Activation Analysis of Soil, Air and Water near the NSLS-II Accelerator Enclosures

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Abstract

The radiation environment in the accelerator enclosures is sufficiently high energy to permit the activation of air inside the accelerator enclosures, soil surrounding the concrete shielding of the enclosures and the coolawent water used for cooling various components of the accelerator system like magnets etc. Bremsstrahlung is generated inside the accelerator enclosures by the radiative interaction of the electron beam with the accelerator components. This bremsstrahlung further interacts with the surrounding air, soil and cooling water to produce radioactive isotopes by the emission of neutrons. These interactions could be direct photo-spallation reactions as in the case of air and water activation or could be secondary interactions of high energy neutrons produced by the photoneutron production. The high energy neutrons produced by the photonuclear interactions will penetrate the concrete shield and produce radioisotopes in the surrounding soil by (n,n), (n,2n) etc. interactions. It should be noted that the past experience at the NSLS and other light sources has demonstrated that the potential for air, water or soil irradiation is quite limited and does not warrant significant radiological considerations.

1. Radioactivation due to Beam Losses at NSLS-II

The activation analysis for the NSLS-II accelerator enclosures has been carried out at four distinct locations where the probability for beam loss is a maximum. These are at the linac momentum slit, at the booster injection septum, at the booster extraction septum and at the storage ring injection region. In each case most conservative point beamloss assumptions are employed. In the linac enclosure, a 50% loss of the accelerated beam at 200 MeV is assumed at the momentum slit. The assumed pulse rate is 10 pulses of 15 nC a minute. At the booster injection septum also a 50% loss of the beam at 200 MeV is assumed. The assumed pulse rate is 5 pulses of 15 nC a minute. At the booster extraction septum a 20% beam loss at 3 GeV is assumed at the rate of 5 pulses of 15 nC a minute. The assumed beam dissipation at the storage ring injection region is 18 nC a minute, with 2 hour beam life time, taking into account for the injection losses at the septum. A continuous operation of 5000 hours at the top-off mode is assumed for the facility.

These estimates of beam losses are conservatively derived and will provide an over-estimate of the actual losses that will take place during normal operation. There will be beam stops in the booster and linac enclosures that will also receive beam during commissioning and tuning periods. Losses at these beam stops have not been included in this analysis, since they will be locally shielded with photon (lead) and neutron (polyethylene) shields.

2. Soil Activation

The potential for soil activation is limited at the electron accelerators, since the main radiation component, bremsstrahlung, is in the forward direction [1] of the electron beam and gets absorbed by the machine components like magnets, absorbers etc. The soil berm is generally at very large angles (almost at right angles) to the forward direction of the electron beam. However there exists a potential for the high energy neutron component to penetrate through the transverse concrete shield and produce radioisotopes in the soil [2]. Some of these isotopes can be leached from the soil by rain water and migrate to the ground water systems. It is important to assess this risk for NSLS-II and determine the potential consequences to the environment.

2.1. Methodology to Estimate Soil Activation

The mechanism of formation of radionuclides in the soil is due to the interaction of high energy neutrons with the elements in the soil. In the present analysis only the high energy neutron component needs to be

considered, because only these neutrons have the penetrating power to escape the concrete shielding. As required by the BNL subject area Accelerator Safety, analysis has been done to estimate the rate of formation of two radioactive isotopes ^{3}H and ^{22}Na in the soil during the routine operation of NSLS-II. Table 1 gives the summary parameters used for this analysis. The cross section values are obtained from the literature [3,4] and the soil composition for the Long Island soil has been provided by the BNL Environmental and Waste Management Services Division [5]. The soil density at the Long Island is taken as 1.6 gm/cm^3.

Parent Nucleus	Weight (%) In soil	^{3}H Production Threshold (MeV)	^{3}H Production Cross section (mb)	^{22}NaProduction Threshold (MeV)	^{22}NaProduction Cross section (mb)
^{16}O	51.3	15.0	3.07		
^{23}Na	0.0196	20.0	6.81	15.0	36.6
^{24}Mg	0.21	25.0	6.48	25.0	28.2
^{27}Al	1.65	25.0	8.23	50.0	14.5
^{28}Si	45.1	50.0	3.54	50.0	14.5
^{39}K	0.063	50.0	3.56		
^{40}Ca	0.058	50.0	3.0		
^{55}Mn	0.012	50.0	2.0		
^{56}Fe	1.44	50.0	1.65		

Table 1 - Soil Composition and Cross Sections used for Activation Analysis Cross Sections for High Energy (>50 MeV) Neutrons.

From Table 1 the following average parameters can be derived.

Weighted average cross section for ^{3}H production	3.35 mbarns
Weighted average cross section for ^{22}Na production	14.56 mbarns
Weighted average of soil atomic mass number	22.17
Atomic Number density of soil	4.35 x 10^{22} atoms/cm^3
Number density of ^{3}H producers (100%)	4.35x 10^{22} atoms/cm^3
Number density of ^{22}Na producers (47%)	2.04 x 10^{22} atoms/cm^3

When the electron beam interacts with the storage ring components or beam stops an electromagnetic shower will be generated within the material due to successive bremsstrahlung and pair production interactions. The ultimate product of these interactions is a large number of low energy gammas, electrons, positrons and a few neutrons. Almost 80% of the neutrons emitted are with a median energy of 2.1 MeV and are effectively attenuated by the concrete shielding. However, the high energy component of the neutrons can penetrate the concrete shielding and cause interactions in the surrounding soil.
The high energy neutron (HEN) component in the transverse direction of the beam loss location on a thick target is provided by Fasso [6] et al. as:

$$1.3 \times 10^{-3} \text{ HEN/ GeV/ electron/ steradian}$$

The neutron flux at the external surface of the concrete shield wall of thickness 'r' cm at a distance of 'R' cm from the source in the transverse direction, without assuming any self shielding for the neutrons in the target can be estimated as:

$$\Phi(0) = \frac{1.3\times10^{-3}}{R^2} N_e \times E \times e^{-r/\lambda} \text{ neutrons/cm}^2\text{.s} \quad (1)$$

where N_e = Number of electrons interacting with the target material / sec
E = Energy of the electron in GeV
R = Distance of the flux point from the source in cm
r = Thickness of the concrete shield in g/cm^2
λ = Attenuation length of HEN in concrete in g/cm^2 (115 g/cm^2)

Table 2 summarizes the electron energy dissipation at various locations under consideration and the resulting neutron fluxes, Φ(0), on the exterior surface of the concrete shield . The concrete shield thickness and the distances to the activation locations from the loss points are taken from the 80% architectural design drawings of the NSLS-II.

As high energy neutrons traverse through the soil a large fraction of neutrons are removed from the soil by the elastic and non-radiative capture reactions. These neutrons do not take part in the activation reactions. The neutron removal cross section for the soil is given in the literature [7] as 0.016 cm^{-1} for the soil density of 1.6 gm/cm^3. The high energy neutron flux in the soil as a function of soil thickness (x) can be written as:

$$\Phi(x) = \Phi(0)\, e^{-\Sigma x} \tag{2}$$

Where Σ is the neutron removal cross section of the soil and Φ(0) and Φ(x) are neutron flux values at a soil thickness of 0 and x cm correspondingly. The neutron flux declines in an exponential manner with the mean path (1/Σ) of neutrons in the soil. In approximately 4.8 mean free paths (300 cm) 99% of the neutrons will be removed from the soil by the removal reactions. The average flux of neutrons for activation reactions in the soil can be written as:

$$\Phi_{av} = \frac{\int \Phi(x)dx}{\int dx} = \frac{\int \Phi(0)e^{-\Sigma x}dx}{\int dx} \tag{3}$$

Where Φ(0) is the flux at the external surface of the concrete shield and the integration is carried out for 300 cm of the soil thickness. Integrating equation (3) for 300 cm of soil and substituting for Σ of the soil, it can be shown that:

$$\Phi_{av}\,(\text{soil}) = 0.2066\, \Phi(0)$$

Beam Loss Locations	Number of electrons lost /sec	Distance of dose point R (cm)	Concrete thickness r (cm)	Neutron Flux Φ(0) ($n/cm^2.s$)	Average Neutron Flux Φ_{av} ($n/cm^2.s$)
Linac momentum slit Floor	7.8×10^9	170	50	2.53×10^1	5.23
Linac momentum slit Lateral wall	7.8×10^9	150	50	3.25×10^1	6.72
Booster injection septum Floor	3.9×10^9	170	50	1.26×10^1	2.61
Booster injection septum Lateral wall	3.9×10^9	150	50	1.63×10^1	3.36
Booster extraction septum Floor	1.56×10^9	170	50	7.59×10^1	1.57×10^1
Booster extraction septum Lateral wall	1.56×10^9	150	50	9.75×10^1	2.01×10^1
Storage Ring Floor	1.87×10^9	188	68	5.14×10^1	1.06×10^1

Table 2 - High Energy Neutron Flux at the Exterior of the Concrete Shield and Average Flux in the Soil.

$$\text{Rate of radioactive atom production in soil} = \Phi_{av}\, N\, \sigma \tag{4}$$

where Φ_{av} = Average high energy neutron flux in the soil ($n/cm^2.s$)
N = Number density of element in the soil producing the radioactive isotope
σ = Neutron cross section of the element for activation (cm^2)

The number of atoms of the radionuclide of interest (n) per unit volume is governed by the following differential equation during the period of irradiation:

$$\frac{dn}{dt} = -\lambda_R n + \Phi_{av} N \sigma$$

The equation has the following solution applying the boundary condition when n = 0, t=0:

$$n(t) = \frac{\Phi_{av} N \sigma}{\lambda_R} (1 - e^{-\lambda_R t}) \quad (5)$$

where λ_R = Decay constant of the radioactive isotope
t = Irradiation time (for NSLS-II operations, a conservative annual irradiation time of 5000 hours is considered).

Thus the specific activity (Bq/cm^3) induced in the soil as a function of time by this nuclide of interest:

$$A(t) = \Phi_{av} N \sigma (1 - e^{-\lambda_R t}) \quad Bq/cm^3 \quad (6)$$

The activity of the radioactive nuclide of interest in the soil is calculated using equation (6) and available cross sections from Table 1.

2.2. Results of Soil Activation Calculations for NSLS-II

Table 3 gives the activity in the soil at various beam loss locations created due to ^{3}H and ^{22}Na by 5000 hours of NSLS II operation.
Using the methodology established in the Accelerator Safety Subject Area, the leachable concentration created in the soil has also been given. Leachability of 100% and 7.5% are assumed for ^{3}H and ^{22}Na correspondingly. A water concentration factor of 1.1 is taken due to the annual rain fall of 55 cm at Long Island, according to reference [8]. It can be mentioned that the soil underneath the concrete floor is not exposed to rain fall and the potential leachability of radioactive isotopes from the soil to the water table at these locations will be minimal.

Beam loss Location	Average HEN Flux in soil Φ_{av} (n/cm^2.s)	^{3}H Soil Activity (Ci/cm^3)	^{3}H Leachable to water (pCi/liter)	^{22}Na Soil Activity (Ci/cm^3)	^{22}Na Leachable to water (pCi/liter)
Linac momentum slit Floor	5.23	0.31×10^{-15}	0.34	0.30×10^{-14}	0.25
Linac momentum slit Lateral wall	6.72	0.40×10^{-15}	0.44	0.38×10^{-14}	0.31
Booster injection septum Floor	2.61	0.62×10^{-15}	0.68	0.56×10^{-14}	0.46
Booster injection septum Lateral wall	3.36	0.80×10^{-15}	0.88	0.77×10^{-14}	0.64
Booster extraction septum Floor	15.7	1.87×10^{-15}	2.06	1.81×10^{-14}	1.49
Booster extraction septum Lateral wall	20.1	2.40×10^{-15}	2.64	2.32×10^{-14}	1.91
Storage ring Floor	10.6	1.27×10^{-15}	1.39	1.23×10^{-14}	1.01

Table 3 - Activity in the soil at various beam loss locations created by ^{3}H and ^{22}Na.

As specified in the Accelerator Safety Subject Areas; we assumed 100% leachability for tritium and 7.5% leachability for sodium. A soil water concentration factor of 1.1 is also provided in the Subject Area. These calculated values are well within the BNL Action Levels of 1000 pCi/l and 20 pCi/l for ^{3}H and ^{22}Na. Therefore no additional engineered safeguards are required.

3. Activation of air in the Accelerator Enclosures

Routine accelerator operations at NSLS-II would generate small amounts of air activation at high beam loss locations due to photoneutron reactions of bremsstrahlung in air. The isotopes produced due to air activation are ^{13}N (half life =10 min), ^{11}C (half life = 20 min) and ^{15}O (half life = 2.1 min). These would be produced

within the accelerator enclosure and attain saturation activity within hours of accelerator operation, but would decay quickly because of the short half lives and remain primarily within the confines of the enclosure. The air activation analysis for the NSLS-II has been carried out at four distinct locations of the accelerator enclosures, as given in section 2, where the probability for beam loss is a maximum. These rates of assumed point beam loss, are also given in section 2.

3.1. Methodology of Air Activation Calculations

The mechanism for formation of radionuclides in the air is the photoneutron interaction of bremsstrahlung with air nuclei. The threshold for (g,n) reactions in air is 10.55 MeV for ^{13}N and 15.67 MeV for ^{15}O. In addition ^{11}C is formed of photospallation of both nitrogen and oxygen. Using the neutron yield expression from reference [9] and taking the effective Z for air as 7.26, the neutron yield in air is given by the expression:

$$Y = 1.21 \times 10^8 Z^{0.66} \text{ neutrons/Joule} = 4.5 \times 10^8 \text{ n/J} \quad (7)$$

Implicit in the release of neutrons, is the formation of an unstable nucleus in air, which may be radioactive. Equation (7) gives only the Giant Resonance Neutron (GRN) yield. The yield of High Energy Neutrons (HEN) increases the total neutron yield by about 6%. But since most of the GRN neutrons are well below the threshold energies mentioned above, using the yield of GRN neutrons alone will give a conservative estimate of the radionuclide formation.

The change in number (n) of radioactive atoms in air per unit time if no ventilation flow is present is:

$$\frac{dn}{dt} = YFW\,(1- e^{-x/\lambda}) - \lambda_R n \quad (8)$$

where W = Beam energy dissipated at the location (watts)
x = Effective air path of bremsstrahlung in the accelerator enclosures (4 meters)
λ = Effective bremsstrahlung attenuation length in air [6] (385 meters)
λ_R = Decay constant of the radioactive nuclide (s^{-1})
F = Fraction of beam energy that converts into bremsstrahlung and escapes into air.

In the case of beam loss at the linac momentum slit and septa, a conservative radiation yield of 50% is assumed (F=0.5) [2]. Attenuation due to the local shielding that will be provided around the high loss points is not taken into account in these calculations. In reality a fraction of this bremsstrahlung will get absorbed in the accelerator components without ever traversing through air.

Solving equation (8) for initial conditions n=0 for t=0

$$n = \frac{WFY}{\lambda_R}\,(1 - e^{-x/\lambda})\,(1- e^{-\lambda_R t})$$

The activity due to these radionuclides is equal to $\lambda_R n$

$$A = WFY\,(1 - e^{-x/\lambda})\,(1- e^{-\lambda_R t}) \quad Beq$$

Since the radionuclides formed have short half lives (< 20 min), saturation is achieved quickly during operation and the activation at saturation becomes:

$$A = WFY\,(1 - e^{-x/\lambda}) \quad Beq \quad (9)$$

Of the total saturation activity, 88% is from ^{13}N, 9.5% is from ^{15}O and 1.9% is from ^{11}C, reference [10].

3.2. Results of Air Activation Calculations for NSLS-II Accelerator Enclosures

Table 4 gives the activity in air at the four specified beam loss locations inside the accelerator enclosures of NSLS-II. The volume of air in each enclosure is also given in Table 3. The air changes in the enclosures cause mixing of the activity in the entire volume of the enclosure. The air in the storage ring is supplemented by 5000 cfm of air to maintain the positive pressure inside the storage ring. An equivalent rate of air volume seeps out from the storage ring. The air is re-circulated in the linac and booster tunnels.

Beam loss location	Beam loss (watts)	Charge loss (nC/s)	Enclosure volume (m^3)	^{13}N (μCi)	^{15}O (μCi)	^{11}C (μCi)	Concentration (μCi/cm^3)
Linac momentum slit	0.25	1.25	473	13.20	1.43	0.28	3×10^{-8}
Booster injection septum	0.125	0.625	1304	6.60	0.71	0.14	5.7×10^{-9}
Booster extraction septum	0.75	0.25	1304	39.60	4.28	0.84	3.4×10^{-8}
Storage Ring injection region	0.90	0.30	7594	47.52	5.13	1.03	7×10^{-9}

Table 4 - Saturation Activity in Air at Various Beam Loss Locations.

The computed concentration of radionuclides in air at various beam loss locations inside the accelerator enclosures for the conservative set of beam loss assumptions is lower than the derived air concentration for occupational computed from DOE order 5400.5. Once the operation is shut down, this concentration will rapidly decrease due to radioactive decay and air ventilation. As has been the case at NSLS and other light sources, it is clear that there will be no problems associated with air activation within the enclosures of NSLS-II accelerators.

We have also used these calculations to estimate the potential dose at the site boundary from air releases to the atmosphere. These calculations have been performed by B. Hooda of the BNL Environmental and Waste Management Services Division using the CAP88 program. He has concluded that the annual dose to a person living at the site boundary from air releases from the NSLS-II operations will be << 0.001 mrem/year and can therefore be considered negligible.

4. Radioactivation of the Cooling Water

Activation of water for cooling the magnets and the other accelerator components may be estimated by the similar method as the estimation of air activation inside the accelerator enclosures. The primary reactions leading to the activation of cooling water are the bremsstrahlung interactions with ^{16}O in water. The most abundant of the radionuclides produced by this process is ^{15}O. Other activation products that are formed include ^{11}C (4.4% of ^{15}O), ^{3}H (at saturation, 2.2% of ^{15}O) and ^{13}N (about 1% of of ^{15}O). ^{15}O has a radioactive half life of 2.05 minutes and attain saturation during a short period of operation. Because of the long half life of ^{3}H (12.3 years), this radionuclide will not attain a substantial fraction of its saturation activity in 5000 hours.

4.1. Methodology to Estimate Cooling Water Activation

The main mechanism for formation of radionuclides in the cooling water is the photoneutron interaction of bremsstrahlung with oxygen nuclei. The threshold for this reaction is 15.67 MeV. In addition some ^{11}C, ^{13}N and ^{3}H are also created due to bremsstrahlung interactions with water. Using the neutron yield expression from reference [9] and taking the effective Z for water as 3.34, the neutron yield in water is given by the expression,

$$Y = 1.21 \times 10^{8}\, Z^{0.66} \text{ neutrons/Joule} = 2.68 \times 10^{8} \text{ n/J} \qquad (10)$$

Implicit in the release of neutrons, is the formation of an unstable nucleus in water, which may be radioactive. Equation (10) gives only the Giant Resonance Neutron (GRN) yield. The yield of High Energy Neutrons (HEN) increases the total neutron yield by about 6%. But since most of the GRN neutrons are below the threshold energies for photoneutron reactions, using the yield of GRN neutrons alone will give a conservative estimate of the radionuclide formation. Assuming the abundance of ^{15}O as 100% for each neutron created, the change in number (n) of radioactive atoms per unit time if the cooling water is in a closed loop is given by:

$$\frac{dn}{dt} = YFW\,(1- e^{-x/\lambda}) - \lambda_R n \qquad (11)$$

where W = Beam energy dissipated at the location (watts)
x = Average path of bremsstrahlung in water (assumed to be 2 cm for the cooling water hoses)
λ = Effective bremsstrahlung attenuation length in water [11] (50 cm)
λ_R = Decay constant of the radioactive nuclide ^{15}O (s^{-1})
F = Fraction of beam energy that converts into bremsstrahlung (assumed to be 50%)

Solving equation (11) for initial conditions n=0 for t=0

$$n = \frac{WFY}{\lambda_R}\,(1 - e^{-x/\lambda})\,(1- e^{-\lambda_R t})$$

The activity due to ^{15}O is equal to $\lambda_R n$

$$\text{Activity} = WFY\,(1 - e^{-x/\lambda})\,(1- e^{-\lambda_R t}) \quad \text{Beq} \qquad (12)$$

Because the half life of ^{15}O is only 2.05 min, it will reach saturation rapidly.
The saturation activity under static conditions will be given by:

$$\text{Saturation Activity} = WFY\,(1 - e^{-x/\lambda}) \quad \text{Beq} \qquad (13)$$

The saturation activity of ^{15}O can be calculated from equation(13). The saturation activity of other nuclides can be calculated from the saturation activity of ^{15}O. It can be noted that ^{3}H will take a long time to attain saturation. In this case equation (12) can be used to calculate the build up of activity of ^{3}H for known period of operation. In the present calculations it is assumed that the cooling water is in a closed system and the total water inventory in the closed loop is ~100,000 gallons (3.78×10^8 cm^3).

4.2 Results of Cooling Water Activation Estimates

Among the accelerator components which require cooling, the storage ring septum is a highest beam loss location. The saturation activity of radionuclides in the storage ring cooling water is estimated using equation (13) for 18 nC of charge dissipation per minute. Table 5 provides the saturation activity of the radio nuclides in the cooling water of the NSLS-II storage ring. As mentioned earlier, ^{3}H will attain saturation only after decades of operation. After 5000 hours of continuous operation, the concentration of ^{3}H will be only 3% of the saturation value.

Beam loss (watts)	Charge dissipation (nC/min)	^{15}O (μCi)	^{11}C (μCi)	^{13}N (μCi)	^{3}H (μCi)
0.90	18	134	5.6	1.2	2.8

Table 5 - Saturation Activities of Radionuclides in the Cooling Water of the Storage Ring (per 100,000 gallons of water).

The computed concentration of radionuclides in the cooling water of 100,000 gallons ($3.78x10^5$ litres) is orders of magnitude smaller than the derived concentration for environmental discharge limits in the DOE

order 5400.5. Once the operation is shut down, concentration of all nuclides, except that of ^{3}H, will rapidly decrease due to radioactive decay of the short lived isotopes.

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