Charge state optimisation for beryllium accelerator mass spectrometry

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ABSTRACT

The yields of $^9$Be charge states ($q = +1, +2, +3$, and $+4$) from injected BeO$^-$ were measured with terminal voltage range from 1 to 5 MV. The yields were determined from the momentum analysed beam currents measured by beam profile monitors, after the 90° injector and analysing magnets. Stripping gas pressure dependency of the transmission efficiency was also determined and the results were studied with SRIM simulations. The effect of stripping medium was studied using both Ar and CO$_2$ gases. The presented results are compared against experimental values found from literature, semi-empirical parametrisation models, and charge-exchange theories.

1. Introduction

Accelerator Mass Spectrometry (AMS) is an ultrasensitive analytical technique in which atoms are counted directly instead of being induced to emit radiation. In this technique, negative ions are formed in the sputtering process in the ion source, extracted and accelerated to high energies in two stages, with charge exchange between stages, separated according to their momentum, charge and energy, and then individually counted after identification to have the correct atomic number and mass. The suppression of the molecular background is one reason for the high sensitivity of AMS. AMS is able to measure concentrations of one atom in $10^{13}$ atoms to an accuracy of about one per cent, using a sample size of the order of one milligram, and a measurement time of one hour.

AMS at University of Helsinki is based on a 5 MV tandem accelerator that routinely performs radiocarbon measurements. The accelerator contains two ion sources and five beam lines and is mainly used for $^{14}$C AMS, ion beam analysis and irradiations. Due to increasing interest in processes of a time scale wider than the limit of radiocarbon dating, for example geological applications, the development of measurement setups for other isotopes is underway. One of the isotopes of cosmogenic origin and with a long half-life is $^{10}$Be ($t_{1/2} = 1.5 \times 10^6$ years). To adapt the existing AMS setup for $^{10}$Be AMS measurements, transmission efficiency was also studied to check the minimum gas thickness requirement for charge state equilibrium conditions. From a practical point of view, it is important to ascertain charge state distributions and to maximise accelerator throughput for precise and efficient Be AMS measurements.

2. Experimental setup and measurements

The University of Helsinki 5 MV tandem accelerator [1,2] presented in Fig. 1, with the gas stripper was used to study transmission of $^9$Be with terminal voltages from 1 MV to 5 MV. This terminal voltage range covers a wide range of operating AMS machines. In the following Be always refers to $^9$Be. Stripping efficiency was measured with CO$_2$ and Ar stripping gases.

The accelerator is comprised of three major sections: 1) the low energy section with two MCGSNIICS (from National Electrostatics Corp.) ion sources and an injector, 2) the acceleration section with low and high energy acceleration with a gas stripper, and 3) the high energy side. The BeO$^-$ ions were extracted from the ion source, accelerated to $45$ keV, and injected to the main accelerator. Beam currents after the injector at the entrance of the accelerator were measured with the Beam Profile Monitor (BPM) and with the Faraday Cup (FC) denoted as BPM_IN and FC_IN respectively.

Charge-exchange at the high voltage terminal was achieved with a gaseous electron stripper. The stripper system is located in the middle of the terminal and is comprised of a tube of diameter 8 mm and of length 546.5 mm. The stripping gas is recirculated through a turbo molecular pump, with a pumping speed of $150$ L/s. The gas is fed in through an inlet in the centre point of the stripping canal and pumped to the ends of the stripping canal. Currently, two gas containers with Ar...
and CO₂ gas are attached to the gas feeding tube. To determine how the transmission efficiency depends on the stripping gas pressure, the system was optimised for +2 charge state and the thermal leak valve was set to high power. During the increase of the stripping gas pressure, the beam was monitored with BPM before and after the accelerating stage. Stripping yields for +2 charge state were extracted from BPM data. The measurement of the beam current corresponding to all charge states was performed by the BPM before the high energy analysing magnet. All the BPMs are of the rotating wire type (NEC model BPM80 or BPM83).

After the acceleration section, the beam is analysed with a 90° magnet. Before the analysing magnet, currents were measured with BPM and FC denoted as BPM_OUT and FC_OUT. Currents of Be beams with different charge states were measured after the high energy analysing magnet just before the beam line switching magnet with BPM and FC denoted as BPM_SW and FC_SW.

An example of the raw data output of a FC_IN and FC_SW from the measurement are shown in Fig. 1a. Beam profile monitors were used to determine beam currents to ensure reliable results regardless of the beam focus point. To calibrate BPM current measurement, the beam was focused to the nearest FC and calibration of BPM was done against the FC current. Mutual calibration between BPMs and FCs remains constant as checked several times during the measurements. The BPM x and y profiles are roughly Gaussian and have a FWHM of 2–3 mm. Example of BPM_SW area dependency of the beam current for charge state +2 is shown in the Fig. 1c. In the Fig. 1b. data is raw BPM data without calibrations. Beam currents determined from BPM signals were calculated averaging beam profile areas in x- and y-directions. Signals from BPMs are digitised and several parameters like areas and mean positions are calculated on the fly with real time digital signal processing units. The use of BPMs allows simultaneous determination of injected and out-coming beam current intensity. In addition, BPMs have larger aperture as compared to FCs, allowing for current measurements not to be affected by beam focus or position. Examples of the calibration between BPM signal and FC current are shown in the Fig. 1c.

3. Calculations and results

Measurements were performed in order to study the charge state distribution of Be in CO₂ and Ar stripper gas (see Figs. 2 and 3). Be transmission of the accelerator normalised to injector particle current during the measurement with CO₂ stripper gas was about 80% and with

Fig. 1. Schematic of the accelerator system at University of Helsinki, showing the components relevant to this experiment (upper panel). Lower panel: (a) Beam currents (electrical) from Faraday cups FC_IN and FC_SW; (b) uncalibrated beam profile monitor signals from BPM_IN, BPM_OUT, BPM_SW; and (c) BPM area response for beam current respectively.

Fig. 2. Results of the absolute charge state yield measurements with CO₂ stripper gas. Dashed curves are calculated from [7,5] Experimental data from Calcagnile et al. [3] are shown for comparison purposes.
Ar about 90%. Transmission dropped steeply for terminal voltages below 1.5 MV. Maximum yield of about 50%, was reached above 3 MV terminal voltages when the +2 charge state was selected, as shown in Fig. 2. The absolute yield is the ratio of particle currents measured by BPsMs.

Results of measurements for the two different stripper gasses as well as a set of published results [3], the only one to our knowledge, of Be charge state yield within a terminal voltage range of 0–5 MV, is presented in Fig. 3. The general trend, which can be seen for each of the charge states is an increase in yield up to a certain terminal voltage and then a fall off thereafter. The higher terminal voltages our machine can reach allows us to access higher charge states, which was not possible in the work by Calcagnile et al. [3], where Ar stripping gas was used and +2 charge state transmission efficiency was measured within 1.9 MV to 2.3 MV terminal voltage range. In terms of yield, their results are similar to ours, albeit slightly higher. With respect to the total yield, we notice a fall off of CO₂ around 1.0–1.5 MV. Measurements were done with 100 nA injected beam currents.

The underlying theory of the average equilibrium charge states for heavy ions in both gas and carbon foil strippers is given in [4–7]. The basic assumption made is that the charge-state distribution follows an equilibrium thickness. The review of charge exchange process by Betz [5] summarises many of the facts and formulae surrounding the charge exchange process of heavy ions in gaseous and solid targets.

Experimental results were compared with semi-empirical models for the average charge state with a Gaussian charge state distribution. The model in this case is based on an approximation and is used together with input from [7] to form the theoretical curves presented in Fig. 3. Another model we compared our charge yield distributions against in Fig. 3 is the work of J. L. Yntema (1974) [4], whose work is based on a parametrisation based on earlier experimental values on gaseous targets reported by Dmitriev and Nikolaev. Our results compare very well against both models, Gaussian distribution approximation [4] and the semi-empirical model [5]. Our results are also in relatively good agreement with the experimental data presented in the literature [3].

The effect of stripper gas density was studied by varying the pressure of the Ar stripping gas (see Fig. 4). Similar pressure dependence was observed for CO₂. Measurements by BPsMs allow us to make simultaneous measurements of the injected beam and the charge state distribution yield.

We checked also the pressure dependence of the angular scattering through a full SRIM simulation, which allowed us to obtain the kinetics of ions leaving the stripper canal. The fraction of ions transmitted through the canal was calculated for a given stripper gas density and acceptance angle of the high-energy section of the accelerating tube from the SRIM output. At some stripper gas pressure, one would expect to get the equilibrium charge state distribution. At higher pressure depending on which charge state is being studied the transmission efficiency drops down. However, if the gas pressure is too high, this would result in decreased transmission partly due to the effects of angular straggling.

Results presented in the Fig. 4 indicate the charge state equilibrium reached at 0.8 Pa gas pressure. In the same figure we include for comparison Be^{all}, which we believe, gives us a rough idea of what a measurement of all charge states would look like. However this particular measurement curve is from BPM_OUT and is proportional to the particle current before the analysing magnet. Maximum BPM_OUT signal is at about 0.7 Pa gas pressure. The maximum at lower pressure than for the +2 charge state can be explained by non equilibrium conditions, the higher yield of lower charge states and neutral atoms.

The general behaviour of the yield curve in Fig. 4 is an increase in yield with stripper pressure up to around 0.6 Pa, and then a decrease in yield thereafter. This is due to increasing angular straggling with increased stripper density, which limits the yield. This also means that there is a decrease in transmission. It is also observable that the experimental measurements drop a bit lower than the simulated line, which might be due to the energy loss in the stripper and thus change in the charge state distribution. No SRIM curve is shown for Be^{all}, due to additional factors which cannot be taken into account in SRIM as this measurement was made simultaneously to the one for Be^{+2}.

4. Conclusion

The optimum terminal voltage for Be AMS with a 5 MV tandem and BeO⁻ injection were found to be above 4 MV terminal voltage and a charge state of +2. A second stripper might be required for the charge state of +2, if there is a high molecular background, as it would be necessary to break up the molecules. With the 5 MV tandem accelerator, despite a lower beam current than for the measured +2 charge state, with a second stripper, a charge state of +3 might be of interest. We found during this study, the total transmission efficiency to be about 80% and 90% for CO₂ and Ar respectively for our system.

The charge-exchange models from [4–7] compares well against experimental results. The results give us an insight into charge exchange cross sections and effective charge states, both of which are relevant in studies and theory of stopping power.
Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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