FIRST RADIOCARBON MEASUREMENTS AT BINP AMS

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Abstract

Present status of the BINP accelerator mass spectrometry (AMS) facility is described. The results of experiments for beam selection and radiocarbon concentration analysis in trial samples are presented.

INTRODUCTION

The AMS is mainly dedicated for dating of archaeological, paleontological and geological samples by measurements of the ratio between carbon isotopes.

The BINP AMS facility [1] includes negative ion source, folded type vertical electrostatic tandem accelerator, magnesium vapors stripper [2], the high-energy and low-energy beam lines with analyzers, time-of-flight final detector [3].

The negative ion beam is horizontally extracted from the ion source. Then the beam is vertically injected into the low energy accelerating tube through injection channel with 90° magnet. The negative ions are accelerated to the positively charged high voltage terminal and stripped to charge state 3+ in magnesium vapors stripper. Then they pass through the 180^0 electrostatic bend and then again are accelerated vertically into the high energy accelerating tube to the ground potential. Then ions are horizontally put to the final detector through high-energy channel with 90° magnet.

The most distinguishing feature of our AMS machine is the use of additional electrostatic separator of ion beam, located inside the terminal. Interfering isobaric molecules are destroyed by collisions in the stripper into the terminal and are selected immediately after the stripping process. It is important to decrease the background from molecular fragments before the second stage of acceleration [4], because the energy of fragments is always less then the ion energy (at this moment). The next important distinguishing feature is magnesium vapours stripper instead of the gas stripper. The gas flow into the accelerator tubes leads to big energy spread in the beam thus limiting the sensitivity and accuracy of spectrometer. The molecular destruction and ion recharging by magnesium are localized into the hot tube of the stripper.

BINP AMS FACILITY MODIFICATIONS

Now the AMS facility created at BINP SB RAS is installed at CCU "Geochronology of the Cenozoic era". The accelerator is placed into underground room with radiation shielding. The inner size of the room is 6 x 6 x 7.5 meters. The basic parts of electronic devices are located outside of the shielding room and connected with accelerator elements. The local equipment of the water cooling, compressed air and gas transfer system has been installed.

The 500 kV terminal voltage was achieved with 1 atm atmospheric air into pressure tank (without insulating gas). The equipment for gas filling and drying was not used, but the silica gel was placed directly into the tank. Initially, the terminal voltage was limited by the water vapor condensate on the cool surface of the gas turbine feeding dielectric tube, located along the accelerator column. This tube is used for terminal turbine feeding by compressed air. The electrical conductivity of condensed water distorts the electric fields, which can induce electrical breakdown. For prevention of water condensation, the lower part of the tube (outside of the tank) was heated. Now the electrical breakdowns are occurred only during the first start after tank closing, as we assume, due to the dust accumulation when the tank is open. Recently, 1 MV terminal voltage was achieved by using low cost air-gas mixture. The tank was pumped to the 0.8 atm air pressure, and then the tank pressure was increased to 1.6 atm by four nitrogen gas-cylinder. The 4 kg of SF_6 gas was added (+0.02 atm) to increase the electrical strength of the mixture. The 1 MV has been achieved without breakdowns.

The multi-cathode (for 24 samples) sputter ion source has been recently manufactured and installed. It's needed for synchronous analysis of the samples and for comparison of the tested samples with the reference one. The negative ions are produced by bombarding graphite target with positive cesium ions. The Cs+ ions are produced on a hot tantalum ionizer (1100^0C) by cesium vapor from the oven (180^0C). The cesium ion beam is focused on the carbon sample placed on the cathode, because the working surface of ionizer is a spherical-shape cup. The copper sample holder has the inner diameter of 2 mm. The holder is water cooled to reduce sample heating. The cesium ions leaving the ionizer are accelerated by 7 kV potential. The negative carbon ions are accelerated by the same potential and extracted through the hole 6 mm in diameter in the center of the ionizer. The power consumption of the ion source does not exceed 250 W. The test sample in ion source is selected by sample wheel rotation. The stepping motor with Pi/25600 rad/step resolution is used for sample changing. The process of rotation is controlled by motor driver and checked by optoelectronic sensor system (at every turnover) and by video camera (online).

The new magnesium vapors stripper has been manufactured and installed. All hot parts of striper are located in vacuum. It prevents corrosion of striper surface by the tank gas mixture. The power consumption is about 50 W.

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The electronics for time-of-flight detector (ToF) was improved. At present, the ToF channel width is 70ps. Moreover, the moment of time for ion detection can be registered with 16 μs channel width. This data is used for calculation of number of detected ions per unit time, allowing to filter the background ions from electrical breakdowns.

EXPERIMENTAL RESULTS

During the experiments, the injection energy of carbon beam was 25 keV. The carbon beam current was about 5 uA. The terminal voltage of tandem accelerator was 1 MV. The 180° electrostatic bend was set to transmit the ions with charge state 3+. The magnesium vapors stripper was heated for obtaining the equilibrium charge state distribution, but not more. The ions transmission of AMS system at this energy is about 10% (includes the stripping yield for 3+ charge state). The ^{14}C ions are counted by ToF detector. The ^{12}C ions are measured in shielded Faraday cups with secondary electron suppression. The current of the mass-12 ions can be measured immediately after the magnet of low energy beam lines (^{12}C- ions) and at the AMS exit ($^{12}C^{3+}$ ions). The vacuum level in the beam line was about 10^{-6} Torr.
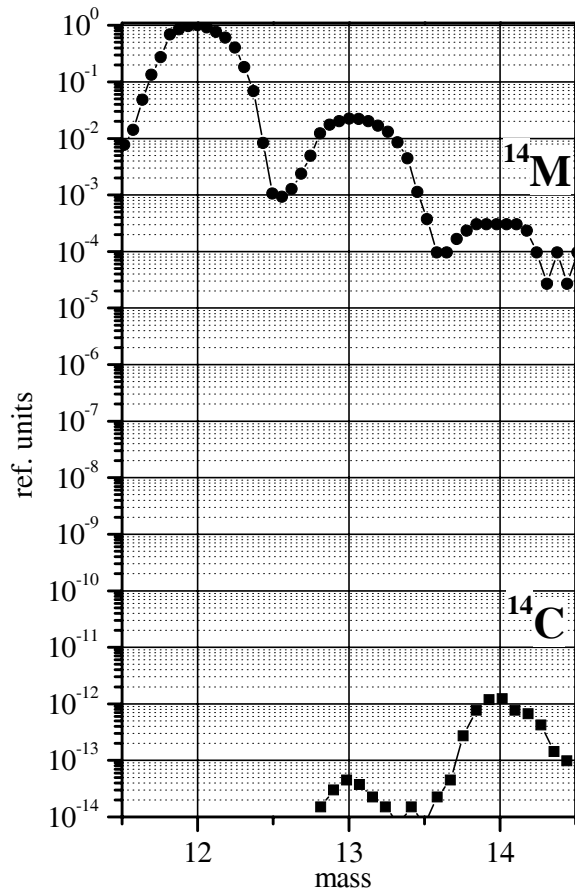

Figure 1: Mass spectrums of the injected (upper curve) and accelerated (lower curve) beams.

The typical mass spectrum of the carbon target before acceleration is shown in Fig. 1 (upper curve). The intensity of the mass-14 peak is more than 10^{-4} per ^{12}C isotope. It is mainly the $^{12}CH_2$ and ^{13}CH molecular currents. The ToF spectrum at the exit of AMS is also shown in Fig. 1 (lower curve). The mass is calculated from ToF channels. The AMS system is tuned for radiocarbon transmission. The molecular background of the mass-14 is suppressed by the destruction process in the magnesium target and then filtered by tandem 180° bend. The small mass-13 peak is also visible in the spectrum, but the mass separation is good enough for radiocarbon measurements.
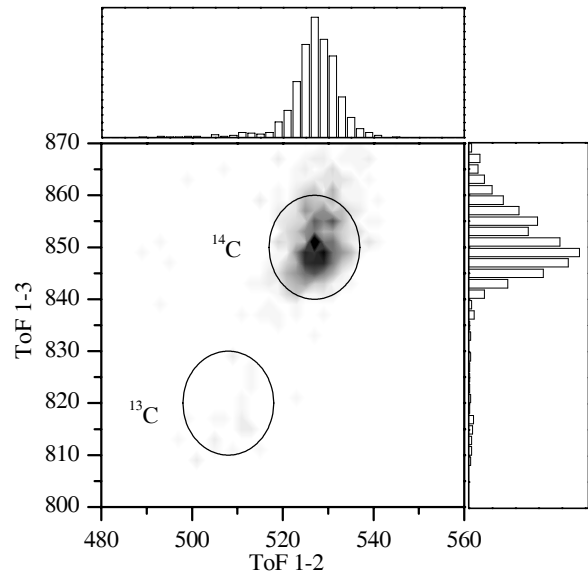

Figure 2: The 2D ToF spectrum at the exit of AMS (70 ps per channel).

The more detailed 2D ToF spectrum with the same AMS tuning is shown in Fig. 2. The solid circles show the locations of the radiocarbon and mass-13 peaks. The particles from mass-14 circle are calculated for radiocarbon concentration determination. The time-of-flight histograms for both ToF distances are also shown in Fig. 2. The peaks separation is bigger about of factor 2 than peak widths (FWHM) for each ToF distance. Such a system of several sequentially positioned detectors on the particles path allows a significantly decrease in the number of random coincidences.

The commercially available carbon fabric is used as test modern sample. The filaments of fabric are pressed into the cathode holder. The carbon fabric is made of organic materials. The radiocarbon isotope ratio of the modern organic matter is about 10^{-12} ($^{14}C/^{12}C$). The graphite MPG is used as test "dead" sample. The radiocarbon concentration in graphite is about 100 times lower than in modern sample.

For radiocarbon concentration analysis, the ^{12}C ions current and ^{14}C ions number are measured for each sample. During the experiments, the $^{12}C^{3+}$ ion current was measured one time of each 400 s interval of radiocarbon

counting. During switching between the isotopes, the magnets settings are changed. The $^{12}C^-$ ions were measured simultaneously with the ^{14}C counting. The process of isotope measuring and sample changing (wheel rotation) is fully automated.
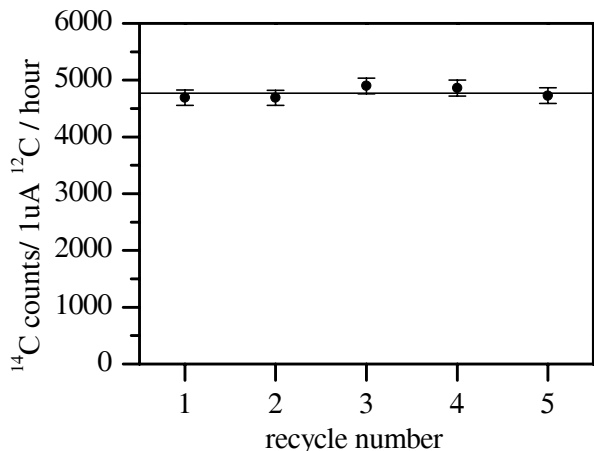

Figure 3: Radiocarbon concentration in modern sample. The sample was measured five times.

For estimation of the reproducibility of measurements, the series of five radiocarbon concentration measurements for one sample is presented at Fig.3. The solid lines show the mean concentration value. The experiment was carried out without rotation of the sample wheel. The ^{14}C counts time is 800 s for each measurement. The statistical uncertainty of radiocarbon registration is about 3% (shown by error bars). It is seen, that the results are in agreement with each other within the error ranges.
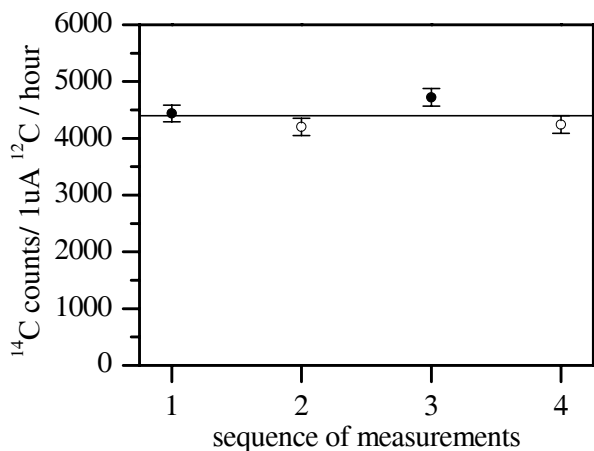

Figure 4: Radiocarbon concentration in two modern samples (measured alternately).

For testing of the reproducibility of measurements after sample wheel turning, another series of measurements is presented in Fig. 4. Here, two modern samples are measured alternately. One can see the results with wheel rotation are similar to ones presented in Fig. 3. The samples were degassed before the measurements by Cs beam. The time of degassing is about 5 min per sample.

The effect is visible by vacuum monitoring. The ion source parameters are not stable during the degas process.

For ion background estimation, the modern sample and "dead" sample are measured alternately. The results are shown in Fig. 5. The data are normalized to the radiocarbon concentration in modern sample. As seen the radiocarbon concentration in "dead" sample is about 1% of the modern sample concentration.
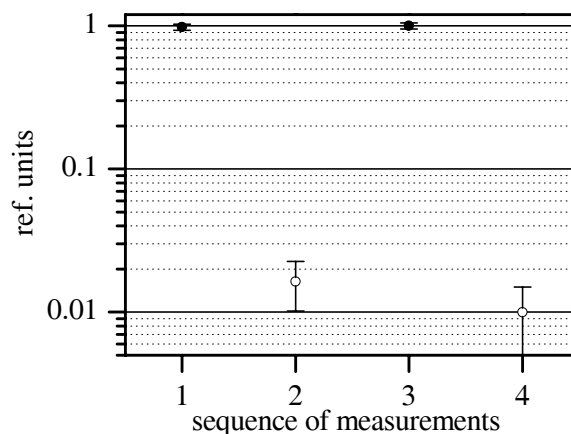

Figure 5: Radiocarbon concentration in the modern and "dead" samples (measured alternately).

For radiocarbon analysis, the samples with large content of carbon /were used. The sample preparation is needed for transformation of natural objects to such samples by combustion and graphitization. We tested more then 100 samples prepared by CCU "Geochronology of the Cenozoic era". The measured background carbon contamination during sample preparation is about 10%. This work will be continued to the reduction of contamination.

The data presented is the first preliminary estimation of BINP AMS facility for radiocarbon dating. The detailed study of the systematic errors and ion background investigation will be done soon.

SUMMARY

The accelerator complex has demonstrated the sustained performance on 1MV running. The reproducibility of first radiocarbon concentration measurements is about 3%. The measured radiocarbon concentration in "dead" sample is about 1% of the modern sample concentration.

ACKNOWLEDGMENTS

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