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Investigation of gas stripping at 4.1 MeV for high mass negative ions

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ABSTRACT

We have used a tandem accelerator to measure charge state distributions at 4.1 MeV for negative ions ranging from carbon to uranium oxide passing through the gases helium, argon and xenon. The gas density was varied over a wide range to observe the onset of equilibrium stripping conditions. Using a 12° electrostatic deflector after the accelerator, charge states from 1+ to 8+ have been measured. For carbon, the charge state is seen to strip progressively from a low charge of around 1+ towards equilibrium with increasing gas thickness. However, for all the other ions, there is a high probability of multiple electron loss in single collisions with the gas, leading to mean charge states greater than 2 even at very low gas stripper pressure. This effect is observed to be gas-independent.

1. Introduction

The overall efficiency of Accelerator Mass Spectrometry (AMS) using tandem accelerators is principally a combination of two factors: first, the efficiency of converting the sample into negative ions in the ion source and, second, the particle transmission through the accelerator. The accelerator transmission is itself a combination of the beam optical transmission, which can be close to 100% with appropriate design, and the yield of the positive charge state selected for analysis. In the analysis of actinides by AMS with ANSTO's ANTARES accelerator [1,2], an accelerator transmission of 4–5% is achieved for the 5+ charge state at 4 MV terminal voltage using argon gas stripping. This is the lowest charge state which can be analysed with the existing analysing magnet (2 m radius, mass-energy product 250). Similar yields are achieved by other laboratories [3–5] using 5+ ions. Higher yields (10–15%) have been achieved using the 3+ charge state at 0.5–1.0 MV with lower voltage accelerators [6,7]; however it is not possible to use such low voltages on higher energy accelerators such as ANTARES.

The present study aimed to investigate gas stripping yields for high mass ions to gain a better understanding of how yields could be improved. Initial experiments have been performed with argon gas as the stripper medium. Alternative gases (helium and xenon) were then tested as there exist earlier data [8] which show contrasting effects from their use. In that paper, helium was shown to have equilibrium yields equivalent to foil stripping at low energy, while xenon gave higher yields of lower charge states. Purser et al. [9] proposed a method to improve transmission for actinides by using a low stripper thickness to enhance the yield of 1+ ions. These ions are then stripped again after acceleration to form higher charge states, thereby destroying molecules and enabling magnetic analysis. Hence a stripper with a high yield of low charge states is also of interest.

Our knowledge of the stripping process is based mainly on a large body of data obtained in the 1960s and 1970s, which have been the subject of reviews [10,11]. Various authors, for example Schiwietz and Grande [12], have used such data to derive semi-empirical formulae for calculation of the charge state distributions (mean charge state and distribution width). These formulae are derived for equilibrium stripping conditions. At equilibrium, multiple charge changing collisions occur, so that the projectile ion has no memory of its original charge state. The majority of the early data is for light ions at high velocity, where a low charge state positive ion is stripped to higher charge states. The theoretical understanding of the stripping process (see [11] and references therein) is built on the assumptions that the ion velocity exceeds the Bohr velocity (orbital velocity of valence electrons, in the target atoms) and that one or at most two electrons are lost (or gained) by the projectile in each collision. In tandem accelerators at lower voltages, such as those used commonly for AMS, these conditions may not be met, especially for high mass projectiles. For example, in the analysis of actinides by AMS, the ion velocities in the stripper are typically less than the Bohr velocity. Also, the initial ion is a negative ion, rather than the positive ions generally used in most charge state distribution experiments. Additionally, for actinides in particular, a molecular ion (such as UO^-) is injected, as this provides a higher yield from the ion source than atomic ions [3,4].

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2. Experimental method

Charge state distributions have been obtained from beam transmission measurements in the ANTARES FN tandem accelerator at ANSTO, using the first part of the actinides AMS beamline [1,2]. Beams emerging from the accelerator are analysed using a 12° electrostatic deflector and brought to focus by an electrostatic quadrupole doublet at a Faraday cup after the deflector (the 'DE Cup', see Fig. 1). The quadrupole is adjusted for each beam and charge state to focus that ion in the cup. The deflector separates ions according to their energy-to-charge ratio (E/q), given by

$$E/q = ((q + M_f/M_i) \times V + E_{inj})/q$$

where V is the terminal voltage, E_{inj} the injection energy, q the charge state, M_i the mass of negative ion injected (which may be a molecular ion) and M_f the mass of the fragment ion measured. For injection of atomic ions, $M_f = M_i$. In most cases, charge states up to 8+ can be resolved by the deflector.

Fig. 1 also shows a schematic of the gas stripper in the terminal of the accelerator. This system has been in operation on ANTARES for 15 years. It is of the recirculating type, similar to others built at that time, with an Alcatel TMP 5400 model turbopump (nominal pumping speed 400 L/s) operating at 89% of maximum speed. Since installation argon gas has been used, supplied from a gas bottle through a thermo-mechanical leak valve. Recently two extra gas bottles were added to this system, each with their own thermo-mechanical leaks, to supply xenon and helium stripper gases. The stripper thickness is controlled by adjusting the flow rate through each thermo-mechanical leak. A direct measurement of the stripper thickness is not available for this system. At the time of the measurements reported here, the vacuum gauge in the stripper chamber was not operational. Vacuum gauges at the entrance and the exit of the accelerator provide some indication of the stripper thickness by way of gas leakage down the accelerator tubes but are not sufficiently sensitive at low gas flow rates. As a consequence, for the data reported here, the voltage applied to the leak valve is used as a proxy for stripper thickness. This setting, although individual for each valve and not linearly related to thickness, was nevertheless found to be highly reproducible and a reliable gauge of stripper thickness. A residual gas analyser connected at the low energy (LE) end of the accelerator provided confirmation of the stripper gas being used and a further indication of stripper thickness from gas leakage to the LE end. By comparison with earlier data on non-equilibrium charge state yields [13], we can say that our experiments are operating in thickness range from about 0.03 to about 1 μg/cm^2.

Yields of each charge state were measured by comparing the beam current measured in the DE Cup to the injected negative ion current ('LE Cup' in Fig. 1). The total positive beam current emerging from the accelerator was also measured, in the 'HE Cup', which served as a means for checking the beam transmission between this point and the DE Cup; it is used below to distinguish different types of particle losses. Individual charge state yields were measured with a precision better than ±5%; repeat measurements at the same settings on separate occasions were performed to verify this. For clarity, error bars have not been included in the figures presented below.

The measurements reported here were all performed with 4 MV on terminal and 100 keV injection energy, giving 4.1 MeV negative ion beam energy at the gas stripper. Beams injected were C^-, Fe^-, FeO^-, I^-, Au^-, ThO^- and UO^-. In cases of molecular ion injection (XO^-), the emerging oxygen positive ions were measured in addition to the higher mass component (X^{n+}); XO^+ beams were also recorded.

With this method of measuring charge state distributions created in the accelerator terminal, neutrals and residual negative ions cannot be measured. Where 'total transmission' is referred to below, this means the total of the yields of positive charge state beams emerging from the accelerator. In cases of molecular ion injection, positive atomic (X^{n+}) and molecular ions (XO^+) are included; oxygen ions are not included in the calculation to avoid the possibility of double counting. 'Mean charge state' refers to the mean of the positive charge states of atomic ions of the element specified.

3. Results and discussion

Fig. 2 shows the mean charge state and total transmission as a function of stripper thickness for argon gas. The total transmission rises at first with increasing thickness, as the proportion of ions which interact with the gas increases. The transmissions reach peak values of 60–80% and then fall as beam losses increase. For carbon, the mean charge state can be seen to evolve from a single collision value of 1.5 to an equilibrium value approaching 3.8. Of the other species, iron oxide appears to show somewhat similar behaviour but the higher mass species do not exhibit the same

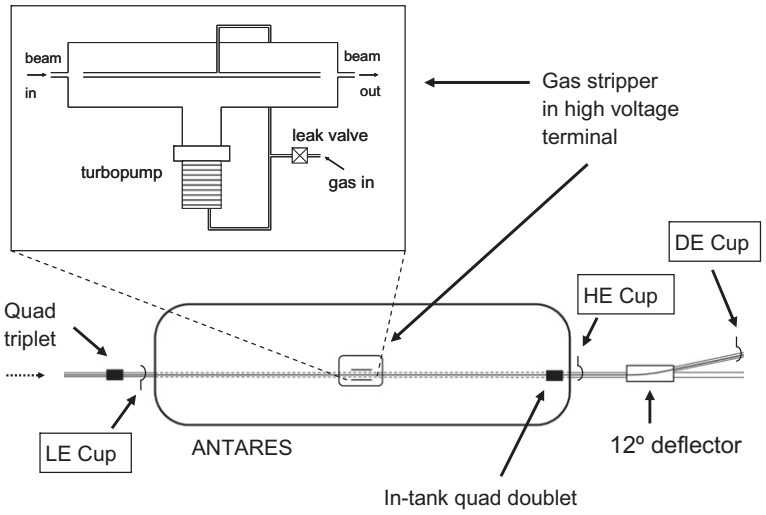

Fig. 1. Schematic of the elements of the ANTARES accelerator used for the current experiments, including an inset of the gas stripper in the terminal. The stripper has a tube length of 690 mm and diameter 9.5 mm.