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Tritium(^{3}H) Activity Measurement by the Liquid Scintillation Counting Method

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Abstract

At a nuclear power plant, environmental radioactivity monitoring is routine work for the radiation safety management. For the environmental monitoring of tritium(^{3}H) activity in water samples, liquid scintillation counting(LSC) method is applied to measure low-energy beta activity from tritium in the samples. The ^{3}H activity is measured using the BECKMAN 5801 system at the KRISS(Korea Research Institute of Standards and Science) for evaluating the lower limits of detection(LLD) of ^{3}H measurement and the measuring capability of low-level ^{3}H activity at four nuclear power plant sites. The LSC systems used for low-level ^{3}H activity measurements at the nuclear power plants are confirmed to satisfy throughout an intercomparison study under the experimental arrangements by the KRISS.

1. INTRODUCTION

Tritium(^{3}H) is a radioactive isotope of hydrogen. It decays with the emission of an electron with mean energy of 5.75 keV to from ^{3}He : $^3H \rightarrow {}^3He + \beta^-$. The maximum energy of beta-particle emitted from tritium decay is 18.6 keV and its half-life is 12.35y(NCRP, 1985).

Quantitative detection of low-level ^{3}H has been considered to be very difficult owing to its low energy and interference of the background(Ishigawa et al., 1979). For the detection of low energy beta-emitting radionuclides such as ^{3}H, the liquid scintillation counting(LSC) method offers potentially a great sensitivity than the gas-filled counting technique because of the capability of the detector to accept a large sample size derived from the high density of the liquid phase(Iwakura et al., 1979). The electron spectrum and decay scheme for tritium is displayed in Fig. 1(Evans, 1958).

It is well known that a fraction of tritium escapes into the effluents of nuclear power plants and enters the environment. Much of the tritium that remains in the environment exists as the tritiated water, HTO and T_2 or HT gas.

At present, there are nine nuclear power plants under the commercial operations in Korea and seven others are additionally under construction by the national energy policy. Accordingly, a real atomic era is expected as we enter the 2000s. Under such circumstances, the KRISS has estabilished an experimental standard procedure for measuring tritium activity in samples from nuclear power plants by the LSC method. By the method, problems related to sample self-absorption, the attenuation of beta-particles by detector windows, and beta-scattering from a detector(Ishigawa et al., 1973) are completely eliminated. These advantages are particularly important for low energy radiation such as beta-particle emitted by tritium. Because tritium is important in chemical and biomedical applications, much of development of the

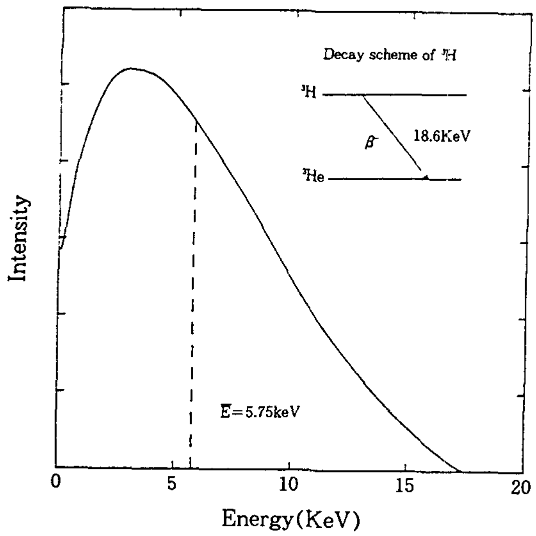

Fig. 1. Electron emission spectrum and decay scheme for tritium

LSC method has taken place in connection with several branches of sciences.

2. MATERIAL AND METHODS

2. 1 Principles

Several texts have been published which throughly review the fundamental principle of liquid scintillation counting(Wang et al., 1975). In the basic theory, liquid scintillation involves the detection and counting of radioactive decay. The radioactive sample to be counted is mixed with a scintillation cocktail which usually contains a solvent, an emulsifier, and a fluor. The cocktail serves to convert the energy of beta- particle emitted during the radioactive decay process of tritium into light, which is subsequently detected by PMTs. The light produced is the result of the following interactions :

$$\text{Radioactive decay} + \text{Solvent} \rightarrow \text{Solvent}^* \quad (1)$$

$$\text{Solvent}^* + \text{Solvent} \rightarrow \text{Solvent} + \text{Solvent}^* \quad (2)$$

$$\text{Solvent}^* + \text{Fluor} \rightarrow \text{Solvent} + \text{Fluor}^* \quad (3)$$

$$\text{Fluor}^* \rightarrow \text{Fluor} + \text{Light} \quad (4)$$

Note : * =excited molecule

To use the LSC method, samples in scintillation cocktails are placed in vials. In the interaction of Eq.(4), the amount of light being emitted from the vial is proportional to the energy of beta- particle. The higher the energy, the more solvent molecules are able to be excited, because they travel farther in the solvent. Therefore, more light is generate.

2. 2 Method

The light in Eq.(4) is directed to photomultiplier tubes(PMTs) which convert it into a measurable electrical pulse. The pulses are analyzed, converted to digital form, and stored in the appropriate channel of a pulse height analyzer(PHA), which corresponds to the beta- particle energy. The data in the PHA can be used to determine the energy of the particles from the sample and the rate in cpm of the radioactive decay in the sample. A simplified block diagram of LSC system shown in Fig. 2.

This LSC system is characterized by a digitized and summed coincidence pulse and designed to provide highly accurate, automated counting of the low- level of radioactivity in the environmental samples. The system is a bench- top, microprocessor- controlled spectrometer for radionuclide measurements.

To use the liquid scintillation instrument, the samples in scintillation cocktail are placed into the sample changer of the instrument. During the operation, the racks are moved in a counter- clockwise direction in the sample changer until reaching the right rearmost position.

The scintillation cocktail for the samples are prepared as follows :

(1) 1L of tolune, 5g of PPO(2,5- diphenyloxazole), and 0.3g of POPOP(1,4- bis- 2- (5- phenyloxazolyl)) are well mixed and dissolved.

(2) 500mL of APE(alkylphenoxy- polyethoxy- ethanol) is added in to the solution of (1) and leave it alone until the transparent state appears.

(3) Put the solution of (2) into the brown- color bottle and store at the cool and dark place for later use.

Meanwhile, the measuring samples are prepared as follow :

(1) Sample quantity is accurately weighed and put into low K- borosilicate glass vial.

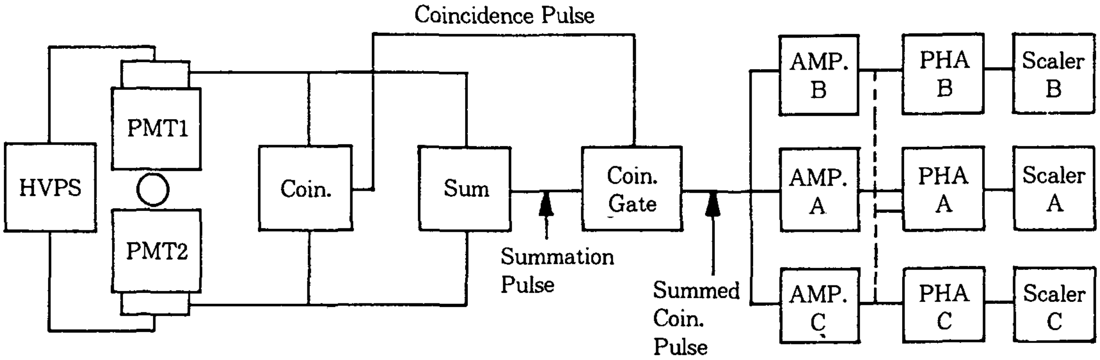

Fig. 2. A block diagram of LSC system(BECKMAN 5801)

(2) For a 20mL vial, the sample quantity of 8mL and the scintillator quantity of 12mL are put together.

(3) The vials containing the measuring sample from the solution of (2) are well shaked and heated in the 50℃ water bath.

(4) The vials of (3) is stored at the cool and dark place for 24 hours before the LSC measurement takes place.

2.3 Measurement

For the tritium activity measurements, a certified reference material(CRM) from the Laboratoire de Metrologie des Rayonnements Ionisants(LMRI), Saclay, France was used for the low-level tritium samples. The tritium activity of the LMRI's CRM was 28.69MBq/L as of August 1, 1991. This CRM solution from the LMRI was diluted with the distilled water to prepare samples for measurement.

Table 1. Comparison of tritium activity (Reference date: August 1, 1991)

Sample	Activity given (Bq/L)	Activity measured (Bq/L)	Difference (%)
1	87.58	85.98	−1.88
2	146.77	151.14	+2.90
3	180.95	183.64	+1.48
4	203.49	206.87	+1.65
5	284.18	278.82	−1.88

By using pycnometer(KRISS made) and microbalance(METTLER M5), five samples were prepared. The tritium activity of each sample is compared in Table 1. in terms of activity given by the LMRI and activity measured by the internal standard method at the KRISS.

Based on this measuring capability of the KRISS, the tritium water samples were distributed to the four nuclear power plants as shown in Table 2.

Table 2. Specifications of tritium water samples

Tritium sample \ Nuclear power plant	A	B	C	D
No. 1(mL)	34.06	31.29	30.32	34.14
No. 2(mL)	33.07	34.02	30.62	34.32
No. 3(mL)	32.72	39.23	32.06	32.67
Blank water(mL)	32.68	33.93	32.83	37.66

In the table, the blank water was provided by the Japanese Center for Analytical Chemistry(JCAC) for this paticular study. The tritium activity concentration of the blank water was 0.5Bq/L at the reference date. Tritium samples were prepared using the tritium CRM(1.123Bq/mL as of August 27,1987) from the National Institute of Standards and Technology(NIST). The activity concentration of each sample was No.1: 8.28Bq/L, No.2: 38.41Bq/L, No.3: 68.43Bq/L, respectively, as of November 1, 1991.

3. RESULTS

The experimental results of intercomparison study conducted under the KRISS's coordination are given in Table 3. The lower the limit of detection(LLD) was calculated from the following equation(IAEA, 1989):

$$C_{min}=\frac{k}{\varepsilon V_2}\sqrt{(R_0/t_s)(1+t_s/t_0)}\,e^{\lambda\Delta t}$$

where

C_{min}: minimum detectable activity concentration
k: coefficient of confidence
R_0: mean counting rate of blank water samples
ε: counting efficiency
V_2: volume of sample of blank water
t_s: summed counting time of the sample vials
t_0: summed counting time of the blank vials
λ: decay constant
Δt: time elapsed from sampling to counting

Table 3. Results of tritium measurements(Reference date: November 1, 1991)

Tritium sample \ Nuclear power plant	A	B	C	D
No. 1(Bq/L)	7.39±1.17	12.47±3.41	12.74±3.41	9.22±6.57
No. 2(Bq/L)	32.50±1.25	33.19±3.49	50.49±4.53	36.03±6.86
No. 3(Bq/L)	58.83±1.33	67.85±3.61	76.91±5.60	70.87±6.87
LLD (Bq/L)	6.61±2.43	7.18±2.45	7.12±2.38	7.97±2.73

4. CONCLUSION

With a commercial liquid scintillation counter, direct scintillation counting is therefore preferable for tritium activity concentrations higher than approximately 20Bq/L.

With reference to the LLD values for the tritium water samples, the Ministry of Science and Technology(MOST) in Korea points out the LLD value of 37Bq/L and the Radiological Environmental Monitoring Program(REMP) in U.S.A. 18.5Bq/L, respectively. As shown in Table 3, the four nuclear power plants have demonstrated their capabilities to be able to measure LLD values well below the LLD value designated by the REMP as well as the MOST. In this regard, the LSC systems used for tritium measurements at four nuclear power plants have been turned out to satisfy the LLD requirements for tritium.

And also, the measured tritium activity concentrations shown an agreement with a reference values.

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