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The ANTARES Accelerator: A Facility for Environmental Monitoring and Materials Characterisation

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SUMMARY ANTARES is a facility dedicated to accelerator mass spectrometry and ion beam analysis. Research programs based on the AMS spectrometer include applications of ^{14}C, ^{10}Be, ^{129}I and other long-lived radionuclides in quaternary science studies, global climate change and environmental monitoring for nuclear safeguards. 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 are under construction, including an AMS beamline for the measurement of actinide isotopes and a heavy ion microprobe for elemental imaging with micron resolution.

1. INTRODUCTION

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, in 1989, the accelerator has undergone a complete refurbishment and upgrade. During the first phase, major items in this transformation have been new spirally-inclined accelerator tubes, a 60-sample high-intensity sputtering source and a high resolution injection magnet (1). Several major elements of the accelerator structure were to improve both the energy stability and the focal properties of the particle beam. The most significant recent upgrades have been the installation of a recirculating gas stripper and a new Pelletron charging system. As a consequence, operations at eight million volts is now possible. Versatility is allowed by multiple beamlines for AMS and IBA applications (Fig. 1)

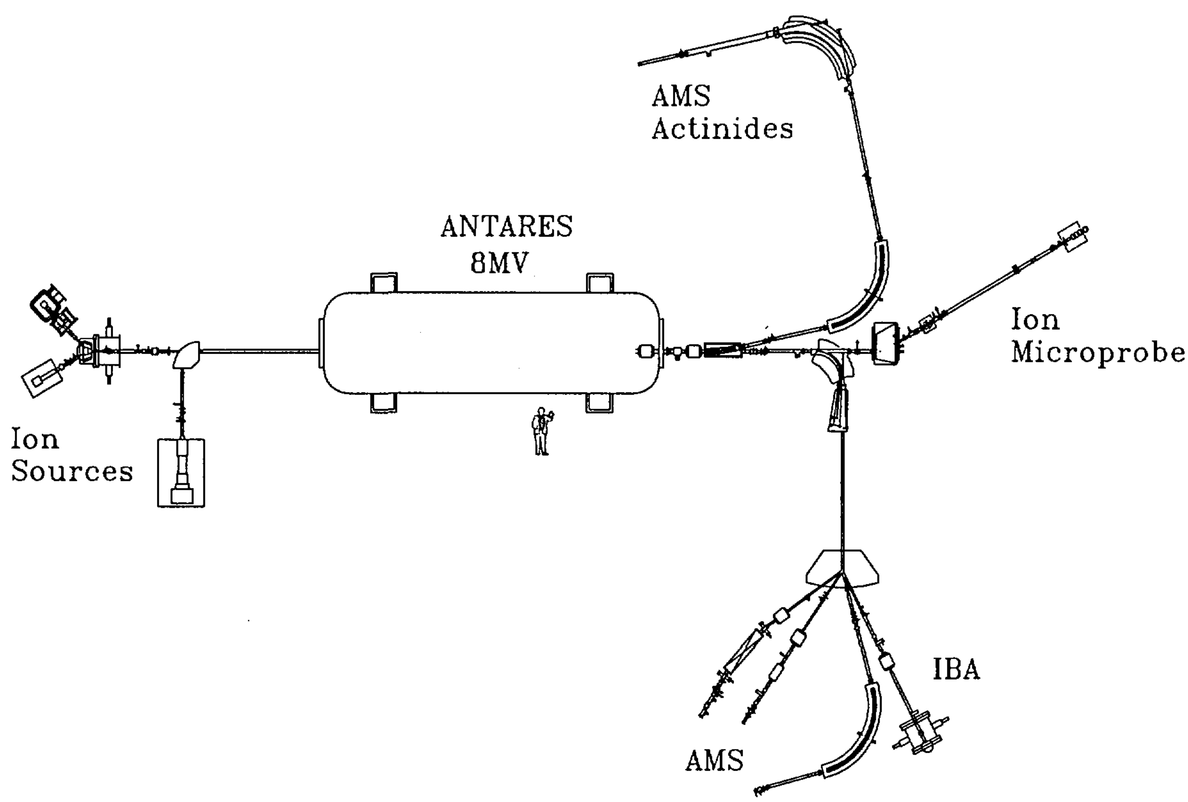

Figure 1 – Layout of the ANTARES facility.

1.1. The ANTARES AMS spectrometer

The ANTARES AMS facility performs ultrasensitive analysis of the long-lived radioisotopes ^{14}C, ^{10}Be, ^{26}Al, ^{36}Cl and ^{129}I.

The chemistry laboratories for 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, we are currently developing methods for the preparation of targets containing tens of micrograms of carbon. Several unknown samples with masses as low as 50 μg carbon have been recently analysed. We are presently expanding 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 are being developed for ^{129}I analyses.

After chemistry processing, AMS targets are loaded in the sputter ion source of the ANTARES accelerator. High precision AMS analysis is carried out by rapid sequential injection of the isotopes of interest, in order to overcome variability in source output and accelerator transmission. 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. Following the acceleration, combinations of magnetic and electric fields select charge, momentum, energy and velocity of the ions. The beamlines dedicated to AMS are equipped with a Wien filter, a 22° electrostatic analyser and a 90° electrostatic analyser, which provide the selectivity necessary to separate the radionuclide of interest. 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 counted, 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.

2. AMS APPLICATIONS

Long-lived radionuclides are used as tracers and chronometers in many disciplines: geology, archaeology, astrophysics, life 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, 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 can measure cosmogenic radionuclides in geological samples up to 10^6 times smaller then those required for conventional techniques, allowing novel applications in geology and environmental science.

2.1 Global climate change

The ANTARES AMS spectrometer is a key facility for reserch programs based on the application of nuclear science and technology to the understanding of natural processes. Some of the projects carried out at ANSTO are discussed in the following.

2.1.1 Long-lived radionuclides in Antarctic ice

Ice cores are providing the best source of preserved 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).

Future research will include studies of the last glacial/interglacial transition, search for a Younger Dryas in the Southern Hemisphere and investigations on changes in CO_2 and CH_4 in the Holocene period, concentrating on particular periods of interest such as the Maunder Minimum.

^{10}Be measurements at annual resolution are planned in cores of known chronology from Low Dome spanning up to several solar cycles to determine the magnitude of the solar modulation and to compare the data with neutron flux records.

An important greenhouse gas is methane, but the anthropogenic contribution to the global methane budget is not well understood. In fact, it is difficult to obtain atmospheric samples prior to the nuclear era for the measurement of ^{14}C and the determination of the methane budget. The large volumes of atmospheric gas trapped in the porous firn overlying the ice offer a solution and a sampling expedition is planned to investigate the feasibility of this approach.

2.1.2 Southern hemisphere glaciation project

Cosmogenic radionuclides such as ^{10}Be, ^{26}Al and ^{36}Cl produced in rocks can provide information on glacial histories in the southern hemisphere. The ANSTO research program is targetting three geographic regions that show distinct glacial formations and deposits: Tasmania, New Zealand and Antarctica. The ANTARES AMS spectrometer is used to analyse glacially polished bedrock surfaces, large erratics and boulders deposited on lateral and terminal moraines and within glacial outlet valleys.

2.1.3 High resolution palaeoclimate records

The aim of this project is to improve the chronologies of key palaenvironmental records in the Asian, Australian and Antarctic regions spanning the last 350,000 years. This project will complete the transect of records for climate change across the southern hemisphere and provide past climate patterns. The research is centred on sediment cores which will be used to reconstruct changes in salinity, temperature, nutrient levels, ice cover and productivity in the region from evidence provided by diuatom records, thorium/polonium ratios, carbon-13 values and grain size analysis. Dating of cores is being carried out using AMS radiocarbon techniques and lead-210 dating.

2.2 Monitoring nuclear activities

ANSTO is involved in IAEA programs, supported by the Australian Safeguards Office, to develop new methods for environmental monitoring for nuclear safeguards.

Nuclear activities such as reactor operation and fuel reprocessing 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 locations from a nuclear reprocessing plant. ANSTO researchers are also analysing ^{129}I and other long-lived radionuclides 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. ION BEAM ANALYSIS

Ion accelerators can provide a variety of high-energy ion beams to probe the structure of materials and their composition. Ions penetrating the surface of a material specimen 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 (ERD) 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. ERD 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. ERD techniques using 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 (5).

4. FUTURE DEVELOPMENTS

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, an ion microprobe and a system for AMS analysis of *heavy* rare isotopes, are under construction and will expand present capabilities.

4.1. The heavy ion microprobe

Ion microbeam analysis uses an ion beam focussed to μm dimensions for elemental imaging of materials surfaces. 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 (6). These first experiments paved the way for the modern nuclear microprobes, characterised by sub-micron lateral resolution (7). A heavy-ion microprobe for surface imaging and depth profiling is presently being developed at ANTARES,. This nuclear microprobe is designed to focus a variety of ion beams, including iodine, to lateral dimensions of about 10 microns or less.

4.2. 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 and 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 new analysing magnet (mass-energy product = 250 MeV.amu) and an electrostatic analyser (ESA). The 90° 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.

5. CONCLUSIONS

ANSTO is promoting an advanced research and development 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.

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