A TRITIUM-IN-AIR MONITOR WITH COMPENSATION AND ADDITIONAL RECORDING OF α-, β- AND γ-BACKGROUNDS

Takahiko Aoyama
Department of Nuclear Engineering
Nagoya University
Nagoya 464-01, Japan

ABSTRACT

Development is described of a computer controlled tritium-in-air monitor using flow-through air proportional counters as the detector. Combining automatic compensation for β- and γ-backgrounds with α-background discrimination, the monitor can detect tritium activity separately from coexisting other radioactive gases. The computer of the monitor calculates and prints out not only tritium concentration in air but also dose equivalent rate by external γ-rays and/or activity concentrations of other radioactive gases than tritium in real-time. Since the monitor has a wide range of 0.005 - 5000 Bq/cm^3 (6 decades) and a short response time of several min, it can be used effectively to monitor tritium in air in fission and fusion reactor environments.

INTRODUCTION

Tritium-in-air monitoring in the environments of heavy water reactor, reprocessing facility, 14 MeV neutron generator and nuclear fusion reactor requires separate detection of tritium from radioactive rare gases or air activation products. Real-time tritium monitors used for occupational radiation protection and emission control must have a high sensitivity to meet legal requirements and a wide range to respond to an accidental release of large amounts of tritium gas.

Various methods have been developed to monitor tritium in the presence of other radioactive gases, which are reviewed by R. A. Jalbert [1]. Of these, the one with the highest sensitivity was considered to be the use of proportional counters with anti-coincidence shielding or pulse-shape discrimination. Conventional proportional counters, however, have the disadvantages of requiring a counting gas and of a short operating range.

The present paper describes a new method of tritium monitoring using flow-through air proportional counters as the detector. The counters need no counting gas other than the sampled air, and the monitor, using the combination of pulse height discrimination, anti-coincidence shielding and background compensation, is possible to detect tritium β-rays alone not only in an external γ-background but also in α- and β-backgrounds originating from other radioactive gases contained in the sampled air. Using this monitor, we can obtain a low detection limit of about 0.005 Bq/cm^3 with a natural background level and a measuring time of 1 min, and a wide range up to 5000 Bq/cm^3 with 6 decades.

The author wish to thank Professor Emeritus T. Watanabe for his continued encouragement and Mr. K. Yanagida for his technical support to fabricate the interface of the computer.

CONSTRUCTION AND PRINCIPLE

Fig. 1 shows the cross sectional view of the detector. The detector consists of four layers of multiwire proportional counters set in a stainless-steel vessel and it is designed so that the air flows uniformly through the counters. The air was sampled with a diaphragm pump from any spot of a room, and dust particles with a size larger than 1 μm were removed from the air with a membrane filter.

The anode of each counter was composed of 62 tungsten wires, 50 μm in diameter and 200 mm long, which were soldered on a glass-epoxy printed circuit board with a wire spacing of 3 mm and were connected to a charge sensitive preamplifier. The cathodes were made of stainless-steel wire meshes, 50 μm in diameter and 0.5 mm apart, which were glued to glass-epoxy frames, and to which a negative high voltage was applied.

Fig. 2 shows the scheme of the monitor. The outer two layers of the counters were used as guard counters to eliminate the γ-background, and the tritium signal was obtained from the inner two layers of the main counters. The counting signals from each counter were fed to a pulse-processing circuit, and separated into three counting rates, i.e. a coincidence rate N_g between the main and guard counters, an anti-coincidence rate N_a derived from the main counters, and a coincidence rate N_c between the main counters.

0018-9499/90/0400-0885$01.00 © 1990 IEEE

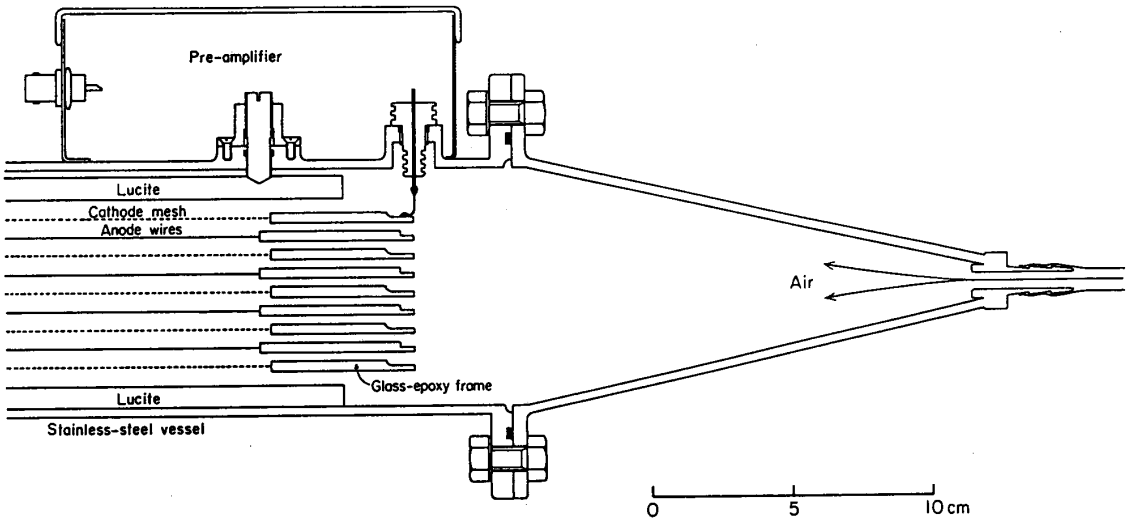

Fig. 1. Cross sectional view of the detector.

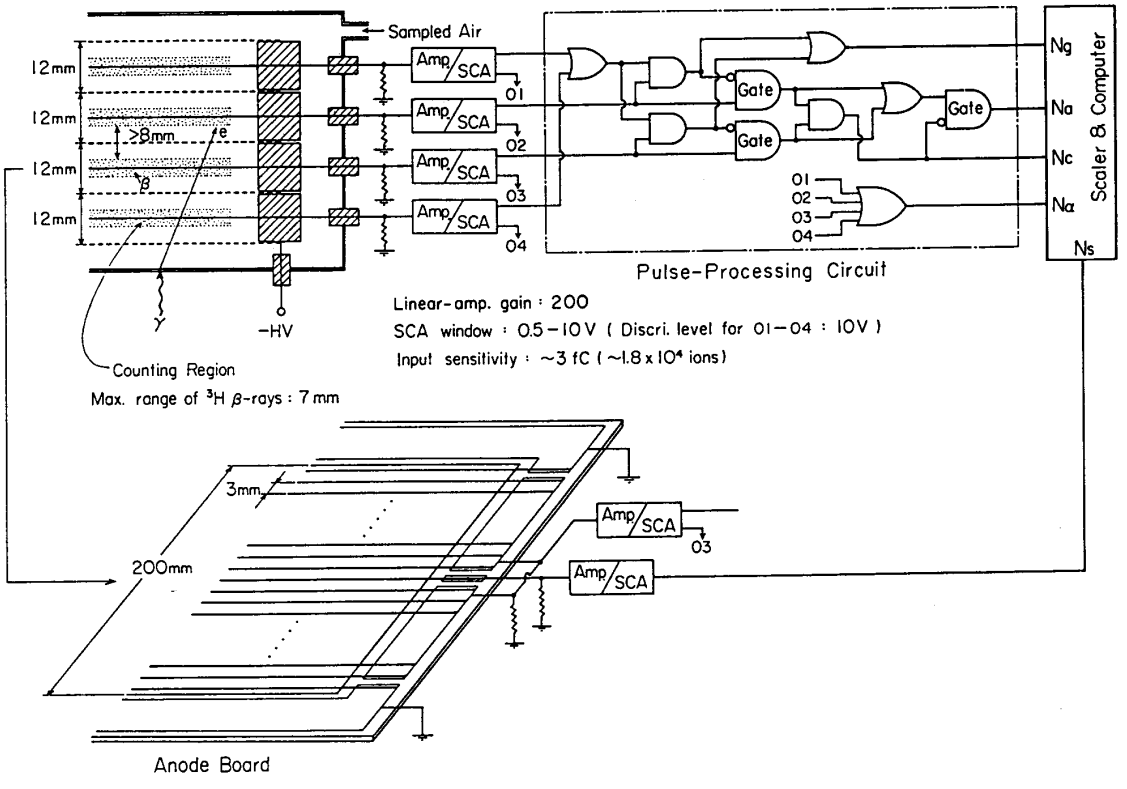

Fig. 2. Scheme of the tritium-in-air monitor.

The anode-cathode distances of each counter were 6 mm. Counting regions around each anode wire (dotted in fig. 2) were found to be localized within 2 mm from the center of the wire in the air at an atmospheric pressure [2], and therefore the regions have gaps larger than 8 mm between them. Since tritium β-rays have short ranges with a maximum of about 7 mm in the same condition, they cannot be counted coincidentally, thus contributing to N_a alone. On the other hand, secondary electrons produced by γ-ray interaction with detector materials and β-rays emitted from other radioactive gases have various ranges, and hence they contributed to all of the rate N_g, N_a and N_c. Since the elimination of the γ-component by the anti-coincidence shielding is incomplete, about half being eliminated from N_a, we estimated the residual γ-component from the ratio N_g/N_c and the sum N_g+N_c, and subtracted it from N_a to produce a net counting rate by tritium β-rays in real-time. A β-component from other nuclides than tritium could be eliminated by the same manner as the γ-component elimination, and an α-component from radon and its daughter nuclides was eliminated by pulse height discrimination, since it produced saturated pulses with a height larger than 10 V. The saturated pulses were summed into an α-counting rate N_α.

One of the 124 anode wires of the main counters was separated and a counting rate N_s was derived from this single anode wire. The rate N_s was used as a substitute for the anti-coincidence rate N_a when saturation of N_a became significant for $N_a > 1.4\times10^4$ cps. This extended the maximum measurable tritium concentration from 50 to 5000 Bq/cm^3.

Due to the localization of the counting regions as described above, this monitor is expected to be free from the tritium memory effect, since β-rays emitted from tritium depositions on the cathode meshes can scarecely reach the counting regions because of a wide gap of about 4 mm between the meshes and the regions. This was verified simply by using tritium gas with a concentration level of some 100 Bq/cm^3 and not for tritium oxide though the latter might be significant.

Data acquisition and detector control were carried out automatically through a personal computer. Data derived from pressure, temperature and humidity sensors installed at the sampled air intake of the detector were used to control the high voltage applied to the detector and to correct the data of the coincidence counting rates as will be described later.

OPERATION AND CHARACTERISTICS

The coincidence rates N_g and N_c were verified to be independent of the presence of tritium in the sampled air, and they were proportional to the intensity of external γ-rays and of β-rays from other radioactive gases. We can therefore estimate the background component included in the anti-coincidence rate N_a from observed values of total coincidence rate N_g+N_c if a compensation factor $k=N_a^b/(N_g+N_c)$ is given beforehand, where N_a^b is the value of N_a for the background.

Both the factor k and the ratio $r=N_g/N_c$ were found to be dependent on the energy of β- and γ-rays. Fig. 3 shows 1/k as a function of r, which was obtained by irradiating γ-rays with various energies and by injecting β-emitting nuclides such as ^{85}Kr and ^{14}C into the detector. The experiment was carried out with a constant appled voltage of 4745 V by filling dry air to the detector at 1 atm and 25°C. It is seen from fig. 3 that 1/k

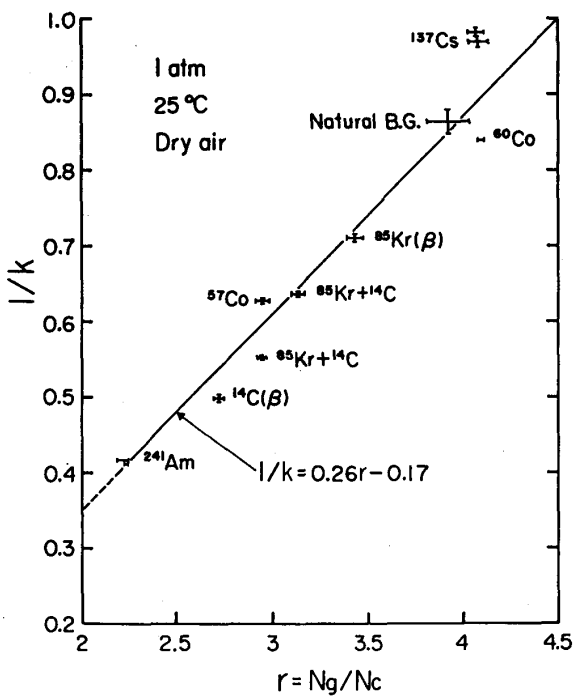

Fig. 3. Relation between 1/k and $r=N_g/N_c$ observed with various β- and γ-sources. The experiment was carried out with a constant applied voltage of 4745 V by filling dry air at 1 atm and 25°C. Error bars are based on the counting statistics alone without including unknown systematic errors.

increased with r linearly within a 10% error for r larger than the minimum value of 2, and that this linear relation could be applied to mixtures of different β-emitting nuclides. The value of k slightly depended on the direction of external γ-rays - about 5% smaller for parallel incidence than for normal incidence on the counter layers. Since the value of r also depended on the direction of γ-rays and for different directions the value of 1/k shifted along the linear relation, the relation is realized independently of the direction of external γ-rays. From fig. 3 we can estimate the factor k independent of the details of the background using observed values of r. The net counting rate N_n by tritium β-rays then is calculated from $N_n=N_a-k(N_g+N_c)$, where N_a, N_g and N_c are the values corrected for accidental coincidences. Incorrect adjustment of the factor k results in the error of the counting rate N_n in the tritium channel, e.g. for 1 Bq/cm^3 of ^{85}Kr a 5% error of the factor k results in an apparent tritium counting rate of 1260 cpm, which is corresponding to 0.08 Bq/cm^3 of tritium concentration.

The compensation factor k was considered to be dependent on the density of the air and therefore on the pressure and the temperature at a constant applied voltage. Fig. 4 shows an applied voltage V[volts] required to keep k constant as a function of pressure P[mb], which was obtained by irradiating γ-rays from ^{137}Cs to dry air at a constant temperature of 25°C. A linear relation between V and P is evident for a pressure range from 960 to 1040 mb. The relation, also using a temperature T[°C] as a variable, is given by

$$V = 838.2 \frac{P}{273.2+T} + 1895. \quad (1)$$

Pulse height analysis at each set of pressure and applied voltage in fig. 4 revealed a constant gas gain, which is consistent with a linear relation between V and P at a constant gas gain observed with conventional proportional counters [3]. The relation of eq. (1) would therefore be independent of β- and γ-ray energies.

Fig. 4 also shows the change with pressure of relative efficiency η for either anti-coincidence or total coincidence counting normalized at 1 atm, which was obtained with each set of pressure and applied voltage. It is seen from fig. 4 that the relative efficiencies η for ^{137}Cs γ-rays and for tritium β-rays are constant within a 1% error for a pressure change of ±30 mb from 1 atm. When applied voltage is adjusted according to eq. (1), a tritium concentration C[Bq/cm^3] can be evaluated using a

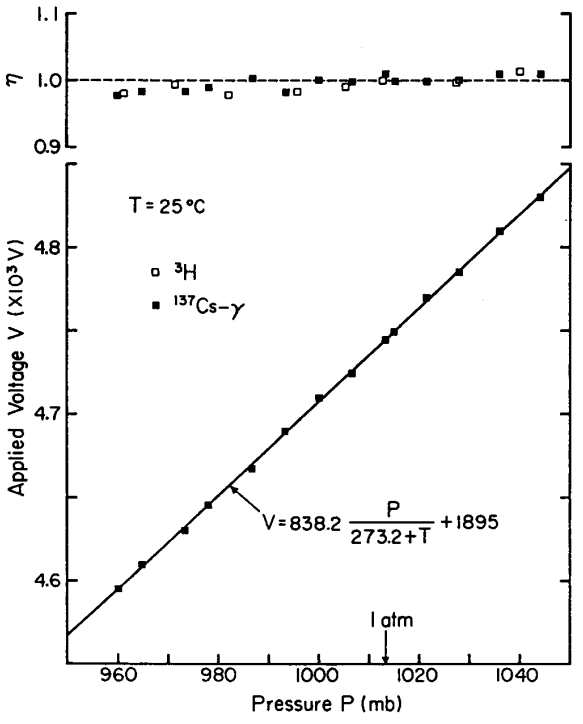

Fig. 4. Applied voltage V required to keep k constant as a function of pressure P, and the change with pressure P of relative efficiency η for either anti-coincidence or total coincidence counting normalized at 1 atm, which were obtained by irradiating γ-rays from ^{137}Cs (full squares) and by injecting tritium gas (open squares) into the detector filled with dry air at 25°C.

calibrating factor f (= 6.12x10^{-5} Bq/cm^3/cpm) independent of pressure and temperature from the equation

$$C = fN_n = f[N_a - k(N_g+N_c)]. \quad (2)$$

When sampled air is humid as is always the case, counting efficiency decreases because of the reduction of the counting regions around the anode wires [4], though the gas gain was found to be constant without depending on the humidity. The decrease of the efficiency was considered to be dependent on the abundance ratio of water molecules in the air or the ratio of a water vapor pressure P_w to a total pressure P. The pressure P_w is given using a humidity H[%] and a temperature T[°C] by the expression

$$P_w = \frac{H}{100} \times 1013 \exp\left(-\frac{5306}{273+T} + 14.33\right). \quad (3)$$

Fig. 5 shows relative counting efficiencies or ratios $(N_g^h+N_c^h)/(N_g+N_c)$ and N_a^h/N_a as a function of the ratio P_w/P, where $N_g^h+N_c^h$ and N_a^h are respectively the total coincidence and anti-coincidence rates for humid air, and N_g+N_c and N_a are those for dry air. It is seen from fig. 5 that the ratio $(N_g^h+N_c^h)/(N_g+N_c)$ decreased linearly with increasing P_w/P while the ratio N_a^h/N_a was kept constant without depending on the energy of irradiated γ-rays. The total coincidence rate $N_g^h+N_c^h$ obtained with humid air must therefore be corrected to the rate N_g+N_c for dry air for the evaluation of the net counting rate N_n in eq. (2). The correction was carried out automatically by replacing the data of temperature and humidity into eq. (3) and by using the relation of fig. 5.

When humidity exceeded 30%, the generation of spurious pulses and therefore the instability of the detector became appreciable because of the occurrence of leakage current through the surface of the insulators constructing the counters [4]. For the present monitor, if the humidity of the sampled air is higher than 30%, it must be lowered to the acceptable level, for instance by mixing dried clean air to the sampled air.

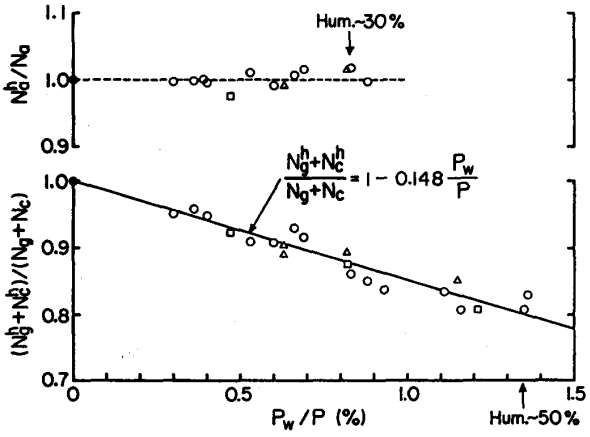

Fig. 5. Dependences of relative efficiencies or ratios N_a^h/N_a for anti-coincidence counting and $(N_g^h+N_c^h)/(N_g+N_c)$ for total coincidence counting on the ratio P_w/P of the water vapor pressure P_w to the total pressure P, which were observed with γ-ray irradiation from ^{57}Co(open triangles), ^{137}Cs(open circles) and ^{60}Co(open squares) having different γ-ray energies.

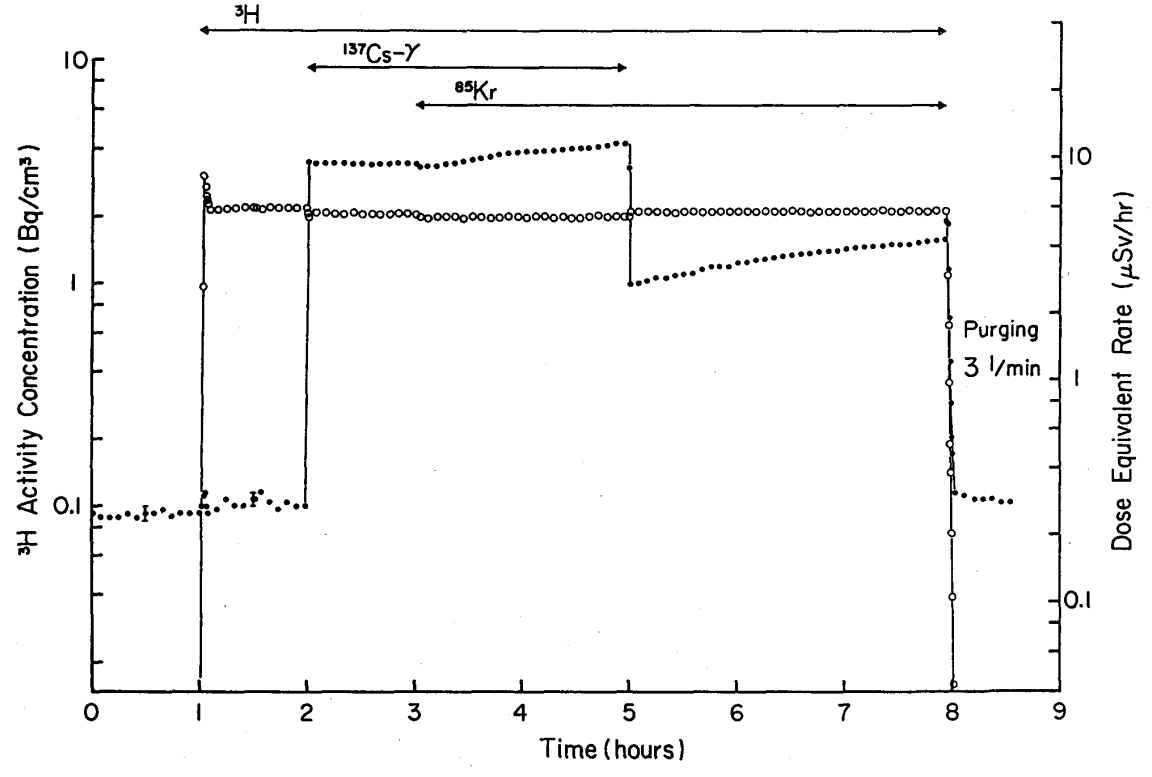

Fig. 6. Tritium monitoring in β- and γ-backgrounds. Tritium activity concentration (open circles) and dose equivalent rate (full circles) are direct printouts from the computer.

REAL-TIME TRITIUM MONITORING IN β- AND γ-BACKGROUNDS

The values of coincidence and anti-coincidence counting rates, and of pressure, temperature and humidity of sampled air, which were acquired every 30 sec, were used as the input data of the computer, and the tritium concentration was calculated using eq.(2) in real-time. β-background from radioactive gases other than tritium cannot be distinguished from the γ-background, and it contributes to corresponding dose equivalent rate D by ^{137}Cs γ-rays or, as a substitute, β-activity concentration by other radioactive gases. The rate D[μSv/hr] is given by

$$D = g(N_g+N_c), \qquad (4)$$

where g (= 5.92×10^{-4} μSv/hr/cpm) is the calibrating factor, and the rate N_g+N_c is that corrected for accidental coincidence and humidity.

For a slow change of tritium concentration and dose equivalent rate, average values of C and D over 5 min are printed out. If the value of C or D suddenly changes by more than three times the standard deviation within 30 sec, both C and D are printed out directly.

Fig. 6 shows a demonstration of tritium monitoring in both β- and γ-backgrounds, where the values of C and D were those directly printed out together with their time from the computer. At 1 hr, the tritium gas was injected into the detector and the air flow was stopped. At 2 hr, ^{137}Cs γ-rays were irradiated externally. At 3 hr, diffusion of ^{85}Kr gas (0.37 Bq/cm^3 at saturation level) into the detector was started from the sampled air intake of the detector. At 5 hr, the ^{137}Cs source was removed, and at 8 hr, tritium and ^{85}Kr gases were exhausted with a flow rate of 3 l/min. After 5 min from the start of exhausting, a tritium concentration C=0 was printed out, which indicated that the product of response time and flow rate was 15 l - about twice as large as a detector volume of 7.8 l. It is seen from fig. 6 that the tritium concentration was calculated correctly independent of the presence of β- and γ-backgrounds.

The minimum net counting rate N_m with which we can detect the presence of tritium activity in sampled air with a 99.7% confidence level is obtained as the net counting rate equal to three times the standard deviation:

$$N_m = N_a - k(N_g + N_c) = 3\sqrt{\frac{N_a}{t} + k^2\left(\frac{N_g}{t} + \frac{N_c}{t}\right)}, \qquad (5)$$

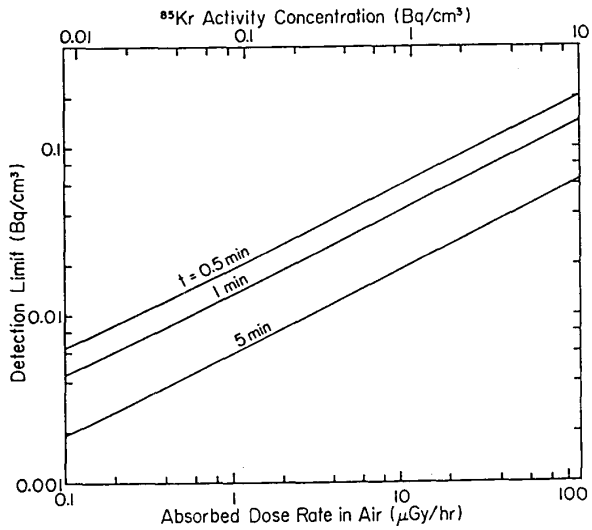

Fig. 7. Detection limits for tritium concentration as a function of absorbed dose rate in air by ^{137}Cs γ-rays or of ^{85}Kr activity concentration, which were calculated for measuring time t of 0.5, 1 and 5 min with typical values of k=1.2 for ^{137}Cs γ-rays and k=1.4 for ^{85}Kr β-rays.

where t is the measuring time. Fig. 7 shows detection limits fN_m for tritium concentration as a function of absorbed dose rate in air by ^{137}Cs γ-rays or of ^{85}Kr activity concentration. It is seen from fig. 7, with a measuring time of 30 sec, we can obtain a low detection limit of less than 0.2 Bq/cm^3 at a maximum acceptable γ-ray dose rate of 100 Gy/hr in air or a ^{85}Kr concentration level of 10 Bq/cm^3.

CONCLUSIONS

We devised a highly sensitive tritium-in-air monitor using flow-through air proportional counters as the detector. The counters need no counting gas other than the sampled air, and the monitor is possible to detect tritium β-rays separately from background radiations by using α-background discrimination and β- and γ-background compensation. Since the monitor has a function to adjust the compensation factor automatically, background compensation was carried out correctly independent of the details of the background radiations.

Data acquisition every 30 sec and detector control were carried out through a personal computer, where the high voltage applied to the counters was adjusted automatically according to the data of the pressure and the temperature of the sampled air. The computer calculated and printed out tritium concentration, dose equivalent rate by external γ-rays or β-activity concentration by other nuclides than tritium, and α-activity concentration in real-time.

Detection limit for tritium concentration was as low as 0.2 Bq/cm^3 with a measuring time of 30 sec under a γ-background with a dose rate level of 100 μGy/hr in air or a β-background from radioactive gases other than tritium with a concentration level of some 10 Bq/cm^3. Since this monitor can also respond to an accidental release of tritium with a concentration up to 5000 Bq/cm^3, it would be used effectively to monitor tritium in work locations of fission and fusion reactors, and of their related facilities.

REFERENCES

[1] R. A. Jalbert, "Monitoring tritium in air containing other radioactive gases," Los Alamos National Laboratory report LA-9514-MS, September 1982.

[2] T. Aoyama, H. Sugiura and T. Watanabe, "Application of air proportional counters to a tritium-in-air monitor," Nucl. Instr. and Meth., vol. A254, pp. 620-626, 1987.

[3] T. Watanabe, C. Mori, T. Aoyama and P. Christmas, "Dependence of the plateau threshold voltage of a 4πβ-proportional counter on filling gas pressure," Nucl. Instr. and Meth., vol. 178, pp. 121-124, 1980.

[4] T. Aoyama, M. Totogawa and T. Watanabe, "Influence of water and ethanol vapors on the operation of air proportional counters," Nucl. Instr. and Meth., vol. A255, pp. 524-530, 1987.