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Original Article

Radioactivity analysis for EPS waste using organic solvents

Jun Woo Bae [a], [**], Hee Reyoung Kim [b], [*]

[a] Korea Atomic Energy Research Institute, 34057, Republic of Korea
[b] Department of Nuclear Engineering, Ulsan National Institute of Science and Technology, 50, Unist-gil, Eonyang-eup, Ulju-gun, Ulsan, 44919, Republic of Korea

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ABSTRACT

In this study, the recovery rates of the dissolution method for radioactivity analysis of expandable polystyrene (EPS) with a liquid scintillation counter (LSC) using tetrahydrofuran (THF), toluene, and acetone as solvents were estimated. The detection efficiency calibration curve for each solvent was derived. Two methods—the volumetric ratio method and the quenching agent method—were used to prepare quench source sets, and calibration curves were derived by linking the data from the two quench source sets. The R^2 value of the calibration curve for THF was found to be 0.984. The relationship between the mass of dissolved EPS and the quench level was estimated: the quench level increased as the mass of dissolved EPS increased. Premix and postmix dissolution methods were tested. The recovery rates using THF with the premix method were 84.9 ± 0.9% and 96.5 ± 1.5% for ^3H and ^{14}C, respectively. Furthermore, the stability of the recovery rate over time when using THF was evaluated. The dissolution method with the premixed solution exhibited a more stable recovery rate over time. The dissolution methods were found to be applicable for analysis using LSC, and THF was found to be the most suitable solvent for the proposed method.

1. Introduction

Tritium (^3H) and radiocarbon (^{14}C) can be easily inhaled or ingested as tritiated vapor (HTO) and radioactive carbon dioxide ($^{14}CO_2$), respectively [1,2]. In particular, ^3H and ^{14}C are generated during the operation of nuclear power plants; thus, HTO and $^{14}CO_2$ are always present within power plants, and they pose the risk of contamination [3,4]. In principle, all items that are carried into a radiation management area are inspected for radioactivity before they are carried out. The inspection is performed using a survey meter or a surface contamination inspector; however, these devices may not be able to detect pure low-energy beta emitters such as ^3H and ^{14}C. HTO and $^{14}CO_2$ pose contamination risks with porous materials, such as expandable polystyrene (EPS; Styrofoam). Upon generation, EPS is generally removed from nuclear power plants; however, EPS waste may be generated during the delivery of equipment or consumables. In these cases, radioactivity analysis is required, and such EPS waste can be considered as clearance-level waste. In particular, the contamination risk may be greater if the pollution is difficult to control, such as during decommissioning of nuclear power plants.

In general, organic compounds such as polystyrene, a raw form of EPS, are pretreated via thermal decomposition and oxidation at high temperatures [5]. In this process, the sample is loaded onto a sample boat and gradually heated at programmed intervals in an air or oxygen atmosphere. However, in case of EPS, high-temperature oxidation poses the risk of explosion and back-pressure owing to the production of volatile organic compounds [6–8]. Thus, heating should be conducted in an even more gradual and programmed manner; however, this would increase the manpower and time required for analysis. In addition, the density of EPS is low; thus, the mass of the sample will be low, and accordingly, the detection limit or minimum detectable activity (MDA) may be high. Therefore, it is necessary to develop an alternative pretreatment method for radioactivity analysis of EPS samples.

In this study, a dissolution method was developed for the analysis of ^3H and ^{14}C, and its applicability was evaluated. Given that EPS is not soluble in diluted inorganic acids or oxidants [9,10], several organic solvents were selected and tested. The efficiency calibration curves were derived based on quenching, which

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[*] Corresponding author.
[**] Corresponding author.

E-mail addresses: junwoobae93@kaeri.re.kr (J.W. Bae), kimhr@unist.ac.kr (H.R. Kim).

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involved mixing the selected solvents with a scintillation cocktail. Moreover, the recovery rates of ^{3}H and ^{14}C were estimated.

2. Materials and methods

2.1. Selection of solvents

Two aspects should be considered for selecting a solvent: solubility and chemical quench. First, the solvent must be able to dissolve EPS completely in a short time. If a long time is required to dissolve EPS, ^{3}H and ^{14}C would be removed during the dissolution process, and the recovery rate would decrease. Furthermore, the ability to dissolve vapor and carbon dioxide is preferred for capturing ^{3}H and ^{14}C, respectively. Second, the solvent should have an appropriate chemical quenching power. If the quenching power is too strong, the detection efficiency would decrease. However, if it is too weak, deriving the calibration curve using a different volume of solvent would be difficult, and it would not possible to derive a quench-efficiency calibration curve unique to the solvent, because other quenching agents would need to be used. Most aromatic hydrocarbons, ethers, and ketones can dissolve EPS. In this study, acetone, tetrahydrofuran (THF), and toluene were selected as representative solvents. Acetone is the most common solvent used for EPS as it can rapidly dissolve EPS. However, acetone is a quenching agent; thus, the detection efficiency can decrease if a liquid scintillator is used [11]. THF is a cyclic ether material that is used to dissolve EPS for further processing [12]. However, since it is volatile, the recovery rate may be limited. Nevertheless, THF exhibits a high capacity to dissolve EPS. Toluene has a strong ability to dissolve EPS and offers a high detection efficiency as well, owing to the low quenching effect where it is even used as a base material for scintillators [13,14].

The suitability of the solvents for measurement when mixed with the scintillator was evaluated, given that all three candidate solvents readily dissolve EPS. The following cases were determined to be unsuitable for measurement: (1) when the solvent is not mixed well with the scintillator and is therefore opaque, (2) when layer separation occurs, and (3) when the quenching effect is excessive.

To verify mixability, 5 mL of each solvent and 15 mL of the scintillation cocktail (Optiphase Hisafe 3 (OH3), PerkinElmer) were mixed in a 20 mL polyethylene (PE) vial and left for 24 h; subsequently, the associated radioactivities of ^{3}H and ^{14}C were measured using a liquid scintillation counter (LSC; Quantulus 1220, PerkinElmer). The LSC was calibrated using unquenched standards (PerkinElmer) with radioactivities of 3,165 Bq and 1,587 Bq for ^{3}H and ^{14}C, respectively. The endpoints of their spectra were 320 Ch and 630 Ch, respectively. Note that the signal pulse height was logarithmically allocated into channels during digital conversion; hence, the difference between the endpoints of the two spectra was 310 Ch. Accordingly, the channel window was set to 50–320 Ch for the measurement of ^{3}H and to 50–650 Ch for the measurement of ^{14}C. The coincidence bias was the trigger threshold level. There are two options for the LSC: low and high. The low bias is suitable for isotopes with low energy, such as tritium, and the high bias is used to improve the figure of merit (i.e., to lower the background counts) by rejecting low-pulse-height signals generated by Cerenkov photons due to fluorescence and the presence of beta particles ^{40}K in a glass vial. In this study, the "low" coincidence bias option was selected for ^{3}H, and "high" was selected for ^{14}C. A blank sample was measured for 90 min to determine the background counting rate. Each radioactive sample was measured for 30 min, and the spectral quench parameter (SQP(E)) was subsequently acquired. The counting rate for the sample with a high level of quench was expected to be ~700 cpm. Considering the number of counts, a counting time of 30 min was sufficient to lower the uncertainty to less than 1%.

SQP is a representative spectral index that indicates the level of quench. It represents the calculated endpoint channel of the spectrum. When photon loss occurs owing to chemical/color quench, the value of SQP changes because the spectrum shifts toward the lower channel. The expression (E) indicates "external," implying that the SQP was acquired using an external radiation source. In the case of tritium, based on experience with conventional tritium analysis in distilled water or nitric acid, the detection efficiency was ~30% when the SQP was 700, and it decreased sharply when the SQP was lower than 700. Acetone, THF, and toluene had SQP values of ~700, 740, and 800, respectively. Although acetone showed the lowest SQP, it was considered a useable solvent because it can be used for LSC measurement when the SQP is ~700.

2.2. Derivation of calibration curve

In general, the calibration curve is derived before analysis using LSC [15]. Radioactivity analysis using LSC is based on the interaction between the liquid scintillator and the ionizing radiation. Photons are produced by the aforementioned interaction and then detected using photomultiplier tubes. The radioactive sample and the scintillator are mixed in a vial, and the molecules of the radioactive sample interfere with the energy transfer from the radiation to the scintillator molecules. More quenching occurs depending on this chemical interference level; thus, the efficiency of the LSC should be corrected according to the degree of quenching. Because the degree of quenching varies with the solvent, it was necessary to derive efficiency calibration curves for each of the three solvents used in the study.

Two methods are available for developing a quench source set. The first involves the addition of a quenching agent, and the second involves the assignment of different volumetric ratios to the solution and scintillator [16,17]. When an aqueous solution such as water or diluted nitric acid is used, the latter method is suitable, given that the quench parameter changes significantly with changes in the solution volume. However, for an organic solution such as toluene, the volumetric ratio does not have a significant influence on the quench parameter. Therefore, in this study, both methods were used to derive the calibration curves for THF and toluene. Owing to the significant influence of acetone on the scintillation property, as mentioned in the previous subsection, the volumetric ratio method was employed for acetone.

The ^{3}H source was prepared by diluting a certified ^{3}H standard solution (Eckert & Ziegler) comprising T_2O diluted in deionized water. The ^{14}C source was prepared by dissolving ^{14}C-labeled glucose (PerkinElmer) in deionized water. Using LSC, the radioactivity concentrations were estimated to be 77.85 Bq/mL and 43.99 Bq/mL for ^{3}H and ^{14}C, respectively. The relative uncertainties of the prepared sources were 2.43% and 1.99%, respectively, at a 95% confidence level.

For the volumetric ratio method, toluene (J.T.Baker) and THF (J.T.Baker) were mixed with the scintillation cocktail in solvent-to-cocktail ratios of 4:16–10:10. Acetone (Showa Chemical Industry) was mixed with the scintillation cocktail in ratios of 2:18–8:12. In the quenching agent method, 1 M nitric acid was used as the quenching agent. Nitric acid was selected because it is widely used for oxidation pretreatment in analysis of ^{3}H and ^{14}C and because it can sufficiently change the quenching level even when used in small amounts. The volume of the scintillator cocktail used was up to 12 mL. The volume of nitric acid added was 0.0–2.0 mL, in intervals of 0.5 mL, and the remaining volume was filled with toluene or THF. Finally, 0.5 mL of the radioactive source was added to each vial, and the vial was shaken to ensure thorough mixing. It should be noted that both water and nitric acid molecules act as quenching agents; thus, the injected volume of the radioactive source was sufficient to reduce the SQP and determine the calibration curve. The detection efficiency (ε) was calculated using Eq. (1):

$$\varepsilon(\%) = \frac{(C_s - C_b)}{A \cdot V_A} \times 100 \tag{1}$$

where C_s, C_b, A, and V_A are the counting rates of the sample and background (cps), the radioactivity concentration, and the volume of injected radioactive source, respectively. All counting vials were measured in triplicates for 15 min using the LSC. The calibration curve was derived through quadratic fitting using the counting rates and the SQP(E) values obtained from the measurements.

Another experiment was performed to verify the relationship between the mass of EPS and SQP(E). First, 50 mL of each solvent was prepared, and 0.75 g of EPS was dissolved in it. Then, 6 mL of the dissolved solution for acetone and 8 mL for THF and toluene were transferred to each counting vial. These steps were repeated three more times, and a total of 3.0 g of EPS was dissolved. The prepared counting vial was measured using LSC. The counting time for estimating SQP(E) was set as 2 min, and the measurement of each vial was performed thrice.

2.3. Estimation of recovery rate

The recovery rate represents the fraction of radioactivity recovered through a given pretreatment method. In this experiment, radioactivity was intentionally injected into the sample, and the sample was assumed to be radioactive. Then, the extent of radioactivity that was recovered through the proposed dissolution method was evaluated. In particular, 3.5 g of EPS added to 100 mL of each solvent. For the dissolution of EPS in the solvents, 2 mL of a radioactive source was simultaneously injected. Moreover, two dissolution methods were compared. The first (Method A) involved dissolution of EPS in the solvent. Then, the dissolved EPS solution was injected into the PE vial, whereas the remaining volume of the vials was filled with the scintillation cocktail. The second (Method B) involved mixing of the solvent and scintillation cocktail in an 8:12 ratio for THF and a 6:14 ratio for toluene and acetone; the mixtures were then used to dissolve the EPS. The mixed solution was stirred for 10 min to complete the dissolution. For both methods, the recovery rate (R) was calculated using Eq. (2):

$$R(\%) = \frac{\textit{Collected radioactivity in vial}}{\textit{Injected radioactivity in sample}} = \frac{\left(\frac{C_s - C_b}{\frac{\varepsilon_{curve}}{100}}\right)}{A \cdot V_A \cdot \frac{m_v}{m_s}} \times 100 \tag{2}$$

where ε_{curve} is the detection efficiency calculated from the derived calibration curve, m_v is the volume of the solution injected into the PE vial, and m_s is the volume of the mixed solution containing the solvent, EPS, and radioactive source. It was noted that the experiments for deriving the calibration curve and the recovery rate were independent. The values of the parameters used in Eq. (1) were not used for determining the recovery rate, and all values were measured separately. The MDA was calculated for each experimental condition using Eq. (3) [18]:

$$MDA = \frac{2.71 + 4.65\sqrt{B}}{T \cdot V \cdot \varepsilon \cdot R \cdot \frac{m_v}{m_s}} \tag{3}$$

where B, T, and V are the background counts; the measurement time (in seconds), where the background counting time is equal to the sample counting time; and the mass (volume) of the sample, respectively.

With regard to THF, additional experiments were conducted to evaluate the stability of the extracted radioactivity. When the dissolution was completed, a portion of the sample was extracted and poured into a vial after 0, 30, 60, 90, and 120 min, and the radioactivity recovery rate variation with respect to time was estimated.

3. Results and discussion

Fig. 1 shows the calibration curves when THF, toluene, and acetone were used as solvents. The filled and hollow markers (both squares and circles) indicate the quenching agent and the volumetric ratio methods, respectively. With regard to THF, as shown in Fig. 1(a), the trends of the detection efficiency and SQP(E) obtained for the two methods were in agreement when the curve was derived using all the data points. Moreover, the R^2 values were 0.986 and 0.987 for ^3H and ^{14}C, respectively. When the volumetric ratio of the THF was increased, SQP(E) decreased. Moreover, when the nitric acid content was increased, SQP(E) decreased. The decrease in SQP(E) can be attributed to the addition of impurities due to the dissolution of EPS. Therefore, to utilize the center of the curve (SQP = ~740), THF:OH3 = 8:12, for which SQP was ~760, was considered a suitable condition for practical application of the dissolution method.

With regard to toluene, as shown in Fig. 1(b), the trend was nonuniform. When the volumetric ratio of toluene increased, both SQP(E) and the detection efficiency increased. However, when the ratio exceeded 8, the detection efficiency decreased, although SQP(E) increased. Given that toluene is hydrophobic, the water (and thus the radioactivity) was not homogeneously mixed in the PE vial; thus, the detection efficiency decreased. Toluene is a substance that can also be used as a scintillator base material; thus, it does not have a significant influence on the detection performance of OH3 [19–21]. Because toluene is hydrophobic, if its content increases, it would not mix well with the aqueous radioactive sources used in the experiment. It was speculated that a small fraction of the radioactive source was physically separated from the scintillation cocktail solution. Therefore, the detection efficiency decreased with increasing volumetric ratio of toluene, although SQP(E) increased. When nitric acid was added in volumes of 0.5 and 1.0 mL, the data points of the calibration curve became outliers, and the detection efficiency decreased, whereas SQP(E) increased. Some studies have noted that the detection efficiencies for ^3H and ^{232}U are low within the pH range of 2–4 [22,23], and the data points were included in this range. With the exception of the outliers, the data points were placed on the calibration curve derived through quadratic fitting. The trend was close to linear as the coefficient for x^2 was of the order of 10^{-4}. As a result, the data in the SQP(E) range of 780–800 were null, and the reliability of the curve was relatively low. The SQP(E) value of the mixed solution was ~790 when the EPS was dissolved in the 6:14 mixture of toluene and OH3 scintillation cocktail. In addition, for ^3H, an R^2 value of 0.997 was obtained; however, for ^{14}C, a low R^2 value of 0.938 was obtained. Considering these values, toluene was found to be unsuitable for practical measurement of dissolved EPS samples. With the exception of that for ^{14}C in toluene, the R^2 values of the fitting curves were greater than 0.98, and the curves were in agreement with the datasets obtained using both the volumetric and quenching agent methods. The hybrid calibration curve for THF can be employed in practical applications as it exhibited high R^2 values for both ^3H and ^{14}C.

With regard to acetone, as shown in Fig. 1(c), the trend was similar to the quench curve in the general LSC analysis. When the acetone content increased, both SQP(E) and the detection efficiency decreased. Given that SQP(E) ranged from 600 to 705, it was suitable to apply the calibration curve because the R^2 values of the

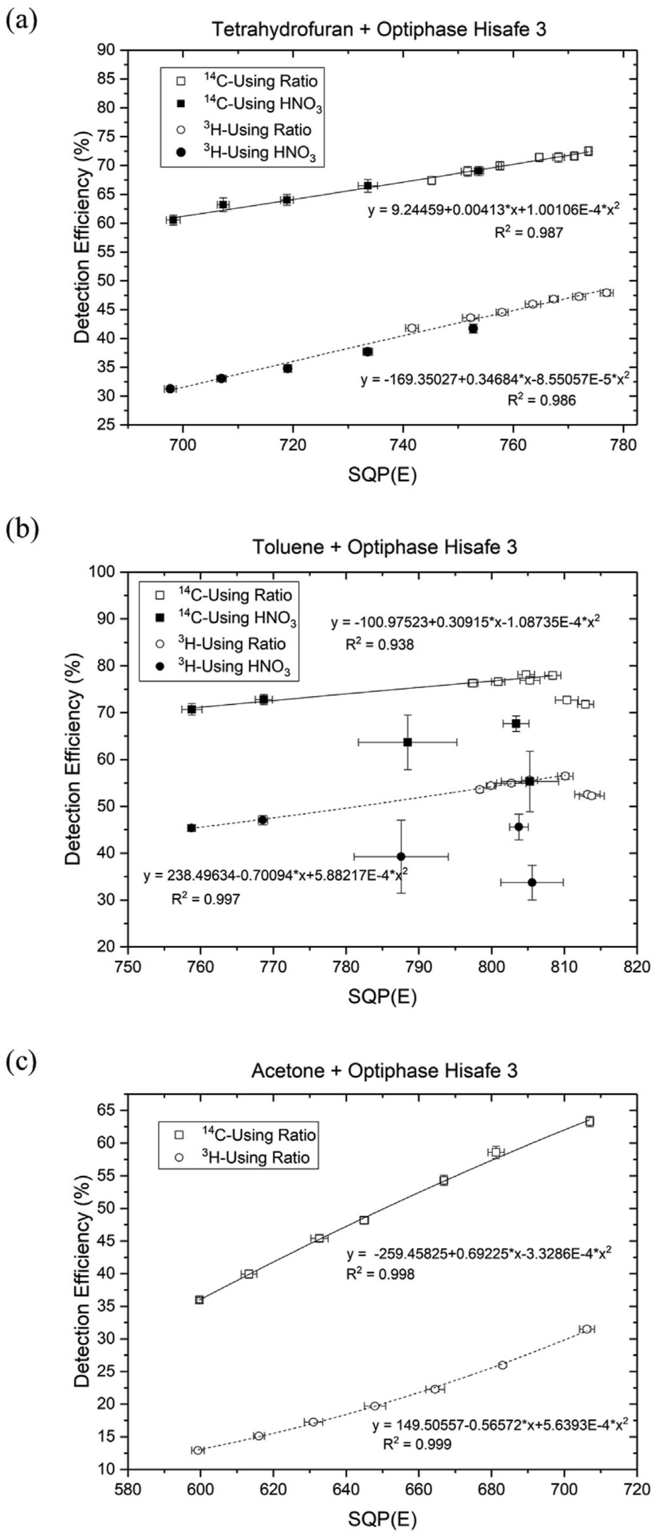

Fig. 1. Detection efficiency calibration curves when (a) tetrahydrofuran, (b) toluene, and (c) acetone were used as solvents.

curves for ^{3}H and ^{14}C were 0.998 and 0.994, respectively. The calibration curve for acetone, unlike those for toluene and THF, showed a quadratic trend because SQP(E) was lower than 700.

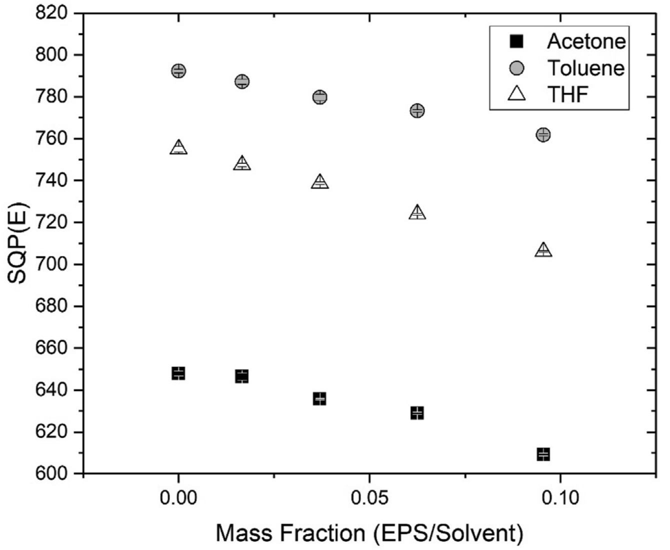

Fig. 2. SQP(E) variation according to mass fraction for three solvents.

Fig. 2 shows the variation in SQP(E) according to the different mass fractions for the three solvents. SQP(E) linearly decreased as the mass fraction increased. SQP(E) decreased by 38.7, 30.5, and 48.9 for acetone, toluene, and THF, respectively. Because 4/3 times more solvent had been injected into the counting vial for THF, this solvent exhibited the largest decrease. SQP(E) decreases because of the absorption of UV light by polystyrene [24,25]. The aromatic ring in polystyrene absorbs the wavelength of ~260 nm and can interrupt the energy transfer to the scintillation cocktail. In addition, commercial EPS, which can be bleached using fluorescent materials can also interrupt energy transfer. Therefore, it is necessary to maintain the same mass fraction of the solvent and the sample during practical analysis. If the group of EPS samples is changed, it is recommended to re-calibrate the detection efficiency.

Table 1 presents the detection efficiency, recovery rate, and MDA for each solvent. The background counting rates for THF, toluene, and acetone were 7.92 ± 0.21, 8.01 ± 0.72, and 7.98 ± 0.34 cpm for ^{3}H and 4.23 ± 0.16, 4.40 ± 0.13, and 4.40 ± 0.04 cpm for ^{14}C, respectively. The background counting rate showed no dependence on the solvents. The absence of data of toluene for Method A can be attributed to the hydrophobic nature of toluene; thus, a nonconstant detection efficiency and recovery rate were observed. Given that pure toluene had not been mixed with ^{3}H or ^{14}C, the detection efficiency and recovery rate could not be estimated. The detection efficiency followed the order toluene > THF > acetone, given that SQP(E) was the highest for toluene and the lowest for acetone. The recovery rate for acetone was relatively low and was the lowest under Method A. Overall, a higher recovery rate was observed for Method B than for Method A, and the scintillation cocktail under Method B acted as a stable mixing agent that could adequately contain the radioactivity. Although the detection efficiency and recovery rate were both high in toluene, the lower MDA in THF was due to the mixing ratio. THF was mixed in an 8:12 ratio, while toluene was mixed in a 6:14 ratio; this allowed for a greater volume of the THF samples to be contained in the PE vials. Toluene was found to be superior in terms of detection efficiency and recovery rate, which have a direct influence on detection. However, the usability of THF was higher in terms of the stability of the amount of solution to be transferred and the reliability of the calibration curve.

The recovery rate was evaluated over time to determine whether a rapid change occurs during the mixing of the EPS sample

Table 1
Detection efficiency, recovery rate, and MDA for each solvent.

Method	Isotope	Solvents	Detection efficiency (%)	Recovery rate (%)	MDA (Bq/g)
Postmix method (Method A)	^{3}H	THF	39.968 ± 0.034	84.7 ± 1.5	0.33
		Toluene	–	–	–
		Acetone	21.360 ± 0.034	68.5 ± 1.9	1.0
	^{14}C	THF	66.23 ± 0.14	95.7 ± 3.0	0.10
		Toluene	–	–	–
		Acetone	50.17 ± 0.24	45.74 ± 0.79	0.36
Premix method (Method B)	^{3}H	THF	37.310 ± 0.072	84.93 ± 0.91	0.35
		Toluene	46.95 ± 0.10	87.8 ± 1.1	0.36
		Acetone	15.465 ± 0.050	80.9 ± 1.9	1.2
	^{14}C	THF	64.579 ± 0.048	96.5 ± 1.5	0.10
		Toluene	69.785 ± 0.051	95.6 ± 1.3	0.12
		Acetone	39.181 ± 0.093	92.9 ± 1.7	0.22

and solvent, dissolution, and transfer to the PE vial, given that the solvents used in this study are volatile. Under the proposed dissolution technique, the total time required for the mixing and transfer to the PE vial was 10 min for Method A and 15 min for Method B, on average, given that Method A exhibited a higher dissolution rate. Fig. 3 shows the recovery rates for ^{14}C and ^{3}H with respect to time for the two dissolution methods. The hollow markers indicate Method A, wherein only the solvent was used to dissolve the EPS, which was followed by mixing with the scintillation cocktail. The filled markers indicate Method B, wherein the solvent and scintillation cocktail were first mixed, following which the solution was used to dissolve the EPS. Method A produced more significant changes in the recovery rate than Method B did. Under Method A, after 120 min, the recovery rate for ^{3}H decreased by 9.35% and that for ^{14}C increased by 8.34%; by contrast, under Method B, the recovery rate for ^{3}H decreased by 3.02% and that for ^{14}C increased by 6.65%.

Mixtures of water and THF have a high evaporation rate [26]. The OH3 scintillation cocktail was mixed thoroughly with water and THF as it is designed for aqueous samples [27,28]. The addition of OH3 reduced the evaporation rate; thus, the recovery rate, especially that for ^{3}H, varied significantly with respect to time. The recovery rate for ^{14}C increased with time as ^{14}C was not removed through evaporation or volatilization. This was because the source was ^{14}C-labeled glucose. If the carbon that constitutes the EPS is contaminated with ^{14}C or a surficial contaminant such as ^{14}C-contaminated lubricant oil, a similar trend will be observed. The small amount of $^{14}CO_2$ gas trapped in the pores of the EPS is considered soluble in toluene, THF, and acetone [29]. If $^{14}CO_2$ is not dissolved at saturation level, it is not removed with the solvent during volatilization. This involves physical dissolution, and not chemical dissolution, in the form of a carboxylate group or carbonate salt; thus, the solution may be sensitive to impact and should be handled as such [30].

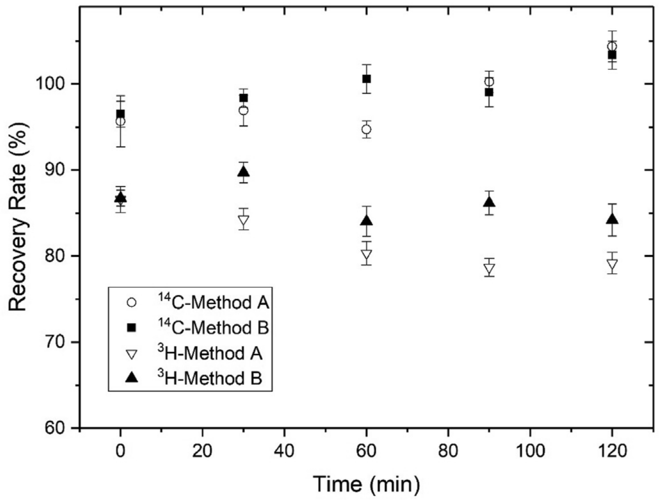

Fig. 3. Recovery rates for ^{14}C and ^{3}H with respect to time for the two dissolution methods.

Owing to the variation in the recovery rates with respect to time under both methods, the solution should be transferred to the PE vial immediately after complete dissolution of the EPS sample. The variations in the recovery rate with respect to time were smaller under Method B than under Method A. Method A, owing to its high dissolution rate, is advantageous a large amount of EPS is dissolved. Method B is recommended when the amount of EPS is the same or smaller than that considered in this study. For example, the dissolution of 3.5 g of EPS in 100 mL of solvent required ~5 min under Method A but 10 min under Method B. However, when 10 g of EPS was dissolved in 100 mL of the solvent, the dissolution time exceeded 30 min for Method B, which may affect the recovery rate. In this case, Method A is preferable. When using Method B to determine the level of radioactivity transferred to the vial, it is necessary to accurately measure the mass of the mixed solution, proceed with dissolution, and record the mass change before and after dissolution.

4. Conclusions

In this study, the recovery rates of the dissolution method for radioactivity analysis of expandable polystyrene (EPS) with a liquid scintillation counter (LSC) using three solvents—tetrahydrofuran (THF), toluene, and acetone—were estimated. EPS exhibited satisfactory solubilities in all three solvents. However, the detection efficiency calibration curve for toluene was found to be unsuitable owing to excessive quenching and a low recovery rate. The relationship between quenching and the mass of the dissolved EPS sample was also analyzed. The dissolved mass affects the quenching level; therefore, maintaining the mass fraction of EPS and the solvent is important for practical analysis. The stability of the recovery rate was estimated for the three solvents as well, and THF was found to be the most appropriate solvent for the proposed dissolution method. The Results of this study can serve as a fundamental database for LSC analysis of organic materials, such as oil and plastic, in addition to EPS. However, the recovery rate of the dissolution method was evaluated with respect to only one type of contamination situation. In future studies, we intend to evaluate the recovery rate using samples similar to those in actual contaminations. The results of such investigations can be applied to radioactivity analysis of waste generated during the operation of nuclear facilities and decommissioning of nuclear power plants.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.net.2021.06.008.

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