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LLNL/UC AMS Facility and Research Program

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Abstract

The Lawrence Livermore National Laboratory (LLNL) and the University of California (UC) now have in operation a large AMS spectrometer built as part of a new multiuser laboratory centered on an FN tandem. AMS measurements are expected to use half of the beam time of the accelerator. LLNL use of AMS is in research on consequences of energy usage. Examples include global warming, geophysical site characterization, radiation biology and dosimetry, and study of mutagenic and carcinogenic processes. UC research activities are in clinical applications, archaeology and anthropology, oceanography, and geophysical and geochemical research. Access is also possible for researchers outside the UC system. The technological focus of the laboratory is on achieving high rates of sample throughput, unattended operation, and advances in sample preparation methods. Because of the expected growth in the research programs and the other obligations of the present accelerator, we are designing a follow-on dedicated facility for only AMS and microprobe analysis that will contain at least two accelerators with multiple spectrometers.

Introduction

The Physics Department of the Lawrence Livermore National Laboratory has completed the initial year of trial runs and research operation of the accelerator mass spectrometry beamline at its Multiuser Tandem Laboratory.[1] The beamline has been funded by and used by Livermore's Physics Department, Nuclear Chemistry Division, Biomedical Sciences Division, Environmental Sciences Division and the Laboratory's Internal Research and Development Program. Partial support was also provided by the Regents of the University of California. UC users in the first year have included staff from the Riverside, San Diego and Berkeley campuses and from the Lawrence Berkeley Laboratory and the Scripps Institute of Oceanography. Collaborators from outside the UC system who have used the facility include staff from Simon Fraser University, the University of Texas, Texas A&M University, San Jose State University, Angelo State University, Woods Hole Oceanographic Institute, and the U.S. Geological Service.

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LLNL built its new tandem laboratory as a versatile facility primarily for ion beam analytical methods. Development of new analytical methods and cost reduction in present applications are primary goals of the facility, as is assistance to users in developing their own sample preparation capabilities as necessary.

The tandem laboratory and AMS spectrometer are operated by the Center for Accelerator Mass Spectrometry of the LLNL Physics Department. Coordination of the LLNL research program with other users of the tandem and access to the facility by UC and non-UC users is under the auspices of the Center.

Research Facility

Floor plan of the LLNL Multiuser Tandem Laboratory is shown in Figure 1. The accelerator is the FN tandem formerly used as an injector at the University of Washington double FN tandem laboratory. Upgrades to the tandem including Dowlish titanium electrode tubes, an NEC pellet charging system, and SF_6 insulating gas have been completed on the accelerator. Control of all accelerator power supplies and measurement of all parameters is through a distributed computer control system.[2] The control, archive, and resetting of all parameters is to 16 bit precision, with adjustable error checking for each parameter.

Three ion source positions are available in the present configuration for AMS experiments. All source positions were designed for sequential injection of isotopes. A Danfysik double focussing, double 90° injection magnet with a floating vacuum tank for fast isotope switching can accommodate two sources. On one position of this magnet, a prototype 60-sample Cs sputtering source purchased from General Ionex has been installed. Performance and modification of this source are discussed in a companion paper at this conference.[3] The second 90° source position will be occupied by a developmental thermal emission source fielded for iodine AMS measurements by PNL collaborators.

The zero degree source position is reserved for a developmental source for tritium and carbon AMS. Sequential injection and selection of elements of interest will be accomplished by a switchable Wien filter in the injection optics.

The primary AMS spectrometer consists of two $ME/Z^2 = 150$ 90° magnets. Apertures of the magnet tanks are 5 cm vertical by 15 cm horizontal. Apertures of all other optical elements are 10 cm diameter. A Danfysik Wien filter is installed between the second magnetic quadrupole triplet and the detector, at present a multianode ionization chamber. Time-of-flight analysis will be added to the spectrometer this year, along with a second Wien filter at the high energy end of the tandem. As originally designed, the spectrometer was unconstrained by other applications of the tandem and optimized for isotopes between ^{10}Be and ^{45}Ca. A pumped stripper canal has been added to the tandem and, with magnet power supply upgrades, will be used for ^{129}I operation. Operation to date has been confined to developing ^{10}Be tunes and extensive ^{14}C research operation. In the latter mode, precision and accuracy of 1% have been attained for Modern materials. The spectrometer and its performance are described in more detail in a companion paper.[4]

The second switching magnet (converted from a TCC CN-15 cyclotron will be installed in the laboratory this year. One side of the magnet has been configured for tritium AMS measurements, allowing tritium, deuterium and hydrogen beams to exit into separate beamlines. A small Wien filter and counter telescope system will be installed on the tritium line, Faraday cups on the other two.

The spectrometer is supported by several sample preparation lines. Two lines using the standard graphitization techniques of Vogel[5,6] are in operation on the LLNL site, one for natural abundance materials and one for ^{14}C-labeled materials. Also onsite at LLNL are Be and Cl preparation laboratories. Graphitization lines are in use at collaborator facilities at Scripps and UC Riverside, and a calcium line is at Riverside as well.

The physical facility housing the accelerator is an unshielded building. Radiation protection is provided by a computer monitored array of 16 photon and neutron detectors that imposes a set of exclusion rules. A 100 m^2 addition to the facility will be built this year to provide additional space for control and data acquisition electronics and to allow more options for radiation protection.

Research Program

Thrusts of the program with the new spectrometer are both scientific and technological. Established research applications of the LLNL divisions include the use of ^{10}Be, ^{14}C, and ^{36}Cl isotopes in geophysics and geochemistry, as both tracers and chromometers. Study of atmospheric mixing processes using ^{10}Be and of hydrology of arid regions using ^{36}Cl, especially near the proposed Yucca Flats repository, will begin in the next year. Measurement of dissolved organic carbon in ocean waters, of primary photosynthetic production in coastal upwelling, and of the rate of production of humus have already been performed using ^{14}C measurements. UC collaborators have already used ^{14}C dates from the spectrometer to date human artifacts and erosional processes. The use of the spectrometer for environmental investigations is expected to expand rapidly as the result of a major UC program being developed to study processes related to global warming.

The high sensitivity of AMS for detection of specific isotopes has stimulated a broad LLNL program to develop new diagnostic tools for use in biomedical, clinical and environmental sciences. We have used ^{14}C tags to measure damage to DNA caused by mutagens at lower doses than previously possible.[7] These measurements will expand into a broad program of measurement of mutagenic and carcinogenic effects at low exposure levels.[8,9] With UCSF collaborators,[8,9] we will perform experiments with ^{41}Ca and ^{45}Ca to develop tools for study of body calcium inventory and loss processes, and kidney function in pediatric patients. Human subjects experiments with the calcium isotopes will begin in mid-1990. Sample preparation for a ^{36}Cl measurement series to infer Hiroshima thermal neutron fluences is in process; preliminary measurements have been performed at the Rochester AMS facility.

The scientific research program is supported by technological work both to reduce the cost of analyses and to develop new capabilities. Computer control has been implemented as part of an effort to develop automated operation, hence lower

cost for the accelerator-related portion of the measurement. The spectrometer has run without adjustment for as long as eight hours, readjustment being required as a result of thermal cycling of the source during the changeout of the 60 sample cassette in the source. In the eight hour cycle, we have measured 60 samples (typically 45 unknowns, 15 calibration standards and backgrounds) to an accuracy equivalent to 1% for ^{14}C in Modern materials. Throughput of 120-150 unknowns per day appears possible, though improvements in system stability are required. Accuracy of 0.2-0.3% appears possible with refinements of the present equipment. At present, the sample throughput of our spectrometer is well beyond our ability to prepare graphite samples with the best conventional method.[5,6] We are exploring more rapid catalytic techniques to prepare samples for the somewhat less demanding measurement regimes of biomedical applications. Requirements for such preparation are outlined in a companion paper.[9]

We will explore the difficulties of performing accurate tritium AMS measurements in the coming year. While conventional wisdom holds that scintillation counting is more attractive than AMS for short-lived isotopes, the combined advantages of low dose and speed of measurement with AMS may offer advantages in biomedical and clinical use. Difficulties are expected in controlling hydrogen loss and isotope fractionation in sample preparation and ion source operation; accelerator and spectrometer requirements appear modest in comparison.

Follow-on Projects

AMS usage is expected to rise to 50% of the accelerator time at our new facility. We project growth in usage well beyond that limit in biomedical and geophysical research, particularly if cost reductions in analysis can be achieved. To meet that need, we plan to propose a dedicated facility for AMS and ion microprobe analysis to be built as a regional facility. Such a dedicated multidisciplinary facility would be sited in a UC Institutes corridor outside the LLNL fence. Research capabilities of such a facility would be in the application of ion beam imaging and elemental analysis techniques and AMS isotopic analysis to a broad range of geophysical and biomedical research problems.

The present facility has served to prototype many systems required for the next. The safety of operating a large tandem in an unshielded structure and the computer control systems required for cost-effective operation have been demonstrated. LLNL has two EN tandems and the necessary magnets in hand to build the next facility on an 18 month schedule. One tandem would be devoted to ion microprobe work and ^{14}C-^{45}Ca AMS measurements in both high throughput and high accuracy modes. The other tandem would be optimized for heavy ion AMS and developmental work. As the Wisconsin EN tandem with SF_6 and pellet chains has run to above 8 MV without tubes,[10] being limited by chain motor power at that point, we feel that operation of these machines at 8-9 MV might be possible. As the present research programs at LLNL and within UC develop, we will determine when to propose such a project.

A separate follow-on would involve developing an integrated and optimized spectrometer for both tritium and ^{14}C measurments for labeling measurements in the clinical environment. Most of the difficult effort required here is in the physical chemistry of sample preparation, accelerator and ion source issues being secondary in fielding an acceptable system for clinical use. If the initial clinical applications demonstrate adequate promise, we will seek support and appropriate corporate partners for development of a prototype clinical system.

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Figure Caption

Figure 1. Floor plan of the Multiuser Tandem Laboratory showing the main elements of the accelerator mass spectrometry system. Elements shown in halftone will be added in the next year.

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Multi-user Tandem Laboratory