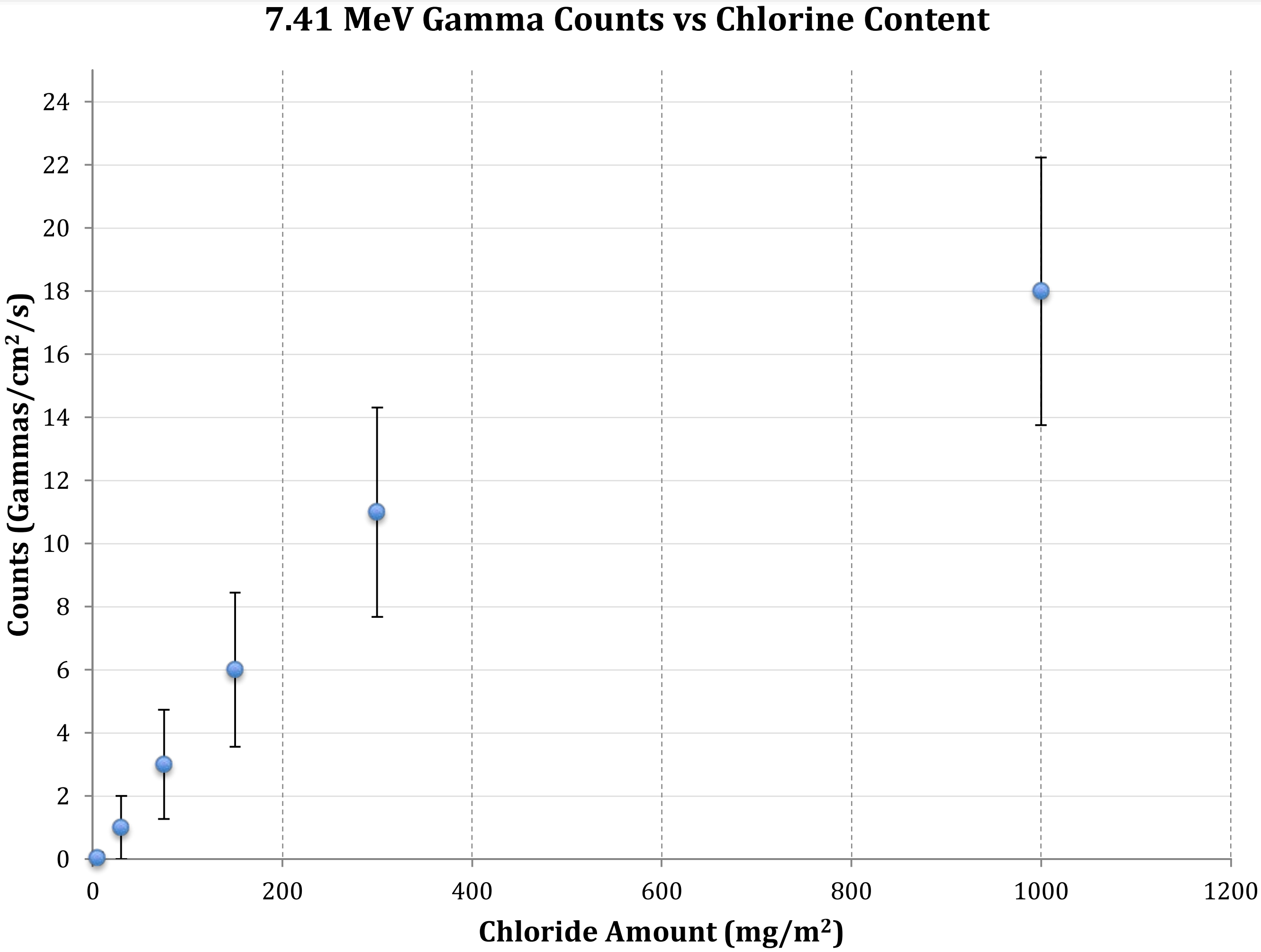


Chloride Detection and Life Prediction of Dry Storage Casks Using PGAA and NAA Techniques

Counts at Outer Concrete Surface — MCNP6

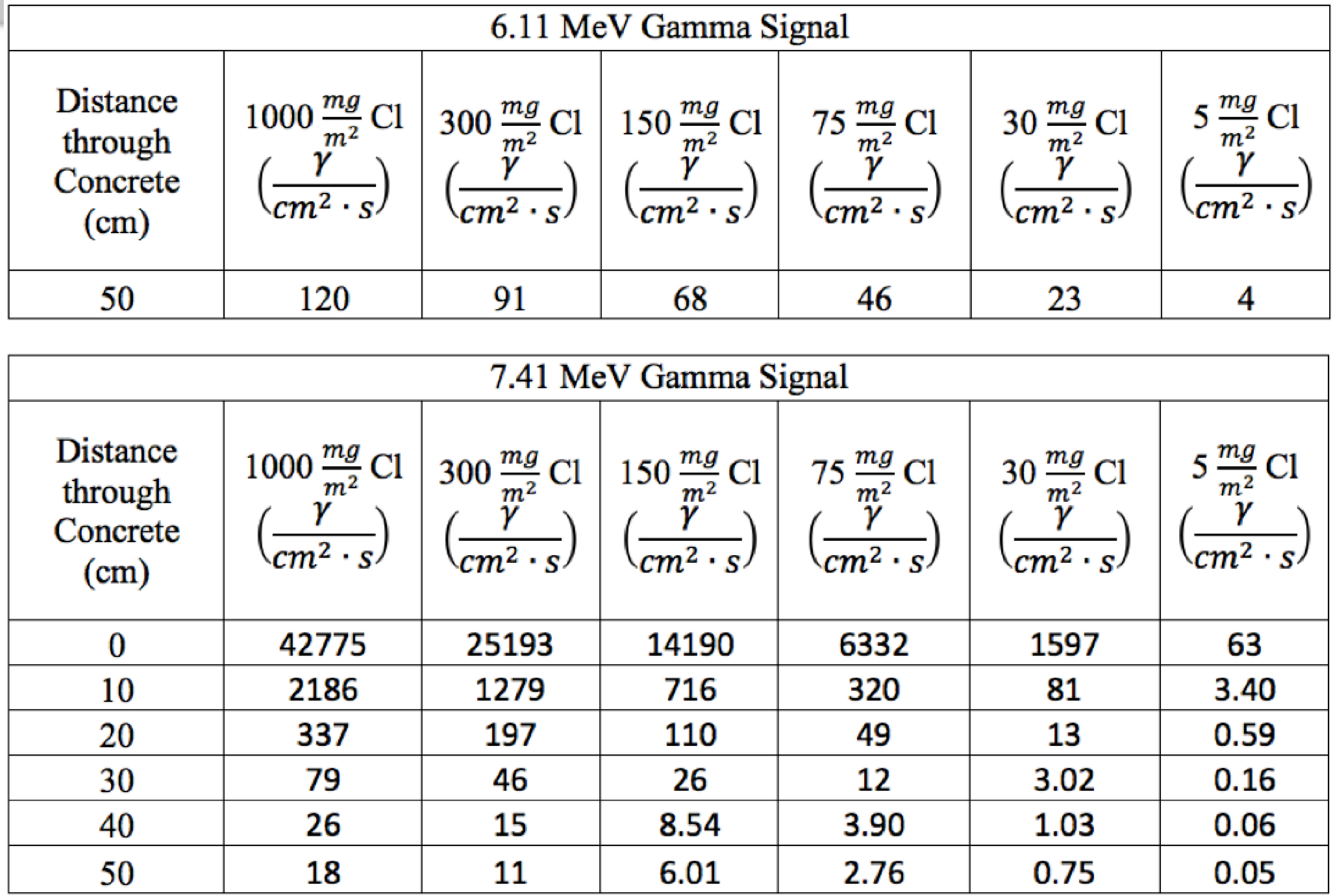


Chloride Detection and Life Prediction of Dry Storage Casks Using PGAA and NAA Techniques Counts at Outer Concrete Surface–MCNP6



Chloride Detection and Life Prediction of Dry Storage Casks Using PGAA and NAA Techniques

Summary

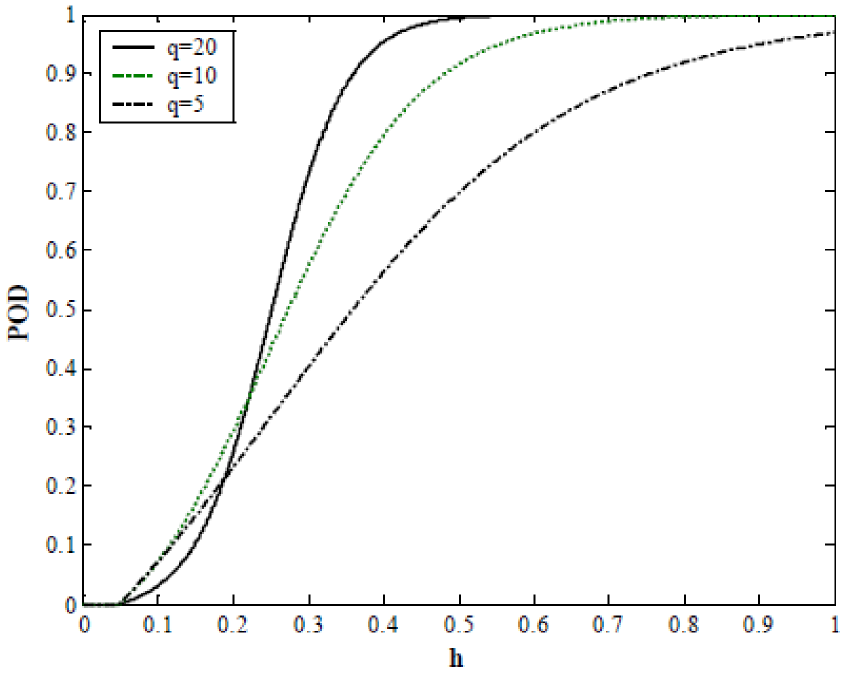


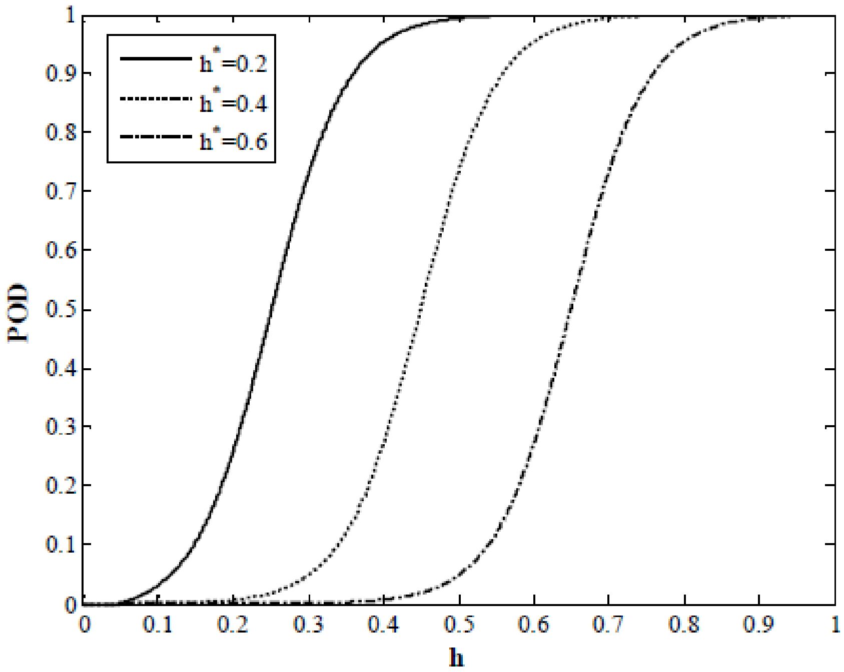
6.11 MeV Gamma Signal						
Distance through Concrete (cm)	$1000\,\frac{mg}{m^2}$ Cl $\left(\frac{\gamma}{cm^2\cdot s}\right)$	$300\,\frac{mg}{m^2}$ Cl $\left(\frac{\gamma}{cm^2\cdot s}\right)$	$150\,\frac{mg}{m^2}$ Cl $\left(\frac{\gamma}{cm^2\cdot s}\right)$	$75\,\frac{mg}{m^2}$ Cl $\left(\frac{\gamma}{cm^2\cdot s}\right)$	$30\,\frac{mg}{m^2}$ Cl $\left(\frac{\gamma}{cm^2\cdot s}\right)$	$5\,\frac{mg}{m^2}$ Cl $\left(\frac{\gamma}{cm^2\cdot s}\right)$
50	120	91	68	46	23	4

7.41 MeV Gamma Signal						
Distance through Concrete (cm)	$1000\,\frac{mg}{m^2}$ Cl $\left(\frac{\gamma}{cm^2\cdot s}\right)$	$300\,\frac{mg}{m^2}$ Cl $\left(\frac{\gamma}{cm^2\cdot s}\right)$	$150\,\frac{mg}{m^2}$ Cl $\left(\frac{\gamma}{cm^2\cdot s}\right)$	$75\,\frac{mg}{m^2}$ Cl $\left(\frac{\gamma}{cm^2\cdot s}\right)$	$30\,\frac{mg}{m^2}$ Cl $\left(\frac{\gamma}{cm^2\cdot s}\right)$	$5\,\frac{mg}{m^2}$ Cl $\left(\frac{\gamma}{cm^2\cdot s}\right)$
0	42775	25193	14190	6332	1597	63
10	2186	1279	716	320	81	3.40
20	337	197	110	49	13	0.59
30	79	46	26	12	3.02	0.16
40	26	15	8.54	3.90	1.03	0.06
50	18	11	6.01	2.76	0.75	0.05

Probability of Detection

$$\mathrm{POD}(h)=\begin{cases}1-\dfrac{1+\exp(-qh^{*})}{1+\exp\left[q(h-s-h^{*})\right]}, & \text{if } h>s\\ 0, & \text{otherwise}\end{cases}$$





Summary and Conclusions

- Key environmental components for assessing the canister sustainability is a Chlorine concentration and detection
- The Monte Carlo Simulation demonstrated that it's feasible to detect and quantified relatively small amount Chlorine on the canister surface at the outer surface of concrete overpack (signal from PGAA of ^{35}Cl).
- Literature review and other related experiments supported our preliminary simulation results and conclusions.

Future Work

- Detectors selection and array around the concrete overpack
- Feasibility study in edifying of other existing elements (i.e.Na,Mg,H,O)
- Residence time of Chlorine on the canister surface (Peak Magnitude and $^{37}Cl(n,\gamma)^{38}Cl$)
- Experiment Design and Performed

Questions

Thank you for your Attention

Zeev Shayer, David Olson, Stephen Liu, Zhenzhen Yu- CSM
Korukonda Murty-NCSU; Djamel Kaoumi -USC; Jonathan Almer and Peter Kenesie-ANL
Charles Bryan and David Enos-SNL
TJ Ulrich and Eric Flynn-LANL
Donald W. Lewis and Jeffery Johns - Westinghouse (former CB&I)

Backup Slides

Gamma Ray Sensors

Sensor material	Detector type	Energy resolution (%)	Density, ρ (g/cm^3)	Fano Factor
Polyvinyl Toluene—PVT	Scintillation	~25*	1.03	(≈1)
Sodium Iodide—NaI(Tl)	Scintillation	5–8	3.67	≈1
Cadmium Zinc Telluride—CZT	Semiconductor	1–10	5.78	<0.2
Lanthanum Bromide—LaBr$_3$	Scintillation	3	5.29	≈1
Germanium—Ge**	Semiconductor	0.25	5.32	0.08

$$FWHM = 2.35k\sqrt{N}$$

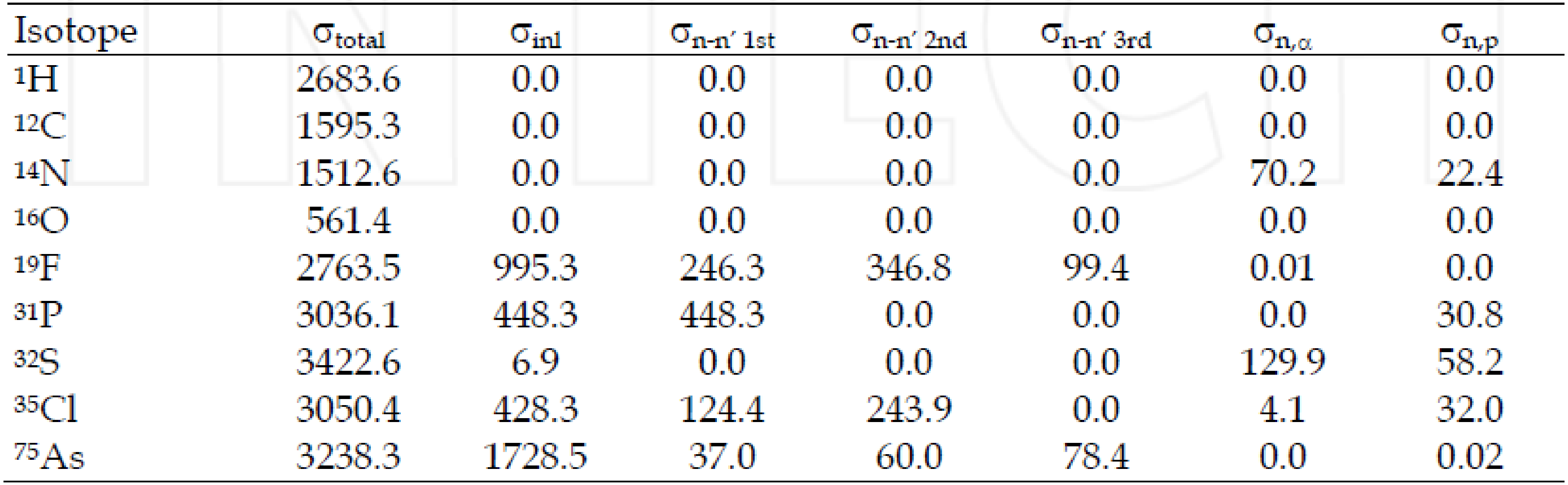
The fractional resolution due to a purely Poisson process is

$$R_{Poisson} \equiv \frac{FWHM}{H_0} = \frac{2.35k\sqrt{N}}{kN} = \frac{2.35}{\sqrt{N}}$$

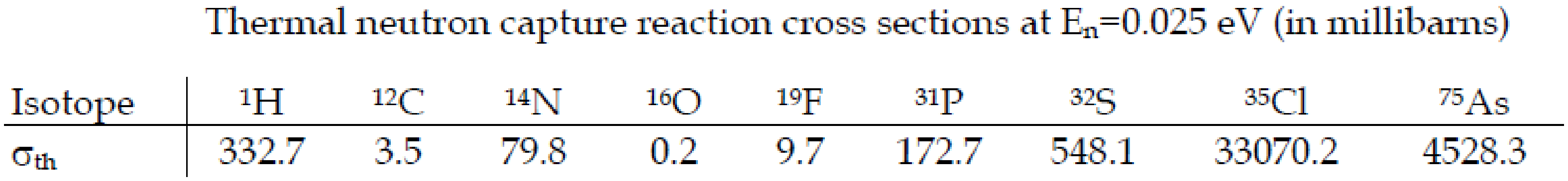
$$F \equiv \frac{\text{observed variance in } N}{\text{Poisson predicted variance in } N}$$

$$R_{Statistical} = 2.35\sqrt{\frac{F}{N}}$$

2.45-MeV neutron induced nuclear reaction cross sections (in millibarns)



Isotope	σ_{total}	σ_{inl}	$\sigma_{n\text{-}n'\ 1st}$	$\sigma_{n\text{-}n'\ 2nd}$	$\sigma_{n\text{-}n'\ 3rd}$	$\sigma_{n,\alpha}$	$\sigma_{n,p}$
^{1}H	2683.6	0.0	0.0	0.0	0.0	0.0	0.0
^{12}C	1595.3	0.0	0.0	0.0	0.0	0.0	0.0
^{14}N	1512.6	0.0	0.0	0.0	0.0	70.2	22.4
^{16}O	561.4	0.0	0.0	0.0	0.0	0.0	0.0
^{19}F	2763.5	995.3	246.3	346.8	99.4	0.01	0.0
^{31}P	3036.1	448.3	448.3	0.0	0.0	0.0	30.8
^{32}S	3422.6	6.9	0.0	0.0	0.0	129.9	58.2
^{35}Cl	3050.4	428.3	124.4	243.9	0.0	4.1	32.0
^{75}As	3238.3	1728.5	37.0	60.0	78.4	0.0	0.02



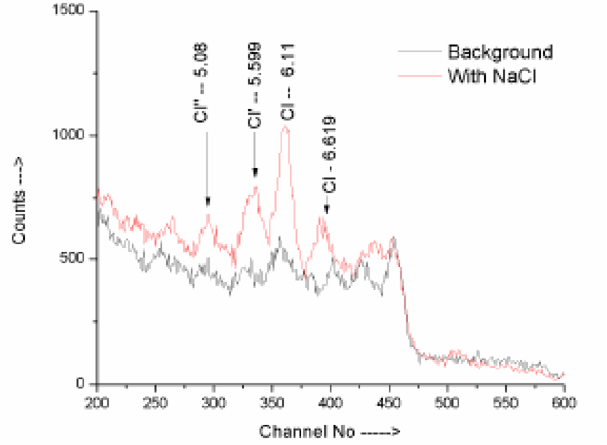
Thermal neutron capture reaction cross sections at E_n=0.025 eV (in millibarns)

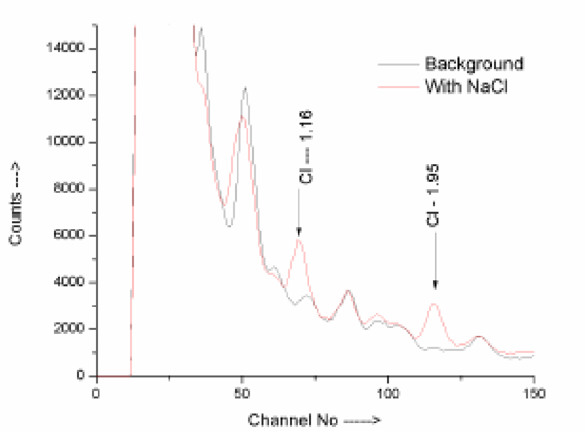
Isotope	^{1}H	^{12}C	^{14}N	^{16}O	^{19}F	^{31}P	^{32}S	^{35}Cl	^{75}As
σ_{th}	332.7	3.5	79.8	0.2	9.7	172.7	548.1	33070.2	4528.3

Investigation of Neutron Based Techniques for the detection of Illicit Materials and explosives

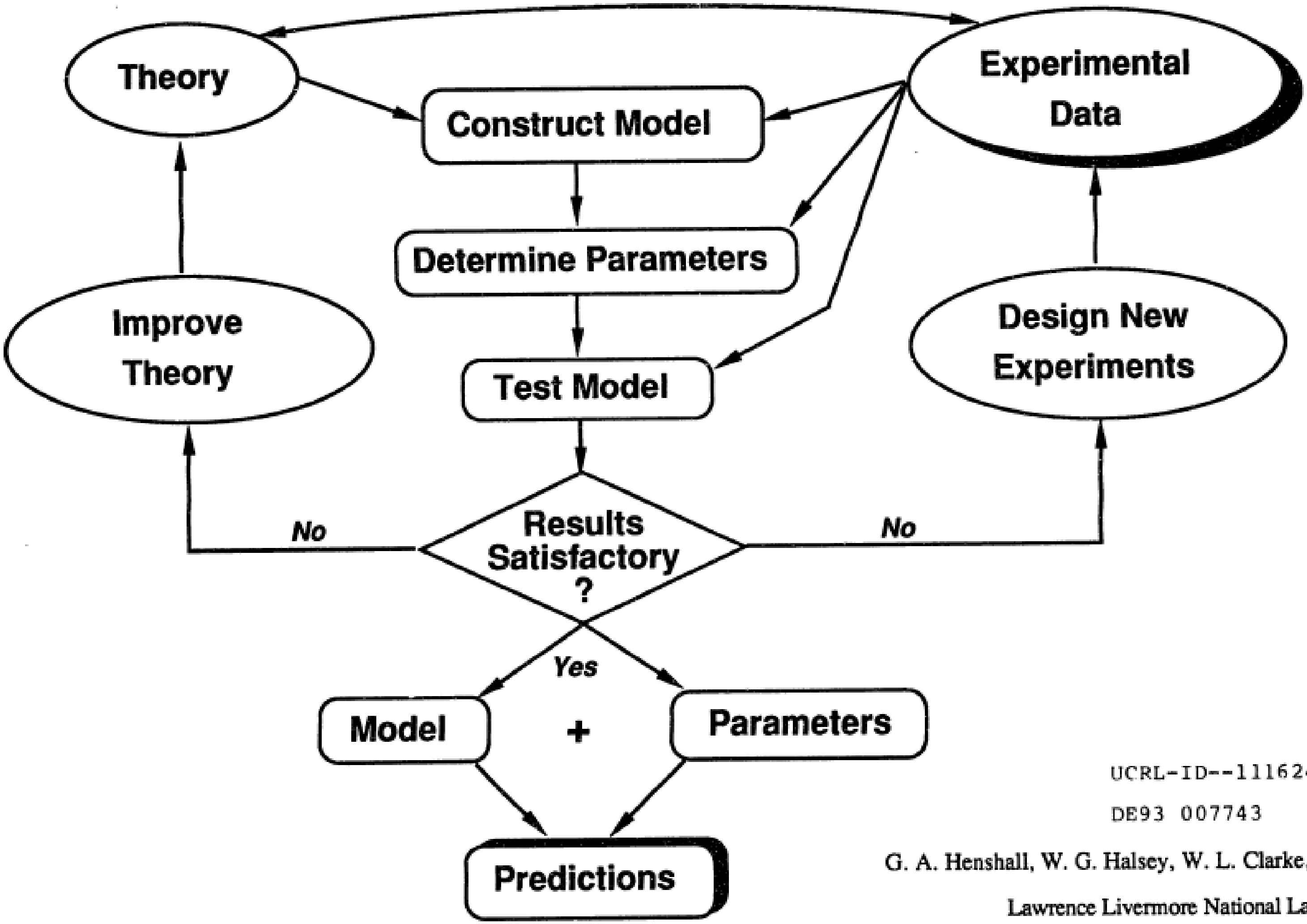
Laser and Neutron Physics Section
Bhabha Atomic Research Centre
Trombay, Bombay, INDIA

The main aim to carry out the detection chlorine (Cl) capture lines in salt is due to fact that chlorine based compounds form part of narcotics. Since Cl has more neutron capture cross-section (43b) than Na the capture gammas of Cl are much more detectable than Na. In a data collection time of 1200 seconds we could detect 4 Chlorine photo-peaks and one of their escape peaks. Figure 20 clearly shows the lines with respect to the background..





Interaction Between Modeling and Experiments



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