Accelerator Mass Spectrometry in Laboratory Nuclear Astrophysics

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Abstract. The extreme sensitivity and discrimination power of accelerator mass spectrometry (AMS) allows for the search and the detection of rare nuclides either in natural samples or produced in the laboratory. At Argonne National Laboratory, we are developing an AMS setup aimed in particular at the detection of medium and heavy nuclides, relying on the high ion energy achievable with the ATLAS superconducting linear accelerator and on gas-filled magnet isobaric separation. The setup was recently used for the detection of the 146Sm process nuclide and for a new determination of the 146Sm half-life (68.7 My). AMS plays an important role in the measurement of stellar nuclear reaction cross sections by the activation method, extending thus the technique to the study of production of long-lived radionuclides. Preliminary measurements of the 147Sm(\(\alpha\),\(\gamma\))146Sm are described. A measurement of the 142Nd(\(\alpha\),\(\gamma\))146Sm and 142Nd(\(\alpha\),\(n\))145Sm reactions is in preparation. A new laser-ablation method for the feeding of the Electron Cyclotron Resonance (ECR) ion source is described.

1. Introduction

The activation method provides a powerful method of integral cross section measurements for nuclear astrophysics. While the method is mostly used by measuring the radiation emitted in the decay of a reaction product, accelerator mass spectrometry (AMS), through direct counting of the nuclides produced, is able to extend the measurements to cases where the half-life of the product is long (\(t_{1/2} > 10\) y) resulting in unmeasurable decay rates or where the radiation emitted with the decay is hard to detect (e.g. electron capture). In recent years, AMS has been widely used in nuclear physics and astrophysics [1]. We review in Section 2 the principle of accelerator mass spectrometry and its use for the determination of nuclear cross sections. In Section 3, we focus on reactions of importance to the understanding of \(p\)-process nuclide production. Preliminary AMS measurements of the 147Sm(\(\gamma\),\(n\))146Sm reaction cross section are described. (\(\alpha\),\(\gamma\)) reactions in the rare-earth region, shown to be sensitive to calculated abundances of \(p\)-process nuclides in this region [2], were studied recently [3,4] by \(\gamma\)- and characteristic X-ray decay counting and show systematic deviations from results of statistical calculations. The case of the 142Nd(\(\alpha\),\(\gamma\))146Sm reaction, in preparation for AMS measurements, is presented and a new technique of sample feeding by laser ablation is described for AMS analysis.

2. Accelerator mass spectrometry and measurements of nuclear reaction cross sections

Radioactive nuclides are detected at ultra-high sensitivity by the technique of accelerator mass spectrometry [5]. AMS is based on individual atom identification and counting, which competes favorably with decay counting (conventional \(\alpha\), \(\beta\), or \(\gamma\) spectroscopy) for half-lives long compared to...
Figure 1: Measurement of the cross section of the $^{26}\text{Mg}(p,n)^{26}\text{Al}(t_{1/2} = 0.72 \text{ My})$ reaction [6], an early application of accelerator mass spectrometry in nuclear astrophysics. The inverse reaction is a destruction process for the $^{26}\text{Al}$ nuclide. The reaction was studied by bombarding $^{26}\text{Mg}$ targets with a proton beam and, after chemical extraction of the $^{26}\text{Al}$ atoms with a stable Al carrier ($\text{Al}^*$) of known amount, the number of $^{26}\text{Al}$ product nuclei and the corresponding cross section was determined through the ratio $^{26}\text{Al}/\text{Al}^*$ measured by AMS.

typical laboratory measurement times. The quantity measured by AMS is the isotopic abundance of a specific rare nuclide relative to a stable isotope (or in case of actinides where stable isotopes do not exist, a neighboring nuclide taken as reference) and in favorable cases the isotopic abundance measured can be as low as $10^{-15}$-$10^{-16}$ with a typical precision of 2-10% (or governed by counting statistics), except for $^{14}\text{C}$ where dedicated facilities achieve subpercent precision [5]. The key in AMS to unambiguous ion identification leading to measurements of these ultra-low abundances is the use of fast accelerated ions: (i) the determination of the atomic number $Z$ of a detected ion by nuclear detection methods leads to discrimination (and sometimes physical separation) of isobaric (same $A$) species. Techniques employed rely on measurement of the differential energy loss $dE/dx$, of the range of an ion in matter or the average ionic charge state ($Z$-dependent) in a low-pressure gas; (ii) the elimination of stable (and possibly overwhelming) molecular ions having same mass number is achieved by stripping in gas or a thin foil (or as described below, in a high charge-state positive ion source), resulting in fragments which are either separated by magnetic and electrostatic analysis or eventually discriminated in the particle detector.

The analysis of these low isotopic abundances in samples with masses typically in the mg range leads to the capability of detecting unambiguously as few as $\sim 10^4$ rare atoms present in the sample without relying on their decay, a sensitivity unparalleled by other techniques. It should be emphasized that this capability is mostly limited to unstable species because chemical impurities usually cause unmanageable background in measurements of stable nuclides. The technique proves powerful for the measurement of nuclear reaction cross sections leading to unstable species. Fig. 1 illustrates the first application of AMS in nuclear astrophysics where the cross section of the reaction $^{26}\text{Mg}(p,n)^{26}\text{Al}$ was determined at energies just above threshold [6]; the inverse process is a destruction reaction of the important radioactive nuclide $^{26}\text{Al}(t_{1/2} = 0.72 \text{ My})$, observed in the last decade by $\gamma$-astronomy.
Figure 2: Identification spectrum of ions accelerated from a fast-neutron irradiated $^{147}$Sm sample (top) and a natural Sm sample (bottom). The $x$-axis represents distance along the focal plane of the gas-filled spectrograph and the $y$-axis is an energy loss signal. $^{146}$Sm produced by the $^{147}$Sm($n,2n$) reaction are observed, well separated from stable isobaric $^{146}$Nd and chemical impurities, transported by the accelerator due to $m/q$ degeneracy. The sensitivity limit determined by the background counts in the $^{146}$Sm region for the natural sample corresponds to an isotopic abundance in the range $^{146}$Sm/$^{147}$Sm $\approx 10^{-12}$.

Measurements of the cross section of nucleosynthesis reactions by AMS have been intensively performed in recent years, especially for $s$-process neutron capture reactions [1]. A few reactions induced by charged-particle have also been performed, e.g. $^{40}$Ca($\alpha,\gamma$)$^{44}$Ti [7], $^{33}$S($\alpha,p$)$^{36}$Cl [8]. The method provides an extension of the conventional activation method for products which are long-lived or whose decay radiation is difficult to detect. Importantly, the measurement is independent of the half-life of the reaction product or branching ratios in the product decay.

While most of the reactions studied via AMS were directed to light or medium mass nuclei, we presently use the Argonne ATLAS facility for the detection of heavier nuclides. The higher energies attainable allow us to extend AMS to the rare-earth region, still discriminating efficiently isobaric nuclides. A sample of the material to be measured is fed into the plasma of an Electron Cyclotron Resonance (ECR) ion source which produces highly-charged positive ions. The process eliminates already at this stage any molecular ions. Following magnetic analysis, the ions are accelerated by linear acceleration in ATLAS to energies of about 8 MeV/u and analyzed in a gas-filled magnetic spectrograph [9]. Ions are dispersed in the spectrograph according to $mv/\langle q\rangle$ where $v$ is the ion velocity (reduced by slowing down in the gas) and $\langle q\rangle$ is an average charge state along the ion trajectory in the gas, separating parasitic ions transported by the accelerator and isobaric nuclides. Fig. 2 shows an identification spectrum showing physical separation of $^{146}$Sm from stable isobaric $^{146}$Nd and from chemical impurities in the sample. The method was successfully used to detect the nuclide $^{146}$Sm and make a new determination of its half-life (68.7 My) [10].
3. \textit{p-process studies in the rare-earth region}

The $^{146}$Sm nuclide, produced like stable $^{144}$Sm by the \textit{p}-process, is particularly interesting in view of its presence in the Early Solar System, established by measurements of isotopic anomalies in its $^{142}$Nd alpha-daughter abundances in meteorites [11]. Unlike stable nuclides whose Galactic history cannot be established, a live radioactive nuclide must have been synthesized a few half-lives before the Solar system formation and as such, $^{146}$Sm had been recognized very early as a cosmochronometer of the \textit{p}-process [12]. The study, both experimental and theoretical, of nuclear reactions responsible for the production of \textit{p}-process nuclei in this region is under intense investigation [13]. It was noted for example that the rate of ($\gamma$,\textit{a}) reactions (or of the inverse ($\textit{a}$,\textit{\gamma}) reactions) impact sensitively on the calculated abundances of $\textit{p}$-process nuclides in the region $A >140$ and $N > 82$ [2]. Moreover, an experimental study of the $^{144}$Sm($\textit{a}$,\textit{\gamma})$^{146}$Gd reaction showed that the energy dependence of the reaction is hard to understand theoretically [14]. While Somorjai et al. [14] used energy-dependent alpha-nucleus optical potentials to reproduce the data, it was recently shown that taking into account coupled channels involving the Coulomb excitation of the nucleus does not require the use of special optical potentials [15].

We are presently applying the $^{146}$Sm detection technique described above to the study of nuclear reactions in this region of nuclides. The photonuclear reaction $^{147}$Sm($\gamma$,\textit{n})$^{146}$Sm was studied by irradiating an enriched $^{147}$Sm target by bremsstrahlung photons. The irradiation experiments were performed at Tohoku University and Kyoto University. Preliminary results for endpoint energies of 50 to 10 MeV, demonstrating the feasibility of the study, are shown in fig. 3.

The experimental study of the reactions $^{142}$Nd($\gamma$,\textit{a})$^{146}$Sm and $^{142}$Nd($\textit{a}$,\textit{n})$^{145}$Sm is in preparation. Fig. 4 shows the results of theoretical calculations for these reactions. Based on the background conditions shown in fig. 4, we estimate that an AMS measurement of $^{146}$Sm production could reach a cross section sensitivity of $\sim20$ mb, corresponding to incident energies down to $\sim11$ MeV. Test irradiations of a natural Nd target and an enriched $^{142}$Nd target (both as oxide Nd$_2$O$_3$, evaporated by Ar sputtering onto a 1.7 $\mu$m Al foil) were performed with an alpha particle beam of 13.5 MeV from the ATOMKI...
facility. Preliminary analysis shows the presence of \(^{145}\text{Sm}(t_{1/2} = 340 \text{ d})\) through \(\gamma\)- and characteristic X-ray activities of the \(^{145}\text{Pm}\) decay product, measured directly from the irradiated targets. After gamma counting, samples were prepared for AMS analysis of \(^{146}\text{Sm}\), using an ad-hoc procedure. The targets are dissolved in a HCl solution together with a known amount (typically 5 mg) of natural Sm carrier and, after transfer to a HNO\(_3\) solution, Al is separated in a chromatographic column of TRU resin.

Figure 4: Hauser-Feshbach statistical model calculations of alpha-induced reactions on \(^{142}\text{Nd}\): (left) NON-SMOKER code [16]; (right) SMARAGD code [17]. Calculations were performed (a) taking into account the set of 40 low-lying excited levels in the \(^{146}\text{Sm}\) compound nucleus from the NNDC compilation (2010) (labeled full NNDC levels) and (b) with no experimental levels, using a theoretical level density.

The Nd fraction of the residual solution is then carefully separated by a Ln-resin column in order to reduce as much as possible presence of isobaric \(^{146}\text{Nd}\) prior to the AMS measurement and the purified Sm fraction (containing \(^{145,146}\text{Sm}\) activation products) is finally eluted from the Ln column. After precipitation and ignition, the final residues are prepared as Sm\(_2\)O\(_3\) oxide samples for AMS. The measurement of the \(^{146,145}\text{Sm}/\text{Sm}\) ratio and the amount of Sm carrier by AMS determine the number of \(^{146,145}\text{Sm}\) products in the irradiations. It is important to note that this determination based on isotopic ratios is independent of chemical or detection efficiency. These samples are presently awaiting AMS analysis.

4. Laser-ablation accelerator mass spectrometry
In parallel, a new method of material feeding to the ECR ion source is developed using laser ablation of the solid sample [18]. Fig. 5 illustrates the layout of the ion source and the laser system. Laser ablation is performed with a 1064 nm Nd:YAG laser with a 15 ps pulse width, variable repetition rate up to 400 Hz and energy up to 5 mJ per pulse with a maximum power of \(3 \times 10^8\) W in the pulse. The target material resides inside the ECR plasma chamber in the injection end and the laser pulses enter the ECRIS through the beam extraction hole and are focused to a diameter of 0.5 mm at target. Target materials are ejected a few cm into the ECR plasma where they are ionized to a distribution of positive charge states. The ablation by the laser beam, well-focused on the sample surface, is expected to reduce the amount of contaminants entering the plasma compared to conventional methods of sample feeding by sputtering or oven evaporation. In a preliminary experiment using a reagent-grade samarium oxide (Sm\(_2\)O\(_3\)) sample, charge currents of \(^{147,152}\text{Sm}^{22,23+}\) in the nA range adequate for AMS analysis were successfully obtained. Figure 6 shows the spectrum of \(^{152}\text{Sm}\) ions produced by laser ablation and identified in the focal plane of the gas-filled spectrograph. In the conditions of the experiment, Sm ion rates are ~7-fold higher with laser ablation (laser on) than with laser off. The intensity of an observed \(^{126}\text{Xe}\) group (transported by the accelerator as \(^{126}\text{Xe}^{19+}\) ions due to a \(m/q\) near degeneracy with \(^{152}\text{Sm}^{23+}\) ions) attributed to residual atmospheric Xe in the ion source plasma chamber (at a pressure of \(10^{-7}\) Torr) is unchanged by laser irradiation of the Sm\(_2\)O\(_3\) sample.
Figure 5: Layout of the laser ablation setup at the ATLAS ECR ion source. The sample is selected and loaded in position at the back edge of the ECR plasma chamber by a multisample changer [19]. The laser beam is directed towards the sample through a zero-degree port of the analysis magnet onto the sample surface co-linearly with the exiting ion beam. Ablation by the fast (15 ps) pulsed laser at a typical energy of 3 mJ/pulse causes a plume of released atoms which penetrates the plasma and gets multiply ionized by the RF accelerated electrons.

5. Conclusion
Accelerator mass spectrometry extends the scope of activation measurements conventionally performed by decay counting to cases of long-lived products or otherwise practically undetectable decay radiations. Although generally more complex, the technique which measures directly the number of nuclides produced by a nuclear reaction, offers also the advantage of being independent of the product half-life or decay branching ratios. The further extension of AMS to heavier elements in the rare-earth region requires higher acceleration energies. The setup at Argonne National Laboratory is being developed for the study of $^{(\alpha,\gamma)}$ reactions, known to be particularly sensitive to the calculations of abundances of $p$-process nuclides in that region. The new technique of laser ablation feeding of sample material shows good promise for reduction of contaminants from the ion source and for use of smaller-size samples.

6. Acknowledgments
This work was supported by U.S. Department of Energy, Office of Nuclear Physics, under Contract No. DE-AC02-06CH11357 and by OTKA grant K108459.
Figure 6: Identification spectra collected in the gas-filled magnetic spectrograph collected with laser ablation on (top) and off (bottom) using a natural Sm$_2$O$_3$ sample. The group of $^{126}$Xe ions is attributed to residual atmospheric Xe in the ion source plasma and its intensity is unchanged by the laser irradiation while the output of Sm ions is enhanced ~7-fold.

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