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ELECTRON AND ION BEAMS, ACCELERATORS

Accelerator Mass Spectrometer for the Siberian Branch of the Russian Academy of Sciences

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Abstract—A concept of an accelerator mass spectrometer intended for the Siberian Branch of the Russian Academy of Sciences is suggested. Preliminary tests have been conducted, and a carbon isotope having a mass of 14 amu has been detected in a charcoal sample.

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Accelerator mass spectrometers (AMSs) are intended for ultrasensitive analysis of the isotopic composition of materials [1]. In this case, the concentration of a radioactive isotope may be as low as 10^{-12}–10^{-14} of the main isotope concentration. At so small concentrations of a radioactive isotope, its content is measured in the ion count mode; that is, each ion is counted.

AMSs date back about 30 years and immediately found wide application, primarily in radiocarbon analysis based on measuring the ^{14}C isotope concentration. This analysis is necessary for proper object dating, environmental monitoring, biomedical monitoring of the human organism by tracing the propagation of radioactive carbon introduced in a very small amount, and some other applications. Today, the first Russian AMS is being designed (Institute of Nuclear Physics, Siberian Branch, Russian Academy of Sciences) [2]. Despite the long history of creating AMSs, the installation being developed offers new possibilities for suppressing the isobar background, which limits the sensitivity of the analyzer.

The sensitivity of mass spectrometers is limited by the background consisting of isobars and molecules, the masses of which are close to that of an isotope to be measured. Background particles may penetrate through filters by interacting with the residual gas and walls of equipment. Scattering, charge exchange, energy loss, fragmentation of molecules, etc., are attendant processes. Acceleration of particles to energies on the order of 1 MeV allows researchers not only to considerably decrease the cross section of particle–residual gas interaction, but also to apply additional methods of selection (such as determination of the nucleus charge and complete fragmentation of molecules passing into the charge state 3+).

SPECIFIC FEATURES OF THE ACCELERATOR MASS SPECTROMETER UNDER DEVELOPMENT

Most AMSs currently available are based on tandem electrostatic accelerators. In these instruments, a test sample is under a low potential outside the tank of the accelerator, which makes it possible to easily change samples. In addition, the use of negative ions at the initial stage of selection sometimes allows researchers to considerably reduce the isobar background. For example, it is known that negative ^{14}N nitrogen ions are unstable [1]. In the simplest case, a tandem accelerator represents two sequential accelerating tubes in which the beam propagates along a straight line.

It is emphasized in the new project that the background flux can be considerably reduced by placing an energy filter in the high-voltage terminal. In the available AMSs, an energy filter is placed either at the entrance to the accelerator, at its exit, or at both. However, these versions differ in filtering efficiency. In fact, although negative nitrogen ions are unstable, nitrogen atoms may fall into the accelerator in the form of nitrogen molecular compounds that generate negative ions. If molecular ions are produced in the ion source, a device placed in the inlet channel of an additional energy filter will substantially reduce this background. However, molecular ions produced from the residual gas in the first accelerating tube may be subjected to acceleration [3]. Upon passing through the first accelerating tube, such ions will have an energy lower than the energy of ions being analyzed, because (i) they lack the injection energy, (ii) the molecules fragment on a charge-exchange target, and (iii) the energy gain in the first accelerating tube is the highest for particles in the charge state 1–.

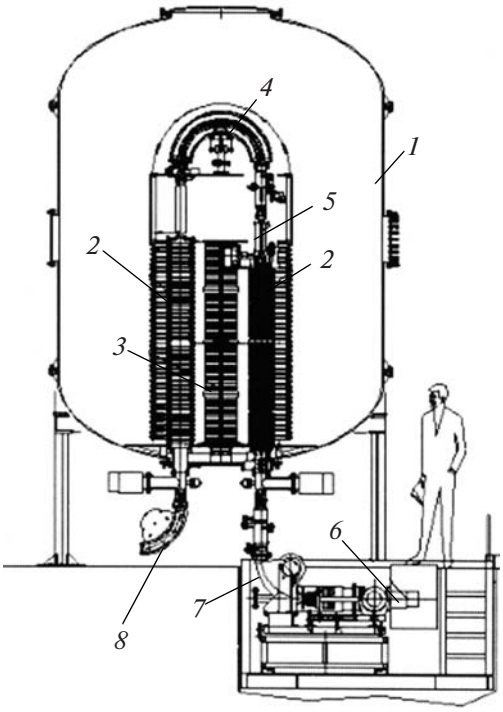

Fig. 1. Accelerator mass spectrometer: (*1*) tank for SF_6, (*2*) accelerating tube, (*3*) electrostatic filter, (*5*) magnesium vapor target, (*6*) ion source, and (*7*, *8*) magnetic filters.

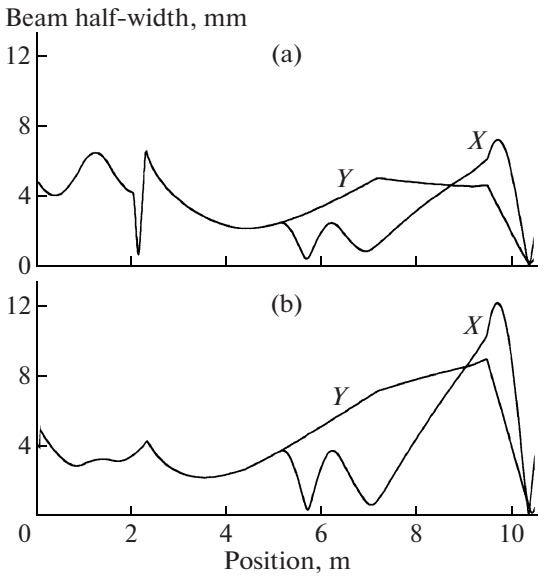

Fig. 2. Calculated transverse sizes of the beam along the AMS. The injection energy, accelerating voltage, and ion charge are (a) 30 keV, 2 MV, and 3+ and (b) 15 keV, 250 kV, and 1+.

Since the energy of background particles in the high-voltage terminal is always lower than the energy of test particles, the former can be efficiently filtered out in energy. The situation changes radically if the nitrogen from molecular fragments freely falls into the second accelerating tube, since ions having fallen into it may compensate for an energy deficiency by passing part of the tube in the charge state higher than that of test particles. Since the energies of test and background particles at the exit from the rectilinear accelerator may be the same, energy filtering becomes less efficient and subsequent separation is possible only using a nuclear detector, which distinguishes the particles that have passed through the material using the difference between energy losses.

Another feature of the project is that charge exchange is perceived to take place on a magnesium vapor target. Such a target does not degrade vacuum conditions outside the heated space. If vacuum conditions degrade (especially in accelerating tubes), the amount of transmitted background particles increases. For ion energies above 1 MeV, charge-exchange foils are not applied, because of the fast erosion thereof. In currently available AMSs, gas targets are used. Gas recirculating systems serve to reduce the gas flow into the accelerating tubes. However, such systems must be equipped with expensive turbomolecular pumps operating in the high-voltage terminal.

DESIGN OF THE ACCELERATOR MASS SPECTROMETER

The AMS assembly is schematically shown in Fig. 1. Negative ions are accelerated in the ion source in the horizontal plane to an injection energy, turn in a magnetic field by 90°, and are accelerated vertically up in the first accelerating tube toward the positive potential of the high-voltage terminal. Then, the ions are stripped to a positive charge state on the magnesium vapor target, reverse, and are accelerated down in the second accelerating tube toward the ground potential. Finally, the ions turn by 90° in the magnetic field and enter into a particle detector.

The variation of the transverse size of the beam calculated for such a motion is shown in Fig. 2. Calculations were performed with the MAD program [4]. Upon turning in the high-voltage terminal, in which radial and horizontal focusings differ, since deflecting plates are cylindrical, the beam is no longer circular.

When the voltage of the high-voltage terminal ranges from 1 to 2 MV, the optics of the instrument remains the same and the injection energy should be raised from 15 to 30 keV. The channel diameter of the charge-exchange target determines the calculated range of the beam's maximal emittance: 3.7–5.2 mm mrad $MeV^{1/2}$ for injection energies of 15–30 keV. For an accelerator voltage of 250 kV and a beam injection energy of 15 keV, the beam is not focused at the entrance to the accelerating tube. The respective maximal calculated emittance equals 1.8 mm mrad $MeV^{1/2}$.

Ions to be analyzed are generated by bombarding the sample with cesium ions. For this purpose, we

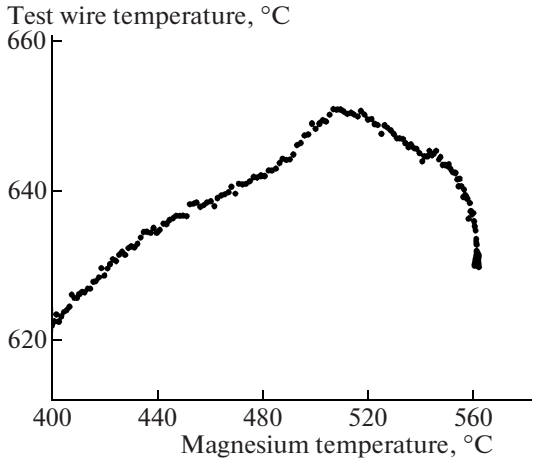

Fig. 3. Test of the magnesium vapor target. The temperature of the wire strained in the stripping channel is plotted vs. the temperature of the magnesium container.

designed a negative ion source in which ions are obtained by sputtering.

$CsCr_2$ pellets are heated approximately to 600°C, and cesium vaporizes. Passing through a tube, cesium vapor reaches a tantalum ionizer heated approximately to 1000°C. Positive cesium ions forming on the surface of the hot ionizer are accelerated and focused on the cathode with a test sample, since the ionizer has the form of a semisphere. The ionizer–cathode voltage is 8 kV. Negative ions knocked out from the sample are accelerated toward the ionizer by the same potential and, having passed through a hole in the ionizer (8 mm in diameter), are accelerated to the ground potential to reach an injection energy.

The accelerator is placed in a stainless steel tank 4.6 m in height and 3.2 m in diameter, which is intended for a pressure of 1.7 atm. Two accelerating tubes and a cascade generator are placed in a 2-m-high guard column with an inner diameter of 1.4 m. The accelerating tube consists of five sections with an electrode spacing of 21 mm. The electrodes are separated by ceramic rings with an inner diameter of 18 cm. The aperture of the accelerating tube channel is 10 cm in diameter. A voltage to the high-voltage terminal is applied from a symmetric cascade generator operating at a frequency of 20 kHz.

All the facilities in the high-voltage terminal are powered from a compressed-air-driven 500-W turbogenerator. Compressed air is supplied from the ground potential through high-resistivity hoses. At the moment, the accelerator is situated on a setup platform without radiation shielding; therefore, tests were conducted at lower-than-ultimate voltages of the high-voltage terminal and without filling the tank with a gas having a high dielectric strength (SF_6). A voltage of 500 kV was reached in air without predrying. Experiments with the beam were carried out at a voltage of 250 kV. Inside the tank (in the high-voltage terminal), radiation detectors were mounted. At a voltage of 250 kV and a beam current of 1 μA, the radiation background inside the tank was about 1 mR/h. Outside the tank, the influence of the beam on the background was not detected because of absorption in the tank wall.

The stripping channel of the target is 30 cm long and has an inner diameter of 6 mm. At the ends of the target, wells for magnesium vapor deposition are provided, which are kept at room temperature. An evacuated container with solid magnesium is placed outside the channel. When the container is heated, magnesium vapor penetrates into the stripping channel through small holes. The target was tested using a tungsten wire strained in the stripping channel.

The conductivity of the wire was measured as a function of its temperature. The temperature of the wire (the temperature was converted from the resistance of the wire) versus the temperature of the magnesium container is shown in Fig. 3. The temperature of the wire first grows because of thermal radiation from the heating stripping channel. Then, when the magnesium vapor pressure increases considerably, the wire starts cooling due to the heat conduction of the magnesium vapor, since the surrounding stripping channel has a lesser temperature. To date, the magnesium target has operated for several hours as part of the AMS.

To properly guide the ion beam, correction of its coordinates and angles at the exit from the ion source is provided by means of four pairs of electrostatic dipoles. The same correction systems are installed in the high-voltage terminal before and after the place where the beam reverses. Initially, the guided beam had only an injection energy (the high-voltage terminal was deenergized) and the tank of the accelerator was open. In this case, one can adjust the positions of the accelerator units directly from the signals of the sensors. The position of the beam was determined using signals from the plates of the electrostatic dipoles.

Ion detection in the count mode was accomplished with a time-of-flight detector. The detector consists of two sensors recording the instant of ion transit. The sensors contain a thin film, and the ions penetrating it cause electron emission. The electrons knocked out of the film are extracted by applying a voltage of 1.5 kV and then are reflected by the electrostatic field toward microchannel plates, where they multiply to a level sufficient for the operation of the discriminator. Finally, the time of flight between the sensors is measured. The time step in time-of-flight measurements is 0.5 ns.

The control and monitoring system of the AMS assembly is fully computerized and uses the CAMAC and ADAM standards and LabVIEW software. During operation, about 100 parameters of the electronics and the readings of the beam-diagnosing sensors are written and memorized.

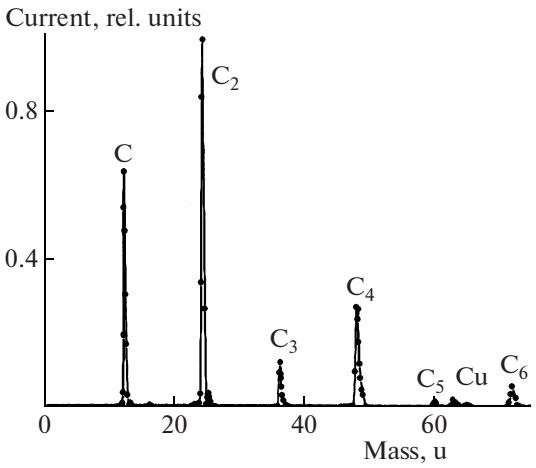

Fig. 4. Mass spectrum of negative ions produced in the sputtering ion source with the graphite sample.

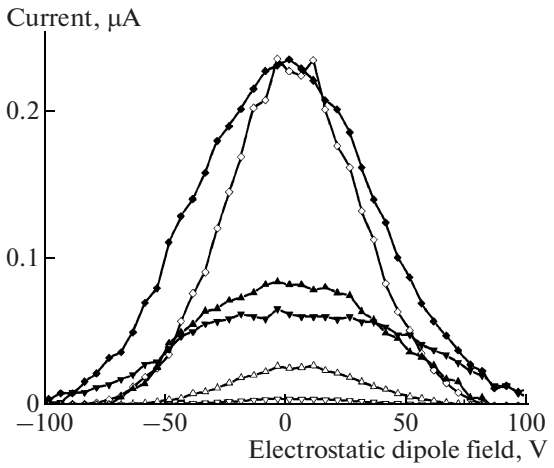

Fig. 5. Variation of the beam current at the exit from the AMS with electrostatic dipole voltage when quarter of the current is picked up from the body of the stripping target (empty circles) and when the beam is displaced in the transverse direction (filled circles). The target temperature characterizing the beam transmission is (◇) 500, (△) 400, and (▽) 350°C.

RESULTS OF EXPERIMENTS WITH THE ION BEAM

A typical ion mass spectrum taken of the graphite sample at the exit from the injection magnet is shown in Fig. 4. The ion current of carbon having a mass of 12 amu (carbon-12) is 3 μA, and the ion energy is 10 keV. Along with carbon ions, cluster ions and copper ions are produced. The cathode is made of copper, and a sample is placed in a hole 2 mm in diameter. At the initial stage of sputtering, the spectrum contains oxygen in an amount comparable to the amount of carbon. If briquette coal is used instead of graphite, the oxygen content remains nearly constant across the thickness of the sample. The spectrum also exhibits hydrogen-containing molecules. The fraction of carbon-14 relative to the amount of carbon-12 is 3×10^{-4}.

If the negative ion beams passes through the stripping target, differently charged ions appear at the exit. The ion charge equilibrium distribution depends on the beam energy. For the optimal charge exchange into the state 3^+, an energy of about 2 MeV is necessary [1]. At a voltage across the terminal of 250 kV, roughly half of the ions are stripped to the charge state 1^+ and approximately the same amount are stripped to the charge state 0 (the fraction of ions in other charge states is insignificant). For example, when the current of the injected negative ion beam is 1 μA, the contributions of ions with different charge states to this current at the exit from the AMS are the following: C^{1+}, 0.2500 μA; C^{2+}, 0.0150 μA; and C^{3+}, 0.0003 μA. The currents were measured with Faraday cups at the exits from the injection magnet and AMS. Note that, to measure the fractions of different charge states at the AMS exit, we measured the field of the output magnet and electrostatic parameters in the high-voltage terminal. When readjusting the output magnet, we took into account that the energy extracted in the second accelerating tube is proportional to the ion charge.

To study the propagation of particles along the AMS channel, we measured the charge on the body of the magnesium target. Corresponding data are presented in Fig. 5. The beam is scanned by the electrostatic dipole. When the dipole is under a zero potential, the beam passes through the stripping channel in an optimal path. The injected beam current equals 1 μA. At a small temperature of the target, when the magnesium vapor pressure is low, the beam current at the exit from the AMS is nearly zero. However, the current picked up from the magnesium target is about 0.25 μA, which suggests that injected negative ions directly hit the body of the target. As the target temperature rises, electrons are stripped away from carbon ions. Stripped electrons fall on the body of the target, and carbon ions with charge 1+ reach the exit from the AMS.

The maximal current of $^{12}C^{1+}$ is roughly equal to 0.25 μA. From the ion charge state distribution, this value corresponds to a current of the carbon beam emitted from the target of 0.5 μA. Since the number of electrons stripped away from each emitted ion is about 1.5, the current picked up from the target is expected to be 0.75 μA. This value is consistent with the signal from the target (see Fig. 5). Thus, the beam leaving the target reaches the exit from the AMS without losses. Losses are observed near the magnesium target: one quarter of the current is lost on its body and one more quarter is lost on the diaphragm 7 mm in diameter that is placed immediately before the target. It should be noted that 250 kV across the high-voltage terminal is not a design value. As follows from the above calculations, with an increase in the voltage of the accelerator,

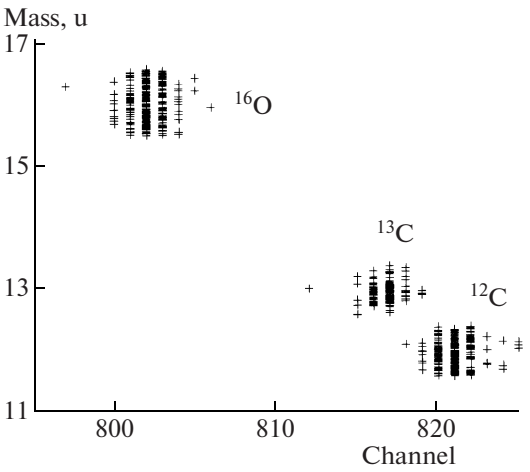

Fig. 6. Mass spectrum at the exit from the AMS under the conditions of scanning by the output magnet. On the abscissa axis, the time of flight of ions in the channels of the time-of-flight detector is plotted.

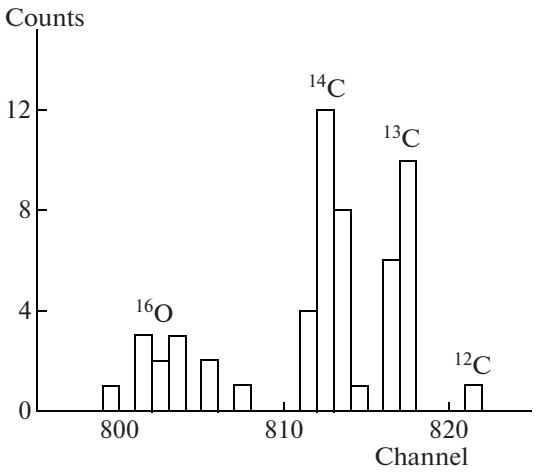

Fig. 7. Record of the ^{14}C isotope present in the charcoal sample. The ordinate axis, number of events; the abscissa axis, the same as in Fig. 6.

the emittance of the beam emitted by the system grows roughly by a factor of 3, so that one can expect lossless propagation of the beam under such conditions.

A typical ion spectrum recorded by the time-of-flight detector is depicted in Fig. 6. Ions passing through the filter of the high-voltage terminal have charge 3+, and their energy at the exit from the AMS slightly exceeds 1 MeV. This spectrum was obtained under the conditions of magnetic scanning at the exit from the AMS. The isotope ^{14}C is absent in the spectrum, since the amount of it is very low. This isotope was recorded in the steady-state count mode, when the system was tuned to passage of the ^{14}C isotope with charge 3^+ (see Fig. 7).

The sample was prepared from briquette charcoal. At an accelerator voltage of 250 kV, the fraction of ions with charge 3+ is small and it is therefore difficult to gain relevant statistics to measure radioactive carbon. At this stage of investigation, we can only notice that $^{14}C^{3+}$ ions can be recorded.

Today, an AMS center with an underground shelter intended for the accelerator is being built up. Under such radiation-safe conditions, the accelerator voltage can be raised to provide optimal charge exchange into the charge state 3+.

CONCLUSIONS

An accelerator mass spectrometer has been designed and installed to conduct preliminary tests at the Institute of Nuclear Physics (Siberian Branch, Russian Academy of Sciences). The basic point in the concept of this mass spectrometer is additional filtering of the isobar background. The mass spectrometer has been tested at a reduced voltage (250 kV). The passage of the beam through the AMS system meets design calculations. The radioactive ^{14}C carbon isotope has been recorded, the concentration of which in currently available samples is 10^{-12} of the ^{12}C concentration.

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REFERENCES

1. C. Tuniz, J. R. Bird, D. Fink, et al., A*ccelerator Mass Spectrometry: Ultrasensitive Analysis for Global Science* (CRC, Boca Raton, 1998), p. 3.
2. N. I. Alinovsky, V. F. Klyuev, E. S. Konstantinov, et al., in *Proceedings of the 9th European Particle Accelerator Conference (EPAC 2004), Lucerne, Switzerland, 2004,* p. 2389.
3. J. Southon, G. Santos, K. Druffel-Rodriguez, et al., Radiocarbon **46** (2), 41 (2004).
4. H. Grote and F. C. Iselin, The MAD program, CERN/SL/90-13 (AP), 1996.