METHOD DEVELOPMENT FOR Pu ISOTOPES AND ^{90}Sr IN SEAFOOD SAMPLES USING ION EXCHANGE/EXTRACTION CHROMATOGRAPHY

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The Fukushima Daiichi NPP accident caused the release of a large amount of radioactive fission products to the marine environment. As a result, the public concerns increase on the seafood stuffs in terms of radionuclide contamination due to its toxicity. This paper presents a quantitative and rapid method of sequential separation of Pu isotopes and ^{90}Sr in seafood samples using anion exchange resin and Sr Spec resin. The procedure combined two separate procedures for Pu isotopes and ^{90}Sr into one simple method. The sample solution was passed through an anion exchange column connected to a Sr Spec column. Pu isotopes were purified from the anion exchange column and ^{90}Sr was eluted from other interfering radionuclides using the Sr Spec column. The measurement of Pu isotopes was carried out using an alpha spectrometer. ^{90}Sr was measured by a low-level liquid scintillation counter. Reproducibility test as well as the test using tracer solutions showed that the method would be very reliable for seafood samples.

I. INTRODUCTION

The Fukushima Daiichi NPP accident occurred on 11 March 2011 caused the release of a large amount of radioactive fission products (*ca.* 770 PBq of ^{131}I and ^{137}Cs) to the marine environment and the radionuclide release is continuing. Marine organisms (plants and animals) are capable of accumulating such radionuclides, as a consequence, ingestion of seafood is the most significant route of radionuclide intake for members of the general public. Some studies have been conducted on the path of radionuclide intake and the long-term effects of radionuclide bioaccumulation in the human body. Since the Fukushima accident, the public concerns increase on the seafood stuffs in terms of radionuclide contamination due to its toxicity. While ^{137}Cs and ^{131}I have been monitored in the seafood stuffs, the monitoring of Pu (^{238}Pu, ^{239,240}Pu) and Sr (^{90}Sr) isotopes, even though both isotopes are controlled by CODEX, have not been widely conducted because of the complexity of sample preparation, radiochemical techniques for radionuclide separation and purification, source preparation, and measurement. As the result, the available information about the bioaccumulation and distribution of plutonium is limited. This study is to establish a rapid and simple radioanalytical method to determine the Pu and Sr isotopes in the seafood stuffs and to evaluate an impact of the Fukushima Daiichi NPP accident on the Korean seafood stuffs.

II. EXPERIMENTAL

II.1. Reagents and Equipment

Plutonium standard solutions were supplied by NIST, USA. Strontium standard solutions were standardised and supplied by KRISS, Korea. Both hydrochloric (37 %) and nitric (65 %) acids were analytical grade purchased from Merck, Germany and all other analytical grade reagents/chemicals were supplied by Sigma-Aldrich, USA. Deionised water (Milli-Q water) was used for all dilutions. Eichrom's anion exchange resin, 100-200 mesh (1-X8, chloride form) and Sr-resin, 100-150 mesh were used for the study. The Perkin Elmer 1220 Quantulus ultra low-level liquid scintillation counter was used for ^{90}Sr measurement. Alpha spectrometry measurement for Pu isotopes were performed using an EG&G Octete-PC alpha spectrometer fitted with 450 mm^2 ruggedised low-background ion-implanted detectors. Gamma spectrometry measurement (^{85}Sr) was carried out using an HPGe detector supplied by ORTEC.

II.2. Sample Preparation

Dried and ignited seafood samples (50 g of fresh weight) were spiked with both ^{242}Pu and ^{85}Sr solutions as an internal yield monitor. The samples were then leached using 8M nitric acid on a hotplate at 90°C for 2 hours. The acid solution was cooled down to room temperature and the residue was filtered using a Whatman No.540 filter paper. The filtered 8M nitric acid solution was placed in a fume cupboard until the radiochemical separation work began.

II.3. Radiochemical Separation

Anion resin (5 cm height) and Sr-resin (4 cm height) columns were prepared using Eichrom's 20 ml & 2 ml empty plastic columns. Both resins were soaked in water overnight prior to the column preparation. Anion resin column was placed on top and Sr-resin column was placed directly under the anion resin column so the acid passed through the anion

resin column could drop into the Sr-resin column. Both columns were pre-conditioned using 25 ml of 8M nitric acid. The radiochemical separation procedure is presented in Fig. 1.

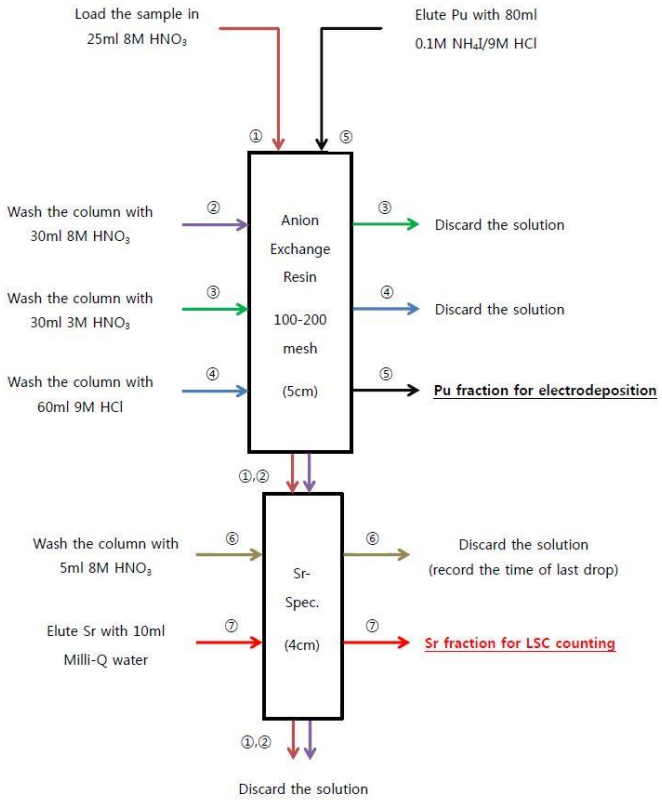

Fig. 1. Sequential separation of Pu and Sr isotopes

II. CONCLUSIONS

A simple and rapid radiochemical separation method was developed using both anion exchange resin and Sr-Spec resin. Reproducibility test as well as the test using tracer solutions showed that the method would be very reliable for seafood samples with the sample size of 50 g (fresh). Chemical recoveries turned out to be high, > 90% for Pu isotopes and > 80% for ^{90}Sr, respectively.

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