PRODUCTION OF LONG-LIVED RADIONUCLIDES ^{10}Be, ^{14}C, ^{53}Mn, ^{55}Fe, ^{59}Ni AND ^{202g}Pb IN A FUSION ENVIRONMENT

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In a fusion reactor and also in future advanced reactor types, long-lived activation products may lead to significant long-term waste disposals and radiation damage. 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}Fe, ^{59,63}Ni; and for heavier isotopes ^{202m}Pb, ^{210m}Bi. Since a few years the technique of accelerator mass spectrometry (AMS) has been applied at the Vienna Environmental Research Accelerator (VERA) facility for the detection of long-lived radionuclides for such studies. In this respect, samples were irradiated with quasi-monoenergetic neutrons at TU Dresden's 14-MeV neutron generator and the van de Graaff accelerator at IRMM. 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. The radionuclides ^{10}Be, ^{14}C, ^{26}Al, ^{55}Fe, ^{210m}Bi and ^{202g}Pb were measured at the VERA facility.

KEYWORDS : long-lived radionuclides, accelerator mass spectrometry (AMS), nuclear fusion, ADS, cross section measurement

INTRODUCTION

Improved and highly accurate nuclear data are urgently required for the design of advanced reactor concepts, like the design of nuclear fusion devices (e.g. ITER), or next generation nuclear power plants (Gen IV, ADS). In a fusion environment particularly long-lived activation products may lead to significant long-term waste disposals and radiation damage. Many of these production cross sections are not well-known, making it difficult to calculate concentration limits [1]. With the high neutron flux, also impurities in structure materials may lead to significant or dominating activations. For such nuclides production cross-sections and induced activities are key parameters for safety and design analysis.

Neutron induced reactions from thermal to fast neutron energies need to be considered. Impurities also play a significant role. The high neutron dose expected allows also multi-step processes to become of importance. Long-lived radionuclides, however, despite being of concern as long-term waste disposal, are difficult to measure in standard activation experiments due to their low activities. This shortcoming can be overcome by using mass spectrometric techniques for measuring directly the number of produced radionuclides rather than their decay. In particular, accelerator mass spectrometry (AMS) has proven to represent a powerful technique in quantifying long-lived radionuclides from activation experiments. In Table 1 some selected longer-lived radionuclides are listed for nuclides in the mass range from Be to the Fe/Ni region (for a more detailed list see e.g. [2,3]). Shown is also the dominant reaction leading to that long-lived nuclide. Interestingly, such radionuclides often are nuclides predestined for accelerator mass spectrometry measurements.

We have continued to measure via AMS critical production cross sections of long-lived radionuclides of relevance to nuclear fusion, and have extended our program to study the production of such nuclides also for neutron environments relevant for ADS and GenIV facilities:

Recently, production of ^{10}Be from neutron irradiation of Be and C, ^{14}C production from ^{14}N(n,p), and in detail ^{55}Fe production was studied at the VERA (Vienna Environmental Research Accelerator) laboratory. AMS measurements for ^{54}Fe(n,γ)^{55}Fe from thermal to MeV neutron energies, for ^{56}Fe(n,2n) and ^{58}Ni(n,α) were performed, demonstrating that AMS allows to generate data with 2–3 % uncertainty. Similarly, the production of ^{53}Mn ($t_{1/2}$ = 3.7 Myr) has been studied in detail. These data, for the first time based on AMS, are sensitive to the total production of ^{53}Mn and indicate that previous data strongly underestimate the production of ^{53}Mn (sec. 4).

Table 1 Some selected long-lived radionuclides produced in a fusion environment and being well-suited for AMS

isotope	half-life	reaction
^{10}Be	1.39×10^{6} yr	^{13}C(n,α)
^{14}C	5730 yr	^{14}N(n,p)
^{14}C	5730 yr	^{nat}O(n,x)
^{26}Al	7.1×10^{5} yr	^{27}Al(n,2n)
^{53}Mn	3.7×10^{6} yr	^{54}Fe(n,2n)
^{53}Mn	3.7×10^{6} yr	^{54}Fe(n,np+d)
^{55}Fe	2.74 yr	^{56}Fe(n,2n)
^{63}Ni	101 yr	^{63}Cu(n,p)
^{63}Ni	101 yr	^{64}Ni(n,2n)
^{59}Ni	76 000 yr	^{60}Ni(n,2n)

NEUTRON IRRADIATIONS

For our measurement programme (table 2), neutron irradiations were performed at different neutron producing facilities, spanning a range of neutron energies from cold neutrons to 21 MeV. Among those, the Research reactor at Budapest, Hungary (cold neutrons) [4], the Atominstitut in Vienna, Austria (thermal neutrons) [5], the 3.7 MV Van de Graaff accelerator setup for keV-neutrons at Karlsruhe [6] (via Li(p,n)), Germany (25 – 520 keV), the 300 kV Cockroft -Walton neutron generator at TU Dresden, Germany (14-MeV neutrons via T(d.n) [7], and the 7-MV Van de Graaff accelerator at IRMM, Geel, Belgium (13 – 21 MeV) [8], generated such neutrons. In this paper we restrict ourselves to activations in the fast neutron energy range from 13 to 21 MeV. Results from other measurements, e.g. neutron capture cross sections of interest to nuclear astrophysics (s-process nucleosynthesis), and in the actinide mass range, relevant for reactor design can be found in [9,10].

AMS MEASUREMENTS

1.1 Vienna Environmental Research Accelerator

Accelerator mass spectrometry represents a mass spectrometric technique based on the use of a (tandem) accelerator. The VERA facility operates a 3-MV tandem accelerator. It represents a state-of-the-art AMS setup which allows to transport all nuclides over the whole mass range from hydrogen up to super-heavy elements; from the negative ion source to the various particle detectors [11]. Among the measured radioisotopes are e.g. ^{10}Be, ^{14}C, ^{26}Al, ^{36}Cl, ^{41}Ca, ^{55}Fe, ^{129}I, ^{182}Hf, ^{202,210}Pb, ^{210m}Bi, ^{236}U and $^{239-244}$Pu, used within a wide range of applications – from archaeology, climate research to nuclear physics and astrophysics.

1.2 Maier-Leibnitz Laboratory - Munich

The AMS setup at Maier-Leibnitz Laboratory (MLL) of the Munich Universities [12] – equipped with a 14-MV tandem – offers, in combination with high particle energies and a dedicated gas-filled magnet system, a unique separation power of rare radionuclides from their isobars. Examples are among others, medium-mass ^{60}Fe, ^{53}Mn, ^{59}Ni, ^{79}Se where isobaric interference from stable ^{60}Ni, ^{53}Cr, ^{59}Co, ^{79}Br, respectively, is strongly suppressed allowing measurements of isotope ratios between 10^{-13} and 1×10^{-16}.

Table 2 Samples recently irradiated at TUD and IRMM and measured via AMS at VERA and at MLL (*see [13]).

reaction	E_n [MeV]	Irradiation / AMS
^{54}Fe(n,np+d)^{53}Mn	13.4 – 14.8	TUD / MLL
^{54}Fe(n,2n)^{53}Fe	13.4 – 14.8	TUD / VERA
^{56}Fe(n,2n)^{55}Fe	13 – 20	TUD / VERA
^{60}Ni(n,2n)^{59}Ni	17 – 19	TÜB / MLL*
^{58}Ni(n,α)^{55}Fe	13 – 20	TUD / IRMM / VERA
^{14}N(n,p)^{14}C	13 – 20	TUD / IRMM / VERA
^{13}C(n,α)^{10}Be	13 – 20	TUD / IRMM / VERA
^{204}Pb(n,3n)^{202g}Pb	18 – 21	IRMM / VERA
^{232}Th(n,x)	18 – 20	IRMM / VERA
^{238}U(n,x)	18 – 20	IRMM / VERA

RECENT MEASUREMENTS applying AMS

1.3 ^{54}Fe(n, np+d)^{53}Mn ($t_{1/2}$ = 3.7 Myr)

The high production rate of ^{53}Mn ($t_{1/2}$ = 3.7 Myr) is of some concern as an activation product since this activity will increase steadily during the lifetime of an operating reactor. In particular, Fe-containing materials are candidate materials for fusion reactor systems: ^{53}Mn can be produced directly via ^{54}Fe(n,np) and ^{54}Fe(n,d). The respective neutron threshold energies are 9 and 6.8 MeV. ^{53}Mn can also be produced via the ^{54}Fe(n,2n)^{53}Fe reaction, and decay of ^{53}Fe ($t_{1/2}$ = 8.5 min). The latter reaction has its threshold at 13.6 MeV neutron energy. This high threshold makes it a very sensitive measure for fusion temperature changes, similar to ^{27}Al(n,2n)^{26}Al [14]. Experimental information for the production of ^{53}Mn is scarce and discordant to calculations (see Fig. 1). Activation experiments in a fusion peak neutron field showed, that for materials like Eurofer,

production of ^{26}Al and ^{53}Mn are the dominant long-term activities, with estimated contributions of 70 % and 27 % to the total dose rate, respectively, however, for ^{53}Mn with an uncertainty of 60 % [15, 16].

Fe metal samples, highly enriched in ^{54}Fe, have been irradiated with quasi-monoenergetic neutrons at TU Dresden's 14-MeV neutron generator [7]. Via the T(d,n)^{4}He reaction, neutrons with energies between 13.4 and 14.9 MeV were produced. Several Fe samples were exposed to neutrons with a total fluence of a few 10^{13} ncm^{-2}. In addition, short-term irradiations were performed to measure the production of short-lived ^{53}Fe (see 4.2)

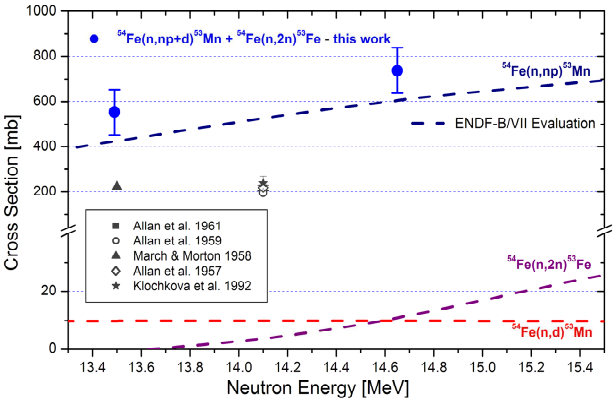

Fig. 1. Excitation function for production of ^{53}Mn around 14 MeV. A recent evaluation [18] disentangles the contribution of the three different reaction channels from ^{54}Fe via (n,np), and the minor contributions from (n,d) and (n,2n) producing the long-lived ^{53}Mn.

The ^{53}Mn content was measured via AMS utilizing the 14-MV tandem accelerator of MLL. In combination with a gas-filled magnet and a multi-anode ionization chamber, a detection limit lower than 10^{-14} for ^{53}Mn/^{55}Mn isotope ratios was achieved [12,17]. Assuming a cross-section value of 600 mbarn, for a neutron fluence ≥ 10^{13} n cm^{-2}, we calculate a ratio ^{53}Mn/^{55}Mn of ≥ 6×10^{-12} - well above background.

Previous measurements for the ^{54}Fe(n,np+d)^{53}Mn reaction [18] have been performed at neutron energies of 13.5 and 14.1 MeV. These measurements were based on the detection of emitted protons and were sensitive to the np-channel only. Their results indicate a constant cross section of about 200 mbarn (Fig. 1). However, they disagree by a factor of 2-3 with recent evaluations (see e.g. ENDF). Evaluations indicate an increasing excitation function with cross-section values between 400 and 650 mbarn for neutron energies between 13.4 and 15 MeV.

Preliminary experimental data obtained in this work are plotted in Fig. 1. Our data, for the first time based on AMS measurements, are sensitive to the total production of ^{53}Mn in such a neutron environment. However, these preliminary data indicate that previous data seem to strongly underestimate the production of ^{53}Mn in this energy range (new data for additional neutron energies are upcoming). Our data are slightly higher than ENDF-VII.

1.4 ^{54}Fe(n,2n)^{53}Fe & ^{56}Fe(n,2n)^{55}Fe

In addition to the direct production of ^{53}Mn from ^{54}Fe, the indirect step via ^{54}Fe(n,2n)^{53}Fe was investigated. Fe samples, highly enriched in ^{54}Fe, were irradiated at TUD to study the strongly energy-dependent (n,2n) cross section dependence between its threshold at 13.6 MeV up to 14.8 MeV. While the total contribution to the decay product ^{53}Mn is fairly small (less than 10 mbarn in this energy range, see Fig. 1), the steeply increasing excitation function is of interest to fusion diagnostics, as the production rate of ^{53}Fe –similar to ^{26}Al– is a sensitive function of the fusion plasma temperature [14]. However, existing data for ^{54}Fe(n,2n)^{53}Fe are strongly varying. In order to test its sensitivity as a temperature monitor, we re-measured that reaction with the goal to produce highly precise data.

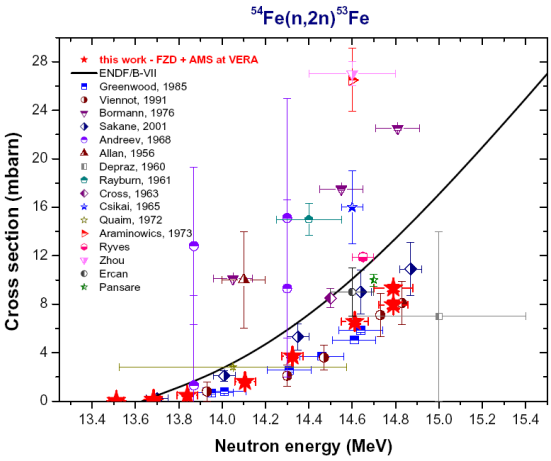

Fig. 2. Excitation function for the reaction ^{54}Fe(n,2n)^{53}Fe around 14 MeV. Some experimental data are shown as symbols, together with ENDF evaluation. Preliminary data from a first irradiation campaign are plotted.

In addition, neutron exposure of natural iron material will produce high a mounts of ^{55}Fe ($t_{1/2}$ = 2.73 yr) via the threshold reaction ^{56}Fe(n,2n)^{55}Fe. Existing experimental data, exhibit relative large uncertainties (Fig. 3). The reaction product, ^{55}Fe, is an excellent candidate for AMS measurements: its background was measured at the VERA laboratory at levels below 10^{-15} for the isotope ratio ^{55}Fe/^{56}Fe; and a measurement precision of better than 2% was demonstrated [19].

We have irradiated natural Fe samples at TUD with neutrons between 13.4 and 14.8 MeV, and at IRMM from 13 to 20 MeV. Some preliminary AMS data from the first irradiation campaign with low energy resolution are shown in Fig. 3. Final results for ^{56}Fe(n,2n) are expected to be

precise at a level of < 4 %.

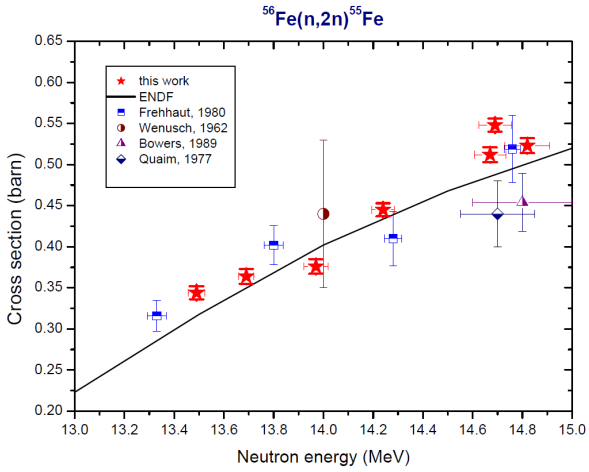

Fig. 3. Excitation function for the ^{56}Fe(n,2n)^{55}Fe around 14 MeV. Selected experimental data are shown as symbols, together with ENDF evaluation. Preliminary data from a first irradiation campaign are plotted. New, precise measurements for neutron energies between 13 and 20 MeV are in progress now.

1.5 ^{204}Pb(n,3n)^{202g}Pb ($t_{1/2}$ = 52.5 kyr)

Lead is of interest both for developments in nuclear physics and for applications. Lead is found everywhere for shielding against γ-radiation, also in neutron environments. In advanced reactors liquid lead is proposed as a possible coolant alternative to sodium or gas. In accelerator driven systems lead is part of the proposed spallation target.

A particular feature of the lead isotopes are low (n,2n) and (n,3n) thresholds. For instance (n,3n) channels are open well below 20 MeV. This has led to experimental results for ^{208}Pb(n,3n)^{206}Pb, ^{206}Pb(n,3n)^{204m,g}Pb and ^{204}Pb(n,3n)^{202m}Pb reactions [8]. Since isomer cross sections are more difficult to model, it is important to have results for the total cross section. The reaction ^{204}Pb(n,3n)^{202g}Pb is an example of a reaction that could not be studied before (no data in EXFOR) and is possible with AMS. The end product ($t_{1/2}$ = 5.25×10^4 yr) is long-lived and emits no gamma-rays. The isobar of Hg does not represent a problem in AMS since Hg is not forming negative ions.

Samples enriched in ^{204}Pb have been irradiated at two different neutron energies (18.5 and 20.5 MeV) at IRMM. The isotope ratio ^{202g}Pb/^{204}Pb produced in the neutron irradiation was measured at VERA. VERA has been upgraded for heavy ion detection recently [11] and is now especially well suited for heavy isotope measurements with increased resolution power. Interfering ions which pass all beam filters are identified with a high-resolution time-of-flight system.

No AMS measurement exists so far for ^{202g}Pb. Experience on the performance at VERA for heavy isotopes has shown that an isotope ratio of several 10^{-12} is required in order to obtain good quantitative results. Such work includes studies on ^{209}Bi(n,γ)^{210m}Bi. Measurements on ^{236}U showed a similar background level [10]. In case of ^{202g}Pb no interference from lower masses is expected. Lead samples enriched in ^{204}Pb were used for these study. The high sensitivity of the AMS technique requires very small sample masses of typically 30 mg.

First measurements indeed demonstrated background-free data, and no ^{202}Pb was detected for un-irradiated Pb samples. Based on these first results; we expect an overall cross-section uncertainty of the order of 5 – 10 %.

SUMMARY

AMS represents a powerful technique with excellent sensitivity for the detection of long-lived radionuclides through ultra-low isotope ratio measurements. The combination of activation and subsequent AMS detection is an independent and complementary technique, and thus depending on different systematic uncertainties compared to standard techniques.

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REFERENCES

[1] D. L. Bowers and L.R. Greenwood, *J. Radioanal. Nucl. Chem.* **123**, 461 (1988).
[2] S. Fetter, E. Cheng, F. Mann, Fus. Eng. Des. **13**, 239 (1990).
[3] R.A. Forrest, Fus. Eng. Des. **81**, 2143 (2006).
[4] http://www.iki.kfki.hu/nuclear/instruments/pgaa_en.shtml
[5] http://www.ati.ac.at/
[6] W. Ratynski and F. Käppeler, Phys. Rev. C**37**, 595 (1988).
[7] K. Seidel et al., *Fusion Eng. Des.* **81** 1211 (2006).
[8] V. Semkova et al., Phys. Rev. C**80**, 024610 (2009).
[9] these proceedings ND1528
[10] these proceedings ND1520
[11] P. Steier et al., *Nucl. Instr. and Meth.* B**268**, 1045 (2010).
[12] M. Poutivtsev et al., *Nucl. Instr. and Meth.* B**268** 756 (2010).
[13] A. Wallner, et al., Proc. Int. Conf. on Nuclear Data for Science and Technology, April 22-27, 2007, Nice, France, eds O. Bersillon et al., EDP Sciences, 1007 (2008).
[14] A. Wallner et al., *Europ. Phys. J.* **A17** 285 (2003).
[15] K. Seidel et al., *Jour. Nuc. Mat.* **307-311** 1037-1041 (2002).
[16] A. Wallner et al., *AIP Conf. Proc.* **769**, pp. 621-624 (2005).
[17] J.M. Schaefer et al., *Earth Planet Sci. Lett.* **251**, 334 (2006).
[18] ENDF/B-VII.0 see: http://www-nds.iaea.org/exfor/endf.htm; and EXFOR see: http://www-nds.iaea.org/exfor/exfor.htm
[19] A. Wallner et al., Nucl. *Instr. and Meth.* B**259** 677 (2007).