RADIOACTIVITY PRODUCED BY IRRADIATING WATER AT HIGH ENERGIES

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Cooling water used in accelerators becomes radioactive due to the spallation of oxygen. This can become a hazard due to the resulting radiation levels near water pipes and heat exchangers and from accidental spills and subsequent contamination of the biosphere. These problems were examined by DeStaebler during the design phase of the Stanford Two-Mile Linear Accelerator. Of particular interest was the radioactivity produced in the beam dumps because of the very high power absorbed in the water. In this report we will summarize some measurements taken on the cooling water during accelerator operation.

Based on DeStaebler's results, Coward calculated for a beam dump in which all the beam is absorbed in water such as Beam Dump A or Beam Dump East, a 1 MW beam would produce saturation activities as follows:

<table>
<thead>
<tr>
<th>Daughter Nuclide</th>
<th>Curies</th>
<th>Half-Life</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{150}$O</td>
<td>35,000</td>
<td>124 seconds</td>
</tr>
<tr>
<td>$^{13}$N</td>
<td>1,390</td>
<td>10 minutes</td>
</tr>
<tr>
<td>$^{11}$C</td>
<td>1,390</td>
<td>20.4 minutes</td>
</tr>
<tr>
<td>$^{7}$Be</td>
<td>260</td>
<td>53.6 days</td>
</tr>
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</table>
In addition there will be a slow buildup of tritium ($T_1 = 12.3$ years) in the water. The buildup of tritium will depend in part on the rate of evaporation or leakage of water and the subsequent addition of makeup water. This activity is initially distributed throughout a water system consisting essentially of 2500 gallons of water passing in series through the beam dump, a heat exchanger and a surge tank. In its present form the water is open to the atmosphere at the surge tank. A bypass demineralizer utilizing a mixed bed ion exchange resin is attached at all times. Because of the expected high radiation levels the surge tanks and heat exchangers are contained in shielded rooms. The room is so designed that any water spilled will drain into the BSY housing and eventually collect in sumps. Very little was known of the chemical form of the radioactive elements since many reactions are possible in the highly ionized environment.

$^7$Be was expected to be the major problem in an accidental spill because of its long half-life. In early operation of the first two sectors of the accelerator, we found that $^7$Be was collecting in a demineralizer used to cool a temporary dump. The $^7$Be was identified by $\gamma$-ray spectroscopy. Measurements of water samples from the cooling loop and of the demineralizer indicated that the demineralizer had removed $> 99.7\%$ of the $^7$Be. The removal may depend on the $pH$ of the water which is 6 - 7 in our system. Within the accuracy of our figures ($\pm 30\%$), the amount of $^7$Be agreed with the prediction of Coward. Subsequent measurements have confirmed that $^7$Be is, indeed, very efficiently removed by the demineralizer and can probably be ignored as a contaminant to the biosphere.
Our next measurements were made on the cooling water system for the tune-up dump with a 7 GeV beam. This dump does not absorb all the beam in water. The fraction absorbed in water was calculated from shower data and our measurements corrected accordingly. We had available a demineralizer which could be operated in full flow or by-passed and turned off. We measured the γ-ray spectrum of the water with a scintillation spectrometer both in the demineralizer and in the water pipe itself. No γ-rays could be seen except for the 0.511 MeV annihilation peak. Any other γ-rays present were down in intensity by at least several orders of magnitude. All of the expected isotopes except ⁷Be are positron emitters with no γ-rays, so identification could be made only by half-life measurements on the annihilation γ-rays. For this purpose a NaI scintillator and an RCL 400 channel analyzer were used in the multiscaler mode. The largest counting time available in this mode was 0.9 seconds per channel or 6 minutes total. This was really too short for good identification of the half-lives involved and was supplemented with ion chambers driving chart recorders. The scintillator was superior in that it could be heavily collimated and was used to look at a single portion of the water system, such as a resin column or a pipe. Figure 1 shows the decay curve expected from the levels of radioactivity predicted by Coward, i.e., the ratio \( \frac{^{150}O}{^{13}N/^{11}C} = 35/1.4/1.4 \). For the first 6 or 8 minutes it gives an apparent half-life of about 140 seconds. Measurements were made at periods when the beam had been off for a long time, and the activity in the water was negligible. When the beam came on, the demineralizer was immediately
allowed to fill for about 30 seconds and then closed off to hold the water sample. The beam was then turned off again. The ion chamber which looked at the entire demineralizer gave the decay curve shown in Figure 2 by the crosses with two attempts made to fit an equation to it of the form

\[ \text{ACTIVITY} = Ae^{-\lambda_1 t} + Be^{-\lambda_2 t} + Ce^{-\lambda_3 t} \]

when \( \lambda_1, \lambda_2, \) and \( \lambda_3 \) are the decay constants of \( ^{15}O, \) \( ^{13}N, \) and \( ^{11}C \) respectively. The best fit was obtained when \( B \) equals zero and the ratio \( ^{15}O/^ {11}C = 35/4.1 \). Also shown is a calculation using ratios of \( ^{15}O/^ {13}N/^ {11}C \) of 35/0.78/3.77 to show the effects of small admixtures of \( ^{13}N \). This measurement cannot be compared directly with Coward's calculation since:

1. Coward calculated the saturation activity. We are measuring water which spent only a short time in the tune-up dump.
2. The water we measured had decayed during the transit time out to the service areas. We measured this time to be about 1\( \frac{1}{2} \) minutes. It would take about \( \frac{1}{2} \) minute to fill the demineralizer tank, so this water has had \( \sim 2 \) minutes decay time at the start of our measurement.

The capacity of the tune-up dump is \( \sim 5 \) gallons. Since the flow rate that night was 26 gallons a minute, the water had an average dwell time of only \( \sim 1/5 \) minute in the tune-up dump. Using the notation of DeStaebler\(^5\), the activity of each isotope would be:

\[ \frac{dn}{dt} = R \left( 1 - e^{-T/t} \right) e^{-t/\tau} \]

where

\( R = \) creation rate = saturation activity
\( T = \) irradiation time
\( t = \) decay time
\( \tau = \) mean life = \( \frac{1}{\lambda} = \frac{T^{\frac{1}{2}}}{.693} \)
For our situation \( T \ll \tau \)

so we can rewrite the above equation as:

\[
\frac{\mathrm{d}n}{\mathrm{d}t} = R \left( \frac{\tau}{t} \right) e^{-\frac{t}{\tau}}
\]

- for \(^{15}O\), \( \frac{\mathrm{d}n}{\mathrm{d}t} = R(15O) \times 0.0342 \)
- for \(^{13}N\), \( \frac{\mathrm{d}n}{\mathrm{d}t} = R(13N) \times 0.0121 \)
- for \(^{11}C\), \( \frac{\mathrm{d}n}{\mathrm{d}t} = R(11C) \times 0.0064 \)

With these relationships we can correct our measured ratio of \(^{15}O/11C\) = \( 35/4.1 \) as follows to obtain the ratio of the saturation activity.

\[
\frac{R(15O)}{R(11C)} = \frac{35}{4.1 \times \frac{0.0342}{0.0064}} = \frac{35}{21.9}
\]

This is far higher than one would expect. There are several possible explanations when we remember that nothing is known so far of the absolute magnitude of the activity, but only the ratios of the \(^{15}O/11C\). Some possibilities are as follows:

1. The cross sections for \(^{15}O\) production are much lower than expected or, conversely, those for \(^{11}C\) production are much higher than expected.
2. The \(^{15}O\) is not getting to our measurement site because of formation and holdup of gaseous oxygen or reaction of the oxygen with the pipes.
3. A nonuniform distribution of activity in the demineralizer.
4. A combination of all of these.
It will not be possible to obtain a better answer to this question until an experiment can be performed under better conditions.

The scintillator and a collimator with a resolution of about 1" in the center of the demineralizer was used to measure the vertical distribution of activity in the demineralizer as shown in Figure 3. The sharp drop-off of the activity at ~16" corresponds to the level of the resin bed. The relatively higher activity near the top of the demineralizer while the beam is on may indicate a small amount of gaseous oxygen in the top of the tank. The relatively higher activity in the lower portion of the demineralizer when the beam was off, may indicate a small amount of holdup of long-lived activity in the resin. These possibilities were supported by measurements of decay rates with the multiscaler. We obtained apparent half-lives of 150 seconds, 225 seconds and 140 seconds at the top, middle and bottom of the demineralizer respectively as shown in Figure 4. The dose rate at the surface of the water pipes was reduced to about 60% by the insertion of the demineralizer. This is not enough to be of practical value and might be of only a temporary nature due to, e.g., an oxidation phenomenon on the resin.

Another series of measurements was taken on the surge tank at A beam dump. It was desired to know the quantity and composition of the radioactive gases released at that point. The surge tank on this dump received the full flow of water. The water splashes over a perforated cone so there is ample opportunity for release of dissolved gases. The surge tank is vented to a vertical pipe with 1200 cu. ft./min. of air
being blown through it. It was discovered that this blower exerted a slight pressure on the surge tank tending to prevent the escape of gases. A purge line was connected to the surge tank to allow a small flow of air to overcome this pressure. This same arrangement was used by other groups studying hydrogen evolution in the water system.

The isotopes expected in the water, i.e., $^{15}$O, $^{13}$N, and $^{11}$C are all short-lived positron emitters and their Maximum Permissible Concentration (MPC) is determined by the immersion dose. The concentration in a semi-infinite cloud which will give 30 mrem in a 40 hour week has been calculated by several authors\textsuperscript{9-10} to be about $4 \times 10^{-7}\mu$Ci/cc. In practice, the dose rate from this concentration will always be lower because of non-homogeneous clouds, cavities caused by buildings, etc. To determine the situation in the end station area we have examined the diffusion of the plume from the 1200 CFM blower with smoke bombs under nighttime conditions (the night is the worst because of the prevalence of atmospheric inversions). We did this with the wind from the west at about 9 mph, from the south at about 9 mph, and in a nearly dead calm. In all cases, the smoke rose to about the roof level of the end stations and then dropped back down filling the end station area. At the same time we made studies of the concentration of radioactive gas in the air in the area between the end station and the corresponding dose rates. The radioactive gas concentration was made with a standard air monitor—a lead shielded tank containing a thin-walled G-M counter. Dose rates were measured with an ion chamber.
These measurements indicated that a concentration of $3 \times 10^{-5}$ μCi/cc would give 30 mrem in a 40-hour week. Because of the geometrical consideration above, this relationship will vary with position and can be considered only a rough guide.

Placing the air monitor so that it sampled the air coming directly out of the 1200 CFM blower stack enabled us to measure the concentration and total activity coming from the surge tank. For higher concentrations, however, it was necessary to replace the G-M counter with a flow through ion chamber. It was also possible to close off the pump to the air monitor and observe the decay rate of the trapped sample. This showed that the only significant components were $^{150}$ and $^{11}$C in the ratio 4/1. Some representative values are shown in the table below.

**TABLE II**

<table>
<thead>
<tr>
<th>Beam Power KW</th>
<th>(Total) Stack Concentration μCi/cc</th>
<th>*$^{11}$C (Only) Concentration in Stack μCi/cc</th>
<th>(Total) Concentration on Pad Between End Stations μCi/cc</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.6</td>
<td>$4.5 \times 10^{-3}$</td>
<td>$9 \times 10^{-4}$</td>
<td>$1.3 \times 10^{-5}$</td>
</tr>
<tr>
<td>10.8</td>
<td>$2.4 \times 10^{-2}$</td>
<td>$5 \times 10^{-3}$</td>
<td>$7.2 \times 10^{-5}$</td>
</tr>
<tr>
<td>21.6</td>
<td>$5 \times 10^{-2}$</td>
<td>$1 \times 10^{-2}$</td>
<td>$2.0 \times 10^{-4}$</td>
</tr>
</tbody>
</table>

* The $^{11}$C concentration was determined from half-life curves.
It is apparent that we would be limited to about 8 kW in order to keep radiation levels within SLAC limits. For a solution to this problem, there appeared to be three approaches as follow:

1. Build a high stack to dissipate the exhaust gases.
2. Seal the system and install a catalytic recombiner to get rid of the evolved hydrogen.
3. Store the exhaust gases and allow the radioactive component to decay before venting them.

The first approach was ruled out because of the probable opposition to an unsightly high stack. The third approach is impractical if it is necessary to allow the 20.4-minute $^{11}$C to decay but might work if the $^{11}$C were in a chemical form that could be removed in some manner and allowed to decay in a condensed state. Several experiments were performed where the exhaust gases could be pulled through a fritted glass bubbler containing various liquids. We had capabilities for measuring the concentrations before and after the bubbler and for observing the decay of the activity trapped in the bubbler. The liquids used in the bubbler were as follows:

**TABLE III**

<table>
<thead>
<tr>
<th>Liquid</th>
<th>Gases Trapped</th>
</tr>
</thead>
<tbody>
<tr>
<td>KOH</td>
<td>$\text{CO}_2$</td>
</tr>
<tr>
<td>Cosorbent</td>
<td>CO</td>
</tr>
<tr>
<td>Potassium Pyrogallol</td>
<td>$\text{O}_2$, unsaturated hydrocarbons</td>
</tr>
</tbody>
</table>
In addition we tried passing the gases through Drierite columns to remove any activity carried in water vapor. The results indicate that substantially all of the $^{11}$C activity is in the form of CO$_2$. Substantially all of the oxygen can be trapped in potassium pyrogallol solution and is probably in the form of oxygen gas, although we cannot be certain that some portion is not in the form of unsaturated hydrocarbons. No measurable activity was trapped on the Drierite.

Based on these data, design studies were made for both recombiner and hold-up systems. We can estimate the degree of reduction needed from the data in Table II. If 21.6 kW gives $2 \times 10^{-4}$ $\mu$Ci/cc, at 1 MW on the Research Pad we will have about $10^{-2}$ $\mu$Ci/cc. Using the calculated MPC of $4 \times 10^{-7}$ $\mu$Ci/cc, we need a reduction factor of $10^5$ assuming our measurements were made under reasonably bad inversion conditions. Using our measured MPC of $3 \times 10^{-5}$ $\mu$Ci/cc, we would need a reduction factor of only $3 \times 10^2$.

It is interesting to note that at 1 MW we would be venting about 80 Ci/min total activity. A CO$_2$ trap would build up to an equilibrium activity of 320 Curies and would give a dose rate of about 43 rem/hr at 10 feet.

Since these measurements were performed, the decision was made to install a catalytic hydrogen recombiner on the high power water systems. With the danger of hydrogen explosions removed, it has been possible to seal these systems and allow the $^{11}$C and $^{15}$O to decay without reaching the atmosphere.
REFERENCES


LIST OF FIGURES

Figure 1 -- Decay of Predicted Radioactive Products of Irradiated Water.

Figure 2 -- Measured Decay of Radioactive Products of Irradiated Water.

Figure 3 -- Distribution of Radioactivity in a Demineralizer.

Figure 4 -- Decay of Activity in Various Segments of a Demineralizer.
FIG 2

X X MEASURED POINTS
O O $^{15}{O}/^{13}{N}/^{12}{C} = 35/0.78/3.77$
\(\Delta\) \(\Delta\) $^{15}{O}/^{11}{C} = 35/4.1$

ACTIVITY—ARBITRARY UNITS

TIME—MINUTES