



AUSTRALIS: AMS for Ultra Sensitive TRAce eLement and Isotopic Studies.

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ABSTRACT

The AMS at the CSIRO HIAF laboratory is being upgraded to enable in-situ measurements of ultra-traces and isotopic-ratios in mineralogical applications. The upgraded system will include a microbeam Cs ion source which is designed to produce better than 50 micrometre diameter Cs beam to enable analyses of monomineralic grains. The Cs primary beam will be mass analysed in order to minimize contamination of the sample. The detection system will be upgraded to enable analyses of elements up to U, at 2 MV terminal voltage for charge states 4 and 5. The system will be known as AUSTRALIS: A.M.S. for Ultra Sensitive TRAce eLement and Isotopic Studies. An overview of the system and the anticipated applications in minerals exploration and mining research are presented.

1. INTRODUCTION

Mass spectrometry in its many forms represents an important cornerstone of the geosciences by producing chemical and isotopic data that provide insight into geological and mineralogical structure and processes. One of the fundamental limitations of any system, particularly for isotopic analysis, is the presence of mass interferences due mainly to oxides, hydrides and a few other complexes. For conventional thermionic sources, chemical separations are usually the prerequisite in order to circumvent or reduce the problem.

Probe techniques (i.e. in situ microanalysis, as opposed to bulk analysis) in mass spectrometry, such as SIMS (Secondary Ion Mass Spectrometry) or ion microprobes, and more recently laser-ablation microprobes, enable analysis of microscopic samples or features, which add another dimension to the information obtained. With these probes, data can be obtained from monomineralic grains, inclusions or microstructures (e.g. zoning) in minerals which are relics of physico-chemical processes of their formation and evolution. In addition these methods obviate the usually laborious mineral separation and chemical extraction. However the problem of interferences has to be addressed.

Very high mass resolution, usually at the expense of efficiency, can be used to solve this problem. Increased efficiency can be obtained by invoking higher order corrections to the beam transport system, enabling large acceptance angle for the mass spectrometers. In ion microprobes, this approach is particularly successful when applied to cases where additional information is available to correct for molecular and isobaric interferences, e.g. for U/Pb and S isotopes. Such information however is not always available for a number of potentially useful chronographs. SIMS has also been applied successfully to determination of trace elements, particularly light elements, many of which are of geochemical interest. For heavy elements, REE represent one such example but their determination is hampered by molecular interference problems.

AMS presents significant progress in the resolution of this problem. Most interference is eliminated by virtue of complete destruction of molecular ions through acceleration to high energies and charge exchange collisions, and isobaric discrimination is achieved through particle identification techniques available at MeV energies. The use of negative ions from the source, and availability of variety of molecular ions themselves can also be exploited to reduce or eliminate interference.

2. AUSTRALIS

The AUSTRALIS system being developed at CSIRO is an AMS system with a microprobing source, and thus fulfils the requirements for in-situ microanalysis at high efficiencies and sensitivities. This

system represents a continuing development of the existing conventional AMS system, based on the 3 MV Tandetron in the HIAF laboratory at North Ryde. Figure 1 shows the schematic of the laboratory, showing the existing system and the AUSTRALIS system. The modified HICONEX 834 source in the present system (used for C14 and Be10 measurements), will be replaced by a new source, and a new high energy analysing system will be constructed.

2.1. The Ion Source

The new source features a microbeam of Cs, which is mass analysed in order to minimise contamination of the sample, essential for example when Rb/Sr, K/Ar chronology are contemplated. It is envisaged that the Cs beam will be of the order of 30-50 microns in diameter, with intensities of the order of 1 micro ampere for an acceptable yield of the secondary ions. In this respect the source will be similar to that being developed at North Texas University [1].

To facilitate the production of the microbeam, the magnetic analyzer for the Cs system should have low aberrations coefficients, and for high intensities should have as large an acceptance angle as possible. The Cs beam will have no more than 10-15 keV energy when analyzed, and thus a magnet with beam product of 2 MeV.amu. is sufficient.

The image of the analyzing magnet for the Cs beam serves as the object of a microbeam lens system that will focus it onto the sample. The extraction geometry for the secondary ions is critical since it is intertwined with the primary beam lens system. The first order effect is the deflection of the primary beam by the secondary ions extraction fields because of the off-normal (45°) entry, compounded with higher order effects that will defocus the primary beam.

The secondary ions are extracted at normal angle and first focussed at some distance from the sample using an einzel lens, to form the object for a spherical ESA. This can be used as a simple double focusing analyser, as well as an energy filter to minimise injection of molecular ions. The image of the ESA in turn is the object for a 90 degree analyzing magnet. This magnet is already emplaced as part of the present AMS system. The various isotopes will be selected for injection into the accelerator by modulating the ion energy to give the same magnetic rigidity, i.e. the standard "bouncing" method.

Other features of the source include a sample viewing system with high ($> \times 100$) magnification, a microstage for sample manipulation with precision of 1 micron. In addition, a provision is made for an electron flood gun to prevent charge build up on the sample, and for a second Cs gun to improve the negative secondary ion yield from insulating samples, e.g. silicates

2.2. Analysing system

The high energy side features a high energy mass spectrometer system suitable for analysis of ions up to the actinides at 2.5 MV terminal voltage.

At the accelerator exit, the beam is focussed by an electrostatic quadrupole doublet to form the object for the analyzing magnet. In the present setup, the focus is at 1.2 m from the doublet. The present 90 degree analyzing magnet has a beam product of only 16, radius 50 cm and 2.5cm gap, and is set to give unity magnification. For the new system, a 90 degree magnet with beam product ~ 120 will be acquired, to enable analysis of mass 240 with 17.5 MeV and 6+ charge state, which is expected to be one of the more abundant charge states. In the new system, in order to maintain a reasonable gap size, the high energy quadrupole will have to be focussed further downstream, but physical constraints imposed by the present analysing magnet limits this point to at most 2.2 m from the quadrupole. According to the beam optical calculation, if the quadrupole is focussed in the x (in plane) direction as required for maximum resolving power, the beam will diverge to as much as 50 mm in the x direction but only ~ 6 mm in the vertical (y axis) at the magnet and thus a 25 mm gap may be sufficient. It is assumed that the beam can be focussed to ~ 1 -2 mm wide at the object point of the magnet.

To detect the various isotopes, it is planned to use a technique whereby the magnet setting is kept constant, and the different isotopes are deflected into the magnet using an electrostatic deflector, such that the exit orbit is on axis. This will require special shaping of the entry pole face to maintain low aberrations. This technique requires the magnet to have a quasi-broad range transmission (± 10 a.m.u. for a central trajectory of mass 240).

The beam is further deflected electrostatically after magnetic analysis. Here the ESA acts only as a deflector rather than a focussing element, since the required distances are impractical otherwise. A quadrupole is used to focus the beam into the detector system. This ESA serves to clean up the background further. The detector system will be the usual proportional counter system, or a non-dispersive ion counting system, depending on the beam intensity.

3. Applications

The AUSTRALIS system will be applied to minerals exploration and mining research, through measurements of ultra traces (concentrations in the sub parts-per-billion range), various stable isotopes and potential chronographs on microscopic samples.

A number of applications will be developed as an extension of current studies of trace element geochemistry by means of the proton microprobe. Whereas the proton microprobe typically has detection limits in the 1-100 ppm, AMS has been shown [3] to be capable of detecting trace elements (demonstrated mainly for the platinum group elements, Au) at sub-ppb level. Such sensitivity is warranted in exploration problems, where it is for example desirable to understand the distribution of trace elements in coexisting phases in the deposit, both for ore genesis studies and deposit delineation in mining operations.

In most geological samples, the micro-PIXE detection limits for trace elements lighter than Fe are generally poor, because of the ubiquitous presence of Fe and the limitations imposed by the use of energy dispersive spectrometers (Si(Li) detectors). Many light elements are important geochemical markers, and in-situ detection of these at sub-ppm levels will become possible. Similarly, for heavy elements there is a gap in sensitivity for the REE in micro-PIXE, and the new system will be able to remedy this situation.

In isotopic studies, AUSTRALIS will enable applications of a few chronographs or tracers previously either tedious and limited to bulk applications, or completely inaccessible. Potentially useful applications include the common Pb, U/Pb (Pb^{207}/Pb^{206}), Rb⁸⁷/Sr⁸⁷, Re¹⁸⁷/Os¹⁸⁷ systems. The particular advantage of the AMS principle can be exploited: in the Rb/Sr case, the interfering isobar (the parent Rb⁸⁷) can be suppressed by selecting the hydride beam from the source. In the Re/Os system, the negative elemental ions of Re is highly suppressed [4].

To balance these optimistic expectations, it should be noted that AMS has not been applied to determination of isotopic ratios to very good precisions (permil range) on a routine basis except for C isotopes measurements. Fractionation effects due to the source, the accelerator and beam transport system, and physical processes involved (e.g. Coulomb explosion, charge changing multiple collisions) can affect and perhaps obscure the effects sought. For this reason some system, e.g. the Sm¹⁴⁷/Nd¹⁴³, where the required precision is in the 0.05 permil range, is unlikely to be feasible. In contrast, fractionation of Re/Os during partial meltings in the mantle, resulting in high ratios in the crust makes it an attractive candidate for a chronometer or tracer. The development of AUSTRALIS presents an exciting challenge to expand the capabilities of AMS.

References.

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Fig. 1. A schematic diagram showing the plan view of the HIAF laboratory. The present conventional AMS system is based on a modified HICONEX 834 source, and the detector system is installed in the 22.5° left beam line after the switching magnet. The "AUSTRALIS" system being developed is shown enclosed in the dashed line. The microprobing ion source will replace the existing HICONEX source. The analysing system will bypass the present high energy analysing magnet.

