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Alpha spectroscopy for *in-situ* liquid radioisotope measurements

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ABSTRACT

Using calculation and SRIM simulations of alpha particle energy spectroscopy, we show that the initial energies and concentrations of alpha-emitting radioisotopes can be measured in-situ in a liquid environment. We quantify the effect on the alpha spectrum of reducing the thickness of the liquid source in front of the alpha particle detector as well as adding a cover material onto the alpha particle detector surface. In all cases, initial energies and concentrations are recoverable from the alpha particle energy spectra. By reducing the thickness of the liquid source, the contribution to the spectrum for low count rate, low energy radioisotopes can be revealed. However, adding a cover on the detector obscures the contributions of these radioisotopes.

1. Introduction

Alpha particle spectroscopy is a standard procedure in the identification and quantification of radionuclides. Typically, it is performed using one of two methods, which both start with chemically isolating the radionuclides. In the first method, the radionuclides are deposited on a disc, and their alpha particle energy spectrum is determined in an alpha measurement chamber. In the second method, the radionuclides are mixed with a scintillating liquid, and the resulting scintillation photon energy spectrum is measured. In both methods, the resulting energy peaks are Gaussian in nature. The centroids of those peaks identify the radionuclides, and the area beneath the peaks corresponds to the total activity and thus amount of the radionuclides [1]. Both techniques are obviously inapplicable for real-time, in-situ monitoring of radionuclides.

However, in nuclear fuel treatment facilities, real-time, in-situ monitoring is a pressing need. Examples of fuel treatment processes include aqueous processing at the La Hague plant in France [2], and pyroprocessing of spent nuclear fuel currently being investigated at Idaho National Laboratory [3]. Aqueous processing is maintained at temperatures above 50 °C, a temperature beyond which Si-based detectors experience degraded resolution [1]. Pyroprocessing is performed in a molten salt which is maintained at about 500 °C, a temperature where Si and other common room-temperature detectors will fail because Si behaves as a metal instead of a semiconductor due to its relatively low bandgap. It may be possible to place a detector in these processing environments, such as inside of a processing tank, in direct contact with the radioisotope-containing liquid, and make alpha particle energy measurements with the detector to identify and quantify radionuclides in that liquid. We have recently shown that detectors made from 4H-SiC, a wide-bandgap semiconductor, can measure alpha particle energy spectra up to 500 °C with a 3 mm diameter circular active area [4]. Detectors such as these may be able to survive in the nuclear fuel treatment facility tanks. However, a proof-of-principle calculation is needed that demonstrates that the alpha particle energy spectra that would be measured in liquid-distributed alpha particle source environments could be used for *in-situ* radioisotope identification and quantification.

The most basic detector-liquid geometry involves inserting a detector directly into the liquid, thus exposing the detector to an effectively semi-infinite, homogenous alpha-emitting source. If electrical and chemical interaction between the detector and liquid source are found to degrade a detector's performance in the bulk reprocessing fluid, it may be necessary to isolate the surface of the detector from the bulk fluid. This may be achieved through either coating an insulating cover on the front surface of the detector, or integrating a microfluidic channel on top of the detector surface, or both. Attempting to measure the concentration of a radioisotope in a liquid using an in-situ alpha particle detector has been investigated at France's La Hague aqueous reprocessing plant [2]. However, no investigation into the mathematical form for the alpha particle spectrum has been presented to date in literature for various relevant source/detector geometries.

Our long term goal is to assess the potential of various source and detector geometries for identifying and quantifying the

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concentration of radionuclides in a liquid for SiC-based alpha detectors. In this paper, we determine alpha particle energy spectra for various source and detector geometries for perfect planar alpha particle detectors, analytically and through simulations. Four source and detector geometries are considered with increasing complexity: Case A—a planar detector in contact with a semi-infinite radioisotope-containing liquid salt; Case B—a microfluidic channel that is infinitely wide, but thin, above a planar detector surface; Case C—a semi-infinite source volume with a cover on the surface of a planar detector; and, Case D—a combination of the geometries of Cases B and C. Mathematical expressions for the alpha particle energy spectra are determined so as to be able to assess the functional dependence of the energy spectra on various geometric parameters, such as the thickness of the cover or the source volume. The validity of the mathematical expressions are confirmed by simulations using the code package SRIM, the Stopping and Range of Ions in Matter [5]. The validation is confirmed for source activities and energies and source and cover materials that are relevant for pyroprocessing in a molten salt which is maintained at about 500 °C in the Mk. V electrorefiner at Idaho National Laboratory [3].

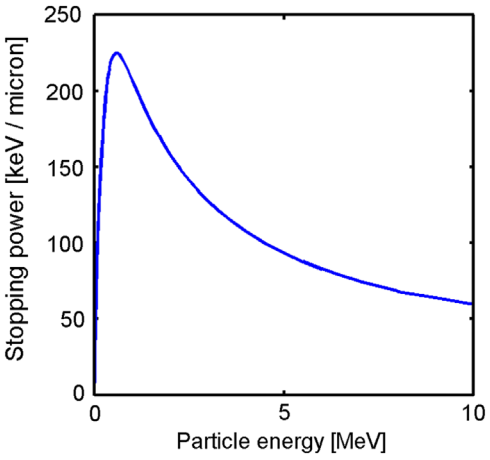

Fig. 1. The stopping power, $S_{salt}(E)$, of molten LiCl–KCl eutectic salt at a density corresponding to 500 °C. This stopping power was calculated using the range table function in SRIM 2011.

2. Analysis—Alpha particle spectra for different salt-cover-detector geometries

The general approach to calculating an alpha particle energy spectrum starts with finding the number of particles crossing a unit area at a given point on the salt-detector interface per unit time, i.e. the particle partial current. For this, we assume that the salt is mixed well enough that all alpha-emitting isotopes can be treated as uniformly distributed in the salt. Furthermore, we note that alpha particles will be emitted isotropically.

The differential (with respect to energy) partial particle current into the detector ("+" direction), can be expressed as

$$\frac{dj_+}{dE_{incident}} = \frac{dr}{dE_{incident}} \frac{s_v}{4\pi} \mu d\Omega, \tag{1}$$

where $E_{incident}$ is the recorded energy of the alpha particles, $dj_+/dE_{incident}$ has units of alpha particles per unit energy per unit area per unit time, $E_{incident}$ is the recorded energy of the alpha particles, r is the radius of a sphere centered on the point detector, dr is the differential thickness of that sphere, s_v is the differential volumetric source term in units of alpha particles emitted per second per volume, μ is the cosine of θ (the angle between the unit vector normal to the planar detector surface at the detector point and the unit vector in the direction from the detector point to the differential source volume), and $d\Omega$ is an element of solid angle associated with a differential surface area on the sphere [6].

In order to calculate the spectrum, one needs to find an expression for r in terms of $E_{incident}$, calculate $dr/dE_{incident}$, and integrate over $d\Omega$ for physically-relevant limits of integration. This calculation is assisted by the fact that alpha particles traveling through a material travel with a well-known relationship between their energy E, and range $R_{mat}(E)$, which is the average distance that particle travels in the material before stopping. This well-known relationship is governed by the well-defined relationship (stopping power) between an alpha particle's instantaneous energy, E, and the energy loss of that alpha particle as it travels through the material, $S_{salt}(E)$. Fig. 1 shows the dependence of $S_{salt}(E)$ on E. As mentioned above, four cases will be examined here, resulting in four different expressions for r versus $E_{incident}$, $dr/dE_{incident}$, and the limits of integration of $d\Omega$.

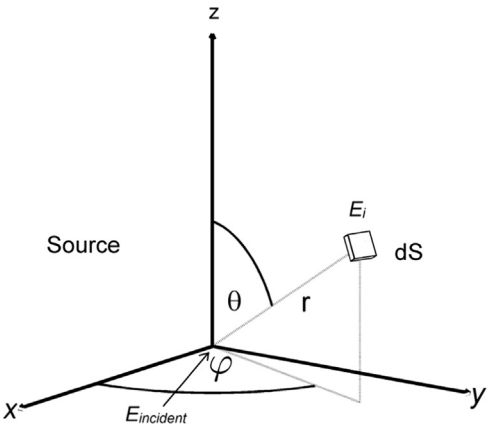

Fig. 2. Geometry for the calculation of Case A, the infinite source spectrum. A uniform isotropically-emitting alpha source is located in the $+n$ volume. The detector plane is the x–y plane through the origin, and a point detector is located at the origin. The differential source term, dS, can be expressed as $dS = s_v r^2 dr d\Omega$, where dr is the differential distance in the direction of r and $d\Omega$ is the differential solid angle.

2.1. Infinite salt thickness, with no protective cover

Case A is where the detector face is placed directly in contact with the semi-infinite alpha-emitting salt, with no protective cover. Fig. 2 shows the geometry for this calculation, where the detector surface lies in the x–y plane through the origin and the source material is the volume in the $+z$ direction.

To find an expression for $dr/dE_{incident}$, we assume that we are investigating only one alpha particle birth (initial) energy, E_i, and note that r corresponds to the distance the alpha particles striking the detector with energy $E_{incident}$ have travelled to get to the detector;

$$r = R_{salt}(E_i) - R_{salt}(E_{incident}), \tag{2}$$

where $R_{salt}(E)$ is the range of alpha particles in the salt at energy E. Fig. 3 illustrates the relationship among r, $R_{salt}(E_i)$, and $R_{salt}(E_{incident})$.

Noting

$$\frac{dr}{dE_{incident}} = \left.\frac{dR_{salt}}{dE}\right|_{E_{incident}}, \tag{3}$$