Radiochemical separation of ^{99}Tc in spent resin generated from Korea NPPs

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1. Introduction

The cumulative increase of low and medium level radioactive wastes is expected to result in capacity problem of temporary storage in some NPPs (Nuclear Power Plants) of Korea around 2008. Radioactive wastes are scheduled to be disposed in a permanent disposal facility in accordance with the Korean Radioactive Wastes Management Program. It is mandatory to list the quantities of several long lived radionuclides (ex. ^{3}H, ^{55}Fe, ^{90}Sr, ^{94}Nb, ^{99}Tc…) when the waste packages are transported from temporary storage in NPPs to disposal facility. Moreover, correlations between the hard-to-measure and more easily measured radionuclides, which we could predict the concentrations of the hard-to-measure radionuclides, must be proposed. Therefore, it is required to establish analytical procedures for a number of radionuclides.

Since ^{99}Tc is a low-energy pure-β-emitter with a long half-life, when it is released to ground-water it could significantly cause the environmental problems [1, 2]. The complete elimination of other radionuclides in the radiochemical separation step prior to β-counting of ^{99}Tc is prerequisite to obtaining its amounts in radioactive wastes. Nowadays, Re is used routinely for chemical yield tracer, because it is well known that Re and Tc chemistries are similar [3].

In this study, the procedure of separating ^{99}Tc from spent resin samples and quantifying them by gas proportional counting (GPC) is presented.

2. Methods and Results

2.1 Acid digestion of spent resin sample [4]

Spent resin samples were pretreated with the acid microwave digestion method in order to obtain homogeneous solution. Around 0.1 g of spent resin was weighed into a TMF vessel and 3 mg of Re as a carrier and 8 ml of HNO_3 were added to it. The microwave digestion procedure was performed in 4 steps : (1) 4 min at 290 watt, (2) 10 min at 400 watt, (3) 1 min at 300 watt, and (4) 10 min at 250 watt. The clear solution was transferred into a 20 ml volumetric flask and diluted to volume.

2.2 Separation and purification of ^{99}Tc

After adjusting the nitric acid concentration of 8 ml portion of the dissolved solution to 0.5 M HNO_3, 0.75 mg of a Re carrier were added, and the solution was stirred well for about 30 minutes.

A disposable column was made out of polypropylene tube and filled with anion exchange resin (AG MP-1, 100 – 200 mesh, Bio-rad) by the slurry-packing method to give a resin bed of 4.5 × 53 mm. The anion exchange resin column was first converted to the nitrate form using 5 M HNO_3. It was preconditioned by passing 3 ml of 0.5 M HNO_3 through it before use. The sample solution was then transferred to the top of the column after filtered through a microcellulose filter (0.45 ㎛, Millipore). The flow rate was maintained at about 0.2 ml/min, using gravity flow. The resin was sufficiently washed with about 20 ml of 0.5 M HNO_3 to remove all the other radionuclides. Only ^{99}Tc was retained on the resin at this point.

The ^{99}Tc was eluted from the column with 5 ml of 10 M HNO_3 and then the eluent was collected in a glass beaker. Finally, the eluate was evaporated to dryness using an infrared lamp. The residue was dissolved in 5 ml of 0.1 M HNO_3 and then ReO_4^- and TcO_4^- were co-precipitated with 0.5 ml of 0.02 M TPAC(tetraphenyl arsonium chloride, $(C_6H_5)_4AsCl$).

2.3 Chemical yield of Re

The recovery of Re at the acid digestion step was investigated by ICP-AES. The recovery of three different spent resin samples obtained from suggested microwave

digestion procedure was 93.1 ± 5.1%. The standard deviations presented were based on n = 3.

Radiochemical yield of Re was determined by weighing the $(C_6H_5)_4AsReO_4$ precipitate. The chemical yield of Re during the analytical separation and purification of ^{99}Tc is 85.9 ± 3.0% (n = 3).

Under the optimal compromised experimental condition for quantification of ^{99}Tc, the overall recovery of Re was shown to be 70.9 ~ 83.0% (Table 1).

2.4 Determination of ^{99}Tc

^{99}Tc was separated and purified from in radioactive spent resins generated from nuclear power plants was assayed by gas proportional counter. Table 1 gives the overall chemical recovery yields and concentration of ^{99}Tc.

Table 1. Analysis of the spent resin samples generated from NPPs

No.	Chemical recovery of Re (%)	^{99}Tc concentration (Bq/g)
1	83.0	1.29
2	83.0	1.20
3	70.9	2.10

3. Conclusion

In order to quantify ^{99}Tc in the spent resin generated from NPPs, we have established the proper sample preparation and radiochemical separation method. Re, which is chemically analogous to Tc, was used as a carrier for ^{99}Tc. The sample solution was made by acid digestion of the spent resin sample. ^{99}Tc was separated from the sample solution using AG MP-1 anion exchange resin, and then determined by GPC. Recovery of ^{99}Tc was 70.9 ~ 83.0% and concentration of ^{99}Tc in spent resin generated from NPPs was 1.20 ~ 2.10 Bq/g.

REFERENCES

[1] A. Aarkrog, Technetium in the Environment, Elsevier, London and New York, p. 419

[2] T. M. Bates, A review of the biological and geochemical behaviour of technetium in the marine environment, Journal of Environ. Radioact., Vol. 3, p. 1, 1986

[3] K. Tagami, Separation of rhenium by an extraction chromatographic resin for determination by inductively coupled plasma-mass spectrometry, Analytica Chimica Acta, Vol. 405, p 227, 2000.

[4] J. J. Lee, H. Y. Pyo, J. S. Jeon, C. H. Lee, K. Y. Jee, P. K. Ji, Determination of major and minor elements in low and medium level radioactive wastes using closed-vessel microwave acid digestion, Korean radioactive waste society, Vol. 2(4), p. 231, 2004