



Effect of ^3H and ^{14}C Interference on the Radioactivity Measurement of ^{129}I in Low and Intermediate-Level Radwastes†

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AJC-13629

The effect of ^3H and ^{14}C interference on the separation and quantification of ^{129}I from iron scraps and heating insulators as a low and intermediate-level radwaste has been studied. The iron scraps and heating insulators were liquidized using an acid digestion pretreatment method. Then, an anion-exchange resin adsorption method was used to separate the ^{129}I and ^{14}C in the pretreated aqueous solution. The ^3H remained in the aqueous phase after the back-extraction and was removed by low-temperature evaporation at 70 °C. Through the chemical separation and evaporation, most of the ^{14}C and ^3H were removed and no radioactivity was detected. In the separation extraction, the average recovery rate of ^{129}I was approximately 80.8 %.

Key Words: Low and intermediate level radwastes, ^{129}I , ^{14}C , ^3H , recovery.

INTRODUCTION

In Korea, researchers have been working on the development of an analytical method to detect α and β -emitters to aid in regulating the transportation and disposal of nuclear waste generated from nuclear power plants. Among these critical nuclides, ^{129}I is produced as a fission product in nuclear reactors and has a long half-life ($t_{1/2} = 1.57 \times 10^7$ y)^{1,2}. The significance of the production of this nuclide has been recognized due to its radio-ecological effects^{3,4}. This radioactive iodine isotope is also difficult to measure directly with a β counter because its specific activity is low (6.4 Bq/ μg)^{5,6}. For long-lived, β -emitting nuclides the requirement of radiochemical purity is stringent and the degree of purity is difficult to measure or control, particularly because many of the impurities are also pure β -emitters⁷.

In addition, ^3H , which is generated along with ^{129}I in a fission and activation process, emits weak-energy β radiation and is not easily removed by an ionic exchange or degasification method because it exhibits strong movement and diffusion⁸. Organic and inorganic ^{14}C is generated in nuclear facilities and inorganic ^{14}C is collected *via* adsorption and precipitation, but organic carbon is easily released into the environment⁹⁻¹⁰. The ^3H and ^{14}C isotopes can interfere with the measurement of ^{129}I because they emit in the same detective energy range as ^{129}I ¹¹.

In this study, iron scrap and heating insulator types of the simulated radioactive wastes were prepared to measure the radioactivity of ^{129}I present in the low- and intermediate-level radwastes. To investigate the interference of ^3H and ^{14}C in the measurement of ^{129}I using a liquid scintillation counter, ^3H and ^{14}C , which were added into a pretreated simulated acid solution, were separated and removed *via* ionic exchange and low-temperature evaporation methods.

EXPERIMENTAL

Reagent grade NaNO_3 (Aldrich) was used as acid digestion agent and KI was added as an ^{129}I carrier. The ^{129}I used to trace the radioiodine was first diluted in 2,138 Bq/mL, $\text{NaI}/0.1 \text{ M Na}_2\text{S}_2\text{O}_3$ (NIST-SRM-4949 c). An anion exchange resin (AG 1 \times 2, 50-100 mesh, Cl^- form, Bio-rad Co.) was used for iodine absorption after it was washed with a dilute acid. The radioactivity of the ^{129}I was measured using a liquid scintillation counter.

Application of the acid digestion method: Approximately 10 g iron scrap and heating insulator of the dry active radwastes were placed into a 500 mL polyethylene bottle into which 20 mL of 7.0 % NaClO and 1 mL standard KI solution (1000 $\mu\text{g}/\text{mL}$) were added. After mixing for 4 h using a shaker and filtering, a portion of the solution was used as a stock.

†Presented to the 4th International Symposium on Applications of Chemical and Analytical Technologies in Nuclear Industries, Daejeon, Korea

Separation of the ¹²⁹I using an anionic exchange resin:

The pH 10 buffer solution (5 mL), 10 mg KI and 5 g anionic resin (AG1-X 4 Resin) were placed into a plastic beaker and stirred with a magnetic bar for 12 h to absorb the iodine. The mixture was then filtered with Whatman # 42 filter paper and the aqueous solution was discarded. The resin was used to fill a polyethylene pipe 10 mm in diameter and 140 mm long. Next, the resin was washed twice with 10 mL distilled water. The iodine was eluted first with 0.2 M NaNO₃ at a flow rate of 0.4 mL/min, then with 2 M NaNO₃. The 10 mL eluted solution was poured into a 50 mL extraction tube and 1 mL HNO₃ and 1000 mg NH₂OH·HCl were added. The iodine was extracted for 3 min with the addition of 10 mL CHCl₃. The aqueous phase of the extraction solution was discarded and only 5 mL of the organic phase was placed into a new extraction tube. After adding 5 mL 0.1 N NaHSO₃ to the separated organic phase in the tube, the solution was back-extracted to the aqueous phase and the 1 mL aqueous phase was removed into a glass vial. The radioactivity was measured with an liquid scintillation counter after mixing with a 14 mL cocktail.

RESULTS AND DISCUSSION

Quench calibration: To determine the reliability for quench correction methods used in liquid scintillation counter, samples with known activity were measured using liquid scintillation counter. To determine the counting efficiency from the quench levels obtained during tritium counting, a 4.86 Bq ¹²⁹I standard (NIST-SRM-4949 c), a 73,059 Bq ³H standard (spec-check, Packard) and a 14,878 Bq ¹⁴C standard (spec-check, Packard) were each placed into 10 scintillation vials. As a quenching agent, nitromethane was also added in the range of 0~150 µL for ¹²⁹I, 0~100 µL for ³H and 0~200 µL for ¹⁴C.

Effect of ¹⁴C and ³H on iodine measurement via liquid scintillation counter: As a radioactive waste, iron scrap and heating insulator produced from a nuclear power plant contain a constant concentration of ¹⁴C and ³H, which may be several tens times higher than that of ¹²⁹I. Therefore, measurement methods of ¹²⁹I activity from a β-emission nuclide such as ¹⁴C and ³H was studied because ¹²⁹I is also a pure β-emission radio-nuclide and is not easy to be separated during pretreatment. Fig. 1 provides the β-spectra for several volatile radioactive nuclides (³H, ¹⁴C and ¹²⁹I) obtained using a liquid scintillation counter. As shown in Fig. 1, because the spectra of ¹⁴C, ³H and ¹²⁹I can overlap in the 250~450 channel band each other, ¹⁴C must be removed for accurate determination of ¹²⁹I.

Separation of ¹²⁹I using an anionic exchange resin:

A flowchart for the analysis process for the separation and quantification of ¹²⁹I in the iron scrap and heating insulator samples is provided in Fig. 2. The aqueous phase eluted by the acid leaching was loaded onto the anionic resin (AG1-X 4 Resin) to adsorb the ¹²⁹I and inorganic ¹⁴C. Then, the ¹⁴C was eluted from the column using a 0.2 M NaNO₃ solution. The ¹²⁹I remained in the column and was eluted using 2 M NaNO₃. Then, the ¹²⁹I in the eluted solution was reduced to I₂ by hydroxylamine hydrochloride (NH₂OH·HCl) and extracted into CHCl₃. The I₂ was back extracted by reduction to I⁻ with sodium bisulfite (NaHSO₃). The ³H was removed by evaporation at a temperature below 70 °C after removing the iodine in the

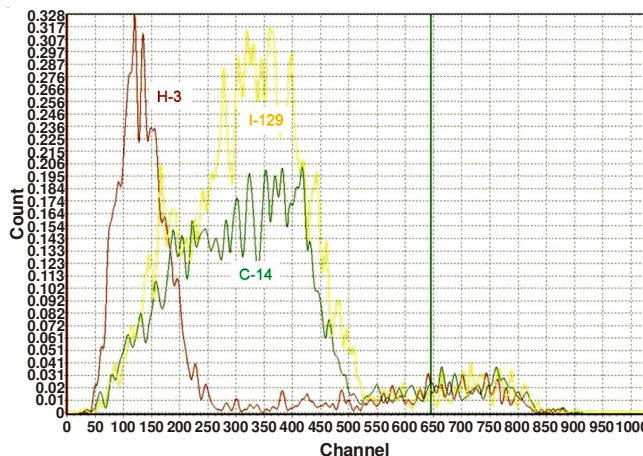


Fig. 1. Spectra of ³H, ¹⁴C and ¹²⁹I as determined by liquid scintillation counting

The affinities of various anions for the resin of AG-1 are in the order :

- I⁻ > phenolate > HSO₄⁻ > ClO₃⁻ > NO₃⁻ > Br⁻ > CN⁻ > HSO₃⁻ > NO₂⁻ > Cl⁻ > HCO₃⁻ > IO₃⁻ > HCOO⁻ > Ac⁻ > OH⁻ > F⁻

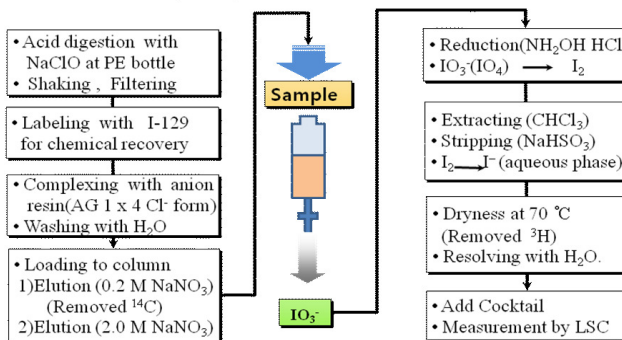


Fig. 2. Chemical procedure for separating ¹²⁹I in low- and intermediate-level radwastes

organic phase to an aqueous phase. The radioactivities for ¹²⁹I before and after ¹⁴C and ³H removal were compared in Tables 1 and 2. As shown in Tables 1 and 2, the evaporation procedure for the ¹⁴C and ³H did not induce co-evaporation of ¹²⁹I at temperatures below 70 °C. Table-3 provides the recovery rates for ¹²⁹I from a solution containing ¹⁴C and ³H standards after

Radioactivity (Bq/mL)			Recovery (%)	Remark
Add ¹²⁹ I	Add ¹⁴ C	Measured ¹²⁹ I		
4.36	0	3.62	80.0	Removed ¹⁴ C
4.36	100	5.01	114.9	Not removed ¹⁴ C
4.36	100	3.49	83.0	Removed ¹⁴ C

Radioactivity (Bq/mL)			Recovery (%)	Remark
Add ¹²⁹ I	Add ³ H	Measured ¹²⁹ I		
4.36	0	3.68	82.40	Remove ³ H
4.36	0	3.52	84.7	No evaporation
0	50	0.04	<0.1	Remove ³ H
4.36	50	18.60	37.2	No evaporation
4.36	50	3.55	81.4	Remove ³ H

TABLE-3
RECOVERY OF ^{129}I ON THE ^3H AND ^{14}C AS MEASURED
BY LIQUID SCINTILLATION COUNTER

Radioactivity (Bq/mL)				Recovery (%)	Remark
Add ^{129}I	Add ^{14}C	Add ^3H	Measured ^{129}I		
4.36	100	50	5.13	117.6	No column elution no evaporation
4.36	100	50	3.52	80.8	Removed ^{14}C , ^3H

separation and removal *via* anion exchange resin column elution or volatilization. As shown in Table-3, the ^{129}I recovery rate for the sample in which the ^{14}C and ^3H were not separated is 117.6%. In contrast, ^{129}I recovery rate measured after removal of the ^3H and ^{14}C was 80.8%.

Conclusion

To quantify ^{129}I , the radionuclide must be radiochemically separated from other β -emitting radionuclides, such as ^{14}C and ^3H because ^{129}I has a relatively low radioactivity in the radwaste compared with ^3H and ^{14}C . To identify the chemically pure separation of ^{129}I , simulated samples were prepared through the addition of ^{129}I , ^{14}C and ^3H to iron scrap and a heat insulator. The ^{14}C and ^3H were largely removed by radiochemical separation and evaporation and no radioactivity from the sample was detected. Through the separation and quantitative analysis experiments, an average recovery rate of 80.8% was achieved for ^{129}I added to the simulated sample.

ACKNOWLEDGEMENTS

This study has been carried out under the nuclear research and development program by the Ministry of Education, Science and Technology of Korea.

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