

The Analysis of U in a Korean Total Diet Sample
by ^{238}Np tracer and RNAA

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Abstract

In order to estimate the degree of intake of U through daily diet, a Korean total diet sample was prepared after the investigation of the amount of consumption of daily diet which is dependent on the age of 20's to 50's. For the analysis of U, RNAA method was applied and NIST SRM 1566a, Oyster Tissue was used as a quality control material. The result of SRM analysis was compared with certified value, the relative error was 15%. The determination of U in a Korean total diet sample was carried out under the same analytical condition and procedure with SRM. It is found that the concentration of U in synthetic Korean total diet was 35.4 ± 4.4 ppb and the amount of daily intake of ^{238}U by diet is 6.98 μg per day. Radioactivity due to the intake of U was estimated to be about 86.7 mBq per person per day and the annual dose equivalent was revealed as 2.11 μSv per person.

1. Introduction

^{238}U is a radioactive element which incurs internal doses by inhalation of airborne particulate matter and by intake of daily diet [1]. Therefore, it is necessary to apply an accurate and precise analytical method for U and important to assess radiation doses from this natural source in the human body.

Even though there are several very sensitive analytical techniques for U, such as isotope-dilution mass spectrometry, neutron-induced track counting or ICP-MS, it was known that Radiochemical Neutron Activation Analysis (RNAA) possesses the considerable advantage of being blank-free [2]. It was the aim of this study to apply RNAA for the determination of U in the Korean total diet sample based on the daily food data of those aged 20 to 50(77 adults) in Korea. The average amount of daily food consumption was 1700g.

In this study, through the determination of the concentration of U in Korean total diet by using ^{238}Np tracer and RNAA, it was intended to estimate the daily intake of U and to assess the annual dose equivalent.

2. Experimental

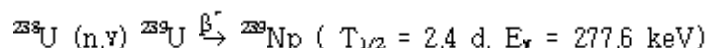
2.1 Sample Preparation

To prepare the Korean total diet for analysis of U, diet samples were purchased in the market and pretreated as in the following procedure : diet samples were mixed by blender for homogenization and freeze dried for 30 hours at -80°C . After freeze-drying, The weight of the total diet was reduced from 1120,0 g to 130,2g. A freeze dried sample was finely ground by using agate mortar. Pretreated Korean total diet sample was prepared into polyethylene capsule washed by 1N HNO_3 for irradiation. NIST SRM 1566a, Oyster Tissue was done as well after drying for 2 hrs at 80°C in oven in accordance with recommended procedures of NIST certificate.

2.2 Sample Irradiation and Radioactivity Measurement

The prepared diet sample was irradiated under the thermal flux of $1,7 \times 10^{13} \text{ n/cm}^2 \cdot \text{sec}$ for 4 hours using PTS(Pneumatic Transfer System) of HANARO research reactor. NIST SRM samples were also irradiated in the same condition. These samples were allowed to cool for more than 3 days. 0,5 ml of ^{237}Np solution ($0,1 \mu\text{Ci}/50\text{ml}$, Isotope Products Laboratories) was irradiated for 10 - 20 min, to produce relevant activity of ^{238}Np ($T_{1/2} = 50,8 \text{ hr}$, $E_{\gamma} = 984 \text{ keV}$). It should be noted that the absolute activity of ^{238}Np is not required, because the chemical yield measurement is based on relative measurements of ^{238}Np in an unprocessed aliquot(as standard) and in the sample fraction separated according to the RNAA procedure.

Uranium was determined by using the nuclear reaction of



All samples were analyzed by estimation of ^{239}Np -activity. For the γ -ray measurement of ^{239}Np , ^{238}Np and other nuclides, HP-Ge semiconductor detector(EG & G ORTEC, GEM25185, 1,85keV resolution at 1332keV of Co-60, Peak to Compton ratio 45 : 1) and 8K MCA(MCB 919, EG & G ORTEC) were used.

2.3 Radiochemical Separation Procedure

RNAA method using anion exchange resin (DOWEX 1 x 8, Cl^- form, 100-200 mesh) was applied for the analysis of U [3]. The stepwise separation scheme of U is presented in Figure 1, subsequent separation procedures are as follows : before sample digestion, added the aliquot of ^{238}Np -solution and then samples were digested in concentrated HNO_3 . Now this digested solution was almost evaporated to dryness, 25 ml of 9M HCl and 0,5 g of hydroxyl amine hydrochloride were added and then warmed gently for 10 minutes to convert Np to Np^{4+} . After preparing the ion exchange column, the digested solution of sample was loaded and Np was eluted with 60 ml of 1M HCl from the column. Eluent was evaporated from 60 ml to about 5 ml and added 7 ml of H_2SO_4 and about 100 mg of K_2SO_4 . The solution was gently warmed and 40 mg of Ba carrier was added dropwise until BaSO_4 precipitate appeared. This precipitate was filtered using

Whatmann 542 filterpaper and gamma-activity of Np was measured finally.

3. Results

For quality control, the concentration of U in NIST SRM 1633a, Oyster Tissue was determined to check accuracy and precision of RNAA method. The analytical value was 113 ± 1 ppb and compared with certified value (132 ± 12 ppb). The relative error was 15%. But considering the uncertainty of the certified value, our value was in good agreement with the certified value. Figure 2 shows gamma-ray spectrum of U analysis in SRM Oyster Tissue and ^{238}Np standard for yield calculation.

As a result of the analysis of Korean total diet sample, the concentration of U was 35.4 ± 4.4 ppb. Figure 3 shows gamma-ray spectrum of U analysis in Korean Total diet sample and ^{238}Np standard for yield calculation. Table 1 shows the analytical results of U in SRM Oyster Tissue and Korean total diet sample. Even though the difference of the chemical yields was identified in Table 1 (especially Korean total diet), analytical values did not fluctuate much, which means an advantage of ^{238}Np tracer method.

Applying the average amount of daily food consumption of 1700g, the U intake by Korean total diet was estimated to be 6.98 μg per person per day. Converting this into radioactivity, the value is about 86.7 mBq per person per day. The effective dose equivalent by ^{238}U intake was known as 6.88×10^{-8} Sv/Bq [1]. Consequently, it can be concluded that the annual dose equivalent of ^{238}U in daily diet is about 2.11 μSv per person. Dietary intakes of U for Japanese have been reported in limited number. The mean daily intake and its standard deviation, 0.71 ± 0.31 μg (8.80 ± 3.84 mBq) per person, were reported from a duplicate portion study covering a wide range of Japan [4]. Our result was estimated to be much higher relative to those of Japanese.

4. Conclusion

From the above results, it was clarified that the RNAA method can be applied selectively for the determination of U in real samples which have ppb level of concentrations. In addition, for the assesment of radiation dose from natural radioactive sources such as U and Th, RNAA will be effectively applied to biological samples.

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Sample	SRM Oyster Tissue			Korean Total Diet		
Run No.	1	2	3	1	2	3
Chemical Yield	44 %	58 %	65 %	21 %	16 %	82 %
Analytical Value	114 ppb	113 ppb	112 ppb	39.1 ppb	37.8ppb	29.2 ppb
Mean ± SD.	113 ± 1 ppb			35.4 ± 4.4 ppb		

Table 1. Analytical results of U in SRM Oyster Tissue and Korean Total Diet

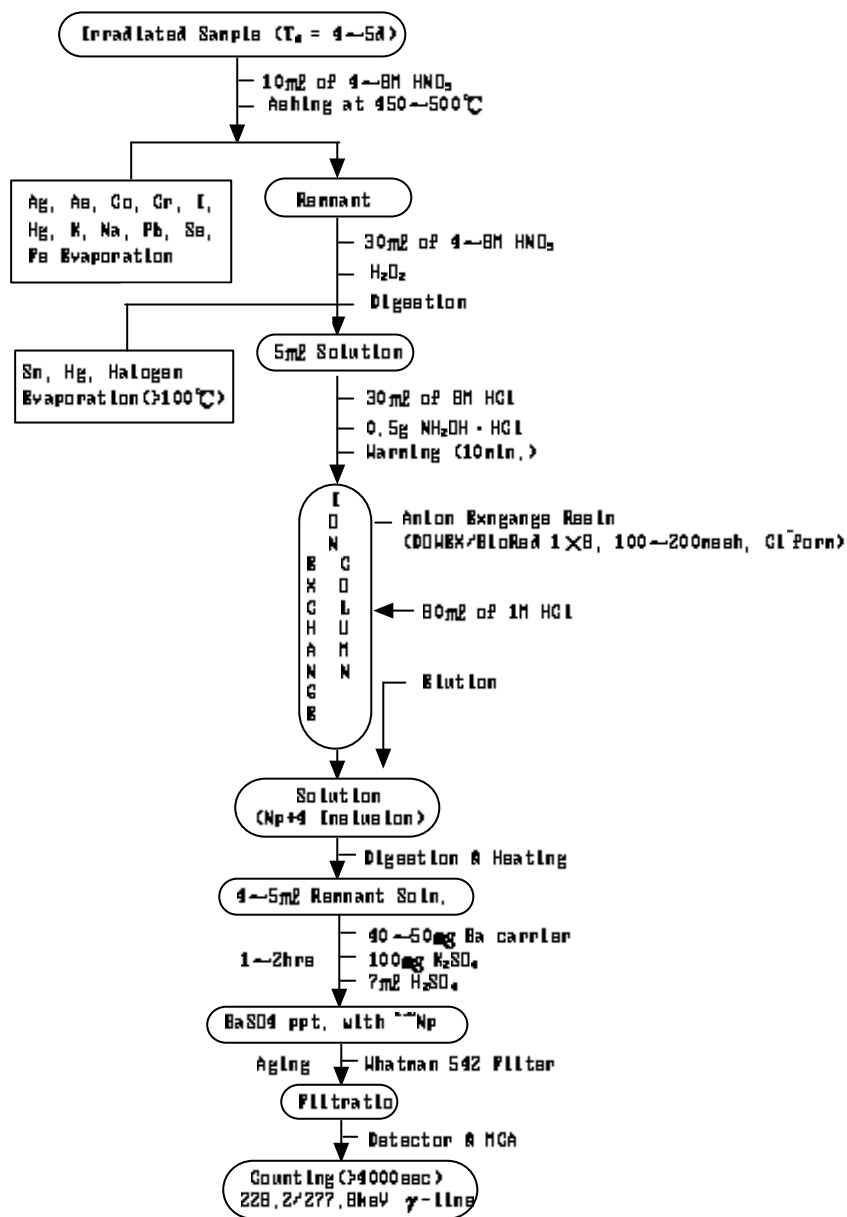


Figure 1. Radiochemical separation scheme for the determination of U(²³⁵Np)

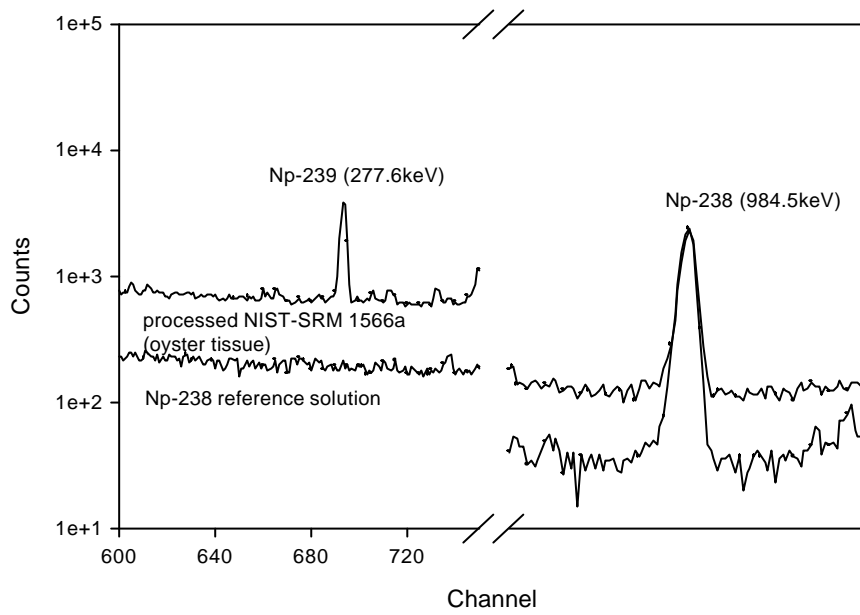


Figure 2. Gamma-ray spectrum of U analysis in SRM Oyster Tissue and ^{238}Np standard

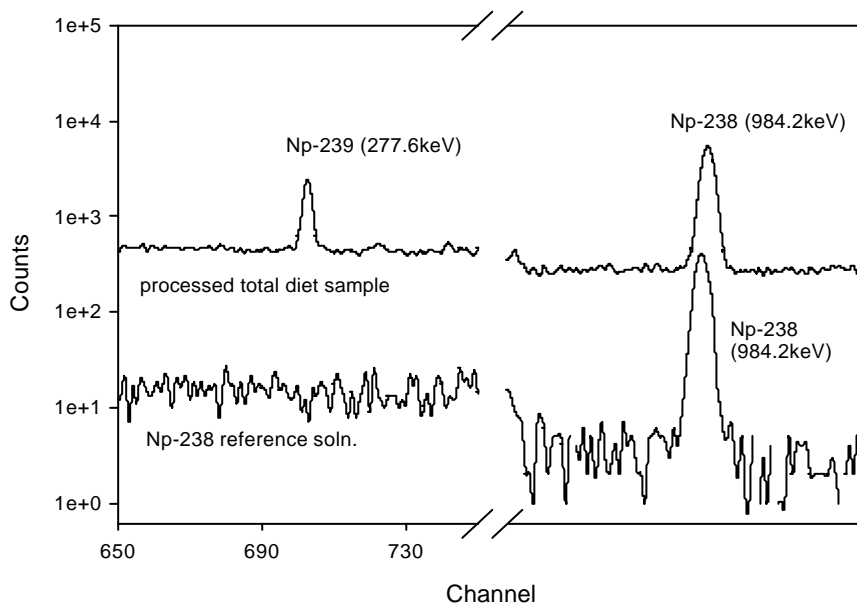


Figure 3. Gamma-ray spectrum of U analysis in a Korean Total Diet and ^{238}Np standard