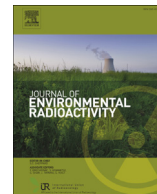




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Accelerator Mass Spectrometry (AMS) in Radioecology

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ABSTRACT

Accelerator Mass Spectrometry (AMS) provides with an excellent sensitivity for the determination of radionuclides in the environment. In fact, conventional radiometric techniques can hardly compete with AMS in the solution of many problems involving the measurement of very low levels of radioactivity in Nature. For that reason, during the last years AMS has become a powerful tool for Radioecology studies.

In this paper a review is done on the evolution of AMS applications to the measurement of environmental radioactivity and, therefore, its contribution to the understanding of radionuclide behavior in Nature. For that, the advantages of using AMS to determine key nuclides as ^{129}I , ^{14}C , Pu-isotopes and others in different natural compartments will be discussed. The content of the paper is illustrated with the contributions to these studies of the Spanish National Center for Accelerators (CNA) AMS systems.

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1. Introduction

One of the main interest in Radioecology is the study of the dynamics and behaviour of radionuclides in the environment, their sources and fate. For that we need to measure radioactivity in Nature. In the majority of the cases at low or very low levels. Radiometric methods solve this problem by the direct measurement of the radiations emitted by the radionuclides as a way of determining its activity. The basis for these methods is the well known Equation.

$$A = \lambda N \quad (1)$$

Being A the activity of the sample, λ the disintegration constant and N the number of radioactive nuclei. There are many examples in the literature which show how these methods have succeeded all along the history of Radioecology. In many cases, however, they cannot give data with reasonable uncertainty, or simply the measurement is impossible, due to the extremely low concentration of the radionuclide or its very long half-life, or both. In these cases it is better to measure N instead of A . That is possible by using mass spectrometry techniques where the application of electric and magnetic fields to a charged beam, that contains the radionuclide of interest, produces a deviation of its trajectory according to its mass and energy.

Accelerator Mass Spectrometry (AMS) is a veteran mass

spectrometry technique which consists of the coupling of a mass spectrometer to an accelerator of tandem type. Its sensitivity has made it a very competitive analytical technique. In fact very long lived radionuclides can be determined with limits of detection close or below to one part in 10^{15} . A brief description of AMS is given in the next Section. After, its application to the measurement to key radionuclides in Radioecology is discussed.

2. Methods

AMS was proposed for the first time by Alvarez and Cornog (1939) which used a cyclotron to identify ^3He in Helium. Under its actual form, however, AMS appears by 1977 (Gove et al. 2010) as a way to solve the limitations of conventional radiometric techniques for ^{14}C dating. After this moment it has rapidly evolved to find application in many disciplines. One of them, the measurement of Environmental Radioactivity.

In Fig. 1 we present a simplified scheme of an AMS system which is well representative of many working systems all along the world. The beam, containing the radionuclide of interest is extracted from the sample in the ion source at a charge state 1^- . In the low energy side of the system, the beam is analyzed for mass with a magnet. After that it is injected in the accelerator tank and accelerated in two steps. First before the stripping and then after the stripping process during which the beam becomes positively charged. The positive beam is analyzed once more for mass and energy at the high energy side of the spectrometer with a magnet and an electrostatic deflector. The selected mass is then driven to the radiation

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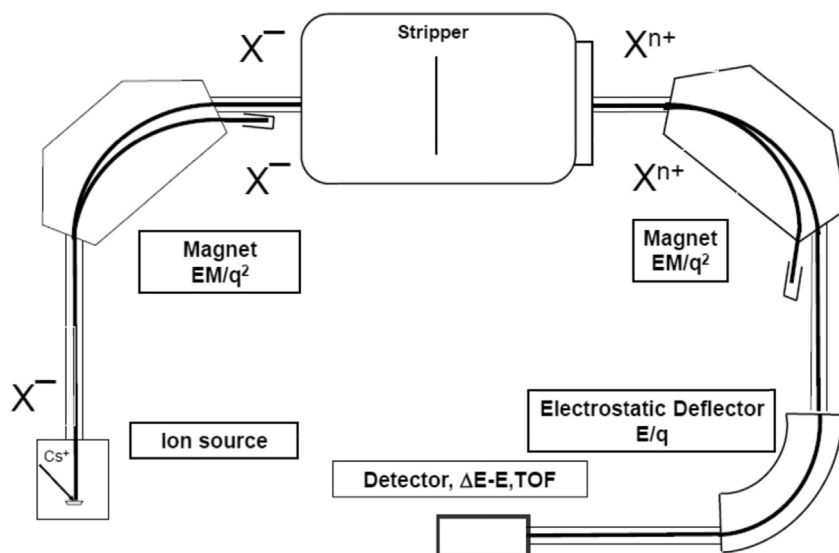


Fig. 1. AMS typical system. It is a scheme of the CNA 1 MV Cockcroft-Walton spectrometer.

detector where the number of radionuclides of interest are counted.

The advantages of AMS regarding conventional mass spectrometry come basically from two special features. First, the dramatic lowering of the molecular background as a consequence of the stripping process, since the majority of molecules are unstable under the resulting strong positive charge states. Second, since the beam is accelerated until energies of the order of MeV/nucleon, it is possible to use nuclear radiation detectors and, consequently, to add to the problem new boundary conditions that permit the discrimination of isobaric species. The use of ΔE -E telescopes make possible the measurement of stopping powers and the application of the Bethe-Bloch equation to distinguish isobars. Or the use of Time of Flight Spectrometers allow the measurement of different masses, even slightly different masses, at the same energy. As an additional advantage, the use of negative ions at the beginning makes a previous selection of the ions injected in the system. This is very convenient for many radionuclides. In fact, the discovery of the instability of $^{14}\text{N}^-$, the main interfering nucleus of ^{14}C , gave place to the development of a new radiocarbon dating technique based on AMS as described above (Gove et al. 2010).

At these conditions the sensitivity one can reach with AMS makes it a really powerful tool to afford many problems in Environmental Sciences and other disciplines. A good review on the Limits of Detection of AMS for many cases can be found in Tuniz and Norton (2008).

The main drawback of the technique has been the need of large facilities. Nevertheless, since the beginning of this century it has been developed the so called Low Energy AMS with uses low terminal voltage accelerators (below 1 MV) and small dimensions machines. An example is the National Center for Accelerators (CNA) AMS system based on 1 MV Cockcroft-Walton tandem accelerator. The dimensions of such spectrometer are relatively small, $3.8 \times 6.3 \text{ m}^2$. Even smaller systems can be fairly competitive in AMS. An example is the Spanish MICADAS, $3 \times 2.5 \text{ m}$ in size, based on a 200 kV van de Graaff accelerator. This system is devoted in CNA to ^{14}C measurements. Both are described in Chamizo et al (2015a).

There is an ample documentation about how AMS can successfully meet Radioecology problems. In practical terms the main advantages of AMS is providing extremely good precision in the measurement for very small samples and using low counting

times. Indeed, g or mg for solid samples or few L or fraction of L for liquid samples are very common in the majority of AMS experiments. Counting times of some 20 min for ^{129}I are enough to obtain uncertainties of <10% for its determination with current methods. Another illustrative example is the case of the measurement of Pu-isotopes in environmental samples where some 15 min of counting gives results with an uncertainty of 5% and the additional advantage of measuring ^{239}Pu and ^{240}Pu separately. This is simply impossible to achieve with conventional radiometric methods.

The application of AMS to the measurement of radionuclides in waters, sediments, biota, etc., i.e. to some relevant problems for Radioecology, needs new radiochemical methods for sample preparation. Indeed, the final chemical form of the radionuclide to be measured has to be compatible with the mass spectrometer ion source.

There are many examples in the literature about such investigations. One of them is the procedure developed at CNA by Chamizo et al. (2008) for the separation of Pu from different matrices based on the use of TEVA resins. The main potential interferents in this case are ^{238}U and Dy isotopes. ^{238}U may reach the detector even in the case the spectrometer is tuned for ^{239}Pu , due to, for instance, instrumental instabilities. $^{160}\text{Dy}^{2+}$ has the same M/q ratio that $^{240}\text{Pu}^{3+}$ and, therefore, can also be counted by the detector. The method provides good decontamination factors that makes possible the measurement of ^{239}Pu down to fg levels. Pu is injected in the spectrometer as Pu oxide.

Another interesting example is that presented by Gómez-Guzmán et al. (2010) also at CNA for the separation of ^{129}I from lichens based on a microwave digestion which significantly diminishes the processing time regarding the conventional leaching or combustion based methods. As a last step of the method, Iodine is precipitated as AgI.

3. Results

The range of applications of AMS and the number of radionuclides that can be measured with it are still expanding. Nowadays, however, many relevant radionuclides for Radioecology can be tried by AMS. A very brief summary of such applications are given in what follows.

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