RADIOACTIVE GAS PRODUCTION FROM THE R.P.I. ELECTRON LINEAR ACCELERATOR*

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Summary

The energy and average power of the R.P.I. electron linear accelerator (Linac) are great enough that radioactive gases are produced. During two years of operation ^{15}O and ^{13}N have been detected, identified, and measured quantitatively. This paper describes the air handling and monitoring systems, measurement of the gaseous activities, concentration guides developed at R.P.I. for these isotopes, and operating procedures used for control of the activity.

Introduction

The design and operation of the R.P.I. electron Linac have been described in detail elsewhere.[1,2] The operational parameters of interest are that it produces electrons over the energy range 5 Mev to 70 Mev, in pulses from 5 ns to 4.5 usec wide, at repetition rates from 1 to 720 pps. The largest observed peak current to date is 2.0 amperes and the highest average beam power is 50 kilowatts. With an electron beam of this intensity, very high gamma ray and neutron fluxes are produced, over 10^7 Rads/sec of gammas in a Bremsstrahlung beam and over 10^{14} neutrons/second. These fluxes of gamma rays and neutrons are of an energy and intensity capable of inducing radioactivity and radiation damage in any exposed material. Reactions producing gaseous activities are considered in this paper while some radiation damage effects are described in another paper at this conference.[3]

Air Exhaust and Filtration System

The accelerator and target rooms at the R.P.I. Linac (Figure 1) are maintained at a pressure slightly below the normal atmospheric pressure of the rest of the laboratory by control of the air intake and exhaust rates. These rooms have a volume of 43,000 cubic feet. Air is exhausted at a maximum rate of ~15,000 cubic feet per minute providing an average of ~1 air change every three minutes. Both intake and exhaust are regulated from the Linac control room. Exhaust air passes through pre filters and absolute filters before leaving the target room. It is ducted up through the Linac shielding and then to the exhaust blowers (Figure 2). It is released from a 20 meter high 3 foot diameter stack at a velocity of about 20 mph. Outside air can be mixed with the exhaust air through another damper located just before the exhaust blowers.

Monitoring Systems

A sample of exhaust gas is drawn continuously from the stack through an isokinetic sampling head. It is passed at a rate of about 5 c.f.m. through a fluid monitor and a moving filter paper particulate sampler. The fluid monitor consists of a 30 litre volume of gas contained in a steel cylinder shielded by 2 inches of lead. This volume of gas is looked at by two thin wall G-M tubes with their outputs connected in parallel. (For gamma ray energy determinations, one of the G-M tubes is replaced by a 2" x 2" NaI crystal and photomultiplier tube.) The count rate of both of these monitors is displayed and recorded on a dual channel pen recorder in the Linac control room. (Figure 2)

Calibration of the observed count rates in terms of radioactivity per volume of gas is made with a mixture of a known activity of ^{85}Kr and a known air dilution. A correction is also made for the change in detector sensitivity from the positron and annihilation gamma ray energy of ^{15}O and ^{13}N to the ^{85}Kr radiation. Wind direction and velocity are also monitored and recorded continuously.

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Air from the target and accelerator rooms and air from the laboratory areas is sampled and fed to another dual channel fixed filter particulate monitoring system located in the accelerator building. Radiation levels are monitored at 6 locations in the laboratory, the target room, accelerator room, klystron modulator room, neutron flight path area, hot laboratory, and change area. The detectors are NaI crystals and phototubes and the information is displayed and recorded in the accelerator control room. Any liquid waste from the radiation areas of the laboratory is carried in a separate drain system to a stainless steel hold up tank. The contents of this tank are measured for radioactivity before disposal.

Radioactive Gases Produced

Before the construction of the first high power electron linear accelerator, studies were made, notably by Petree[4] at the National Bureau of Standards, of the reactions and activities that could be produced in air. It was predicted that ^{15}O and ^{13}N could be formed in amounts that would require control. This prediction has been confirmed.

The nuclear reactions which are responsible for the generation of these isotopes are ^{16}O (γ,n) ^{15}O and ^{14}N (γ,n) ^{13}N. They have thresholds of 15.6 and 10.5 Mev respectively and giant resonance energies of 23.5 and 22.5 Mev respectively. ^{15}O is a positron emitter with a half life of 2.05 minutes. ^{13}N is also a positron emitter with a ~10 minute half life.

The isotopes were identified in target room air samples and in exhaust stack gas by radioactive decay half life determination and by gamma ray spectra. The decay of a sample of irradiated air is shown in Figure 3. The curve is well fitted by the sum of two exponential decays, with 2 minute and 10 minute half lives. A gamma ray spectrum of a sample of irradiated air is shown in Figure 4. It shows a strong peak at about 0.51 Mev corresponding to the annihilation radiation from the positrons from the decay of ^{15}O and ^{13}N. No other radioactive gases have been detected. Their activities, if present, must be more than two orders of magnitude lower than the ^{15}O and ^{13}N.

Concentration Guides

Following identification of the two gases ^{13}N and ^{15}O, attempts were made to determine recommended concentration guides for these gases for both radiation workers and the general population. At that time, neither the U.S. Atomic Energy Commission nor the International Committee on Radiation Protection had put forth recommended concentration limits for these isotopes. Calculations were made at R.P.I. using methods described by the I.C.R.P.[5] This calculation takes into account decay energy and half life of the isotope, absorption cross sections for the radiation, and organs of reference and other biological parameters. The results of these calculations were the following concentration guides.

Isotope	Occupational(40 hr)	Population
^{13}N	2.3×10^{-6} uc/ml	5×10^{-8} uc/ml
^{15}O	2×10^{-6} uc/ml	6×10^{-8} uc/ml

These calculations were made independently by the AEC with essentially the same results and they have been accepted by the New York State Department of Health.

To determine maximum allowable activity for stack discharge, the population limit for the radioactive gases was used and a meteorological calculation based on Sutton's equation[6] was performed. This takes into account wind direction and velocity, height, and velocity of exhaust as well as radioactivity of the discharge. The result of this calculation was an upper limit for stack discharge of 1.2×10^{-4} uc/ml for the mixed ^{15}O and ^{13}N activity. This calculation has also been verified by the AEC and accepted by the New York State Department of Health.

Operating Experience and Procedures

The concentrations of radioactive gases produced by the Linac depends on electron beam energy and average power and on the air path of the Bremsstrahlung. At high power and high energy and with a long gamma ray air path, gas activities will exceed the limits for discharge by several oders of magnitude. The operating procedure used at the R.P.I. Linac is that the scientist in charge of each experiment must maintain radioactive discharge below the above limits.

The plans for accomplishing this, accelerator operating conditions, targets and shielding are subject to review by the health physicist and a committee of other scientists. The techniques for maintaining discharge below the set limits may involve operating the accelerator at low energy, low beam current or low repetition rate. More often they include shielding to limit the air path for the gamma rays or a vacuum chamber surrounding the target to exclude air from the γ-ray path. With a very diversified experimental program using the accelerator under many different conditions, each of these techniques has been used. For example, by keeping the gamma ray air path to less than 10 cm with heavy shielding, it is possible to operate at 25 Kw average beam power at 55 Mev without exceeding the prescribed limits for stack discharge. As another example, at electron beam energies below 30 Mev, a gamma ray air path of 5 meters does not produce radioactive gas in excess of the limits. This is because relatively little of the gamma ray flux has an energy above the giant resonance energies for the photonuclear reactions producing the gaseous radioactivity.

Ozone and Oxides of Nitrogen

A high power electron linear accelerator can also produce ozone and oxides of nitrogen. Both the electron beam and the gamma rays can induce formation of O_3 from the oxygen in the air. Maximum permissable concentrations for these compounds have been established.[7] The presence of these compounds is relatively easy to detect, but a reliable absolute measurement of their concentration is difficult. These molecules are unstable and recombine to form normal air. The rate of recombination depends on mixing of the air and temperature, among other things. The recombination can be catalyzed by a variety of agents.

Concentrations of 10 parts per million of ozone have been detected in the target room but concentrations in other parts of the target room at the same time were lower by a factor of 10. Concentrations in the stack discharge have not been measured above 1/10 part per million.

A study is underway investigating ozone monitors. The possibility of a catalyst in the exhaust system to speed up recombination of ozone and oxides of nitrogen is also being studied. At present, the limitations on electron beam air path and gamma ray air path placed by the radioactive gas production also serve to limit the ozone production. Concentrations in the environment have not been detected above prescribed limits.

References

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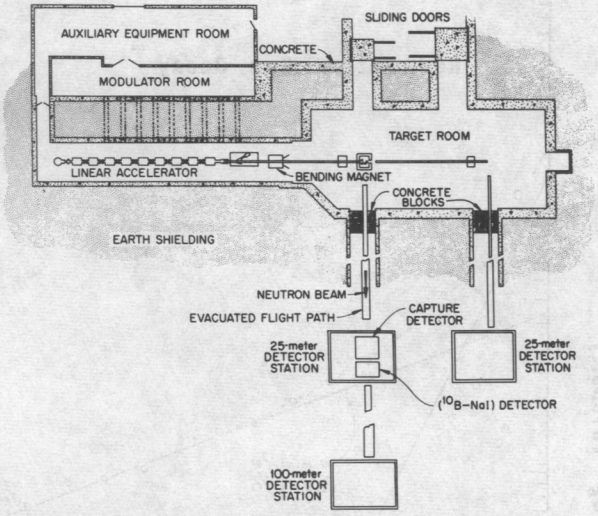

Fig. 1. R.P.I. Electron Linear Accelerator and Target Room.

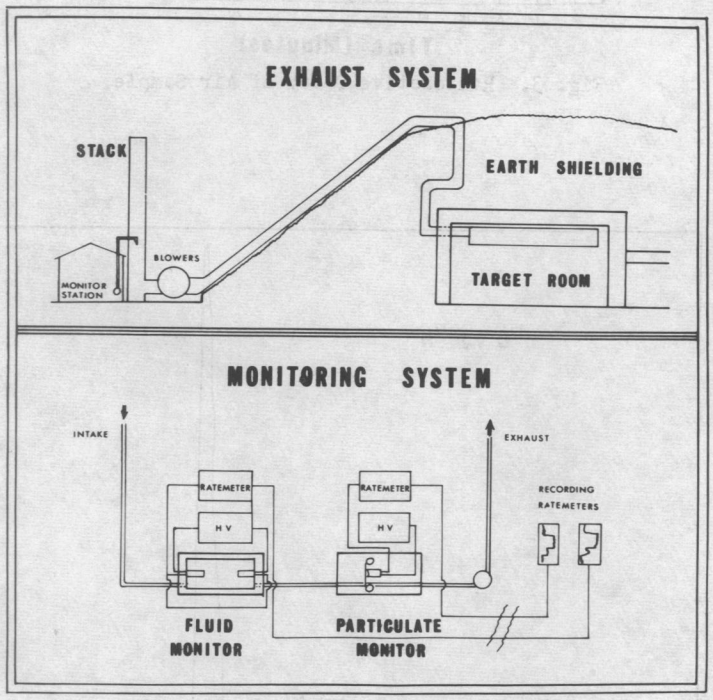

Fig. 2. Exhaust System and Monitoring System.

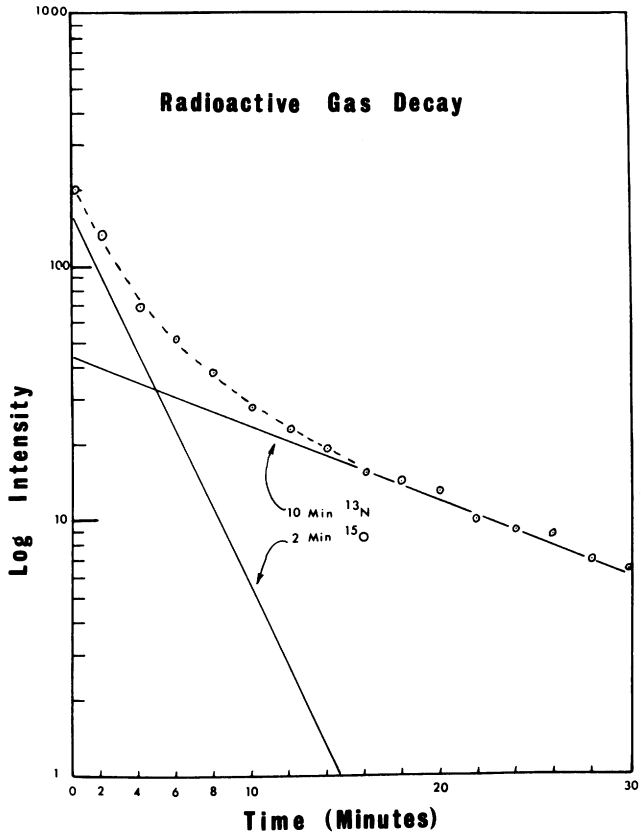

Fig. 3. Radioactive Decay of Air Sample.

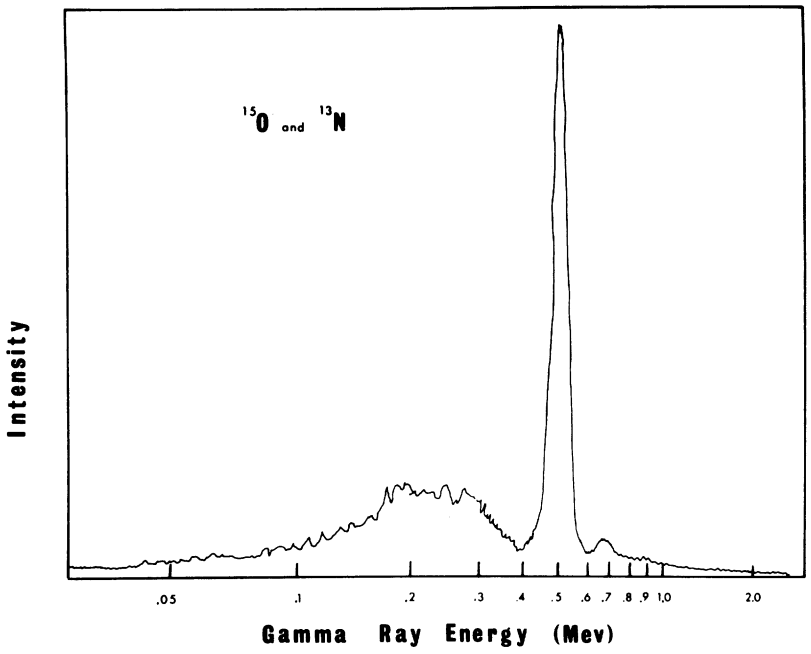

Fig. 4. Gamma Ray Spectrum of Air Sample.