Potential of AMS for Quantifying Long-Lived Reaction Products

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Abstract. Accelerator mass spectrometry (AMS) represents a powerful technique for the detection of long-lived radionuclides through ultra-low isotope ratio measurements. In many cases, counting atoms rather than decays yields much higher sensitivities. The potential of AMS will be demonstrated on typical radionuclides of interest with half-lives between some tens of years up to a hundred million years. The precise measurement of the $^{27}Al(n,2n)^{26}Al$ excitation function will be exemplified. Lack of information exists for a list of nuclides as pointed out by nuclear data requests. A brief overview on detection limits and some applications for selected long-lived radionuclides is given.

1. INTRODUCTION

A variety of nuclides are produced both in nature (e.g., cosmogenic) and in man-made fission and fusion devices. Induced radioactivity in structural materials of fission and fusion reactors, particularly long-lived radionuclides, may lead to a significant amount of long-term nuclear waste [1-3]. In addition, long-lived radionuclides can also serve as diagnostic tools for different applications [1,4]. For such nuclides, production cross-sections as well as total induced activities are key parameters for safety and design analyses. Therefore, well-established data on their production rates are highly desired.

The quantification of such long-lived radionuclides can in principle be performed utilizing either decay counting, or counting directly the number of atoms under investigation, or via detection of the prompt radiation associated with its production. AMS represents an analytical technique for the detection of some specific nuclides through direct counting with the potential of quantifying isotope concentrations down to levels of 10^{-15} and below. For typical sample masses (≈mg), AMS offers a tremendously higher sensitivity compared to the decay counting method, which obviously is the consequence of the long half-life of those radionuclides.

2. AMS SYSTEMS: VERA AND GAMS

AMS deals mainly with long-lived radionuclides, which are also of some concern and importance in fusion technology, mainly with respect to radioactive waste disposal. In combination with use of negative ions and the dissociation of molecules at the stripper of the tandem accelerator, AMS offers an excellent sensitivity. The actually measured parameters are isotope ratios, i.e., rare versus stable isotope contents in a sample. Detection limits of lower than 10^{-15} have been achieved for several nuclides, which allow the detection of these isotopes at natural concentrations. For a more detailed description of AMS the reader is referred to review articles, e.g., [5-8].

The Vienna Environmental Research Accelerator (VERA) represents a state-of-the-art AMS facility based on a 3-MV tandem accelerator [9-11] that provides the ability for quantifying nuclides without interfering stable isobars over the whole mass range. Some of the measured radioisotopes are ^{10}Be, ^{14}C, ^{26}Al, ^{129}I, ^{182}Hf, ^{236}U, and ^{244}Pu (Table 1a). Complementary to "small" AMS laboratories such as the VERA Lab., there are AMS facilities that provide high particle energies: For example, the Accelerator laboratory in Munich, which comprises a 14-MV tandem accelerator in combination with a gas-filled

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TABLE 1a. Basic features of some radionuclides measured at VERA [10].

Radionuclide	Half-Life (yr)[1]	Overall Efficiency[2]	Detection Limit[3]	Precision	Remark
^{10}Be	(1.52±0.05)*10^6	5x10^{-5}	<5x10^{-14}	< 5%	isobar ^{10}B
^{14}C	(5730±40)	2x10^{-2}	<3x10^{-16}	< 0.5 %	no stable isobar[5]
^{26}Al	(0.71±0.02)*10^6	1x10^{-4}	<6x10^{-16}	< 1.0 %	no stable isobar[5]
^{129}I	(15.7±0.4)*10^6	1x10^{-2}	2x10^{-14}	2 %	no stable isobar[5]
^{182}Hf	(8.90±0.09)*10^6	1x10^{-4}	1x10^{-11}	5 %	isobar ^{182}W
^{236}U	(23.42±0.03)*10^6	not measured yet	<<6x10^{-11}	5 %	no isobar
^{244}Pu	(80.0±0.9)*10^6	>4x10^{-5}	--[4]	5 %	no isobar

TABLE 1b. Basic features of some radionuclides measured with the GAMS setup [12].

Radionuclide	Half-Life (yr)[1]	Overall Efficiency	Detection limit	Precision	Remark
^{41}Ca	(1.03±0.05)*10^5	1x10^{-4}	<2x10^{-15}	8 %	CaH_3^-, no isobar[5]
^{53}Mn	(3.74±0.04) *10^6	2x10^{-5}	<1x10^{-14}	8 %	isobar ^{53}Cr
^{59}Ni	(7.6±0.5)*10^4	1x10^{-4}	1x10^{-13}	8 %	isobar ^{59}Co
^{60}Fe	(1.5±0.3)*10^6	1x10^{-4}	1x10^{-16}	8 %	isobar ^{60}Ni
^{63}Ni	(100.1±2.0)	1x10^{-5}	2x10^{-14}	8 %	isobar ^{63}Cu
^{244}Pu	(80.0±0.9)*10^6	>5x10^{-5}	--[4]	8 %	no isobar

[1]See [25-27].
[2]Means atoms detected with a particle detector per atoms in the sample.
[3]Given as ratio, radionuclide relative to stable nuclide.
[4] ^{244}Pu measurements are assumed to be background-free.
[5]No stable negatively charged isobar is extracted from the source (e.g., $^{14}N^-$ is not stable).

magnet system (GAMS). This combination offers among the best suppression of isobaric background, in particular in the medium-mass range: Isotopes like ^{41}Ca, ^{53}Mn, ^{59}Ni, ^{63}Ni, and ^{60}Fe (see Table 1b) are measured in a wide range of applications [12].

3. NUCLEAR PHYSICS AND AMS

A variety of nuclear data request lists exist (e.g., from the NEA [13]). Large-scale nuclear data programs are, e.g., HINDAS for cross-section measurements in the 20–200 MeV energy region and beyond on Fe, Pb, U [14-16], applying partly the AMS technique too (see also [17,18]). Another somewhat competing program is n-TOF [19] for cross sections measurements from 1 eV to 250 MeV.

Clearly, studying long-lived radionuclides or reaction-products with an unfavourable decay scheme coincides with the potential of AMS. The production of long-lived activities, in particular long-lived fusion and fission products, may lead to a significant amount of long-lived nuclear waste and radiation damage in fusion devices. Many of these production cross sections are not well known, making it difficult to calculate concentration limits [3]. With the high neutron flux in a fusion reactor, impurities in structure materials may also lead to significant or dominating radioactivities. The impact of supra-thermonuclear neutrons (higher-energy neutrons) has to be taken into account. In addition 'sequential (x,n) reactions' can produce important radionuclides not achievable in one-step neutron interactions [20], e.g., the ^{23}Na(α,n) reaction producing ^{26}Al.

The shape of excitation functions may exhibit some specific features, which in principle make such a reaction suitable as a diagnostic tool in a fission or fusion environment. For example, a useful reaction for retrospective reactor dosimetry is represented by the ^{62}Ni(n,γ)^{63}Ni reaction [21]. Common detection of ^{63}Ni requires wet chemistry, whereas AMS allows in principle the direct use of the sample. Some 60 years after the explosion of the Hiroshima and Nagasaki atomic bombs, long-lived radionuclides produced therewith are still present and can be detected via AMS. This fact is used for the retrospective neutron dose reconstruction of survivors and helps to clarify neutron dosimetry (e.g., [22]).

4. PRECISE MEASUREMENT OF THE ^{27}Al(n,2n)^{26}Al EXCITATION FUNCTION

A detailed measurement of the ^{27}Al(n,2n)^{26g}Al reaction cross sections was recently performed in the near-threshold region (13.5-19 MeV), and its possible applicability for ion temperature measurements [1,4] was investigated [23].

Al-containing materials and Si carbide are candidate materials for fusion-reactor systems. The production of the long-lived radionuclide ^{26}Al ($t_{1/2}$=7.1*10^5 a) is therefore of considerable interest to the fusion reactor program since this activity will increase steadily via the ^{27}Al(n,2n) reaction and the two-step process ^{28}Si(n,np+d)^{27}Al(n,2n). The ^{27}Al(n,2n) reaction is expected to vary strongly with neutron energy above threshold (E_{th} = 13.55 MeV). An accurate description of the excitation function is

necessary to estimate the production of ^{26}Al in a typical D-T fusion environment.

The energy region around the threshold of the ^{27}Al(n,2n)^{26}Al reaction (E_{th}=13.55 MeV) is covered by the neutrons produced in a D-T plasma. Depending on the ion-temperature of the plasma, the distribution of these neutrons varies strongly and in a characteristic way, and consequently the production rate of ^{26}Al depends on the ion temperature, which makes this reaction sensitive to temperature changes [1,4].

To investigate this excitation function, Al samples were irradiated with fast neutrons under different geometries at four laboratories [23] and the amount of ^{26}Al produced in this way was analyzed by the AMS technique with the Vienna Environmental Research Accelerator (VERA). The determination of the cross sections is directly deduced using the isotope ratio (N_{26}/N_{27}) and the neutron fluence Φ_n. With the present setup of VERA [11], a background of lower than $6*10^{-16}$ for the ^{26}Al/^{27}Al ratio is achieved, which corresponds in this case ($\Phi_n \approx 10^{14}$ n/cm²) to a (n,2n) cross section of about 0.01 mb. Several AMS run series were performed resulting in the measurement of a total number of almost 400 Al samples with unknown ^{26}Al/^{27}Al ratios. In this way accurate cross-section data were obtained and cross sections considerably closer to the threshold than in previous investigations could be measured. In Fig. 1 final cross-section data are plotted. A comparison with previous

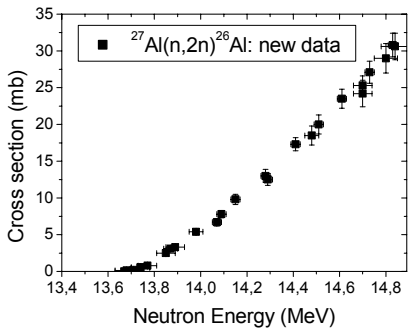

FIGURE 1. New cross–section data for the ^{27}Al(n,2n)^{26g}Al excitation function near threshold. The ^{26}Al produced was quantified via AMS at VERA [23].

data shows a substantial improvement in the knowledge of this excitation function. The new data allow a general estimation for its possible usefulness as a monitor to temperature changes in a DT-fusion plasma. For *thermal* plasmas, temperature changes of the order of 5% to 10% should be detectable. A *non-Maxwellian* fusion plasma leads to different neutron spectral distributions that depend on the observation angle. Making use of this behaviour, the ^{27}Al(n,2n)^{26}Al reaction becomes extremely sensitive for changes in the ion temperature of a non-thermal DT fusion plasma in case of backward viewing [23].

5. LONG-LIVED RADIONUCLIDES: DETECTION LIMITS, APPLICATIONS

Tables 1a and 1b list basic features of some radionuclides of interest in AMS: Their half-lives span a range from 100 years (^{63}Ni) to 80 million years (^{244}Pu). Overall efficiency is the fraction of particles in a sample that can be counted with the particle detector. The next column depicts the detection limit, either determined by isobaric interferences or by the instrumental background. Also listed is the precision of the AMS measurement if not limited by counting statistics. The nuclides presented in Table 1 do not form a complete list. In addition, new technical developments for isobar suppression are ongoing with the goal of implementing the 'medium mass' isotopes e.g., ^{36}Cl and ^{41}Ca at VERA as well as 'higher mass' isotopes like ^{146}Sm and ^{182}Hf at Munich.

In the following, a few examples of reactions with sparse cross-section data will be discussed for isotopes, which may be quantified via AMS (Table 2). ^{14}C depicts a neutron poison for thermal neutrons in fission reactors, but it is also clearly one of the major activation products in fusion processes. Looking at the EXFOR database [24], it is surprising, that the dominating reaction for the production of natural ^{14}C, ^{14}N(n,p)^{14}C, lists only one – almost 30 years old – experimental value above a neutron energy of 7 MeV. ^{14}C may also be produced by fast neutrons via oxygen, either from ^{17}O(n,α) or from ^{18}O(n,n´α), reactions for which no EXFOR entry is found above 1 MeV, or none at all, respectively.

The ^{28}Si(n,t)^{26}Al reaction (E_{th}=16.7 MeV) may serve as a tool for fusion plasma diagnostics with respect to α slowing down studies. No experimental data are available for this reaction. However, neutron fluences of the order of a few times 10^{11} n/cm² should result in enough ^{26}Al atoms to be quantified with AMS. Another activation product is ^{182}Hf, whose production from W may be of waste disposal concern.

Besides Al, other structural materials also suffer from the same problem as ^{26}Al (see sec. 4). Prominent long-lived fusion products will be found in the medium-mass range, i.e., Cu, Fe, and Ni containing materials will steadily accumulate e.g., the radionuclides ^{53}Mn, ^{60}Fe, and ^{59,63}Ni. ^{53}Mn will be produced either directly from ^{54}Fe via the (n,np+d) reaction or to a lesser extent indirectly via the beta-decay of ^{53}Fe ($T_{1/2}$= 8.5 min). Besides its drawback as an activation product, ^{53}Mn may also be of interest in plasma diagnostics. Similar to the ^{27}Al(n,2n)^{26}Al

TABLE 2. Selected nuclides whose production may be quantified via AMS.

Radionuclide	Reaction	x/s [1] (mb)	Particle Fluence [2] (cm^{-2})	Sample Mass / Isotope Ratio[3]	Energy Range
^{14}C	$^{14}N(n,p)$	10	$1x10^{11}$	≈ mg N	> 7 MeV
^{14}C	$^{17}O(n,\alpha)$, $^{18}O(n,n\alpha)$	5	$1x10^{11}$	≈ mg $^{17,18}O$	> 2 and 6.6 MeV
^{26}Al	$^{28}Si(n,t)$,	1	$5x10^{11}$	100 mg Si	E_{th}=16.7 MeV
^{53}Mn	$^{54}Fe(n,np+d)$	100	$2x10^{12}$	50 mg ^{nat}Fe	E_{th}= 6.75 MeV
^{59}Ni	$^{60}Ni(n,2n)$	100	$1x10^{13}$	$1x10^{-12}$	E_{th}=11.58 MeV
^{63}Ni	$^{62}Ni(n,\gamma)$	10	$1x10^{13}$	$1x10^{-13}$	30 keV
^{63}Ni	$^{62}Ni(n,\gamma)$	$1.5x10^{4}$	$1x10^{11}$	$1x10^{-12}$	thermal

[1] x/s means cross section; the value represents an estimation of the expected value only.
[2] Fluence necessary for estimated 100 counts detected in AMS, or to result in a ratio 10 times above AMS background.
[3] Sample mass in combination with particle fluence gives the number of produced radionuclides; the isotope ratio follows directly from cross section and fluence.

reaction, the $^{54}Fe(n,2n)^{53}Fe$ excitation function has a threshold at 13.63 MeV, which makes it sensitive to temperature changes too [4]. However, the knowledge of this excitation function is still very poor. The production of ^{59}Ni via the (n,2n) reaction on ^{60}Ni was measured twice for one neutron energy at 14.8 MeV [2,24]. However, their value deviates from each other by a factor of 4. Finally, the $^{62}Ni(n,\gamma)^{63}Ni$ reaction at stellar temperatures (30 keV) is of interest for neutron capture nucleosynthesis models, which (see above) is also of interest for fission reactor dosimetry [21].

AMS clearly allows the quantification of such radionuclides. The potential of ultra-low isotope-ratio measurements is reflected in a wide range of applications ranging from nuclear physics to environmental and life sciences.

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