

DETERMINATION OF ^{129}I USING LIQUID SCINTILLATION COUNTING

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ABSTRACT. We developed a method for the determination of ^{129}I in environmental samples by liquid scintillation counting. Prior to analysis, ^{125}I was added as a chemical yield tracer, and aqueous samples were heated in 160 mL of 0.4N $\text{K}_2\text{Cr}_2\text{O}_7$ in 48% H_2SO_4 , and then distilled after adding 16 mL of 30% H_3PO_3 and 1 mL of 30% H_2O_2 . The distilled iodine was extracted into toluene, then back-extracted into the water phase with 1 mL of 4M NaOH solution. The water phase was dried and transferred into a 20-mL LSC Teflon[®] vial with 4 mL of distilled water and then mixed with 16 mL of Ultima Gold[™] LLT. ^{129}I and ^{125}I were simultaneously counted by a Quantulus 1220[™] LSC at room temperature. The activities of ^{129}I and ^{125}I were calculated by the 3-over-2-fitting method in the EasyView[™] software (Wallac, PerkinElmer). Counting efficiencies of ^{125}I and ^{129}I were 35.1–38.1% and 75.9–81.1%, respectively. The chemical yields of ^{129}I during distillation and during solvent extraction were between 65% and 95%. Analytical results of ^{129}I in the spiked water samples were in good agreement (within 5.3% relative error) with the known concentration of ^{129}I .

INTRODUCTION

^{129}I is regarded as a very important isotope due to its long-term dose from accidental releases and fallout from atomic bomb tests, its high mobility in stored radioactive waste (Buraglio et al. 2000), and its use as a new geochemical tracer in environmental science (Santschi and Schwehr 2004). The inventory of ^{129}I in surface environments has been significantly increased by anthropogenic releases over the past 50 yr (Buraglio et al. 2000). ^{129}I is an emitter of low-energy beta, gamma, and X-rays and is generally measured by accelerator mass spectrometry (AMS) or neutron activation analysis (NAA) (Hou et al. 2001). However, these methods require very expensive equipment and facilities and a time-consuming chemical separation. Therefore, we present a method for determining ^{129}I by liquid scintillation counting in order to simplify the determination of ^{129}I in environmental samples released during nuclear accidents.

MATERIALS AND METHODS

A trial water sample spiked with ^{129}I was prepared. KI and ^{125}I were added as an iodine carrier (1 mg as I⁻) and a radiological tracer of ^{129}I , respectively. The sample was heated with 160 mL of 0.4N $\text{K}_2\text{Cr}_2\text{O}_7$ in 48% H_2SO_4 and distilled after adding 16 mL of 30% H_3PO_3 and 1 mL of 30% H_2O_2 . The distilled iodine was transferred into 30 mL of 1.0M LiOH solution (Cox and Pickford 1992). The LiOH solution was acidified to pH 2 with concentrated HNO_3 and was transferred into a separation funnel. Unidentified materials (yellow color) in the water phase were extracted into toluene, which was discarded, and then new toluene was added into the separation funnel. The iodide in the water phase was oxidized by adding 1 mL of 30% H_2O_2 , and then extracted into the toluene (color changed to pink in toluene phase). The iodine was back-extracted into the water phase by adding 1.0 mL of 1.0M NaOH and 5 mL of distilled water. The water phase was then dried and transferred into a 20-mL LSC Teflon[®] vial with 4 mL of distilled water and subsequently mixed with 16 mL of Ultima Gold[™] LLT LSC cocktail. ^{129}I and ^{125}I were simultaneously counted by a Quantulus 1200[™] at room temperature (see Figure 1).

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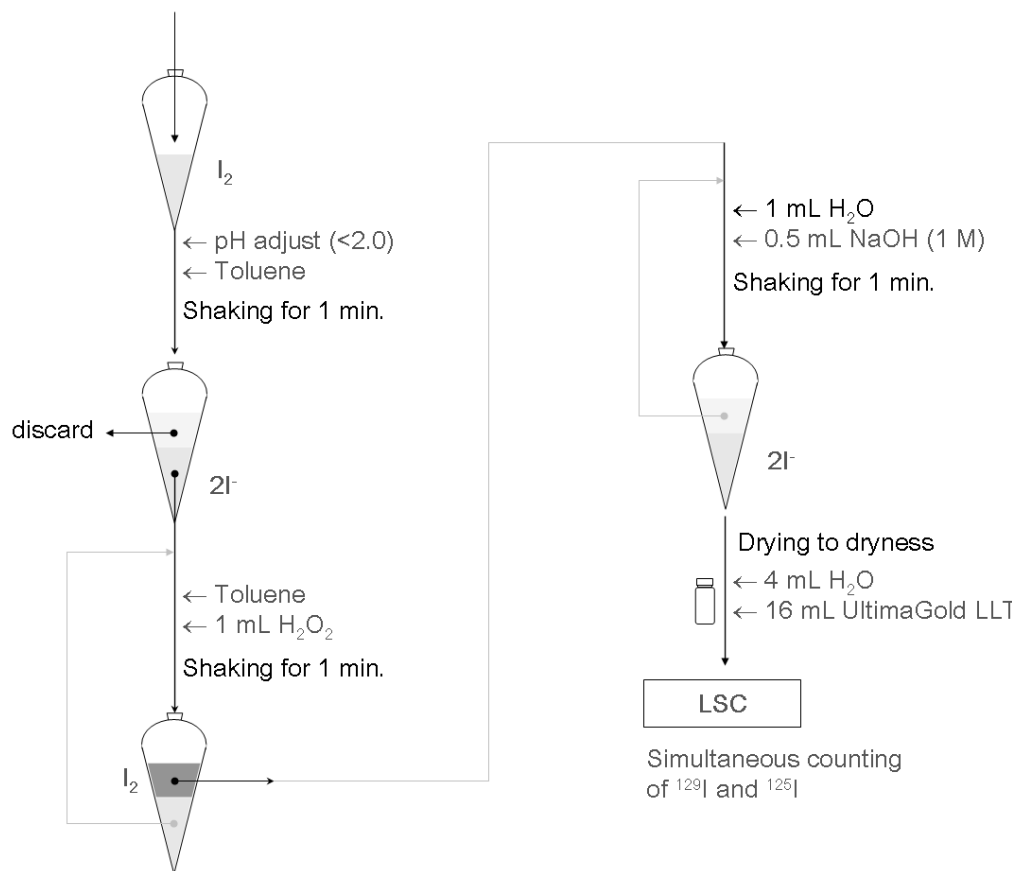


Figure 1 Separation procedure for ^{129}I applied in this study

The activities of ^{129}I and ^{125}I were calculated by the 3-over-2-fitting method using EasyView™ software (Wallac, PerkinElmer). Window settings for 3-over-2-fitting were 100–200, 201–330, and 331–500 channels.

RESULTS AND DISCUSSIONS

In this study, ^{125}I was used as a radiological tracer for ^{129}I , and ^{125}I and ^{129}I were simultaneously counted with LSC. In addition, toluene was used as an extraction solvent, instead of the carbon tetrachloride that is generally used for iodine solvent extraction, due to the possibility that the residual CCl_4 in the counting sample would act as a strong quencher for scintillation counting. NaOH used for back-extraction prevented iodine loss during evaporation and enabled the dried sample to be easily dissolved in 4 mL of water and then mixed with Ultima Gold LLT. However, at low temperatures, phase separation occurred in the sample and cocktail solution mixture. To avoid this separation, counting of ^{125}I and ^{129}I was performed at room temperature.

Figure 2 shows LSC spectra of simultaneously counted ^{125}I and ^{129}I . Count rates were calculated with EasyView software. As seen in Figures 3 and 4, the counting efficiencies of ^{125}I and ^{129}I were 35.1–38.1% and 75.9–81.1%, respectively.

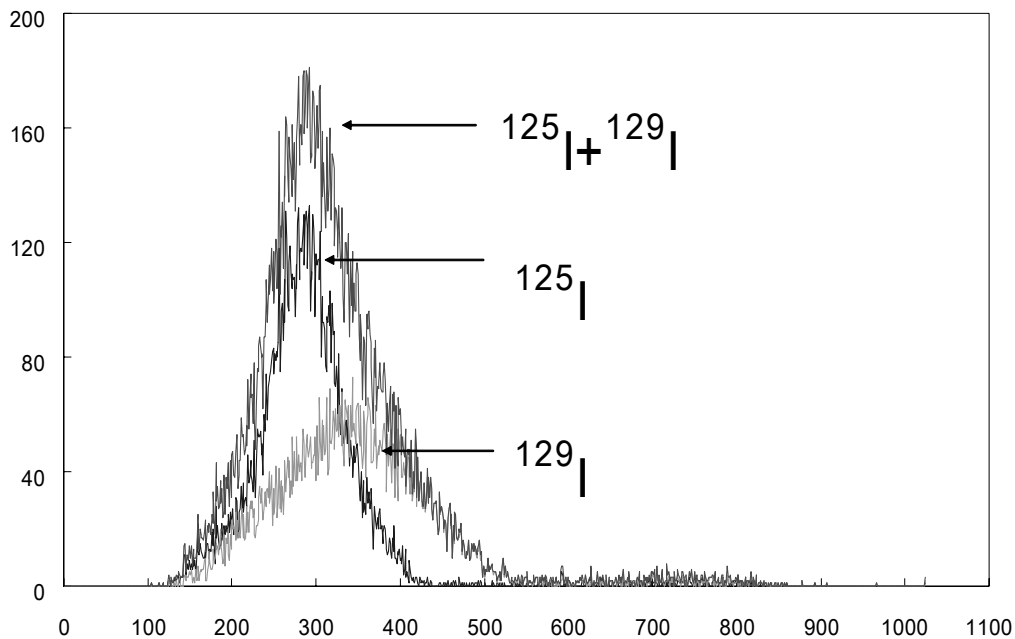


Figure 2 LSC spectra of ^{125}I and ^{129}I

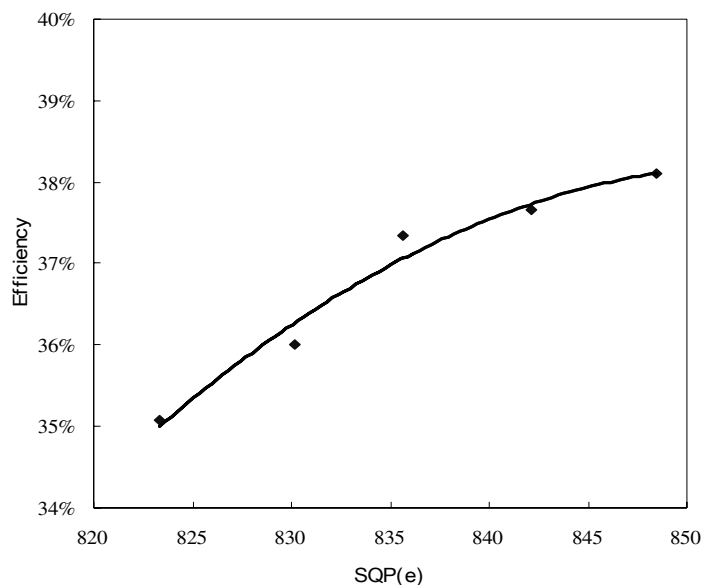
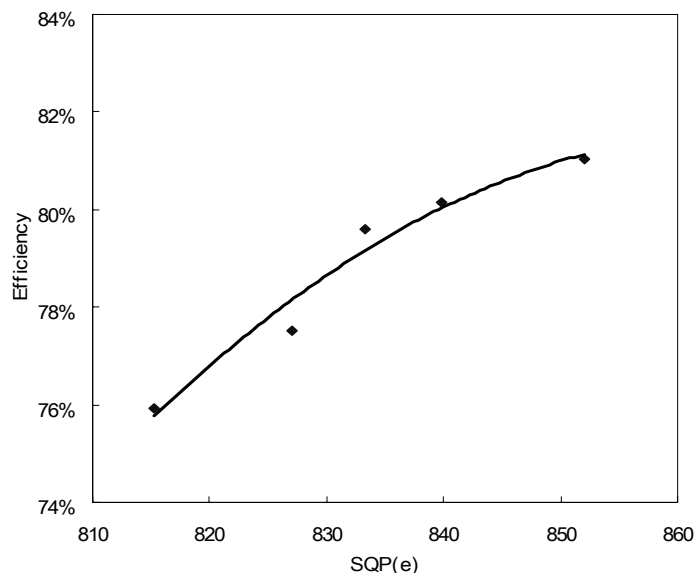


Figure 3 Counting efficiencies of ^{125}I

The chemical yields of ^{129}I during distillation and during solvent extraction were between 65 and 95%. The analytical results of ^{129}I in the spiked water samples were in good agreement with the known concentration of ^{129}I within 5.3% relative error (Table 1). The lower limit of detection (LLD) in the spiked sample was ~ 10 mBq.

Figure 4 Counting efficiencies of ^{129}I Table 1 Analytical results of ^{129}I in the trial samples.

Spiked water samples	Tracer (^{125}I) (Bq)	Added ^{129}I (Bq)	Measured ^{129}I (Bq)	Ratio (measured/added)
A	5.91	3.21	3.040 ± 0.01	0.947
B	5.89	1.61	1.540 ± 0.01	0.958
C	5.93	0.345	0.341 ± 0.005	0.990
D	5.77	0.165	0.173 ± 0.003	1.053

We studied a method for determining ^{129}I by liquid scintillation counting. It is difficult to measure ^{129}I in ordinary environmental samples using this method, but we expect that the method can be used for measuring ^{129}I due to radiological accidents.

REFERENCES

- Buraglio N, Aldahan A, Possnert G. 2000. Analytical techniques and applications of ^{129}I in natural water. *Nuclear Instruments and Methods in Physics Research B* 172:518–23.
- Cox RJ, Pickford CJ. 1992. Determination of I-129 in vegetable samples by inductively coupled plasma mass spectrometry. *Journal of Analytical Atomic Spectrometry* (7):635–40.
- Hou X, Dahlaard H, Nielsen SP. 2001. Chemical speciation analysis of ^{129}I in seawater and a preliminary investigation to use it as a tracer for geochemical cycle study of stable iodine. *Marine Chemistry* 74:145–55.
- Santschi PH, Schwehr KA. 2004. $^{129}\text{I}/^{127}\text{I}$ as a new environmental tracer or geochronometer for biogeochemical or hydrodynamic processes in the hydrosphere and geosphere: the central role of organo-iodine. *Science of the Total Environment* 321(1–3):257–71.