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Rapid alpha spectrometry from liquids doped with ^{209}Po and ^{241}Am using simplified sample processing

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HIGHLIGHTS

- Rapid filtration/evaporation of liquid samples is possible for alpha spectrometry.
- Radionuclides may behave differently in simplified sample processing.
- Shape of the alpha peaks may depend on the nuclide present in the sample.
- Spectrum unfolding programs should account for different peak shapes.

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ABSTRACT

Rapid detection of alpha-particle emitting radionuclides from liquids doped with ^{209}Po and ^{241}Am was investigated. These nuclides may mimic or be among those that could be used as threat agents in malevolent actions such as deliberate poisoning of refreshments. The liquids investigated here by using a semiconductor alpha detector in vacuum were coffee, beer and apple juice. Methods for the sample processing were rapid evaporation of the liquids and filtration. Both methods produced samples, which gave alpha spectra that could be readily analyzed by an unfolding program. The doped radionuclides were easily identified from the spectra, but the samples were too thick for reliable activity determination. Determination of the activity ratios is possible if the peak shapes are equal. However, radionuclides may behave differently during the sample preparation, which must be accounted for in the activity ratio determination.

1. Introduction

Rapid methods to detect alpha-particle emitting radionuclides from liquid foods are of importance in nuclear security missions, such as in the Litvinenko case (Harrison et al., 2007), when malicious use of alpha-particle emitting radionuclides is suspected. Rapid methods are also relevant in a nuclear incident when a large number of individuals may be exposed to a set radionuclides released into the environment. However, in conventional alpha spectrometry the radiochemical sample treatment may be too tedious and slow, especially when the radionuclide identification is of concern. In a nuclear security mission, the measurements are usually done in order to prevent malicious actions, whereas in a nuclear incident the measurements are typically done after the incident in question. Although development of viable and fast measurement techniques is essential in both cases, we focus here on prevention measurements.

Simplified methods have been developed to detect, identify and determine alpha-particle emitting radionuclides either directly from the liquid (Egorov et al., 2005) or by using robust procedures for the sample processing (for example Kiliari and Pashalidis, 2010; Vesterbacka and Pöllänen, 2010; Semkow et al., 2009; Pöllänen et al., 2005). The data acquisition is usually done in a vacuum, although in-situ measurements at ambient air pressure are also possible (Pöllänen et al., 2012).

Alpha spectrometry equipment, basically similar to those used in a laboratory for high-resolution measurements, have been mounted in moving vehicles (Hoffman et al., 2011; Smolander and Toivonen, 2004) but detailed radiochemical sample processing cannot be performed in these mobile systems. Simplified methods are necessary in field measurements or when the sampling and sample manipulation are to be done by a person who is not familiar with sample processing techniques.

In this short communication, we investigate the possibility to rapidly detect and identify alpha-particle emitting radionuclides doped into different refreshments using two simple methods: evaporation of the liquid with subsequent measurement of the

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Table 1
Activity concentrations of ^{209}Po and ^{241}Am doped in the liquids and data acquisition times, t_{acq}, for the evaporation residues, filters and evaporation residues after the filtration. The measurement times are here much longer than might be in a real case (for example in a nuclear accident) because of the low activities of the samples.

Liquid	^{209}Po [Bq/mL]	^{241}Am [Bq/mL]	Evaporation residue t_{acq} [h]	Filter t_{acq} [h]	Residue after filtration t_{acq} [h]
Coffee	1.1	1.1	20.4	70.3	27.9
Beer	1.3	1.1	20.4	70.3	27.9
Apple juice	0.8	1.2	20.4	70.3	27.9

residue, and filtration of the liquid with subsequent measurement of the filter and the residue. These methods can be used in the field for screening the samples before more sophisticated radiochemical analyses in a fixed laboratory, but they can also be applied for radionuclide identification and even for activity ratio determination. Development of straightforward procedures is of importance for practitioners operating in the field. Preliminary data presented here may also be valuable for investigators developing simplified sample processing tools or spectrum unfolding programs.

2. Materials and methods

The liquids used in the investigation were coffee, beer, and apple juice (Table 1). Results for the water doped with (or containing) different radionuclides were reported elsewhere (Keevil et al., 2011, Vesterbacka and Pöllänen, 2010). Here, alpha-particle emitting radionuclides doped into the liquids were ^{209}Po and ^{241}Am, mimicking nuclides that may be used in deliberate poisoning of the refreshments. Emission energies of their main alpha peaks are 4883 keV and 5486 keV, respectively. Since liquids cannot be measured in a vacuum, the samples from each liquid type were prepared using two simple methods as follows (full

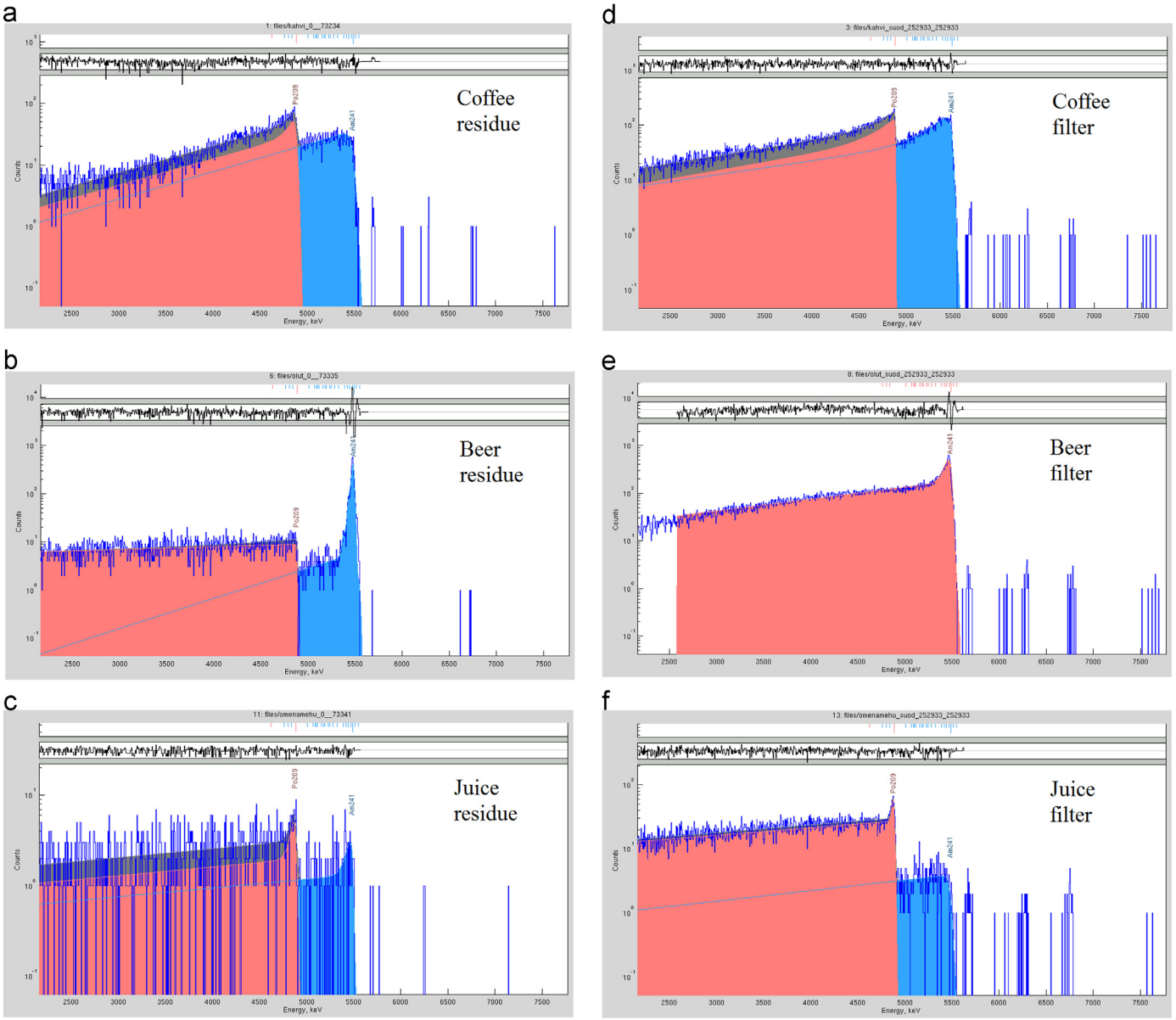

Fig. 1. Alpha spectra containing peaks of ^{209}Po and ^{241}Am measured from samples of coffee, beer and apple juice. Spectra from the evaporated residues are presented in the left and spectra from the filters are in the right. The spectra were analyzed using ADAM (snapshots from the screen). The reduced residual is presented at the top of each figure.