EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Letter of Clarification to the ISOLDE and Neutron Time-of-Flight Committee

Measurement of the neutron capture cross-sections of ^{53}Mn at EAR-2
CERN-INTC-2014-012, INTC-P-408

<div style="text-align: right">[Thursday, June 05, 2014]</div>

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Abstract:

Discussing the experiment proposal CERN-INTC-2014-012, INTC-P-408 the INTC asked in the minutes of the 46th meeting of the INTC (CERN-INTC-2014 - INTC-046) for some clarifications for a final decision.
We will address in this letter of clarification the three main topics mentioned in the INTC minutes: (i) a more detailed scientific justification, (ii) the radioprotection issue using the off-line mass separator setup and (iii) the more precise time estimation of use the on-line ISOLDE setup in off-line mode.

Requested protons:
- 6 shifts at ISOLDE on-line setup in off-line mode (no protons)
- 3.5×10^{18} protons on n_TOF target

Experimental Area: EAR2

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i) Scientific justifications investigating the ^{53}Mn(n, γ) reaction

The understanding of the origin of short lived radionuclides like ^{26}Al, ^{44}Ti or ^{53}Mn is one of the major goals of cosmochemistry [1]. Signals carried by a least modified and chemically unaltered class of components such as calcium- and aluminium-rich inclusions or chondrules are essential to constrain the conditions during the formation of our Sun [2]. In addition, they can establish a chronology of the first million years of developments in the solar system [3].

There are different production routes for short lived radionuclides during the development of massive stars. Aluminium-26 and Iron-60 e.g. originates most probably in young solar systems from the outlet of Wolf-Rayet stars [3], but this production path is rather unlikely to explain the initial content of ^{53}Mn in the early solar system [4]. Another possibility to produce cosmogenic radionuclides would be via spallation reactions induced by high energetic galactic radiation. The observation of ^{60}Fe in deep sea ferromanganese sediments [5] and its interpretation as signal of freshly injected matter ejected from a nearby super nova initiated the search of other short lived cosmogenic radionuclei on Earth. In [6] it was deduced, based on model calculations from [7] and [8], that a super nova explosion in a distance of 15 pc to 120 pc would introduce a significant signal to terrestrial archive. It is clearly shown in Fig. 1 that the signal of ^{53}Mn exceeds more than one order of magnitude the expected contribution of the production in spallation reactions with the galactic background radiation. Therefore, nowadays it is commonly assumed that ^{53}Mn like ^{44}Ti will be produced dominantly by explosive episodes of star development.
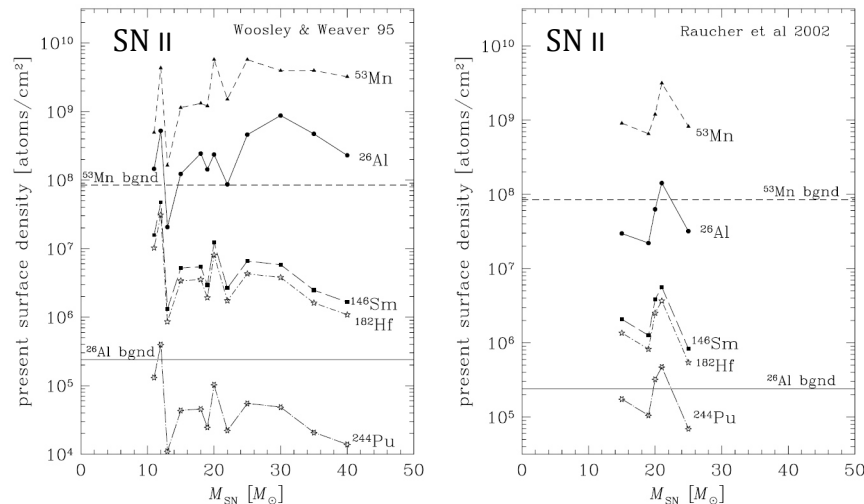

Fig. 1: Predicted terrestrial surface densities as function of the progenitor mass for different short lived radionuclides. In addition the expected input originating from the galactic background is indicated. (Figure taken from [6])

Model calculation of the production of short lived radionuclides in core collapsed super novae showed that a tuning of the model parameters can provide the observed initial content of ^{26}Al and ^{41}Ca, but results in an over production of the initial abundance of ^{53}Mn ([9], [10]). A common cure for that problem will be the use of adjusted mass cuts excluding the innermost layers containing the majority of synthesized ^{53}Mn from the ejection process [11]. An improved understanding of the involved nuclear reaction is mandatory for a better understanding of processes in super novae.

In the outer shells of core-collapse super novae conditions are present where photodisintegration of heavy elements play an important role for the production of p-nuclei, the so called γ–process (see e.g. [12]). Starting from a stable seed nuclei the γ–process evolves with a series of (γ, n) reactions till the neutron binding energy exceeds the proton binding energy or the α-particle separation energy. At such break-point - typically a closed neutron-shell - the development path is deflected to isotopes with a lower atomic-number. According to the particular conditions these photo-disintegration reaction are in competition with neutron, proton or α-particle induced reactions.

In [13], hydrodynamic simulations were combined with nuclear network calculations for p-process nucleo-synthesis for massive white dwarf thermonuclear super novae. It was shown, that in general, using a solar metallicity almost all p-nuclides heavier than nickel are produced in similar amounts like ^{56}Fe if normalized to the solar abundances. These calculations ([13], [14]) revealed also that in such scenarios the production of ^{54}Fe as a light p-nuclide is in direct competition with the neutron capture of ^{53}Mn (see Fig. 2).
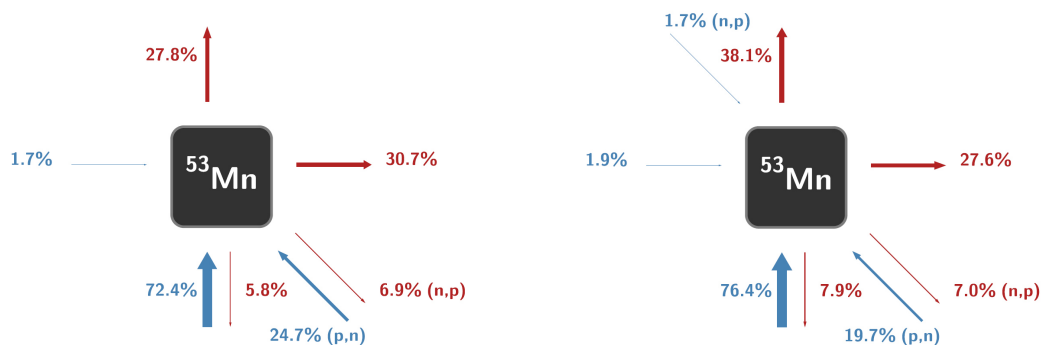

Fig. 2: Integrated ^{53}Mn production in SN Ia for different pre-explosion abundance patterns ([13], [14]) emphasizing the importance of the (n, γ) channel (figures adapted from [13], [14])

In a parameter study [15], the dependence of the final mass fraction of short lived radio-nuclides from the peak temperature and peak after the re-bounce shock was investigated. Here, the initial electron fraction Y_e, i.e. the ratio of the proton number to the total nucleon number before the super nova ignition, was used as a fixed parameter. The shock wave generates temperatures from 4×10^9 K to 10^{10} K and density variations of one or two orders of magnitude. The value of the peak density can be constrained due to the observation of ^{60}Fe as super nova ejecta on Earth to be in the range of 10^5 g/cm^3 to 5×10^6 g/cm^3 (see Fig. 3). In contrast to ^{44}Ti, the final mass fraction of ^{53}Mn is sensitive to the assumed initial Y_e value. In a more neutron rich environment the production of ^{53}Mn will be massively enhanced. The determination of the ejected amount of ^{53}Mn can be used to constrain this parameter, provided that the neutron reactions to and from ^{53}Mn are well determined.

In both discussed cases a more detailed knowledge on possible reaction paths both regarding production and destruction of ^{53}Mn is needed. In the frame of an ongoing project to gain exotic radionuclides from accelerator waste this includes a re-determination of the half-life is envisaged [16], as well as the ^{53}Mn(n, γ) cross section which can be investigated using the in ERAWAST provided amounts of this isotope. The enhanced neutron flux in the n_TOF EAR-2 is expected to enable a measurable signal in the range from 25 meV to 10 keV, covering the expected region of the first resonances. In addition we anticipate extending this region possibly up to 100 keV.
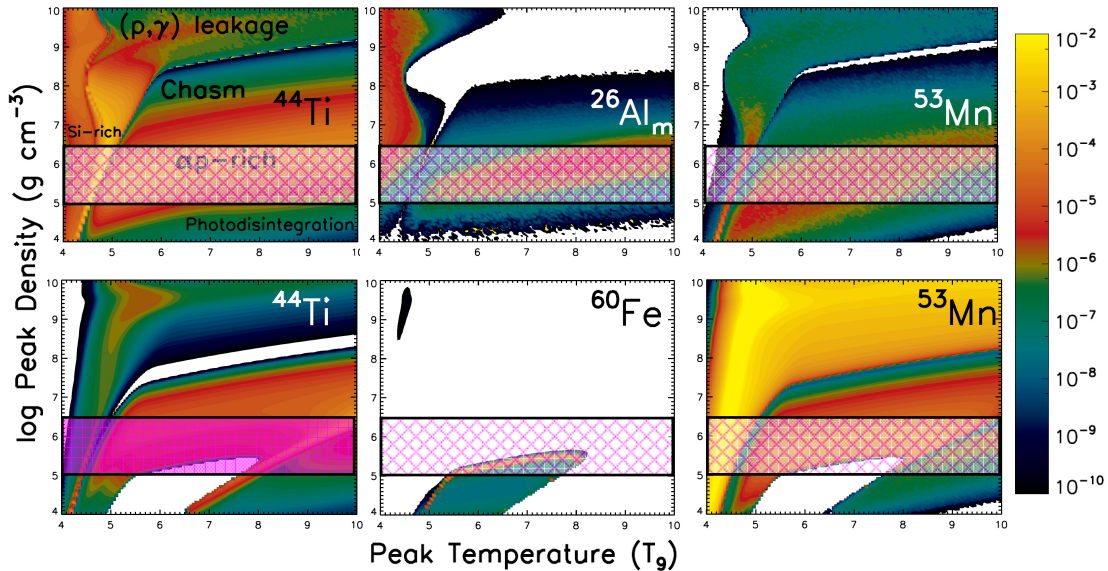
Fig. 3: Final mass fractions in the peak temperature–density plane of a core collapsed super nova. The top row shows parameter studies for Y_e=0.52. The bottom row shows results for Y_e=0.48, where the production of ^{53}Mn is strongly enhanced. (figures taken from [15])

ii) Radiation safety issue using the off-line setup

According to the Swiss radiation protection ordinance [17] (in the following called StSV) the work with radioactive substance must be performed in a way guarantying that the exposure of human beings is as low as possible and does not exceed the dose rate limit for occupationally radiation-exposed persons. Therefore, the radiation safety limits according to the StSV must be taken into account for the preparation and execution of experiments as well as for the post processing of the samples.

The body dose rate limit of non-monitored persons outside of controlled zones must not exceed 20 µSv/week (StSV article 59 paragraph 1). In the case that persons will not stay permanently in that region a limit of 100 µSv/week must be respected. Taking into account a working time of 40 h per week the activity per 01.06.2015 of the proposed experiment will respect these.

We will here discuss the problems rising from the use of the ISOLDE off-line ion-source test setup for a mass separation of ^{53}Mn using the chemically separated stock solution. In Tab. 1 judgment parameters of the StSV and our expectations of both relevant isotopes are compiled. The total available activity of ^{53}Mn is less than the free limit and can therefore from the Swiss radiation protection law treated as non-radioactive.

The amount of ^{54}Mn, a by-product of the spallation reactions with the steel specimens will drop below the license limit at the end of this year. From this time the StSV does not any more demand the exclusive handling inside a controlled zone (StSV article 69 paragraph 1). Never the less, general radiation safety rules for handling, transport or disposal of radioactive substances must be fulfilled.

The main quantity of the used substances will be remaining in the extraction structure of the ion-source (more than 90%). This part, a cylindrical structure 5.0 cm length 2.5 cm diameter made out of tantalum, will be extracted and used for recovery of ^{53}Mn at PSI. The

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remaining radioactive parts of these extraction structures will be afterwards conditioned for a final disposal and temporarily stored at PSI.

Tab. 1: Radiation safety Limits according the StSV and expected activity of ^{53}Mn and ^{54}Mn in the proposed experiment

Radiation safety parameter	^{53}Mn	^{54}Mn
Free Limit (LE)	300 kBq	10.0 kBq
License Limit (LA)	100 MBq	4.0 MBq
Dose equivalent at a depth of 10 mm in 1.0 m distance ($H_p(10)$) per LA	<0.1 µSv/h	0.5 µSv/h
Surface contamination out side controlled areas (CS)	1 kBq/cm²	100 Bq/cm²
Activity in sample per 01.06.2015	173 kBq	2.7 MBq
Dose equivalent Hp(10) per 01.06.2015	<0.0017 µSv/h	0.34 µSv/h
Release into ion beam-line per 01.06.2015	4.3 kBq	81 kBq
Contamination of inner surface* per 01.06.2015	0.00009 CS 0.09 Bq/cm²	0.016 CS 1.6 Bq/cm²

* approximated inner surface of ion-beam line ≈ 50000cm²

The release of activity into the ion-beam line would be the most problematic if a dismantling of the off-line device will be performed within the next 5 years. The total introduced activity of ^{54}Mn is expected to be about 68 kBq, taking into account an ionisation yield of 2.5% of manganese. This activity will drop below the free limit according to StSV at the end of 2017. In the case of an early disassembling the ion-beam line or the parts of it exceeding surface contamination limit would have been temporarily stored until the remaining activity is decayed below the free limit (LE).

We agree with the opinion of the INTC that this might be in some cases inconvenient and enhance the efforts to handle parts of this setup. All these aspects exclude the use of the off-line ion-source test setup to perform the envisaged mass separation.

iii) Use of ISOLDE on-line separation setup

The use of the ISOLDE on-line separator was proposed from the INTC due to the expected radiation safety difficulties connected with the ^{54}Mn content in the prepared stock solution.

We must apologize here for a mistake in our estimation of the separation time. The presented value of 10 days to separate the amount of 1×10^{17} atoms of ^{53}Mn was calculated taking erroneous into account a beam-transmission efficiency of 2.5%. In reality this parameter is well above 85%. The limiting process will be the ionization of manganese inside the FEBIAD source, which reaches a value of 3.0%. The corrected parameters of the mass separation are summarized in Tab. 2. These vales leads to a separation time of 5 h per 1×10^{17} atoms of ^{53}Mn, i.e. in total 25 h for the needed 5×10^{17} atoms of ^{53}Mn in the final sample. We expect that additional time will be needed for setup and tuning the ion-source. Therefore, in total 48 h beam time of the ISOLDE on-line setup without protons are requested to perform the mass separation of the manganese sample.

We will underline that this is the first attempt of a collaboration between ISOLDE and n_TOF. The possibility to produce mass separated samples of rare isotopes would not only ehnace the quality of the obtained results but also enables in some cases the use of specific isotopes as in the present case proposed.

Tab. 2: Expected performance parameters for an off-line separation using the ISOLDE on-line separation facility.

Parameter	Value
Cr-Mn vapor pressure ratio at 1250 K	$1.5{\times}10^{-4}$
Ionization yield at 1250 K	3.0%
FEBIAD output current	100 µA
Ion-transmission in on-line separator	>85%
Output of Mn+	$6.2{\times}10^{14}$ pps
Mass suppression Δmass = 1	$> 10^3$
Mass suppression Δmass > 1	$> 10^4$
Separation time per 10^{17} atoms ^{53}Mn	5 h
Separation time per $5{\times}10^{17}$ atoms ^{53}Mn	25 h
Remaining content vanadium	$8.7{\times}10^4$ atoms
Remaining content chromium (only ^{53}Cr)	$2.4{\times}10^{12}$ atoms
Remaining content manganese (only ^{55}Mn)	$5.8{\times}10^{15}$ atoms
Remaining content iron	$4.9{\times}10^9$ atoms

Summary of requested protons:

- 6 shifts at ISOLDE on-line setup in off-line mode (no protons)
- $3.5{\times}10^{18}$ protons on n_TOF target for usage in EAR-2

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