Neutron Activation
and
Activation Analysis

General

General

Many nuclear reactions produce radioactive products.

The most common of these reactions involve neutrons:

Neutron + Target Nuclide   →   Activation Product

General

Important Applications/Issues Associated with Neutron Activation

1. Neutron Activation Analysis (NAA)

This is an extraordinarily powerful technique for identifying and quantifying various elements (and nuclides) in a sample.

2. Neutron Fluence Rate (Flux) Measurements

Neutron fluence rates in reactors or other neutron sources can be measured by exposing targets (e.g., metal foils) to the neutrons and measuring the induced activity.

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General

Important Applications/Issues Associated with Neutron Activation

3. Dosimetry Following Criticality Accidents

The induced activity in objects or individuals following a criticality accident can be used to estimate the doses to these individuals.
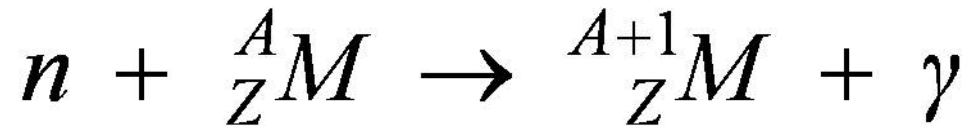
4. Hazards from Induced Activity

Induced radioactivity in the vicinity of intense neutron sources can constitute an exposure hazard. Examples of such sources include reactors, accelerators and, of course, nuclear explosions.

General

Neutron Capture

The most important reaction is neutron capture:

$$n + {}^{A}_{Z}M \rightarrow {}^{A+1}_{Z}M + \gamma$$

Thermal neutrons are most likely to be captured.

The target nuclide is usually, but not necessarily stable.

If the product is radioactive, it is likely a beta emitter.

The gamma ray, referred to as a prompt gamma or capture gamma, is typically of high energy.

General

Neutron Capture Example:

$$n + {}^{50}_{24}Cr \rightarrow {}^{51}_{24}Cr + \gamma$$

This is an exception to the generalization that the activation product is a beta emitter. Cr-51 decays by electron capture!
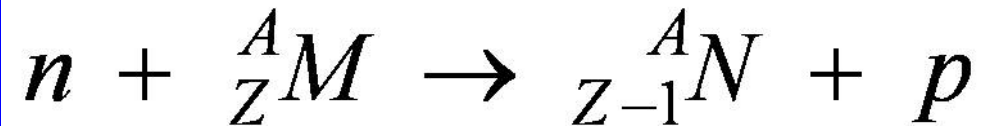
The major prompt gamma rays:

749 keV produced 11.0% of the time
8512.1 keV produced 6.16% of the time
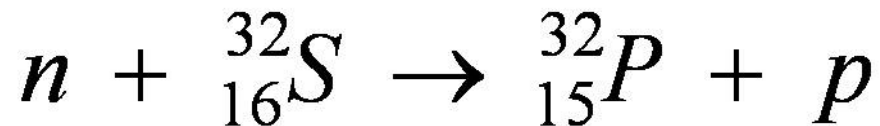8484.0 keV produced 4.54% of the time

General

Neutron-Proton Reaction

Another potentially important reaction is the n-p reaction:

$$n + {}_{Z}^{A}M \rightarrow {}_{Z-1}^{A}N + p$$

The n-p reaction is most likely for fast neutrons and target nuclides with low atomic numbers.

Example:
$$n + {}_{16}^{32}S \rightarrow {}_{15}^{32}P + p$$

Factors Affecting Activation

Factors Affecting Activation

General

The following factors need to be considered:

1. Stability of the reaction product
2. Half-life of the activation product
3. Radiation emitted by the activation product
4. Cross section of the target nuclide
5. Abundance of the target nuclide
6. Neutron fluence rate (and mass of target element)

The following section will consider each of these in turn using the element hydrogen as an example.

Factors Affecting Activation
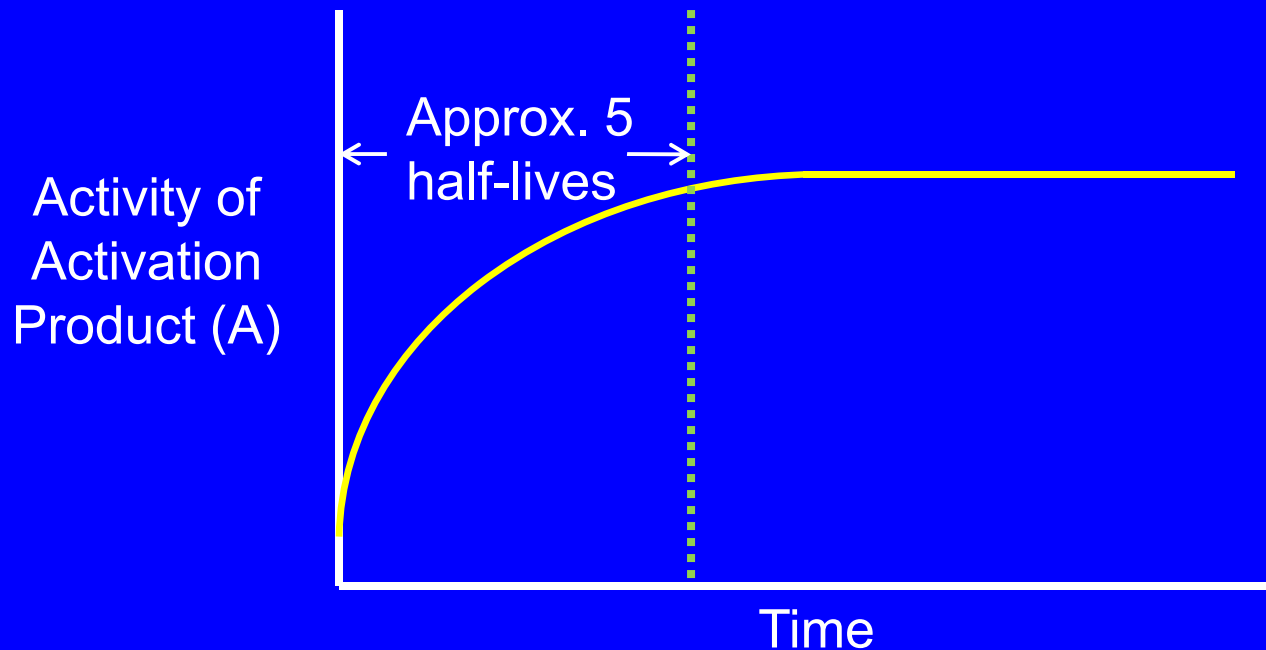## 1. Stability of the Activation Product

If H-1 absorbs a neutron, it simply becomes H-2 which is stable.

However, when H-2 (deuterium) absorbs a neutron, it becomes H-3 (tritium) which is radioactive. This is the reaction of importance. Examples would be the production of tritium at heavy water reactors and tritium in the oil of pumps at accelerators.

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Factors Affecting Activation

2. Half-life of the Activation Product

If the exposure of the target to neutrons is continuous, the activation product activity increases and eventually reaches a maximum equilibrium activity when the rate of production equals the rate of decay. As an approximation, this equilibrium is achieved after the neutron exposure has continued for five half-lives of the activation product.

Activity of Activation Product (A)

Approx. 5 half-lives

Time

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Factors Affecting Activation

2. Half-life of the Activation Product

For situations when the exposure is brief, the shorter the half-life of the activation product, the greater its activity at the end of activation.

The long half-live of tritium (12.3 years) means that short exposures to neutrons will produce almost no activity.

Factors Affecting Activation

3. Radiation Emitted by Activation Product

The radiation emitted by the activation product will affect the hazard associated with its production and the methods by which it can be detected.
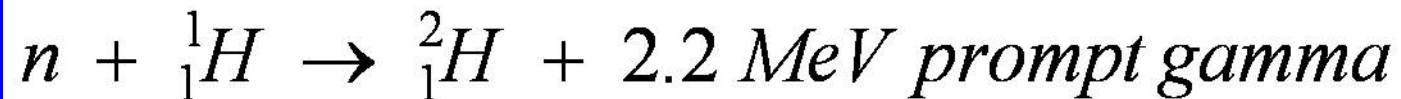

The beta particles emitted by tritium are of such a low energy (18.6 keV max) that they cannot be detected by conventional survey instruments. In addition, there is no gamma ray emitted when tritium decays.

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Factors Affecting Activation
3. Radiation Emitted by Activation Product

The chart of the nuclides does not tell us this, but a 2.2 MeV prompt gamma is produced every time H-1 absorbs a neutron to become deuterium.

$$n + {}_{1}^{1}H \rightarrow {}_{1}^{2}H + 2.2 \; MeV \; prompt \; gamma$$

Prompt gamma rays like this tend to be of high energy and therefore hard to shield.

At the same time, the detection of 2.2 MeV gamma rays is an indication that there is a neutron source nearby.

Factors Affecting Activation
4. Cross Section of the Target Nuclide

The cross section of a reaction reflects the probability of the reaction. The latter depends on the energy of the neutrons and the identity of the target nuclide.

For neutron reactions, the cross section is symbolized with the Greek letter sigma (σ).

The subscript indicates the specific reaction, e.g.,:

σ_γ is the cross section for neutron capture

σ_p is the cross section for the n-p reaction

Factors Affecting Activation
4. Cross Section of the Target Nuclide

Cross sections are expressed in cm^2 or barns.

One barn = 10^{-24} cm^2

In general, one barn is a fairly large cross section.

Unless indicated otherwise, the cross sections on the chart of the nuclides are in barns.

Factors Affecting Activation
4. Cross Section of the Target Nuclide

The capture cross section for H-1 is fairly large (0.33 barns) but that for H-2 is small (0.52 millibarns).

As such, neutron capture by an atom of H-2 to produce tritium has a low probability.

The greater absorption of neutrons by H-1 than H-2 is why heavy water can be considered a superior moderator
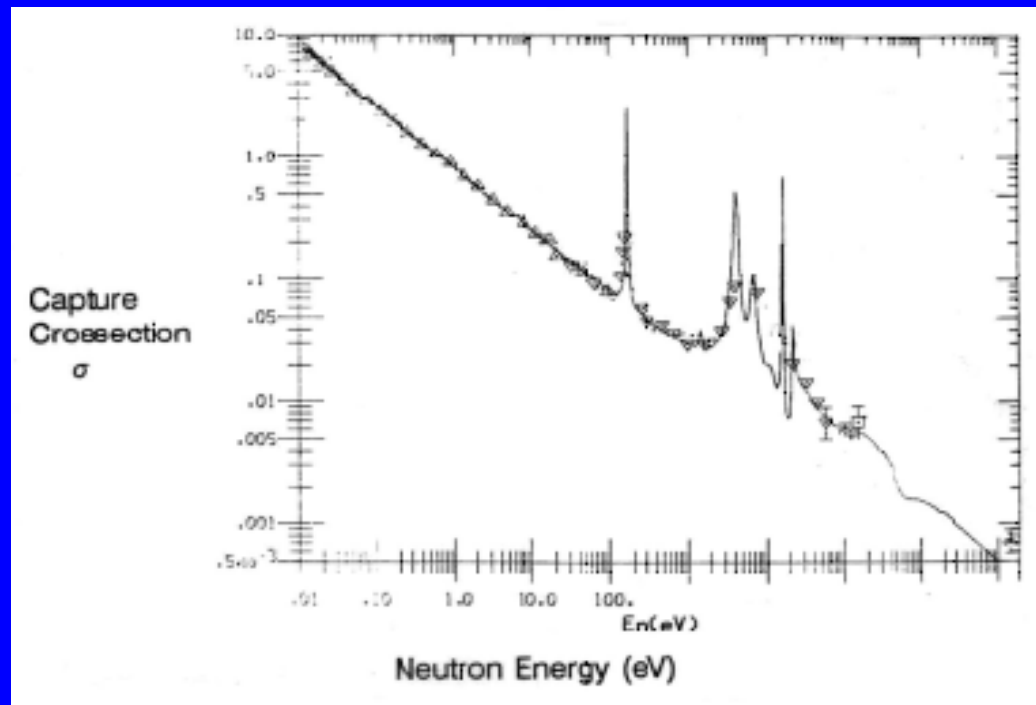
Factors Affecting Activation
4. Cross Section of the Target Nuclide

H1 1/+	H2 1+	H3
99.985	0.015	12.3 a
		β^- .0186 no γ
σ_γ .333, .150	σ_γ .52 mb, .23 mb	$\overline{\sigma}_\gamma < 6\,\mu$b
1.00782503	2.01410178	E .01860

The chart of the nuclides indicates two cross sections for each reaction. The first is the cross section for thermal neutrons (0.333 barns for H-1) – this is the most important cross section. The second (0.150 barns for H-1) is the resonance capture cross section.

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Factors Affecting Activation
4. Cross Section of the Target Nuclide

The above figure shows the neutron capture cross section for vanadium-50 as a function of neutron energy. The spikes at the higher energies are referred to as resonance. This occurs when the neutron energies match up with excitation levels of the target nucleus.

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Factors Affecting Activation
5. Abundance of the Target Nuclide

Towards the top of the box of a stable nuclide, the chart indicates the percent abundance – the percent of the element's mass that consists of the nuclide.

The above shows that only 0.015 percent of natural hydrogen (by mass) is H-2. This means that there is very little H-2 in hydrogen that would serve as a target for tritium production.

Factors Affecting Activation

6. Neutron Fluence Rate and the Mass of the Target Element

This is somewhat self evident, but the greater the neutron fluence rate, the greater the induced activity of the activation product.

In addition, the greater the mass of the target element (and the number of target atoms), the greater the induced activity of the activation product.

Neutron Activation Analysis

Neutron Activation Analysis (NAA)

General

Neutron activation analysis is a powerful technique for identifying and quantifying elements (and nuclides). Its advantages include the fact that NAA is:

- A multi-element technique -- many elements can be analyzed simultaneously.

- Sensitive. Most elements can be analyzed to 1 ppm and many can be analyzed many orders of magnitude below that.

- Fast. The only processing of the sample routinely employed is drying and mixing.

Neutron Activation Analysis (NAA)

General

Its advantages include the fact that NAA:

- Nondestructive. Aside from the production of some short lived activity, the sample is chemically and physically unchanged after the analysis.

- Especially effective with large solid samples (e.g. soil) that are difficult to analyze by other means.

Neutron Activation Analysis (NAA)

How NAA Works

NAA typically works as follows:

1. The sample is exposed to neutrons

2. The activated sample is analyzed by gamma spectroscopy; the activation products are identified and "quantified".

3. The identity and quantity of the target nuclides/elements are deduced.

Neutron Activation Analysis (NAA)

How NAA Works – Example

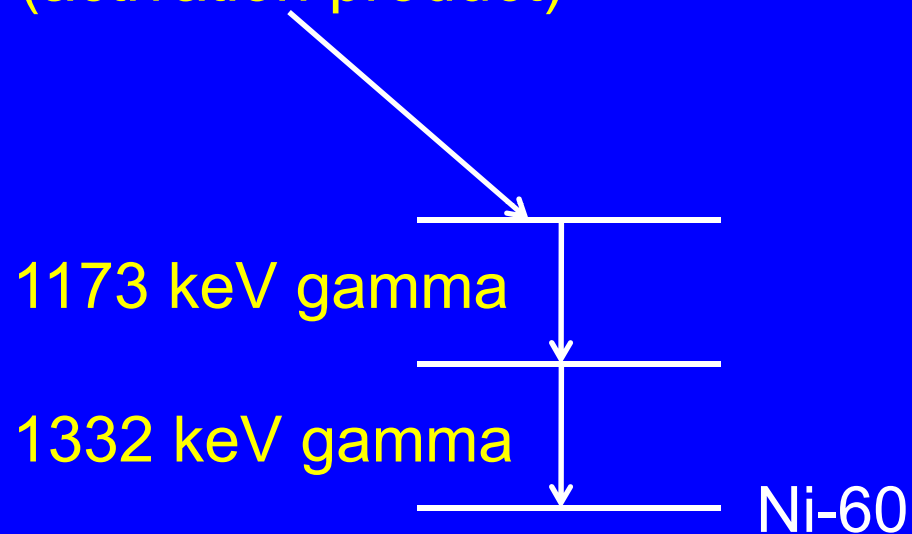Example of using NAA to identify a nuclide (and element) in a sample:

The sample in question is exposed to neutrons for a period of time.

After the irradiation, the activated sample is analyzed by gamma spectroscopy. Two peaks show up on the spectrum: one indicating a gamma ray at 1173 keV and the other a gamma ray at 1332 keV.

What is the target nuclide?

Neutron Activation Analysis (NAA)

How NAA Works – Example

Co-59   +   n   →   Co-60
(stable target)   (activation product)

1173 keV gamma

1332 keV gamma

Ni-60

The target nuclide is Co-59, the only stable nuclide of cobalt!

Neutron Activation Analysis (NAA)

Predicting the Sensitivity of Neutron Activation Analysis (NAA)

Neutron activation analysis does not identify and quantify all nuclides or elements equally well.

The following discussion will consider the sensitivity of neutron activation analysis for two elements: iron and manganese.

In any sample containing iron, all the stable nuclides of iron will be present.

Predicting the Sensitivity of NAA - Iron

Fe54	Fe55 $^{3/-}$	Fe56	Fe57 $^{1/-}$	Fe58	Fe59 $^{3/-}$
5.9	2.73 a	91.72	2.1	0.28	44.51 d
	ϵ no γ				β^- .466, .271, ··· γ 1099.2. 1291.6, ···
δ_γ 2.3, 1.3	σ_α 17E1	σ_γ 2.6, 1.4	δ_γ 2.5, 1.6	σ_γ 1.2, 1.5	δ_α < 10
63.939613	E .231	55.934940	56.935396	57.933276	E 1.565

Beginning with Fe-54 serving as the target nuclide, neutron irradiation produces Fe-55 (the activation product).

Although radioactive, Fe-55 has a long half life (2.73 years). For significant production of Fe-55, very long exposure periods would be required.

Even if it could be produced in quantity, Fe-55 decays exclusively by electron capture (ϵ); no gamma rays are emitted with which we could detect and identify its presence.

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Predicting the Sensitivity of NAA - Iron

Fe54	Fe55 ³/⁻	Fe56	Fe57 ¹/⁻	Fe58	Fe59 ³/⁻
5.9	2.73 a	91.72	2.1	0.28	44.51 d
	ε no γ				β⁻ .466, .271, ··· γ 1099.2. 1291.6, ···
σ_γ 2.3, 1.3	σ_α 17E1	σ_γ 2.6, 1.4	σ_γ 2.5, 1.6	σ_γ 1.2, 1.5	σ_α < 10
63.939613	E .231	55.934940	56.935396	57.933278	E 1.565

The next stable nuclide of iron that we might be able to detect is Fe-56.

Unfortunately, the Fe-56 in the sample is simply converted to another stable nuclide, Fe-57.

Similarly, any Fe-57 present in the sample will be converted into a stable product, Fe-58.

Predicting the Sensitivity of NAA - Iron

If NAA is going to detect the presence of iron with any degree of sensitivity, it must be done by converting Fe-58 into Fe-59.

The half life of Fe-59 (44.51 days) is a bit long but it's the only option.

Fe-59 emits gamma rays at 1099.2 and 1291.6 keV with good intensities, 56% and 43% respectively.

The cross section of the target, Fe-58, is decent: 1.2 barns.

Predicting the Sensitivity of NAA - Iron

The problem lies in the low abundance of the target, 0.28%.

As such, the detection of trace quantities of iron requires high neutron fluence rates.

Predicting the Sensitivity of NAA - Manganese

Mn53 7/− 3.7E6 a	Mn54 3+ 312.2 d	Mn55 5/− 100	Mn56 3+ 2.578 h	Mn57 5/− 1.45 m	(0)+ Mn58 3+ 3.0 s ←→ 65 s
ε nσγ	ε γ 834.8 δγ < 10	σγ 13.5, 14.0	β⁻ 2.84, 1.04,··· γ 846.8, 1810.8, 2113.1,···	β⁻ 2.55,··· γ 122.1, 14.4, 692.0,···	β⁻ 6.1,··· β⁻ 3.8,··· γ 1447− γ 810.8, 2227 1323.1, 459.2,···
	E 1.377		E 3.696	E 2.692	E 6.32

We now consider the case of manganese.

The target abundance is great. Manganese has only one stable nuclide, Mn-55, with an abundance of 100%!

Mn-55's cross section for neutron capture is high:13.3 barns. Mn-56, the activation product, has an ideal half-life of 2.578 hours and emits a gamma ray at 846.8 keV. The latter's intensity is 99%.

As a result, manganese is extraordinarily easy to detect..

Neutron Activation Analysis (NAA)

Quantifying an Element via NAA

There are two general approaches to quantitative neutron activation analysis:

1. Use a standard (also referred to as a comparator or monitor).

2. Without a standard. Use equations to estimate the mass of the target element based on the measured activity of the activation product.

   Since this approach uses relatively simple mathematics to describe a complex situation, it has a number of problems that are avoided when a standard is used.

Neutron Activation Analysis (NAA)

Quantifying an Element via NAA by Using a Standard

1. A standard containing a known mass of the element of interest (m_S) is irradiated with neutrons for time (t_a)

2. After activation, the sample is counted for time t_c on a gamma spec system. The time between the end of activation and the start of the count is t_w.

3. The net counts (C_S) are determined for the main photopeak produced   by the activation product.

4. A calibration factor (F) is determined relating the target element mass in the standard to the photopeak counts.

Neutron Activation Analysis (NAA)

Quantifying an Element via NAA by Using a Standard - Example:

A standard soil sample is obtained from the National Institute of Standards and Technology (NIST) at great expense. According to the calibration sheet it contains:

$$10.0 \pm 0.4 \text{ mg Mn}$$

The standard is exposed to neutrons for 6 minutes.

The activated sample is counted 0.5 minutes after activation.
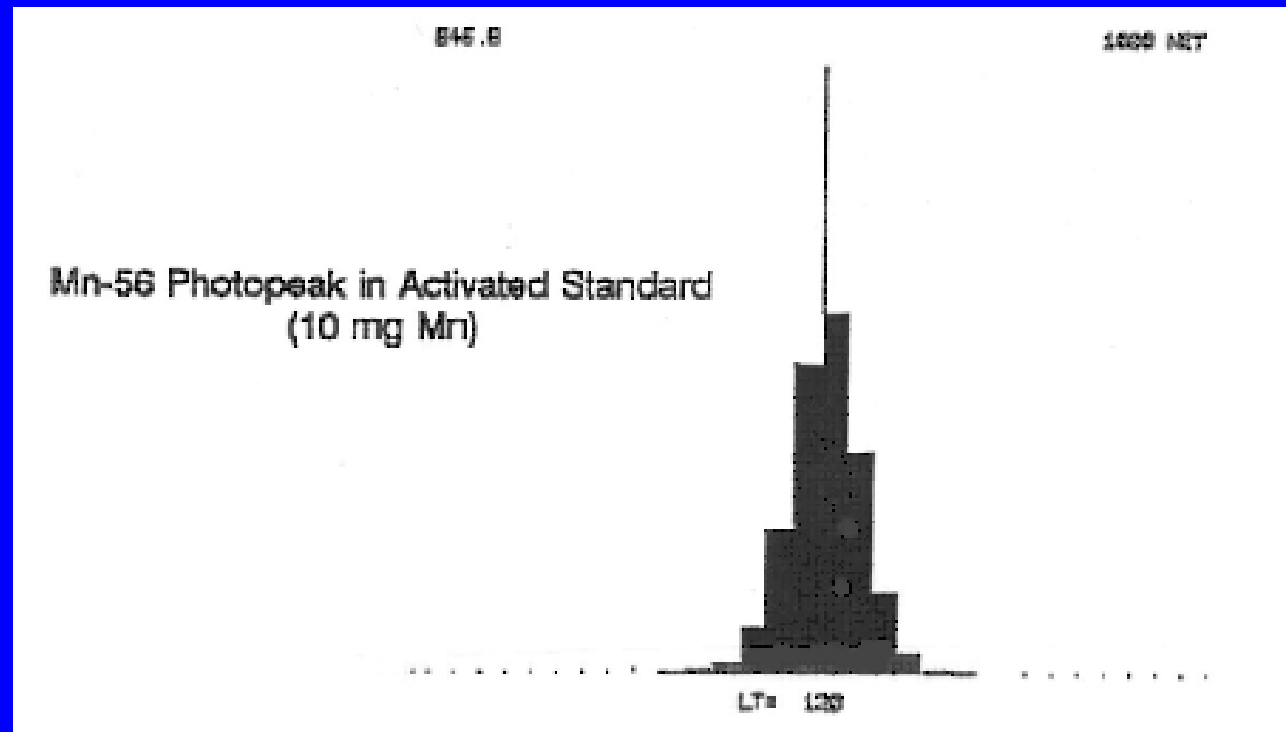
The count is for 2 minutes using a germanium detector.

Neutron Activation Analysis (NAA)

Quantifying an Element via NAA by Using a Standard - Example:

A photopeak appears for the Mn-56 847 keV gamma ray.

A region of interest is set up about the peak and the net count is 1000.

Mn-56 Photopeak in Activated Standard (10 mg Mn)

Neutron Activation Analysis (NAA)

Quantifying an Element via NAA by Using a Standard - Example:

The calibration factor is calculated as follows:

$$F = \frac{m_S}{C_S}$$

$$= \frac{10\,mg\ of\ manganese}{1{,}000\ counts}$$

$$= 0.01\,mg\ of\ manganese\,/\,count$$

Neutron Activation Analysis (NAA)

Quantifying an Element via NAA by Using a Standard - Example:

Now an unknown sample is analyzed. While not essential, it is best to handle the unknown and standard the same way:

The unknown is activated for 6 minutes.
The time between activation and counting is 0.5 minutes.
The count time is 2 minutes.

If enough manganese is present to exceed the lower limit of detection, a discernible photopeak will show up at 847 keV. Assuming that such a peak yields a net count of 500:

Neutron Activation Analysis (NAA)

Quantifying an Element via NAA by Using a Standard - Example:

If such a peak yields a net count of 500, what is the mass of the manganese in the unknown?

846.8   500 NET

Mn-56 Photopeak in Activated Unknown
(mg Mn = ?)

LT= 120

Neutron Activation Analysis (NAA)

Quantifying an Element via NAA by Using a Standard - Example:

The mass of the manganese in the unknown (m_U) is calculated as follows:

$$m_U = F \ x \ C_U$$
$$= 0.01 \ x \ 500$$
$$= 5 \ mg \ of \ manganese$$

It couldn't be easier!

Neutron Activation Analysis (NAA)

Quantifying an Element via NAA Without a Standard

The following equation gives the activity (A) of the activation product at the start of the count:

$$A = N \sigma \phi (1 - e^{-\lambda t_a}) e^{-\lambda t_w}$$

N is the number of target atoms (e.g., Mn-55)

σ is the thermal neutron capture cross section of the target nuclide (cm^2)

ϕ is the thermal neutron fluence rate ($n/cm^2/s$)

λ is the decay constant of the activation product (min^{-1})

t_a is the activation time (e.g., min)

t_w is the time between the end of activation and the start of the count (e.g., min)

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Neutron Activation Analysis (NAA)

Quantifying an Element via NAA Without a Standard

In neutron activation, we are primarily interested in determining the amount of the target nuclide in the sample.

As such, the appropriate form of the previous equation is:

$$N = \frac{A}{\sigma \, \phi \, (1 - e^{-\lambda t_a}) \, e^{-\lambda t_w}}$$

A is the activity of the activation product at the start of the count (Bq). It is usually determined by gamma spectroscopy.

Neutron Activation Analysis (NAA)

Quantifying an Element via NAA Without a Standard

The mass of the target nuclide (e.g., Mn-55) in the sample is:

$$\text{mass of the target nuclide} = \frac{N \ x \ M_i}{6.02 \ x \ 10^{23}}$$

M_i is the isotopic mass of the target nuclide (grams/mole)

$6.02 \ x \ 10^{23}$ is Avogadro's number (atoms per mole)

Neutron Activation Analysis (NAA)

Quantifying an Element via NAA Without a Standard

The mass of the target element (e.g., manganese) in the sample is:

$$\text{mass of the target element} = \frac{\text{mass of target nuclide}}{f}$$

f is the fractional abundance of the target nuclide (not the percent abundance), e.g., the fraction of the mass of manganese that is Mn-55.

Neutron Activation Analysis (NAA)

Quantifying an Element via NAA Without a Standard - Problem

The preceding equations are reasonably straightforward if the neutrons are all of the same energy and the cross section for neutrons of that energy is known.

In many situations, thermal neutrons are so predominant that they are the only ones that need to be considered. Furthermore, capture cross sections are often much larger for thermal neutrons than neutrons of higher energy.

Another potential problem is a non-uniform fluence rate throughout the sample volume. If the cross sections are large, the fluence rate can be lower in the center of the a physically large sample than near its surface.

Measurement of Neutron Fluence Rate

Measurement of Neutron Fluence Rate

General

The following equation can be used to calculate the neutron fluence rate if the neutrons are predominantly of the energy of interest (e.g. thermal) and if the capture cross sections for the other neutron energies are small.

$$\phi = \frac{A}{N \, \sigma \, (1 - e^{-\lambda t_a}) \, e^{-\lambda t_w}}$$

For example, a certain mass of vanadium (i.e., a known number of V-51 atoms) is irradiated for a known period (t_a). A specified time after irradiation (t_w), the activity of V-52 in the foil (A) is determined by gamma spectrometry and this equation used to predict the neutron fluence rate.

Criticality Accident Dosimetry

Criticality Accident Dosimetry

Initial Screening

The neutrons emitted during a criticality accident might result in sufficient activation of the body's sodium that it can be detected with a GM or NaI detector:

$$n_{th} \; + \; Na\text{-}23 \; \rightarrow \; Na\text{-}24$$

There is quite a bit of the target nuclide, Na-23, in the body and it has a large capture cross section for thermal neutrons. The activation product, Na-24 has a half life of 14.96 hours and emits high energy gammas.

Immediately after the criticality, approximately 50% of the induced activity is Cl-38. After 2 and 4 hours, the contribution drops to 10% and 1% respectively.

Criticality Accident Dosimetry

Initial Screening

A quick sort of the victims can be accomplished by placing a side window GM or NaI against the victim's abdomen and having them bend over the probe to improve the counting geometry. Another approach is to place the detector under the armpits.

Any increase in count rate is indicative of an exposure.

Several different rules of thumb have been proposed for evaluating the neutron dose. Some of these are indicated on the following slide.

Criticality Accident Dosimetry

Initial Screening

As a very crude quantitative estimate, an increase of 0.01 mR/hr (0.10 uSv/h) above background indicates a fast neutron exposure of 1 rad (0.01 Gy).

If the instrument reads out in mR/h, the neutron dose can be estimated by using the following relationship:

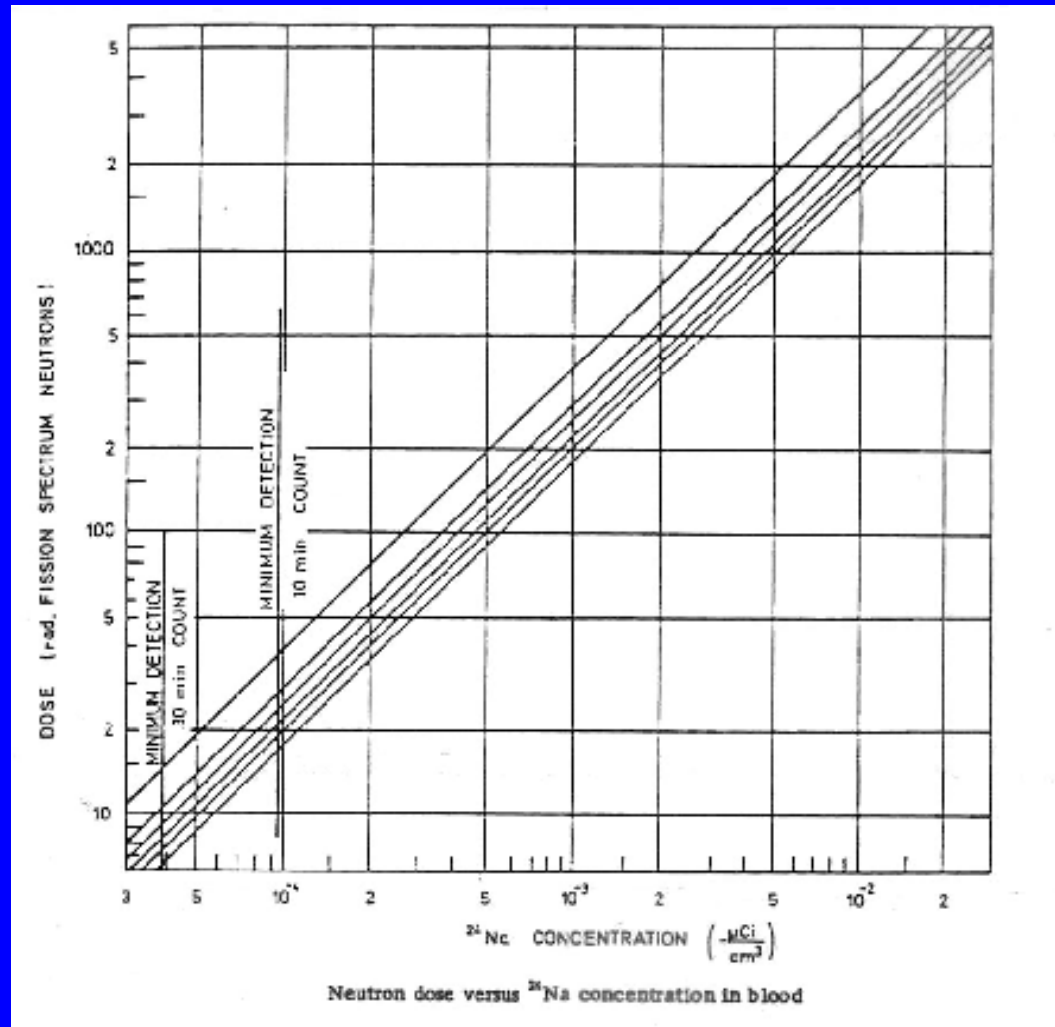Dose (rad)　　=　　8000 X/body weight in pounds

where X is the exposure rate in mR/h

" 70 counts per minute above background equals one rad of neutron dose." (Ricks et al 2002).

Criticality Accident Dosimetry

More Detailed Analysis

By measuring the sodium-24 in the blood, back-calculating the activity to the time of the accident, and using a graph like that to the right, it is possible to estimate the neutron dose.

Neutron dose versus ^{24}Na concentration in blood

Hazards Associated with Neutron Activation

Hazards Associated with Neutron Activation

General

After the source of neutrons has been removed, the hazard associated with the activation products can persist.

For example, following a reactor shut-down it is necessary to wait before entering containment to permit the short-lived activation products to decay. The activation product Co-60 is a particular problem because it has a long half-life and emits high energy (penetrating) gammas. For the most part, it is produced by activation of the cobalt present in valves in the coolant lines. Approximately 50% of the radiation exposures at a nuclear power plant are due to Co-60. Other activation products of importance in reactors include Cr-51, Co-58, Mn-54, Fe-59, and H-3.

Hazards Associated with Neutron Activation

General

High energy (fast) neutrons are often produced in large quantities in the vicinity of accelerator targets.

Although the induced activity decreases by one half in the first 10 minutes after shut-down, long-lived activation products may persist.

Common activation products near the target include: Mg-27, Na-24, Cu-62 and Al-28.

Tritium, which is an internal hazard, can also be produced in a large quantities, especially in pump oil, e.g. ion pumps.

N-16 can be produced in the air near the target and in the cooling water supply by the activation of 0-16.