

Acceleration Mass Spectrometer of the Budker Institute of Nuclear Physics for Biomedical Applications

S. A. Rastigeev^a, A. R. Frolov^a, A. D. Goncharov^a, V. F. Klyuev^a, E. S. Konstantinov^a,
L. A. Kutnyakova^b, V. V. Parkhomchuk^a, and A. V. Petrozhitskii^a

^a*Budker Institute of Nuclear Physics, Siberian Branch, Russian Academy of Sciences, Novosibirsk, 630090 Russia*

^b*Institute of Archeology and Ethnography, Siberian Branch, Russian Academy of Science, Novosibirsk, 630090 Russia*
e-mail: S.A.Rastigeev@inp.nsk.su

Abstract—An accelerator mass spectrometer (AMS) made at the Budker Institute of Nuclear Physics (BINP), Siberian Branch, Russian Academy of Sciences, is installed in the Geochronology of the Cenozoic Era Center for Collective Use for the carbon 14 dating of samples. Distinctive features of the BINP AMS include the use of a middle energy separator of ion beams, magnesium vapor target as a stripping target, and a time-of-flight telescope with thin films for accurate ion selection. Results of experiments measuring the radiocarbon concentration in test samples with radiocarbon labels for biomedical applications are presented.

DOI: 10.1134/S1547477114050240

INTRODUCTION

Accelerator mass spectrometry (AMS) is a super-sensitive technique for the radiocarbon dating of materials. In recent years, AMS centers have become used actively to conduct biomedical research. The principle by which the research is conducted is based on the addition of radiocarbon labels into the biopreparation introduced into living systems with the further recording of such labels in biomedical samples using AMS. The high sensitivity of AMS makes it possible to introduce a minimum of radiocarbon labels to perform the analysis. Thus, the generally accepted radioactivity of radiocarbon in preparations seeking approval for use in humans is 100 nCu, which is much less than the minimal significant activity of a radiation source according to Russian radiation standards. However, even using such a small number of radiocarbon labels, the AMS sensitivity exceeds the sensitivity of liquid chromatography routinely used in biomedical research by factors of hundreds.

Performing an AMS analysis of natural samples, the maximum concentration of radiocarbon is found in the present objects ($^{14}\text{C}/^{12}\text{C} \sim 1.2 \times 10^{-12}$) decreasing by a factor of 2, when the age of the object increases by 5730 years, which corresponds to the half-life of radiocarbon. It should be noted that the concentration of radiocarbon in the objects formed in the 1960s exceeds the present level by a factor of 1.5–2 due to the additional formation of radiocarbon during the testing of nuclear weapons. Carrying out biomedical research using AMS, radiocarbon labels are needed to increase the radiocarbon concentration in a

sample analyzed by a significant value taking the possible natural fluctuation of radiocarbon in living systems into account. Due to the inhomogeneity of the distribution of the biopreparation introduced to a living organism and the necessity of following the changes in time, the radiocarbon concentration in a certain sample could exceed the natural one by several orders of magnitude. Thus, the AMS should provide the possibility of measuring the concentrations of radiocarbon of a higher level when compared to radiocarbon dating. Even when analyzing natural objects, the memory effect is of importance when a small part of radiocarbon from an analysis of a previous sample is revealed during the analysis of the following sample. It is clear that increasing the range of concentrations of radiocarbon enhances this effect. Moreover, in contrast to the purposes of dating objects, where the determination of the age of an object is a complete task, during biomedical research, the analysis of tens and hundreds of samples is required. This is due to the necessity of collecting statistics on different species to follow a change of radiocarbon concentration with time, in different organs of the organism, under different conditions, etc. Thus, it is necessary to provide the possibility to load a greater number of samples into the ion source of an AMS. Moreover, it is desirable to reduce both sample preparation time for AMS analysis and cost of this process. The simplification of sample preparation results in an increase in the number of admixtures in the sample. The presence of nitrogen and hydrogen leads to the appearance of molecular isobars of radiocarbon in the ion beams; thus, the

AMS should reliably separate radiocarbon and accompanying isobars.

The conception of the BINP AMS [1, 2] provides additional (when compared to the analogs outside Russia) possibilities for reliably isolating and detecting radiocarbon. The BINP AMS uses a magnesium vapor charge-exchange target [3], which makes it possible to localize a place both for the ion charge exchange and disintegration of molecular ions in the region of hot surfaces of the target. Thus, growth in the energy spread in the beam that makes it possible for background ions to overcome selection stages in the electromagnetic field is prevented. Moreover, in the BINP AMS fragment, ions are screened with respect to the energy immediately after the breakup of the molecules, which is very effective, since only at this moment the energy of the fragment ions is always less than that of the ions itself [4, 5]. The application of the time-of-flight telescope made in the BINP [6, 7], which makes it possible to reliably detect radiocarbon ions at the final stage of the selection, is also of importance.

ANALYSIS OF THE SAMPLES WITH AN INCREASED CONCENTRATION OF RADIOCARBON USING THE BINP AMS

To test the possibilities of the BINP AMS in regards to analyzing samples with an increased concentration of radiocarbon, a sputter ion source with a wheel rotation for 23 positions was used. Graphitized samples are arranged in a circle, the distance between them being 7 mm and the sample diameter being 2 mm. Using a step motor, the sample wheel could rotate into place a sample chosen under a cesium beam. When sputtering the sample with cesium, negative carbon ions are drawn from the sample by the voltage applied and analyzed in the AMS. However, a significant part of the atoms is sputtered, "sticking" to surrounding surfaces. Taking into account the fact that, analyzing biomedical samples, radiocarbon concentration could differ by many orders of magnitude, getting even an insignificant part of the carbon sputtered from one sample to the region of another sample might lead to a significant distortion of the results. Thus, there is a diaphragm of 6 mm set before the sample wheel at a distance less than a millimeter in the BINP AMS. Such a diaphragm makes it possible to cover remaining samples from carbon, when the sample chosen is analyzed. Moreover, though the diameter of the cesium beam at the sample surface is about 0.5 mm, a small part of cesium knocks ions out also from a larger area. Taking into account the fact that carbon falling onto the diaphragm would be common when different samples are analyzed, its diameter is chosen to be sufficiently large. It should be noted that, if radiocarbon ions of

external origin having an appropriate energy are drawn from the ion source, then it is impossible to filter them out; however, taking the level of the influence of such a process into account is sufficiently easy. To do it, it is necessary to add to a sample wheel of the ion source a sample having a low concentration of radiocarbon and compare it with the value measured. If in AMS the reliable selection and detection of radiocarbon ions are performed, then an increased concentration of radiocarbon in the measurements is evidence of contamination in the ion source (or that of unreliable data on the concentration of radiocarbon in the sample). Before measurements, the sample wheel is slowly rotated under the cesium beam to remove contaminations from the sample surfaces capable of distorting the results of the measurements.

The radiocarbon ions that are knocked out go through all stages of selection in the AMS and, finally, are detected in the time-of-flight telescope at the exit of the AMS. For every ion passing the time-of-flight detector, the time of travel of each of two straight sections and absolute time of getting to the detector are recorded. The projections of a typical three-dimensional picture of such a recording is given in Fig. 1a, where the times of flight of ions of two time-of-flight intervals, TOF12 and TOF13 (one channel of the detecting system is equal to 70 ps) and time of arrival of ions with the resolution of 16 μ s are given. The principles of particle detection using the times-of-flight of two intervals are well-known. However, to increase the reliability of the measurements, recording the time it takes an ion to get into a time-of-flight telescope is of importance. Using high-voltage electrostatic elements, there is always a possibility that a breakdown occurs. Herewith, due to the change in the voltage, a short-term failure of the filtering property of the AMS occurs, resulting in background ions appearing at the exit of the AMS. If the time of arrival of an ion to a time-of-flight interval is recorded, then such a process is clearly seen (Fig. 1b); it is easily diagnosed due to the sharp increase in count per time unit (Fig. 1 presents a histogram of events with a resolution of 10 ms) so, when the data obtained are processed, it is removed from the count of radiocarbon ions. It should be noted that such events in a device that has been well tested are quite rare; nevertheless, not taking them into account leads to a decrease in the reliability of measurements using the device.

To test the ability of the AMS to record an increased level of a count, samples with a carbon concentration 10 and 100 times exceeding the natural level in living systems were put into the sample wheel of the ion source. The source of radiocarbon labels was represented by methanol enriched in radiocarbon. The detection was performed at an ion energy of about 4 MeV in a charge state of +3. The disintegration of

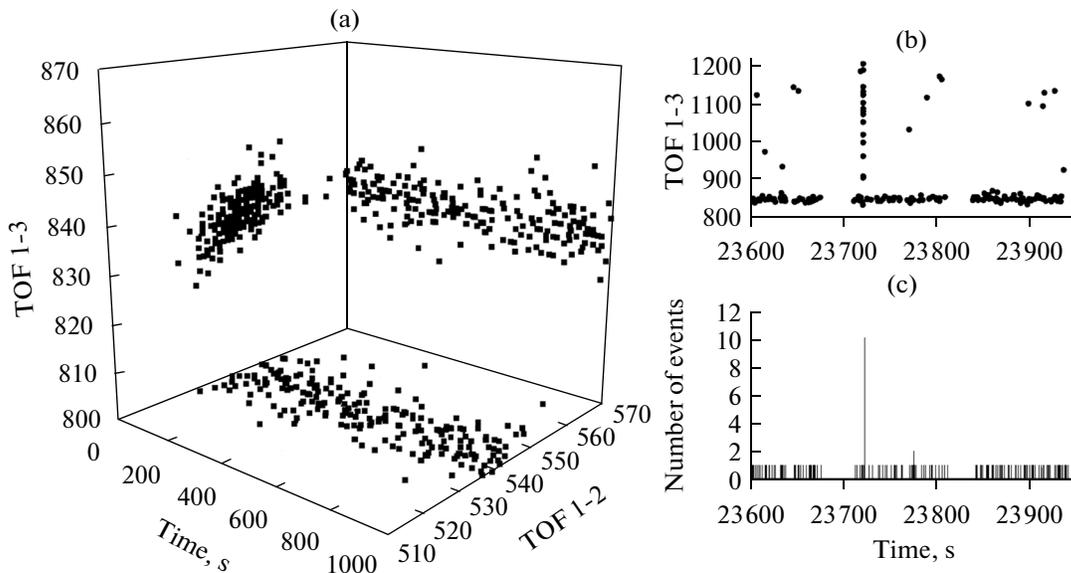


Fig. 1. Projections of a three-dimensional recording of ions (a) and recording of high-voltage breakdowns (b, c) by a time-of-flight telescope.

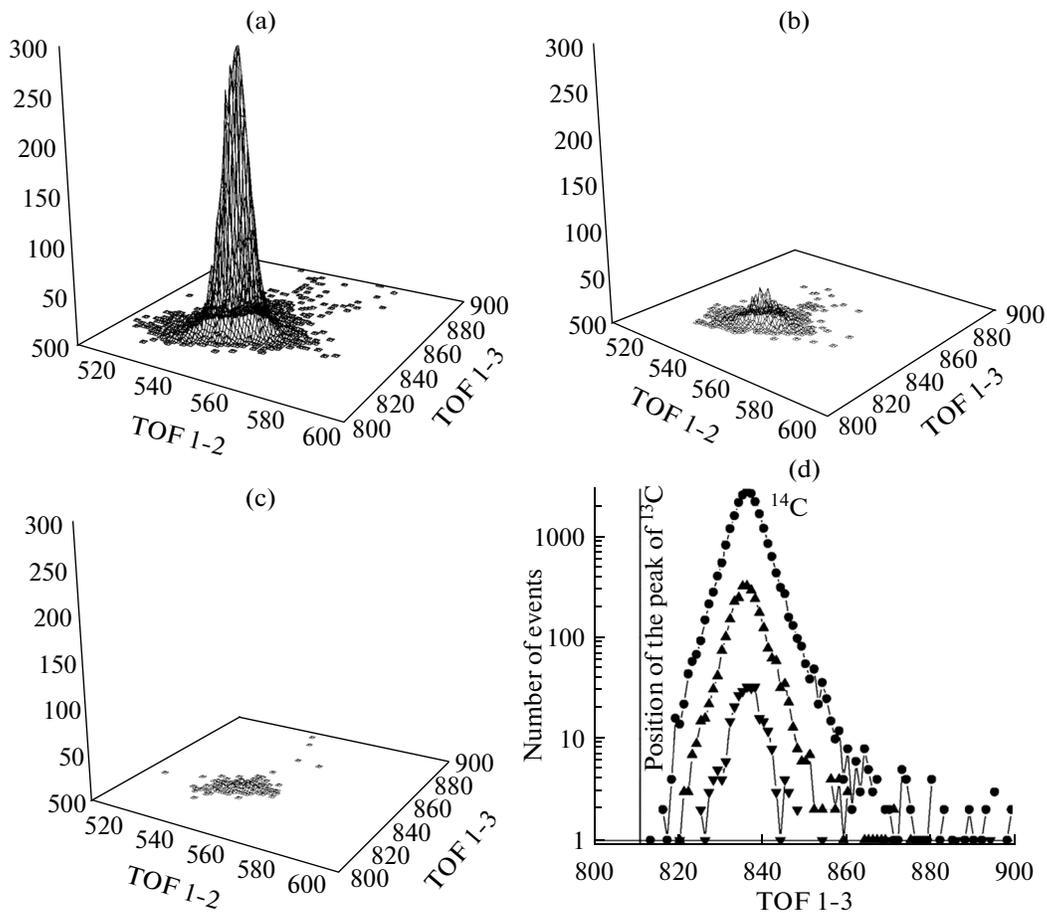


Fig. 2. Two-dimensional histograms of recording radiocarbon in samples with the natural concentration level (c) and with labels introduced that increase the radiocarbon concentration by factors of 10 (b) and 100 (a); a one-dimensional histogram of such concentrations (d) with the location of the nearest isotope ion background indicated.

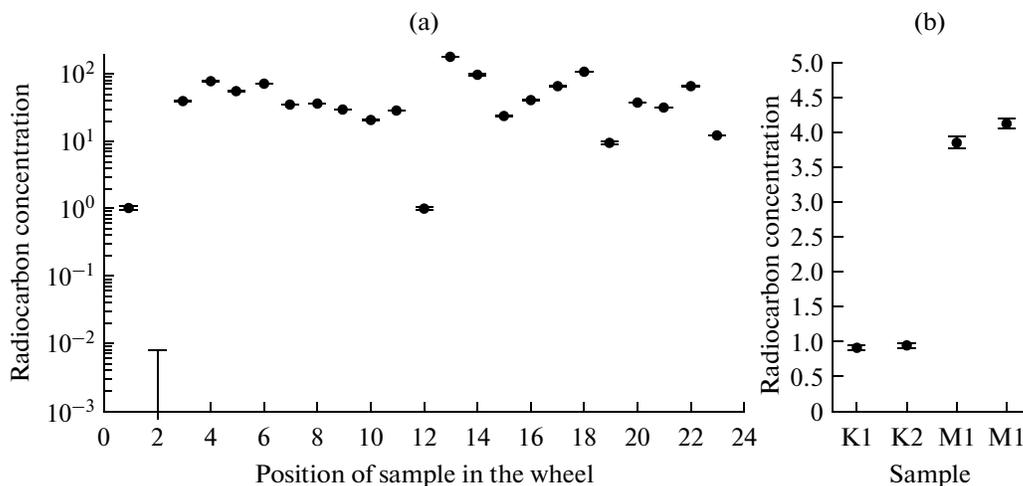


Fig. 3. The radiocarbon concentration measured in the AMS in samples enriched in radiocarbon (a) and an example of an AMS analysis of laboratory mice (b).

the molecular ion background was performed at a magnesium vapor target at a temperature of 580°C , which corresponds to the achievement of the equilibrium distribution with respect to ion charges at the exit of the target. Figure 2 shows good distance between the radiocarbon peak and the nearest peak of ion background (carbon with a mass of 13 u) for all concentrations of radiocarbon.

To test the degree of contamination of some samples when other samples are measured, samples with the natural level of radiocarbon in living systems (samples of carbon fiber at the positions 1 and 12 in Fig. 3a), sample with a low concentration of radiocarbon (graphite MPG at the position 2 in Fig. 3a), and series of samples with a radiocarbon concentration 10–100 times exceeding the natural one (the other samples in Fig. 3a) were put into the sample wheel of the ion source. The data given in Fig. 3a correspond to the number of radiocarbon ions in samples normalized to a current of carbon with a mass of 13 u measured at the exit of the AMS by a Faraday cup with a statistical measurement error presented. The mean value in carbon fibers is taken as 1. From Fig. 3a it could be seen that the concentration of radiocarbon measured from the graphite MPG is less than 1% (a mean value of the radiocarbon concentration in the graphite MPG, when samples are dated, is 0.2%). Thus, it could be concluded that performing measurements using the AMS the effect of the samples with increased concentration of radiocarbon on those with a natural one is insignificant. It should be noted that, graphitizing biomedical samples, the graphite MPG subjected to the similar procedure of combustion and graphitization in the same reactors has a significantly greater value of contamination of 2–3%.

At present, coworkers at Novosibirsk State University (L.Ph. Gulyaeva, et al.) jointly began the first studies in the field of biomedicine. We give as an example the radiocarbon concentration measured in an AMS in the kidneys of labeled mice (M1 and M2 in Fig. 3b) and in those of control ones (C1 and C2 in Fig. 3b). The labeled laboratory mice were administered with methyl alcohol containing radiocarbon labels with a radioactivity of 10 Bq (which corresponds to the natural radioactivity of 1/4 of a usual banana). In 3 h, the kidney samples were taken (3 mg of carbon in each). From Fig. 3b it could be seen that the radiocarbon concentration in the mice labeled significantly (four times) exceeds that in the control mice despite the small number of radiocarbon labels.

CONCLUSIONS

The data presented in the paper demonstrate that the BINP AMS makes measurements of the concentration of radiocarbon labels in biomedical samples and the reliable detection of radiocarbon from accompanying ion background possible.

ACKNOWLEDGMENTS

This study was supported by the Federal Special Purpose Program, state contract no. 14.512.12.0004, and Siberian Branch, Russian Academy of Sciences, integration project no. 106.

REFERENCES

1. N. I. Alinovskii, A. D. Goncharov, V. F. Klyuev, S. G. Konstantinov, E. C. Konstantinov, A. M. Kryuchkov, V. V. Parkhomchuk, M. V. Petrichenkov, S. A. Rashtigeev, and V. B. Reva, "Accelerator mass spectrometer

- for the Siberian Branch of the Russian Academy of Sciences,” *Tech. Phys.* **54** (9), 1350–1354 (2009).
2. S. A. Rastigeev, A. R. Frolov, A. D. Goncharov, V. F. Klyuev, E. S. Konstantinov, L. A. Kutnykova, V. V. Parkhomchuk, and A. V. Petrozhitskii, “Development of the BINP AMS complex at CCU SB RAS,” *Probl. At. Sci. Technol.* **79** (3), 188 (2012).
 3. V. F. Klyuev, V. V. Parkhomchuk, and S. A. Rastigeev, “A magnesium vapor charge-exchange target for an accelerator mass spectrometer,” *Instrum. Exp. Tech.* **52** (2), 245–248 (2009).
 4. V. V. Parkhomchuk and S. A. Rastigeev, “Analysis of the ion background in an acceleration mass spectrometer of the Siberian Division of the Russian Academy of Sciences,” *Tech. Phys.* **54** (10), 1529–1533 (2009).
 5. V. V. Parkhomchuk and S. A. Rastigeev, “Ion selection in accelerator mass spectrometer at the budker institute of nuclear physics,” *Phys. Part. Nucl. Lett.* **9** (4–5), 406–409 (2012).
 6. N. I. Alinovskii, E. S. Konstantinov, V. V. Parkhomchuk, A. V. Petrozhitskii, S. A. Rastigeev, and V. B. Reva, “A time-of-flight detector of low-energy ions for an accelerating mass-spectrometer,” *Instrum. Exp. Tech.* **52** (2), 234–237 (2009).
 7. V. V. Parkhomchuk, A. V. Petrozhitskii, and S. A. Rastigeev, “Thin-film detector for ion registration in accelerator mass spectrometers,” *Phys. Part. Nucl. Lett.* **9** (4–5), 448–451 (2012).

Translated by E. Berezhnaya