CERN-INTC-2014-012 / INTC-P-408
15/01/2014

EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

Measurement of the neutron capture cross-sections of ^{53}Mn at EAR-2

January 14th 2014

R. Dressler[1], C. Guerrero[2,3], F. Gunsing[3,4], St. Heinitz[1], T. Mendonca[3], D. Schumann[1], T. Stora[3], R. Reifarth[5], A. Wallner[6,7], C. Weiss[3,8], and the n_TOF Collaboration.

[1] *Paul Scherrer Institute, Villigen, Switzerland*
[2] *Universidad de Sevilla, Spain*
[3] *European Organization for Nuclear Research (CERN), Geneva, Switzerland*
[4] *Commissariat de l'Energie Atomique (CEA) Saclay - Irfu, Gif-sur-Yvette, France*
[5] *Goethe University Frankfurt, Frankfurt, Germany*
[6] *University of Vienna, Vienna, Austria*
[7] *Australian National University, Canberra, Australia*
[8] *Vienna University of Technology, Vienna, Austria*

Spokesperson: Rugard Dressler (rugard.dressler@psi.ch)
Technical coordinator: Oliver Aberle (oliver.aberle@cern.ch)

Abstract:

We propose to measure the neutron capture cross sections of ^{53}Mn at the Experimental Area 2 (EAR-2) of the n_TOF neutron time of flight facility at CERN. This will be the first ever determination of the ^{53}Mn excitation function. These data will influence the models of explosive stage of star evolution and will serve as experimental input data to improve nuclear reaction codes.

The ^{53}Mn target will be manufactured in the frame of the ERAWAST project at PSI. A chemical separation of manganese out of irradiated steel samples will deliver a stock solution containing 5×10^{19} atoms of ^{53}Mn. Due to the high amount of ^{55}Mn, the stock solution can not be used directly to prepare a target. An additional depletion of ^{55}Mn must be performed in a subsequent mass separation using the ISOLDE off-line ion-source test setup. The final target will contain 5×10^{17} atoms of ^{53}Mn and less than 1×10^{16} atoms of ^{55}Mn.

The aim of this experiment is the determination of the neutron capture cross sections of ^{53}Mn from thermal neutron energies to neutron energies of about 100 keV.

Requested protons: 3.5×10^{18} protons on target

Experimental Area: EAR-2

1 Introduction and motivation

Presently available experimental techniques are able to determine neutron capture cross sections at astrophysical interesting energies with a precision level of 1% as indicated in [1]. However, such results have been obtained for a few relevant and stable isotopes only. A large number of reactions where the cross section is only known with an uncertainty level of more than 10% have to be reinvestigated in the future. This task is one of the aims of the n_TOF facility. In Fig. 1 an overview on the uncertainties of the neutron capture cross sections at stellar neutron energies is depicted [2]. It should be pointed out that these data represent maxwellian averaged cross sections at a stellar neutron energy of 30 keV and that the situation becomes even worse for entire excitation functions, cross sections at lower neutron energies or in general for radioactive isotopes. A precise knowledge of the nuclear reaction rates involved in the production and destruction of short lived cosmogenic radionuclides is needed for a better understanding of the physics involved in the explosive stages of star evolution. In addition, more reliable determinations of the half-lives of involved radionuclides are fundamental ingredients to understand the nucleosynthesis in the Universe (see e.g. [3]).

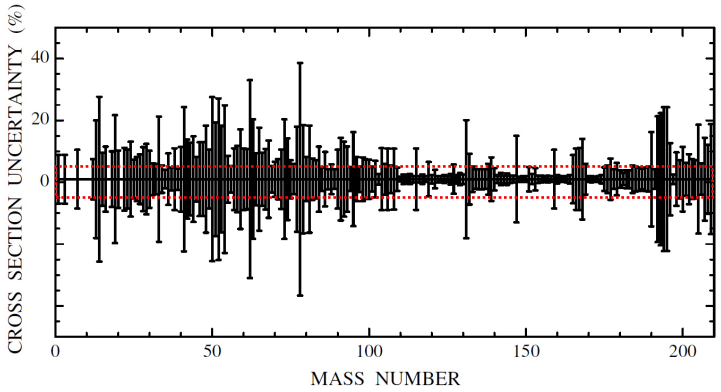

Fig. 1: Uncertainties for the (n; γ) cross sections at stellar neutron energies. The 5% uncertainty level is indicated by dashed red lines (figure reproduced from [2]).

^{53}Mn ($t_{1/2}$ = 3.7 My [4]) was discovered by Wilkinson and Sheline [5] in 1955 using proton induced reactions on highly enriched ^{53}Cr targets. However, the produced amount in the reaction ^{53}Cr(p; n)^{53}Mn was not sufficient for the determination of neutron capture cross sections in activation experiments or the determination of the half-life.

The main part of ^{53}Mn in the Universe is formed as decay product of ^{53}Fe produced in core-collapse supernovae by explosive Si-burning conditions (see e.g. [6]). Consequently, the main source of the stable isotope ^{53}Cr observed in the solar system is the decay of ^{53}Mn. In other types of supernovae explosions (e.g. thermonuclear supernovae or supernovae type-Ia) ^{53}Mn can be directly produced in an efficient way (see [7 – 9]). In 1960 Shedlovsky [10] showed that iron meteorites contain ^{53}Mn in an amount of about 5×10^{11} atoms per gram meteoritic material. This finding made it possible to perform investigations of the nuclear properties of ^{53}Mn as well as nuclear reaction studies in the late 1960th and 1970th. First measurements of the neutron capture cross section at thermal neutron energy using up to 10^{13} atoms of ^{53}Mn extracted from meteorites were carried out by H.T. Millard [11] in 1965 yielding $\sigma_{th} \approx 170$ b and by R. Wölfle et al. [12] in 1972 resulting in a value of $\sigma_{th} = (66\pm7)$ b. The experiments reported in [11, 12] are the only ones performed so far to determine neutron capture cross sections of ^{53}Mn. These measurements correspond to neutron energy of

25 meV, while the region of resolved resonances and the neutron energies of astrophysical interest are in the range up to several 100 keV.

Measurements of neutron capture cross sections as well as other nuclear reaction cross sections of ^{53}Mn will improve our knowledge about the nuclear structure and nuclear reaction mechanisms affecting the production of ^{53}Mn in different phases of the star development. In addition they will serve as input data for nuclear reaction codes like TALYS and enhance their prediction strength. Fig. 2 shows all experimental neutron capture cross sections of ^{53}Mn together with predictions available in nuclear data libraries as EAF-2010 [13], and various TENDL versions [14]. It is obvious that all of the predictions used the value of Wölfle as an anchor point. Moreover, the energy of the resolved resonance region in TALYS calculations did not only change dramatically between 2008 and 2010, but in the 2012 even the shape and strength of individual resonances has been modified by changing corresponding parameters. All these features based so far only on general assumptions about the nuclear structure and observed general trends across the chart of nuclei, but not on really measured data of ^{53}Mn.

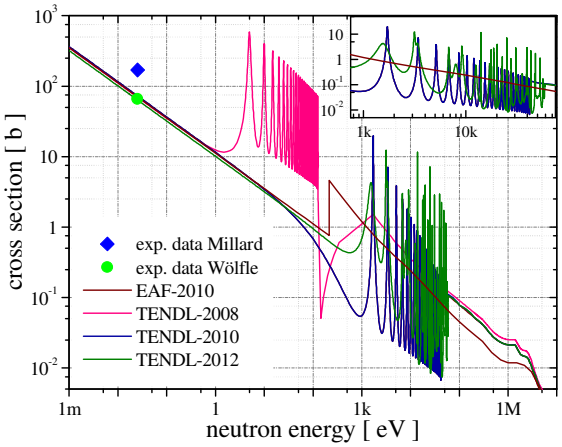

Fig. 2: Comparison of all known experimental data of the ^{53}Mn(n;γ) reaction (blue diamond [11]; green circle [12]) with published model calculations (wine line EAF-2010 [13]; pink, royal blue and olive green lines TALYS predictions [14])

In order to measure experimentally relevant reaction rates for unstable isotopes, one of the fundamental requirements is their availability in sufficient amounts. In the past, samples containing short-lived cosmogenic radionuclides were mainly obtained from meteoritic material, limiting the available amount to maximal 10^{13} atoms. However, these nuclei can as well be produced on earth via spallation reactions induced by high-energy protons or neutrons. Therefore, the exploitation of long-term irradiated materials (e.g. construction components, collimators, shielding materials, targets, and beam dumps) stemming from high-energy accelerator devices, which would usually be treated as radioactive waste, may alternatively serve as a source to obtain such valuable exotic radionuclei.

At the Paul Scherrer Institute, which operates one of the most powerful proton accelerators world-wide with an output power of about 1.4 MW [15], the production and extraction of some of these rare isotopes was demonstrated [16, 17]. The attempt to use these materials as a potential source of long-lived isotopic material was started several years ago, when the ERAWAST (**E**xotic **R**adionuclides from **A**ccelerator **Was**te for **S**cience and **T**echnology) initiative was launched.

2 Experimental setup

One of the most favorable sources for gaining considerable amounts of ^{53}Mn are stainless steel samples from the SINQ Target-Irradiation Program (STIP) at PSI. This material research program was initiated in 1996 with the objective to study fundamental mechanical properties and radiation damage effects of structure materials under conditions representative for high-flux nuclear reactors and high-power spallation sources. Detailed descriptions of the SINQ and the STIP program can be found in [18, 19]. Once the material investigations are completed, the samples are no longer needed and, therefore, considered as waste. In a recently published paper [20] we report on the use of irradiated STIP samples to extract ^{44}Ti, ^{26}Al and ^{53}Mn for experiments in nuclear astrophysics.

After finalizing all chemical treatments and purifications [20], a stock solution will be available to produce a target containing 5×10^{17} atoms ^{53}Mn on 0.2 mm pyrolytic graphite. The diameter of the deposited material will be about 20 mm.

The experiment will be performed at the n_TOF Experiment Area 2 (EAR-2). We expect a gain of a factor 27 in neutron flux for this type of experiments compared to Experimental Area 1 (EAR-1) based on the design study [21]. Contrarily, the possibility to resolve resonances in the energy region above 10 keV will be reduced due to the shorter time of flight path and consequently a deteriorated resolution function. Therefore, the identification of individual resonances may be hindered for neutron energies exceeding 10 keV. Results of simulation comparing the expected signals recorded in EAR-1 and EAR-2 using the capture cross sections of the TENDEL-2012 compilation [14] and assuming the whole requested proton dose applied to the n_TOF target are displayed in Fig. 3. But even in the case that the resonances cannot be resolved; the data can be treated in a way to obtain average cross sections in this region.

The prompt γ-rays emitted after the neutron capture will be detected using a set of two C_6D_6 detectors perpendicular oriented to the neutron beam axis in a face-to-face geometry about 9.0 cm upstream from the capture target. The performance of a similar setup at EAR-1 was proven in several experiments see e.g. [22, 23]. The neutron sensitivity and thus the background from scattered neutrons is minimized in this configuration. The C_6D_6 detector setup at EAR-2 will be further optimized exhibit an even more reduced neutron sensitivity compared to the setup used at EAR-1 (for more details see [24]). In contrast to the EAR-1 a somehow extended halo of the neutron beam must be taken into account. Therefore, the C_6D_6 detectors will be placed at a distance of about 4.0 cm from the center of the neutron beam axis. This will reduce the absolute efficiency to detect γ-rays to about 18% instead of 20% reached in EAR-1 for a similar setup at a distance of 2.5 cm.

Fig. 4 shows the expected event yield of individual constituents of the target during the whole requested beam time. The calculations were performed using an energy resolution of 100 bins per energy decade and taking into account the target thickness, the neutron flux and the detection efficiency as well as the target composition according to the abundance of the chemical elements present in the target. The used neutron capture cross sections were taken from the ENDF-B/VII.1 [25] data compilation with exception of ^{53}Mn where the TENDL-2012 [14] predictions were used. The ratio of scatter and capture cross section is for all isotopes well below 100. Thus, no additional problems evaluating the recorded data in the envisaged energy region have to be expected.

The possibility to extract cross section values from the recorded data depends mainly on the total detection efficiency of the experimental setup and, equally important, on the background of scattered neutrons and γ-rays from the entire installations in EAR-2. Monte-Carlo simulations showed a significantly elevated neutron scattering background compared to EAR-1. This background will additionally complicate measurements with very low target amounts in the region above 10 keV. First measurements performed during the commissioning phase of EAR-2 can clarify whether the background exhibits a serious problem performing measurements in the energy region above 10 keV or not. Besides the background generated from the experimental setup in EAR-2 the used target will be the main source of background events both from scattered neutrons and unwanted capture reactions. Special care must be taken concerning the possibilities to exclude sources of background events in the sample (see Appendix for details). From this it follows that only after a subsequent mass separation a sufficient purity of the sample material can be reached to make the sample suitable to produce a target for measuring the capture cross sections of ^{53}Mn.

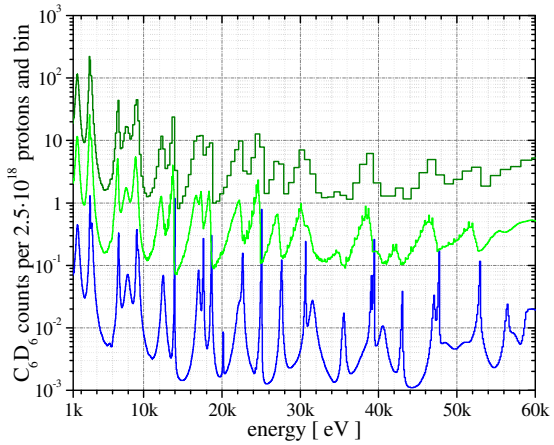

Fig. 3: Expected signals from ^{53}Mn at the n_TOF EAR-1 and EAR-2. Displayed are the simulated counts recorded with two C_6D_6 detectors originating from 5×10^{17} atoms ^{53}Mn. Given are the signals in EAR-1 (blue line), EAR-2 (green line) using 1000 bins per energy decade. The gain factor of 27 in the applied neutron flux is clearly visible. The different time of flight path length leads to significant changes in the expected time resolution function. Therefore, resonance kernels cannot be well extracted in the energy region above 10 keV. However, the disadvantage using 1000 bins per energy decade will be that the main part of the resolved resonate region will fall below one count per bin and therefore, will not be detectable. Hence, in addition the signal in EAR-2 is displayed using only 100 bins per energy decade (olive green line). In this case all resonances produce a measureable signal, but for sharp resonances the counts will fall in only one of a few bins and it will not be possible to disentangle close together resonances.

3 Objectives and beam time request

The goals of this proposal are:

- To measure the neutron capture cross sections of ^{53}Mn in the range from 25 meV to 100 keV
- To identify the resonance region and determine kernels of resolved resonances up to 10 keV.

The experiment at n_TOF EAR-2 can then be conducted fall 2015 or spring 2016. In total 3.5×10^{18} protons on n_TOF target will be required to conduct this experiment.

Summary of requested protons: 3.5×10^{18} protons on n_TOF target

In Tab. 1 the distribution of the requested protons for the main measurement as well as for reference and background measurements is given.

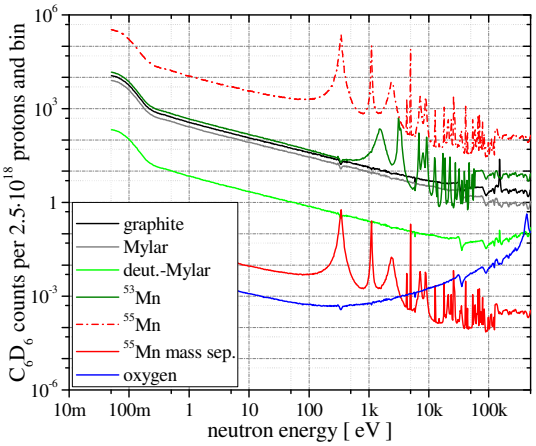

Fig. 4: Expected signals using the ^{53}Mn target at the n_TOF EAR-2. Displayed are the simulated counts recorded with two C_6D_6 detectors. Given are the signals from ^{53}Mn (olive green line), ^{55}Mn (red lines), 0.2 mm pyrolytic graphite backing (black line), 2.0 µm Mylar backing (gray line), 2.0 µm deuterated Mylar backing (green line) and the oxygen content if fully oxidized compounds are deposited on the backing (blue line). The red dashed dotted line represents the expected signal from ^{55}Mn present in the stock solution without an additional mass separation (for more details see Appendix).

Tab. 1: Number of protons requested for determination of ^{53}Mn(n; γ) cross section.

sample	Purpose	protons
5×10^{17} atoms ^{53}Mn (on 2.0 µm D-Mylar foil)	^{53}Mn(n; γ)	2.5×10^{18}
1×10^{18} atoms ^{197}Au (on 2.0 µm D-Mylar foil)	^{197}Au (n; γ)	2.0×10^{17}
7.4×10^{21} atoms ^{197}Au (on 0.1 mm Au foil)	saturated resonance analysis ^{197}Au (n; γ)	2.0×10^{17}
1×10^{21} atoms ^{55}Mn (on 2.0 µm D-Mylar foil)	Background from ^{55}Mn(n; γ)	2.0×10^{17}
2×10^{21} atoms ^{12}C (0.1 mm D-Mylar ø 20 mm)	background from backing	4.0×10^{17}
none	background w/o beam	0
	total dose	3.5×10^{18}

References:

[1] Bao Z Y et al (2000) Atomic Data and Nuclear Data Tables **76** 70
[2] Käppeler F (2007) Nucl. Instr. and Methods **B 259** 663
[3] José J, Iliadis C (2011) Rep. Prog. Phys. **74** 096901
[4] Honda M, Imamura M (1971) Phys. Rev. C 4 1182
[5] Wilkinson J R and Sheline R K (1955) Phys. Rev. **99** 752
[6] Thielemann F K, Nomoto K, Hashimoto M (1996) Astrophys. J. **460** 408
[7] Thielemann F K, Nomoto K, Yokoi K (1986) Astron. Astrophys. **158** 17
[8] Travaglio C et al (2011) Astrophys. J. **739** 93
[9] Kusakabe M, Iwamoto N, Nomoto K (2011) Astrophys. J. **726** 25
[10] Shedlovsky J P (1960) Geochim. Cosmochim. Acta **21** 156
[11] Millard Jr H T (1965) Science **147** 503
[12] Wölfle R, Herr W, Herpers U (1972) Radiochim. Acta 18 207
[13] EAF-2010 JEFF 3.0/3.1: Sublet J-Ch, et al. (2010) The European Activation File: **CCFE-R 05**
[14] TENDL: "TALYS-based Evaluated Nuclear Data Library"
TENDL-2008 release 17.11.2008: Koning A J, Rochman D (2008) **JEFF-DOC 1262**
TENDL-2010 release 08.12.2010: Rochman D, Koning A J (2010) **JEF/DOC 1349**
TENDL-2012 release 11.01.2013: Koning A J, Rochman D (2012) Nuclear Data Sheets **113** 2841
[15] Wagner W et al (2009) Nucl. Instrum. Methods Phys. Res. **A 600** 5
[16] Schumann D, Neuhausen J (2008) J. Phys. **G 35** 014046
[17] Schumann D, et al. (2009) Radiochim. Acta **97** 123
[18] Dai Y, Bauer G S (2001) J. Nucl. Mater. **296** 43
[19] Dai Y et al (2005) J. Nucl. Mater. **343** 33
[20] Dressler R, et al. (2012) J. Phys. G: Nucl. Part. Phys. **39** 105201
[21] E. Chiaveri and the n_TOF Collaboration (2012) "Proposal for n_TOF Experimental Area 2" (CERN-INTC-2012-029/INTC-O-015)
[22] Guerrero C, et al. (2013) Eur. Phys. J. **A 49** 27
[23] Lederer C, et al. (2013) Phys. Rev. Lett. **110** 022501
[24] P. Mastinu et al., "New C_6D_6 detectors: reduced neutron sensitivity and improved safety", CERN-n_TOF-PUB-2013-002 (2013)
[25] ENDF-B/VII.1: Chadwick M B, et al. (2011) Nuclear Data Sheets **112** 2887

Appendix: Target preparation

We will use irradiated STIP samples to extract the needed amount of ^{53}Mn for the proposed experiment. In total 38 selected samples (about 60 g steel material) were available for chemical treatments. Assuming a similar production rates for ^{53}Mn and ^{54}Mn (see Tab. 2) a total content of 3×10^{19} atoms ^{53}Mn in the entire set of STIP samples is expected to be available.

Tab. 2: Specific activity and content of radionuclides of the steel samples irradiated in the STIP program (data taken from [20])

isotope	spec. activity Bq/g	10^{16} atoms per g
^{26}Al	5.8	0.019
^{44}Ti	4.7×10^{6}	1.28
$^{54/(53)}$Mn*	1.2×10^{6} *	48.5**
^{60}Co	1.2×10^{6}	0.029

* activity of ^{54}Mn on 13.5.2011; ** number of ^{54}Mn atoms back calculated to end of beam at 17.12.1999

The original samples contained 0.25% vanadium, 9.5% chromium and about 0.5% manganese in addition to the bulk material iron (more details can be found in [20]). At present days 17 pieces of the STIP samples are already processed. The main part of matrix elements is separated; moreover in the case of 10 of these samples an additional purification was performed to additionally deplete the chromium amount in the solution. The overall chemical yield of manganese for the whole separation and purification procedure was determined to be about 70%. The contents of all refractory metals with exception of manganese will be at least four orders of magnitude reduced compared to there initial values due to the applied chemical separation procedures.

The main part of the background events recorded during experiments in EAR-2 will originate from the remaining refractory elements in the stock solution. These are 3.3×10^{21} atoms ^{55}Mn, 5.7×10^{19} atoms iron and 1.8×10^{19} atoms vanadium from the bulk material but also isotopes of elements produced during the proton irradiation. Fig. 5 displays simulations of the outcome of an experiment using a target with 5×10^{17} atoms ^{53}Mn prepared from this stock solution taking into account the amount of refractory elements and oxygen. It is clear that the amount of ^{55}Mn will overwhelm the ^{53}Mn signal. But also the other refractory metals will disturb the ^{53}Mn signal significantly. Only after a subsequent mass separation a sufficient purity of the sample material can be reached to make the sample suitable to produce a target for measuring the capture cross sections of ^{53}Mn.

The off-line ion-source test setup of ISOLDE can be used to perform such a mass separation and deplete the content of ^{55}Mn and other steel components with several orders of magnitude. This separation procedure will be established in collaboration with the ISOLDE target development group. The off-line ion-source can operate at pressures up to 10^{-2} mbar which makes operational temperatures of 1250 K possible. Thus, the output current of the ion-source can reach 6.2×10^{14} single charged ions per second. The transmission of the setup will be about 10% while the suppression of neighbouring masses will reach 10^{3} and of next neighbouring masses 10^{4}. Under these operational conditions 10^{17} atoms ^{53}Mn can be separated within 10 days. Special care must be taken to design all processes in a way that

the material not transferred to the mass separator part can be reprocessed and repeatedly introduced into the separation setup to enhance the overall yield. In the presented proposal the total efficiency to transfer ^{53}Mn into the final sample is expected to be about 2.5% whereas the depletion of mass 55 is in the range of 4 orders of magnitude or higher. After the mass separation the contribution of ^{55}Mn to the recorded measurement signal will be very small compared to the ^{53}Mn signal (see Fig. 4).

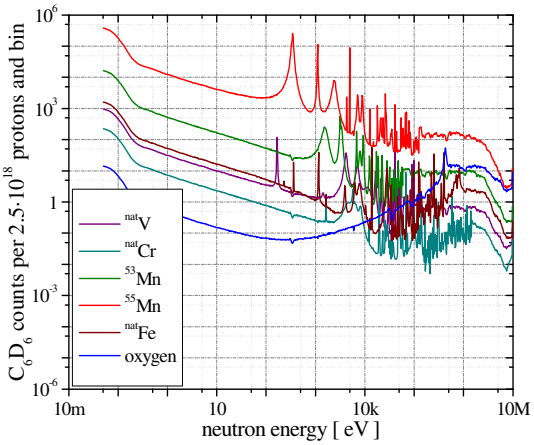

Fig. 5: Expected signals using a target prepared from an untreated Mn stock solution at the n_TOF EAR-2 (same as Fig. 4). Given are the signals from ^{nat}V (purple), ^{nat}Cr (dark cyan blue), ^{53}Mn (olive green), ^{55}Mn (red), ^{nat}Fe (wine red) and the oxygen content of the entire elements in the most common oxidation state (blue).

After finalizing all chemical treatments and purifications, a stock solution will be available to produce a ^{53}Mn target on 0.2 mm pyrolytic graphite. Additional sources of background will originate from the used backing. Therefore, alternative target backings were investigated and compared to a 0.2 mm pyrolytic graphite sheet to optimize the signal to background ratio. Fig. 4 shows simulation results for a 0.2 mm pyrolytic graphite sheet and 2.0 µm Mylar as well as 2.0 µm 98% deuterated Mylar. The use of ordinary Mylar will reduce the background by approximately 30%; whereas the use of highly deuterated Mylar will reduce this background component by about a factor 50. Highly deuterated Mylar is commercial available, e.g. as granular material. Unfortunately, there is no supplier known for thin deuterated Mylar foils. However, the production of such thin foils from granules raw materials should be feasible in a laboratory scale.

Objectives of target preparation

The goals of the target preparation are:

- To produce of a stock solution containing about 3×10^{19} atoms ^{53}Mn
- To process the stock solution using the ISOLDE off-line setup to deplete the ^{55}Mn content by 4 orders of magnitude
- To prepare a target containing 5×10^{17} atoms ^{53}Mn on a thin deuterated Mylar foil.

All preparative tasks for manufacturing the target will be performed in 2014/2015.