

RADIOACTIVITY PRODUCED AND RELEASED FROM WATER AT HIGH ENERGIES*

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Introduction

Water is used to cool accelerator components, beam transport components and beam dumps at SLAC. The accelerator is composed of thirty sectors. Each sector has three cooling water systems: accelerator, klystrons and waveguides. Two of these systems, accelerator and waveguide, provide water to areas where activation is possible. There are six cooling water systems for the research and beam switchyard areas. These systems supply water for magnets, collimators, targets and beam dumps. All of these systems supply water to areas where activation is probable.

The primary isotopes formed in water are ^{15}O , ^{11}C and ^7Be . ^{15}O and ^{11}C are positron emitters with no X-ray emission. ^7Be is an electron capture isotope with a 0.478 MeV X-ray 10% of the time. The radioactivity presents operational problems due to the high radiation levels near the water pipes and heat exchangers. The beam transmission through the accelerator is excellent so there is little water activation. We typically find radiation levels less than 0.5 mr/hr at these heat exchangers which are unshielded. Higher levels (4 mr/hr) are found near the accelerator analyzing stations. Water systems for cooling the positron source produces levels as high as 400 mr/hr with a 100 kW incident electron beam on the e^+ source. This system is shielded. The radiation levels near surge tanks and heat exchangers for high power dumps are very high (120 R/hr with 170 kW beam). These systems are shielded and locked. The surge tanks for high power cooling water systems are sealed to prevent the release of gaseous radioactivity to the atmosphere.

There are some power absorbing devices on an unshielded heat exchanger. The activated water travels through a 6" diameter pipe for 500 feet. We typically measure 30 mr/hr at 4 feet with a 7 kilowatt incident beam. Since radiation levels fall proportional to the reciprocal of the distance rather than distance squared the pipe represents an infinite line source.

Radioactivity Produced in Water

During the design phase of SLAC, DeStaebler examined the radiation problems associated with radioactivity produced in water.¹⁻⁴ Coward,⁵ using DeStaebler's results, calculated the saturation activity produced by a one-megawatt electron beam absorbed

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in a water beam dump (Table 1).

TABLE 1

<u>Nuclide Produced</u>	<u>Activity (Curies)</u>	<u>Half-Life</u>
^{15}O	35,000	124 sec
^{13}N	1,390	10 min
^{11}C	1,390	20.4 min
^7Be	280	53.6 days

In addition to the isotopes listed in Table 1 there will be a slow buildup of tritium which will depend in part on the rate of evaporation, leakage from the system, and the subsequent addition of makeup water. A slow buildup of tritium in the water systems for the high power dumps has been observed. Concentrations of $3 \times 10^{-2} \mu\text{Ci/cc}$ have been measured.

There are several other isotopes which are produced, but they are not considered here since their half-lives are so short, e. g., ^{14}O , ^{16}N . These have been identified in a special experiment at SLAC.⁵ The water was transferred quickly from the irradiation point to the measuring point (a few seconds elapsed time). At SLAC, these isotopes are not important, because it takes several minutes for the water to reach the outside of the shield.

^7Be was expected to be the major problem in an accidental water spill because of its 53-day half-life. During the sector test period, ^7Be was found collecting in a demineralizer. The isotope was identified by gamma spectroscopy and water samples showed that more than 99% had been removed by the demineralizer.⁷ Later measurements by Busick⁸ confirmed that ^7Be is very efficiently removed by mixed bed resins. This reduces the problem to one of handling the resin containing radioactivity, since periodic regeneration is required.

The other isotopes (^{15}O , ^{13}N and ^{11}C) are short-lived positron emitters and present an external radiation problem due to the 511 keV annihilation photons. Since ^7Be is effectively removed by the demineralizer and the ^{15}O half-life is so short, the ^{11}C activity will determine the time before an area can be entered and the water system repaired.

During the beam pulse (in addition to radioactivity), high concentrations of free radicals are produced in the irradiated water. These radicals recombine to form H_2 , O_2 , H_2O_2 and others. The gases are released from the water in the surge tanks and the hydrogen concentration can reach an explosive level if means of removal are not provided. Two methods of removing hydrogen were considered: (1) sealing the surge tank and adding a catalytic recombiner or (2) using a purge tank and delayed exhaust to allow ^{15}O to decay or separate ^{15}O and ^{11}C from the air chemically.

In the remainder of the paper we will discuss the measurements used to determine the feasibility of method No. 2. The important questions are: what is the $^{15}\text{O}/^{11}\text{C}$ ratio in exhaust gases and what is the gross concentration? If the ^{15}O activity is many orders of magnitude higher than ^{11}C a simple holdup method could be used. If ^{11}C concentration is high then it would require a long holdup and chemical separation would have to be used.

$^{15}\text{O}/^{11}\text{C}$ Ratio in a Water Dump

The beam enters the dump and travels through 10 radiation lengths of water. Water-cooled copper plates absorb the remaining power. Figure 1 shows a schematic flow diagram for the water system. Water flows from the dump into a surge tank for out-gassing,

then it goes to the heat exchanger and back to the dump. A small fraction of the water is bypassed through a demineralizer. The dump contains 2000 gallons of water. The flow rate through the dump is 400 gal/min. This gives an overall average dwell time of five minutes for the water inside the dump. Figure 2 shows a decay curve for activity in the heat exchanger after an erratic high power run. The power level was reduced and held fairly constant at 40 kW for an hour before the beam was turned off. The travel time from the dump to the heat exchanger is calculated to be about one minute, using the pipe diameter and flow rate.

The activity of each isotope can be expressed as

$$-\frac{dn}{dt} = R(1 - e^{-T/\tau})e^{-t/\tau} \quad (1)$$

where

R = production rate = saturation activity
 T = irradiation time
 t = decay time
 τ = mean life

The ratio from Fig. 2 is 9/1. This ratio must be corrected for irradiation time and decay time to give a 13/1 ratio of saturation activities. The ratio is a factor of two lower than predicted by Coward. However, this value of 13/1 agrees well with the ratio of 12/1 for the absolute yields of ^{15}O and ^{11}C reported by Gorbunov et al.⁹ Kramer et al.,¹⁰ has reported a ratio of 14/1 for integrated cross sections for photoproduction of ^{15}O and ^{11}C up to 65 MeV. V. Di Napoli et al.,¹¹ has reported a 14/1 ratio for the (γ, n) to $(\gamma, 2n3P)$ reactions in ^{12}C from 300 MeV to 1 GeV. The ratio production from ^{16}O is probably similar to that from ^{12}C .

$^{15}\text{O}/^{11}\text{C}$ Ratio in a Copper-Water Dump

The $^{15}\text{O}/^{11}\text{C}$ ratio was measured in a water-cooled copper dump. A demineralizer, which could be used full flow or bypassed and turned off, was the sampling vessel. Figure 3 shows the decay curve and two attempts to fit a curve of the form

$$Ae^{-\lambda_1 t} + Be^{-\lambda_2 t} + Ce^{-\lambda_3 t}$$

where λ_1 , λ_2 , and λ_3 are the decay constants of ^{15}O , ^{13}N and ^{11}C , respectively. The best fit is $A = 35$, $B = 0$ and $C = 4.1$.

We have not identified ^{13}N in water. Bishop¹² has found a ^{13}N production cross section of 1.2 MeV mb. The cross section for ^{11}C production reported by Kramer et al., is 3.4 times larger. This makes it difficult to separate as shown in Fig. 3.

The capacity of the dump is five gallons and flow rate is 26 gal/min. The travel time from the dump to the detector is measured at 1.5 minutes. Equation (1) can be used to express the activity of each isotope. Since the irradiation time is short, the equation reduces to

$$-\frac{dn}{dt} = R\left(\frac{T}{\tau}\right)e^{-t/\tau} \quad (2)$$

Then the measured ratio of 35/4.1 can be corrected to give a ratio of 1.6/1 for the saturation activities. This is much lower than expected from calculations and measurements on the water dump, previously discussed. The weak point here is not knowing how the isotopes will behave in a highly ionized environment. One can postulate ^{15}O being held in gas pockets or reacting chemically with the copper plates and pipes. This would require a six-minute holdup, to adjust the ratio to that measured in the water beam dump. This is an unusually long time. At present we cannot explain the difference between the two systems. Further measurements will be needed to explain this point.

The flow diagram is shown in Fig. 1. The water flows directly from the dump to the surge tank where the water splashes over a perforated cone for out-gassing. The tank is vented to a vertical pipe where the gases are mixed with 1200 ft³/min of air and exhausted to the atmosphere. Table 2 shows the vent concentration measured at three power levels by exhausting an air sample through an ionization chamber. Also shown in Table 2 is the concentration measured in the research yard with a portable G. M. gas sampler.

TABLE 2

Beam Power kW	(Total) Stack Concentration $\mu\text{Ci/cc}$	* ¹¹ C (Only) Concentration In Stack $\mu\text{Ci/cc}$	Total Concentration In Research Yard $\mu\text{Ci/cc}$
3.6	4.5×10^{-3}	9×10^{-4}	1.3×10^{-5}
10.6	2.4×10^{-2}	5×10^{-3}	7.2×10^{-5}
21.6	5×10^{-2}	1×10^{-2}	2.0×10^{-4}

* The ¹¹C ratio was determined from half-life curves.

The water inside the beam dump will be in a highly ionized state and the newly formed carbon 11 atoms will have an opportunity to combine with oxygen and hydrogen atoms to form hydrocarbons and carbon dioxide. The surge tank is open to the atmosphere so the water is probably saturated with CO₂ and O₂. The CO₂ and O₂ will act as a carrier for the ¹¹C and ¹⁵O activity that may be in the form of CO₂ and O₂. There are standard chemicals for selectively absorbing CO₂, O₂ and CO from air.¹³ These chemicals can be used as scrubbers to reduce the activity released to the atmosphere. Tests were conducted to establish which chemicals would significantly reduce the gaseous radioactivity. The chemicals used are listed in Table 3 along with the gases trapped and isotopes identified in each medium. Figure 4 shows a schematic diagram of the equipment used in these measurements. We measured the concentration before and after the scrubber and observed the decay rate of the activity in each scrubber.

TABLE 3

Collecting Medium	Gases Trapped	Isotopes Detected
KOH	CO ₂	¹¹ C
Potassium Pyrogallol	O ₂	¹⁵ O
Cosorbent	CO	Traces of ¹¹ C
Drying Agent	H ₂ O	Traces of ¹⁵ O

The efficiency for trapping ¹¹C by KOH can be calculated using ¹⁵O/¹¹C ratio from Figs. 5 and 6. In order to calculate the efficiency, we assumed that the fraction removed by the scrubber is independent of concentration. The ratio of ¹⁵O/¹¹C before the scrubber is 4/1 (Fig. 5) and after the scrubber is 56.5 (Fig. 6). By adding the two scrubbers containing KOH, the travel time was also increased by one minute therefore ¹⁵O concentration must be corrected for this decay time.

The equation for the ¹⁵O/¹¹C ratio after the scrubber is

$$\frac{{}^{15}\text{O out}}{{}^{11}\text{C out}} = \frac{{}^{15}\text{O in } e^{-\lambda t}}{(1-F)^2 {}^{11}\text{C in}} \quad (3)$$

where

- $^{15}\text{O in} = ^{15}\text{O concentration before the scrubber}$
 $^{11}\text{C in} = ^{11}\text{C concentration before the scrubber}$
 $^{15}\text{O out} = ^{15}\text{O concentration after the scrubbers}$
 $^{11}\text{C out} = ^{11}\text{C concentration after the scrubbers}$
 $F = \text{fraction of } ^{11}\text{C removed in each scrubber}$
 $\lambda_1 = \text{decay constant for } ^{15}\text{O} = \frac{0.693}{T_{1/2}}$
 $t = \text{travel time through scrubbers}$

$$\frac{180}{3.2} = \frac{4 \text{ } ^{11}\text{C in} e^{-\lambda t}}{(1-F)^2 \text{ } ^{11}\text{C in}} \quad (4)$$

$$F = 0.78$$

78% of the ^{11}C is removed by each scrubber containing KOH. Also the efficiency for trapping ^{11}C can be calculated from the ratio of the activities trapped in the scrubbers. The following equation can be written:

$$\frac{A_1}{A_2} = \frac{F(^{11}\text{C in})}{F(1-F)^2 \text{ } ^{11}\text{C in}} = \frac{1}{1-F} \quad (5)$$

where

- $A_1 = ^{11}\text{C activity in scrubber one} = 3300 \text{ c/6sec}$
 $A_2 = ^{11}\text{C activity in scrubber two} = 800 \text{ c/6sec}$
 $F = \text{fraction of } ^{11}\text{C removed in each scrubber}$
 $\frac{1}{1-F} = \frac{3300}{800}$
 $F = 0.76$

These two methods of calculating the trapping efficiency agree very well.

The efficiency for trapping ^{15}O can be calculated by using the measured currents from the ionization chambers before and after adding two scrubbers: one containing KOH and the other K pyrogallol. By adding these two scrubbers the current was reduced by a factor of five. The following equation can be written:

$$\frac{I_1}{I_2} = \frac{^{15}\text{O in} + ^{11}\text{C in}}{^{15}\text{O in} (1-F_1)e^{-\lambda t} + (1-F_2) \text{ } ^{11}\text{C in}} \quad (6)$$

where

- $I_1 = \text{current before scrubbers}$
 $I_2 = \text{current after scrubbers}$
 $F_1 = \text{fraction of } ^{15}\text{O removed in K pyrogallol scrubber}$
 $F_2 = \text{fraction of } ^{11}\text{C removed by KOH scrubber}$
 $^{15}\text{O in} = ^{15}\text{O concentration before scrubber}$
 $^{11}\text{C in} = ^{11}\text{C concentration before scrubber}$
 $\frac{5}{1} = \frac{5 \text{ } ^{11}\text{C in}}{(1-F_1) \text{ } ^{15}\text{O in} e^{-\lambda t} + (1-F_2) \text{ } ^{11}\text{C in}}$
 $\lambda = ^{15}\text{O decay constant} = 0.693/T_{1/2}$
 $T = \text{travel time through scrubbers.}$

The equation can be solved for F_1 .

F_2 is 0.78 from Eq. (4).

F_1 , the fraction of ^{15}O removed by the potassium pyrogallol scrubber, is 0.73.

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The degree of reduction needed to decrease the concentration in the research area can be estimated assuming our measurements were obtained under strong inversion conditions. Before measurements were taken a smoke bomb was fired so the dispersion of gases from the vent could be observed. If the concentration at 21 kW is extrapolated to a beam power of 1 megawatt, the concentration would be $\sim 10^{-2} \mu\text{Ci/cc}$.

The ICRP¹⁴ equations can be used to calculate the concentration in an infinite cloud to deliver 100 mrem/wk.

$$\text{MPC}_a = \frac{2.6 \times 10^{-6}}{\Sigma(E)} \mu\text{Ci/cc}$$

MPC_a = maximum permissible concentration ($\mu\text{Ci/cc}$) in a large cloud of gas that will deliver 0.1 Rem/wk.

(E) = effective energy per disintegration.

$Qf = 1$.

If the positron and annihilation radiation are considered in calculating the whole body dose a concentration of $2 \times 10^{-6} \mu\text{Ci/cc}$ would be needed. However, an MPC_a can be calculated separately for each component. The annihilation radiation will be whole body exposure and positrons will be considered as a skin dose.

This calculation shows that a concentration of $4.03 \times 10^{-6} \mu\text{Ci/cc}$ for ^{15}O is needed to deliver a 0.1 Rem/wk whole body dose and a concentration of $2.05 \times 10^{-5} \mu\text{Ci/cc}$ would deliver 0.6 Rem/wk to the skin.

The size of the sphere must be considered in deciding which MPC_a to use. The radius of the hemisphere filled with ^{15}O ($2.05 \times 10^{-5} \mu\text{Ci/cc}$) necessary to deliver a +B dose to the skin at the center of the hemisphere in excess of thirty times the photon whole body dose can be calculated. The photon dose at the center of an infinite hemisphere is

$$\sum_{i=0}^{\infty} D_{\text{Ci}} \propto \int_0^{\infty} e^{-\mu_a r} \propto \frac{1}{\mu_a}$$

where

μ_a = absorption coefficient for air

D_{Ci} = dose rate at center of infinite hemisphere

r = radius of hemisphere

The dose at center from a finite hemisphere is

$$\sum_{i=0}^r D_{Cr} \propto \frac{1}{\mu_a} - \sum_{i=r}^{i=\infty} e^{-\mu_a r}$$

$$\sum_{i=0}^r D_{Cr} \propto \frac{1}{\mu_a} - \frac{e^{-\mu_a r}}{\mu_a}$$

$$\frac{D_{Cr}}{D_{Ci}} = \frac{1/\mu_a - (e^{-\mu_a r}/\mu_a)}{1/\mu_a} = 1 - e^{-\mu_a r}$$

$$\text{for a finite radius } D_{Cr} = \frac{1}{30} D_{\beta^+}$$

$$\text{for an infinite radius } D_{Ci} = \frac{1}{1.2} D_{\beta^+}$$

$$\frac{D_{Cr}}{D_{Ci}} = \frac{1.2}{30} = 1 - e^{-\mu_a r}$$

for ^{15}O , $r = 12.0$ meters.

For a sphere with a radius less than 12.0 meters the skin dose would be the limiting factor. If people are inside buildings the higher MPCa could be used. However if people are outside, the lower value should be used. SLAC uses a guideline of 1.5 Rem/yr for whole body exposure. This means a concentration of $1.2 \times 10^{-6} \mu\text{Ci/cc}$ should be used to estimate the reduction factor.

Using a concentration of $1.2 \times 10^{-6} \mu\text{Ci/cc}$, a reduction factor of 1.6×10^4 would be needed to reduce the ^{15}O concentration to an acceptable level. If the ^{11}C could be reduced by 3×10^3 , a holdup of 28 minutes would reduce the ^{15}O concentration to acceptable levels.

This removal storage and venting scheme was rejected in favor of a positive sealing and hydrogen recombining system. Using this technique the potential exposure from this source of gaseous radioactivity has been eliminated.

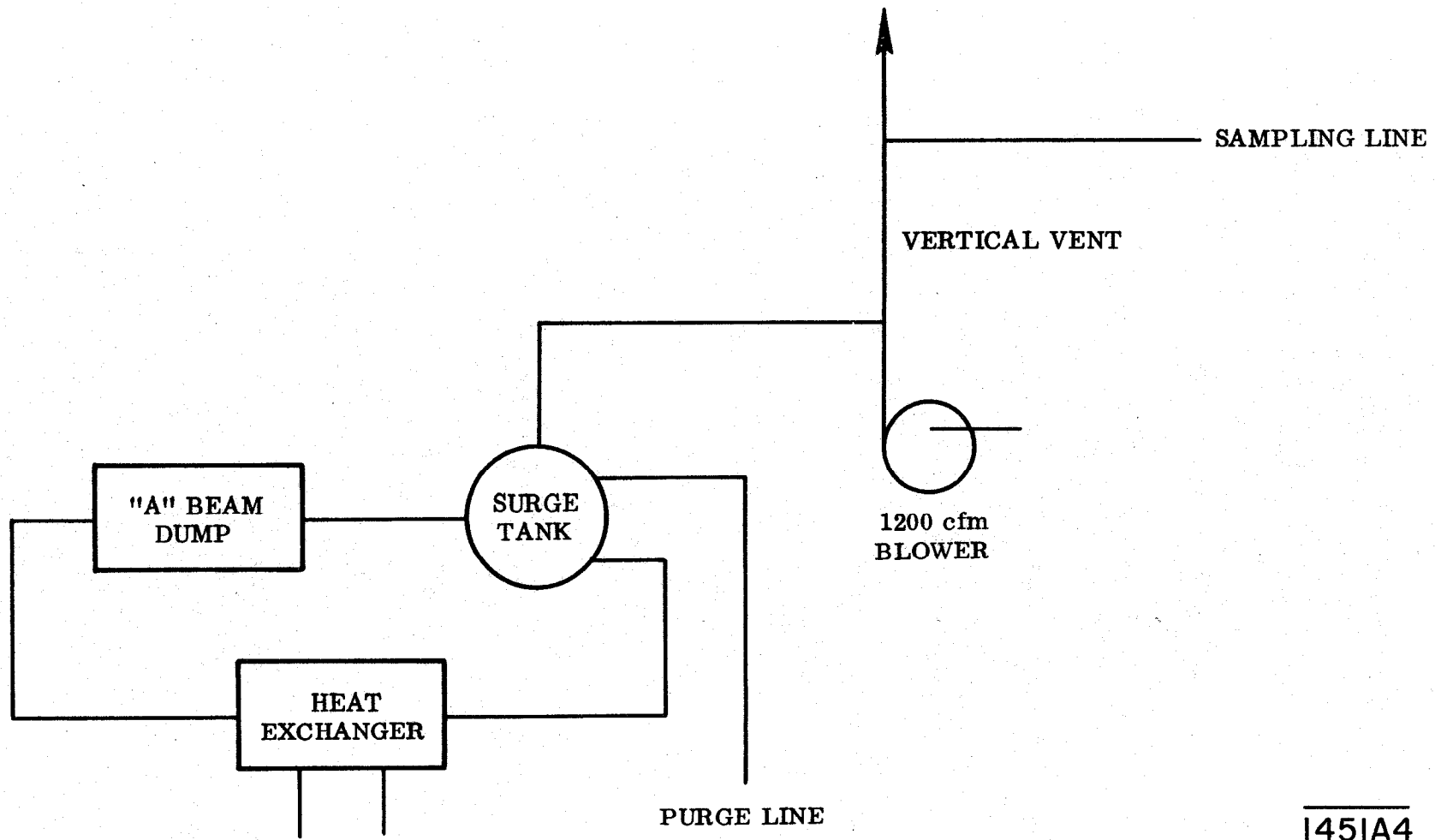
Summary

The $^{15}\text{O}/^{11}\text{C}$ ratio has been measured in two beam dump cooling water systems: (1) a water dump where 90% of the electromagnetic shower develops in water and (2) a water-cooled copper dump. The activity ratio ($^{15}\text{O}/^{11}\text{C}$) in each system is the same but corrected saturation activities are considerably different: 13/1 as compared to 1.6/1. We have not been able to explain the apparent difference. This will require measurements under carefully controlled conditions.

The gaseous activity released from the surge tank can be trapped in potassium hydroxide and potassium pyrogallol solutions. We have assumed that efficiency for removal of these isotopes is independent of concentration. As the concentration changes the fraction removed by a single pass will also change. We did not determine the functional relationship between removal efficiency and concentration during these tests.

REFERENCES

1. H. C. DeStaebler, Activation of the Cooling Water, Report No. SLAC-TN-61-19, Stanford Linear Accelerator Center, 1961.
2. H. C. DeStaebler, Activation in Water in Pipes Under the Accelerator, Report No. SLAC-TN-62-74, Stanford Linear Accelerator Center, 1962.
3. H. C. DeStaebler, Photon-Induced Residual Activity, Report No. SLAC-TN-63-92, Stanford Linear Accelerator Center, 1963.
4. H. C. DeStaebler, Tritium Production in Water, Report No. SLAC-TN-64-6, Stanford Linear Accelerator Center, 1963.
5. D. H. Coward, Stanford Linear Accelerator Center, Private Communication, 1964.
6. J. M. Wyckoff of The National Bureau of Standards, and T. M. Jenkins of Stanford Linear Accelerator Center, Private Communication, 1968.
7. R. C. McCall, Activation of Water During Beam Test, Report No. SLAC-TN-66-4, Stanford Linear Accelerator Center, 1966.
8. D. D. Buisck, ^7Be Build-Up in a Large Water Beam Dump System at Stanford Linear Accelerator Center, Report No. SLAC-PUB-521, 1968.
9. A. N. Gorbunov, V. A. Dubroxina, V. A. Osipova, V. S. Silaeva and P. A. Cerenkov, Investigation of the Photo-Effect in Light Nuclei, Soviet Phys. JETP: 42, 747-757, (March 1962).
10. K. Kramer, H. V. Buttlar, A. Goldmann and B. Huber, Cross Sections of the Reactions $^{16}\text{O}(\gamma, X) ^{13}\text{N}$ and $^{16}\text{O}(\gamma, X) ^{11}\text{C}$ and Yield of $^{12}\text{C}(\gamma, t)$ Up to 55 MeV, Zeitschrift für Physik: 207, 1-13 (1967).
11. V. Di Napoli, F. Dobicci, O. Forina, F. Salvetti and H. G. De Carvalho, Photo-nuclear Reactions in Carbon at Energies Between 300 MeV and 1000 GeV, Nuovo Cimento, Series X, : 55B , 95-106 (March 1968).
12. G. R. Bishop, University of Glasgow, Private Communications, 1969.
13. L. F. Hamilton and S. G. Simpson, Quantitative Chemical Analysis, 11th ed., pp. 465 and 466, Macmillan Publishing Company, New York, New York, 1958.
14. Committee II on Permissible Dose for Internal Radiation, Radiation Protection, Report No. ICRP-PUB-2, pp. 22 and 28, Pergamon Press, New York, New York, 1959.



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Fig. 1

Schematic water flow diagram for "A" beam dump.

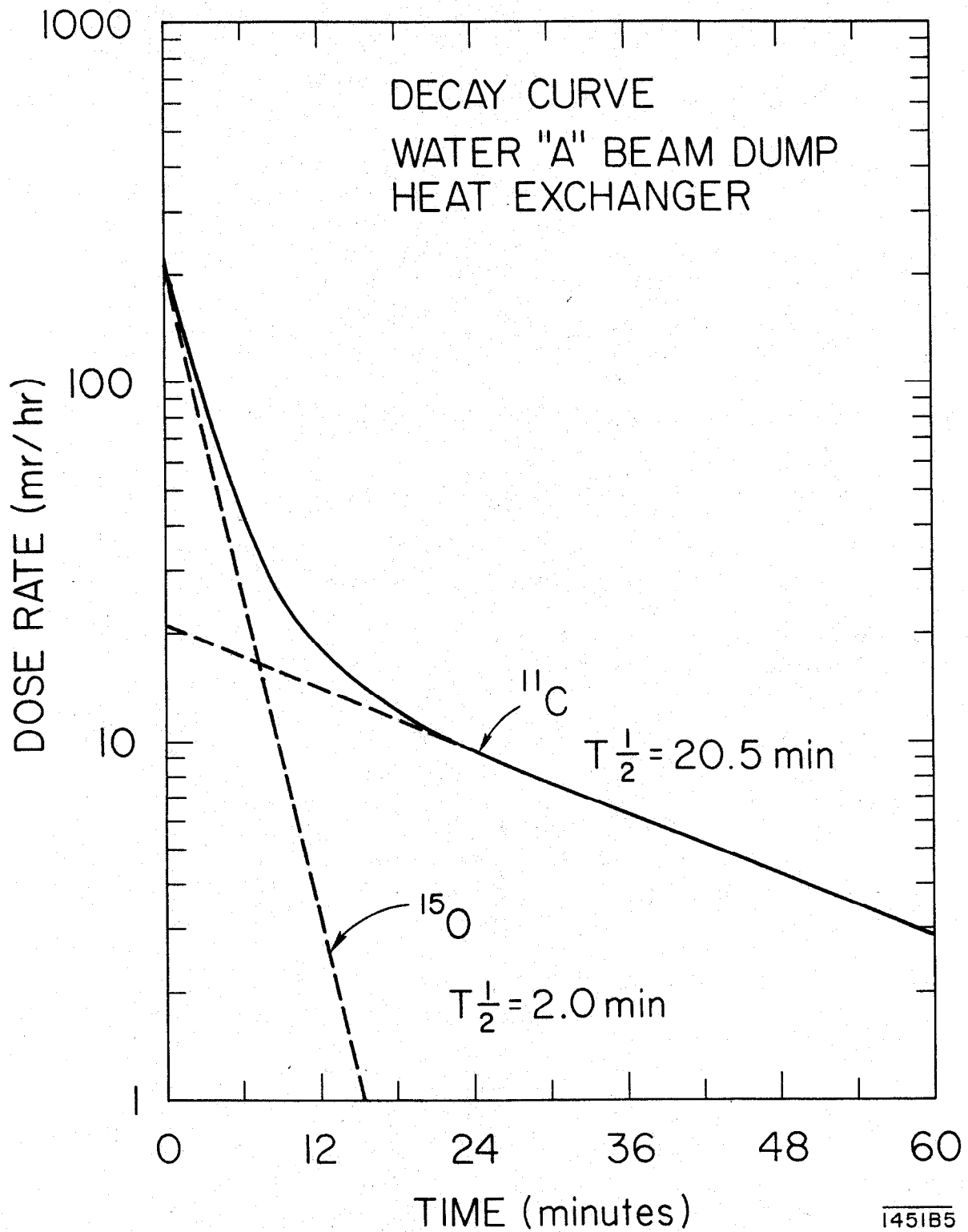


Fig. 2

Decay curve—activity in "A" beam dump heat exchanger.

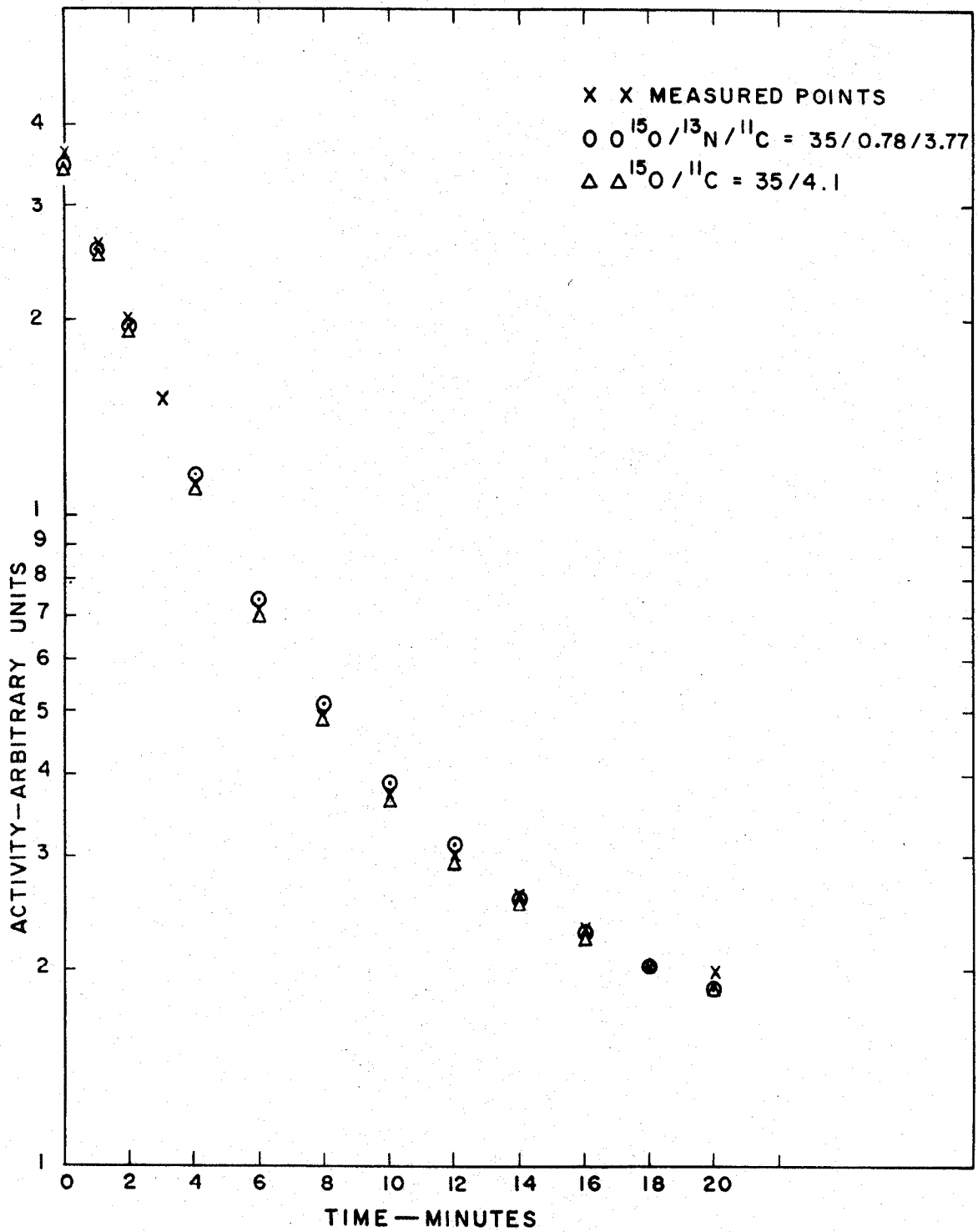
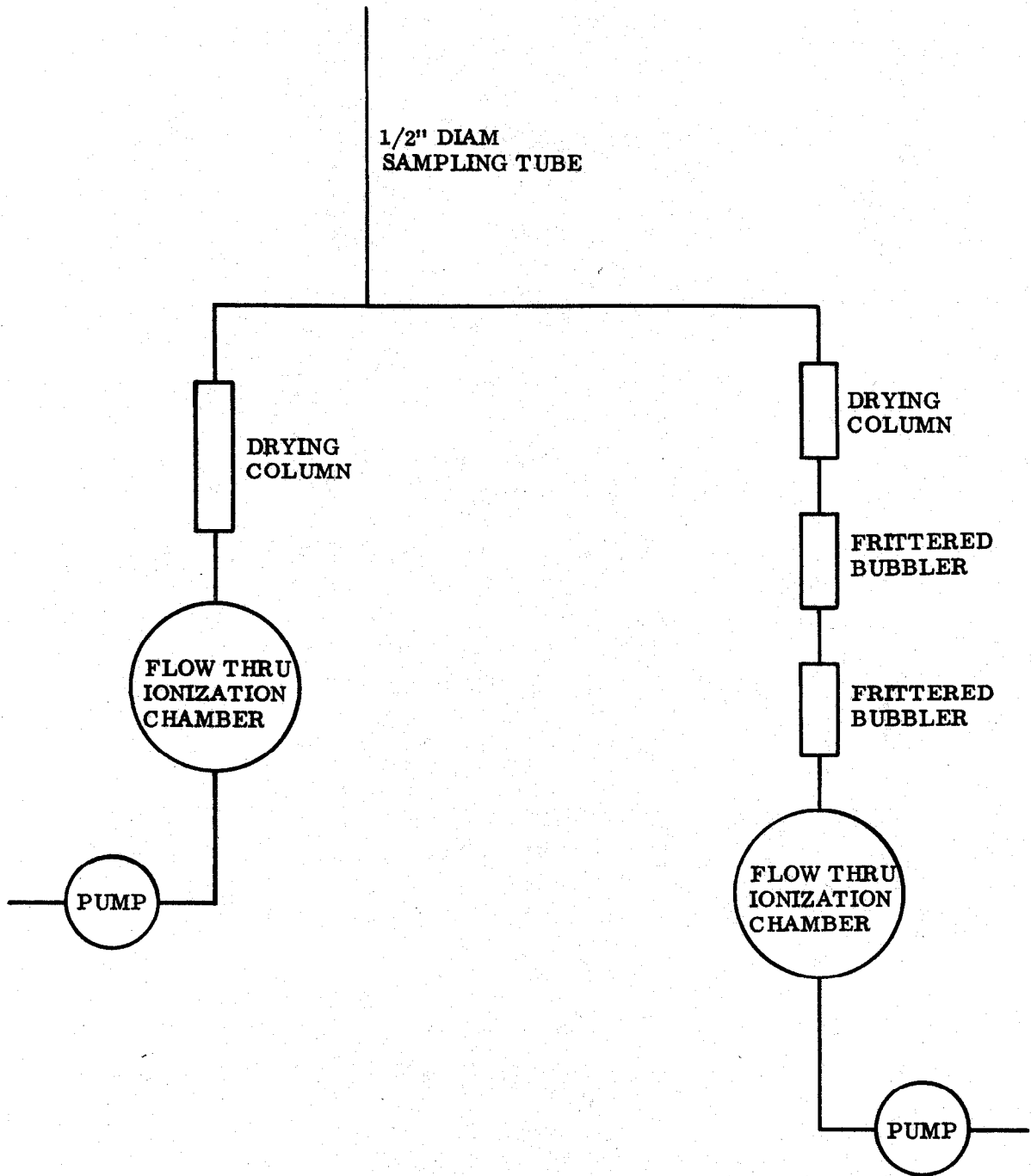


Fig. 3

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Decay curve-activity in cooling water for copper beam dump.



EXPERIMENTAL ARRANGEMENT

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Fig. 4

Schematic showing scrubber and monitor arrangement.

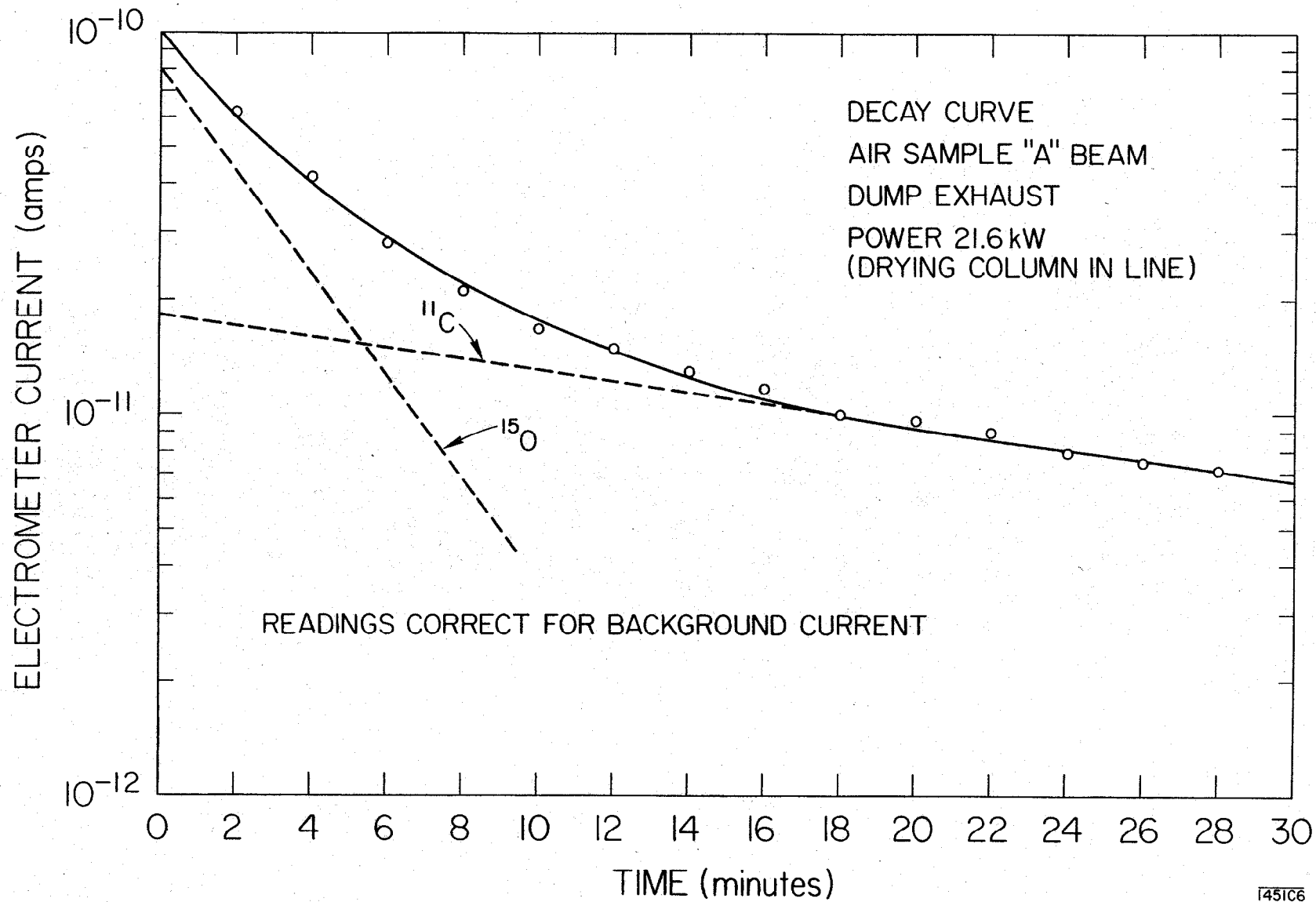


Fig. 5

Decay curve-air sample before scrubbers.

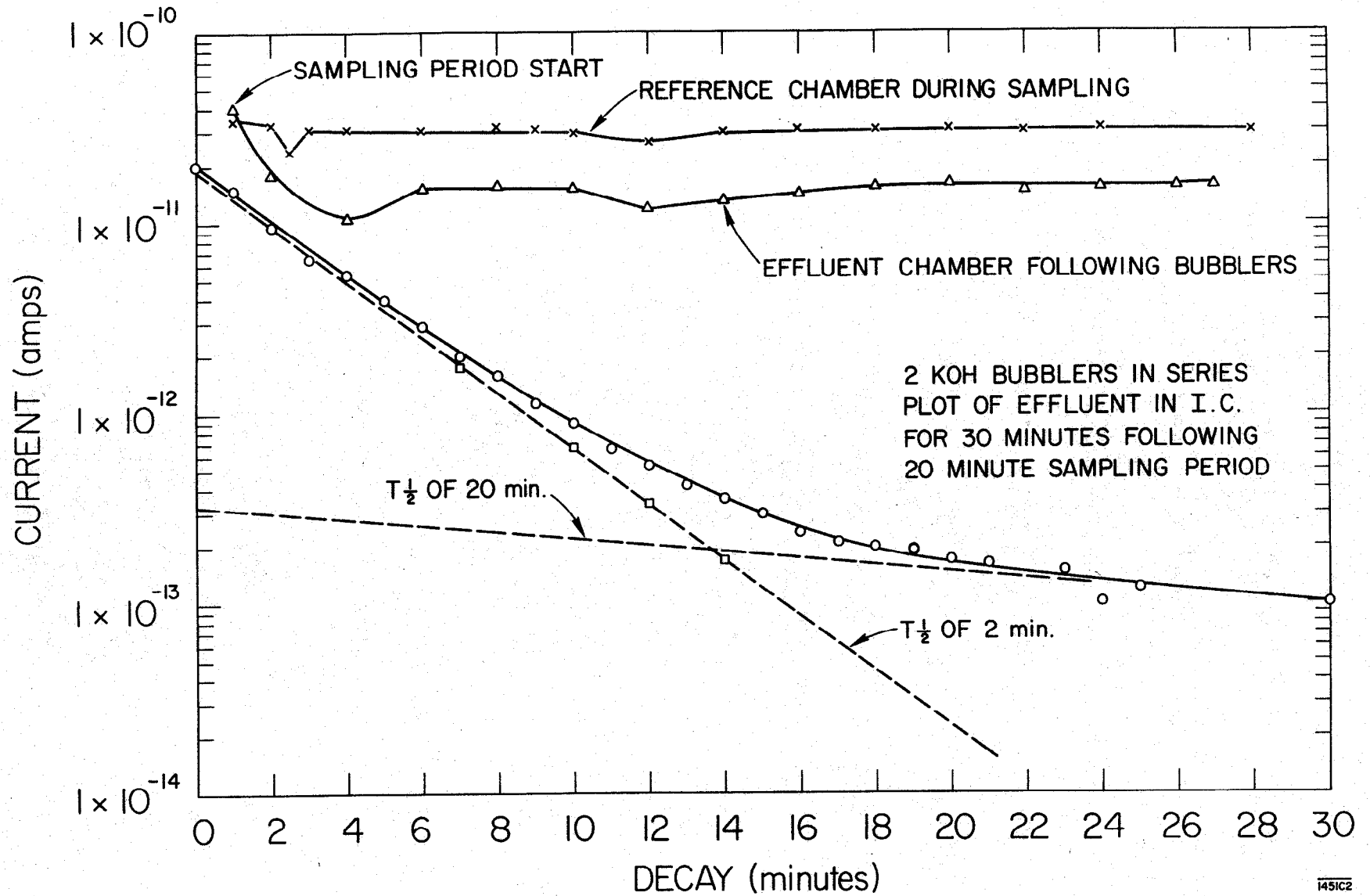


Fig. 6

Decay curve-air sample after scrubbers.