



## APPLICATION OF ACCELERATOR MASS SPECTROMETRY TO NUCLEAR SAFEGUARDS

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### Introduction

Improvements in the safeguards system in the last ten years have focussed on verifying the completeness of declarations made by States. Environmental sampling has been introduced, for samples taken within and around declared facilities. To provide assurance of the absence of undeclared facilities, the concept of wide area environmental sampling (WAES) has been proposed and studied [1]. WAES is based on the assumption that operation of undeclared nuclear facilities will result in the routine or accidental release small amounts of radionuclides, for example, certain long-lived fission and activation products. Key signatures of reprocessing include  $^{85}\text{Kr}$ ,  $^{90}\text{Sr}$ ,  $^{134,137}\text{Cs}$ ,  $^{129}\text{I}$  and Pu isotopes. Although not recognised in earlier work,  $^{236}\text{U}$  also has great potential, as it has higher yield in uranium irradiation than any fission product.

The expected levels of the WAES signature radionuclides are very low and pose considerable challenges to sampling and detection techniques. In the cases of  $^{129}\text{I}$  and  $^{236}\text{U}$ , Accelerator Mass Spectrometry (AMS) offers the most sensitive detection method available. While  $^{129}\text{I}$  has been measured for a number of years by AMS, at ANSTO we have pioneered the measurement of  $^{236}\text{U}$  in environmental materials by this method [2].

### Accelerator Mass Spectrometry

AMS (see ref. [3]) differs from other kinds of mass spectrometry in accelerating ions to energies in the range 10-100 MeV. AMS measures isotopic ratios, by counting the ions of the rare radioisotope while measuring the beam current of the corresponding stable isotope. With the use of a Tandem accelerator, the method has three key properties:

- (i) negative ions are injected; this eliminates stable isobars of elements which do not form negative ions; for example,  $^{129}\text{I}$  is free from xenon interference;
- (ii) interfering molecular ions are destroyed by stripping ions to high charge states in the high voltage terminal; for example  $^{236}\text{U}$  detection is free of  $^{235}\text{UH}$ ;
- (iii) at high energies, ions can be positively identified through precise measurement of their energy, rate of energy loss and time-of-flight; for example  $^{129}\text{I}$  can be distinguished from multiply-scattered  $^{127}\text{I}$  ions.

As a result, AMS has a very high abundance sensitivity: in some cases, isotopic ratios can be measured with a detection limit as low as  $10^{-15}$  (for example, radiocarbon, where the  $^{14}\text{C}:^{12}\text{C}$  ratio is measured in samples containing about 1mg carbon). In the case of uranium, where typical environmental samples yield about  $1\mu\text{g}$  of uranium, the current detection limit is  $10^{-9}$  for the  $^{236}\text{U}:^{238}\text{U}$  ratio. This corresponds to a detection limit of around 1femtogram for  $^{236}\text{U}$ . AMS has also been shown to have this level of sensitivity for Pu isotopes and  $^{237}\text{Np}$  [4].

## Iodine-129

Prior to AMS, few measurements of  $^{129}\text{I}$  in the environment had been performed due to the difficulty of its detection. Using AMS, very low concentrations of  $^{129}\text{I}$  are being measured in the environment, contributing to an understanding of its global background variability and transport, and facilitating its use as a monitor of nuclear activity. It has been shown to occur naturally, with an isotopic ratio  $^{129}\text{I}:^{127}\text{I}$  in the range  $10^{-11}$  to  $10^{-14}$ . However, anthropogenic sources of  $^{129}\text{I}$  now dominate in all environments exposed to global fall-out. It has been detected as originating from many sources, including reprocessing facilities, accidents at nuclear facilities and nuclear weapons tests [5]. Some of these have resulted in general global fall-out of  $^{129}\text{I}$ , while other studies have shown local and regional distribution of this isotope from specific sources.

## Uranium-236

$^{236}\text{U}$  is produced naturally, at very low levels, in uranium ores due to the low neutron flux existing in ore bodies. The  $^{236}\text{U}:^{238}\text{U}$  ratio has been found by AMS to be in the range  $10^{-10}$  to  $10^{-11}$  by measuring milligram-sized samples [6]. These low ratios contrast sharply with nuclear fuel, in which the  $^{236}\text{U}:^{238}\text{U}$  ratio builds up to  $10^{-2}$  to  $10^{-4}$ . Such high ratios relative to the naturally-occurring ratio make its measurement a promising signature of irradiated uranium contamination in the environment.

Recent results for environmental samples show that in areas of expected contamination  $^{236}\text{U}:^{238}\text{U}$  ratios are elevated relative to sites remote from sources of contamination [2]. For example, sediments from the Irish Sea close to the Sellafield reprocessing site exhibit ratios of  $10^{-5}$  to  $10^{-6}$ , about one thousand times higher than sediments from areas unaffected by direct sources of contamination. Similar contrasts are seen for terrestrial samples taken from close to, and far away from, sources of contamination, including Chernobyl, a nuclear test site and reprocessing facilities.  $^{236}\text{U}$  is also present in depleted uranium as a result of the use of recycled uranium in enrichment plants.

## REFERENCES

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