A New Measurement Method of Tritiated Water with No Radioactive Waste- for Monitoring Post -15050

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ABSTRACT

A new measurement method has been developed by using a conventional liquid scintillation counter (LSC) with plastic scintillator pellets (PS-pellets) which is used as an alternative material of liquid scintillator. The measurement efficiency (%=cpm/dpm×100) of PS-pellets with a conventional LSC is as high as using a liquid scintillator. Measurement efficiency of tritiated water was more than 50% by using PS-pellets for LSC without liquid scintillator. Qualitative and quantitative analysis were possible by using PS-pellets for LSC, because the PS-pellets method showed good linearity between the activity and count rate. Also, an end point of a spectrum showed a maximum energy of a radionuclide when one channel estimated 0.5 keV. The PS-pellets were reusable by dipping them in 5% soapsuds of hot water for more than overnight except for a case of measuring for a long elapsed time. So, no radioactive waste of liquid scintillator and PS-pellets is produced with this method. We designed a new field monitor and a monitoring post.

INTRODUCTION

A LSC technique is a superior method to detect and measure beta-ray emitters because of its 4-π detection. It means high measurement efficiency is invested with a LSC for low energy beta emitters. On the other hand, a LSC technique has a demerit of production of radioactive liquid fluid wastes (WASTE) because of using organic liquid scintillator. So, some researchers studied no-waste product methods like plastic scintillator (PS) use, where they used as an alternative method to liquid scintillator [1-4]. Unfortunately, these experiments were not so successful because all these methods did not achieve high measurement efficiency, especially for tritiated water.

Measurement of tritium labeled compounds was successful by using PS-sheets (PS-sheets method) [1]. The measurement efficiency using PS sheets was as high as using liquid scintillator. Also, an identification of radionuclide was possible by the PS sheets method because there was no chemical quenching of the fluorescence. Furthermore, the PS sheets method produced no WASTE. However, PS-sheets method did not work appropriately for unstable compounds like tritiated water due to the nature of vaporizing. Tritiated water measurement is very important for our environmental safety. So a measurement method with high measurement efficiency for a short measurement time is required, especially in field monitoring.

We studied a new measurement method for tritiated water with a new type PS and achieved high measurement efficiency. We report a new PS method and propose a future design of monitoring.

EXPERIMENTAL

A LSC used was Tri-Carb3110TR (PerkinElmer) and the measurement time was 2 min for each vial. Radioactive sample used was mainly tritiated water (Water, [H-3] Biological Grade: Moravek Biochemicals, Ins. 925 MBq g^{-1}). The radioactive concentration of sample was approx. 240 kBq/mL. Mainly, the radioactivity of each vial was approx. 1.2 kBq with 5μL.

Figure 1 shows a diagram of the new measurement method (PS-pellets method). Key points of the method are as follows: 1. Tritiated water is put in a low potassium glass vial which is filled with plastic scintillator pellets (PS-pellets: EJ-200, G-tech Co. Japan) for LSC measurement. 2. The size of the pellet is approx. 3 mm in a length and 3 mm in a diameter. 3. Soon after the sample is applied to the pellets, the vial is screwed a lid on, which has a cone gum inner to avoid leaks (uGV2, Meridian Biotechnologies Ltd.).

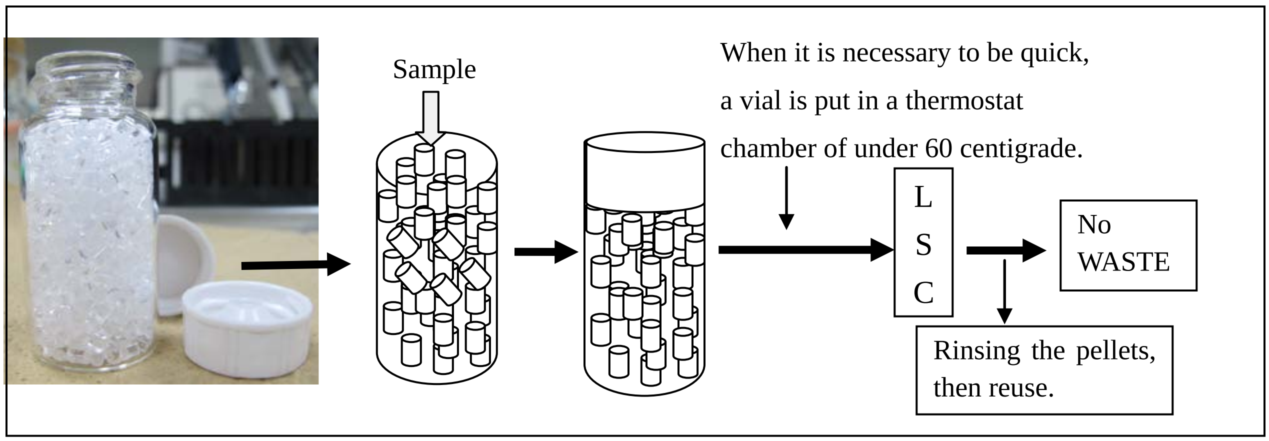

Fig.1 A diagram of the measurement procedure.

For the PS pellets method by using LSC, 5 factors were studied as follows: 1) Relationship between measurement efficiency and elapsed time, 2) Relationship between count rate and activity, 3) Detection limits calculated with the back ground counts of PS pellets, 4) Possibility of reuse of the pellets and 5) Possibility of qualitative analysis. For the efficiency calculation, the activity (dpm) was measured with liquid scintillator (LS: ACS-2, GE Healthcare Life Sciences Japan) because a standard vial was not prepared until then. Measurement efficiency (Eff.) and detection limit count rate (n_D) were defined as follows [5]:

$$\text{Eff. (\%)} = \text{cpm/ dpm} \times 100 \qquad \text{(Eq. 1)}$$

$$n_D\,(\text{cpm})=\frac{k^2}{2}\left[\frac{1}{t_S}+\sqrt{\frac{1}{t_S^2}+\frac{4n_B}{k^2}\left(\frac{1}{t_S}+\frac{1}{t_B}\right)}\,\right] \qquad \text{(Eq. 2)}$$

where k is a coverage factor corresponding to a confidence interval of 95% when k equals 2. The t_S, and t_B are measurement time of the sample and back ground, respectively. Also the n_B is the count rate of back ground. A detection limit activity concentration (A_D) was defined as:

$$A_D\ (\text{dpm mL}^{-1}) = n_D/\ \text{Eff}\ /100\ /\ \text{sample volume (mL)}. \qquad \text{(Eq. 3)}$$

RESULTS AND DISCISSIONS

Relationship between measurement efficiency and elapsed time: Fig. 2 shows measurement efficiency of 5 μL tritiated water with LS and PS pellets. The measurement efficiency after 4 h applied to PS pellets was higher than that of LS. When waiting 4 h is too long, the time becomes short for 10 min to put assemblages into a thermostat chamber of under 60 centigrade. It is considered that 5 μL tritiated water is necessary for 4 h at room temperature to vaporize and scatter in the 20 mL vial. Also the measurement efficiency agreed well among 5 vials by using cone gum inner lids. On the other hand, normal aluminum-cork inner lids caused leak even when silicon tape was used on a screw as a seal. So the key of the PS-pellets method is to avoid leaking among measurement. For this purpose, the lid with a cone gum inner was effective.

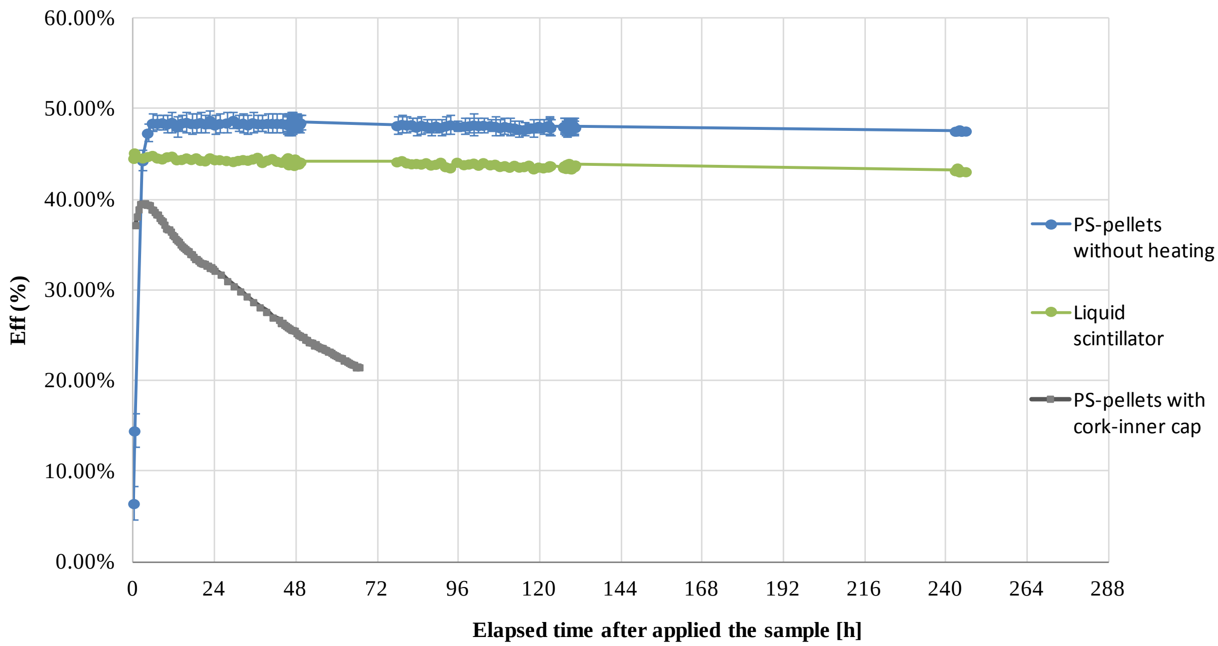

Fig. 2. Relationship between measurement efficiency and elapsed time.

Relationship between count rate and activity: Fig. 3 shows the good linearity between the count rate and activity of tritiated water. In this case, the sample used was 50 μL and the activity was from 76 to 6600 Bq mL^{-1}. When the sample used was 50 μL, the measurement efficiency was approx. 25% for every concentration. It means that the PS-pellets method can use quantitative analysis, however, it is necessary to use same volumes among samples because the measurement efficiency depends on the total volumes applied to the PS-pellets.

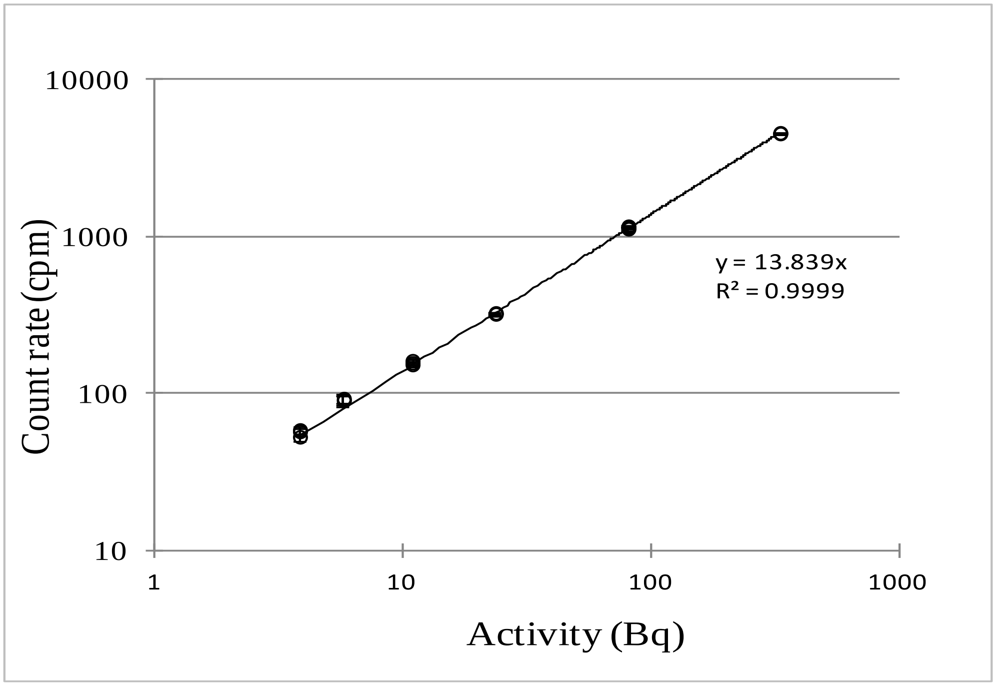

Fig.3. Relationship between count rate and radioactivity by using PS-pellets:

Detection limits: Fig.4 shows the detection limits calculated with the above equations by using the

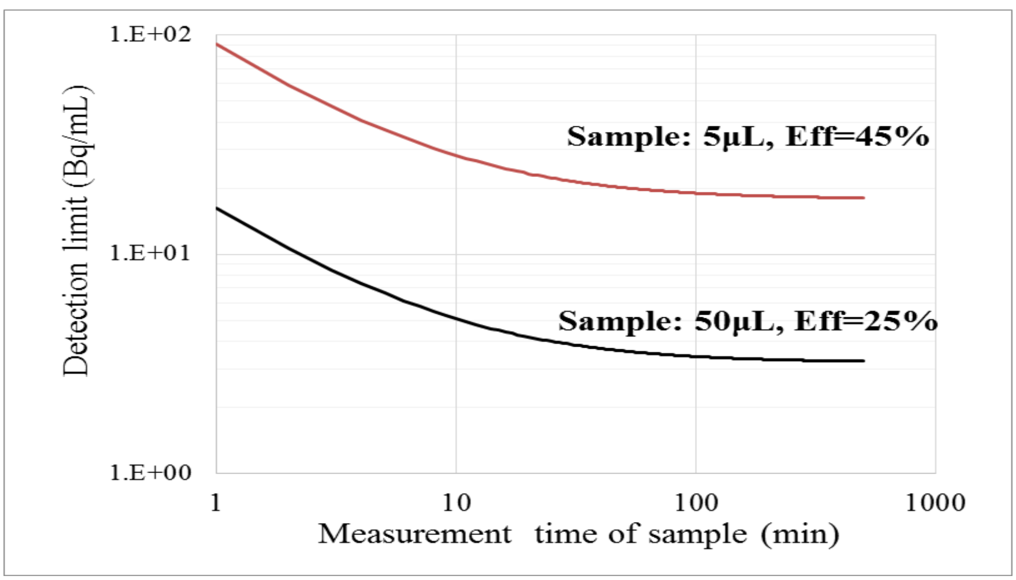

Fig.4. Detection limits of tritiated water with the PS-pellets method.

background (BG) of 15 cpm. When using k=2, the measurement time of BG was 10 min and measurement efficiency was 45% for 5μL of sample, 28 Bq/mL of tritiated water was measurable with 10 min by PS-pellets in a vial for LSC. Also, 5 Bq/mL measurement was possible when 50 μL tritiated water applied to the PS-pellets with the same condition. It means the PS-pellets is possible to detect the regulation limit of Japan: 60 Bq/mL. On the other hand, it is the limit of the detection by using a 20 mL vial for conventional LSC. So, a new larger scale detector is necessary to measure more volume and decreasing a detection limit for this PS-pellets method.

Possibility of reuse of the pellets: The PS-pellets were rinsed under the running water, and put in a beaker with 5 % soapsuds (DECON 90, Decon Laboratories Limited.) for overnight, which method was effective to reuse the PS-sheets. However, several ten thousand cpm became a hundred cpm for the PS-pellets. So, the soapsuds was warmed approx. 40 centigrade, the assemblage cpm became several ten cpm. However, the BG of PS-pellets was approx. 15 cpm. We have a plan to shake PS-pellets for longer time.

Possibility of qualitative analysis: Fig.5 shows spectra of the PS-pellets method (average of 3 spectra) and that of LS. The spectrum with LS was shifted to low energy region because of quenching and the maximum energy showed approx. 12 keV where 0 count started continuously. On the other hand, all 3 spectra with PS-pellets were not shifted and they showed their maximum energy of 18.5keV. It means a quantitative analysis is possible by the PS-pellets method except for mixture of radionuclides.

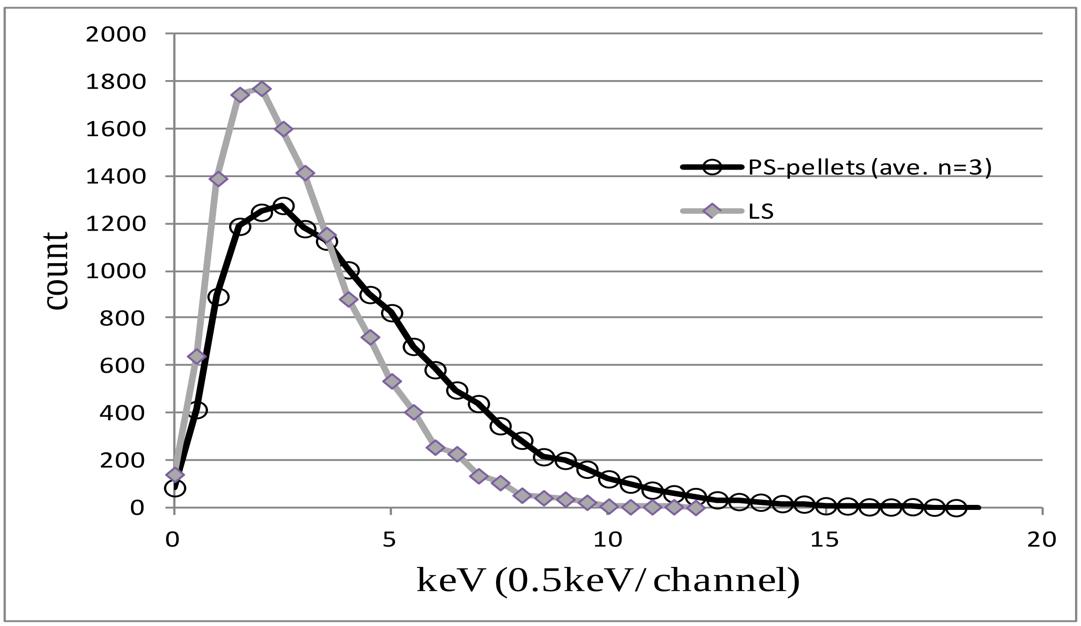

Fig.5. Spectra of tritiated water measured with the PS-pellets method and liquid scintillator.

Factors mentioned above worked for other compounds and radionuclides: e.g. H-3 or C-14 labeled sodium acetic acid.

FUTURE PLANS

Two types of new field monitoring equipments are designed: A) is like a handheld dosimeter and B) is large scale equipment which is a monitoring post. A mechanism of A) is nearly equal to a conventional LSC. However, a container which is filled with PS-pellets can be set on/off to equipment and the volume of it is more than 100 mL. Also, a distance between the container and detectors of 2 photomultiplier tubes (PMs) is nearer than that of a conventional LSC. The container and PMs are put in a dark box with a thin shield. The box is able to bring to a field like a handheld dosimeter.

Fig.6 shows a schematic diagram of B) as a monitor and the volume of a container of PS-pellets is more than 1 L. It connects with a sample box and a pump to circulate samples or cleaning water.
Because measurement efficiency of the PS-pellets method depended on the volume of containers (sample volume), equipments of different scales need to be studied for the best conditions as a next step.

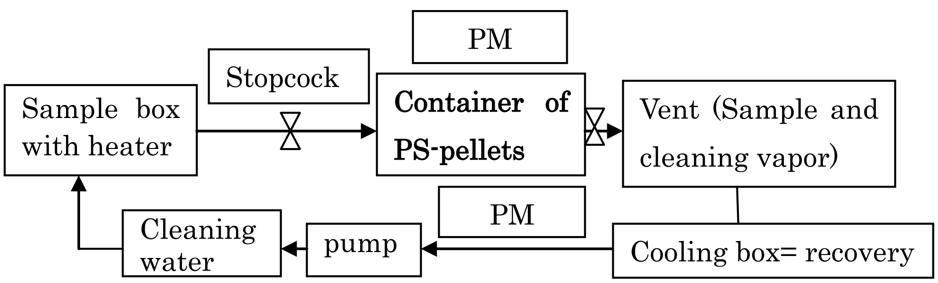

Fig 6 Schematic diagram of a monitoring post.

CONCLUSION

Without liquid organic waste fluid, tritiated water was measurable by using plastic scintillator pellets (the PS-pellets method) with a liquid scintillation counter. The measurement efficiency of the PS-pellets method was as high as that of liquid scintillator. Reproducibility of the pellets measurement was good. Also, the PS-pellets method was able to identify radionuclides with their spectra. The pellets are reusable to be soaked in a hot water for around 10 min with a shaker. The pellets are able to use other tritium labeled compounds and other radionuclides. The PS-pellets method is suitable to a handheld dosimeter for a new field detector and a monitoring post for environmental researches.

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