Development of an Automated Extraction Chromatography for the Rapid Analysis of Radionuclides

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1. Introduction

Radioactive wastes related to plant decommissioning come from the structures of the plant which have been either irradiated or contaminated with radioactive isotopes. The characterization of these wastes plays an important role from a waste management point of view. The proper and accurate determination of radioactivity is essential to evaluate the contamination level for possible decontamination, and for the classification of radioactive waste produced by various facilities. The radionuclides in these wastes may be ^{3}H, ^{14}C, ^{36}Cl, ^{41}Ca, ^{60}Co, ^{55}Fe, ^{63}Ni, ^{90}Sr, ^{99}Tc, ^{129}I, ^{133}Ba, ^{137}Cs, ^{152}Eu, ^{154}Eu and some transuranics. Of these radionuclides, gamma emitters are easy-to-measure radionuclides and can be relatively simply determined by gamma spectrometer. However, the beta and alpha emitters such as ^{55}Fe, ^{63}Ni, ^{99}Tc, ^{90}Sr and transuranics have to be separated and purified prior to radiometric or mass spectrometric detection [1]. To effectively determine the radioactivity of these hard-to-measure radionuclides using LSC, Alpha Spectrometer or ICP-MS, it is necessary to remove all interfering radionuclides and the matrix components in samples. Interfering nuclides are typically removed using extraction chromatographic methods under a gravitational flow or using a vacuum box system. However, these separation methods have time-consuming and labor-intensive radiochemical separation processes.

In this study, the detailed designs of automated extraction chromatographic (AEC) systems are described for the rapid and reproducible separation of radionuclides in radioactive waste samples.

2. Methods and Results

The AEC systems have been designed and fabricated in our laboratory. The Labview based virtual instruments have also been developed for the control of the AEC systems. Performance of the AEC systems has been evaluated by the separation of ^{99}Tc, ^{90}Sr, ^{55}Fe, thorium and uranium isotopes.

2.1 Design of AEC systems

Two types of AEC system have been developed with different extraction column arrangement. The schematic diagrams of the AEC systems with single and tandem column arrangement are shown in Fig. 1.

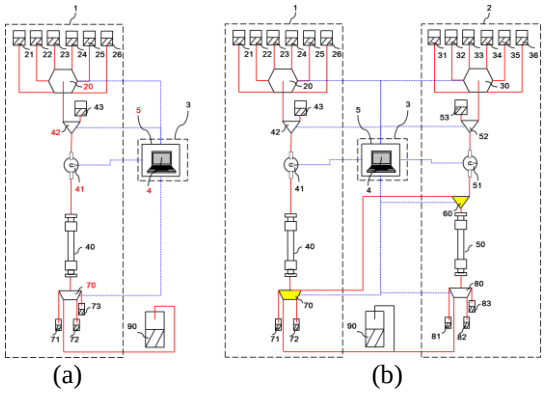

Fig. 1. Schematic diagram of AEC systems for the separation and purification of radionuclides: (a) single column arrangement, (b) tandem column arrangement.

The AEC systems consist of a data acquisition device, peristaltic pumps, solenoid valves and columns. The data acquisition device is connected to a computer via USB port. The data acquisition device has two types of DAQ board: the one with 64 digital inputs/outputs is used to actuate solenoid valves and the other with 16 analog outputs is used to control the flow rate of the peristaltic pumps. Three types of solenoid valve are used for AEC systems with single and tandem column arrangements. A 6-inlet port flow selection valve (20, 30) is used to select different chemical reagents. The 2-inlet port flow selection valve (42, 52, 60) is used to select a sample or a chemical reagent, and to connect two columns as a tandem arrangement. The 4-way flow distribution valve (70, 80) is used to collect purified solution and to connect two columns as a tandem arrangement. The AEC system with a tandem column arrangement, designed and built in our laboratory, is given in Fig. 2. This system has been commercialized as the name of automated sequential radionuclides separator (ASRS). The ASRS consists of three parts: ASRS-CS, ASRS-MC and ASRS-SS. The ASRS-CS means automated sequential radionuclide separator-control software. The ASRS-MC stands for automated sequential radionuclide separator-main controller, which is built in the ASRS-SS. The ASRS-SS means automated sequential radionuclide separator-sequential chromatographic separator. The ASRS-SS consists of five parts: reagent bottle part, sample tube part, column part, pump part, and elution tube part. The size of the ASRS-SS is W810 mm × D505 mm × H505 mm. This system can handle simultaneously 8 samples with single

column arrangement and 4 samples with tandem column arrangement in a single run.

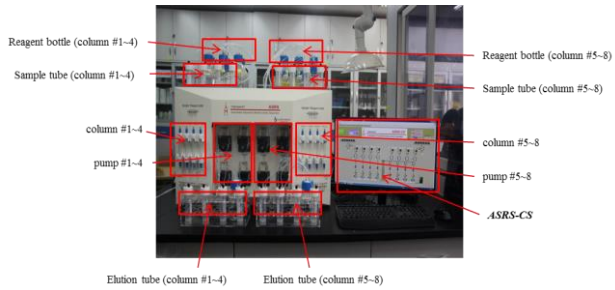

Fig. 2. A photograph of the AEC system

2.2 Design of Labview based virtual instruments

Control software developed on the Labview platform is a virtual instrument to control AEC system by means of manipulating the most important input parameters through the graphical user interface on the computer screen. The virtual instrument shown in Fig. 3 consists of three windows for interfacing the software and AEC systems. The first and second windows are used for calibrating the flow rate of peristaltic pumps. The final window is used to execute an automated radiochemical procedure for separating radionuclides from the interfering elements and matrix components. The input parameters in the virtual instruments are the volumes, flow rates and selection of reagents for executing an automated radiochemical procedure.

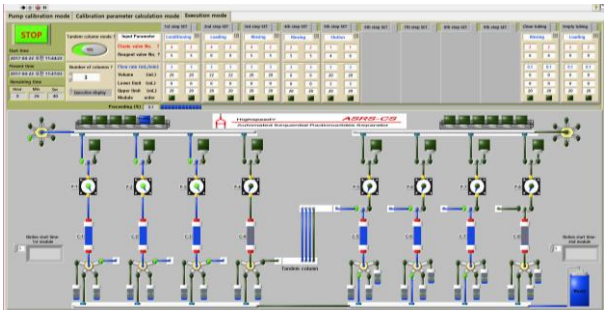

Fig. 3. A Labview based virtual instrument of the AEC system with the tandem column arrangement.

2.3 Performance of AEC systems

The AEC systems with a single column arrangement have been successfully applied for the measurement of ^{99}Tc in groundwater [2] and $^{89/90}$Sr in milk [3]. An AEC system with a tandem column arrangement was recently developed and applied for the sequential separation of ^{55}Fe and ^{90}Sr in spiked water samples [4]. Performance of the AEC system with a tandem column arrangement was also evaluated by the sequential separation of thorium and uranium isotopes in spiked water samples. The AEC system uses TEVA resin for purification of thorium and UTEVA resin for purification of uranium isotopes from the interfering radioactive nuclides. The mean chemical recoveries shown in Fig. 4, determined by ICP-OES, of thorium and uranium isotopes separated from the spiked water samples were 92% (n=4) for uranium and 97% (n=4) for thorium in a tandem column arrangement. In a single column arrangement with the same AEC system, the mean chemical recoveries were 97% (n=4) for uranium and 98% (n=4) for thorium. The results obtained for four independent measurements showed a quite good reproducibility with the relative standard deviation values of less than 2%.

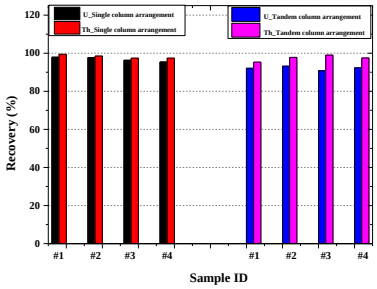

Fig. 4. Recoveries of U and Th obtained by using AEC system with single and tandem column arrangements.

3. Conclusions

The AEC systems, designed and built in our laboratory, were successfully applied to separation and purification of radionuclides in various samples. Compared to conventional radionuclide separation methods carried out manually, AEC system is faster, less labor-intensive and expected to be widely used as a powerful and convenient tool for the chemical separation and purification of the hard-to-measure radionuclides in various radioactive wastes related to plant decommissioning.

REFERENCES

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