A Study on the Optimization of Experiment Conditions for Enhancement of Tc Recovery from LILWs

Hyeon Jeong Oh[1,*], Jung Bo Yoo[1], Sang Jin Lee[2], Sang Hoon Lee[2], Hong Joo Ahn[1]

[1]*Nuclear Chemistry Research Division, Korea Atomic Energy Research Institute, 34057, 111 Daedeok-daero-989, Yuseong-gu, Daejeon, Republic of Korea; *E-mail: ohj@kaeri.re.kr*
[2]*Radioactive Waste Project Division, Korea Radioactive Waste Agency (KORAD), 780-050, 89 Bukseong-ro, Gyeongju, Republic of Korea*

In the present work, the ideal experiment conditions to obtain the high recovery of Tc (Technetium) from low-and intermediate radioactive wastes were performed by controlling the TPAC (Tetraphenylarsonium Chloride) concentration, the amount of TPAC and reaction time. It is therefore to assume that TPAC is one of the most effective chelating reagents with Tc. Radiochemical yield of Re was determined by weighing the $(C_4H_5)_4AsReO_4$ precipitation; when 2.5 mL of 0.02M TPAC for 60 min under room temperature were added, the recovery rate of Tc had the highest value (99.3±1 %).

I. INTRODUCTION

Currently, radioactive wastes in Korea are classified into two categories depending their activity and the level of heat generation as low- and intermediate level wastes and high level wastes. In case of low- and intermediate level wastes, the amount of radwastes generated from the nuclear power plants (NPPs) have reached approximately over 90,000 drums as of March, 2015, and are still being stored at NPP sites. Therefore, the safe and effective management of radwastes are the national mission needed for maintainable generation of nuclear power [1].

Technetium-99 (^{99}Tc) is an artificial radioactive nuclide with a long half-life of 2.1 x 10^5 years, which is generated by the thermal neutron fission of ^{235}U and ^{239}Pu. ^{99}Tc is a weak beta emitter with 0.29 MeV, but secondary X-rays may become important with larger amount of the isotope. ^{99}Tc is one of the most significant radionuclides in safety assessment of radioactivity in the environment, as well as in decommissioning of nuclear facilities and management of nuclear waste [2]. As the chemical behaviors of Tc and Rhenium (Re) both are similar, Re was used as a tracer of Tc [3]. Therefore, chemical separation and determination of Tc are necessary. Until now, numerous research results to determine ^{99}Tc have been developed by various approaches such as solvent-extraction, precipitation, ion-exchange process and extraction chromatography.

Herein, we not only investigated the effective separation conditions of Tc by adjusting the concentration of TPAC, amount of TPAC and reaction time but also determined a Tc nuclide by weighing the $(C_4H_5)_4AsReO_4$ precipitation. An important benefit of this research is that ^{99}Tc can be separated effectively compared to the previous experimental conditions.

II. EXPERIMENTAL SECTION

All chemicals were purchased from commercial suppliers and used without further purification unless otherwise stated. Anion exchange resin, AG MP-1 (100-200 mesh) in chloride form was manufactured by Bio-Rad Laboratories, USA. TPAC (97 %) was obtained from the Aldrich Chem. Co, USA). Nitric acid (65 %), ethanol (99 %) and acetone (99 %) were purchased from MERCK (Germany). The Re (Rhenium) standard solution with 10 mg/mL were purchased from Accustandrad, USA. DI-water was used a Milli-Q plus Ultra-Pure Water System (Millipore). In brief, 5 mL of 0.1M nitric acid and 0.3 mL of Tc standard solution were taken in a 15 mL of centrifuge tube. After vigorous hand-shaking for several times, 0.5 mL of ethanol and TPAC as a precipitation reagent were added drop-wise and stirring continued for another 10 sec to precipitate the Re with TPAC ($(C_4H_5)_4AsReO_4$). The as-prepared $(C_4H_5)_4AsReO_4$ compounds were centrifuged at 3,500 rpm for 5 min and washed with 5 mL of 0.1M nitric acid and deionized water several times. White precipitate formed was dried by using Infrared lamp (250 W) for 30 min. Then, the dried solid product was transferred into a copper planchet which was washed by acetone. The recovery of $(C_4H_5)_4AsReO_4$ was calculated using the equation:

$$\text{Recovery of Re} = \frac{(C_4H_5)_4AsReO_4\ weight\ (mg)\ X\ 0.294}{Re\ contents\ (total\ 3\ mg)}$$

III. RESULTS & DISCUSSION

III.A.1. Effect of Different TPAC Concentration

To investigate the effect of different TPAC concentration, diverse concentration of TPAC were added on the as-prepared solutions. The concentration ranges were changed from 0.005M to 0.01M, 0.015M and 0.02M, respectively. Fig.1 (a) shows the TPAC concentration-dependent chemical yield of Tc. When the TPAC concentrations were increased, the chemical yield values were similarly increased. The chemical yields of 0.005M, 0.01M, 0.015M and 0.02M TPAC were 85.8±2 %, 88.3±2 %, 95.2±2 % and 99.3±1 %, respectively.

III.A.2. Effect of Different Precipitation time

In order to study the influence of precipitation time, the recovery of the Tc after the chromatographic separation stage was investigated by controlling the reaction time which was performed from 0 min to 60 min. Until 30 min, no dramatic chemical yield was obtained (Fig. 1(b)). However, more than 90 % of Tc recovery was showed over 30 min. When the separation step was performed for 60 min, the Tc recovery was reached about 100 %. The chemical yields of 30 min, 40 min, 50 min and 60 min were 36.2±3 %, 82.1±2 %, 93.6±1 % and 97.0±5 %, respectively.

III.A.3. Effect of Different TPAC amount

TPAC amount-dependent chemical yields of Tc are given in the right panel of Fig. 1(c). A notable feature is that the chemical yield of Tc increases with increasing the amount of TPAC. The chemical yields of 0.5 mL, 1.0 mL, 1.5 mL, 2.0 mL and 2.5 mL were 86.2±1 %, 89.4±4 %, 92.3±3 %, 95.1±1 % and 98.5±5 %, respectively. This suggests that the amount of TPAC is a key role on the recovery of Tc.

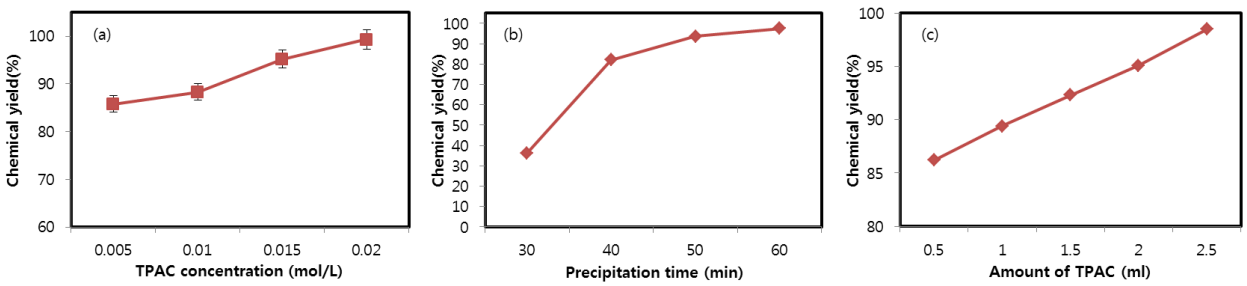

Fig 1. Plots of Tc recovery values depending on the (a) TPAC concentration, (b) Precipitation time, and (c) Amount of TPAC

IV. CONCLUSIONS

In this work, the effective separation conditions to obtain the high recovery of Tc from low-and intermediate radioactive wastes were performed by adjusting the TPAC concentration, the amount of TPAC and reaction time. As the chemical behaviors of Tc and Rhenium (Re) both are similar, Re was used as a tracer of Tc. Radiochemical yield of Re was determined by weighing the $(C_4H_5)_4AsReO_4$ precipitation; when 2.5 mL of 0.02M TPAC for 60 min under room temperature were added, the recovery rate of Tc had the highest value (99.3±1 %). An important benefit of this research is that ^{99}Tc can be separated effectively compared to the previous experimental conditions.

ACKNOWLEDGMENTS

This work was supported by the Ministry of Science, ICT, and Future Planning (MSIP) of South Korea.

REFERENCES

1. Nuclear Safety and Security Commission, NSSC Notice 2015-4: Regulation on Safety Classification and Applicable Codes and Standards for Nuclear Reactor Facilities, the Nuclear Safety and Security Commission, Seoul, Korea, 2015
2. K. Shi, X. Hou, P. Roos and W. Wu, "Determination of technetium-99 in environmental samples: A review", *Analytica Chimica Acta*, **709**, 1 (2012)
3. K. Tagami, "Separation of Rhenium by an Extraction Chromatographic Resin for Determination by Inductively Coupled Plasma-Mass Spectrometry", *Analytica Chimica Acta*, **405**, 227 (2000)