

# PRODUCTION OF RADIOACTIVITY BY PARTICLE ACCELERATORS

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### I. INTRODUCTION

At all accelerators with energies greater than some tens of MeV, induced radioactivity results whenever beams interact with accelerator or beam transport components. Typically these interactions occur at such spots as injection and extraction points and beam splitting stations. Losses at these points are not desirable, and great efforts are often required to reduce them. Beam losses also occur at collimators, scrapers, target areas and beam dumps; these losses are deliberate and cannot be reduced. Consequently, these are usually the most radioactive areas in the accelerator laboratory, and work near them is the largest source of radiation exposures at all laboratories. It is therefore necessary to be able to anticipate the magnitude of the problems involved in such work.

While these loss points are common to all accelerators, the magnitude of the resulting problems depends on many factors unique to each accelerator: The type of particle accelerated, the particle energy, and the geometry and composition of the items being struck. I shall deal with these considerations in turn. What follows is only a general introduction for those not actively involved in this area. The literature should be consulted for details.<sup>1</sup>

### II. MECHANISMS OF ACTIVATION.

A. Proton Accelerators.

The extranuclear hadron cascade process which has been discussed in previous lectures is the process which produces the major fraction of the induced radioactivity at proton accelerators. Each high energy particle which interacts with a nucleus may be absorbed and/or many knock some nuclei out of the struck nucleus. Additional high energy particles may also be created in the collision. If the resulting nucleus is highly excited, it will de-excite by "boiling off" evaporation neutrons. The entire nuclear reaction process is known as "spallation". Each such reaction is called a "star" because of the many secondary particles radiating from it.

The resulting nucleus may be stable or radioactive. The probability, or cross section, of producing a particular nuclide depends on the target nucleus and on the energy of the incident particle. These cross sections are best determined from experimental data; if such data is lacking, an empirical formula of Rudstam gives a good approximation to cross sections which vary over several orders of magnitude.<sup>2</sup>

The particles in the cascade continue to propagate and decay or interact until their energy drops below the threshold for nuclear reactions; this is usually between 10 and 50 MeV. However, for some nuclides, neutron capture is an exoergic reaction which can occur at all energies and has a large cross section for thermal neutrons. (The radioactivation of concrete occurs principally by thermal neutron capture on  $^{23}$ Na to produce 15-hour  $^{24}$ Na.)

The excellent book by Barbier contains information on many cross sections which are relevant to radioactivation and describes how to calculate induced activity levels from such data. I will discuss some details of these calculations later. First I would like to discuss some simple rules of thumb which can be used for most "back of the envelope" calculations.

Rule 1: The dose rate D (R/hr) at a distance r (meters) from a "point source" of gamma rays is given in terms of the source strength S (Curies) and the photon energy  $E_{\gamma}$  (MeV), by<sup>3</sup>

 $\dot{D} = \frac{k\gamma}{r^2} \approx \frac{\Sigma E\gamma}{2.2} \frac{S}{r^2}$ (1)

Rule 2: In many common materials, about 10% of the nuclear interactions produce a radionuclide with a lifetime longer than a few minutes.

Using these rules, we can for instance calculate the dose rate near a target a tenth of an interaction length long. (If it were much longer, we would have to make a large correction for secondary interactions in the target.) Assume a beam of  $10^{11}$  protons/sec has struck the target for several months long enough for many of the radionuclides produced to reach their saturation levels. Of the  $10^{11}$  protons/second incident, one tenth interact and a tenth of the interactions yield radionuclides of interest. The resulting decay rate is  $10^9$ /sec, or  $10^9/3.7 \times 10^{10} = 27$  mCi. (1 Curie  $\equiv 3.7 \times 10^{10}$  disintegrations/second.) Assuming each decay produces a 1 MeV photon, the dose rate half a meter away is

 $\dot{D} = \frac{1 \text{ Mev} \cdot 0.027 \text{ Ci}}{2.2 \cdot (0.5 \text{m})^2} = 0.049 \text{ R/hr} = 49 \text{ mR/hr}$ 

This activity will decay with time in a way which this simple model cannot predict.

Another useful rule relates the total number of stars produced by a single proton in the entire cascade to the incident proton (or hadron) energy:

Rule 3: In a cascade, a proton produces four stars for each GeV of kinetic energy.<sup>4</sup>

Thus a beam of  $10^{12}$  400 GeV protons/sec (= 0.16 µA, or 64 kW) produces a total of 4 x 400 x  $10^{12}$  = 16 x  $10^{14}$  stars/sec in its beam dump. If 10% of the stars yield a radionuclide of any importance, then the total amount of radioactivity in the dump is

$$\frac{(0.1 \text{ dis/star}) \quad (16 \times 10^{14} \text{ stars/sec})}{3.7 \times 10^{10} \text{ dis sec}^{-1} \text{ Ci}^{-1}} = 4 \text{ kCi}.$$

### B. Electron Accelerators

The electron-photon shower, the process whereby high energy electrons interact in bulk matter, is conceptually similar to the hadron shower just described. The principal differences lie in the type of particles which propagate, and in the interactions which produce them. When the initiating electron or photon has an energy greater than several GeV, the predominant interactions are:<sup>5</sup>

1. Bremsstrahlung, in which an  $e^{\pm}$  radiates a photon in the electric field of a nucleus;

2. <u>Pair Production</u>, in which a photon turns into an electron-position pair in the field of a nucleus.

At lower energies - some tens to a hundred MeV, depending on the material - the following processes for reducing the electron or photon energies become important:

3. <u>Electron-Electron Collisions</u> (ionization), in which an incident electron elastically scatters from an atomic electron, resulting in two lower energy electrons;

4. <u>Compton Scattering</u>, in which photons eleastically scatter from atomic electrons, transferring some of their energy to the electrons.

The longitudinal development of the shower is characterized by a quantity known as the "radiation length", X<sub>o</sub>. It is the

distance which the average energy of an electron or photon is reduced by a factor of e. Since the bremsstrahlung and pair production cross sections are proportional to  $Z^2$ , X is approximately proportional to  $Z^{-2}$ . Radiation lengths for common materials range from 35 cm for light elements to 6.5 cm for lead.

The number of electrons and photons in the shower increases exponentially with depth (as  $e^{X/X_0}$ ), with additional electrons and photons being produced by pair production and bremsstrahlung. This continues until the average particle energy is below the "critical energy" for that material; at this point ionization and Compton scattering play a dominant role in removing energy from the shower, and particle multiplication ceases. Because of random variations in each interaction, the cascade dies off only slowly when the mean particle energy reaches the critical energy. The electron shower is therefore characterized by a rapid exponential rise to a maximum, followed by a slower fall.

The shower consists of equal numbers of positrons, electrons, and photons, but only the photons play a significant role in producing nuclear reactions. Most significant are the  $(\gamma, n)$ ,  $(\gamma, 2n)$ ,  $(\gamma, p)$ ,  $(\gamma, pn)$  and  $(\gamma, \alpha)$  photonuclear reactions.

In a shower initiated by an electron of energy E, the number of photons with energies between E and E+dE is

$$dN(E) \propto \frac{E_{\circ}}{E} dE$$

(2)

The total track length g(E) is the total distance travelled by all photons in the shower which have energies between E and E+dE. It is given by<sup>6</sup>

g (E) 
$$dE = 0.57 \frac{E_{o}}{E} X_{o} dE$$
 (gm/cm<sup>2</sup>) (3)

The track length is multiplied by the number of target nuclei per grams and by the reaction cross section to obtain the number of photonuclear reactions initiated by photons of energy E. We then integrate over all possible photon energies to obtain the number of photonuclear reactions in the cascade (the "giant resonance yield"):<sup>7</sup>

$$Y = 0.57 \frac{E_{o}X_{N_{o}}}{A} \int_{0}^{E^{o}} \frac{\sigma(E)}{E^{2}} dE$$
(4)

Two factors combine to allow an approximation to the integral. First, the photonuclear cross sections are dominated by the "giant resonance" phenomenon which occurs between 20 and 50 MeV; other processes occur less frequently. In addition,

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the photon spectrum is a sharply falling function of energy. The denominator ( $E^2$ ) may be taken to be equal to its value ( $E_m^2$ ) at the cross section maximum, and moved outside the integral:

$$Y = 0.57 \frac{E_{o}X_{o}N_{o}}{A E_{m}^{2}} \int_{0}^{E_{o}} \sigma(E) dE$$
(5)

Barbier presents curves of integrated cross sections as a function of target mass for the five photonuclear reactions of interest. His most useful curves are those in which he has combined all the factors in eq. 5 except for the incident electron energy. These are reproduced as fig. 1. To find the number of reactions produced in a shower due to an incident electron of energy  $E_o$ , one multiplies the giant resonance yield (read from the appropriate graph) by  $E_o$ .

As an example, we calculate the total activity created in an iron target (assumed to be 100% <sup>56</sup>Fe) by a 1  $\mu$ A, 20 GeV electron beam (20 kW of power). The most significant reaction is <sup>56</sup>Fe( $\gamma$ ,pn) <sup>54</sup>Mn. The yield for this reaction is  $3\times10^{-6}$  per MeV. The incident beam is

$$\frac{10^{-6} \text{ Coul/sec x 2 x 10^{4} MeV}}{1.6 \text{ x 10^{-19} Coul}} = 1.25 \text{ x 10^{17} MeV/sec}$$

The reaction rate is then

 $Y = 1.25 \times 10^{17} MeV/sec \times 3 \times 10^{-6} / MeV = 3.8 \times 10^{11} sec^{-1}$ .

When the 300-day half life  ${}^{54}$ Mn has been saturated, this will be a total activity of 10 Ci. This is about 1/10 of the activity produced in an iron dump by a proton beam of the same power.

C. Relation between hadron and electron showers.

While hadron and electron showers were just discussed separately, they are in fact connected. In hadron-initiated interactions, about one third of the pions produced are uncharged. These promptly decay into two high-energy photons, which start electron showers. At 400 GeV, about half of the incident proton beam energy is dissipated in the form of electron showers. However, electron showers are much less effective than hadron showers, per MeV of energy, at producing radioactivity, so that this effect may be ignored in hadron cascades. Conversely, the photonuclear reactions which occur in electron showers liberate nucleons from the struck nuclei. However, since most of these reactions occur near 50 MeV, the escaping nucleons do not have much energy. They are therefore not likely to interact further except by neutron capture, and whether this yields a significant amount of radioactivity depends on the specific materials involved.

III. CALCULATIONAL TECHNIQUES; COMPARISON WITH EXPERIMENT.

In order to provide more accurate, and thus more useful information concerning dose rates due to activation, we must have more or less detailed information concerning the cascade source term, nuclear reaction data, and radiological data. By these terms I mean:

cascade source term: information concerning the spatial distribution and spectra of the particles in the hadron or electron shower which produces the radioactivation of the object of interest ("target").

nuclear reaction data: reaction cross sections for various nuclei in the "target" being transformed into other, radioactive nuclei by the particles in the cascade.

radiological data: nuclear lifetimes, decay schemes, transport of  $\beta$ 's and  $\gamma$ 's out of the activated object (i.e., self-shielding) and flux-to-dose conversion factors.

There are several calculational approaches. Alsmiller and co-workers at Oak Ridge use the most involved technique: they Monte Carlo the cascades, including the intra-nuclear details of each reaction, and then carefully calculate the transport of the decay  $\gamma$ -rays out of the material.<sup>8</sup> An example of the results obtained from such a calculation is shown in fig. 2.

A different approach is used by Barbier<sup>1</sup>. He separates the problem into two parts: the user is given the task of determining the flux of activating particles (hadrons or photons), while Barbier provides information on residual dose rate for a unit activating flux. Barbier derives his cross section data from an empirical formula of Rudstam<sup>2</sup> (in the case of spallation reactions), or from his own smooth fits to measured photonuclear cross sections. Experimental values of nuclear lifetimes and decay characteristics are entered into the calculations. Transport of  $\beta$  and  $\gamma$  rays out of the material and conversion to dose rate is done in an approximate analytic way. The results may be presented in terms of what Barbier calls the "danger parameter". This is the dose rate in a cavity inside an infinite volume of

radioactive substance of uniformly distributed activity produced by unit flux (1 particle/cm<sup>2</sup> sec). Curves of danger parameters for different materials are shown in fig. 3, which are similar to conventional cooling curves. The danger parameter is a function of irradiation and cooling time because isotopes with different lifetimes will saturate and decay differently with time.

We can use one of these curves to determine the dose rate in a real situation involving a thick source by using the following relation:

$$\dot{D} = \frac{\Omega}{4\pi} \cdot \Phi \cdot d$$

The dose rate at any point ( $\dot{D}$ ) is obtained by multiplying the danger parameter (d) by the fractional solid angle the source subtends when seen from the point of interest ( $\Omega$ ), and by the activating flux ( $\Phi$ ) at the surface of the object.<sup>9</sup> (The activity several  $\gamma$  absorption lengths into the body does not contribute much to the external dose rate because of selfshielding by the activated object.)

It is possible to obtain the hadron flux at the surface of the object from a Monte-Carlo calculation run for that purpose, or from a collection of "standard" cases.<sup>10</sup> If this is done, one must be careful because many Monte Carlo programs have a low-energy cutoff, below which they cease to follow particles. Since this cutoff may be higher than the thresholds of many activation reactions, use of the "flux" predicted by the Monte Carlo calculation will give an erroneously low value for the activation. Correction for this effect may be made if the ratio of the true flux to the "Monte Carlo flux" is known. This ratio is material-dependent, and to the extent that the spectrum has not reached an equilibrium shape, it is position dependent as well.

Rearranging the factors:

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$$D(t_i, t_c) = \Sigma_{\mu} A_{\mu} \{ \exp(-t_c/\tau_{\mu}) - \exp(-[t_i+t_c]/\tau_{\mu}) \}$$
$$= D(\infty, t_i+t_c)$$

Thus the dose rate sought is the difference between the dose rates for the case of an infinite irradiation time and two hypothetical cooling times: one equal to the actual irradiation time, the other equal to the actual irradiation plus cooling times. It is far easier to make predictions of induced radioactivity than it is to verify those predictions, thus experimental data is hard to come by. Figure 4 shows measurements and calculations made by Barbier and Cooper at CERN.<sup>11</sup> The average difference between prediction and measurement is about a factor of two. Other comparisons show similar agreement. When known cross sections and thin targets are used, the agreement is even better.<sup>12</sup>

#### IV. APPLICATIONS

Life is not really as simple as one might believe from what I have said up until now. First, calculations by different authors disagree with each other, often by factors of two. Second, real life sometimes does not reproduce any author's calculation. (Fortunately, things seem to cool off faster in real life than they do in calculations.) Figure 5 shows a calculation from Oak Ridge, with a number of measured curves added. The curve labelled "accel" is based on the measured cooling of the average dose rate around our 6-km main ring. Our accelerator has been operating for 3 years with essentially constant losses, thus the curve ought to be comparable to the one labelled "one year"; but it falls about a factor of 3 below it for long cooling times.

The curve labelled "neutrino" represents the decay of an area near the target of our dichromatic (narrow band) neutrino target train after a run of 8 months. The beam intensity varied by a factor of about two over this period. Again, the components cool much faster than one would predict.

Finally, the curve marked "AGS" represents the dose rate near the AGS slow external beam splitter.<sup>13</sup> This unit has been operating for many years, and its decay agrees reasonably well with the predictions for long irradiations. The most obvious explanation for the disagreement of observations and calculations is that possibly a lot more short-lived activity than is expected (or a lot less long-lived activity) is actually being produced. I have no data to substantiate or refute this conjecture, however.

As can be seen from the various graphs of the danger parameter, different materials vary widely in their relative hazards due to activation. Aluminum is preferred to iron because of its lesser activation - the only major long-lived activity produced from aluminum is  $^{22}Na$ . For shielding purposes,  $CaCO_3$  marble - is excellent for the same reason; in this case the cross section for  $^{22}Na$  production from calcium is even lower. Elements above calcium, however, are capable of readily producing longlived activity.<sup>14</sup>

Sodium, on the other hand, is notorious for causing problems because of its high thermal neutron capture cross sections, producing <sup>2</sup> Na. The effects of even small amounts of sodium in concrete have been studied extensively both theoretically and experimentally.<sup>15</sup> Briefly, concrete containing one percent of sodium by weight produces enough <sup>2</sup> Na activity to approximately double the radiation level in the machine enclosure the first day after machine turn-off. Since the major source of sodium in concrete is the aggregate, a proper choice of aggregate, such as limestone, can eliminate this problem.

Another subject which has been studied at some length is the question of radioactivation of air in target and accelerator enclosures.<sup>16</sup> It has been our experience, and that of others, that because of the short half-lives involved (20 min <sup>11</sup>C, is the longest) there is no internal exposure hazard. There is an <u>external</u> exposure hazard, but that is smaller than, and no different from the hazard due to radioactivated accelerator components in the same area.

Both proton and electron accelerators produce radioactivity in water when their respective showers cross water cooling paths in beam dumps. Both types of machines are capable of producing many kilocuries of short-lived activity. The SLAC accelerator has a water beam dump, and in fact produces such large activities.<sup>17</sup> Proton accelerators, on the other hand, have less beam power, and their dumps have only small water channels. As a result, our water activity problems are much less severe.

Under normal circumstances, the radioactivation of the water which cools targets and beam dumps presents several distinct problems. The first is gamma exposure from short-lived radionuclides in the water as it passes through pipes and heat exchangers. At SLAC there are dose rates of many R/hour near such items; the corresponding levels at Fermilab are tens of mR/hour. The second problem is the deposition of <sup>7</sup>Be in the deionizing columns needed to maintain the water resistivity. These deionizers will also collect radioactive corrosion products from piping and targets. Besides being sources of external exposure, the activity in these columns must be disposed of in an appropriately safe way. The third problem is that the build-up of large quantities of tritium in the water presents a possible hazard in case of water leaks. These hazards may be minimized and confined by having separate, small volume cooling systems for target systems and beam dumps, by insisting on high quality piping, etc., and by periodically draining the water from the cooling loops for disposal under controlled circumstances.

Where earth is used for hadron shielding it will become activated. This may present difficulties if it is later planned

Our experience has been that soil activation may be a problem only around the most radioactive target areas. My own rule of thumb is that soil activation need be considered as a possible source of groundwater contamination only for those objects which are so radioactive that they cannot be easily serviced by hand. Since this problem is of limited applicability, I will not dwell on it here; the literature should be consulted for further details.

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Fig. 1 Giant resonance yield per electron per MeV of energy for fine photo nuclear reactions. (From Ref. 1.)



Fig. 2 Contribution of various radionuclides to the total photon dose rate outside a 40.6 cm radius iron beam stop after an infinite irradiation by 200 GeV protons. From T. A. Gabriel and R. T. Santoro, ORNL-TM-3945 (1972).



Fig. 3. "Danger parameter" for various materials used in accelerator construction. From Ref. 1



Fig. 4 Measured and calculated cooling curves for several different materials. From T. W. Armstrong, et al., ORNL-TM-2498.



Fig. 5. Calculated cooling curves for various irradiation times for iron struck by high energy protons. (From T. W. Armstrong, et al., ORNL-TM-2498). The curve labeled "accel" is the measured average cooling curve for the Fermilab main ring after three years of operation. The curve labeled "neutrino" is for a neutrino target train after eight months of use. The curve labeled "AGS" is for an extraction splitter in use for many years at the BNL AGS.