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Determination of Iodine-129 in Low Level Radioactive Wastes – 13334

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ABSTRACT

For the radioactivity determination of ^{129}I in the radioactive wastes, alkali fusion and anion-exchange resin separation methods, which are sample pretreatment methods, have been investigated in this study. To separate and quantify the ^{129}I radionuclide in an evaporator bottom and spent resin, the radionuclide was chemically leached from the wastes and adsorbed on an anion exchange resin at pH 4, 7, 9. In the case of dry active waste and another solid type, the alkali fusion method was applied. KNO_3 was added as a KOH and oxidizer to the wastes. It was then fused at 450 ℃ for 1 hour. The radioactivity of the separated iodine was measured with a low energy gamma spectrometer after the sample pretreatment. Finally, it was confirmed that the recovery rate of the iodine for the alkali fusion method was 83.6±3.8%, and 86.4±1.6% for the anionic exchange separation method.

INTRODUCTION

Recently, in Korea, an analytical method development for alpha and beta emitters has been conducted with regard to the regulation of transportation or the disposal of nuclear waste generated from nuclear power plants. One of these critical nuclides is ^{129}I, which is produced by nuclear fission in nuclear reactors, and has a long half-life ($t_{1/2}$ = 1.57 x 10^7 y) [1]. The importance of the control of this nuclide has been recognized, owing to its radio-ecological effect [2,3]. ^{129}I, however, is difficult to measure directly with a beta counter due to its very low specific activity of (6.4 Bq/μg) [4]. Generally, concentration of difficult-to-measure (DTM) radionuclides, such as ^{129}I, contained in nuclear wastes generated from nuclear power plants can be predicted from the scaled factor ($^{129}I/^{137}Cs$, $^{129}I/^{60}Co$) by measuring the radionuclide instead, such as ^{60}Co, ^{137}Cs which are relatively easier to measure. Therefore, the activity of ^{129}I has to be measured accurately in order to calculate the scaling factor (radioactivity ratio of ^{129}I and ^{137}Cs) in relation to the activity of the standard nuclide. The latter will then be used as described above. However, the radioactivity ratio of ^{129}I to ^{137}Cs varies widely, owing to many different classes of radioactive waste forms generated from nuclear power plants. Consequently, it can be concluded

that a trustworthy chemical analysis is required to determine the characteristic scaling factors for each plant [5,6].

In this report, the simulated rad waste was prepared for the measurement of the radioactivity of ^{129}I present in rad waste of the power plants [7,8]. The radioactive iodine tracer (^{125}I, $t_{1/2}$=60.14d) was added into the simulated solution, and the recovery rate was measured to establish its analytical procedure. In addition, the chemical recovery rate of iodine during the extraction and stripping process was obtained to compensate for the final recovery rate loss of the whole process.

RESULT AND DISCUSSION

Application of the Alkali fusion method

In the alkali fusion method, isolated I- is oxidized to I2 and eventually fused when KOH, KNO_3, and KI are added to solid sample and heated in a furnace. The dried solid sample was placed into a platinum crucible, and KNO_3 (oxidant) as well as KOH (fusion agent) were added; the standard KI 1.0 mL (μg/mL) was then added. After mixing, the furnace temperature was adjusted to 100-120 ℃ for drying. The temperature was then maintained at 200 ℃ for approximately 1 hour and was then raised slowly to 450 ℃. The fused solid was dissolved in a dilute acid for a stock solution. The flow chart for the alkali fusion method and assay process of sample is showed in Figure 1.

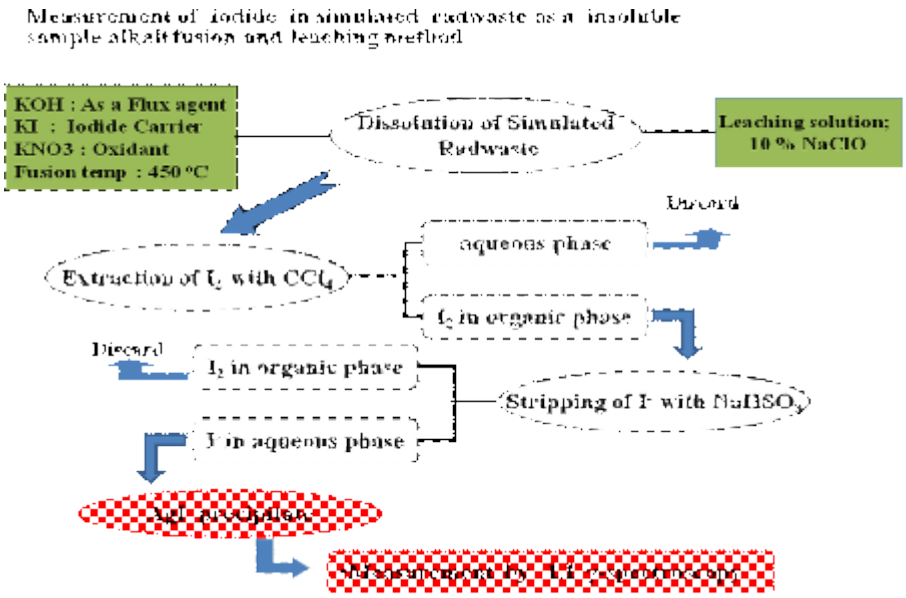

Fig. 1. Chemical procedure for analysis of ^{129}I in radioactive wastes as an insoluble sample.

Application of Adsorption column elution method

The nonvolatile oxidation products are eluted easily from the resin (AG 1 x2, 50-100 mesh Cl^- form) owing to their relatively low affinity compared with other anions such as Cl^-, $Br^-$and NO_2^-, HSO_3^-, and NO_3^-.

After adding the $NH_2OH.HCl$ and the reduction, the I_2 remaining in the elution solution was extracted by adding organic solvent ($CHCl_3$) and then back-extracted with $NaHSO_3$ to an aqueous layer in the form of I^- to determine the recovery rate. The flow chart for the adsorption method and column elution process of sample is showed in Figure 2.

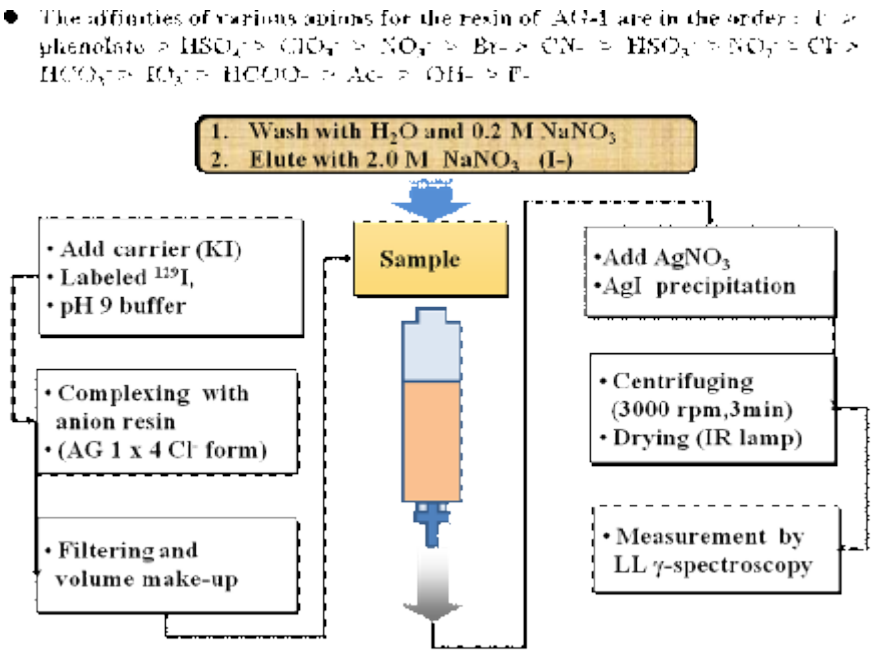

Fig. 2. Chemical procedure for analysis of ^{129}I in radioactive wastes as an aqueous sample.

CONCLUSIONS

An analytical procedure and a chemical separation technique were studied for ^{129}I by two different sample pretreatment method. The recovery rate was measured by adding ^{125}I as an iodine tracer to the simulation sample to measure the radioactive concentration of ^{129}I in a rad waste. The radioactivity of the separated iodine was measured with a low energy gamma spectrometer after the sample pretreatment. The recovery rate of the iodine for the alkali fusion

method was 83.6±3.8%, and 86.4±1.6% for the anionic exchange separation method.

REFERENCE

1. R. Seki, E. Kmura, T. Takahashi, N. Ikeda, "Distribution and behaviour of long-lived radioiodine in soil, "Journal of Radioanalytical and Nuclear Chemistry, Articles 138, 1, 17-31 (1990).
2. J. S. Edmonds and M. Morita, "The determination of iodine species in environmental and biological samples", Pure & Appl.Chem., 70. No 8. 1567-1584 (1998).
3. H. Inoue, M. Kagoshima, "Removed of ^{125}I from radioactive experomental waste with an anion exchange paper membrane", Applied Radiation and Isotopes, 52, 1407-1412 (2000).
4. L. C. Bate and J. R. Stokely, "Determination of ^{129}I in mixed fission products by neutron activation analysis", ORNL/TM-7449 (1980).
5. J. H. Chao, C. L. Tseng, C. J. Lee, C. C. Hsia, S. P. Teng, "Analysis of ^{129}I in radwaste by NAA, "Applied Radiation and Isotopes, 51, 137-143 (1999).
6. J. N. Smith, K. M. Ellis, L. R. Kilius, "^{129}I and ^{137}Cs tracer measurements in the arctic ocean", Deep-Sea Research **I**, 45, 959-984 (1998).
7. J. Kysela, M. Zmitko, V. A. Yuranov, V. F. Tiapkov, " Primary coolant chemistry in VVER units", Nuclear Engineering and Design**,** 160**,** 185-192 (1996).
8. I. Kuleff, S. Zotschev, G. Stefanov, "Determination of the ^{129}I content of the primary coolant of nuclear power reactors", Journal of Radioanalytical and Nuclear Chemistry, Articles, **97,** 1, 73-79(1986).