COUNTING RARE ATOMS FOR NUCLEAR ASTROPHYSICS*

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Accelerator Mass Spectrometry (AMS) is today the most selective and sensitive analytical techniques stellar-nucleosynthesis for determination of isotopic abundances ranging down to below 10^{-15}. The principle and examples of the method are described. We now apply the technique to measurements of nuclear cross sections of stellar-nucleosynthesis reactions leading to long-lived nuclides. Recent measurements and experimental results for the important ^{62}Ni$(n,\gamma)^{63}$Ni ($t_{1/2}$ = 100 yrs) and ^{40}Ca$(\alpha,\gamma)^{44}$Ti (59 yrs) reactions are presented.

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1. Introduction

Accelerator Mass Spectrometry (AMS) is today the most selective and one of the most sensitive analytical methods for the determination of isotopic abundances in the sub-nano range. By isotopic abundance, we mean here the ratio of the number of atoms of a rare long-lived radionuclide to that of an abundant other species. The latter is usually a stable isotope of the same element or in some cases (*e.g.* for actinides) another nuclide taken as reference. The selectivity of the AMS technique is characterized by the unambiguous identification of both the mass number A and the atomic number Z of the radionuclide. The final result of an AMS measurement is therefore an abundance ratio $r = R/M$ between the number of a rare nuclide R to a major nuclide species M in a sample of material. The dynamic range of measurable ratios r which characterizes the sensitivity of AMS, lies typically between 10^{-7} to 10^{-15}, depending on the properties of the rare species detected. It is hard to grasp intuitively a ratio value as small as 10^{-15} and a concrete example may help to conceive its size: it is equivalent

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to the size of one grain of sand *versus* the area of 1000 km^2 of sand dune, as it exists for example in Namibia. This phenomenal sensitivity opens for AMS experimental opportunities in practically all areas of science by making possible measurements previously unachievable and in a sense, fulfilling Galileo Galilei's plea: "Measure what can be measured and make measurable what is not so".

As an example, and certainly not unique among AMS laboratories around the world, of the diversity of scientific fields which can be investigated by AMS, Table I lists the various radionuclides and fields which have been investigated at the Hebrew University [1], using principally the 14UD Pelletron Laboratory at the Weizmann Institute (Israel). In this talk, we review recent results in Nuclear Astrophysics where AMS is used to measure cross sections of important reactions leading to long-lived nuclides by counting the number of atoms produced in a laboratory experiment.

2. Accelerator Mass Spectrometry and nuclear cross section measurements

The AMS analytical method consists in the following methodological steps:

(a) *Ionization of the material*: the AMS analysis makes use almost exclusively of electrostatic tandem accelerators into which negative ions extracted from a material sample, usually in a Cs-sputter ion source [2], are injected. In some cases, the probability of negative-ion formation is exploited to suppress or reduce formation of contaminant isobaric ions. The use of positive ions for AMS is necessary in the case of elements whose electron affinity is negative, leading to unstable negative ions normally incompatible with tandem accelerators. In such cases (*e.g.* ^{39}Ar detection [4,5]) or when heavy ions need to be accelerated to very high energies for efficient isobaric discrimination (see below), high-charge state positive ions have been produced in Electron Cyclotron Resonance (ECR) ion sources and positive-ion accelerators (linac, cyclotron) used.

(b) *Acceleration and ion analysis*: a key feature of AMS is the acceleration of ions following their extraction and a first mass analysis. In tandem accelerators, acceleration is followed by an important stripping step where molecular ions are dissociated (one of the major advantages of AMS compared to conventional low-energy spectrometry) and ions are stripped of orbital electrons to positive ions and further accelerated. A magnetic and/or an electrostatic (or velocity) analysis follows the ion acceleration.

TABLE I

List of radionuclides detected and fields of applications studied at Hebrew University via Accelerator Mass Spectrometry (AMS) [1].

Radio-nuclide	Half-life (year)	Stable isobars	Negative ion	Sample matrix	Sens. ratio	Sens. (atoms)	Application
^{7}Be	0.15	^{7}Li	BeO^-	BeO	1×10^{-14}	4×10^{5}	nuclear astrophysics
^{10}Be	1.5 M	^{10}Be	BeO^-	BeO	1×10^{-14}	4×10^{5}	paleo-archeology
^{14}C	5730	^{14}N	C^-	C	1×10^{-14}	3×10^{4}	paleo-archeology
^{26}Al	720 k	^{26}Mg	Al^-	Al_2O_3	1×10^{-14}	1×10^{6}	geophysics, botany
^{36}Cl	301 k	^{36}Ar, ^{36}S	Cl^-	AgCl	1×10^{-15}	1×10^{6}	hydrology
^{41}Ca	105 k	^{41}K	CaH_3^-	CaH_2	1×10^{-15}	1×10^{7}	paleo-archeology, biomedicine
^{44}Ti	59.2	^{44}Ca	Ti^-	TiO_2	1×10^{-14}	1×10^{7}	nuclear astrophysics
^{59}Ni	760 k	^{59}Co	Ni^-	Metal	1×10^{-13}	1×10^{7}	cosmochemistry
^{90}Sr	28.5	^{90}Zr	SrH_3^-	SrH_2	1×10^{-13}	2×10^{7}	environmental science
^{129}I	15.7 M	^{129}Xe	I^-	AgI	1×10^{-14}	5×10^{5}	environmental science
^{236}U	23.4 M	None	UO^-	U_3O_8	1×10^{-11}	3×10^{8}	geophysics
^{236}U	23.4 M	None	UO^-	Fe_2O_3	—	1×10^{6}	environmental science
^{244}Pu	80.6 M	None	PuO^-	Fe_2O_3	—	4×10^{5}	cosmochemistry, astrophysics

(c) *Detection and identification*: the detection and counting stage is accompanied by identification of the ions by nuclear methods. Measurements of the stopping power dE/dx, the range or mean charge state in matter and of time of flight (together with the constraints on mass and charge set by the ion analysis system) determine the atomic number and mass number of the ion respectively, resulting in unambiguous ion identification. Most importantly, isobaric nuclides (ions of same mass number A whose relative mass differences are in the 10^{-5} range) are separated or discriminated at this stage. Fig. 1 shows an example of identification of ^{41}Ca (a long-lived Ca isotope, $t_{1/2} = 1.04{\times}10^5$ yrs [5]), produced cosmogenically in nature or artificially by neutron activation) at very high sensitivity, where artificially produced ^{41}Ca is used as a biomedical tracer to monitor Ca metabolism in a human patient [6]. The stable isobaric impurity ^{41}K is well discriminated by the detection system.

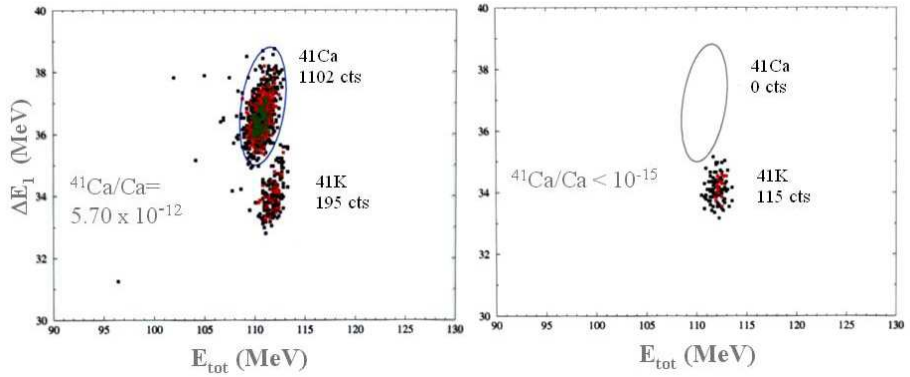

Fig. 1. Two-dimensional spectra of energy loss (ΔE_1) *versus* total ion energy $E_{\rm tot}$ measured for: (left) calcium extracted from a biomedical sample from a human patient into which a dose of ^{41}Ca ($t_{1/2} = 1.04 \times 10^5$ yrs) was injected as a tracer of Ca metabolism [6]. The long half-life of ^{41}Ca eliminates all radiation risks and allows indefinite monitoring of Ca; (right) natural calcium sample in which ^{41}Ca abundance is lower than 1×10^{-15}. One count in the ^{41}Ca window would correspond to an abundance ^{41}Ca/Ca $= 1 \times 10^{-15}$. Clear discrimination of the rare ^{41}Ca radionuclide from ^{41}K stable isobaric impurities is observed in the spectra.

Assuming an abundance of a rare nuclide in the $r = 10^{-13}$ range, typical of many AMS measurements and often surpassed, in a milligram sample ($\sim 10^{19}$ atoms for an element of medium mass number), the number of atoms which can be detected by AMS and quantitatively determined is of the order of $N_R = rN_M \sim 10^6$ atoms. This value allows us to determine the yield of a nuclear reaction well down to cross sections at the μbarn level, especially suited to low-energy astrophysical nucleosynthesis reactions, by

directly counting via AMS the number of nuclides produced in a nuclear activation experiment. This new method of nuclear cross section measurement is related to the conventional method of nuclear activation where the production cross section of a nuclide is determined by its decay activity but here, the number itself of nuclides produced is measured and not their decay. The following differences between the two methods are noticeable: *(i)* the half-life of the product is irrelevant in the AMS determination, extending the activation method to very long-lived nuclides which could not be measured by decay activity; *(ii)* in contrast with decay measurements, the AMS counting is independent of the decay scheme and branching ratios and of reaction mechanism (direct, resonance, compound nucleus formation); *(iii)* the AMS counting is however inapplicable in the case of a stable product whose production in a nuclear reaction cannot normally be discriminated from naturally occurring chemical or isotopic impurities. We present below recent measurements based on this technique for astrophysical nucleosynthesis reactions.

3. The weak s-process reaction ^{62}Ni(n,γ)^{63}Ni

The "weak s-process" path of neutron captures is believed to occur in the He-burning core region of a massive star. Neutrons, created there principally by the ^{22}Na(α,n)^{25}Mg reaction, thermalize at temperatures $T \sim 2{-}3{\times}10^8$ K ($\sim$ 30 keV)and are captured by heavy-element "seeds" of a previous star generation. The mechanism is responsible for synthesis of most elements with $60 < A < 90$. Theoretical nucleosynthesis calculations [7] show however an overestimate by a factor between 5 and 30 for nuclides in this mass region. The ^{62}Ni(n,γ)^{63}Ni ($t_{1/2}$ = 100 yrs) reaction behaves as one of the critical gateways for the weak s-process through which the whole process flows and experimental knowledge of this cross section is considered important to the quantitative understanding of heavy-nuclide synthesis in stars.

The steps of our measurement [8] consisted in:

(i) Activation of an enriched metallic ^{62}Ni target at Forschungszentrum Karlsruhe by neutrons produced by the ^{7}Li(p,n)^{7}Be reaction at an incident energy just above the reaction threshold. The neutron spectrum produced has been shown [9] to simulate well a flux of "quasi-stellar" neutrons in thermal equilibrium at a temperature of 25 keV. Activation of a target by these neutrons allows one therefore the direct measurement of an astrophysical Maxwellian-averaged cross section (MACS).

(ii) Measure the isotopic abundance $r = {}^{63}\mathrm{Ni}/{}^{62}\mathrm{Ni}$ by AMS. Due to the severe isobaric interference of ^{63}Cu impurities, unavoidable in an experimental system, the AMS experiment was performed at high energy

with the ATLAS superconducting linac at Argonne National Laboratory into which highly-charged positive Ni^{15+} were injected and accelerated to 9.2 MeV/u. In this configuration, any molecular ions are destroyed in the ECR process of high charge state ionization [10]. Isobaric separation was achieved using a gas-filled magnetic spectrograph where isobaric ions are spatially dispersed due to their different mean ionization charge state during the passage through a low-pressure gas [11]. Fig. 2 illustrates the isobaric ion identification spectrum from which the counting of the ^{63}Ni ions was performed in a set of repeat measurements, leading to a laboratory cross section (averaged over the quasi-stellar neutron spectrum $n(E)$) of

$$\sigma_{\rm exp} = r/\langle \Phi_{\rm exp} t\rangle = \int_0^{E_\infty} \sigma(E) n(E) dE / \int_0^{E_\infty} n(E) dE = 23.9 \pm 2.3 \text{ mb}\,,$$

where $\langle \Phi_{\rm exp} t\rangle$ denotes the neutron fluence, monitored during the activation.

(iii) The MACS $\langle\sigma\nu\rangle/\nu_T$ is finally derived from the measured value $\sigma_{\rm exp}$ by the expression $\langle\sigma\nu\rangle/\nu_T = (2/\pi^{1/2}) f_{\rm inf}\sigma_{\rm exp}$ [9], where $f_{\rm inf}$ expresses a correction to the laboratory cross section for energies $E > E_{\rm co}$ above the maximum energy of the neutron spectrum. The value of $f_{\rm inf}$, calculated with the JENDL neutron cross section library [12] is 0.95 ± 0.01. Our final value of the MACS of ^{62}Ni is 25.5 ± 2.6 mb at 25 keV (experimental neutron spectrum) and 20.9 ± 2.1 mb after extrapolation to 30 keV (using again the JENDL energy dependence [12]).

This result has been confirmed by two recent experiments, using a time-of-flight [13] and an independent AMS activation [14] measurement technique, respectively. As shown in [8], the value of the ^{62}Ni(n,γ)^{63}Ni reaction impacts very strongly on the yield of the weak s-process.

4. The explosive-nucleosynthesis ^{40}Ca(α,γ)^{44}Ti reaction

The interest in the explosive-nucleosynthesis ^{44}Ti ($t_{1/2} = 59$ yrs) nuclide has sprung in the last ten years after the observation of its live radioactivity from the Cas A supernova (SN) remnant by various γ-ray space observatories [15,16]. The observed ^{44}Ti is produced during the expansion stage of the core-collapse SN during multiple α captures. It poses presently an interesting challenge to SN hydrodynamical and nucleosynthesis models because the yield (160 ± 60 μSolar Mass) deduced from the γ-photon flux (using the known distance of Cas A from Earth and the ^{44}Ti laboratory half-life) is

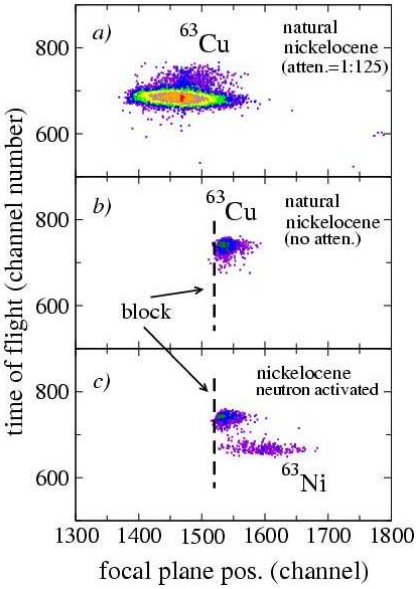

Fig. 2. Time-of-flight *versus* position in the focal pane of the gas-filled Enge split-pole spectrograph for ^{63}Ni and ^{63}Cu impurities, extracted from: (a) and (b) a natural Ni sample; (c) the quasi-stellar-neutron activated sample (see text). The sample used was chemically converted to the organo-metallic compound nickelocene which proves to reduce Cu interference in the ECR ion source [8]. The separation observed along the focal plane (horizontal axis) between ^{63}Ni and ^{63}Cu is due to the gas-filled magnetic spectrograph action on the ions having different mean charge state. In (b) and (c), the extremely intense Cu group is partly blocked by a Ta stopper whose position along the focal plane is adjustable.

much larger than estimated theoretically. One of the key reactions entering the estimate and the one directly responsible for ^{44}Ti SN production is the α capture ^{40}Ca(α,γ)^{44}Ti reaction. Experimental knowledge of this reaction was limited to the identification of several resonances [17] in the range of relevant energies. We initiated the measurement of this cross section, at energies ranging to lower energies, by AMS counting of ^{44}Ti nuclei.

The activation reaction was performed in reverse kinematics by bombarding a gas cell filled with high-purity He with an intense ^{40}Ca ion beam [18]. The activation was performed in two different runs (at different energies) at the ATLAS linac (Argonne) and at the 14UD Pelletron laboratory (Weizmann Institute). In both experiments, fast ^{44}Ti ions forward-recoiling from the α capture are implanted in a Cu catcher which serves also as beam stop to the incident ^{40}Ca beam. After activation, the catcher is chemically etched in a HNO_3 solution containing a known amount n_c of natural Ti, acting as

carrier for the ^{44}Ti implanted atoms. The number n_{44} of ^{44}Ti produced in the activation is then determined by the relation $n_{44} = r_{44}n_{\rm c}$ where r_{44} is the ^{44}Ti/Ti abundance measured by AMS. Importantly, n_{44} is independent of efficiencies of either chemical processing or accelerator transmission during the AMS measurement since only isotopic ratios are involved. Fig. 3 shows identification spectra of ^{44}Ti produced in the α activations in different energy ranges. The cross section of the ^{40}Ca(α,γ)^{44}Ti reaction, averaged over the energy range determined by the thickness and specific energy loss dE/dx of the ^{40}Ca beam in the He gas target, is directly derived from n_{44} and determined as 8.0 ± 1.1 μb over the range $E_{\rm cm} = 0.58$–1.16 MeV/u.

The above experimental value of the average cross section is larger by a factor 2 to 2.5 than derived from previously known resonances and indicates the existence of either additional resonances or closely spaced energy states in ^{44}Ti. Additional resonances have indeed been identified in a recent experiment [19] performed at TRIUMF in which ^{44}Ti recoils from a ^{4}He gas target bombarded by a ^{40}Ca beam are analyzed and detected online in the DRAGON recoil mass spectrometer. Both experiments support the conclusion that the ^{44}Ti yield in explosive nucleosynthesis is larger than calculated in estimates based on the previous experimental information. The new yields are yet smaller than needed to account for that of the Cas A SN and it seems that other physical processes (*e.g.* highly aspherical explosions) must have contributed in this case.

5. Perspectives and conclusion

We are presently engaged in a program of detection of the ^{146}Sm p-process nuclide, one of the heaviest nuclide to be detected and separated from its stable isobar. ^{146}Sm ($t_{1/2} = 1.03 \times 10^8$ yrs), belonging to a family of nuclides present in the Early-Solar System and today extinct, is an important geochronometer for the Solar-System and planetary formation. Study of the cross sections of reactions leading to ^{146}Sm are relevant to the study of the p-process in this region of nuclides and will be useful to determine α-nucleus optical potentials at low energies, for which experimental information is very scarce.

Selectivity and efficiency are two complementary properties whose interplay determines the sensitivity of an analytical technique at ultra-trace levels. Accelerator mass spectrometry is at present unsurpassed among existing analytical methods in its selectivity between ion species and belongs to a family of techniques which, owing to their sensitivity, continuously drive research to new horizons. Laboratory measurements of astrophysical processes and phenomena require and successfully exploit the development of such advanced experimental methods.

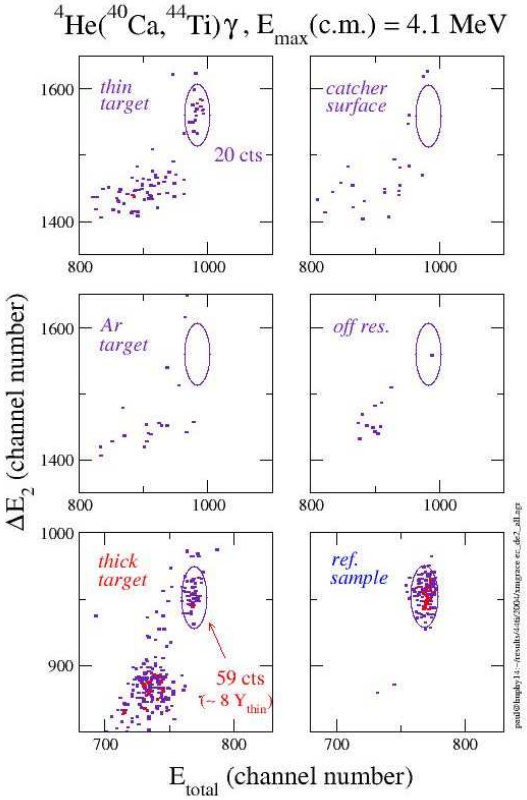

Fig. 3. Identification spectra of ^{44}Ti produced via the ^{40}Ca(α,γ) ^{44}Ti reaction in reverse kinematics by ^{40}Ca bombardment of a He gas target at an incident energy of $E_{\rm cm}$ = 4.2 MeV and implanted in a Cu catcher (see text). Thin and thick target refer to He gas thicknesses which integrate the yield over strong closely spaced resonances at $E_{\rm cm} \sim 4.1$ MeV (top left) or over the range $E_{\rm cm}$ = 4.2 to 2.1 MeV (bottom left), respectively. No ^{44}Ti are observed when etching was done only through a shallow depth (top right) below catcher surface ($\leq 2\mu$m), showing deep implantation as expected ($\sim 5\mu$m) from the ^{44}Ti recoil energy, or for Ar gas replacing the He target (middle left). Bombarding at an energy off the strong resonances (middle right) shows a yield an order of magnitude smaller than the on-resonance measurement (top left). The bottom right spectrum corresponds to a sample of known ^{44}Ti abundance used as reference.

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