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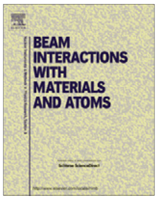

Rafter radiocarbon sample preparation and data flow: Accommodating enhanced throughput and precision

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ABSTRACT

New Zealand's National Isotope Centre, including the Rafter Radiocarbon Laboratory, has upgraded its AMS system from a 6 MV EN-Tandem AMS to a new generation 0.5 MV NEC Extended Compact AMS (XCAMS) system. Current precisions represent approximately an order of magnitude improvement during the AMS era – from 1–2% more than 20 years ago to ~0.2% today. The year since ^{14}C operations were shifted from the EN-Tandem to XCAMS has seen a 40% increase in ^{14}C measurements without reaching full capacity of the new AMS system. Increased throughput and precision require improvements to systems used for the preparation of samples and reporting of results. Our approaches to these challenges are outlined here. They include the development of an automated combustion and CO_2 collection system, and consideration of sources of error or bias including calibration and background correction. Monitoring for any observable changes in reported values associated with the transfer from the EN-Tandem to XCAMS has also been a high priority, and no issues with the continuity of AMS results have been identified.

1. Introduction

The Rafter Radiocarbon Laboratory has relied heavily on AMS for over 20 years, and has now transferred its operation to a new generation 0.5 MV XCAMS system from NEC (see Fig. 1 for brief description). The previous generation 6 MV EN-Tandem AMS was heavily modified over time [1] to greatly improve routine precision, but the new system provides routine ~0.2% (2‰) precision, typically exceeding the best performance ever delivered from the EN-Tandem. Given that 1–2% precision was common early in the AMS era when many lab procedures and calculations were developed and standardized [2], current precisions represent approximately an order of magnitude improvement during the AMS era. The transition to the new XCAMS therefore represents an important opportunity to update procedures and calculations. We outline our progress and philosophy on this topic, with a focus on re-evaluating assumptions and identifying inconsistencies. Changes to operations are also required to efficiently accommodate increased throughput– the year following the transfer of ^{14}C operations from the EN-Tandem to XCAMS has seen a 40% increase in ^{14}C measurements without reaching full capacity. To accommodate increased numbers of samples and standards, we found the development of an automated combustion and CO_2 collection system to be of value.

2. Accelerator mass spectrometry systems

In its final state, the 6 MV EN-Tandem AMS incorporated a range of improvements [1]. These included a new detector beamline with a Wien velocity filter to reduce interferences and background, replacement of the belt-driven charging system with a Pelletron system to improve voltage stability, and a fast beam bouncer on the injector magnet combined with offset Faraday cups for online measurement of $^{13}C/^{12}C$. Despite meeting performance targets for the upgrades, the aging components of the EN-Tandem itself and the highly customized electronic infrastructure were prone to unexpected downtime. A replacement Compact AMS system, incorporating an extended beamline enabling ^{10}Be and ^{26}Al measurement was ordered from NEC in November 2008. Apart from the extended beamline, the new XCAMS system is nearly identical to the NEC CAMS-500 systems for ^{14}C measurement [3–5], with up-to-date performance improvements. It includes

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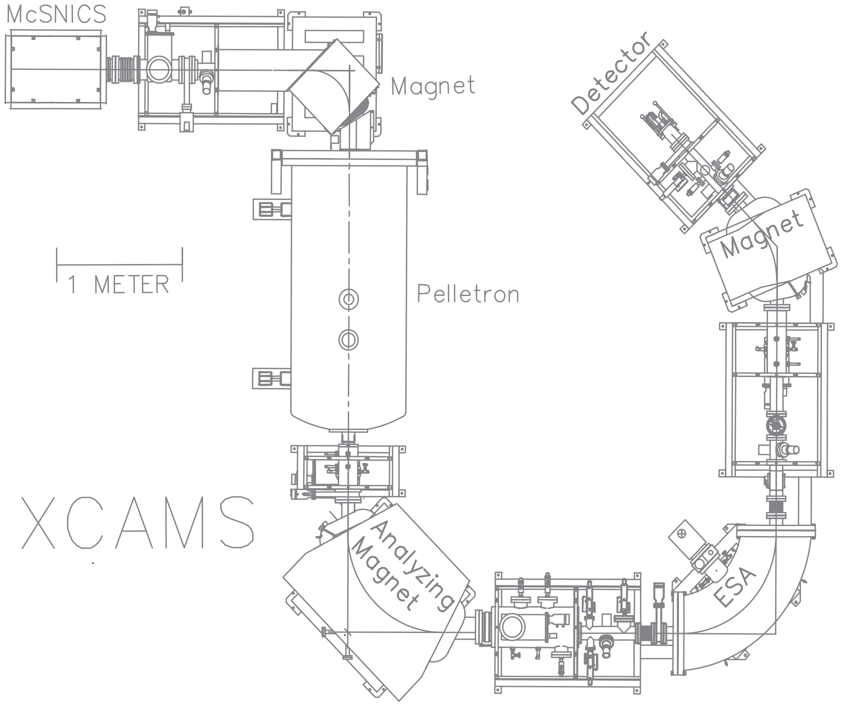

Fig. 1. The Extended Compact Accelerator Mass Spectrometry (XCAMS) system. Briefly, the system is a 0.5 MV 1.5SDH-1 manufactured by the National Electrostatics Corporation (NEC; Madison, Wisconsin, USA). From the MC-SNICS ion source, through the Pelletron, Analyzing Magnet and Electrostatic Analyser (ESA), the system is effectively identical to other Compact AMS systems from NEC (see text). The extension, including the final magnet enables a detection limit for ^{10}Be similar to higher acceleration AMS systems [6].

the MC-SNICS Cs-sputter source with spherical ionizer, and is based on the 1.5SDH-1 Pelletron system (Fig. 1).

The XCAMS system was assembled and tested at NEC's facility in Madison (Wisconsin, USA), including optimization for ^{10}Be measurement [6]. It was subsequently disassembled and shipped in December 2009. The system was unpacked and reassembled at GNS Science in Lower Hutt, New Zealand, beginning 26 January 2010. GNS Science accepted the system based on ^{14}C, ^{10}Be and ^{26}Al performance on 5 March 2010. Design, acceptance and further performance optimisation for the XCAMS system will be reported more thoroughly elsewhere, with a focus on ^{10}Be and ^{26}Al. Briefly, for ^{14}C acceptance testing, XCAMS achieved 0.54 μA $^{13}C^{+}$ beam current, a machine background of 3.4×10^{-16} ($\sim$66 ky), $^{14}C/^{13}C$ and $^{13}C/^{12}C$ standard deviations of 0.23% and 0.11%, respectively ($n = 5$), on modern graphite samples.

Below, we report specifically on results obtained mainly during a phase from 5 March to 30 June 2010 when graphites of adequate size, prepared from secondary standards and unknowns in our laboratory, were split and measured on both the EN-Tandem and XCAMS. The similarity of both systems, including 40-position Cs sputter ion sources and $^{13}C/^{12}C$ measurement capability, meant that no major changes to data processing were required. Calibration to the primary standard continued using our custom-built CALAMS software, which accepts results obtained from both AMS systems. Briefly, CALAMS contains a graphical user interface allowing outliers to be easily excluded. The software uses chi-squared test statistics to account for the variance among cathodes of the primary standard, and of all runs across all cathodes. If variance between runs within cathodes or primary standard variance is greater than expected based on counting statistics, the certainty interval reported for each cathode is expanded accordingly. Following import of data to the laboratory database (Microsoft Access) background corrections were made based on long-term means for each sample type. We are aware that this background correction procedure imposes a limit on our reporting of absolute performance (e.g. secondary standards), but we decided not to alter reporting procedures for the period represented by the results reported below because intercomparison of systems was our main objective.

3. Automated combustion and CO_2 collection system

The Rafter Radiocarbon Laboratory and GNS Science's Stable Isotope Laboratory have collaborated on the implementation of a system for the routine collection of CO_2 from radiocarbon samples (Fig. 2). Organic samples, typically between 0.5 and 1.0 mg C, are weighed into tin capsules and combusted using a Europa ANCA elemental analyzer (EA) interfaced to a Europa 20–20 isotope ratio mass spectrometer (Sercon Ltd., Cheshire, UK). The reactor is held at 1020 °C and receives an O_2 pulse following each sample drop. The reactor is packed with chromium oxide, copper oxide and silver wool, and subsequently removes nitrogen oxides using copper at 680 °C and removes water using magnesium perchlorate. With the column switch valve set to use the CO_2 collection device, helium carrier gas flows through a split as previously developed for interfacing an EA with an IRMS and cryogenic CO_2 collection for ^{14}C [7]. In our system, one percent of the CO_2 flows through a 40° isothermal capillary GC column (0.32 mm diameter, 30 m length, PLOT active phase) and onto the IRMS. The remaining 99% of the CO_2 is trapped cryogenically and transferred to one of twelve pneumatically operated sample bottles for subsequent graphitization and ^{14}C AMS analysis. The CO_2 collection system is interfaced to the EA-IRMS and is controlled by LabVIEW on a separate computer.

Testing of the apparatus has involved two stages. In the first stage, the $^{14}C/^{12}C$ ratios of 109 standards and background materials were compared to values established by pre-existing lab practice of conventional sealed tube combustion, with the goal of showing that results from the two methods were statistically indistinguishable. Results for ANU Sucrose (IAEA C6) are shown as an example in Fig. 3. In addition, blanks were shown to have average conventional