

PROGRESS AT THE SEOUL NATIONAL UNIVERSITY AMS FACILITY

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ABSTRACT. The accelerator mass spectrometry facility at the Seoul National University (SNU-AMS) was completed in December 1998 and a report was presented at the Vienna AMS conference in September 1999. At the conference, we described the basic components of our accelerator system and reported the results of the performance test. Since then, extensive testing of the accuracy and reproducibility of the system has been carried out, and about 200 unknown samples have been measured so far. We obtained a precision of 4‰ for modern samples, and an accuracy of approximately 40 yr was demonstrated by analyzing samples that were previously dated with a conventional technique and by other AMS laboratories. We present these results here, together with detailed descriptions of our data-taking and analysis procedures.

INTRODUCTION

We have been operating our new accelerator mass spectrometry (AMS) facility during the past year. It has been quite a challenging task to maintain an AMS machine like ours in a country with little available expertise in accelerator handling. In early 1999, we began to analyze samples provided by outside users. During one year's operation, about 200 unknown samples have been measured so far. In the meantime, background and chemical blank studies have been carried out. At present, the oldest age we obtained for the samples treated by our sample preparation is 41 ka. This will be improved in the near future by introducing a new combustion method using an element analyzer. In this contribution, we present general laboratory procedures, such as sample preparations, data taking, age calculation, and calibration. In addition, we discuss important issues related to the accuracy and quality of measurements emerging from the Seoul National University AMS facility (SNU-AMS).

Immediately following the acceptance tests (Kim et al. 2000), an additional performance test was made using QA (quality assurance) samples from Beta Analytic. Table 1 shows the results of this test, which are in very good agreement; 6 out of 7 are within an error of 1σ . Although the machine produced stable and reliable results, there have been many technical problems as shown in Figure 1. Most of them were minor in nature and were easily corrected in a day or two. However, it is worth mentioning two major repair works, which resulted in a long machine downtime.

The first was a failure of the scan mechanism of the 846 ion source. The cause of this problem turned out to be moisture collecting around the Syltherm cooling lines and emerging into the servomotor unit, which caused erratic functioning. It took us considerable time and effort to locate the source of the problem. Several hardware items, such as the CPU of the main computer, the power supply of the source computer, servomotor controllers, and also a servomotor, had to be replaced. It took three months to complete repairs.

The second major repair concerned the Ar gas stripper system. The motor-generator system was out of order. It supplies electric power to the turbo molecular pump, which circulates Ar gas in the stripper canal. The pressure tank had to be opened, and the bearings assembly had to be replaced. This repair required a period of three weeks of pressure tank opening.

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Table 1 Results of SNU-AMS measurements for the samples provided by Beta Analytic

Sample ID	BP (1 σ uncertainty)	
	Beta's QA value	SNU-AMS results
SE1-1	145.2 (0.6) ^a	142.9 (0.6) ^a
SE1-2	41,800 (1300)	42,200 (700)
SE1-3	8970 (50)	9100 (120)
SE1-4	12,810 (50)	12,900 (110)
SE1-5	34,980 (520)	35,000 (330)
SE1-6	1130 (40)	1160 (40)
SE1-7	23,060 (130)	22,900 (600)

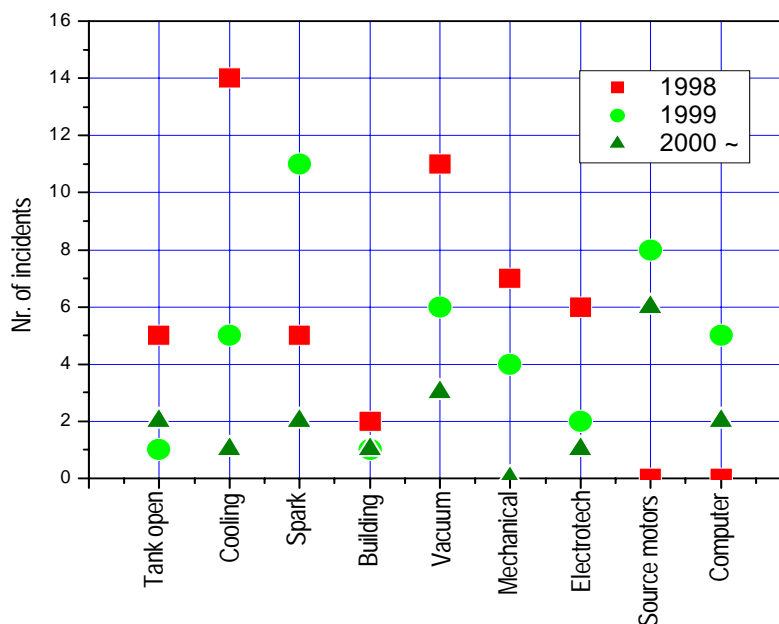
^apMC value

Figure 1 Overview of SNU-AMS technical problems

SAMPLE HANDLING AND MEASUREMENT PROCEDURE

Routine samples such as charcoal, wood, and grain were first examined for contamination: plant roots were removed, and pieces of charcoal were picked by hand or sieved out of the soil matrix. The wood samples were cut into pieces 10–40 microns in size, and the charcoal was crashed into 10–60-micron-sized pieces. Then, after cleaning in an ultrasonic bath to remove adherent particles, the standard AAA (acid-alkali-acid) method (Olsson 1980) was applied as a further pretreatment.

Peat samples were, after physical cleaning, subjected to 1 M of HCl at 80 °C for 24 hr to remove contaminants such as carbonates, fulvic acids, etc. Next, alkali soluble extracts were obtained by applying 0.5% NaOH at room temperature for another 24 hr. These extracts were filtered and precipitated by applying 1 M of HCl at 80 °C for 3 hr. Finally, the solid precipitate was cleaned with cold distilled water and dried in the oven for 8 hr at 120 °C.

Ten to 20 mg of pretreated sample material was placed in a quartz tube containing approximately 1 g CuO. Silver wire was added as a scrubber for sulphur, and the tubes were evacuated by a sorption pump to approximately 2×10^{-3} mb and sealed with a glassblowing torch. For complete combustion of the sample to CO₂, the sealed samples were heated in a muffle furnace at 850 °C for 2 hr. After combustion, the quartz tubes were broken inside a flexible bellows, and the CO₂ was transferred cryogenically to transport vessels of volume 11 cm³. The typical combustion yield is 400 mb CO₂, which corresponds to approximately 3 mg of elemental carbon.

The catalytic reduction method (Vogel et al. 1984) is now a standard method used by many AMS ¹⁴C laboratories. Our system is adopted from the Leibniz Labor, Kiel, Germany (Nadeau et al. 1997). The CO₂ is reduced to carbon by H₂ using Fe as a catalyst at 550 °C in a 3/8" quartz reaction tube. The Fe is preheated and activated by oxidation with 0.5 atm. O₂ at 500 °C for 30 min and subsequently reduced with 0.5 atm. H₂ at 400 °C during 30 min. The sample CO₂ is cryogenically transferred from the transport vessel into the reactor, pre-evacuated to approximately 1×10^{-3} mb by a turbo-molecular pump, and H₂ is introduced into the reactor to the total pressure which is 2.5 times that of CO₂.

The reaction is initiated by placing a preheated oven at 630 °C over the reactor. A cold trap of -20 °C cooled by a Peltier element removes the water vapor from the reaction. The complete reaction takes about 8 hours.

The samples are mixed with Fe powder, and pressed into a 2-mm-diameter cylindrical hole in the Al target holder. Using targets made this way, the 846 Ion source produces approximately 20 μA C, and analyzed ¹³C beams of about 200 nA in the Faraday cup after the 110° magnet. The targets are measured twice at nine different positions around a 1 mm diameter circle for 40 seconds per position. Such measurements are repeated three times per target. The standard deviations are calculated from the statistical results for these three measurements. This measuring time is usually sufficient to obtain ±40 yr 1σ uncertainty for modern samples. Samples prepared from Oxalic-II are used as a standard. We measure one standard per three unknown samples.

The ¹²C and ¹³C currents are measured in Faraday-cups placed just after the 110° magnet in the high-energy beam line of our AMS system. The ¹⁴C³⁺ ions are detected in the ionization chamber, simultaneously with ¹²C and ¹³C taking advantage of the recombinator system built for this new generation Tandemron AMS system (Purser et al. 1988; Gott dang et al. 1995). The conventional radiocarbon age, BP, is obtained from the measured ¹⁴C/¹²C ratios, for samples and standards. Isotopic fractionation is determined from ¹³C/¹²C ratios for samples and standards, using the formulas as given by Stuiver and Polach (1977) and, more recently, by Mook and van der Plicht (1999).

For background correction, the ¹⁴C/¹²C ratio of the chemistry blank is subtracted from the ¹⁴C/¹²C ratio of both the standards and samples. Our chemistry blank are prepared from fossil wood.

RESULTS

Reference Samples

We use Oxalic II as the standard. In addition, we also use IAEA C3 (cellulose) and C5 (wood) reference material (Rozanski et al. 1992) for quality control of our sample preparation chemistry. Their ¹⁴C contents as measured in our laboratory are 129.39 ± 0.76 pMC and 23.81 ± 0.39 pMC, while their recommended values are 129.41 ± 0.06 pMC and 23.05 ± 0.02 pMC. Two additional archaeological samples have assured us of the quality of our sample preparation; for example, a charcoal sample from a Bronze Age archaeological site from the southern part of Korea, Daipyung-ri, was

previously measured by ISOTRACE, the Canadian Center of Accelerator Mass Spectrometry at the University of Toronto, Canada as 2280 ± 60 BP (sample ID TO-6770). The same sample was re-measured in our laboratory just after machine installation, yielding an age of 2360 ± 50 BP (sample ID SNU99-01), showing excellent agreement. Another charcoal sample was submitted to us in the very early stage of our AMS operation. It was again a charcoal sample gathered at a well-known archaeological site discovered recently in suburban Seoul, an ancient military fortress dating from 550 AD. Our measured result for this sample was 1490 ± 50 BP (sample ID SNU99-02), again demonstrating good agreement.

Results on these samples established the reliability of our ^{14}C dating procedure and its capability to perform routine dating. Since that time about 200 samples of archaeological and geological interest have been measured. An exemplary result from our regular batch is shown in Figure 2. In the figure, the instrumental error, i.e., internal error is always less than the external error, which indicates that no significant systematic errors are contributing to the measurement. BP errors of ± 40 are easily obtained with the normal measuring procedure mentioned in the previous section.

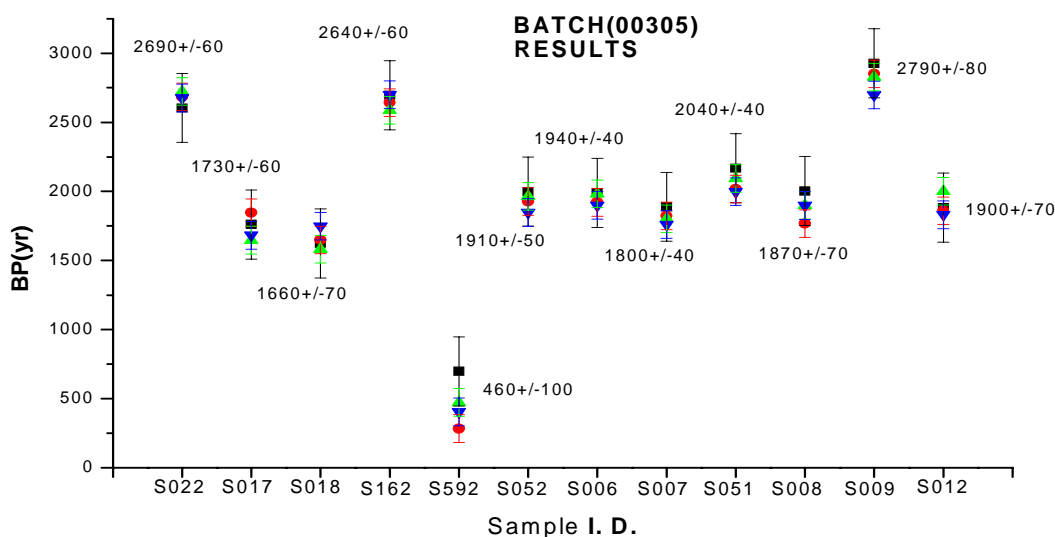


Figure 2 The results from typical batch run. For each sample, of which the ID is denoted in the abscissa, measurements were made three times and the resulting BP values are shown in the ordinate. Standard deviations followed by [+/-] are obtained by procedures described on page 164.

Background

The chemical blank value we obtained thus far by using fossil wood materials is 0.6 pMC, which corresponds to an age of approximately 41 ka. On the other hand, the machine background investigated by using a blank Al target is approximately 70 ka. We also evaluated our standard sample with respect to Oxalic-I prepared by Beta Analytic, and observed that the pMC value of our standard sample, Oxalic-II, prepared by our chemistry is 133.4, which is a little too low compared to the recommended value of 134.06 (Stuiver 1983). To improve this situation and also push the limit of our BP measurement beyond 50 ka, we are planning to adopt the method of *dynamic flash combustion* system utilizing a commercially available Element Analyzer system, as has been successfully employed at other AMS laboratories (Morgenroth et al. 2000; Aerts-Bijma et al. 1997).

CONCLUSION

SNU-AMS is now fully operational, and the basic laboratory procedures are well established. However, improvements are required in the area of sample preparation. Further lowering backgrounds from the present level will probably be achieved by introducing a new combustion system. The throughput will also be increased with this system, together with a multiple reduction line, a 10-fold parallel system, which is under construction at our laboratory.

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REFERENCES

- Aerts-Bijma ATH, Meijer HAJ, van der Plicht J. 1997. AMS sample handling in Groningen. *Nuclear Instruments and Methods in Physics Research B* 123:221–5.
- Gott dang A, Mous DJW, van der Plicht J. 1995. The HVEE ^{14}C system at Groningen. *Radiocarbon* 37(2): 649–56.
- Kim JC, Lee CH, Kim IC, Park JH, Kang J, Cheoun MK, Kim YD, Moon CB. 2000. A new AMS facility in Korea. *Nuclear Instruments and Methods B* 172:13–7.
- Mook WG, van der Plicht J. 1999. Reporting ^{14}C activities and concentration. *Radiocarbon* 41(3): 227–39.
- Morgenroth G, Kerscher H, Kretschmer W, Klein M, Rechel M, Wrzosok I. 2000. Improved sample preparation techniques at the Erlangen AMS-facility. *Nuclear Instruments and Methods in Physics Research B* 172: 416–23.
- Nadeau MJ, Schleicher M, Grootes PM, Erleneuser H, Gott dang A, Mous DJW, Sarnthein JM, Willkomm H. 1997. The Leibniz-Labor AMS facility at the Christian-Albrecht-University, Kiel, Germany. *Nuclear Instruments and Methods in Physics Research B* 123:22–30.
- Olsson IU. 1980. ^{14}C in extractives from wood. *Radiocarbon* 22(2):515–24.
- Purser KH, Smick TH, Litherland AE, Beukens RP, Kieser WE, Kilius LR. 1988. A third generation ^{14}C accelerator mass spectrometry. *Nuclear Instruments and Methods in Physics Research B* 135:284–91.
- Rozanski K, Stichler W, Gonfiantini R, Scott EM, Beukens RP, Kromer B, van der Plicht J. 1990. The IAEA ^{14}C Intercomparison Exercise 1990. *Radiocarbon* 34(3):506–19.
- Stuiver M, Polach HA. 1977. Discussion: reporting of ^{14}C data. *Radiocarbon* 19(3):355–63.
- Stuiver M. 1983. Buisness meeting. *Radiocarbon* 25(2): 793–5.
- Vogel JS, Nelson DE, Southon JR. 1987. ^{14}C background levels in an accelerator mass spectrometry system. *Radiocarbon* 29(3):323–33.