ACCELERATOR MICROANALYSIS

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Abstract

The ANTARES tandem accelerator at the Lucas Heights Science and Technology Centre is a facility dedicated to accelerator mass spectrometry and ion beam analysis. Research programs based on accelerator mass spectrometry include applications of the long-lived radioisotopes ^{14}C, ^{10}Be, ^{26}Al, ^{36}Cl and ^{129}I in environmental and quaternary science. Ion beam analysis methods based on elastic recoil detection are used for the *in-situ* determination of specific elements or isotopes in surface materials. New analytical systems include an accelerator mass spectrometry beamline for the measurement of actinide isotopes and a heavy ion microprobe. These capabilities will allow the development of novel applications in nuclear safeguards, materials and life sciences.

1 INTRODUCTION

Tandem electrostatic accelerators have been developed forty years ago to produce high-energy ion beams for nuclear physics research. A major shift towards the use of these accelerators in the analysis of materials composition and structure for scientific and industrial applications has been witnessed in the last two decades. Advanced facilities for ion beam analysis (IBA) and accelerator mass spectrometry (AMS) have been constructed at several nuclear physics laboratories around the world. Three tandem accelerators are used in Australia for a variety of interdisciplinary applications. The 3 MV Tandetron at the HIAF/CSIRO laboratory in Sydney, with a proton microprobe and a SIMS-AMS facility, is mainly applied for minerals research; the 14UD Pelletron at the Australian National University in Canberra is a facility dedicated to heavy-ion nuclear physics, but it is also used for AMS research; and the 10-MV ANTARES accelerator at the Lucas Heights Science and Technology Centre, is a facility presently dedicated to materials and environmental science.

The ANTARES accelerator is based on the FN tandem accelerator originally built by High Voltage Engineering for Rutgers University (New Jersey, USA). Since its arrival in Australia, the accelerator has undergone a complete refurbishment and upgrade. Major items in this transformation are: new spirally-inclined accelerator tubes, a Pelletron charging system, a 59-sample high intensity sputter source, a high resolution and high rigidity injection magnet and a fast sequential isotope injection [1]. Versatility is allowed by multiple beamlines for AMS and IBA applications (Fig. 1).

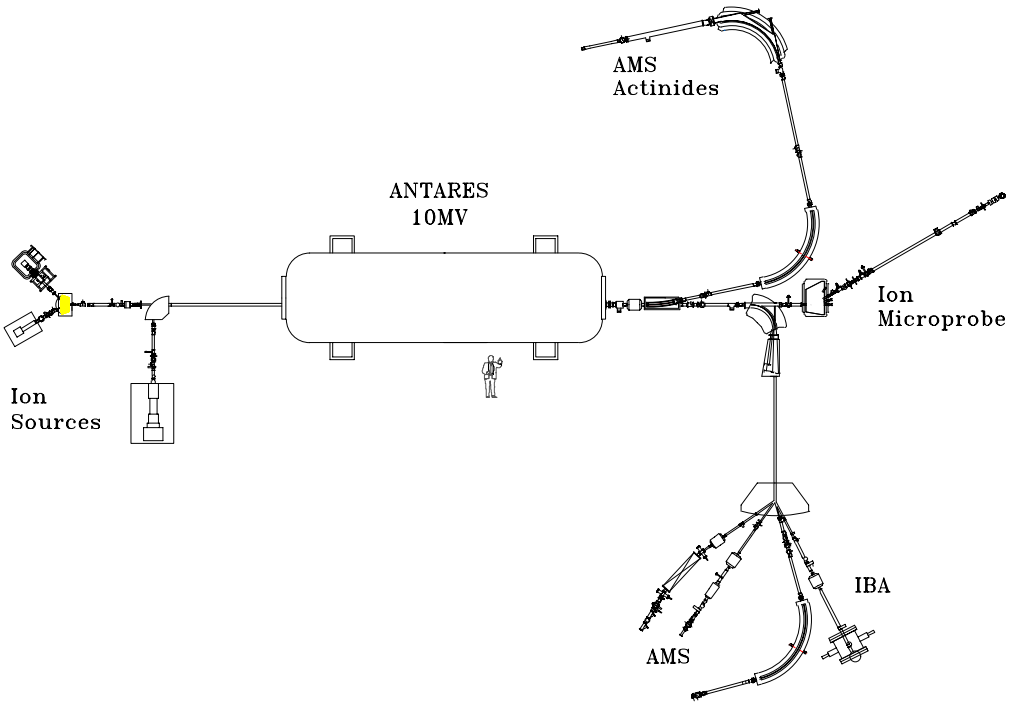

Figure 1: Layout of the ANTARES facility

2 AMS

The AMS method is based on the use of an ion accelerator and its beam transport system as elements of an ultra-sensitive mass and charge spectrometer [2]. Multiple selection stages for energy, momentum, velocity and atomic charge plus final identification of nuclear mass and charge with an ion detector make possible measurements of isotopic ratios some six or seven orders of magnitude smaller than is possible with conventional mass spectrometry. The high isotopic selectivity of AMS enables a dramatic reduction of the backgrounds that plague mass spectrometry: molecular and isobaric interferences and tails of abundant neighbouring masses. For instance, AMS allows an isotopic sensitivity of one part in 10^{15} for ^{14}C, ^{10}Be and other radionuclides occurring in nature at ultra-trace levels. The efficiency of AMS in the detection of long-lived radionuclides is 10^6 - 10^8 times higher than decay counting: the size of the sample required for analysis can be 10^4 - 10^6 times smaller with measuring times 100 times shorter.

In the last 20 years, AMS systems have been developed at more than 40 laboratories for the detection of low-abundance radionuclides in environmental, archaeological and biomedical samples. Electrostatic tandem accelerators are the optimum choice for a variety of AMS applications. Small tandems (2-3 MV) have been specifically designed for ^{14}C analysis. These relatively low-energy tandems can also be used to detect other long-lived radionuclides, such as ^{10}Be, ^{26}Al and ^{129}I or stable isotopes. Larger tandem accelerators, originally used for nuclear physics research, are used to analyse a variety of rare radionuclides, including ^{36}Cl and ^{41}Ca.

2.1 AMS at Lucas Heights

The chemistry laboratories for AMS target preparation are an integral part of the ANSTO AMS facility. Samples containing 0.2 mg or more of original carbon are processed routinely for radiocarbon analysis. The current ^{14}C chemistry background for 1 mg carbon is about 0.2-0.3 percent of modern carbon. With the increasing demand for measurement of extremely small samples in our Antarctic research program, we developed methods for the preparation of targets containing tens of micrograms of carbon. Unknown samples with masses as low as 15 μg carbon have been analysed. We have recently expanded the capabilities of the chemistry laboratories to encompass the preparation of ^{10}Be, ^{26}Al and ^{36}Cl from a variety of environmental samples. Procedures for the extraction of iodine from water, sediments, soils and biota have been developed for ^{129}I analyses.

AMS targets are loaded in the sputter ion source of the ANTARES accelerator. Rapid sequential injection of the isotopes of interest is performed by *bouncing* the chamber of the injection magnet and is essential to obtain high-precision in the measurement of the isotopic ratios. Following injection into the accelerator, negative ions are attracted by the positive voltage at the terminal and thereby accelerated to high energies (e.g. 6 MeV during ^{14}C analysis), at which point they pass through a gas or a foil stripper located at the terminal. The same positive voltage then further accelerates the multi-charged positive ions on the terminal. After the acceleration, combinations of magnetic and electric fields select charge, momentum, energy and velocity of the ions. The 3 beamlines presently dedicated to AMS are equipped with a Wien filter, a 22^o electrostatic analyser and a 90^o electrostatic analyser. The final identification of the rare radionuclide is performed in the ion detector by measuring one or more of the following parameters: total energy, stopping power, range and velocity. Depending on the isotopes to be analysed, a variety of detectors are available for this final stage at the ANTARES AMS spectrometer, including a multi-anode ionisation chamber, a Bragg detector and a time-of-flight system.

3 AMS APPLICATIONS

Long-lived radionuclides are used as tracers and chronometers in disciplines such as geology, archaeology, astrophysics, biomedicine and materials science. Low-level decay counting techniques have been developed in the last half-century to detect the concentration of cosmogenic, radiogenic and anthropogenic radionuclides in a variety of specimens. The radioactivity measurement for long-lived cosmogenic radionuclides, such as ^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl and ^{129}I, is made difficult by low counting rates and in some cases the need for complicated radiochemistry procedures and efficient detectors of soft beta particles and low energy x rays. AMS measures cosmogenic radionuclides in natural samples up to 10^6 times smaller then those required for conventional techniques, allowing novel applications in geology and environmental science. Some of the projects carried out at ANSTO are discussed in the following.

3.1 Global Climate Change

Ice cores are archives of atmospheric air from which to reconstruct levels of greenhouse gases over recent centuries to millennia. Ice cores from Law Dome, East Antarctica, characterised by high accumulation rates but minimal summer melting, provide an unparalleled time resolution through the Holocene and possibly beyond. In

addition, air extracted from the firn permits direct comparison of entrapped trace gas concentrations with modern records. One of the problems is that recent CO_2 growth rate variations are difficult to interpret due to the smearing of ice-core signals induced by the diffusion of air in the firn. In collaboration with the CSIRO Division of Atmospheric Research, ANSTO researchers recently succeeded in using the ^{14}C "bomb spike" to determine the age spread and age of CO_2 in Antarctic ice and firn [3]. Another objective of this collaborative program is to determine the fossil contribution to the global methane budget from the analysis of $^{14}CH_4$ trapped in Antarctic firn before the nuclear age.

A second collaborative program is based on the use of *in-situ* produced cosmogenic radionuclides to determine the chronology and extent of glacial evolution in the southern hemisphere. ^{10}Be, ^{36}Cl and other AMS radioisotopes produced by secondary cosmic rays are analysed in polished glacial bedrock, moraine boulders and till deposits to determine exposure ages, regional erosion rates and uplift rates. These data will help to unravel the timing of glacial and interglacial cycles, ice volume and position, landscape geomorphology etc.

3.2 Monitoring nuclear activities

Nuclear activities introduce into the environment long-lived radionuclides such as ^{129}I and ^{36}Cl. AMS is the analytical technique of choice for the practical analysis of these radionuclides in natural specimens [4]. Isotopic concentrations of 10^6 atoms per gram can be detected in samples taken from a variety of environmental materials such as water, air, soil and biota. In collaboration with the IAEA, the ANSTO AMS group has recently analysed ^{129}I in waters and sediments collected by IAEA inspectors at various distances from a nuclear reprocessing plant. ANSTO researchers have also analysed ^{129}I, ^{14}C and ^{36}Cl in water specimens from the Mururoa lagoon, contributing to an international project aimed at determining the environmental impact of the underground nuclear tests in the Pacific atolls.

3.3 Biomedicine

AMS provides a method for analysing long-lived isotopes of elements for which metabolic and toxicological information is not available. Aluminium, for example, is now considered to be a toxic element, whose accumulation has been identified as the cause of disease states in chronic renal failure patients. Although still a highly controversial issue, aluminium has also been implicated in the aetiology of Alzheimer's disease. Yet, aluminium compounds have been used since last century in water treatment. Without an appropriate radioisotope and being a monoisotopic element, conventional studies of aluminium metabolism have been restricted to large dose quantities of stable aluminium and as such do not reflect normal physiology. Detection via AMS of the long-lived radioisotope ^{26}Al administered at ultra-trace levels and thus with negligible radiation damage can provide a new avenue to understand the role aluminium plays in biological systems. The ANSTO AMS group has detected the presence of ^{26}Al in the brain tissue of Wistar rats gavaged with drinking water containing 70 Becquerel doses of ^{26}Al [5].

3.4 Archaeology

Archaeological projects based on radiocarbon dates performed at ANTARES include detailed research into the probable antiquity of the rock art of Chillagoe and Laura in North Queensland and the Kimberley in Western Australia. A variety of materials are being analysed: pigments, oxalate minerals, silica coatings, plant fibres, carbonised plant matter, fatty acids, beeswax and mud-wasp nests. A number of sample processing techniques have been explored in these studies including low-pressure plasma techniques and laser extraction methods. Intercomparisons between ^{14}C dating and thermoluminescence or optically stimulated luminescence dating are carried out within some of the aforementioned projects. Two archaeological projects are discussed in the following.

The Kimberley rock art sequence is likely to be one of the longest and most complex in the world. On the basis of superimposition and differential weathering, Australian archaeologists have constructed a very detailed rock art sequence of which the major phases include Pecked pits, Irregularly Infilled Animals, Bradshaws, Clawed Hand figures and Wandjinas. This sequence depicts major changes in Aboriginal culture, ideology and local fauna over time. Fieldwork in the region started in 1994 with the aim of providing absolute dates for the Kimberley rock art sequence. Small samples of pigments, beeswax and associated mineral crusts have been collected. AMS dating of these samples has provided the first age estimates for the well-known Bradshaw painting style. Mudwasps, which overlie or underlie Kimberley rock paintings, can be dated by Optically Stimulated Luminescence (OSL), providing minimum or maximum ages for rock paintings. ANSTO provided AMS determinations for some of the same samples, thus allowing comparison between results from two very different dating techniques [6].

Ngarrabullgan or Mount Mulligan, located some 100 km northwest of Cairns, north Queensland, Australia, is a large table top mountain bordered by 300 m high cliffs along most of its periphery. Different vegetation can be noted on the top of Ngarrabullgan and in the savanna woodlands that surround the mountain. Ngarrabullgan Cave, on the top of Ngarrabullgan, is one of the earliest

radiocarbon dated archaeological sites in Australia (37,000 yr BP). The deposits at this site show very low erosion and a near-total absence of territorial vertebrate fauna. Our study shows that intensive use of the mountain started around 5000 yr BP, after 27 millennia of total abandonment. In this work we have also obtained the first paired ^{14}C/OSL determination for pre-30 ka archaeological deposits [7].

4 ION BEAM ANALYSIS

Heavy-ion accelerators provide a variety of high-energy ion beams that can be used to probe the structure of materials and their composition. Ions penetrating the surface of materials lose energy by ionization processes caused by the Coulomb interactions between the projectile and the target electrons and also by nuclear scattering. The range of ions in materials is short, with a relatively well defined end point. By comparison, x-ray and neutron beams are attenuated according to an exponential law and sample a much greater amount of material. Ion beams are applied to trace element determinations using the characteristic x-rays produced in the ionization process. Nuclear reactions, including elastic and inelastic scattering or Coulomb excitation, are useful to identify specific elements and nuclides present in the sample. Concentration measurements of individual elements or isotopes as a function of depth are determined using narrow nuclear resonances and energy loss of ions as they penetrate the material.

An IBA system based on elastic recoil detection analysis (ERDA) is available on one of the ANTARES beamlines. In elastic scattering processes, the target atoms gain momentum in a forward direction. By detecting the atoms leaving the target in a forward direction, information on the concentration and depth distribution of various elements can be obtained. Measurements of time of flight and energy provide unambiguous identification of the knocked-on nuclides. ERDA is being used by ANSTO groups in studies related to Synroc. This material has a high leach resistance in aqueous media, but it is difficult to use H_2O to measure the hydrogen incorporated in leached Synroc, due to the ubiquitous presence of hydrogenous surface contamination. The use of D_2O provides a more sensitive and reliable method for this kind of studies. ERDA techniques with heavy ion beams are used at ANSTO to study deuterium depth penetrations and concentrations in Synroc samples and to evaluate the chemical reactions taking place during the dissolution of this material at different temperatures [8].

5 NEW FACILITIES

As described in the previous sections, the ANTARES accelerator is presently used for the AMS analysis of commonly used long-lived radioisotopes and for the IBA characterisation of materials surfaces. Two new facilities, a heavy ion microprobe and a system for AMS analysis of actinide isotopes, are presently being commissioned.

5.1 The heavy ion microprobe

Ion microbeam analysis uses an ion beam focussed to μm dimensions for elemental imaging. This can be performed by using secondary radiation induced by the primary ion beam, such as x-rays and nuclear reaction products, or by using the energy loss of transmitted primary ions. Pioneering studies with proton microbeams (50 μm diameter) were performed in the mid sixties at the Lucas Heights 3-MV accelerator [9]. These first experiments paved the way for the modern nuclear microprobes, characterised by sub-micron lateral resolution [10]. A heavy-ion microprobe for surface imaging and depth profiling has been recently constructed at ANTARES. This nuclear microprobe has been recently used to focus C, Cl and I beams to lateral dimensions of less than 10 microns.

5.2 AMS of actinides

A new facility is being constructed at ANTARES to analyse rare heavy radionuclides, such as ^{236}U, ^{229,230}Th and ^{244}Pu, in natural samples with ultra-high sensitivity. The main use of this facility will be for the ANSTO program in environmental monitoring for nuclear safeguards. An electrostatic quadrupole doublet has been installed on the high-energy end of the accelerator to provide mass independent focussing of the beam at an external gas stripper, where stripping to higher charge states will allow rejection of molecular fragments having similar M/Q. Momentum and E/Q analysis will be performed with a 90^o magnet (mass-energy product = 250 MeV.amu) and a 90^o electrostatic analyser (ESA). The spherical ESA, manufactured by Danfysik, has a radius of 2.5 m and a nominal maximum rigidity of E/Q = 7.6 MV and an energy dispersion of 5000 in the image plane.

6 CONCLUSIONS

ANSTO is promoting an advanced program in a variety of topics of high international significance such as global climate change and environmental monitoring for nuclear safeguards. Other research projects are related to the processing of novel materials for use in functional devices. The analytical facilities available at ANTARES provide essential capabilities for the development of this program.

7 ACKNOWLEDGMENTS

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8 REFERENCES

[1] C. Tuniz, D. Fink, M.A.C. Hotchkis, G.E. Jacobsen,, E.M. Lawson, A.M. Smith and Q. Hua: 'Research and measurement program at the ANTARES AMS facility', Nucl. Instr. and Methods in Phys. Res. 123 (1997)73.

[2] C. Tuniz, J.R. Bird, D. Fink and G.F. Herzog: 'Accelerator Mass Spectrometry: ultrasensitive analysis for global science', CRC Press, LLC, 1998.

[3] V.A. Levchenko, R.J. Francey, D.M. Etheridge, C. Tuniz, J. Head, V.I. Morgan, E.M. Lawson and G.E. Jacobsen: 'The ^{14}C "bomb spike" determines the age spread and age of CO_2 in Law Dome firn and ice', Geophysical Research Letters 23 (1996) 3345.

[4] C. Tuniz and M.A.C. Hotchkis: 'Accelerator Mass Spectrometry to identify signatures of nuclear activities', Proc. International Workshop on the Status of Measurement Techniques for the Identification of Nuclear Signatures, eds. C. Foggi and F. Genoni, Geel, Belgium, 1997, 149-157.

[5] J. Walton, C. Tuniz, D. Fink, G.E. Jacobsen and D. Wilcox: 'Uptake of Trace Amounts of Aluminium into the Brain from Drinking Water', Neurotoxicology 16 (1995) 2.

[6] R. Roberts, G. Walsh, A. Murray, J. Olley, R. Jones, M. Morwood, C. Tuniz, E.M. Lawson, M. Macphail, D. Bowdery and I. Nauman: 'Luminescence dating of rock art and past environments using mud-wasp nests in northern Australia', Nature 387(1997) 696.

[7] B. David, R. Roberts, C. Tuniz, R. Jones and J. Head: 'New optical and radiocarbon dates from Ngarrabulgan Cave, a Pleistocene archaeological site in Australia: implications for the comparability of time clocks and for the human colonisation of Australia', Antiquity 71(1997) 183.

[8] N. Dytlewski, E.R. Vance, and B.D. Begg: 'Energy-recoil analysis of deuterium incorporated in Synroc by reaction with D_2O at 120 and 190°C', Journal of Nuclear Materials 231(1996) 257.

[9] B.K. Mak, J.R. Bird, and T.M. Sabine: 'Proton microanalysis', Nature 211(1966) 738.

[10] M.B.H. Breese, D.J. Jamieson, P.J.C. King: 'Materials analysis using a nuclear microprobe', John Wiley & Sons, Inc., New York, 1996.