

Accelerator Mass Spectrometer with Ion Selection in High-Voltage Terminal

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Abstract—The folded electrostatic tandem accelerator with ion selection in a high-voltage terminal is the basis of accelerator mass spectrometry (AMS) at the BINP. Additional features of the BINP AMS are the target based on magnesium vapors as a stripper without vacuum deterioration and a time-of-flight telescope with thin films for reliable ion identification. The acceleration complex demonstrates reliable operation in a mode of 1 MV with 50 Hz counting rate of $^{14}\text{C}^{+3}$ radiocarbon for modern samples ($^{14}\text{C}/^{12}\text{C} \sim 1.2 \times 10^{-12}$). The current state of the AMS has been considered and the experimental results of the radiocarbon concentration measurements in test samples have been presented.

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INTRODUCTION

The AMS is a supersensitive method for the detection and highly accurate quantitative estimation of rare long-lived isotopes. The AMS sensitivity is incomparably higher than any other methods, since by the AMS analysis a direct extraction of atoms from the substance under study and a counting of single rare isotopes is carried out. The high sensitivity of this method makes it possible to analyze the samples of milligram masses.

The AMS technique is widely used in archeology and geology for the radiocarbon dating and composition of the time scales of natural depositions, as well as in other spheres of sciences like biomedicine, ecology, astrophysics, material engineering, criminalistics, etc.

ION SELECTION USING BINP AMS

When a radiocarbon analysis is carried out, about 10^8 molecular isobars composed of hydrogen and stable carbon isotope fall on every radiocarbon ion. The basic task by the creation of the AMS complex is the extraction of the needed isotope from the satellite ion background at a level of 10^{-12} – 10^{-15} ion/ion background. The functional scheme of the accelerator mass spectrometer (AMSr) at the BINP is shown in Fig. 1.

The samples are sputtered by a cesium beam in an ion source [1, 2]. The ions knocked out from the sample are pulled by the electric-field positive potential and, as a result, a beam of negative ions with an energy of 25 keV, which consists of atoms and molecules of the sample under study, is formed at the ion source

output. The ions which are formed pass a system of selection in a dipole magnetic field. The main function of this selection stage is a substantial suppression of the masses differing from the chosen one by 1 amu. After the low-energy selection stage, negative ions with a rare isotope are accelerated to an energy of 1 MeV to the high-voltage terminal of the tandem accelerator. In addition, the beam of isobar ions, which exceeds in its intensity the studied one by many orders of magnitude, and the background ion beam of various masses which overcame the first selection stage are accelerated. When the ion beam is passed through the recharging target of a large enough thickness, an ion beam of all the charges is formed as a result of capture and detachment of electrons independently from the initial ion charge in the beam. In the BINP AMS complex, the target based on magnesium vapors is used as a recharging target [3]. Such a target does not deteriorate vacuum beyond the heated volume. The deterioration in the vacuum conditions, especially in the accelerating tubes, causes an increase in the passed background particles [4, 5]. One of the most important results of the analysis at this stage is the possibility of the collapse of the molecular ions, especially isobar ones, which are close in their mass to the extracted ions, and, as a result, difficult to be separated in the magnetic fields. In contrast to the atomic ions, the maximum charge state achieved by molecular ions is equal to +2; by detachment of an additional electron, the molecules are falling apart. Thus, the initial atomic ions, ions of atomic fragments of the molecules in various charge states, the initial molecules, and molecular fragments of the initial molecules in

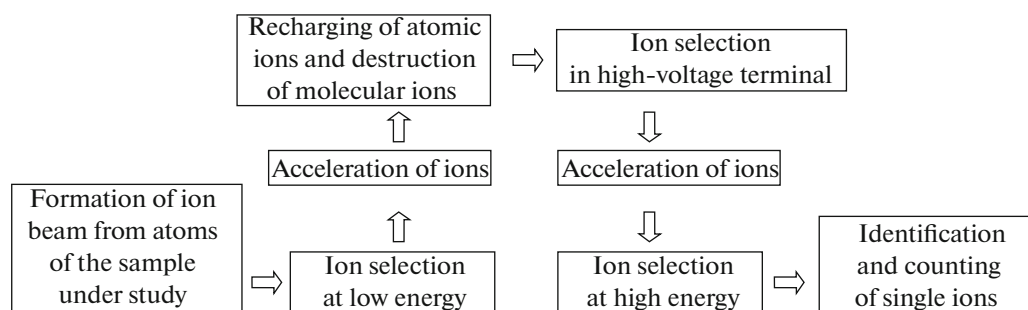


Fig. 1. Functional scheme of the BINP AMSr.

charge states not exceeding $2+$ will be present at the recharging target output. The ions which reach the terminal in a charge state of -1 will have the maximum kinetic energy; part of the ions will recharge into a charge state of 0 by acceleration and will possess lower kinetic energy in the high-voltage terminal region. The kinetic energy of molecular fragments will also be lower than that of the molecules proportionally to the fraction of the total mass. The presence of the ion selection system in the high-voltage terminal region is one of the distinguishing features of the BINP AMS. In a usual system, the whole ion flow transfers into the second acceleration stage after the recharging target. Since the ions at the input to the second accelerating tube will be in different charge states, the accumulated energy will be different for the ions proportionally to their charges. Ion recharging in a residual gas results in the fact that the ions will be in a wide range of the kinetic energies, but not only with those determined by the ion initial charge. At the output of the second accelerating tube, the energy of the molecular ion fragments can be higher, lower, or even equal to the energy of the very molecular ion (while at the input it is always lower), which deteriorates the selection. The electrostatic bending system is located in the high-voltage terminal of the BINP AMSr, which makes it possible to select ions in a charge state of $3+$ and to separate molecular ion fragments. After the second acceleration stage, the ions pass the selection system in

a dipole magnetic field, which serves to separate the background ions differing by 1 amu in their mass that passed the previous selection stage. At the last stage of AMS analysis, ion identification is carried out in the time-of-flight detector, which counts single ions [6, 7]. Not only times of ion flight, but also any single ion arrival moment are registered, which makes it possible to separate a short-time ion background caused by electrostatic breakdowns. Figure 2 shows the mass spectrum of the time-of-flight detector for the carbon filament (the concentration of radiocarbon is at a level of the natural one in living organisms) and for the fine-grain dense graphite MPG.

It can be seen from Fig. 2 that radiocarbon ions are reliably identified by the time-of-flight detector. A procedure of sample preparation is carried out by AMS analysis of natural samples. The samples are purified from impurities and “burned” in vacuum with oxygen admixture; then carbon from carbon dioxide that was formed is catalytically deposited onto iron powder. The iron–carbon powder is pressed into pistons to be placed in the AMSr ion source. The sample preparation procedure makes it possible to obtain the currents of negative carbon ions from any natural samples, which are comparable to those obtained from graphite at the expense of thermal conductivity improvement and carbon concentration increase. However, by the sample preparation procedure, a pollution of the samples with “outside” carbon occurs. Currently, a minimum level of radiocarbon concentration in the prepared samples amounts to 5×10^{-3} compared to that in living organisms, i.e., by about 2.5 times higher than in MPG graphite. Such a level of pollution (radiocarbon concentration) corresponds to a radiocarbon age of about 43 000 years.

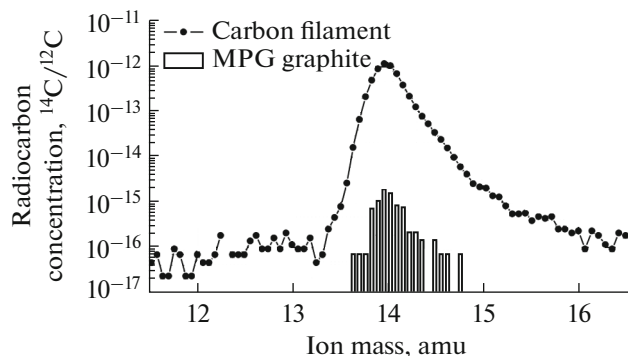


Fig. 2. Mass spectrum at the AMSr output.

ANALYSIS OF USER'S SAMPLES ON BINP AMSr

The sputtering source of BINP AMSr is equipped with a reel for 23 samples. A remote change of samples is implemented by a step motor, which makes it possible to rotate the samples with a discreteness of $1/51$ 200 of the wheel rotation. Commonly, 20 graphitized samples are settled in the ion source reel in order to measure the radiocarbon concentration. Two samples of carbon filament and a sample of MPG graphite

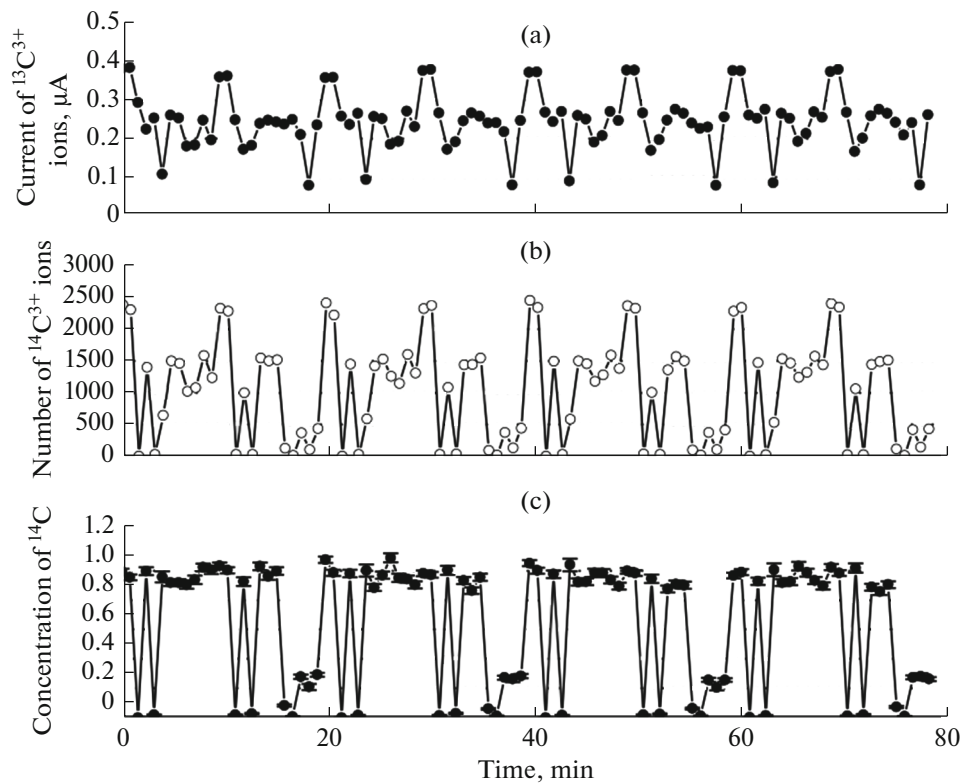


Fig. 3. Dependences of the current of $^{13}\text{C}^{3+}$ ions (a), the number of radiocarbon ions (b), and the radiocarbon concentration (c) on time obtained by the AMS analysis of the samples.

with the known concentration of radiocarbon are placed in the ion source reel in order to control and normalize the process of sample measurements. We note that the control samples do not require the sample preparation procedure and are placed in the ion source reel in their natural form.

When the samples are analyzed by AMS, it is necessary to determine the radiocarbon concentration; therefore, in addition to the reliable extraction and counting of radiocarbon ions, the measurement of the ion current of the stable carbon isotope and its normalization to the ion current from the samples with the known radiocarbon concentration are made. In BINP AMSr, in order for various isotopes to pass the dipole magnet of a low-energy part of AMS, a change in the energy of the ions formed in the ion source is carried out so that the ion impulse would not depend on its mass. A change in the magnetic field is inexpedient because it is a significantly longer process, which, in addition, is characterized by hysteresis effects of the residual field. Simultaneously, matching changes in the focusing and beam correction before the input to the accelerating tube and in the voltage at the electrostatic fold are carried out. The radial aperture of the dipole magnet at the AMSr output makes it possible to transport isotope ions without any rearrangements. A rare isotope is counted by the detector of particles, while the current of a stable isotope is

measured at the Faraday cylinder with a radial bias. Currently, by AMS analyzing carbon samples, 80% of time is devoted to the accumulation of the radiocarbon statistics; 15% is consumed by the switching to a stable isotope and back and by the measurement of the stable isotope current. The rest (5%) of the time is expended on the reel rotation or sample change.

When the samples are degasified and AMSr is adjusted, the measurement of the radiocarbon concentration in the samples is carried out. Once the desired sample is placed under the cesium beam, an alternate measurement of the stable isotope current and counting of the number of ions of a stable isotope are carried out. Four switches between isotopes are made in total. Each time, a rare isotope is collected for 10 s, and twice the current of a stable isotope is measured. For control samples of carbon filaments, such a procedure is repeated twice, while for the rest samples it is made once. After this, the reel is rotated for a new sample and the measurement of the new sample is made. When all the samples in the reel are measured, the measurement of samples on the next reel turn is carried out, etc. Figure 3 shows the dependences of the number of radiocarbon $^{14}\text{C}^{3+}$ ions, current of $^{13}\text{C}^{3+}$ ions, and the radiocarbon concentration on time by AMS sample analysis according to the algorithm that we have described. The presented values of the number of the radiocarbon ions correspond to the summary

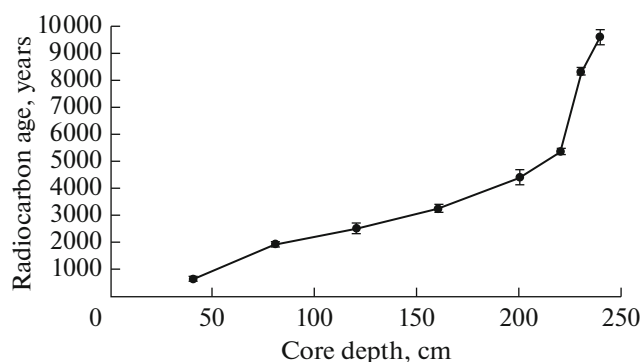


Fig. 4. Radiocarbon ages of bottom sediments, depending on the core depth.

value for four measurements, i.e., to 40 s. Figure 3 corresponds to the measurement of four reel rotations; the duration of a measurement for one rotation amounts to a little less than 20 min. The rate of radiocarbon counting from carbon filaments is about 60 Hz, which corresponds to a value of 2400 in Fig. 3b. By such a counting rate, a statistical accuracy of 1% is achieved for a time of four rotations of the reel with the samples. However, as can be seen from Fig. 3a, the ion current of the prepared samples can be significantly lower than that of carbon filaments; therefore, usually the total time required for an analysis of samples using BINP AMSr amounts to 4 h, which corresponds to 12 rotations of the reel with the samples. A double measurement of the carbon filament is used in order to increase the accumulation of statistics on these reference samples and, respectively, decrease the total statistical error in the AMS analysis, because the normalization is made to the filament samples. Furthermore, the coincidence in the measurement results for carbon filament doublets (within statistical accuracy) proves the absence of transitional processes by measurements because the first measurement is performed directly after placing the sample under a cesium beam, while the second one comes after the first measurement, i.e., about 1 min after placing the sample under the cesium beam. Two samples of carbon filament are placed in the reel with the samples. The coincidence of the measurement results for these samples indicates that the process of the AMS analysis is successful. When the AMS analysis is over, the radiocarbon age of the samples is calculated by normalizing to the average radiocarbon concentration in carbon filaments. As an example, Fig. 4 presents the radiocarbon age for the bottom sediments of Lozhka Lake (Vengerovskii

raion, Novosibirsk oblast, Russia) as a function of the core depth according to the dating results obtained on BINP AMSr using the algorithm described above. For the last year we have carried out the analysis of more than 700 user samples for studies in various spheres of sciences.

CONCLUSIONS

A routine analysis of carbon samples is carried out at BINP AMSr. By the reliable identification of radiocarbon ions, the rate of statistics accumulation for modern samples amounts to about 50 events per second.

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