Neutron Activation Analysis and Dosimetry

based on reports by R. Burrows, A. Mohagheghi, and D. Ward:
Personnel Nuclear Accident Dosimetry at Sandia National Laboratories
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Presentation Outline

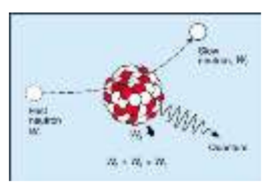
- **Review of Basic Concepts**
  - **neutron discovery**
  - **interaction modes**
  - **activation analysis**
  - **neutron dosimetry**
- **Sandia Personnel Neutron Dosimeter**
- **Nuclear Accident Dosimeter Intercomparison Exercises**

Review: Neutron Discovery

- 1920: *Rutherford* conceived the possible existence of the neutron
- 1920s: Model of the atomic nucleus as composed of protons and electrons – development of quantum mechanics
- 1930: Ambartsumian and Ivanenko found that the nucleus cannot consist of protons and electrons – a neutral particle must be present
- 1931: *Bothe* and Becker found that energetic alpha particles fell on certain light elements, an unusually penetrating radiation was produced – gamma?
- 1932: Curie and Joliot showed that if this unknown radiation fell on paraffin, it ejected protons of very high energy
- 1932: Chadwick showed that the gamma ray hypothesis was untenable and suggested that the new radiation consisted of uncharged particles of approximately the mass of the proton

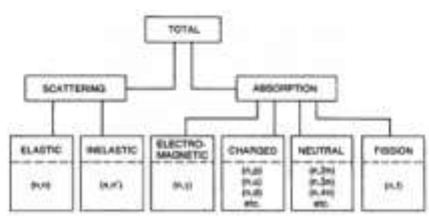
Source: http://en.wikipedia.org/wiki/Neutron

Review: Interaction Modes



- Elastic and Inelastic Scattering - most common for fast neutrons
- Radiative capture with gamma emission (neutron activation) - most common for slow neutrons.
  - Example: Co-59 + n → Co-60 + γ
- Capture with charged particle emissions: (n,p), (n,d), (n, α), …
- Fission

Energy	Neutron Type
< 0.1 eV	Thermal
0.1 < E<1 eV	Epithermal
1 eV < E < 1 KeV	Resonance
E > 1 KeV	Fast



Review: Activation Analysis

If a sample is placed in a neutron field, radiative capture reactions will occur:

$$A + n \rightarrow \gamma_p + B \xrightarrow{\lambda} C + \gamma$$

The target atoms (A) capture neutrons to form the radioactive species B, which then decay to product C. The rate of transformation, R, is given by:

$$R = N\Phi\sigma$$

Where N is the number of the target atoms,
- ϕ is the neutron flux (neutrons/cm^2-sec),
- σ is the cross section for the reaction (cm^2).

The equation above assumes:
- N is remains constant (i.e. the number of the target atoms transformed is very small compared to the total);
- ϕ remains constant throughout the target.

The net change in the number of transformed atoms, B, is:

$$\frac{dN_B}{dt} = \text{Creation rate} - \text{Loss rate} = N\Phi\sigma - \lambda N_B$$

Therefore:

Review: Activation Analysis - Example

$$\lambda N_B = A_B = N\Phi\sigma\left(1 - e^{-\lambda t}\right)$$

Where A_B is the activity of the sample at the end of the irradiation. The quantity $N\phi\sigma$ is called the saturation activity, because it represents the maximum activity obtainable when the sample is irradiated for a long time compared to the half-life of the product, i.e. $t \gg (\ln 2/\lambda)$.

Example: An unknown sample containing Na-23 is irradiated for 24 hours in a neutron flux of 10^5 n/cm^2-sec. The cross section for $^{23}Na(n,\gamma)^{24}Na$ is 0.167 barns. The activity of the sample is measured at the end of the irradiation and the ^{24}Na activity is found to be 293 Bq. How much Na is in the sample? Assume ^{23}Na is the only Na isotope.

$$N = \frac{A}{\Phi\sigma(1 - e^{-\lambda t})} = \frac{293/\text{sec}}{10^5 \frac{n}{cm^2.sec} X 0.167 X 10^{-24} \frac{cm^2}{n}\left(1 - e^{-\frac{0.693 X 24}{15}}\right)}$$

N=2.619X10^{22} atoms

M= 2.619X10^{22} atoms X (23 gm/mole) X (1mole/6.02x10^{23} atoms)

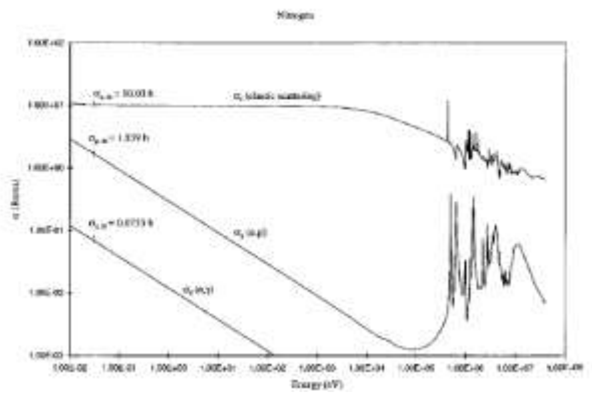
M $\simeq$ 1.00 gm.

Review: Neutron Dosimetry

<u>**Dose is proportional to energy deposited in the body by radiation**</u>

Target atoms: H, O, C, N, Na, Cl

Interaction type and probability depend on neutron energy:



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  - **interaction modes**
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- **Sandia Personnel Neutron Dosimeter (PNAD)**
  - **Objectives**
- **Nuclear Accident Dosimeter Intercomparison Exercises**

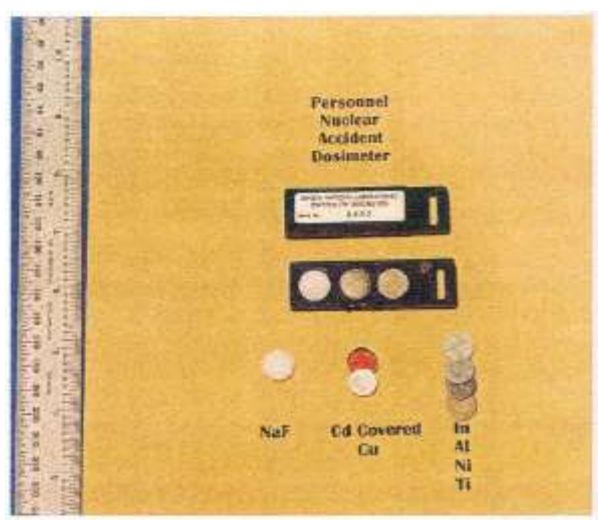
Determine Neutron Spectrum

Table 2. Dosimetry Reactions

Material	Reaction	Half-Life	E_γ (keV)	Gamma Yield (%)	Threshold (MeV)
Al	$^{27}Al(n,\alpha)^{24}Na$	15 h	1368.633	100.0	8
Ti	$^{47}Ti(n,p)^{47}Sc$	3.4 d	159.381	67.9	2
Ni	$^{58}Ni(n,p)^{58}Co$	70.9 d	810.775	99.4	3
Cu	$^{63}Cu(n,\gamma)^{64}Cu$	12.9 h	1345.77	0.47	Epithermal
In	$^{115}In(n,n')^{115m}In$	4.36 h	336.241	45.9	1
	$^{115}In(n,\gamma)^{116m}In$	54.4 m	1293.54	84.4	Thermal[b]
Au[a]	$^{197}Au(n,\gamma)^{198}Au$	64.8 h	411.80	95.5	Thermal
Na	$^{23}Na(n,\gamma)^{24}Na$	15 h	1368.633	100.0	Epithermal

[a] Gold foils used only in FNADs
[b] Not used for dosimetry purposes, but used for Quick Scan purposes, see Appendix B.

Layout of the SNL PNAD



PNAD Elements Data

Table 1. Data Regarding PNAD Elements[a]

PNAD Element	Diameter (inches)	Thickness (inches)	Approximate Wt. (g) $\pm 1\sigma$	Purity (%)
NaF pellet[b] Cadmium[c] covered	0.500	0.100	0.406 ± 0.007	50% NaF by weight[e]
Copper foil	0.406	0.032	$0.610^{d} \pm 0.003$	99.9
Titanium foil	0.500	0.036	0.525 ± 0.004	99.9
Nickel foil	0.500	0.032	0.835 ± 0.006	99.9
Aluminum foil	0.500	0.032	0.271 ± 0.001	99.9
Indium foil	0.500	0.010	0.228 ± 0.003	99.9

Determine Neutron Fluence

Table 3. Neutron Fluence Conversion Equations For PNAD

Reaction	E_γ used for Analysis	Fluence Conversion Equations[a]
$^{115}In(n,n')^{115m}In$	336 keV	$\Phi = \frac{A}{5.985x10^{-12}\sigma}$
$^{47}Ti(n,p)^{47}Sc$	159 keV	$\Phi = \frac{A}{6.076x10^{-14}\sigma}$
$^{58}Ni(n,p)^{58}Co$	810 keV)	$\Phi = \frac{A}{2.167x10^{-14}\sigma}$
$^{27}Al(n,\alpha)^{24}Na$	1369 keV	$\Phi = \frac{A}{7.707x10^{-12}\sigma}$
$^{63}Cu(n,\gamma)^{64}Cu$	1346 keV	$\Phi = \frac{A}{2.661x10^{-12}\sigma}$
$^{197}Au(n,\gamma)^{198}Au$	411 keV)	$\Phi_T = \frac{A}{2.454x10^{-11}}$
$^{23}Na(n,\gamma)^{24}Na$	1369 keV	$\Phi = \frac{A}{4.986x10^{-12}\sigma}$

Absorbed Dose

Neutron Dose = Neutron Fluence * Conversion Factor

Table A- 1. Dosimetry Data for PNAD Materials and Dose Conversion Factors for Selected Neutron Spectra

Spectrum-Averaged Cross Section (barns)	Spectrum Type #1 WATT	#2 GODIVA (Taken from Ing and Makra, 1990)	#3 1/E + WATT	#4 ZPR-6 4Z+ Thermal	#5 FISSION through 10 cm Carbon	#6 FISSION through 90 cm Water	#7 20 cm Li-H Slab	#8 Graphite Test Lattice	#9 FERMI Reactor Test
$^{63}Cu(n,\gamma)^{64}Cu$	0.0107	0.0130	0.194	0.112	0.734	0.127	0.0955	0.184	0.231
$^{115}In(n,n')^{115m}In$	0.183	0.129	0.0412	0.0420	0.0903	0.138	0.185	0.00571	0.000436
$^{58}Ni(n,p)^{58}Co$	0.104	0.0649	0.0230	0.0233	0.0417	0.160	0.155	0.00231	0.000141
$^{47}Ti(n,p)^{47}Sc$	0.0176	0.0113	0.00391	0.00400	0.00739	0.0257	0.0253	0.000419	0.0000262
$^{27}Al(n,\alpha)^{24}Na$	0.000686	0.000468	0.000142	0.000168	0.000209	0.00509	0.00302	0.000010	0.00000140
$^{23}Na(n,\gamma)^{24}Na$	0.000278	0.000338	0.617	0.0239	0.0138	0.0324	0.0267	0.233	0.00780
$^{197}Au(n,\gamma)^{198}Au$	0.0774	0.103	173	29.3	19.6	46.6	1.44	82.7	11.8
$\overline{K}$ $\frac{mrad}{n/cm^2}$	2.81	2.42	0.652	0.985	1.67	2.03	2.22	0.141	0.255
$\overline{D}$ $\frac{mrad}{n/cm^2}$	3.30	2.77	0.762	1.05	1.90	2.43	2.66	0.160	0.229
$\overline{DE}$ $\frac{mrem}{n/cm^2}$	31.7	28.7	7.68	11.6	19.8	20.1	22.9	1.91	3.17
$\overline{D_\gamma}$ $\frac{mrad}{n/cm^2}$	0.215	0.237	0.222	0.294	0.263	0.247	0.231	0.118	0.352

Example Calculation: Activity Measurement Using a HPGe Detector

Table D- 1. Measured PNAD Data[a,b]

Reaction	Measured Specific Activity (Bq/g)	Measured Specific Activity (μCi/g)	Uncertainty in Measured Specific Activity (%)[c]
$^{115}In(n,n')^{115m}In$	1270	0.0343	21.4
$^{47}Ti(n,p)^{47}Sc$	1.19	0.0000322	29.3
$^{58}Ni(n,p)^{58}Co$	1.81	0.0000489	18.0
$^{27}Al(n,\alpha)^{24}Na$	10.74	0.00290	24.0
$^{63}Cu(n,\gamma)^{64}Cu$	3340	0.0903	21.6

[a] Data obtained from run #2, SHEBA High Power, NAD23
[b] Reference dose was 108 rads (to tissue, from neutrons), uncertainty not reported
[c] Uncertainty is 2σ

Example Calculation: Neutron Dose

From table A-1, Spectrum #16 was selected as the best estimate of the incident neutron spectrum:

- Spectrum-weighted cross-section for In: 0.12 barns
- Fluence to dose conversion factor: 2.29 nrad/(n/cm^2)

$$\Phi = \frac{0.0343}{5.985x10^{-12}\,x(0.120)} = 4.78x10^{10}\,n/cm^2$$

$$Dose = (4.78x10^{10}\,n/cm^2)(2.29\frac{nrad}{n/cm^2})x10^{-9}\frac{rad}{nrad}$$
$$= 109\ \text{rad}$$

Reference Dose = 108 rad

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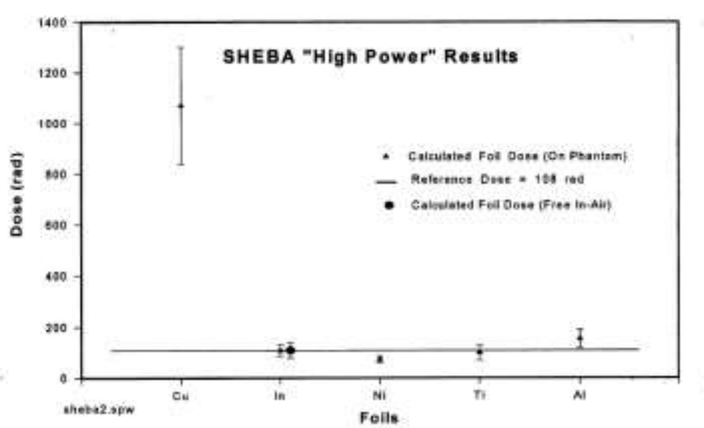
Los Alamos Neutron Accident Dosimetry Exercise 1996
Summary Performance Results

Table 4. Comparison of Measured Dose to Reference

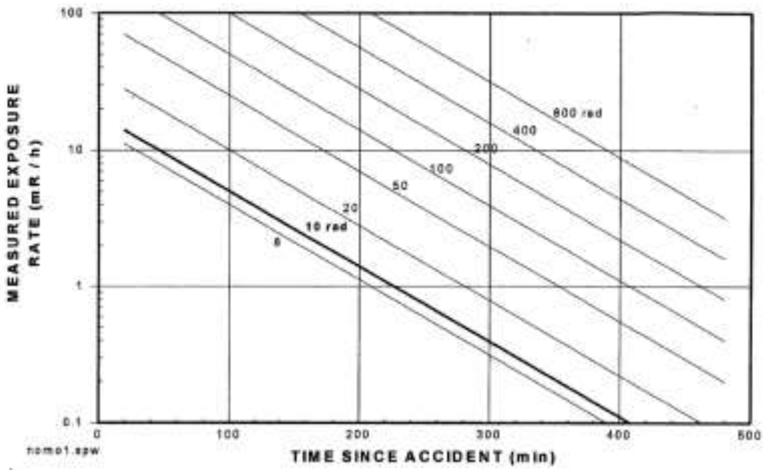
Neutron Source	Reference Neutron Dose (rad)	Reported Neutron Dose (rad)	Bias[a] (%)	Spectrum No. Used (from Table A)
SHEBA high power	108	114	6	16
SHEBA low power	11	12	9	16
SHEBA free-run	94	99	5	16
Bare Godiva	200	272	36	2
Godiva through 12 cm Lucite	26	32	23	27, 28[b]
Godiva through 20 cm concrete	34	32	-6	24
Godiva through 13 cm steel	N/A[c]	48	N/A	

[a] Bias = 100*[1-Reported/Reference)]
[b] Neutron absorbed dose calculated using interpolated values from spectrums #27 and #28.
[c] Reference dose not made available to NAD23 participants.

Los Alamos Neutron Accident Dosimetry Exercise
Detailed Results for a Single Test

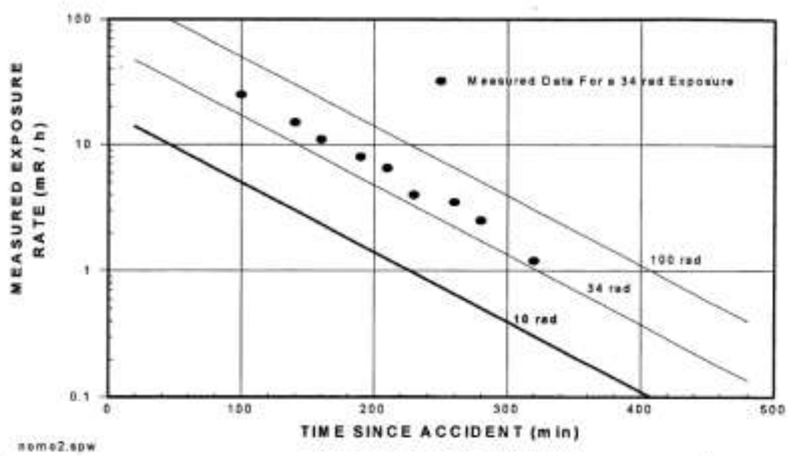


PNAD Quick Scan Procedure



PNAD Quick Scan Procedure: Example Measurement

The PNAD was located on a 20 cm X 20 cm X 10 cm Lucite phantom and was exposed to a Godiva spectrum which passed through 20 cm of concrete.



CALIBAN Neutron Accident Dosimetry Exercise 2010
Example of Performance Results

Table 3: Dose Results - Pulse #1

Distance & Location	PNAD #	Quick-Scan[1] Triage Est. (rad)	SNL: D_n From[2] PNAD Foils (rad)	SNL: D_γ From TLD (rad)	SNL: Total (rad)	CEA Valduc: D_n* (rad)	CEA Valduc: D_γ* (rad)	CEA Valduc: Total* (rad)
4 m, Phantom Front	0137	122	148±56.1	64 ± 10	218±40.8	170	40	210
	0085	120	158±55.8					
4 m, Phantom Side	n/a	n/a		58 ± 14				
	n/a	n/a						
4 m, Phantom Back	0336	92	28.8±27.5	58± 6	95±22.2			
	0143	93	44.4±32.9					
2 m, Free In-Air	0344	68	417±74	72± 14	505±55.8	510	70	580
	0364	70	450±78.6					

[1] See Appendix B for supporting data. Assumed spectrum is a bare Godiva (spectrum #12, Ward et al. [1996])
[2] See Appendix C for supporting data. The average of the D_n values from the PNADs foils were used to calculate the total dose. Stated uncertainty is 2σ.
* Delivered values reported by CEA Valduc.

References

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- Evans, The Atomic Nucleus, McGraw Hill.
- Knoll, Radiation Detection and Measurement, Wiley.
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- Useful Web Sites:
  - National Nuclear Database: http://www.nndc.bnl.gov/index.jsp
  - Radionuclide Decay Data: http://hps.org/publicinformation/radardecaydata.cfm

Thank you for your time

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