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Reusable selective sensing-substrate for ultrasensitive and rapid detection of uranium radioisotopes

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ABSTRACT

An analytical method based on a fabricated analytical disc was developed using a phosphonate-immobilized quartz substrate for the rapid analysis of uranium. Phosphonate groups selectively adsorb uranium dissolved in a solution through the formation of stable complexes. The uranium concentration and isotopic ratios in the sample can rapidly be determined by measuring the radioactivity of the adsorbed uranium on the analytical disc using alpha-particle spectrometry. The chemical structure and elemental composition of the fabricated disc were analyzed by XPS, and the adsorption properties were evaluated as functions of pH of a sample solution, reaction time, and adsorbate concentration, as well as reusability and selectivity, using alpha-particle spectrometry, ICP-OES, and ICP-MS. An analytical method was developed based on these properties and validated with prepared uranium solutions by assessing the chemical recovery yield, relative error, and relative standard deviation. The developed method is expected to be used for on-the-spot uranium analysis.

1. Introduction

Alpha-particle spectrometry is widely and routinely used for the analysis and detection of isotopic ratios and radioactivity due to advantages that include high accuracy, high counting efficiency, high sensitivity, and low cost [1,2]. Achieving good radiochemical separation of the target nuclides from other interfering α-emitters with similar radiation energies is the key to the accurate detection of target-nuclide radioactivity using alpha-particle spectrometry [3]. The commonly used radiochemical separation method includes processes such as co-precipitation, dissolving the residue in acid, anion-exchange separation, and electrodeposition, [4,5]. However, the sophisticated and time-consuming method cannot provide rapid analysis of samples at sampling sites. In this study, a conceptually new method for the rapid analysis of uranium was developed using a highly selective analytical disc to address above issue. Instead of using the traditional radiochemical separation processes, our method used an adsorption process based on a functional-group-immobilized circular substrate (analytical disc) that can selectively adsorb and chemically separate U(VI).

Among functional groups that have selective affinities for U(VI) [6,7], the phosphonate group has attracted significant amounts of attention owing to its strong resistance to radiation, excellent stability, and high complexing ability for U(VI) [8–20]. Phosphonate is a functional group in UTEVA®, which is widely used as an extraction and chromatography polymer resin by virtue of its strong ability to separate U(VI) [21]. In addition, it is eco-friendly and nontoxic, and exhibits rapid adsorption kinetics for U(VI) [22,23]. Phosphonate groups selectively adsorb U(VI) by forming stable complexes with anions, such as NO_3^- and Cl^- in solution [24]. While using a chelate formed from a polymer composite containing the required functional group is a general way of increase adsorption capacity [25], alpha particles lose energy with increase of adsorption-layer thickness due to their high interacting abilities, which results in peak tailing leading to lower spectral resolution in the alpha spectrum, thereby compromising the ability to accurately determine the uranium isotopic ratios [26]. By designing a monolayer adsorption process, the alpha spectral resolution of the analytical disc can be maximized for detecting adsorbed uranium.

By replacing traditional chemical separation with an adsorption process, chemical separation for uranium analysis of a water sample can be completed within 30 min. In addition, because much less

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