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INDUCED RADIOACTIVITY IN BEVATRON CONCRETE RADIATION SHIELDING BLOCKS*

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Induced Radioactivity in Bevatron Concrete Radiation Shielding Blocks[†]

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Abstract

The Bevatron accelerated protons up to 6.2 GeV and heavy ions up to 2.1 GeV/amu. It operated from 1954 to 1993. Radioactivity was induced in some concrete radiation shielding blocks by prompt radiation. Prompt radiation is primarily neutrons and protons that were generated by the Bevatron's primary beam interactions with targets and other materials. The goal was to identify the gamma-ray emitting nuclides ($t_{1/2} > 0.5$ yr) that could be present in the concrete blocks and estimate the depth at which the maximum radioactivity presently occurs. It is shown that the majority of radioactivity was produced via thermal neutron capture by trace elements present in concrete. The depth of maximum thermal neutron flux, in theory, corresponds with the depth of maximum induced activity. To estimate the depth at which maximum activity occurs in the concrete blocks, the LAHET Code System was used to calculate the depth of maximum thermal neutron flux. The primary beam interactions that generate the neutrons are also modeled by the LAHET Code System.

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Table of Contents

1.0 Introduction

The Bevatron is a synchrotron accelerator facility that operated from 1954 to February 1993. It produced proton beams up to 6.2 GeV and heavy ion beams up 2.1 GeV/amu. When the Bevatron ion beam was incident upon an accelerator component such as a target or beam stop, prompt radiation was emitted from the component. Shielding was required to protect personnel and equipment from the prompt radiation generated by the primary ion beam. The Bevatron facility used large concrete blocks for the majority of prompt radiation shielding, but prompt radiation can induce long-lived radioactivity in concrete. The primary radiological concern associated with radiation from the blocks is long-lived ($t_{1/2}$>0.5 yr) radionuclides that emit gamma-rays upon decay.

Quantifying radioactivity in the concrete blocks can be achieved by surface surveys and by analyzing concrete samples using nuclear spectroscopy methods. The goal of this paper was to identify the long-lived radionuclides that could be present in Bevatron concrete blocks and estimate the depth of maximum activity. Estimates for the depth of maximum activity will indicate the depths at which concrete samples for activity analysis should be removed; thus eliminating the possibility that activity deep in the blocks will go undetected.

Estimating the depth (distance from the block surface) at which peak activity occurs in the concrete blocks requires information about the prompt radiation inducing the radioactivity. The prompt radiation is primarily neutrons and protons generated by the primary beam's interactions with targets, beam dumps, plunging magnets, and beam line apparatus. The majority of the activity in the concrete blocks was probably induced through thermal neutron absorption (i.e. radioactive isotopes are created by certain stable isotopes in the concrete capturing a neutron). The magnitude of induced activity at a specified concrete depth should be proportional to the magnitude of the thermal neutron flux at that depth during accelerator operation. Therefore, the depth of maximum activity can be estimated by calculating the depth of maximum thermal neutron flux.

The neutron production and depth of maximum thermal neutron flux were calculated by the LAHET Code System. This is a Monte-Carlo based computer program that can simulate neutron production and transport from approximately 10 GeV to thermal energies. Neutron production was modeled by simulating 4.0 GeV protons incident upon an iron target located in a cylindrical void. Concrete shielding is modeled by surrounding the cylindrical void with a concentric concrete cylinder. The thermal neutron flux is calculated as a function of radius in the concrete cylinder. Thus, the depth of maximum activity in a concrete block is modeled by calculating the radius in the concrete cylinder at which the thermal neutron flux is maximum.

Quantification of the actual radioactivity levels in the concrete blocks was not investigated. To calculate true activity levels requires a detailed history of Bevatron operation and exact location of each concrete shielding block, but detailed historical information is not available.

2.0 Bevatron and Concrete Block Background Information

The Bevatron is a weak focusing synchrotron that started operation in 1954 as a 6.2 GeV proton accelerator. It is composed of four iron quadrant magnets of 50 feet radius to the center of the aperture (Figure 2.0). Straight sections 20 feet long separate the quadrants. Prior to 1962 all targets were located within the iron magnet quadrants or in the straight sections. Concrete blocks encircled the accelerator to provide shielding for the prompt radiation generated in the targets and other components. The iron quadrant magnets themselves also provided prompt radiation shielding. Magnet steel is 2.5 to 4.0 feet thick. Concrete wall thickness varied from 5 to 10 feet with the greatest thicknesses in the vicinity of target locations. The blocks were stacked approximately 16 feet high.

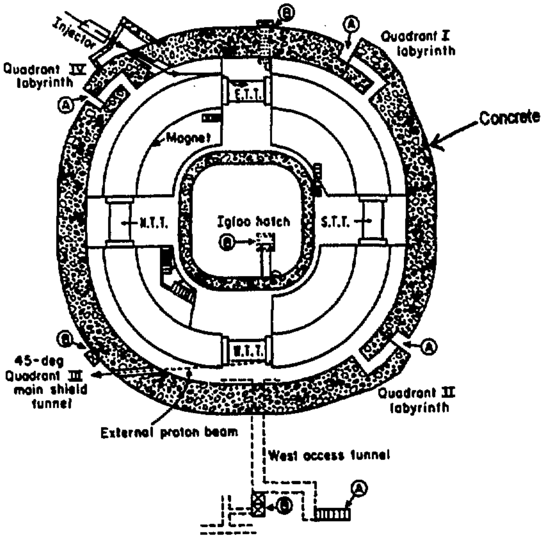

Figure 2.0 Plan view of the original Bevatron 6.2 GeV proton accelerator facility. From *Accelerator Health Physics*, H.W. Patterson, R.H. Thomas [Pat 73].

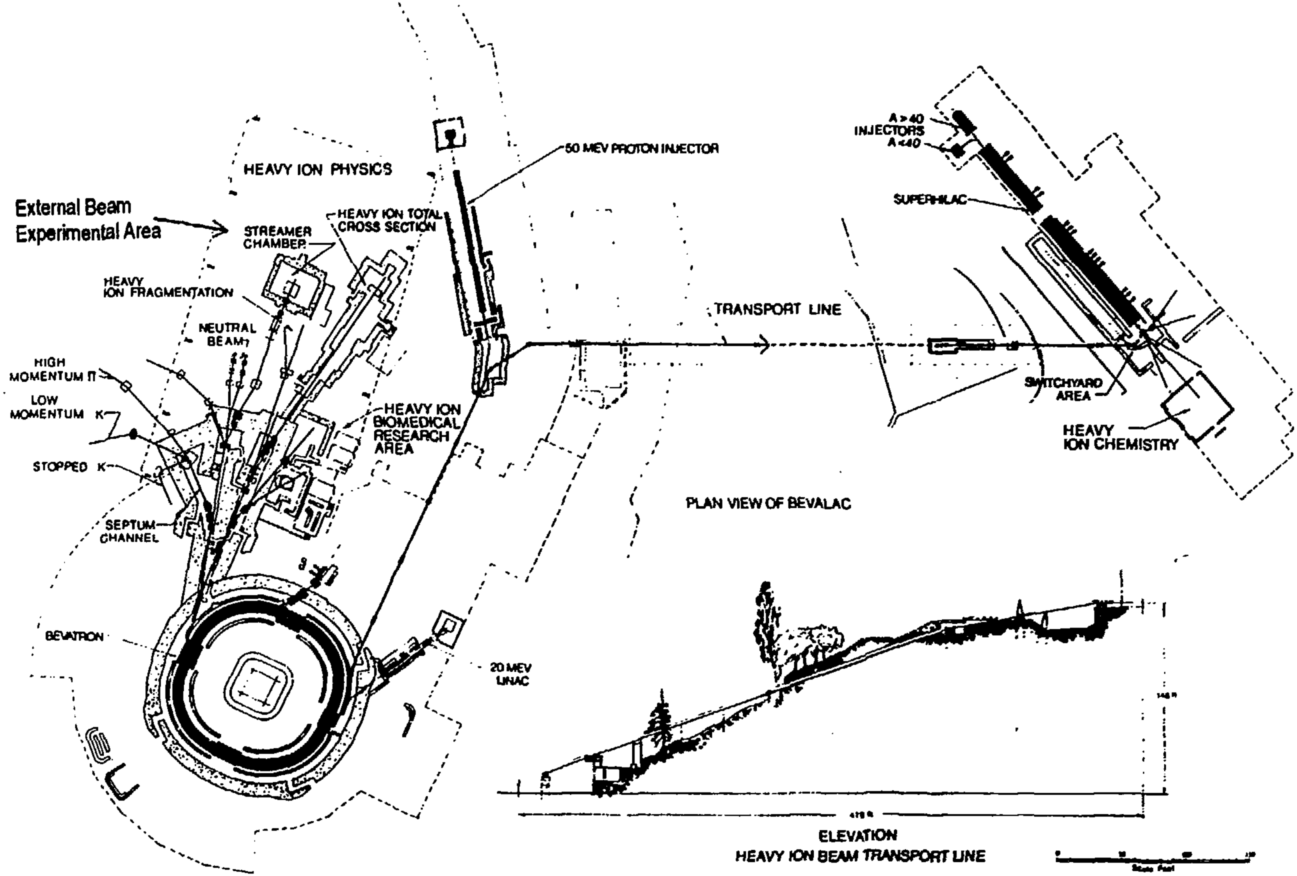

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Figure 2.1 The Bevatron accelerator facility after the External Particle Beam System installation (1962) and SuperHILAC connection (1974). From Bevatron/Bevalac User's Handbook, Biology and Medicine [BE'/1 77].

In June of 1962, the Bevatron ceased operation to permit extensive modifications and improvement. A primary modification was installation of the External Particle Beam System and the associated Experimental Area (Figure 2.1). The additions and modifications to the concrete shielding system included:

1) Increasing the concrete wall thickness to 10 feet at all locations around the accelerator.
2) Installing two courses of blocks made from heavy concrete (3.5 g/cm^3, 225 lbs/ft^3) centered on the median plane of the magnets. Each course of blocks was 4 feet high.
3) Installing a complete concrete roof shield over the accelerator that was 7 feet thick, 2.4 g/cm^3,150 lbs/ft^3
4) *Providing concrete shielding blocks for the new external beam facility.*

Figure 2.2 shows a cross-sectional view of the concrete radiation shielding around the accelerator ring after the 1962 improvements. This configuration was used with only minor alterations up to the time of the Bevatron's shut-down.

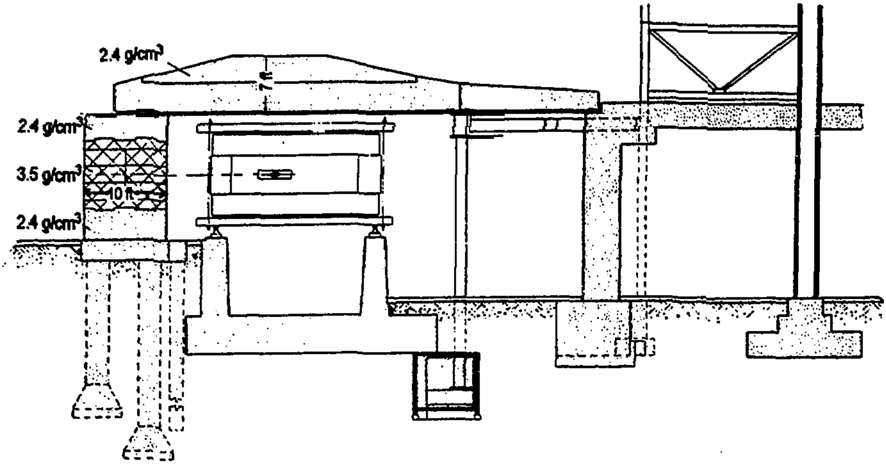

Figure 2.2 Bevatron ring concrete cross-section after 1962 improvements. From *Bevatron Improvements*, E.J. Lofgren, W.D. Hartsough [Lof 63].

After resuming operation in early 1963, most targets were located in the External Beam Experimental Area. There is some evidence that targets were located in the Bevatron ring after 1963, but it appears that this was an uncommon mode of operation. Therefore, after 1963, most of the concrete blocks exposed to prompt radiation were located in the External Beam Experimental Area and in the vicinity of the east and west straight sections where the plunging magnets were located. The plunging magnets were used for diverting the ion beam from the Bevatron ring into the External Beam Experimental Area.

Up to 1971, the Bevatron accelerated only protons. Beginning in 1971, the Bevatron was used to accelerate heavy ions. Starting in 1974 the SuperHILAC linear accelerator was used to inject heavy ions into the Bevatron. The SuperHILAC's pre-acceleration allowed acceleration by the Bevatron of all elements up to uranium with energy up to 2100 MeV/amu. When the SuperHILAC was used as the Bevatron injector, the complete accelerator facility was termed the Bevalac.

Presently there are approximately 4000 concrete blocks at the Bevatron facility. This includes the roof blocks over the accelerator ring, the blocks encircling the accelerator ring, and the blocks in the External Beam Experimental Area. They range in weight from 500 lbs to 60,000 lbs with an average weight of about 21,000 lbs. The total weight of blocks is estimated at 42,000 tons (84×10^6 lbs). The block densities range from 150 lbs/ft^3 to over 300 lbs/ft^3, but the majority are normal density (2.4 g/cm^3,150 lbs/ft^3). Table 2.0 provides concrete block densities with the percentage of total blocks for each density [Gou 94].

Concrete Density (lbs/ft^3)	Percentage of Total Blocks
150	58 %
225	29 %
>225	13 %

Table 2.0 Density of concrete blocks at the Bevatron facility with the percentage of total blocks at each density.

3.0 Induced Radioactivity in Concrete

When a high energy ion beam is incident upon accelerator components, nuclear collisions cause neutrons and protons to be emitted from the component as prompt radiation. At the Bevatron, prompt radiation was generated mainly in targets, beam dumps, plunging magnets, and beam line apparatus. The energy of the prompt neutrons and protons ranged from thermal up to the energy of the ion beam. Personnel and equipment were shielded from the prompt radiation by concrete blocks, but concrete is susceptible to activation by prompt radiation. The prompt radiation generated in accelerator components can induce both long and short lived radioactivity in normal and heavy weight concrete. The mechanisms that produce radioactivity in concrete can be divided into two parts. These are high energy spallation reactions and low energy neutron absorption.

The induced radioactivity at the surface of the concrete blocks is primarily gamma-rays emitted during the decay of radionuclides that were generated by spallation or thermal neutron absorption. In addition to gamma-rays, beta particles (electrons and positrons) are emitted by approximately 75% of the radioactive isotopes in concrete generated by prompt radiation. However, because of the short range of beta particles relative to gamma-rays, the dose from beta particles at the surface is small relative to the dose from gamma-rays [Sul 92]. For this investigation, only the radioisotopes that produce gamma-rays are identified. If a nuclide emits only positrons upon decay, it would also be identified to include the gamma-rays from positron annihilation. Radionuclides that emit only electrons (β^- decay) upon decay would not be identified.

Because of proton beam intensity compared to heavier ion beam intensity and the number of years since proton beams were produced, only the radioisotopes generated in the blocks with half-lives greater than 0.5 years are relevant at this point in time. The intensity (ions/pulse) for proton beams was 2 to 3 orders of magnitude greater than for beams composed of heavier ions [BEV2 77]. Consequently, proton beams produced prompt radiation at levels 2 to 3 orders of magnitude greater than beams of heavier ions. This difference in prompt radiation production between protons and heavier ions indicates activity was induced when the Bevatron was accelerating protons, but after 1974 the Bevatron produced primarily heavy ion beams. Therefore, most activity was induced more than twenty years ago. Radionuclides with half-lives short compared to twenty years would not contribute to activity at this point in time. Consequently, only radioisotopes with half-lives greater than 0.5 year were considered relevant for this investigation.

3.1 High energy induced radioactivity

When high energy (E>~20 MeV) prompt radiation impinges on concrete, nuclei are converted to different isotopes through spallation. Spallation occurs when a high energy particle collides with a nucleus ejecting neutrons, protons, or groups of nucleons from the struck nucleus. The isotope remaining after the interaction can have an atomic weight of any value up to that of the struck nuclei. Normal concrete is composed primarily of Si, O, Al, Fe, Ca, H, and Na. Because Fe is the element in concrete with the largest atomic mass (A), any isotope up to Fe can be produced through spallation. Examination of the chart of nuclides reveals 15 isotopes with A values less than Fe that are long-lived ($t_{1/2}$>0.5 yr) radionuclides. However, only 4 emit gamma-rays upon decay. These nuclides are listed in Table 3.0.

Radioisotope	half-life	gamma energy (MeV)	decay mode	parent isotope for spallation
^{26}Al	7.3x10^5 yr	1.809, 0.511,.....	β^+, EC	^{27}Al, Si
^{22}Na	2.6 yr	1.275, 0.511,.....	β^+, EC	^{23}Na, ^{27}Al, Si
^{54}Mn	312 d	0.835	EC	Fe
^{44}Ti	52 yr	0.078, 0.068	EC	Fe

Table 3.0 Long-lived radionuclides produced by high energy spallation in normal composition concrete.

Manganese-54 and titanium-44 (^{54}Mn, ^{44}Ti) can only be generated by spallation of iron which is 1-2% by weight (not including rebar) in normal concrete. Sodium-22 can be produced from ^{23}Na, ~2%; ^{27}Al, ~4%; and Si, ~30% by weight in normal concrete. Aluminum-26 can be produced from ^{27}Al and Si. The cross-section for any one of these spallation reactions is about the same, on the order of 1 mb. Therefore, the amount of a particular isotope produced through spallation is directly dependent upon the amount of the parent isotope/isotopes present in the concrete. Because iron (parent for ^{54}Mn and ^{44}Ti) is present in small quantities compared to Si, ^{27}Al, and ^{23}Na (parent isotopes for ^{22}Na and ^{26}Al) in *normal composition concrete*, ^{54}Mn and ^{44}Ti will be produced in insignificant amounts compared to ^{22}Na and ^{26}Al.

Approximately equal quantities of ^{22}Na and ^{26}Al would be expected in concrete exposed to high energy prompt radiation. However, the much longer half-life of ^{26}Al compared to ^{22}Na indicates that its specific activity (Ci/g) would by extremely low compared to the specific activity for ^{22}Na. Because of the long half-life for ^{26}Al and because of the small amount of parent isotope for ^{54}Mn and ^{44}Ti, ^{22}Na is the only long-lived radionuclide produced by spallation expected to be detected in normal weight concrete blocks at the Bevatron.

Heavy weight concrete is from 20% to 70% iron by weight. Consequently, the blocks at the Bevatron made from heavy concrete could contain detectable levels of radiation from ^{54}Mn and ^{44}Ti.

3.2 Low energy neutron induced radioactivity

Several trace elements present in concrete become long-lived radioisotopes upon absorption of thermal neutrons, and they also emit gamma-rays upon decay. The trace elements are cobalt-59, cesium-133, europium-151, and europium-153. Table 3.1 provides the pertinent data about the trace elements in concrete that become long-lived radioisotopes upon thermal neutron absorption [Sul 92].

radioisotope	half-life	decay mode	gamma-ray energy (MeV)	parent isotope	σ_{th} for parent (barns)
^{60}Co	5.3 years	β^-	1.33, 1.17	^{59}Co	16
^{152}Eu	13.5 years	β^-, EC	0.121, 1.41,....	^{151}Eu	5900
^{154}Eu	8.6 years	β^-	0.123, 1.27,....	^{153}Eu	1500
^{134}Cs	2.1 years	β^-	0.658, 0.089,..	^{133}Cs	27

Table 3.1 Long-lived radioisotopes produced in concrete by trace elements absorbing thermal neutrons.

These four trace elements are present in only minute amounts compared to the parent elements for spallation. However, their neutron capture cross-sections are 5 to 8 orders of magnitude larger than the cross-section for radioisotope production through spallation. This huge cross-section for thermal neutron capture allows production of radionuclides by neutron capture to dominate radionuclide production in concrete.

4.0 Activity Vs Depth Calculations

The depth of peak activity in the Bevatron concrete blocks can be estimated by considering the case for low energy neutron absorption separate from the case for radioisotopes produced by spallation. However, because the cross-section for producing radionuclides through spallation is extremely small, most activity was produced through low energy neutron absorption. Therefore, the depth at which radioactivity from ^{60}Co, ^{152}Eu, ^{154}Eu, ^{134}Cs is maximum corresponds with the depth of peak activity. The elements that become radioisotopes upon thermal neutron absorption (^{59}Co, ^{151}Eu, ^{153}Eu, ^{133}Cs) are uniformly dispersed in concrete. Consequently, the magnitude of activity at a specified depth is only a function of the thermal neutron flux at that depth during accelerator operation (i.e. the greater the thermal neutron flux the greater the neutron absorption by ^{59}Co, ^{151}Eu, ^{153}Eu, ^{133}Cs). To establish the depth of maximum activity, the depth of maximum thermal neutron flux was calculated using a particle transport computer code.

4.1 Modeling the Bevatron

At the Bevatron, neutron production and the resulting induced activity depended on many variables including: type and thickness of material in which neutrons were generated, beam dynamics and energy, type and thickness of material between the neutron source and the concrete, distance from the neutron source to concrete, and block prompt irradiation history. For determining the depth of maximum activity, no attempt was made to study and model all the different possible situations at the Bevatron. One simple model was used that incorporates common and essential features of concrete exposure to prompt neutrons.

The LAHET Code System [LAH 89] was used to model the neutron production and to estimate the depth in concrete of maximum thermal neutron flux. It is a Monte-Carlo based computer program that computes the results of high energy particle interactions and transports particles including neutrons and protons.

The parameters used for the LAHET Code System to model the neutron production and concrete shielding are as follows:

a) 4.0 GeV protons for source ions.
b) protons incident upon a cylindrical iron target.
c) target 30 cm in diameter by 100 cm long.
d) target located in cylindrical void.
e) cylindrical void surrounded by concentric concrete cylinder.
f) normal composition, normal weight concrete (Table 4.0).

Element	Weight %
H	0.87
O	50.10
Na	1.34
Mg	0.34
Al	4.65
Si	29.6
Ca	7.58
Fe	2.75
total %	97.2

Table 4.0 Concrete composition used to model the concrete radiation shielding blocks at the Bevatron. From *Concrete Activation Experiment at the Bevatron*, W.S. Gilbert, et al. [Gil 68].

Protons were used as the source ion because they generated a much greater flux of neutrons compared to heavier ions, and protons were the only ion beam used for approximately 20 years of Bevatron operation (1954 to 1974). Proton energy varied, but approximately 4.0 GeV was a common energy [Cow 94].

The target diameter of 30 cm in the model is probably much larger than an actual target used at the Bevatron. The large diameter was chosen to model the situation with steel located between the neutron source and the concrete shield. If the target was located in the Bevatron ring, the iron magnets would have provided shielding before the neutrons reached the concrete blocks. Also, in the External Beam Experimental Area some target caves were lined with thick steel plates to provide prompt radiation shielding. For modeling the Bevatron, the target acts as the neutron source. Also, the large target diameter models the situation with some steel between the neutron source and the concrete blocks.

The low energy (0.01 eV-1.0 eV) neutron fluence (n/cm^2) was calculated as a function of radius in a concrete cylinder that surrounds the void in which the iron target is located (Figure 4.0). In Figure 4.0, the Fluence Determination Zone is the section of the concrete cylinder in which neutron fluences were determined. Neutron fluence in this cylinder section corresponds to the neutron fluence at 90 degrees to the beam line.

Areas with high levels of prompt radiation would have been enclosed by side walls, ceiling, and a floor made of concrete. Placing the target in the cylindrical void surrounded by concrete is the method used to model the concrete enclosure around a prompt radiation source. Modeling the concrete geometry around a neutron source is important because the concrete enclosure itself can scatter and contain the neutrons within the shielding walls. Thus, the enclosure around the neutron source can increase the neutron flux at the surface of the concrete.

To investigate the relationship between the total concrete surface area around the target and the depth of maximum neutron fluence, the neutron fluence versus concrete depth was calculated for three different void diameters (100 cm, 400 cm, 600 cm). Altering the void radii also changes the distance from the concrete surface to the neutron source. However, this should only affect the overall fluence magnitude because of the 1/r^2 law, not the depth of maximum fluence.

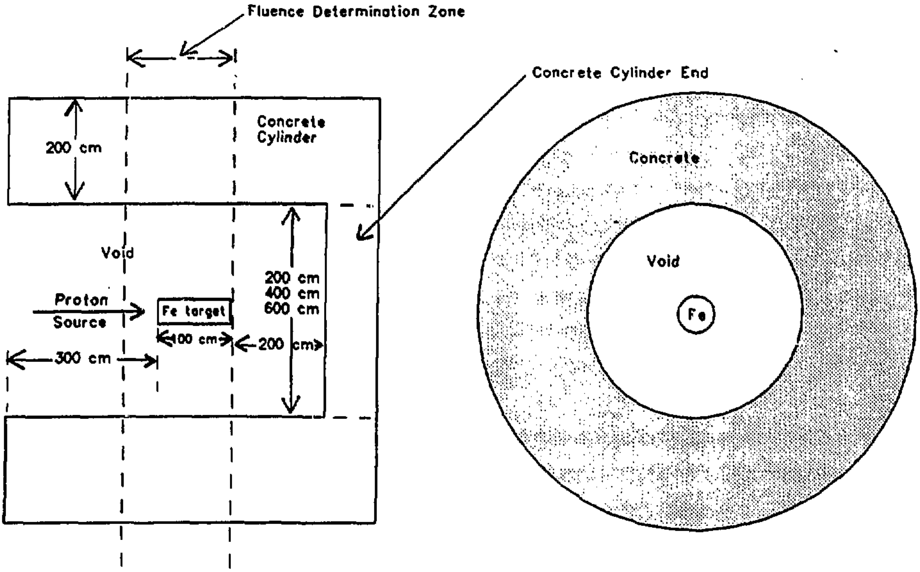

Figure 4.0 Geometry used in the LAHET Code System to calculate low energy neutron fluence in a concrete cylinder that surrounds an iron target, bombarded with 4.0 GeV protons.

4.2 Results

A graph that plots the neutron fluence versus concrete depth is presented in Figure 4.1. The source protons are 4.0 GeV, and the neutron energy range is from 0.01 eV to 1.0 eV.

As Figure 4.1 indicates, the fluence rises slightly from the surface to peak at about 6 cm (2.5 in). The peak at depth instead of near the concrete surface is because there are actually two sources of thermal neutrons. One source is thermal neutrons entering the concrete surface, and the other source is fast neutrons thermalized in the concrete. This thermalization of fast neutrons produces the increase as depth increases.

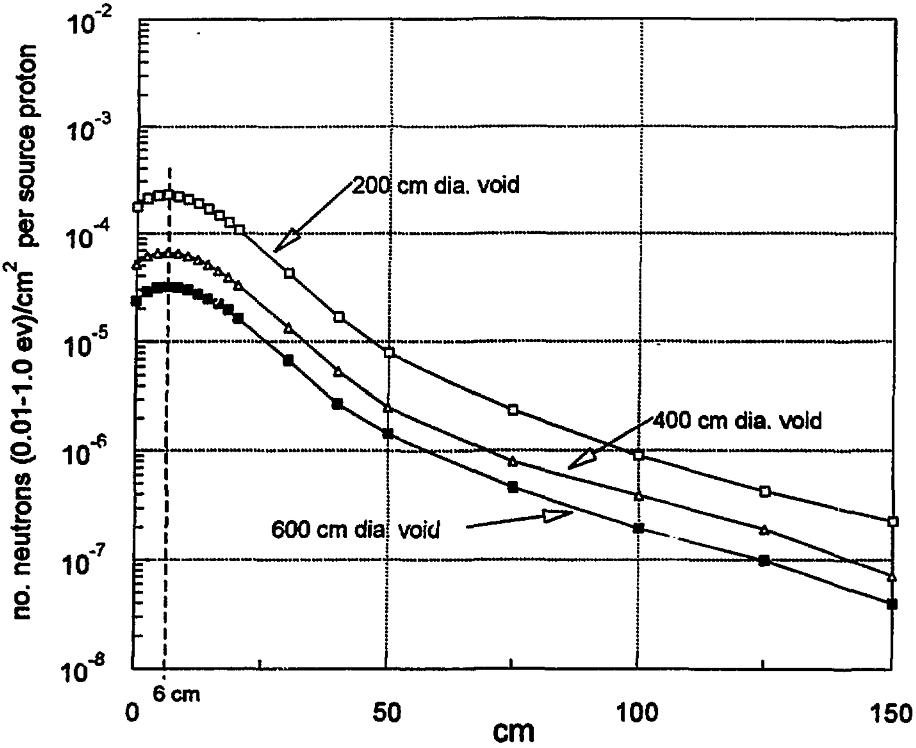

Figure 4.1 Neutron fluence as function of concrete depth at 90 degrees to 4.0 GeV proton beam line as calculated by the LAHET Code System. Protons incident upon Fe cylinder, 30 cm dia. by 100 cm long.

The fluence at 6 cm (i.e. the maximum) is about 10 % greater than at 2 cm (~1 in), and the fluence at 16 cm (6.5 in) is about the same as at the surface. Therefore, for practical purposes, the fluence is essentially constant from the surface to about 16 cm. For depths greater than 16 cm, the fluence decreases approximately exponentially. If the fluence as a function of depth represents activity as a function of depth as theorized, then a concrete sample from 2 cm that is analyzed for activity will represent the activity up to about 16 cm within about 10 % accuracy.

For all three void diameter cases, the depth of peak fluence and the ratio of peak fluence to near surface fluence is the same. This indicates that the activity as a function of depth is not affected by the total concrete surface area of the enclosure as modeled for this investigation.

5.0 Results Compared to Experimental Data

The best available comparison is provided by the concrete radioactivity analysis done for the Princeton-Pennsylvania Accelerator (PPA). Located at Princeton University, the Princeton-Pennsylvania Accelerator was a 3 GeV proton synchrotron that operated from 1962 to 1972. A radiological assessment of the long-lived activity induced in the concrete shielding was performed in 1987 [PPA 87]. Two of the assessment's primary goals coincided with the goals of this paper. These goals were to determine the radioisotopes present in the concrete shielding and determine the activity as a function of depth in the *concrete.*

At the location of highest surface radioactivity, a concrete core sample was analyzed for activity. The core was removed from the floor of the synchrotron ring in a target area. Figure 5.0 plots the specific activity (μCi/g) versus depth for each detected isotope, and Figure 5.1 plots the total specific activity versus depth.

The isotopes detected in the PPA concrete core were ^{22}Na, ^{152}Eu, ^{154}Eu, and ^{60}Co. These are the same isotopes expected in Bevatron concrete blocks exposed to high levels of prompt radiation with the exception of ^{134}Cs. The radioisotope ^{134}Cs was not detected at PPA, but it could be present in Bevatron concrete blocks.

As Figure 5.0 indicates, the activity is primarily from ^{152}Eu, and ^{60}Co which are *both produced by thermal neutron absorption. The contribution to activity from the* spallation product ^{22}Na is insignificant compared to activity produced by thermal neutron absorption. Therefore, as theorized, the activity in the Bevatron concrete blocks is probably generated by trace elements absorbing thermal neutrons.

A table comparing the results from Figure 5.1 with the calculations for the Bevatron is presented in Table 5.0. The total activity versus depth from the PPA analysis corresponds well with the estimates based on thermal neutron fluence versus depth as calculated by the LAHET Code System. The PPA analysis and the calculations both indicate a slight increase in activity from the surface up to a peak at a few inches with activity at 5.5 inches equal to the activity at approximately 1 inch. This correlation between PPA analysis and the theoretical

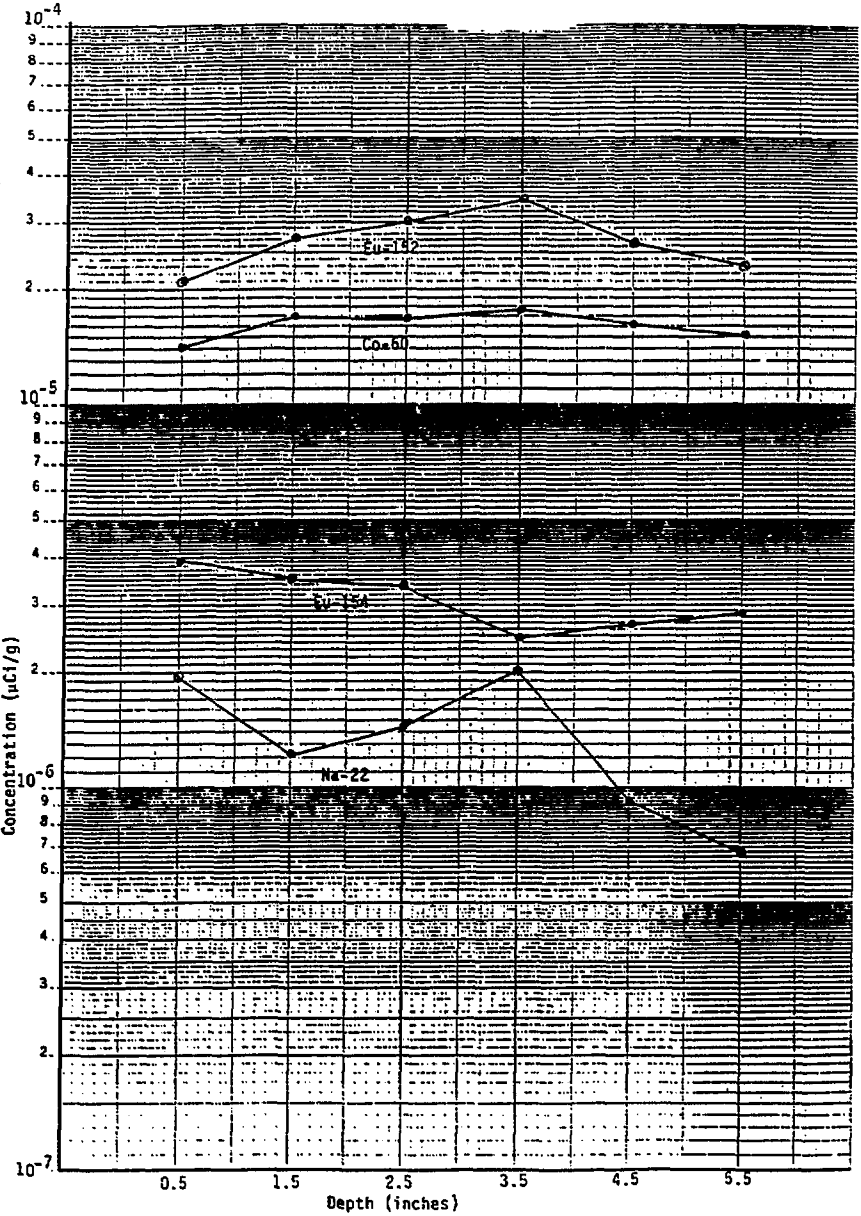

Figure 5.0 Specific radioisotopic concentration versus concrete depth for each radioisotope detected in the PPA concrete core sample [PPA 87].

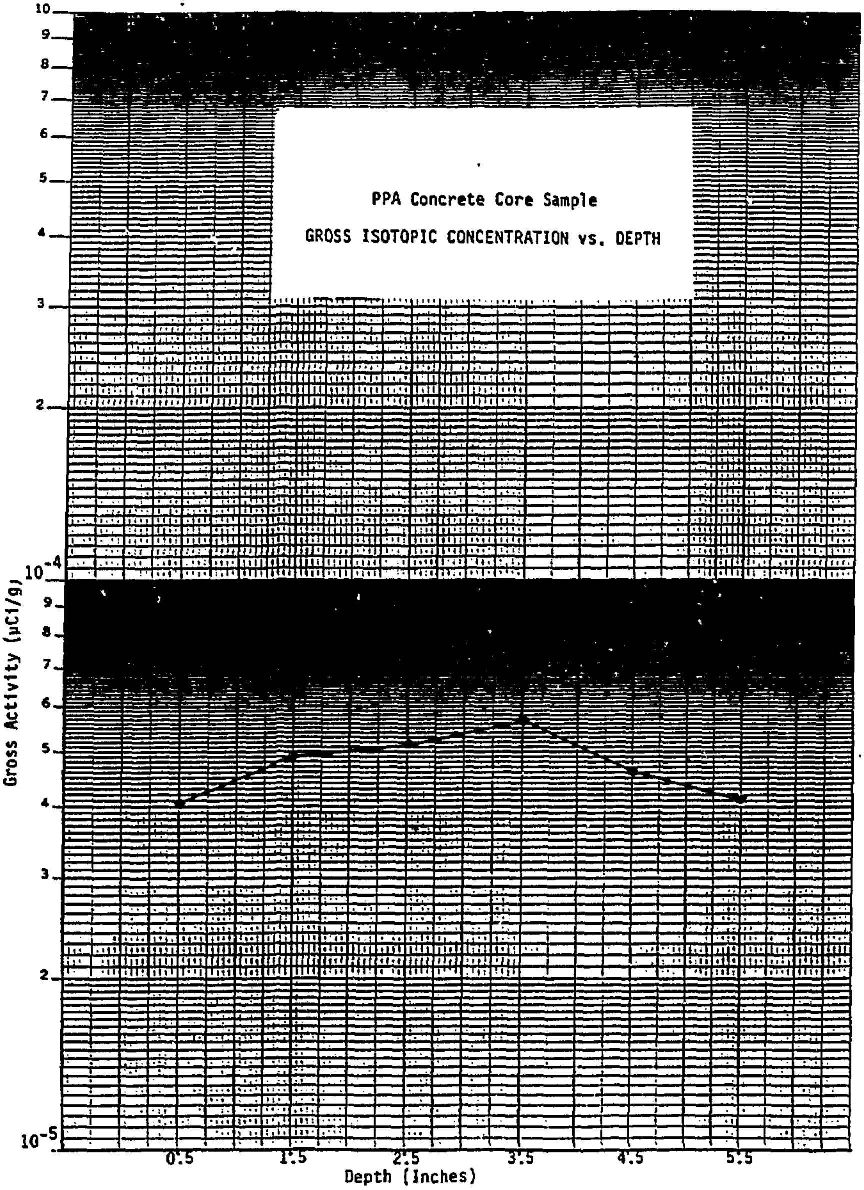

Figure 5.1 Total specific activity versus concrete depth. PPA concrete core sample [PPA 87].

estimates indicates that activity versus depth in Bevatron concrete blocks can be estimated by thermal neutron fluence calculated as a function of depth by a Monte Carlo transport code.

	PPA (based on µCi/g)	Bevatron calculations (based on thermal neutron fluence)
Depth of maximum activity	3.5 inches	2.5 inches
(max activity)/(activity at 1 in.)	1.22	1.10
(activity at 1 in)/(activity at 5.5 in)	1.0	1.0

Table 5.0 Results from Princeton-Pennsylvania Accelerator concrete analysis [PPA 87] compared to the theoretical results based on thermal neutron fluence versus depth in concrete shield.

6.0 Summary

The concrete radiation shielding blocks at the Bevatron facility could contain radioactivity induced by prompt radiation. However, of the approximately 4000 concrete blocks, only a small percentage would have been exposed to enough prompt radiation to generate detectable levels of induced radioactivity. Because of the Bevatron's operating history, only the radionuclides with half-lives greater than 0.5 year are important for present contribution to activity.

Activity in the Bevatron concrete blocks was probably generated by trace elements capturing thermal neutrons. These trace elements are ^{133}Cs,^{59}Co,^{151}Eu, and ^{153}Eu. For blocks composed of normal weight concrete, the radioactive spallation product ^{22}Na could also be present at detectable levels, but at levels insignificant compared to radioisotopes generated by thermal neutron absorption.

The LAHET Code System predicted the thermal neutron flux peaked at about 6 cm (2.5 in) from the surface of the concrete blocks. Using the theory that thermal neutron flux as a function of depth corresponds with the induced activity as a function of depth in the concrete, the activity should also peak at about 6 cm. However, the peak activity is probably only about 10 % greater than the specific activity at the concrete surface based on the thermal neutron fluence profile. The thermal neutron fluence versus depth calculations indicate that the activity is essentially constant from the surface to about 16 cm (6.5 in) with exponential decrease in activity beyond 16 cm. Therefore, if samples are removed from the blocks for quantifying the specific activity, then for practical purposes a concrete sample from near the block surface (~2 cm) will represent the specific activity levels from the surface to about 16 cm.

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