Ultra-sensitive Detection of Nuclear Signatures in Support of IAEA Safeguards

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Summary

The International Atomic Energy Agency (IAEA) applies a range of ultra-sensitive detection techniques to provide assurance that Member States are in compliance with their safeguards agreements. Environmental samples are collected which can contain minute traces of nuclear material or other evidence. Careful analysis of these samples reveals the nature of the activities undertaken in the vicinity of the sampling point. This paper reviews the analytical techniques that are being applied.

To ensure that the IAEA has access to the best available methods, samples are distributed to a group of qualified laboratories around the world for analysis. The Accelerator Mass Spectrometry facility at the Australian Nuclear Science and Technology Organisation (ANSTO) is part of this select group of laboratories, and is the only AMS facility currently accredited with the IAEA. AMS provides the highest sensitivity available for detection of particularly useful signature radioisotopes, including ^{129}I, ^{236}U and plutonium isotopes.

Introduction

Traditional safeguards methods have been applied by the IAEA using verification measures based on nuclear material accountancy and containment and surveillance. These methods verify the *correctness* of the declaration of nuclear material inventories made by Member States. The challenge for the safeguards system is to be able to verify the *completeness* of declarations made by States. In particular, the system must be able to provide assurance of the *absence of undeclared nuclear material and activities*, and be able to provide that assurance with a high level of confidence. Also, it must be able to provide assurance of the *absence of undeclared facilities*. The new *strengthened* safeguards system, developed by the IAEA over the past twelve years, shifts focus from safeguards implementation at the facility level to the State as a whole. Information is obtained from a variety of sources: from States themselves, inspection activities, open sources, satellite imagery and environmental sampling. In the following we will focus on the latter source of information.

Samples taken from within and around declared facilities are analysed. Present and past activities undertaken at a facility disperse and leave behind minute traces of nuclear material and other evidence. The elemental and isotopic composition of these traces, or the presence of certain radioisotopes, act as signatures of the particular activities undertaken. The results are compared to the signatures that would be expected to result from the declared activities of the facility. Any anomalies found are subject to further investigation and inquiries. To provide assurance of the absence of undeclared facilities, the concept of wide area environmental sampling (WAES) has been studied [1] and is being further evaluated.

Environmental Sampling

Since 1996, environmental samples in the form of cotton surface wipes (swipes) have been taken from within a large number of enrichment plants and facilities with hot cells [2]. A range of analytical methods is engaged to study the material collected. The isotopic ratios of uranium and plutonium are of most interest, as this gives information on degree of enrichment and/or fuel burn-up. The first step involves screening by high resolution gamma spectroscopy, performed at the IAEA's Safeguards Analytical Laboratory at Seibersdorf near Vienna. Detailed further analyses are then undertaken either at Seibersdorf or through distribution of samples among a group of expert laboratories around the world. These laboratories must undergo a rigorous accreditation procedure to qualify for membership of the Safeguards Network of Analytical Laboratories (NWAL).

Bulk analysis of swipes is undertaken using X-ray fluorescence, isotope dilution thermal ionisation mass spectrometry (TIMS) and other techniques. Very high sensitivity can be achieved – with TIMS, isotopic analysis of samples containing a total of less

than 1pg of nuclear material can be performed. However, the nuclear material often exists on the swipes as an array of minute particles, and bulk analysis provides only the average isotopic composition. Information about the extremes of isotopic composition, which is of particular interest for nuclear safeguards, may be lost in the bulk analysis.

The most powerful technique available to elucidate the present and past activities of a facility is particle analysis: the isotopic analysis of individual particles picked up on a swipe. The particles are typically a few microns diameter. First it is necessary to identify the 'hot' particles – those containing a predominance of nuclear material – then each hot particle is analysed individually, to determine the U and Pu isotopic ratios.

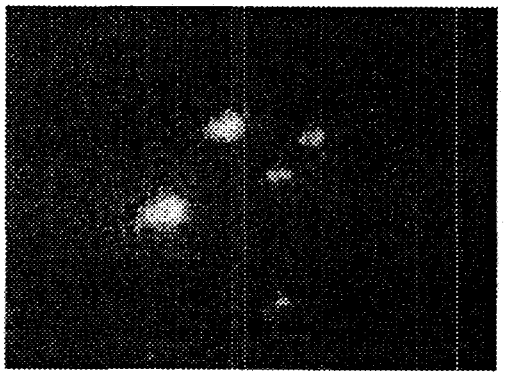

Figure 1: SIMS image of uranium-238 obtained by rastering a <10μm beam spot across a 150μm field of view. Particle sizes range from 6-15um.

Two methods are used for particle analysis. In the first, particles are removed from the swipe and deposited on a track-etch polymer (Lexan), which is then irradiated in a reactor. Particles containing a high proportion of fissionable material are identified visually by their high track density; these particles are then transferred individually on to rhenium filaments for isotopic analysis by TIMS. This method has high sensitivity, high precision, and high cost.

In the second, less costly method, secondary ion mass spectrometry (SIMS) is used both to find the particles, in scanning mode, and then to perform the isotopic analysis on particles identified as containing uranium or plutonium. ANSTO participated in an inter-laboratory comparison of SIMS analysis capabilities for uranium particles; Figure 1 shows a scan of one of the test samples. A technique has also been developed whereby scanning electron microscopy (SEM) is used to locate the hot particles prior to transfer to and re-location in SIMS, where the isotopic analysis is performed [3].

Wide Area Environmental Sampling (WAES)

It is a considerable challenge for the IAEA to be able to provide assurance of the absence of undeclared facilities. Such facilities may be located anywhere where the necessary infrastructure exists. The first obstacle is a political one: under standard safeguards agreements, the IAEA is only permitted to inspect and take samples at or around declared facilities. Under the Additional Protocol to Safeguards agreements, much more extensive inspections are permitted. Parties to the non-proliferation treaty are being encouraged to sign up to the Additional Protocol; Australia was the first signatory back in 1997.

There is also a considerable technical challenge posed by the problem of undeclared facilities. A combination of theoretical studies and field trials has been undertaken to evaluate the possibilities. A report prepared for the IAEA in 1999 [1] concluded that the most promising technical approach would involve the use of networks of high volume air samplers. Air filters can reveal signatures of nuclear activities through high sensitivity bulk analysis. As with swipe analysis, isotopic ratios of U and Pu are of particular interest. Other signatures have also been considered. A small reprocessing operation, for example, would release certain long-lived fission and activation products including ^{85}Kr, ^{90}Sr, ^{134,137}Cs, ^{129}I and Pu isotopes [1].

At ANSTO we have pursued the use of the ^{129}I and ^{236}U as particularly promising signatures for use in WAES. ^{129}I can be detected at extremely low levels by Accelerator Mass Spectrometry (AMS – see below), although this was not appreciated by the authors of the IAEA report [1]. Field trials and other studies [4] have demonstrated the potential of ^{129}I. In addition, we have developed a unique capability for detection of ^{236}U in environmental media [5], and demonstrated its usefulness as a signature of reprocessing. Using AMS allows the detection of minute traces of irradiated uranium, which contains ^{236}U, against a background dominated by natural uranium, which is virtually free of ^{236}U [6].

Accelerator Mass Spectrometry (AMS)

The AMS laboratory at ANSTO is currently the only AMS facility which is accredited as a member of NWAL. It qualified on the basis of its quality-assured measurement capability for ^{129}I and ^{236}U.

AMS (see ref. [7]) 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}I is free from ^{129}Xe interference;

(ii) interfering molecular ions are destroyed by stripping ions to high charge states in the high voltage terminal; for example ^{236}U detection is free of ^{235}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}I can be distinguished from multiply-scattered ^{127}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}C:^{12}C ratio is measured in samples containing 1mg carbon or less). In the case of uranium, where typical environmental samples yield about 1μg of uranium, the current detection limit is 10^{-9} for the ^{236}U:^{238}U ratio. This corresponds to a detection limit of around 1 femtogram for ^{236}U.

To illustrate the high sensitivity of AMS, Figure 2 shows the results for a sample from a recent inter-laboratory comparison exercise, NUSIMEP-2 (Nuclear Signatures Interlaboratory Measurement Evaluation Programme), run by the Institute for Reference Materials and Measurement [8]. The sample contained 0.1μg of uranium and had a 236:238 ratio of 10^{-7}. This ratio proved to be below the detection limit of all other participating laboratories, who were using a variety of mass spectrometry (ICP-MS, TIMS) and radiometric methods. While AMS can eliminate interference from molecular species, ICP-MS and TIMS must rely on background subtractions to correct for molecules. The number of measurements with inaccurate results for this sample indicates problems

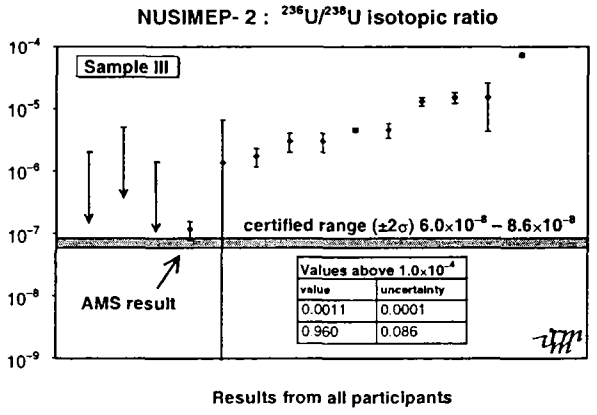

Figure 2: All results for Sample III in the NUSIMEP intercomparison including our AMS result.

with such corrections, or possibly problems with cross-contamination between samples.

AMS has also been shown to have femtogram-level sensitivity for Plutonium isotopes and ^{237}Np [9]. We are now extending our capabilities at ANSTO to include Pu isotopes, with a view to providing the IAEA network with higher sensitivity bulk analysis of swipe samples. At present, there is sometimes insufficient material picked up on swipes for analysis by methods such as TIMS, leaving the IAEA unable to draw conclusions from those samples.

Discussion

In recent times, the role of the IAEA in nuclear safeguards has become a subject of high importance in international affairs and a subject of intense public interest. ANSTO's technical and scientific expertise with a cutting-edge technique – AMS – has enabled us to assist the IAEA in its most vital mission. We have fulfilled this role by originating the ideas, such as the use of ^{236}U in WAES, by doing the necessary research and development, and now by providing a unique analytical service as part of the IAEA's network of analytical labs.

The effectiveness of environmental swipe sampling is now well established. However, further improvements are possible and we are developing higher sensitivity Pu detection for this purpose. Wide area environmental sampling continues to be evaluated through field trials and other sampling. Air sampling systems were deployed in Iraq [10] by the IAEA Action Team.

The techniques discussed in this paper can also be applied to other situations where the presence and nature of minute traces of nuclear or radioactive material may be evidence of unauthorised activities. This area of "nuclear forensics" was the topic of a recent IAEA conference [11] which brought together scientists and other experts in these areas. Particle analysis and isotopic ratio measurements have been used in combination with a range of other techniques in a number of cases of theft and illicit trafficking of nuclear material.

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