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ELECTRON AND ION BEAMS, ACCELERATORS

Analysis of the Ion Background in an Acceleration Mass Spectrometer of the Siberian Division of the Russian Academy of Sciences

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Abstract—The mechanisms of ion background passage in an acceleration mass spectrometer are investigated. It is shown that the background level is controlled by the interaction between ions and residual gas atoms. An effective method is proposed for suppressing the ion background.

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INTRODUCTION

An acceleration mass spectrometer (AMS), which is being developed at the Budker Institute of Nuclear Physics, Siberian Division, Russian Academy of Sciences [1], is intended for isotope analysis of carbon. The concentration of the radioactive carbon isotope with a mass of 14 u in experimental samples is at a level of 10^{-12} relative to the mass of the main isotope. The sensitivity of an AMS is limited by the background of "alien" ions that have passed through all selection stages of the analyzer [2]. Nitrogen ^{14}N ions and molecular ions $^{13}CH_2$ or ^{13}CH with close masses form the basis of the background flow. The separation of these ions using electromagnetic fields separating their trajectories is complicated in the case of a small mass difference $\Delta m/m < 10^{-4}$. Apart from the difference in masses, it is also important to take advantage of different features of interaction of atoms with matter, as well as different abilities of atoms and molecules to form stable ions with a chosen charge state. Processes of scattering, ionization, electron capture, and energy loss during the interaction with a residual gas in the vacuum channel limit the selection potentialities.

Each element being selected has its own role. The general pattern of separation of 14-u carbon isotope can be described as follows. Electric and magnetic filters are intended for separating ions with a mass difference of 1 u and higher. Negative carbon ions are generated in the ion source, while negative nitrogen ions are unstable. The first spectrometer directs all ions with a mass of 14 u to the acceleration column, while molecular ions dissociate in the charge-exchange target. The initial number of 14-u molecular ions is controlled by residual hydrogen left after graphitization of the sample and usually does not exceed 10^{-3} relative to ^{12}C.

All particles that cannot be filtered out by the above methods are analyzed in the nuclear detector. It has become possible to use detectors in nuclear physics since ions can be accelerated to energies of several megaelectronvolts. Additional separation can be carried out using deceleration of ions in a gas or in films. Acceleration mass spectrometers based on a tandem-type electrostatic accelerator are used most often. In such systems, the sample is at a low potential, which is required for routine everyday replacement of samples being analyzed.

Ion selection in a high-voltage terminal of the accelerator is not envisaged in existing AMS systems. The complex being developed at the Budker Institute of Nuclear Physics ensures filtration of a "fragment" of molecular ions by an electrostatic turn directly in the region of their formation. The motion of ions in such an AMS can be described as follows. Negative ions are accelerated in the horizontal direction in the ion source up to the injection energy and then are turned through 90° in a magnetic field and accelerated in the upward direction in the first accelerating tube towards the positive potential of the high-voltage terminal. Ions are "stripped" to a positive charge in the magnesium vapor target, are turned through 180° in the electron field, and then are accelerated in the downward direction in the second accelerating tube toward the earth potential. The accelerated ions are turned through 90° in the magnetic field and enter the particle detector along the horizontal.

The accelerator is not equipped as yet with radiation protection; for this reason, the analysis of the ion background was carried out at 250-kV voltage of the

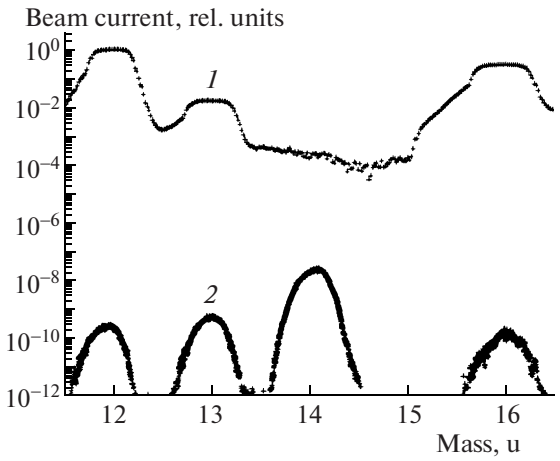

Fig. 1. Mass spectrum of (*1*) injected and (*2*) accelerated beams.

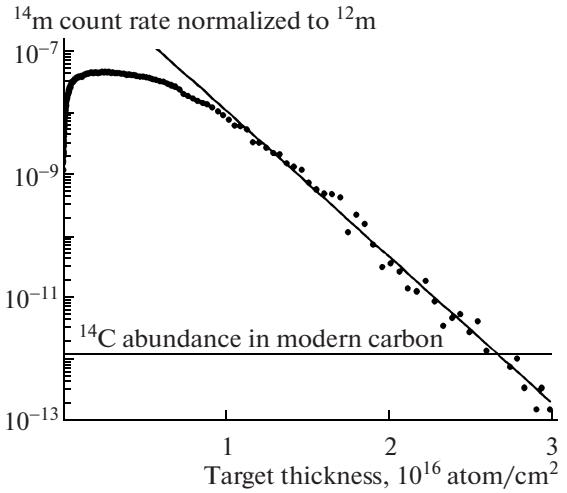

Fig. 2. Destruction of molecular ions during their passage through a magnesium-vapor target. The number of events with a mass of 14 u normalized to the number of events with a mass of 12 u is laid on the ordinate axis, while the target thickness is laid on the abscissa axis. The level of carbon isotope concentration with a mass of 14 u in a modern "living" sample is indicated.

high-voltage terminal. For an injection energy of 10 keV, the beam energy at the AMS outlet is 760 and 1010 keV for ions with a charge of 2+ and 3+, respectively. After the passage through the magnesium-vapor charge-exchange target, the fraction of ions with the 2+ charge is 3% and those with the 3+ charge is 0.05%. The terminal voltage is planned to be elevated to 2 MV to increase the fraction of ions with the 3+ charge to 50%.

ANALYSIS OF ION BACKGROUND

Figure 1 shows a typical mass spectrum of a graphite sample. The spectrum of negative ions (curve *1*) was recorded from the Faraday cylinder at the injection magnet output. It can be seen that, apart from carbon ions, a large number of oxygen atoms are present. The natural concentration of the 13-u carbon isotope is approximately 1.1% of the concentration of the main isotope. However, the measured concentration of the isotope with a mass of 13 u is approximately 2%. This can be explained by the fact that ^{13}C atoms and ^{12}CH molecules cannot be separated by the injection magnet. The lower spectrum in Fig. 1 corresponding to ions with a charge of 2+ was recorded by a semiconducting detector at the AMS outlet. The injection magnet is tuned in this case to the passage of a mass of 14 u, the electrostatic turn is adjusted to a charge of 2+, and scanning is carried out by a magnet at the accelerator outlet. Current peaks are approximately of the same height, but the background of masses of 12, 13, and 16 u is suppressed by from seven to nine orders of magnitude. The peak of the 14-u mass in the spectrum does not correspond to the carbon isotope of mass 14 u, since the ^{14}C concentration in industrial graphite is several orders of magnitude lower than the level of 10^{-12} in modern samples. The peak corresponding to a mass of 14 u in the spectrum is the molecular background. It is well known that molecular ions dissociate during the interaction with matter [3]. For a stronger destruction of molecules with the 2+ charge, the target thickness should be increased.

Figure 2 shows the dependence of the number of 14-u molecular ions on the magnesium-vapor target thickness. The thickness of the target was recalculated from tabulated values of temperature and pressure for an effective length of 30 cm [4]. The number of molecular ions decreases exponentially with increasing target thickness.

Formally, the molecular background can be suppressed by many orders of magnitude. However, an increase in the target thickness leads to particle losses due to scattering. The maximal thickness of the target in Fig. 2 is an order of magnitude larger than the thickness required for charge exchange of ions to a charged state of 2+. For such a target thickness, the number of atomic ions halved. It should be noted that, with ions in the 2+ charge state, the lithium background problem arises. Lithium molecular ions with a mass of 14 u pass through the inlet channel and are accelerated towards the high-voltage terminal. In the charge-exchange target, molecules dissociate. Each atomic lithium ion corresponds to half the energy, and upon charge exchange into 1+, further motion in the AMS is equivalent to the motion of carbon ions with a mass of 14 u in a charge state of 2+. Such background lithium ions at the AMS outlet have an energy half that of carbon ions; however, these ions may arrive in pairs after the disintegration of molecules. Such a background cannot be separated by measuring the particle energy; the separation can be carried out only from different energy losses of the lithium and carbon atoms in a substance. The concentration of lithium atoms in

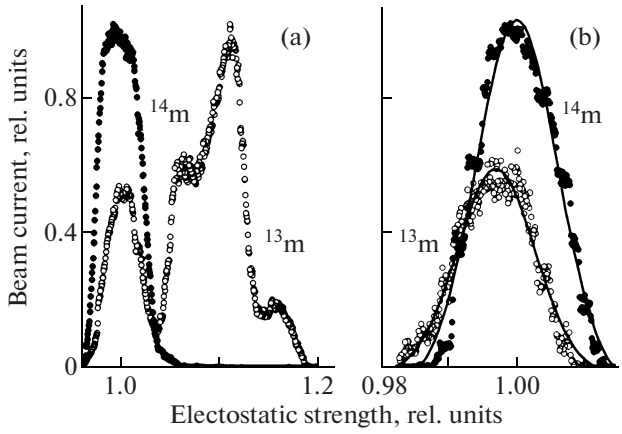

Fig. 3. Passage of the ion background with a mass of 13 u: (a) scanning of the beam by the electrostatic component of the Wien filter at the outlet of the injection magnet; (b) beam scanning by electrostatic turn in the high-voltage terminal. The beam current is laid on the ordinate axis.

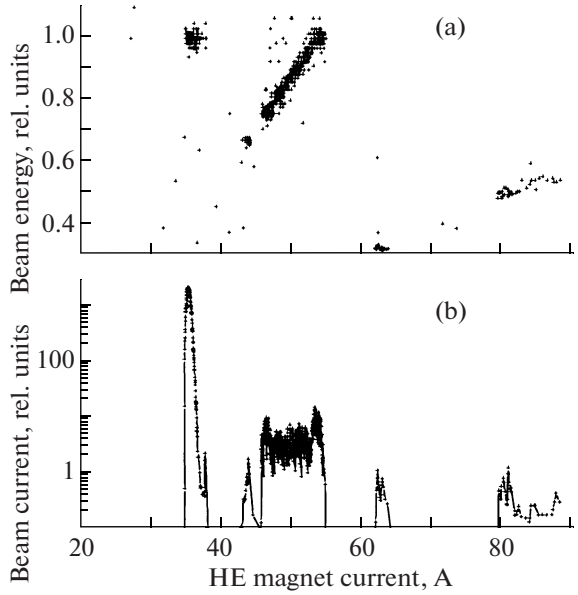

Fig. 4. Dependence of (a) energy and (b) intensity of accelerated ions on the magnet current at the AMS exit.

the sample being analyzed depends on the sample and the degree of its purity. When charge states of 1+ and 3+ of the carbon ion are used, this mechanism of passage of lithium ion does not operate. Molecular ions in the 1+ charge state disintegrate not so easily, and the target thickness should be increased still further.

Let us consider the mechanism of passage of background ions with a mass of 13 u (the corresponding peak can be seen in scanning by the magnetic filter at the AMS outlet). For this purpose, a Wien filter selecting ions by their velocity is added at the outlet of the injection magnet.

Figure 3a shows the dependence of the ion background of mass 13 and 14 u passing though the accelerator on the force of the electrostatic component of the Wien filter. The injection magnet is tuned to a mass of 14 u, while the magnet at the AMS outlet is tuned to the mass of the background ions being measured. It can be seen that a part of ions with a mass of 13 u has the same velocity at the outlet of the injection magnet as that of ions with a mass of 14 u. Analysis of the beam energy at the high-voltage terminal shows (Fig. 3b) that 13-u ions have an energy 790 eV lower than the energy of 14-u ions. This approximately corresponds to 1/14 of the injection energy.

Thus, ^{13}CH molecular ions pass through the injection magnet, disintegrate, and then are accelerated. Since the disintegration of molecules occurs at the injection energy and not in the high-voltage terminal, a fragment of an ion with a mass of 13 u has a close energy and cannot be filtered out by the electrostatic turn (see Fig. 3b).

Ionization and electron trapping by ions in the accelerating tube are responsible for a considerable energy spread of background ions. The energy acquired by an ion in the accelerating tube is proportional to the ion charge. If an ion experiences charge exchange in the accelerating tube, the energy of the accelerated ion changes. Figure 4 illustrates the effect of these processes for 12-u carbon ions. The figure shows the energy and intensity of ions at the AMS outlet as functions of the output magnet current. The energy was measured by a time-of-flight detector. The AMS was adjusted for the passage of 12-u ions with a charge of 3+; the voltage at the terminal was 250 kV and the injection energy was 10 keV. The energy of "regular" ions at the AMS outlet was 1010 keV. From this value, an energy of 250 keV is acquired in the first accelerating tube in which negative ions are accelerated.

After charge exchange in the magnesium target, ions with a charge of 3+ and an energy of 260 keV optimally make the electrostatic turn and then acquire an energy of 750 keV in the second accelerating tube. Such ions pass through the outlet magnet at a magnet current of approximately 37 A (Fig. 4). However, for a higher magnet current, a large number of "irregular" ions with various energies can be seen. For a magnet current of 54 A, ions have the same energy as the main beam. These are the ions that have captured an electron during the interaction with the residual gas at the outlet of the second accelerating tube. A magnet current of 47 A corresponds to the ions that have captured an electron at the inlet of the second accelerating tube; the energy of such ions is 760 keV. A magnet current in the range 47–54 A corresponds to ions that have captured an electron in the second accelerating tube. Accordingly, the energy of such ions ranges between 760 and 1010 keV, depending on the region of interaction with the residual gas. Such processes also occur in the first accelerating tube, but not all ions can make the electrostatic turn after the charge exchange. For

example, a current of 44 A corresponds to ions that have lost an electron in the first accelerating tube, in which their energy was approximately 173 keV. The ions possessing such energy and recharged into the 2+ ions in the magnesium target make the electrostatic turn of the high-voltage terminal tuned to ions with the 3+ charge and an energy of 260 keV. After that, the ions in a charge state of 2+ are accelerated in the second accelerating tube and have an energy of 673 keV at the AMS outlet. Analogously, ions in a charge state of 1+ pass at a magnet current of 63 A and have an energy of 337 keV. For magnet currents exceeding 80 A, ions with the 3+ charge that have made a turn in the high-voltage terminal capture two electrons in the second accelerating tube. The minimal energy of such ions is 510 keV.

Molecular ions in a charge state of 3+ are unstable; thus, all molecules with a mass of 14 u disintegrate. Fragments of molecules differ in mass by 1 u or more and can easily be filtered out in filters with magnetic and electric fields. Each additional filter suppresses such a background by several orders of magnitude. The situation with nitrogen is different. It was noted above that negative nitrogen ions are unstable. Nevertheless, nitrogen enters the system in the form of molecular fragments. Naturally, such molecules have a mass of not 14 u, but at least 15 u. If such ions are formed in the ion source, they can be filtered out even in the inlet channel. However, negative molecular nitrogen ions can be formed from nitrogen molecules of the residual gas in the vacuum system. Nitrogen ions trapped in the acceleration regime in the first accelerating tube may pass through the entire system. The most important matter is that atomic nitrogen ions at the AMS outlet may have the same energy as that of the 14-u carbon isotope being measured. Filters with electric and magnetic fields mounted at the AMS outlet become ineffective. The presence of nitrogen background involved in the acceleration regime was noted in [5]. It was observed in [5] that the background level depends on the beam intensity; i.e., the beam provokes the formation of negative molecular oxygen ions. The AMS design proposed here has a high degree of suppression of nitrogen background primarily due to the energy filter located in the high-voltage terminal.

The existing AMSs have no filter behind the first accelerating tube, which makes them vulnerable to the passage of nitrogen. Let us consider the most probable mechanism of passage of nitrogen ions, in which the energies of nitrogen ions and 14-u carbon ions at the AMS outlet are equal. The nitrogen ions engaged in the acceleration regime have an energy lower than the energy of the carbon ions at the outlet of the first accelerating tube. Since nitrogen ions formed as a result of disintegration of molecules have only part of the energy of the molecule, such ions did not possess the injection energy and could be formed not near the inlet of the accelerating tube. The maximal acceleration in the first accelerating tube is ensured for an ion charge of 1–, and carbon ions have already acquired such a charge; for this reason, there are no mechanisms for compensating the energy deficiency for nitrogen ions at the first acceleration stage.

Since the energy of nitrogen ions in the high-voltage terminal is always lower than the energy of carbon ions, the energy selection by the filter is very effective. At the second sate of acceleration in the AMS, incompletely stripped positive ions are analyzed, since the fraction of ions with a charge equal to the charge of the nucleus for a terminal voltages of several megavolts is extremely small. Usually, carbon ions in a charge state of 3+ are used; the fraction of such ions in the charge distribution is maximal at an energy of approximately 2 MeV and is about 50%. Ions with a charge exceeding 3+ are accelerated more strongly and, hence, may compensate the energy deficit at the first acceleration stage due to acceleration in the second accelerating tube. For example, if nitrogen ions are stripped in the target to 4+, are accelerated in the second accelerating tube, and capture an electron from the residual gas when their energy becomes equal to the energy of carbon ions, the values of energy of nitrogen and carbon ions at the AMS outlet are equal.

For typical parameters of the process, including an electron capture cross section of 10^{-15} cm^2 (1 MeV corresponds to $^{14}N^{4+}$ at H_2 [6]), a vacuum of 10^{-4} Pa, and a length of the charge-exchange regions of 1 cm, the electron capture probability is 3×10^{-5}. Thus, only a single electron capture on the residual gas is required, which makes the passage of nitrogen ions with "regular" energy quite probable. Such a mechanism of nitrogen passage is blocked in the AMS system proposed here. Nitrogen ions can make the electrostatic turn of the high-voltage terminal only if they have a smaller charge and after charge exchange in the first accelerating tube, like ions at a current of 44 A in Fig. 4.

To attain the same energy as that of carbon ions, nitrogen ions must change their charge several times upon interaction with the residual gas in the second accelerating tube. Nitrogen ions must change their charge state from 2+ to 4+ and then to 3+ again, which will ensure the equality of the energy and charge state of nitrogen and carbon ions at the outlet of the AMS. Thus, to pass the nitrogen background that cannot be suppressed by electromagnetic fields, fourfold charge exchange of ions at the residual gas is required, not a single charge exchange as in AMS systems without ion filtration in the high-voltage terminal.

Another feature of our project is the use of a magnesium-vapor target instead of a gas target for charge exchange. Such a target does not degrade vacuum beyond the volume being heated. For an ion energy on the order of 1 MeV, charge-exchange foils are not used because of their rapid destruction. Worsening of the vacuum conditions increases the probability of interaction of ions with the residual gas and, as a conse-

quence, the probability of passage for background ions.

CONCLUSIONS

Our analysis of the passage of the ion background demonstrates a strong effect of interaction of ions with the residual gas. An effective method is proposed for suppressing the isobaric background of nitrogen ions by an energy filter mounted after the first stage of ions acceleration and by using a charge-exchange magnesium-vapor target, which improves the vacuum conditions. The efficiency is ensured due to the fact that the energy of nitrogen ions after the first acceleration stage is always lower than the energy of the carbon isotope with a mass of 14 u being measured.

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