THE DEVELOPMENT OF ACCELERATOR BASED MICRO IBA TECHNIQUES FOR THE STUDY OF ENVIRONMENTAL SAMPLES AND MATERIAL CHARACTERISATION.

David Cohen, Rainer Siegel, Ed Stelcer, Mihail Ionescu, David Garton

Institute for Environmental Research,
ANSTO, Locked Bag 2001 DC, Kirrawee, NSW 2232, Australia
David.cohen@ansto.gov.au

17 May 2010

The Australian Nuclear Science and Technology (ANSTO) is a research establishment of around 950 people located approximately 30 km south west of Sydney, Australia. ANSTO has several research institutes, including Bragg, Radiopharm, Materials and Environment. These institutes alone include about 300 research and technical support staff. ANSTO's major neutron facility is the Open Pool Australian Light water reactor (OPAL). It is a 20 MW pool reactor using low enriched uranium fuel, and cooled by water. It is a multipurpose facility for radioisotope production, irradiation services and neutron beam research.

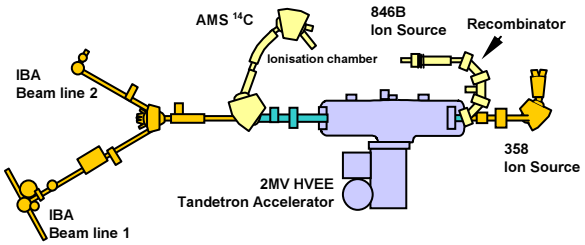
Accelerator Facilities at ANSTO

ANSTO also operates two positive ion accelerators, the 2MV Tandetron, STAR and the 10MV Tandem, ANTARES. There are 3 ion sources and 3 high energy beamlines on STAR and 3 ion sources and 5 beamlines on ANTARES. Both accelerators operate 1 shift a day for approximately 250 days per year (5 days week).

STAR was fully commissioned for sensitive Ion Beam Analysis (IBA) and Accelerator Mass Spectrometry (AMS) operation in February 2005 while ANTARES is an older accelerator which was purchased from Rutgers University, USA in 1989. Both machine have been extensively upgraded and are considered 21st century accelerators. With these two machines we can accelerate most ions in the periodic table from hydrogen to uranium with energies from a few hundred keV to over 100 MeV. This provides comprehensive access to all major

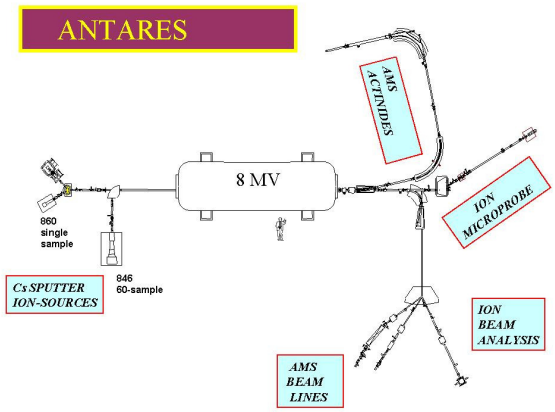
IBA and AMS methods of analysis covering a very broad range of scientific disciplines from Archaeometry to Zoology.

STAR accelerator



STAR beamlines

ANTARES high energy end



ANTARES beamline layout

The accelerator facilities average more than 100 users a year, using the IBA and AMS capabilities on both machines. They perform R&D for all universities around Australia, through the Australian Institute for Nuclear Science and Engineering (AINSE) which provides funding, and commercially for research and environmental organisations (CSIRO, EPA's) and Australian industry. The accelerators are used by all 37 Australian Universities through AINSE. There is considerable external usage of accelerator facilities. For commercial and AINSE users we charge daily fees for these facilities, bringing in external revenue for ANSTO. Commercial revenue tends to fluctuate from year to year and depend strongly on obtaining major grants with industry. It is important to maintain a balance between commercial and R&D outputs. We aim for 40:60 split commercial to R&D research.

Current research themes for accelerators at ANSTO include, radiation dosimetry and micro-dosimetry, nuclear methods for microspectroscopy, mapping and characterisation, climate

change, AMS for nuclear safeguards and nuclear forensics, IBA-Interface Engineering and AMS Biomed.

Recent accelerator based research achievements at ANSTO include**:-**

- The Project has introduced into Australia for the first time Positive Matrix Factorisation (PMF) techniques using IBA analysis methods for determining source fingerprints and their contributions to fine particle pollution in Australia. This is now being rolled out to environmental bodies and Local Councils as a new tool for pollution management and control.
- The Project has developed new ion beam induced charge (IBIC) tools, using the heavy ion microprobe on ANTARES, to test micron sized nuclear detectors used in radiation therapies for cancer treatment.
- In collaboration with the Vietnamese Institute for Nuclear Science and Technology the Project has just finalised (December 2009) a major study using nuclear techniques to characterise air pollution in Vietnam. This study reports for the first time on long term systematic fine particle pollution measurements at Hanoi, Vietnam between 2001 and 2008. The research has been published in international journal Atmospheric Environment.
- A new collaboration involving the University of California Davis, the Australian National University, Monash University and ANSTO has been successfully established to characterise Aeolian dust that is transported from central Australia across the east coast to the Great Barrier Reef. All filters collected are being characterised using the accelerator based ion beam analysis (IBA) techniques at ANSTO. The aim is to quantify the effects of long range transport of dust on the Barrier Reef systems.
- The Project has applied synchrotron XANES and EXAFS analysis techniques in conjunction with heavy ion microprobe analyses to better understand the mechanisms involved in the process of heavy metal hyper-accumulation in native Australian plant systems.
- Heavy ion irradiation techniques have been used to modify the electric properties of polymers creating carbon clusters and making the polymers conductive or semi-conductive. Using high energy iodine beams at low fluences, we have been able demonstrate the directional effect of this conductivity. This may in the future provide a new way to make contacts on semiconductors or new types of sensors.

Scientific Scope and Work Plan

Characterization techniques such as Particle Induced X-ray Emission (PIXE), Particle Induced Gamma Ray Emission (PIGE), and Rutherford Backscattering (RBS) have been developed individually and applied quantitatively for many years at ANSTO for both thick and thin samples. All these techniques rely on MeV ions from low to medium positive ion accelerators.

A current focus of our research is on the development of PIXE, PIGE, ERDA and RBS for the characterisation of fine particle of ambient air pollution collected on continuously on Teflon

strips over several months rather than the normal 24 hrs individual filter collection. These strips have the potential to obtain data with hourly resolution over several months by using IBA techniques to quantify microgram samples of particulate matter with submicron diameters. We are currently developing IBA techniques and the samplers to provide these strips for three particle size fractions, (2.5-1.15)μm, (1.15-0.34)μm and (0.34-0.1)μm and to characterize them for over 20 different elemental species. Our current heavy ion microprobe is capable of producing 2D elemental maps with several micron resolution. We will also be developing a confocal heavy ion microprobe system which uses a swept 5μm beam spot size to generate X-rays which are focused to 20 μm diameter using a polycapillary lens system on an X-ray detector. The intersection of these two beams systems produces a small volume in space which can then be swept though a sample to produce 3D images of the sample with micro resolution.

The 2nd and 3rd stages of this size fraction separation are submicron, below the current spot size of our existing heavy ion microprobe system and we will be using the Australian Synchrotron (AS) Facility in Melbourne to perform this part of the research. This AS facility has 0.1μm resolution for its SXRF elemental 2D mapping systems and is capable of studying both the chemical state (XANES, EXAFS) and the chemical composition of individual fine particles.

It is expected that the proposed research program will develop methods of collecting fine particles on strip filters which are capable of collecting data over several months with better than daily resolution for use by Australian air pollution authorities, providing detailed information for the management of fine particle pollution in Australia.

The proposed research program will develop specific ways to integrate the PIXE, PIGE, ERDA and RBS techniques into one package for characterisation of ambient air in the micron and submicron diameter particle range. It will develop new confocal heavy ion microprobe facilities and capabilities not previously available to Australian Universities for 2D and 3D mapping in materials analysis and environmental studies. It is expected that we will provide new, unique quantified time resolved data on the long range transport of fine dust from major Australia desert regions across the east coast of Australia. We will develop a software package for quick viewing of energy spectra that allows quick elemental mapping for both PIXE and RBS as well as other spectroscopic techniques.

Year 1:

- Develop the 3 DRUM strip sampler for operation in the field using Teflon strips
- Test the 3 DRUM sampler under field conditions for 6 months
- Develop and automate the laser black carbon analysis system for the Teflon strips for each of 3 size fraction below 2.5μm
- Develop the simultaneous PIXE, PIGE, ERDA and RBS analysis systems for use on Teflon strips with better than 24 hr resolution.
- Establish calibration methods for IBA techniques and verify them using standard reference materials
- Develop a first version of data viewer that allows mapping of PIXE, RBS and IBIC data from microprobes.
- Design confocal microprobe and provide first test of the principle of 3D mapping using a confocal X-ray lens.
- Sample for 12 months long range dust transport across Australian east coast using the 3 size fraction DRUM sampler
- Design and select the end-station components
- Report results

Results to Date

We only commenced this Project in January 2010 so have very little to report in relation to accumulation of actual data acquired. However some progress has been made to date, including;

- Development of an ANSTO specific 3 stage PM2.5 DRUM sampling unit which is now operational in the field.
- Commenced tests on new Teflon strips to replace polycarbonate and Mylar strips used previously by other groups in USA. This will enable total hydrogen measurements to be made on each strip and hence total organic content of filters to be estimated.
- Established sampling sites in and around desert regions in Australia
- Established a sampling site in the Australian Barrier Reef on Heron Island to monitor dust from Australian deserts
- Identified current PM2.5 sampling sites which cover 2000 km length of the Australia east coast which can be used to tract dust storms from central Australia.

- Commenced work on appropriate software to analyse DRUM strip filters using IBA techniques.

Work Plan for Subsequent Years

In the second and third years we plan to:-

Year 2:

- Use accelerator based IBA techniques to characterize 3 size fraction DRUM sampling over 12 month period
- Apply for access to the Australian Synchrotron micro beam XRF facility to perform elemental mapping of DRUM strips
- Use the Australia Synchrotron submicron beam facilities to characterize and analysis submicron particles on as many strip filters as possible collected in the previous year.
- Assess and analyse the IBA and Synchrotron data collected to date compare with IBA data on same filters
- Extend data analysis tool to the use with synchrotron data.
- Design and construct 3 dimensional sample stage for use on confocal microprobe and design software for control and data acquisition.
- Design the data acquisition system
- Select and integrate the software control
- Report results

Year 3:

- Apply positive matrix factorization techniques to IBA and Synchrotron result to determine pollution source fingerprints and their contributions.
- Apply HYSPLIT back trajectory methods to follow major transboundary dust events across the Australian east coast
- Publicise new confocal microprobe and ERDA systems and nuclear analytical capabilities around Australian University Departments
- Obtain support from Environmental EPA's, industry and mining groups for placement of new DRUM samplers and use of IBA techniques.
- Publish data in high impact journals
- Report results

Acknowledgements

We would like to acknowledge the considerable and valuable assistance of David Garton in the development of the DRUM sampler, Ed Stelcer, Armand Atanacio with IBA analyses on STAR, Rainer Siegele for significant software and IT assistance and all the technical crew running the accelerators making this project possible.