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Nuclear Data from AMS & Nuclear Data for AMS – some examples

A. Wallner^{1,2}, M. Bichler³, T. Belgya⁴, K. Buczak¹, I. Dillmann⁵, O. Forstner¹, R. Golser¹, F. Käppeler⁶, A. Klis⁷, G. Korschinek⁸, A. Krasa⁹, W. Kutschera¹, C. Lederer¹, A. Mengoni¹⁰, M. Paul¹¹, A. Plompen⁹, A. Priller¹, V. Semkova¹², P. Steier¹

¹ VERA Laboratory, Faculty of Physics, University of Vienna, Währinger Strasse 17, A-1090, Austria

² Department of Nuclear Physics, Australian National University, Canberra, ACT 0200, Australia

³ Atominstitut, Vienna University of Technology, Austria

⁴ Dep. of Nuclear Research, Institute of Isotopes, Hungarian Academy of Sciences

⁵ GSI Helmholtzzentrum für Schwerionenforschung GmbH, Planckstraße 1, D-64291 Darmstadt, Germany

⁶ Karlsruhe Institute of Technology (KIT), Campus Nord, Institut für Kernphysik, Karlsruhe, Germany

⁷ Institut für Kern- und Teilchenphysik, TU Dresden and FZ Rossendorf, Germany

⁸ Maier-Leibnitz-Laboratory, Faculty of Physics, TU Munich, James-Franck-Strasse, Garching, Germany

⁹ European Commission, Joint Research Centre, Institute for Reference Materials and Measurements, Geel, Belgium

¹⁰ Agenzia nazionale per le nuove tecnologie, l'energia e lo sviluppo economico sostenibile (ENEA), Bologna, Italy

¹¹ Racah Institute of Physics, Hebrew University, Jerusalem, Israel

¹² NAPC Nuclear Data Section, International Atomic Energy Agency, A-1400 Vienna, Austria

Abstract. We summarize some recent cross-section measurements using accelerator mass spectrometry (AMS). AMS represents an ultra-sensitive technique for measuring a limited, but steadily increasing number of longer-lived radionuclides. This method implies a two-step procedure with sample activation and subsequent AMS measurement. Applications include nuclear astrophysics, nuclear technology (nuclear fusion, nuclear fission and advanced reactor concepts and radiation dose estimations). A series of additional applications involves cosmogenic radionuclides in environmental, geological and extraterrestrial studies. There is a lack of information for a list of nuclides, as pointed out by nuclear data requests. An overview of some recent measurements is given and the method is illustrated for some specific neutron-induced reactions.

1 Introduction

Nuclear data – i.e. nuclear reaction and nuclear structure data – are an essential ingredient for nuclear based research. Production of radionuclides through nuclear reactions needs to be well-known for many applications. Cross sections are one basic nuclear property. They are of importance in nuclear physics applications (nuclear technology), space technology, nuclear astrophysics (nucleosynthesis, cosmo-chemistry, meteorites) and medical applications (hadron therapy, radiation dose measurements). Moreover, production rates are *the* quantity for cosmogenic nuclides, which are utilized in a wide range of applications: for ¹⁴C-dating, geological and environmental studies. Depending on the nuclide and research topic, particle energies between thermal and up to GeV's are relevant. As a consequence different types of nuclear reactions, from thermal neutron capture to heavy ion spallation reactions might be the dominant reaction channels.

For specific reactions accelerator mass spectrometry (AMS) offers a powerful tool to measure cross sections. The advantage compared to decay counting is independent of the half-lives of the reaction products. The combination of activation and subsequent AMS measurement was applied for a range of measurements where off-line decay counting is difficult or impossible due to long half-lives of reaction products or due to the absence of suitable γ -ray transitions. The interested reader will find a comprehensive summary of AMS in nuclear physics and astrophysics up to 1990 in [1] and in the subsequent AMS proceedings [2]. AMS was also used for measuring half-life values of long-lived radionuclides [3,4]; and also for the search of super-heavy elements [1,5,6] and for other exotic rare nuclides [1].

In this paper we will focus on nuclear reaction data and in particular on cross-section measurements. In the following we will restrict ourselves to the production of long-lived radionuclides, i.e. where the technique of AMS has its great advantage: AMS represents an ultra-

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sensitive technique for measuring a limited, but steadily increasing number of longer-lived radionuclides. The advantage compared to conventional mass spectrometers like TIMS, ICPMS, is its unsurpassed sensitivity for the abundance of specific nuclides.

Counting atoms directly via mass separation and particle acceleration with charge exchange completely suppresses molecular interference and for a few cases isobaric interference is completely excluded as well, and hence highest sensitivities are obtained. A disadvantage is that AMS needs often elaborate sample preparation and is a costly technique; and like all mass spectrometric techniques it is sample destructive.

2 Comparison of ‘activation + AMS’ with other techniques

In general, cross section measurements can be classified into two complementary techniques: online and offline methods (see Fig. 1). The online method makes use of the detection of the prompt and characteristic radiation associated with the production of a specific nuclide, or selectively detects the reaction product itself by means of the recoil separator technique. Typical examples of experimental facilities in this “direct mode” are, -among many others- e.g. the DRAGON setup at TRIUMF (combined with recoil separator technique [7]) and LUNA at Gran Sasso [8] for charged particle induced reactions; and e.g. in Europe GELINA [9] and the n_TOF facility at CERN [10] for studying neutron-induced reactions.

The second and independent offline method makes use of the activation technique, with sample irradiation and subsequent measurement of the reaction product. After the irradiation the number of produced radioactive nuclei can be quantified either by decay-counting or by mass spectrometric methods. This method is mostly restricted to radioactive products; however, it represents a very sensitive technique due to potential long irradiation periods. Long-lived radionuclides have often been inaccessible to decay counting techniques, e.g. because of low activity or an unfavorable decay scheme.

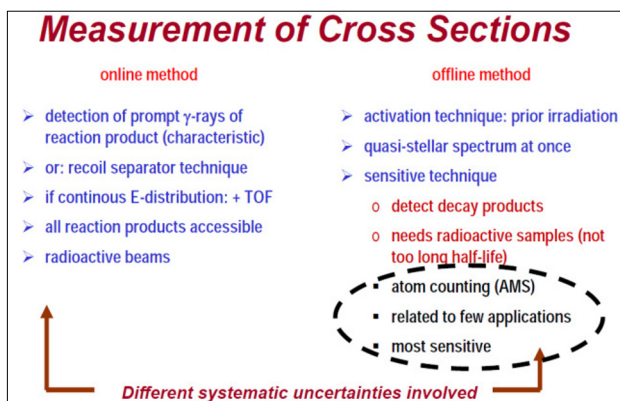


Figure 1: Comparison of online and offline methods. Quasi-stellar spectra are used in nuclear astrophysics (see 4.1).

AMS as an off-line method represents an independent and complementary method to online measurements. A

comparison of AMS data allows studying systematic contributions to the total uncertainty associated e.g. with online methods, which otherwise are hard to quantify.

3 Sample activation & subsequent AMS - a tool to measure cross sections

Due to our interest in long-lived reaction products we do not face any limitations from radioactive decay during typical sample activations. Neutrons, protons, α particles or γ 's etc. are usually produced at dedicated irradiation facilities. However, in some cases reactions were studied in inverse kinematics where a beam of heavy ions is directed into a gas cell containing the lighter reaction partner (e.g. He) [11,12]. Usually, after activation, the irradiated sample has to be converted into a chemical form to suit the subsequent AMS measurement. If the reaction product is of the same chemical element, the isotope ratio of produced radionuclide and the stable target nuclide can directly be measured with AMS. In case of producing a different element, however, a well-known amount of spike (a stable nuclide of the same element as the freshly produced radionuclide) needs to be added and relative to the spike the unknown radionuclide is measured again as isotope ratio.

Neglecting radioactive decay, and with N_r the number of produced radionuclides and N_0 the number of target nuclides in the sample, the isotope ratio N_r/N_0 , generated in an activation experiment is directly related to the cross section σ for a specific reaction and to the neutron fluence Φ

$$\frac{N_r(E)}{N_0} = \sigma_E \cdot \Phi_E$$

This equation is true for a specific particle energy E . If the particle energy is not mono-energetic (see Fig. 2) the integral cross section over the experimental energy distribution is obtained, i.e. a spectrum-averaged cross section will be measured. As an example, the neutron capture reaction $^{54}\text{Fe}(n,\gamma)^{55}\text{Fe}$ can be written as

$$\sigma_E(n,\gamma) = \frac{^{55}\text{Fe}(E)}{^{54}\text{Fe}} \cdot \frac{1}{\Phi_E}$$

This equation contains two simple quantities for calculating the cross-section value: (1) the isotope ratio which is measured in our case using AMS; and (2) the particle fluence, which can either be monitored as a current in case of charged-particle induced reactions, or relative to a well-known monitor reaction or also through the target's intrinsic activity.

AMS is a relatively new method for measuring cross sections. Paul and co-workers first introduced AMS to cross-section measurements in nuclear physics for the $^{26}\text{Mg}(p,n)^{26}\text{Al}$ reaction [13]. Since about 15 years a boost in measurements for nuclear physics and astrophysics applications was observed. AMS laboratories with an active program within the last decade for cross-section measurements include TU Munich [14], ETH Zurich [15], Caserta [16], ANL [17], Purdue [18], Notre Dame [19], the ANU [20], CIAE [21] and the VERA facility [22] at the Univ. of Vienna.

4 Cross section measurements

In the following some applications are listed which (1) either use AMS for measuring nuclear data, or (2) where nuclear data of radionuclides are essential input parameters required for AMS applications. We will summarize and exemplify this technique for some recent cross-section measurements – largely performed at the Vienna Environmental Research Accelerator (VERA) laboratory. We will restrict ourself mainly to neutron-induced reactions. In section 4.4 and 4.5 when discussing applications that require high particle energies, reference to additional recent work is given. Such a selection of AMS measurements can only be a small excerpt of the overall number of AMS-cross section measurements. For additional work the reader is referred to complementary publications, e.g. [1,2, 11-21, 23-26].

We have studied neutron-induced reactions for applications in nuclear astrophysics (see section 4.1), nuclear fusion (sec. 4.2) and fission (sec. 4.3). Advanced nuclear systems (sec. 4.4) and production of cosmogenic nuclides (sec. 4.5) require knowledge of high-energy reaction data, which is often solely based on pure calculations, however, some experimental programs were dedicated for such applications [27,28].

Table 1. List of *neutron* producing facilities with subsequent AMS measurements performed recently

irradiation facility	research area	neutron energy range
KIT	NA, NT	25 – 500 keV
IKI	NP, NT	thermal, cold
IRMM	NT, AA	0.5 – 22 MeV
TUD	NT, AA	13.4 – 14.8 MeV
ATI	NP, NT	thermal

Abbrev: Karlsruhe Institute of Technology (KIT, Germany), Center for Energy Research (IKI Budapest, Hungary), Institute for Reference Materials and Measurements (IRMM, Belgium), TU Dresden (TUD, Germany) Atominsttit (ATI, TU Vienna, Austria); research areas comprise nuclear astrophysics (NA), nuclear physics properties (NP), nuclear technology (NT) and AMS applications (AA).

For our studies, thermal neutrons and cold neutrons were produced at the reactors at the Atominsttit of the Vienna University of Technology [29], and the Budapest Research Reactor (IKI, Institute of Isotopes, Hungarian Academy of Sciences) [30]. Neutrons with an energy distribution peaking at 25 keV were produced through the ${}^7\text{Li}(p,n)$ reaction with protons with energy closely above the reaction threshold ($E_p=1912$ keV, see [31]). The experimental neutron spectrum (see Fig. 2) reported by [31] is a good approximation to our experimental conditions. This neutron spectrum produced from the ${}^7\text{Li}(p,n)$ reaction was recently confirmed in new measurements [32, 33] which is of great importance for nuclear astrophysics (see sec. 4.1). Higher neutron energies, usually with a broad energy spread, were

produced from higher-energetic protons. Cross section measurements via neutron activations in the fast neutron energy range were performed in cooperation with TU Dresden (TUD), utilizing their neutron generator [34], i.e. producing 14-MeV neutrons via the (d,T) reactions; and higher energy neutrons were produced via the Van de Graaff facility at IRMM, Geel, [35] with neutron energies between 13 and 22 MeV. Via the T(p,n) reaction neutrons with energies between 0.6 and 3 MeV and via (d,d) neutrons up to 5 MeV energy were generated at IRMM as well. Simultaneous irradiations of samples at different angles allowed utilizing different neutron energies.

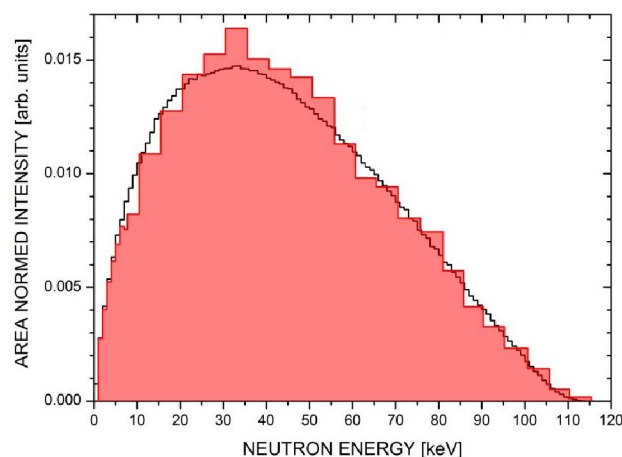


Figure 2: Example of neutron energy distribution produced at KIT for nuclear astrophysics applications. The black line is a calculation based on the specific irradiation geometry [36], the histogram is a measured spectrum by Ratynski & Käppeler [31].

Depending on the cross-section value, isotope ratios of radionuclide to the target nuclide between 10^{-10} and 10^{-15} were produced in such activations. After the irradiations, the subsequent AMS measurements were performed at AMS facilities, like the VERA facility in Vienna or at TU Munich.

4.1. Nuclear Astrophysics

Nuclear data are a key ingredient for nucleosynthesis calculations and for our understanding of the chemical evolution in our galaxy. AMS is a relatively new method for measuring cross sections of relevance to nucleosynthesis. It was first applied to laboratory experiments in nuclear astrophysics some ten years ago for studying the production of ${}^{44}\text{Ti}$ from ${}^{40}\text{Ca}(\alpha,\gamma)$ [37,38], of ${}^{26}\text{Al}$ via ${}^{25}\text{Mg}(p,\gamma)$ [39] and for neutron capture reactions in the regime of s-process environments, e.g. ${}^{62}\text{Ni}(n,\gamma)$ [40]. More specifically, AMS has been applied for neutron-capture studies of ${}^{40}\text{Ca}$, ${}^{62}\text{Ni}$, ${}^{58}\text{Ni}$, and ${}^{78}\text{Se}$ [41-43] and in a series of recently completed measurements of ${}^9\text{Be}$, ${}^{13}\text{C}$, ${}^{35}\text{Cl}$, ${}^{54}\text{Fe}$, ${}^{209}\text{Bi}$ and of ${}^{14}\text{N}(n,p)$ [44-46]. All these neutron-induced measurements made use of “keV neutrons” (see Fig. 2) which were produced at Karlsruhe Institute of Technology (KIT) [31] via the ${}^7\text{Li}(p,n){}^7\text{Be}$ reaction. In a proper irradiation geometry a sample can be irradiated

with neutrons whose integrated cross section is directly related to a Maxwell-Boltzmann averaged cross section at an effective temperature of 25–30 keV (quasi-stellar spectrum) [31-33]. Such a setup was developed at KIT for direct cross section measurements and for activations with subsequent decay counting of the produced activities. In this way a comprehensive set of measurements relevant to s-process nucleosynthesis was performed at KIT [47]. We also produced protons with higher energies as compared to the 1912 keV energy required for “25 keV” (quasi-stellar) cross sections. In this way neutrons with correspondingly higher energies were generated, for our studies up to 500 keV.

4.2. Nuclear Technology - Fusion

The production of long-lived radionuclides as activation products is of concern for a fusion environment since they may lead to significant long-term waste disposal and radiation damage [48-52]. For such nuclides production cross-sections, total induced activities and the knowledge of their decay pattern are key parameters for safety and design analyses. Many of these production cross sections are not well-known, making it difficult to calculate concentration limits. Some prominent long-lived activation products comprise ^{10}Be , ^{14}C , and ^{26}Al ; in the medium-mass range the radionuclides ^{53}Mn , $^{55,60}\text{Fe}$, $^{59,63}\text{Ni}$; and for heavier isotopes $^{202\text{m}}\text{Pb}$, $^{210\text{m}}\text{Bi}$. Since a few years we have applied AMS for measuring such production cross sections as this long-lived reaction products correspond to the radionuclides measured in AMS.

In a recent measurement program, samples were irradiated with quasi-monoenergetic neutrons at TU Dresden's 14-MeV neutron generator and at the Van de Graaff accelerator at IRMM simulating a fusion environment. After the activations the samples were prepared for isotope ratio measurements via AMS. Production of long-lived ^{53}Mn and ^{59}Ni was measured via AMS utilizing the 14-MV tandem of the Maier-Leibnitz-laboratory, TU Munich. Radionuclides ^{10}Be , ^{14}C , ^{26}Al ,

^{55}Fe , $^{210\text{m}}\text{Bi}$, and $^{202\text{g}}\text{Pb}$ were measured at the VERA facility [53].

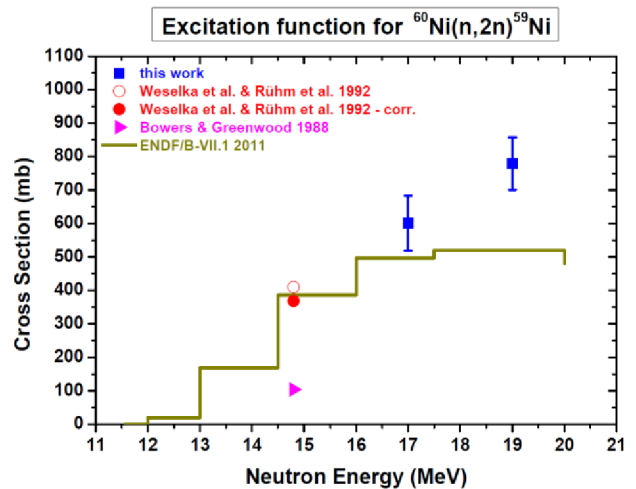


Figure 3: Comparison of AMS data (this work) with previous work [48,54] and a recent evaluation ENDF/B-VII.1 (line) [55].

One of those radionuclides is ^{59}Ni with a half-life of 76,000 years. Its main production in a fusion environment is the (n,2n) reaction on stable ^{60}Ni with some additional contributions from neutron capture reactions on ^{58}Ni via lower energy neutrons. Experimental data for the production of ^{59}Ni via the (n,2n) reaction are strongly discordant or completely missing (prior to our work only two, but discrepant data at 14.8 MeV were available, see Fig. 3) [48,54]. We have performed new measurements utilizing AMS at the TU Munich and plan new measurements at the ANU in the energy range between 14 and 20 MeV. First results are plotted in Figure 3 (squares). Our experimental data at 17 and 19 MeV neutron energy indicate an increasing excitation function up to 19 MeV, which is not expected from the semi-theoretical approach in the evaluated data-file ENDF [55]. Thus, we expect a higher production of long-lived ^{59}Ni in a fusion environment than previously indicated.

4.3. Nuclear Technology - Fission

Table 2. Summary of recent activations at IKI [30], KIT (Karlsruhe Institute of Technology, [31] and IRMM [35] for the study of neutron-induced reactions of $^{\text{nat}}\text{Th}$ and $^{\text{nat}}\text{U}$ samples via neutron activation and subsequent AMS measurements at VERA.

sample	reaction	product	facility	E_n	AMS-nuclide	$t_{1/2}$ (AMS-nuclide)
$^{\text{nat}}\text{U}$	$^{238}\text{U}(n,\gamma)$	^{239}U	IKI, KIT	thermal, 25, 426 keV	^{239}Pu	24100 yr
$^{\text{nat}}\text{U}$	$^{238}\text{U}(n,\gamma)$	^{239}U	IRMM	0.5 – 3 MeV	^{239}Pu	24100 yr
$^{\text{nat}}\text{U}$	$^{235}\text{U}(n,\gamma)$	^{236}U	IKI, KIT	thermal, 25, 426 keV	^{236}U	23.4×10^6 yr
$^{\text{nat}}\text{U}$	$^{235}\text{U}(n,\gamma)$	^{236}U	IRMM	0.5 – 3 MeV	^{236}U	23.4×10^6 yr
$^{\text{nat}}\text{U}$	$^{238}\text{U}(n,3n)$	^{236}U	IRMM	17 – 21 MeV	^{236}U	23.4×10^6 yr
$^{\text{nat}}\text{Th}$	$^{232}\text{Th}(n,\gamma)$	^{233}Th	IRMM	0.5 – 3 MeV	^{233}U	1.59×10^5 yr
$^{\text{nat}}\text{Th}$	$^{232}\text{Th}(n,2n)$	^{231}Th	IRMM	17 – 21 MeV	^{231}Pa	32760 yr
$^{\text{nat}}\text{Th}$	$^{232}\text{Th}(n,4n)$	^{229}Th	IRMM	17 – 21 MeV	^{229}Th	7880 yr

Improved and highly accurate nuclear data are urgently required for the design of advanced reactor concepts. This demand holds for minor actinides but also for the main fuel materials. Existing data for neutron induced reactions have been obtained e.g. by time-of-flight techniques via detection of the prompt capture γ -rays. A difficulty in experiments on actinides is the safe discrimination against the strong γ -background from the competing fission channel. If the reaction product is radioactive, decay counting techniques can be applied for measuring the amount of produced radionuclides. In case of long-lived reaction products direct counting techniques in combination with neutron activation might be utilized. Such long-lived radionuclides might either be the direct product of a reaction, or a decay product of a directly produced short-lived nuclide.

Recent measurements of neutron-induced reactions on actinides by combining neutron activation and subsequent AMS measurements at the VERA laboratory [22] were performed within the European EFNUDAT [56] and EUFRAT programs [57]: neutron activations were performed at neutron producing facilities at KIT [31], IRMM [35], and the Center for Energy Research, Budapest (former IKI) [30]. As shown in Table 2 neutron capture studies on the main actinides ^{232}Th , ^{235}U and ^{238}U are performed applying AMS: in case of $^{235}\text{U}(n,\gamma)^{236}\text{U}$ no chemistry is required and the material can be directly used in AMS for $^{236}\text{U}/^{235}\text{U}$ isotope ratio measurements. Some long-lived radionuclides listed in Table 2 are decay products of the directly produced short-lived nuclides: e.g. ^{239}Pu and ^{233}U from ^{239}U and ^{233}Th , respectively. For a quantitative measurement, ^{239}Pu and ^{233}U had to be separated from the $^{\text{nat}}\text{U}$ and $^{\text{nat}}\text{Th}$ bulk material by dedicated actinide chemistry.

Within EFNUDAT two projects were performed with the goal to determine simultaneously the neutron capture cross sections of ^{235}U and ^{238}U from $^{\text{nat}}\text{U}$ samples [58] via neutron irradiations at thermal (cold) and keV-neutrons: ^{235}U and ^{238}U were activated in well defined, intense neutron fields at the Van de Graaff accelerator of KIT and the Budapest Research Reactor. The reaction product ^{236}U and the decay product of ^{239}U , ^{239}Pu , were subsequently counted by AMS at VERA.

Within EUFRAT two additional projects with irradiations at IRMM are underway for studying (1) neutron capture on ^{232}Th and ^{238}U with neutron energies between 0.5 and 3 MeV; and (2) fast neutron induced reactions on $^{\text{nat}}\text{Th}$ and $^{\text{nat}}\text{U}$ samples (see Tab. 2). Different neutron energies were achieved from simultaneous irradiations at several angles relative to the incoming beam.

The AMS measurements are finished for those samples activated with lower neutron energies (thermal to 426 keV), and are in progress for the samples irradiated with energies between 0.5 and 3 MeV. The fast neutron-induced reactions were studied at different AMS laboratories (ETH Zurich, ANSTO/Sydney, VERA/Vienna). These measurements are finished as well and data analysis is in progress now.

4.4. Nuclear Technology - Advanced nuclear systems

Concepts for hybrid reactors, e.g. accelerator-driven systems or accelerator-driven transmutation of nuclear

waste, combine a ‘powerful’ particle accelerator with a reactor. Particles with energies between a few hundred MeV and GeV will be impinging on a spallation target. For such systems nuclear data are required for much higher energies than in conventional systems. Note, similar particle energies are also obtained with particle accelerators which are in use for hadron therapy in cancer treatments. Clearly, in this energy region there is need for more experimental data. Therefore, dedicated research programs were initiated, e.g. HINDAS (High and Intermediate energy Nuclear Data for Accelerator-driven Systems, supported by the European Commission), to obtain a thorough understanding and complete modelling of nuclear reactions in the 20-2000 MeV region. To achieve this, an ambitious experimental and theoretical programme was launched which included also a series of AMS measurements for quantifying production of long-lived radionuclides from spallation [59, 60].

4.5. Environmental, geological and extra-terrestrial applications

The concentrations of rare radionuclides in our environment provide unique information. The measurement of such radionuclides allows tracing anthropogenic activities or environmental processes. Man-made radionuclides will enter the environment via different processes, e.g. from nuclear weapons tests, as accidental local fallout products, from nuclear-fuel reprocessing plants, or from industrial or medical applications. Production in anthropogenic processes needs to be understood which allows distinguishing their signature from cosmogenic or in-situ production.

Cosmogenic nuclides are produced via interactions of primary and secondary cosmic rays with matter, i.e., meteorites, planetary surfaces, interstellar dust particles, and planetary atmospheres. As such data are needed for p-, n-, α - and μ -induced reactions. For these studies the particle flux, particle energy spectra and the cross-sections are a basic ingredient. Reaction mechanisms include low-energy neutron capture, but spallation reactions become dominating for higher energetic particles (MeV and above).

Only very few experimental data are available, in particular for neutron-induced reactions, mainly due to the difficulty to generate well-defined and intense beams for nuclear reaction experiments. Exemptions were dedicated measurement campaigns as e.g. listed in Ref. [24-28, 59, 60] (see also ref. therein). Recently, neutron beam-lines with quasi mono-energetic neutrons in the energy range up to 200 MeV became available, (see [61-63]).

5. Summary

The combination of activation and subsequent AMS detection of long-lived radionuclides represents a complementary method to online particle detection techniques and also to conventional decay counting. Sample activation in combination with AMS depends on different systematic uncertainties compared to these other techniques. It has been applied for cross section measurements for applications ranging from nuclear astrophysics, nuclear physics and nuclear technologies to those utilizing cosmogenic nuclides.

The advent of new powerful irradiation facilities, e.g. FRANZ (Frankfurt Neutron Source at the Stern-Gerlach Zentrum [63]), SARAF (Soreq Applied Research Accelerator Facility, [64]) or powerful reactors [17] will provide new exciting possibilities for AMS.

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