

# ERAWAST – NUCLEAR CHEMISTRY FOR NUCLEAR SCIENCES

*Dorothea Schumann for the ERAWAST collaboration*

*Paul Scherrer Institute Villigen, Switzerland*

## INTRODUCTION

Five years ago, the first ESF-sponsored exploratory ERAWAST workshop was organised at PSI. ERAWAST means: Exotic Radionuclides from Accelerator Waste for Science and Technology. The initiative was aimed to explore the possibilities to isolate rare exotic radionuclides from existing accelerator waste of high-power accelerator facilities on the one side and identify potential users for those isotopes on the other side. In total, 30 participants from 12 countries decided to create an international collaboration and established a working program. Not all of these, partially very ambitious plans could be realized. But now, after five years, it is time to have a look to the achievements as well as to define the focus of activities for the near, and also for the long-term future.

## ISOTOPES AND THEIR SOURCES

Exotic isotopes can be chemically separated from exposed components of the high-power accelerator facilities of PSI and/or from other sources (nuclear research reactors, other accelerator facilities). Since the components are hit with protons and secondary particles up to 590 MeV, the entire spectrum of radionuclides with mass numbers up to one unit higher than the target mass, have to be expected.

The following sources containing some examples for rare exotic isotopes are currently available at PSI:

SINQ cooling water	$^7\text{Be}$ , $^{22}\text{Na}$ , $^{54}\text{Mn}$
Copper beam dump	$^{44}\text{Ti}$ , $^{53}\text{Mn}$ , $^{26}\text{Al}$ , $^{60}\text{Fe}$
Graphite targets	$^7\text{Be}$ , $^{10}\text{Be}$
Steel samples from the STIP program	$^{44}\text{Ti}$ , $^{53}\text{Mn}$ , $^{26}\text{Al}$
Lead samples from SINQ targets	$^{207}\text{Bi}$ , $^{182}\text{Hf}$ , $^{148}\text{Gd}$

## CHEMICAL SEPARATION TECHNIQUES

For the chemical separations, classical radiochemical techniques like ion exchange, co-precipitation and liquid liquid extraction are applied. Very often, these methods have to be modified due to the very low amount of the desired isotopes in comparison to the contaminations and matrix elements. In several cases, the procedures have to be performed in hotcells with automated systems or remote controlled, due to the high dose rate of the components.

## FIRST RESULTS

In the previous 5 years, several materials had been used at PSI to separate isotopes for certain successful experiments. The following list gives an overview on the achievements and the used material.

$^{207}\text{Bi}$ from lead	calibration source [1]
$^{44}\text{Ti}$ from copper	calibration source [2]
$^7\text{Be}$ from graphite	precise measurement of the half-life [3]
$^{60}\text{Fe}$ from copper	target for neutron capture cross section [4]
$^{60}\text{Fe}$ from copper	determination of the half-life [5]
$^{63}\text{Ni}$ from LANL	neutron capture cross section at n_TOF [6]
$^7\text{Be}$ from SINQ water	environmental studies [7]
$^{68}\text{Ge}$ activation in SINQ	AMS measurements [8]

## ISOTOPES NOW OR SOON AVAILABLE FOR PLANNED EXPERIMENTS IN THE NEAR FUTURE

$^{44}\text{Ti}$	50 MBq for target FZR
$^{44}\text{Ti}$	100 MBq for radioactive beam (CERN, TRIUMF)
$^{53}\text{Mn}$	$\sim 10^{18}$ atoms for the half-life measurement (PSI)
$^7\text{Be}$	1 GBq for $^7\text{Be}(n,\alpha)$ (SOREQ Nuclear Center)
$^{60}\text{Fe}$	$\sim 10^{15}$ atoms for the re-measurement of the half-life (PSI and Uni Vienna)
$^{60}\text{Fe}$	$\sim 10^{15}$ atoms for a target for thermal neutron capture cross section (Uni Vienna)
$^{63}\text{Ni}$	several MBq for the re-measurement of the half-life
$^{26}\text{Al}$	150 Bq
$^{10}\text{Be}$	80 $\mu\text{g}$

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# EXOTIC RADIONUCLIDES EXTRACTION FROM PROTON IRRADIATED COPPER BEAM DUMP AND SINQ COOLING WATER

*Marin Ayranov, Dorothea Schumann, Rugard Dressler, Niko Kivel*

*Paul Scherrer Institute, Villigen, Switzerland*

## INTRODUCTION

In the frame of the ERAWAST initiative (Exotic Radionuclides from Accelerator Waste for Science and Technology) which started at PSI in 2006 [1], two examples concerning the big-scale separation of exotic radionuclides are presented in this work. One source for gaining these isotopes are samples from an irradiated copper beam dump, containing  $^{44}\text{Ti}$ ,  $^{60}\text{Fe}$ ,  $^{36}\text{Cl}$ ,  $^{32}\text{Si}$ ,  $^{29}\text{Al}$ ,  $^{53}\text{Mn}$ , and  $^{59}\text{Ni}$ . For the separation of  $^{7,10}\text{Be}$  the cooling water of the spallation neutron source (SINQ) of PSI was used.

## COPPER BEAM DUMP

After dismantling of a copper beam dump from the pions cancer therapy station, 500 grams of highly-active copper chips were collected as a source of exotic radionuclides. The analyses performed by gamma spectrometry, Liquid Scintillation Counting (LSC) and Accelerator Mass Spectrometry (AMS) estimate that about 100 MBq  $^{44}\text{Ti}$ , 500 kBq  $^{53}\text{Mn}$ , 7 kBq  $^{26}\text{Al}$ , 8 MBq  $^{59}\text{Ni}$  and 5 kBq  $^{60}\text{Fe}$  are available in the collected copper chips [2]. Due to the high activity of  $^{60}\text{Co}$ , approx. 5 GBq in total, the separation should be implemented using appropriate shielding, e.g. in a hot cell. The aim was to develop a simple, selective, efficient, and easy to accommodate for remote manipulation procedure for the separation of  $^{26}\text{Al}$ ,  $^{59}\text{Ni}$ ,  $^{53}\text{Mn}$ ,  $^{44}\text{Ti}$  and  $^{60}\text{Fe}$  from gram amounts of the copper beam dump.

The separation procedure is a combination of selective precipitation and ion exchange. Copper as the main matrix element interferes with the separation of all elements of interest. For this reason, the copper matrix is selectively precipitated as  $\text{CuS}$  by saturation with  $\text{H}_2\text{S}$ . Further the rare radionuclides were separated by gradient elution on a Dowex 1x8 anion exchange column. Al and Ni, which are not retained on the anion-exchanger, are additionally separated by cation-exchange and extraction chromatography. The full scale system is installed in a hot cell where samples with high activity can be handled.

In order to evaluate the reliability and functionality of the system extensive tests have been done. During the test period 15 g in total of the proton irradiated copper beam dump were processed for separation of  $^{26}\text{Al}$ ,  $^{59}\text{Ni}$ ,  $^{53}\text{Mn}$ ,  $^{44}\text{Ti}$  and  $^{60}\text{Fe}$ . Total 6 MBq  $^{44}\text{Ti}$ , and  $4 \cdot 10^{16}$  atoms  $^{60}\text{Fe}$  are separated in n.c.a. form. The procedure manages as well the generated liquid wastes containing high level of  $^{60}\text{Co}$  activity, reducing significantly their volume.

## SINQ COOLING WATER

$^7\text{Be}$  as well as  $^{10}\text{Be}$  are key radionuclides for investigations of several astrophysical processes and phenomena. One of the "hot topics" is the half-life of  $^{10}\text{Be}$ , where the literature values differ from  $1.34\text{-}1.51 \cdot 10^6$  a [3, 4]. Additional measurements are, therefore, urgently needed. One possibility is the use of LSC for the determination of the activity and Inductively Coupled Mass Spectrometry (ICP-MS) for measuring the number of atoms. The calibration of the mass discrimination of the ICP-MS requires at least two isotopes of known amounts, and since Be has only one stable isotope ( $^9\text{Be}$ ),  $^7\text{Be}$  can serve as the second marker.

Other applications of  $^7\text{Be}$  are the study of key reactions concerning the solar neutrino flux, in particular the reaction  $^7\text{Be}(p,\gamma)^8\text{B}$  [5] and the measurement of the cross section of the  $^7\text{Be}(n,\alpha)\alpha$  reaction and the problem of primordial  $^7\text{Li}$  [6].  $^7\text{Be}$  is produced in considerable amounts in the cooling water ( $\text{D}_2\text{O}$ ) of the SINQ facility at PSI by spallation reactions on  $^{16}\text{O}$  with the generated fast neutrons. By-products can be nearly neglected, so that this cooling water establishes an ideal source for highly active  $^7\text{Be}$ -samples.

An ion exchange filter containing 1 liter of the mixed-bed ion exchanger LEWATIT was installed as a by-pass for the cooling water into the cooling loop of SINQ for 6 weeks. The collected activity of  $^7\text{Be}$  was in the range of TBq. Due to the high dose rate a separation system was designed and installed in a hot-cell. After a cooling period of 12 months the ion-exchanger was transferred in the hot cell and  $^7\text{Be}$  stripped with 1 M HCl accompanied with main impurities of  $^{22}\text{Na}$ ,  $^{54}\text{Mn}$ , and  $^3\text{H}$ . Further  $^7\text{Be}$  was purified by cation exchange on a Dowex 50x8 column. As result, 10 GBq of highly pure  $^7\text{Be}$  sample that can be used as a second standard for the  $^{10}\text{Be}$  half-life determination was prepared.

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# SEPARATION OF $^{44}\text{Ti}$ , $^{26}\text{Al}$ , AND $^{53}\text{Mn}$ FROM IRRADIATED STAINLESS STEEL

M. Bunka, M. Ayranov, D. Schumann, Y. Dai

Paul Scherrer Institute, Villigen, Switzerland

## INTRODUCTION

$^{44}\text{Ti}$ ,  $^{54}\text{Mn}$  and  $^{26}\text{Al}$  are attractive for several scientific applications. Proton-irradiated steel samples at PSI contain those isotopes. For the extraction and purification of  $^{44}\text{Ti}$ ,  $^{26}\text{Al}$  and  $^{54}\text{Mn}$  from the complex stainless steel matrix a multistep separation system was developed and optimized [1, 2].

## MATERIALS AND EXPERIMENTAL

To study radiation damage effects on construction materials induced by high-energy protons and spallation neutrons, several experiments have been done at PSI using special positions within the targets of the Swiss spallation neutron source (SINQ). The first SINQ target-irradiation program (STIP I) was running from July 1998 to December 1999. More than 1500 samples of different types of structure materials were irradiated in this experiment [3].

Our materials – martensitic steels (Optifer, Optimax A and Optimax C) originates from these first STIP experiment. There are 38 samples weighting from 1 g to 2 g and containing about 300 MBq  $^{44}\text{Ti}$ , 70 MBq  $^{54}\text{Mn}$  (as a radioactive marker for the unknown amount of  $^{53}\text{Mn}$ ), 70 MBq  $^{60}\text{Co}$  as well as  $^{26}\text{Al}$  in the range of a few kBq.

These martensitic steels contain about 88% Fe, 9,4% Cr, less than 1% Mn, Mo and Ni. Optimax A and Optimax C contain less than 0.01% stable Ti. To separate the desired elements Mn, Al and Ti from all other elements a multistep separation has been developed (Fig. 1).

After the dissolution of steel in 8 M HCl and a few mL of conc.  $\text{HNO}_3$ , extraction with methyl-isobutyl ketone is performed to separate the great part of iron and molybdenum. Next step is the precipitation with concentrated  $\text{NH}_3$ . Then the precipitate containing Fe, Ti, Cr and Mn is dissolved in HCl and precipitated with a mixture of sodium hydroxide and hydrogen peroxide. In the solution stay chromium and aluminium. To separate  $^{26}\text{Al}$  from Cr the solution is made acidic, Fe(III) as carrier is added to the solution and Al(III) and Fe(III) are precipitated with conc.  $\text{NH}_3$ .

The precipitate from  $\text{NaOH} + \text{H}_2\text{O}_2$  precipitation containing Fe, Mn and Ti is dissolved in 8M HCl and extraction with methyl-isobutyl ketone is repeated. For the determination of

the elements  $\gamma$ -spectrometry is used.

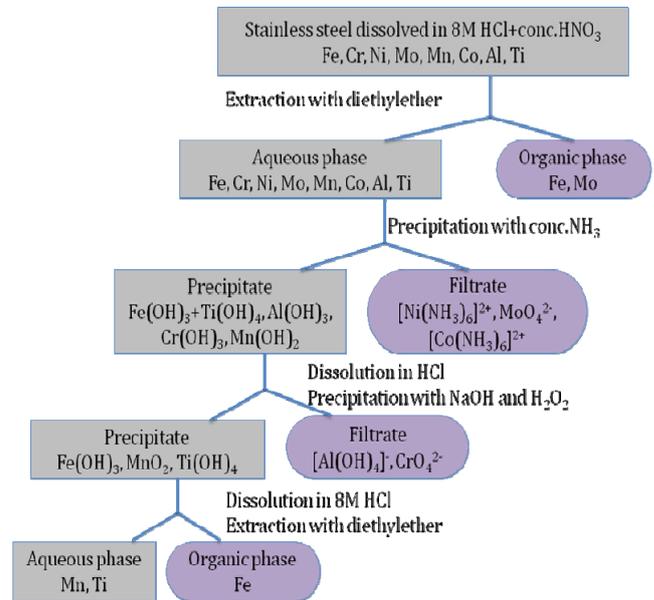


Fig. 1: Separation scheme

The final separation of Ti and Mn was performed using ion exchange chromatography. Overall chemical yields for the entire separation process of 50–60% for Mn and about 90–95% for Ti were reached.

At the moment, about 100 Bq  $^{26}\text{Al}$ , 10 MBq  $^{54}\text{Mn}$  and 60 MBq  $^{44}\text{Ti}$  are already separated, which corresponds to about one forth of the total amount of available samples. We expect to have the remaining samples ready till end of this year. However, the STIP-program is a long-lasting material research program (currently STIP6 is running), therefore sample material containing the desired isotopes with a factor of at least one order of magnitude higher can be expected to be available in the future.

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## ACCELERATOR WASTE, ACCELERATOR MASS SPECTROMETRY AND MORE

*Gunther Korschinek*

*Physik Dept. TUM, Germany*

Exotic radionuclides extracted from accelerator waste are a unique treasure in different fields of science. One example is accelerator mass spectrometry (AMS) where the availability of accurate calibration standards is a prerequisite for any measurement. In many cases standards, determined by their half-life and the activity, are employed. However, these half-lives have in most cases not the precision needed, therefore the isotopic ratios, used in AMS, get as well these uncertainties. In those cases conventional mass spectrometry might help because of their much higher accuracy achievable than in AMS. But these methods have less sensitivities and therefore a high isotopic concentration is needed, that however can be found e.g. in an accelerator beam dump, which got a long built up time for these isotopes. In the past, because of a close collaboration between the scientific groups at the PSI and us, we were

able to determine a very precise half-life of  $^{60}\text{Fe}$  [1,2], but also accurate  $^{60}\text{Fe}$  standards have been produced. The talk will cover past achievements but also follow-up projects for other long-lived radionuclides will be presented. Besides these applications, exotic radionuclides might serve as well as small monoenergetic neutrino sources of high intensities. There is a recent indication for the existence of so-called sterile neutrinos different from muon, tau, and electron neutrinos. This could be confirmed by the help of a strong mono energetic neutrino source as it is for example  $^{55}\text{Fe}$ . A possible measurement will briefly be outlined.

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## A HIGH-INTENSITY SOURCE OF STELLAR-ENERGY NEUTRONS FOR RARE-NUCLIDE ACTIVATIONS

*Michael Paul*

*Racah Institute of Physics, Hebrew University, Jerusalem, Israel  
(for the LiLiT collaboration)*

The recent progress in high-intensity linear accelerators requires development of targets capable of sustaining their beam power. The issue is particularly acute for the generation of accelerator-based neutrons via the  $^7\text{Li}(p,n)$  reaction due to the poor thermal properties of solid lithium. We are commissioning the high-power windowless Liquid-Lithium Target (LiLiT) designed to dissipate a beam power up to several kW. The forced-flow jet (3-10 m/s) of liquid lithium serves both as neutron-producing target for a proton beam (1.9-2.5 MeV, 2-4 mA) and as power beam dump. The LiLiT setup is presently tested with a 20-kV electron

gun in the same power range and will be installed at the Soreq Applied Research Accelerator Facility (SARAF). With a  $1910 \pm 15$  keV proton beam, the expected neutron energy distribution is well fitted by a Maxwellian flux at  $kT=28$  keV with an energy cutoff of  $\sim 140$  keV and the expected intensity is  $2.3 \times 10^{10}$  n/s.mA. Neutron activations of selected nuclides relevant to stellar nucleosynthesis are planned. The combination of the high-intensity neutron source and future availability of radioactive-nuclide targets (e.g.  $^{53}\text{Mn}$ ,  $^{59}\text{Ni}$ ,  $^{60}\text{Fe}$ ) through the ERAWAST project is particularly interesting.

## NEW EXOTIC AND NON-STANDARD RADIONUCLIDES IN AMS

*A. Wallner<sup>1,2</sup>, K. Buczak<sup>1</sup>, A. Plompen<sup>3</sup>, D. Schumann<sup>4</sup>, V. Semkova<sup>3</sup>*

*<sup>1</sup>VERA Laboratory, Faculty of Physics, University of Vienna, Austria, <sup>2</sup>Australian Nuclear Science and Technology Organisation, PMB 1, Menai, NSW 2234, Australia, <sup>3</sup>Neutron Physics Unit, EC-JRC-IRMM, Geel, Belgium, <sup>4</sup>Paul Scherrer Institute, 5232 Villigen PSI, Switzerland*

Five isobaric interferences represent one of the major limitations in mass spectrometry. In the very few cases in AMS where nature allows isobaric-free measurements, lowest background levels are accessible. Such conditions are given, if the isobar does not form stable negative ions either as atomic ion, or by choosing a suitable molecular species; well-known examples are e.g.  $^{14}\text{C}^-$ ,  $^{26}\text{Al}^-$ ,  $^{129}\text{I}^-$ , or  $^{41}\text{CaH}_3^-$ , respectively, and also for the unstable isotopes in the mass range above Bi.

In this work, we will present a few additional cases where isobaric interference is completely excluded, among them  $^{55}\text{Fe}$ ,  $^{68}\text{Ge}$  and  $^{202}\text{Pb}$ . Applications will be discussed where the exceptional sensitivity of AMS offers important insights to such different fields like nuclear astrophysics, nuclear physics and general physics issues.

VERA, a dedicated AMS facility, based on a 3-MV tandem, featuring high mass resolution in combination with efficient background suppression and an automated measurement procedure, allows to transport all nuclides from hydrogen to the actinides through the system up to the detector stations. Such a facility is well suited for developing the tuning and measurement procedures for new and non-standard isotopes.

We will demonstrate the actual measurement limits of such radionuclides for the VERA facility and results for selected applications. In order to generate final values AMS usually relies on the parallel measurement of reference materials. We will discuss the production of such materials for these non-standard AMS nuclides.

## MEASUREMENTS POSSIBILITIES WITH RADIOACTIVE SAMPLES AT CERN n\_TOF FACILITY

*E. Chiaveri*

*on behalf of the n\_TOF Collaboration, CERN, Switzerland*

The neutron time-of-flight facility n\_TOF at CERN, Switzerland, operational since 2000, combines a very high instantaneous neutron flux, an excellent TOF resolution, a low intrinsic background and a wide range of neutron energies, from thermal to GeV neutrons.

These characteristics provide a unique possibility to perform neutron-induced capture and fission cross-section measurements, for applications in nuclear astrophysics and in nuclear reactor technology.

The most relevant measurements performed up to now and foreseen for the future will be presented in this contribution.

The overall efficiency of the experimental program and the range of possible measurements achievable with the construction of a second experimental area (EAR-2), vertically located 20 m on top of the n\_TOF spallation target, might offer a substantial improvement in measurement sensitivities and will open the possibility to measure neutron-induced cross-section on rare and unstable samples, which can be obtained only in low amounts.

A feasibility study of possible realisation will be also presented.

## RADIOACTIVE ION BEAMS AT ISOLDE, CERN: NEW DEVELOPMENTS AND NEEDS

*Thierry Stora*

*ISOLDE Target and Ion Source Development Team, CERN, Switzerland*

The ISOLDE facility has been operating at CERN for more than 40 years. It is one of the first facilities to deliver radioactive ion beams based on the Isotope Mass Separation OnLine technique. The number of available radioisotopes covers today more than 1000 of 70 different chemical elements. This figure in constant progression results from a combination of a development program related to the techniques used to generate the beams on the one hand, and a versatile 1.4 GeV proton primary driver, the Proton Synchrotron Booster, on the other hand, that can exploit a wide range of different nuclear reactions. The developments in target and ion sources are prioritized according to the physics program at the Group for Upgrade of ISOLDE meetings.

The facility is also undergoing an important upgrade program, HIE-ISOLDE, and will benefit from the ongoing CERN injectors' complex renovation project, first with the start-up of Linac 4, followed by the PSB upgrade.

In parallel, the new mode of operation of the CERN injectors' complex to fulfill the needs of the LHC has shifted from an operation/maintenance yearly cycle towards a bi- or tri-yearly cycle; this induces new constraints, and needs, for the ISOLDE facility. In that respect new prospects for the production of beams of long-lived isotopes are thus under investigation for off-line operation periods, to cover a so-called "winter physics" program.

## PLAN TO MEASURE THE NEUTRON CAPTURE CROSS-SECTION OF $^{60}\text{Fe}$ WITH COLD NEUTRONS AT THE PGAA FACILITY IN MUNICH

*S. Söllradl<sup>1</sup>, L. Canella<sup>2</sup>, P. Kudejova<sup>2</sup>, Zs. Reva<sup>2</sup>, R. Dressler<sup>3</sup>, D. Schumann<sup>3</sup>, M. Ayrano<sup>3</sup>, A. Türler<sup>1</sup>*

*<sup>1</sup>Univ. Bern & PSI, <sup>2</sup>Forschungsneutronenquelle Heinz Maier-Leibnitz, <sup>3</sup>Paul Scherrer Institut*

During the maintenance break of the Research Reactor the PGAA (prompt gamma activation analysis) instrument has also been upgraded at FRM II in Munich. The shielding was improved and an automated system for changes between a collimated flight tube and an elliptical neutron focus were introduced.

At the beam line of the PGAA facility cold neutrons with

the average energy of 1.83 meV are available with a flux of up to  $6.07 \cdot 10^{10} \text{ n s}^{-1} \text{ cm}^{-2}$ . An experiment is planned, to measure the neutron capture cross-section of  $^{60}\text{Fe}$  in a low mass, enriched sample. The presentation will give an overview about the latest changes of the instrument, a short introduction about PGAA and an outline of the planned project to measure the neutron capture cross sections of  $^{60}\text{Fe}$  extracted from a former PSI Copper beam dump.

## IS IT POSSIBLE TO STUDY THE $^{44}\text{Ti}(\alpha, p)^{47}\text{V}$ REACTION WITH A RADIOACTIVE TARGET?

*Daniel Bemmerer, Tariq Al-Abdullah<sup>1</sup>, Rugard Dressler, Dorothea Schumann<sup>2</sup>*

*<sup>1</sup>Helmholtz-Zentrum Dresden-Rossendorf, Germany, <sup>2</sup>Paul Scherrer Institute Villigen, Switzerland*

The radioactive nuclide  $^{44}\text{Ti}$  is believed to be produced in the  $\alpha$ -rich freezeout preceding supernova explosions. The  $\gamma$ -lines from its decay have been observed in space-based  $\gamma$ -observatories for the Cassiopeia A supernova remnant. The rates of the nuclear reactions governing the production and destruction of  $^{44}\text{Ti}$  should therefore be known with precision. The  $^{44}\text{Ti}(\alpha, p)^{47}\text{V}$  cross section has so far been

studied only in inverse kinematics, with radioactive  $^{44}\text{Ti}$  beams. These data do not reach the astrophysically relevant energies. A feasibility study is currently underway to determine whether the reaction can also be studied in direct kinematics, using a  $^{44}\text{Ti}$  target, an  $\alpha$  particle beam and particle detectors. Preliminary results and an outlook will be given.

## (n, $\gamma$ ) REACTIONS AT THE FRANKFURT NEUTRON SOURCE “FRANZ”

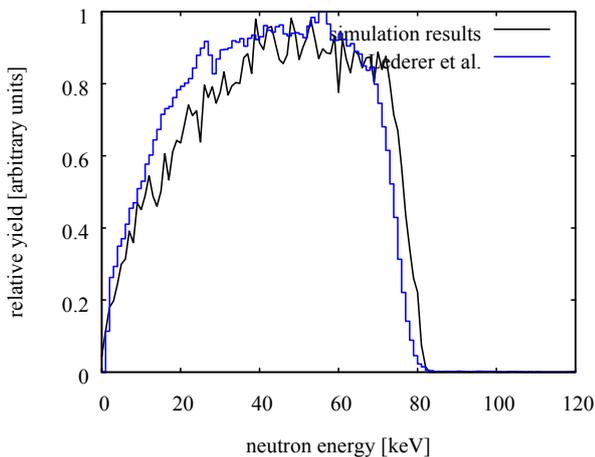
Stefan Schmidt<sup>1</sup>, Gitai Feinberg<sup>2</sup>, Oliver Meusel<sup>1</sup>, Michael Paul<sup>2</sup>, Ralf Plag<sup>1</sup>, Rene Reifarth<sup>1</sup>, Kerstin Sonnabend<sup>1</sup>

<sup>1</sup>University of Frankfurt, Germany - <sup>2</sup>University of Jerusalem, Israel

### (n, $\gamma$ ) REACTIONS AND FRANZ

With modern observational techniques, refined theoretical models and experimental possibilities through large accelerators, the understanding of nucleosynthesis inside stars has become a major topic for astrophysics. An important ingredient for understanding observed element abundances are neutron capture cross section data. Recent accelerator facilities have become able to reproduce the neutron flux inside Red Giant stars via activation techniques, therefore enabling the experimental determination of (n,  $\gamma$ ) cross sections of nuclei relevant to the slow neutron capture process, or s-process. Isotopes produced in this process are not very far off from the valley of stability, yet (n,  $\gamma$ ) cross sections have almost exclusively been measured for stable isotopes [1]. In comparison to stable samples, especially short-lived nuclei require higher neutron fluxes in order to overcome the background radiation from the decay. Together with other facilities around the world the Frankfurt Neutron Source at Stern-Gerlach-Zentrum, FRANZ, currently under construction at Goethe University, Frankfurt, Germany, aims at investigating unstable samples: in comparison to previous generation sources, like the Forschungszentrum Karlsruhe (FZK) Van de Graaff accelerator, FRANZ increases the neutron flux by three orders of magnitude.

### NEUTRON PRODUCTION VIA ${}^7\text{Li}(p,n){}^7\text{Be}$



**Fig. 1:** Simulated and measured spectrum for 1.912 MeV ( $\pm 1$  keV). Only neutrons emitted under  $30^\circ$  are

The FRANZ facility will employ the  ${}^7\text{Li}(p,n){}^7\text{Be}$  reaction in order to supply neutrons to subsequent measurements. When integrated over the full solid angle, the distribution of

neutrons produced at  $E_p = 1.912$  MeV has been shown to resemble a Maxwellian distribution at  $kT = 25$  keV [2], which coincides with the typical temperature inside Red Giants. Additionally, neutrons created at this energy are collimated in forward direction. For  $E_p = 1.912$  MeV, i.e. 30 keV above the reaction threshold, the cone of the

emitted neutrons has an opening angle of  $120^\circ$  [2]. The highest neutron fluxes are available in activation experiments where the sample is directly attached to the neutron production target. However, this approach does not allow the measurement of the neutron energy distribution. This can be compensated either experimentally by using time-of-flight measurements instead of activation, thereby sacrificing neutron beam intensity, or theoretically by numerical simulation of the neutron distribution. Similar to PINO [3], a code has been written that converts a six-dimensional proton distribution into a six-dimensional neutron distribution taking into account kinematic collimation, energy loss due to particle stopping as well as angle-dependent  ${}^7\text{Li}(p,n)$  cross sections from [4]. Comparing the predicted spectrum for a given angle to measurements conducted at Physikalisch-Technische Bundesanstalt at Braunschweig, Germany [5] yields that the simulation can indeed predict the neutron energy spectrum very well (Figure 1).

### NEUTRON SAMPLES

With the start of FRANZ, many isotopes of astrophysical interest will be investigated. Among them,  ${}^{10}\text{Be}$  with a half-life of  $1.5 \cdot 10^6$  years is important because its (n,  $\gamma$ ) cross section can also be measured via Coulomb dissociation, thereby providing an alternate way to verify the cross section data [1]. The  ${}^{10}\text{Be}(n,\gamma){}^{11}\text{Be}$  reaction is part of reaction networks relevant to the r-process. The  ${}^{204}\text{Tl}$  isotope exhibits a much shorter half-life of only 3.78 years, yet its capture cross section is very important as it determines the production of  ${}^{204}\text{Pb}$  [6].

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# USING BEAMS OF RECLAIMED $^{44}\text{Ti}$ TO EXPLORE THE MECHANISM OF CORE COLLAPSE IN SUPERNOVAE

*Jennifer Fallis<sup>1</sup> and Alex Murphy<sup>2</sup> for the ERAWAST- $^{44}\text{Ti}$  collaboration*

*<sup>1</sup>TRIUMF, Canada, <sup>2</sup>School of Physics, University of Edinburgh, UK*

Core collapse supernovae are remarkable astronomical events, exhibiting a combination of extreme temperature, density and energy seen nowhere else. They are central to the formation of many heavy elements, and known to be the engines behind many if not all gamma ray bursts. Not surprisingly, there is immense interest in attempting to understand the physics that drives them. However, this is made extremely difficult, both by the complexity of the explosion and the fact that the key processes are occurring deep beneath the surface. Major fundamental uncertainties remain, for example, the explosion mechanism itself and whether or not core collapse supernovae are the site of the r-process.

One of the few methods by which the explosion mechanism might be studied in a reasonably direct way is through comparison of the amount of  $^{44}\text{Ti}$  observed by satellite (via its beta-delayed gamma-ray emission), to the amount predicted to have been generated in the explosion. The importance of  $^{44}\text{Ti}$  lies in the expectation that it is synthesised in the alpha-rich freeze out that occurs in the shock-heated silicon layer that lies just above the detonating core [1]. This is also the location of the so-called mass cut, that is, the boundary between material that is successfully ejected and that which falls back on to the proto-neutron star. Gamma-rays from material that falls back will be unable to escape the dense environment and thus cannot be observed. Hence, comparison of the observed to the predicted production provides a measure of the location of the mass cut. The mass cut is a key hydrodynamic property of supernova models, and constraining this would be of immense help in finally understanding the explosion mechanism.

For a qualitative comparison between the observed and expected  $^{44}\text{Ti}$  to be achieved, the models of core collapse supernova require better nuclear physics input. The et al. [2] explored which nuclear reactions had most impact on the  $^{44}\text{Ti}$  abundances produced in core-collapse, finding that there were relatively few reactions that contributed to the overall uncertainty in  $^{44}\text{Ti}$  production:  $^{40}\text{Ca}(\alpha,\gamma)^{44}\text{Ti}$ ,  $^{44}\text{Ti}(\alpha,\gamma)^{48}\text{Cr}$ ,  $^{44}\text{Ti}(\alpha,p)^{47}\text{V}$ , and  $^{45}\text{V}(p,\gamma)^{46}\text{Cr}$ , together with the ubiquitous  $^{12}\text{C}(\alpha,\gamma)^{16}\text{O}$  and triple- $\alpha$  reactions. A similar study was published recently by Magkotsios *et al.* [3], which finds a similar list of reactions with the three most important listed as:  $^{44}\text{Ti}(\alpha,p)^{47}\text{V}$ ,  $^{40}\text{Ca}(\alpha,\gamma)^{44}\text{Ti}$ , and  $^{45}\text{V}(p,\gamma)^{46}\text{Cr}$ . Also listed, and of possible importance for the tests discussed here, is the reaction  $^{44}\text{Ti}(p,\gamma)^{45}\text{V}$ . A significant finding of both studies is that, aside from the few reactions listed in the papers, the quantity of  $^{44}\text{Ti}$

synthesised was found to be almost completely insensitive to the variations in other reaction rates. Hence, good knowledge of the rates of these few reactions at the relevant temperatures would represent a major step towards a deeper and more robust understanding of core collapse supernovae.

The  $^{12}\text{C}(\alpha,\gamma)^{16}\text{O}$  and triple- $\alpha$  reactions have a wide role in astrophysics and have been the focus of numerous studies, and the  $^{40}\text{Ca}(\alpha,\gamma)^{44}\text{Ti}$  reaction has already been studied using the DRAGON facility at TRIUMF [4]. The focus of this paper are future measurements of  $^{44}\text{Ti}(\alpha,p)^{47}\text{V}$  and the other  $^{44}\text{Ti}$  reactions. Measuring these reaction rates however are far from easy, with one primary difficulty being the acquisition of sufficient  $^{44}\text{Ti}$ , as it cannot currently be produced in sufficient quantities at ISOL facilities. Despite this fact, the cross section of  $^{44}\text{Ti}(\alpha,p)^{47}\text{V}$  has been measured at several energies by Sonzogni et al [5] using a beam of  $^{44}\text{Ti}$  produced in an offline ion source. Due to the limited beam intensity and small  $^{44}\text{Ti}(\alpha,p)^{47}\text{V}$  cross section, however, these measurements were unable to reach astrophysical energies.

Through the ERAWAST project however, it becomes possible to acquire sufficient quantities of previously produced  $^{44}\text{Ti}$  to create the required beams using the TRIUMF offline ion source (OLIS). This would allow for a measurement of the  $^{44}\text{Ti}(\alpha,p)^{47}\text{V}$  reaction rate to be made using the TUDA apparatus. If the efficiency of beam production from OLIS proves to be sufficient, a study of the two other  $^{44}\text{Ti}$  destruction reactions  $^{44}\text{Ti}(\alpha,\gamma)^{48}\text{Cr}$  [2] and  $^{44}\text{Ti}(p,\gamma)^{45}\text{V}$  [3] could also be undertaken at TRIUMF using the DRAGON facility. There is currently on no experimental data on either of these two reactions.

This research will be a joint effort between the TRIUMF Nuclear Astrophysics and Edinburgh Nuclear Physics groups. Support from the Science and Technology Facilities Council, UK has been secured, and Stage 1 approval has been granted by the TRIUMF Experiments Evaluation Committees (EEC). This has allowed us to begin preliminary beam development tests.

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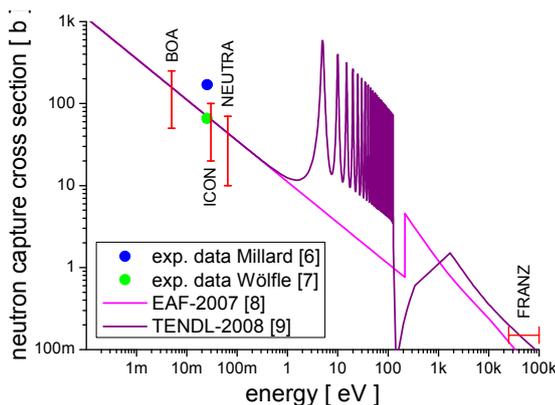
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# PLAN TO MEASURE THE HALF-LIFE AND NEUTRON CAPTURE CROSS SECTION OF $^{53}\text{Mn}$

R. Dressler

Paul Scherrer Institut, Villigen, Switzerland

Short-lived cosmogenic radio-nuclides with half-lives of less than hundred million years (100 Ma) (e.g.  $^{10}\text{Be}$ ,  $^{26}\text{Al}$ ,  $^{36}\text{Cl}$ ,  $^{53}\text{Mn}$ ,  $^{60}\text{Fe}$ ,  $^{59}\text{Ni}$ ) are produced via neutron capture in explosive states of the star development (e.g. super novae explosions or thermally pulsing low mass asymptotic giant branch stars), or via spallogenic processes induced by high energetic protons or neutrons. It is well known that short-lived cosmogenic radio-nuclides were present in the early solar system [1, 2]. Physically relevant parameters in stellar models such as neutron density, temperature, and pressure can be tested and constrained using reliable production rates in neutron capture reactions. Improvements of available data will be essential for the understanding of the galactic stellar evolution of successive stellar generations [3].  $^{53}\text{Mn}$  was discovered by Wilkinson and Sheline [4] in proton induced reactions on enriched  $^{53}\text{Cr}$  targets in 1955. However, the amount of  $^{53}\text{Mn}$  was not sufficient to use it for neutron activation experiments. In 1960 J.P. Shedlovsky [5] showed iron meteorites contain  $^{53}\text{Mn}$  in an amount of about  $5 \times 10^{11}$  atoms per g iron. Using manganese extracted from such meteorites first measurements of the neutron capture cross-section at thermal neutron energy were carried out by H.T. Millard [6] in 1965 giving  $\sigma_{\text{th}} \approx 170$  b and by R. Wölfle et al. [7] in 1972 obtaining  $\sigma_{\text{th}} \approx 66 \pm 7$  b. So far no other measurements of the neutron capture cross-section of  $^{53}\text{Mn}$  were performed. Nuclear reaction calculations (available in nuclear data libraries as EAF-2007 [8], and TENDL-2008 [9]) use the value of  $66 \pm 7$  b [7] for further predictions.



**Fig. 1:** Comparison of known neutron capture cross-section of  $^{53}\text{Mn}$  with model calculations. In addition the neutron energies available at the BOA, ICON, NEUTRA, and FRANZ facilities are indicated.

We intend to start a series of experiments determining the neutron capture cross section of  $^{53}\text{Mn}$  using thermal-, cold-, and ultra cold neutron beams available at several experimental installations at the PSI [10 – 13] as well as neutrons with a quasi stellar energy spectrum available at the Frankfurt Neutron Source “FRANZ” at the Stern-Gerlach-Center

(SGC), Frankfurt, Germany [14, 15]. In Fig. 1 known neutron capture cross-section of  $^{53}\text{Mn}$  are displayed together with different model predictions [8,9] and the mean neutron energies at the BOA, ICON, NEUTRA (PSI), and FRANZ (SGC) facility.

Shortly after the discovery of  $^{53}\text{Mn}$  few papers were published discussing different approaches to determine the half-life of this isotope. Till the beginning of the 70<sup>th</sup> the obtained values varied from 2 Ma to 11 Ma. Finally, the works of Honda [16], Wölfle [17], and Heimann [18] give a consistent value of 3.8 Ma with an uncertainty of about 10%. In the present literature the value of  $(3.7 \pm 0.37)$  Ma taken from [16] is generally accepted and used. Due to the limited amount of  $^{53}\text{Mn}$  in iron meteorites these experiments were carried out with samples containing  $2.5 \times 10^{11}$  to  $1.3 \times 10^{13}$  atoms of  $^{53}\text{Mn}$  limiting the precision of the measurements. Unfortunately, the above discussed data are contradictory with other observations. Nyquist showed in [19] that the isotopic ratio of  $^{53}\text{Mn}$  to  $^{55}\text{Mn}$  of different meteoritic objects is in disagreement with the deduced age using the  $^{206}\text{Pb} - ^{207}\text{Pb}$  chronometer. The obtained data suggest that the half-life of  $^{53}\text{Mn}$  should be in the range of about 4.8 Ma, i.e. significant higher than the accepted value.

A high sensitivity of cross section and half-life measurements in our experiments can be achieved due to the fact that the available amount of  $^{53}\text{Mn}$  in accelerator waste is in the order of  $10^{18}$  atom per g matrix material [20]. We expect that several samples containing more than  $10^{17}$  atoms can be produced at PSI. The goal of our experiments is to determine the neutron capture cross sections and the half-life of  $^{53}\text{Mn}$  with relative uncertainties of less than 5%.

Our measurement will clarify if the contradictions of Nyquist [19] are caused by a significantly too short value of the half-life of  $^{53}\text{Mn}$  or by other processes. Such processes could be nuclear reactions which produce or consume  $^{53}\text{Mn}$  in meteoritic materials. In the case of a significant higher half-life of  $^{53}\text{Mn}$  similar conclusion as in the case of  $^{60}\text{Fe}$  could be drawn adjusting our view of the early stage of the solar system. Chronometers like  $^{53}\text{Mn} - ^{53}\text{Cr}$  or  $^{26}\text{Al} - ^{26}\text{Mg}$  play an important role for dating events in the solar system. It was shown in a recent work of Bovier and Wadhaw [21] that the solar system is about 2 Ma older than expected so far by dating calcium-aluminium-rich inclusions in meteorites using the  $^{206}\text{Pb} - ^{207}\text{Pb}$  chronometer.

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## REACTIONS INDUCED BY RADIOACTIVE Be BEAMS.

*Alessia Di Pietro*

*INFN-Laboratori Nazionali del Sud, Catania, Italy*

The three Beryllium isotopes have very peculiar structures, namely:  $^9\text{Be}$  is a Borromean weakly bound nucleus ( $S_n=1.67$  MeV), with a well developed  $\alpha$ - $\alpha$ -n cluster structure (see e.g. [1]). With an additional nucleon and due to pairing  $^{10}\text{Be}$  in its ground state is equally deformed but much more bound ( $S_n=6.81$  MeV) than  $^9\text{Be}$  and has an equivalent  $\alpha$ - $\alpha$ -n-n cluster structure. Finally,  $^{11}\text{Be}$  is a one neutron halo nucleus whose core is  $^{10}\text{Be}$  and its binding energy is only  $S_n=503$  keV [2]. Reaction induced by these isotopes are of interest for both nuclear structure and nuclear reaction investigations at low bombarding energies around the Coulomb barrier.

I will present results obtained at Rex-Isolde and LNS Catania concerning different reaction channels for the collisions  $^{9,10,11}\text{Be}+^{64}\text{Zn}$  at energy close to the Coulomb barrier. The analysis of elastic scattering shows a damped

elastic angular distribution for the collision induced by the  $^{11}\text{Be}$  halo nucleus when compared to the ones induced by  $^{9,10}\text{Be}$ . Correspondingly, the total reaction cross-section extracted for  $^{11}\text{Be}+^{64}\text{Zn}$  is more than a factor of two larger than for the other two systems. It will be shown that such an enhancement of the total reaction cross-section with  $^{11}\text{Be}$  is due to the presence of strong transfer/break-up channels. Moreover a discussion on possible experiments that can be performed using a post-accelerated  $^{10}\text{Be}$  beam produced in batch-mode will also be done.

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## SEARCH FOR MUONIC ANALOG OF NEUTRINOLESS DOUBLE BETA DECAY IN TITANIUM

*R. Mohapatra<sup>1</sup>, K. Zuber<sup>2</sup>*

*<sup>1</sup>University of Maryland, <sup>2</sup>TU Dresden*

Muons and electrons behave in remarkably similar ways as far as all their properties go except for the mass. This fact is known as electron-muon universality. Search for tiny deviations from this behaviour could hold a clue as to why their masses are so different and to the broader question of lepton flavor puzzle. Understanding neutrino masses and mixings among different flavors observed during the past decade has certainly added an urgency to this kind of search since the underlying dynamics may throw light on their

origin. More than a decade ago, it was proposed that search for the muonic analog of neutrinoless double beta decay [1] i.e.  $\mu^- + \text{Ti} \rightarrow \mu^+ + \text{Ca}$  may provide important information on this question. In this talk we would present the physics case to encourage to perform such an experiment at PSI.

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## DESTRUCTION OF THE COSMIC GAMMA-RAY EMITTER $^{26}\text{Al}$ BY NEUTRON INDUCED REACTIONS

*P.J. Woods<sup>1</sup>, F. Käppeler<sup>2</sup>, C. Lederer<sup>3</sup>*

<sup>1</sup>*University of Edinburgh, <sup>2</sup>Karlsruhe Institute of Technology, <sup>3</sup>University of Vienna*

The nucleus  $^{26}\text{Al}$  was the first radioactive isotope detected in the interstellar medium through its characteristic gamma-ray line. It showed clear evidence of ongoing nucleosynthesis in the galaxy. Most recent observations by the RHESSI and INTEGRAL satellites suggest a galaxy wide distribution of  $^{26}\text{Al}$  material produced in high mass Wolf-Rayet stars. Recent experiments [1] have indicated key resonances for the destruction of  $^{26}\text{Al}$  via the  $(p,\gamma)$  reaction in Wolf-Rayet stars [1], but the most important destruction mechanisms in this environment is expected to be  $^{26}\text{Al}(n,p)$  and  $^{26}\text{Al}(n,\alpha)$  reaction channels, particular the  $(n,p)$  reaction [2].

The  $^{26}\text{Al}(n,p)$  reaction has been studied previously but as discussed in detail in [2] an inconsistent picture emerges demanding new experimental data. Previous studies have been limited by the material in the production target ( $\sim$  a few  $10^{15}$   $^{26}\text{Al}$  atoms, typically). We propose the production

of a new target by the ERAWAST approach where masses of  $\sim 10^{18}$  atoms  $^{26}\text{Al}$  may be feasible [3]. With such a target the reaction  $^{26}\text{Al}(n,p)$  could be studied using the time of flight technique, for example at the n\_TOF/CERN facility or other time-of-flight facilities providing a highly intense neutron beam. The charged particle reaction products would be detected by introducing a sensitive silicon strip detection system, which could also be used for further studies of  $(n,p)$  and  $(n,\alpha)$  reactions.

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## MEASUREMENT OF THE $^{93}\text{Zr}$ CAPTURE CROSS-SECTION AT n\_TOF FACILITY AT CERN

*G. Tagliente for the n\_TOF collaboration*

*INFN Bari, Italy*

The capture cross section of Zr isotopes is important for aspects related to nuclear technologies. Due to its small neutron capture cross section in combination with the favorable chemical and mechanical properties it is a well suited cladding material for nuclear fuel elements in all reactor types. In addition, the capture cross section for the radioactive isotope  $^{93}\text{Zr}$  ( $t_{1/2} = 1.5 \cdot 10^6$  yr) is important with respect to nuclear waste transmutation. In the High Priority Nuclear Data Request List of the Nuclear Energy Agency (NEA/OECD) a 5% accuracy is requested for the  $(n,\gamma)$  cross section of this long-lived fission product (LLFP) in the entire energy region from thermal to 20 MeV.

For these reasons, the  $(n,\gamma)$  cross sections of all stable isotopes and of  $^{93}\text{Zr}$  have been measured at n\_TOF, yielding accurate results for the stable isotopes [1-4], a prerequisite for the correct analysis of the data obtained with a  $^{93}\text{Zr}$  sample of low enrichment. The preliminary analysis of the  $^{93}\text{Zr}$  capture cross section is particularly interesting as it shows a difference of 35% with respect to the only previous measurement. However, the strict radioprotection rules of CERN required that the  $^{93}\text{Zr}$  sample had to be encapsulated in a double-walled container made of aluminum and titanium. The large background from neutron interactions with the container and a significant  $\gamma$ -ray contamination of the neutron beam limited the

sensitivity of the experiment, so that only the energy region below 7 keV could be investigated in this first measurement.

The n\_TOF facility was recently upgraded to allow measurements of unsealed radioactive samples. Furthermore, thanks to a new moderator and cooling system, the background of the facility has been reduced by more than a factor of 10. With these new features the measurement of this important reaction can now be extended well beyond 100 keV. The results obtained so far and future plans for improving the accuracy of the  $^{93}\text{Zr}(n,\gamma)$  cross-section will be presented.

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## THE STELLAR HALF-LIFE OF $^{44}\text{Ti}$ – SOLUTION OF THE CASSIOPEIA A PROBLEM?

*Iris Dillmann*

*Justus-Liebig-Universität Giessen and GSI Helmholtzzentrum für Schwerionenforschung Darmstadt, Germany*

The influence of the high temperatures in the stellar plasma on lifetimes and decay modes of isotopes is many-fold. Isotopes, which are stable under terrestrial conditions, can become unstable under stellar conditions, and vice versa. Two prominent examples for the different behavior under these different conditions are the bound-state  $\beta$ -decay (acceleration of the decay rate) and the hindrance of orbital-electron capture of highly-ionized nuclei.

Bound-state  $\beta$ -decay rates of  $^{163}\text{Dy}$ ,  $^{187}\text{Re}$ , and  $^{207}\text{Tl}$  were for the first time experimentally determined at the experimental storage ring ESR at the GSI Helmholtzzentrum für Schwerionenforschung in Darmstadt/ Germany [1,2,3]. The investigation of the orbital-EC of highly ionized ions in the ESR was for the first time carried out with H- and He-like  $^{140}\text{Pr}$  and  $^{142}\text{Pm}$  with time-resolved Schottky Mass Spectrometry [4,5,6].

The removal of all electrons from EC-isotopes leads to stable isotopes - if no free electron capture is possible. The most prominent example is  $^7\text{Be}$ , which has a terrestrial half-life of 53 d, which extends to 98 d under the conditions in the core of the Sun. According to theory, with only one electron left (H-like), the half-life should be twice as long as for the neutral case. With 2 electrons (He-like) the half-life should be still 9/8 of the neutral half-life. However, experimental determinations of the EC-half lives of H-like  $^{140}\text{Pr}^{59+}$  and  $^{142}\text{Pm}^{61+}$  showed that the decay is instead accelerated [4,5,6].

One Supernova isotope in this context which gained a lot of attraction in the last years is  $^{44}\text{Ti}$  ( $t_{1/2}=60$  y). Calcula-

tions showed that the late bolometric light curve ( $>1500$  d after maximum luminosity) is dominated by  $^{44}\text{Ti}$  decay [7]. Due to its short half-life no influence from Galactical Chemical Evolution is expected, thus detected  $^{44}\text{Ti}$  is of pure origin from one Supernova. However, gamma-ray astronomy has up to now identified only one clear source of  $^{44}\text{Ti}$  in the Cassiopeia A Supernova remnant, which exploded  $\sim 330$  years ago. BeppoSAX and COMPTEL flux measurement indicate that  $2.5 \times 10^{-4} M_{\odot}$  of  $^{44}\text{Ti}$  was produced, but this mass yield cannot be reproduced by SN models.

One possible reason for this discrepancy could be that up to now for all kind of calculations the terrestrial half-life of  $^{44}\text{Ti}$  was used instead of its stellar half-life. Simulations of the evolution of SN remnants [8] showed a time- and position dependence of highly ionized (up to H-like nuclei) over several 100 years after the SN explosion, which call for the use of stellar half-lives in these SN model simulations.

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## THE SEARCH FOR SUPERHEAVY ELEMENTS IN NATURE

*W. Kutschera, F. Dellinger, O. Forstner, R. Golser, A. Priller, P. Steier, A. Wallner, G. Winkler*

*Vienna Environmental Research Accelerator (VERA), Faculty of Physics – Isotope Research, University of Vienna  
Vienna, Austria*

The detection of traces of heavy nuclides in nature beyond any known species became an intriguing possibility when nuclear theorists in the 1960s predicted a region of increased stability in the sea of nuclear instability, which has since been called the “island of stability” around  $Z = 114$  and  $N = 184$ . In order to find such nuclides in nature - commonly called Superheavy Elements (SHE), several conditions must be fulfilled: (i) they must have been produced under suitable stellar conditions, most likely by the r-process in supernova explosions, (ii) they must be long-lived enough ( $> 100$  million years) to survive in detectable quantities since the solar system formed some 4.6 billion years ago, (iii) they must have physical and chemical properties similar to known elements in order to follow them through their geochemical and geophysical history, (iv) they must have properties which allow one to detect them with methods developed for the ultra-sensitive detection of known nuclides, and (v) the detection methods must allow one to perform an unambiguous identification of the new nuclide.

Although numerous attempts have been performed over the years, there is no firm evidence for the existence of long-lived SHEs in nature except for the claims of the group of Marinov et al. from the Hebrew University of Jerusalem. This group has performed high-resolution mass spectrometry with Inductively Coupled Plasma Sector Field Mass Spectrometry (ICP-SFMS) in thorium and gold. They reported evidence of long-lived neutron-deficient thorium isotopes at the abundance level of  $(1-10) \cdot 10^{-11}$  [1] and of

$10^{-12}$  for a SHE nuclide with  $A = 292$  and  $Z \sim 122$  [2] in thorium. In gold they found evidence for the existence of neutron-deficient roentgenium isotopes at an abundance level of  $(1-10) \cdot 10^{-10}$  [3]. These claims could not be confirmed by experiments with Accelerator Mass Spectrometry (AMS) at the tandem accelerator in Munich [4] and at the Vienna Environmental Research Accelerator in Vienna [5-6], even though the sensitivity was orders of magnitude higher. An extensive AMS search for 30 nuclides around  $A = 300$  in Pt, Au, Pb, and Bi with abundance limits between  $10^{-13}$  to  $10^{-16}$  [7] did also not result in any positive evidence for SHEs. Although it is difficult to evaluate Marinov’s experiments without having participated in them, we are forced to conclude that unknown artifacts may have affected the ICP-SFMS experiments.

A brief outlook of these and other SHE searches in nature will be discussed.

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## DETERMINATION OF THE $^{60}\text{Fe}$ HALF-LIFE – A SUCCESSFUL COLLABORATION IN ERAWAST

*Georg Rugel*

*Technische Universität München, Garching, Germany*

The radionuclide  $^{60}\text{Fe}$  is an important nuclide in nuclear astrophysics. Its half-life has been determined with a sample from a copper beam dump at PSI. After characterization of the beam dump and an intense chemical preparation the final sample material was measured to determine the half-life. This was done with an activity measurement in Munich and a number of  $^{60}\text{Fe}$  atoms measurement at PSI. This results in a half-life of  $2.62 \pm 0.04$  Myr [1].

Some of the important aspects of the work will be reported.

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## HALF-LIFE DETERMINATION BY MEANS OF MULTICOLLECTOR INDUCTIVELY COUPLED PLASMA MASS SPECTROMETRY (MC-ICP-MS)

*Niko Kivel, Ines Günther-Leopold*

*Paul Scherrer Institute, Villigen, Switzerland*

Recently the half-life of several nuclides has been determined by means of ICP-MS and an adequate nuclear counting technique with remarkable low uncertainties. Especially the high precision of MC-ICP-MS has helped to improve the quality of the data. The half-life of  $^{60}\text{Fe}$  could be determined with almost 12 times lower uncertainty than the last reported value. Further new values with higher precision for the half-life of  $^{10}\text{Be}$  and  $^{79}\text{Se}$  have been published recently. Current activities are undertaken to improve the uncertainty of the  $^{166\text{m}}\text{Ho}$  half-life, with the aim to produce highly precise reference emitters for gamma-spectrometry.

The trend to facilitate ICP-MS either as additional technique or as the only means of the determination of the number of atoms is based on the improvements in detection power and the availability of higher amounts of the nuclides of interest. So was the determination of the  $^{60}\text{Fe}$  half-life only possible because a rather large amount of  $6 \cdot 10^{15}$  atoms was available for the project.

The presentation will focus on the general capabilities of MC-ICP-MS in half-life applications and summaries the benefits as well as the drawbacks of the technique.

### THE HALF-LIFE OF $^{60}\text{Fe}$ REVISITED

*W. Kutschera<sup>1</sup>, K. Buzcak<sup>1</sup>, O. Forstner<sup>1</sup>, R. Golser<sup>1</sup>, A. Priller<sup>1</sup>, P. Steier<sup>1</sup>, A. Wallner<sup>1</sup>, D. Schumann<sup>2</sup>, R. Dressler<sup>2</sup>, G. Wallner<sup>3</sup>, M. Bichler<sup>4</sup>, G. Steinhauser<sup>4</sup>, P. Collon<sup>5</sup>, M. Bowers<sup>5</sup>, K. Chamberlin<sup>5</sup>, M. Couder<sup>5</sup>, W. Lu<sup>5</sup>, D. Robertson<sup>5</sup>, M. Troy<sup>5</sup>, A. Stoltz<sup>6</sup>, S. Austin<sup>6</sup>, I. Ahmad<sup>7</sup>, J. Green<sup>7</sup>, D. Graczyk<sup>8</sup>, M. Paul<sup>9</sup>*

*<sup>1</sup>Vienna Environmental Research Accelerator (VERA), Faculty of Physics – Isotope Research, University of Vienna, Vienna, Austria, <sup>2</sup>Laboratory for Radio- and Environmental Chemistry, PSI, Villigen, Switzerland, <sup>3</sup>Institute for Inorganic Chemistry, University of Vienna, Vienna, Austria, <sup>4</sup>Atominstut, Technical University of Vienna, Vienna, Austria, <sup>5</sup>Physics Department, University of Notre Dame, Notre Dame, Indiana, USA, <sup>6</sup>National Superconducting Cyclotron Laboratory, Michigan State University, East Lansing, Michigan, USA, <sup>7</sup>Physics Division, Argonne National Laboratory, Argonne, Illinois, USA, <sup>8</sup>Chemistry Division, Argonne National Laboratory, Argonne, Illinois, USA, <sup>9</sup>Racah Institute of Physics, Hebrew University of Jerusalem, Jerusalem, Israel*

The first attempt to measure the half-life of  $^{60}\text{Fe}$  started 30 years ago with an unsuccessful proposal to use  $^{60}\text{Fe}$  from the LAMPF beam stop. Therefore,  $^{60}\text{Fe}$  was produced by spallation of copper with 191-MeV protons at the Brookhaven Linear Isotope Producer (BLIP). After extraction and purification of the  $^{60}\text{Fe}$  material, a half-life of  $(1.49 \pm 0.27) \cdot 10^6$  yr was determined from an AMS measurement of the isotopic abundance with the ATLAS accelerator at Argonne National Lab and a gamma-ray activity measurement with a Ge detector [1]. 25 years later a much larger amount of  $^{60}\text{Fe}$  was extracted from a copper beam stop at PSI having been bombarded for many years with 590-MeV protons [2]. The combination of an ICP-MS measurement for the isotopic ratio of the  $^{60}\text{Fe}$  material at

PSI with a  $\gamma$ -ray activity measurement at the TU München resulted in a significantly longer half-life value of  $(2.62 \pm 0.04) \cdot 10^6$  years [3].

Since an accurate  $^{60}\text{Fe}$  is of considerable interest for a number of astrophysical questions, the current discrepancy clearly asks for additional measurements. These are indeed on the way at different places, and preliminary results will be reported as they are available at the time of the meeting.

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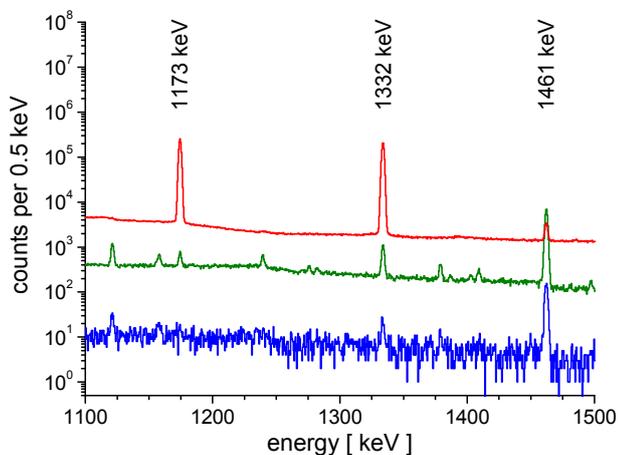
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## REPETITION OF THE RE-MEASUREMENT OF THE $^{60}\text{Fe}$ HALF LIFE

*M. Ayranov, M. Bunka, R. Dressler, I. Günther-Leopold, St. Heinitz, N. Kivel, S. Lüthi, D. Schumann, T. Stowasser, A. Vögele, D. Wittwer*

*Paul Scherrer Institute, Villigen, Switzerland*

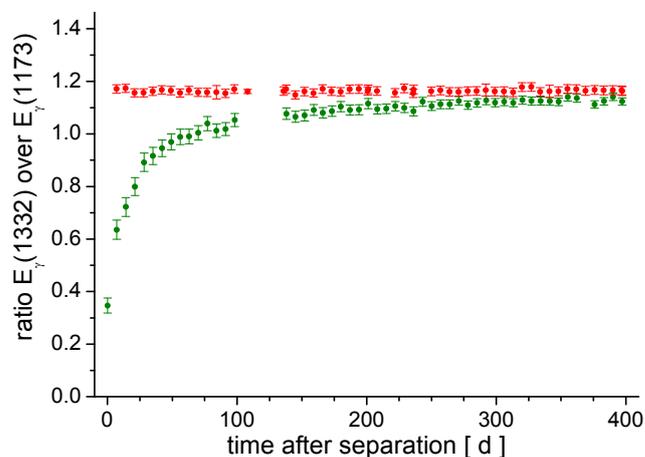
The recently performed re-determination of the  $^{60}\text{Fe}$  half-life of  $(2.62 \pm 0.04)$  Ma [1] showed a deviation from the so far accepted value  $t_{1/2} = (1.49 \pm 0.27)$  Ma [2] of more than 1 Ma. This discrepancy demands a repetition of the measurement. Therefore, a new  $^{60}\text{Fe}$  sample was prepared recycling the target used for the determination of the neutron capture cross section [3]. The  $^{60}\text{Fe}$ , fixed on a carbon backing [4], was dissolved in 7 M HCl and  $^{60}\text{Co}$  was separated by extracting the iron into methyl isobutyl ketone and back extraction into 0.1 M HCl. The procedure was repeated several times. The solution was divided into four parts. Two samples are currently used for the  $\gamma$ -measurement of the half-life determination, one at the University of Vienna, the other one at PSI. The third sample is used for the determination of the number of atoms per gram liquid and the last part is used as target material for the measurement of the thermal neutron capture cross section. About 80% of the original target material could be recovered during the chemical procedures, i.e. a total number of  $3.5 \times 10^{15}$  atoms  $^{60}\text{Fe}$  is expected in each of the samples used for the  $\gamma$ -measurements and  $1.7 \times 10^{15}$  atoms  $^{60}\text{Fe}$  in the sample used for the MC-ICP-MS and for the thermal neutron capture cross section measurements, respectively.



**Fig. 1:**  $\gamma$ -spectra in the region of the two main  $\gamma$ -lines of  $^{60}\text{Co}$ . From top to bottom: 48 h measurement of  $^{60}\text{Co}$ - $^{241}\text{Am}$  reference sample (red, digital stabilizer on), first 160 h measurement of  $^{60}\text{Fe}$  sample (green, digital stabilizer hold), and snap shot of this measurement after 4 h. The  $\gamma$ -lines of  $^{60}\text{Co}$  and  $^{40}\text{K}$  are indicated.

The  $^{60}\text{Fe}$  solution used for  $\gamma$ -spectroscopic investigations at PSI was transferred into a serum vial (11 mm outer diameter) and filled up to 4 ml using diluted acid (0.1 M HCl). A HPGe well detector with blind hole well (crystal length 53 mm, diameter 60 mm; core length 40 mm, diameter 19 mm; active volume  $\sim 132 \text{ cm}^3$ ) is used for the measurements. The serum vial fits tight into the well opening. A reference sample containing  $(157.6 \pm 3.4)$  Bq  $^{60}\text{Co}$  and

$(7.3 \pm 1.6)$  Bq  $^{241}\text{Am}$  (with  $k=2$  expanded standard uncertainties, reference date 2.10.2009) is measured for 4 h alternating to the 160 h measurements of the  $^{60}\text{Fe}$  sample serving to fix the gain and offset of the spectroscopic hardware using a digital stabilizer. The activities of  $^{60}\text{Co}$  and  $^{241}\text{Am}$  are determined using a total efficiency calibrated detector at PSI. Therefore, the uncertainties of these values are rather large. The first measurement of the  $^{60}\text{Fe}$  sample started in August 2009. Fig. 1 shows a measurement using the  $^{60}\text{Co}$ - $^{241}\text{Am}$  reference source, the completed first measurement of the  $^{60}\text{Fe}$  (started 4 h after finishing the last purification) and a snap shot 4 h after start of this measurement. It can be clearly seen that the activity of  $^{60}\text{Co}$  was very low after the last purification. The initial  $^{60}\text{Fe}$  and  $^{60}\text{Co}$  activity was deduced to be 35 Bq and 35 mBq, respectively, from a fit of the time dependence of the  $\gamma$ -lines of  $^{60}\text{Co}$ . The uncertainties of these values are about 10%, depending on the uncertainty of the  $^{60}\text{Co}$  activity in the reference source and the reached quality of the peak area determination and the resulting fit quality. The additional population of the 1332 keV line via a 0.24% branch of  $^{60m}\text{Co}$  leads to an enhanced count rate of this  $\gamma$ -line compared to the  $^{60}\text{Co}$  reference measurement. This is depicted in Fig. 2 via the count-rate ratios of the 1173 keV and the 1332 keV  $^{60}\text{Co}$   $\gamma$ -line.



**Fig. 2:** Count-rate ratios of the 1173 keV and the 1332 keV  $^{60}\text{Co}$   $\gamma$ -line of the  $^{60}\text{Co}$ - $^{241}\text{Am}$  reference source (red) and the  $^{60}\text{Fe}$  sample in dependence of the time after the last chemical separation.

This behavior can be used to determine the total  $^{60}\text{Fe}$  activity in much shorter counting periods.

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## MEASUREMENT OF THE ${}^7\text{Be}+p$ CROSS SECTION USING THE RECOIL MASS SEPARATOR ERNA

*L. Gialanella*

*Second University of Naples and INFN-Naples for the ERNA collaboration*

The cross section of the radiative capture reaction  ${}^7\text{Be}+p$  plays an important role in our understanding of the solar neutrino spectrum observed on Earth. Several experiments were performed using a proton beam impinging on a  ${}^7\text{Be}$  radioactive target, whereas discrepancies between the results of different experiments possibly limit the overall accuracy. The pioneering work of the NaBoNA (Naples Bochum Nuclear Astrophysics) collaboration during the 90's first exploited an alternative approach based on a recoil mass separator in combination with an hydrogen gas target and a  ${}^7\text{Be}$  ion beam at the TTT3 Tandem of the University of Naples.

Recently this effort has been repeated at the Oak Ridge National Laboratory using the Daresbury recoil mass separator. Unfortunately, these experiments could not achieve the necessary precision to significantly contribute to an accurate determination of the stellar rate of  ${}^7\text{Be} + p$ , mostly due to the weak beam intensity.

In 2010 a new experiment has started, based on the recoil mass separator ERNA (European Recoil separator for Nuclear Astrophysics) installed at the CIRCE laboratory in Caserta, Italy, where an intense  ${}^7\text{Be}$  ion beam is routinely produced. The experiment is presented and the expected improvements compared to previous work are discussed.

## MEASUREMENT OF THE NEUTRON CAPTURE CROSS SECTION OF THE S-PROCESS BRANCHING $^{63}\text{Ni}$ AT N\_TOF/CERN

C. Lederer<sup>1</sup>, C. Massimi<sup>2</sup>, M. Barbagallo<sup>3</sup>, F. Belloni<sup>4</sup>, E. Berthoumieux<sup>5</sup>, M. Calviani<sup>5</sup>, D. Cano-Ott<sup>6</sup>, N. Colonna<sup>3</sup>, I. Dillmann<sup>7</sup>, K. Fraval<sup>4</sup>, C. Guerrero<sup>5</sup>, F. Gunsing<sup>4</sup>, F. Käppeler<sup>8</sup>, A. Mengoni<sup>9</sup>, R. Reifarth<sup>10</sup>, D. Schumann<sup>11</sup>, G. Tagliente<sup>3</sup>, J.L. Tain<sup>12</sup>, A. Wallner<sup>13</sup>, V. Vlachoudis<sup>5</sup>, and the n\_TOF Collaboration

<sup>1</sup>University of Vienna, Austria, <sup>2</sup>INFN Bologna and University of Bologna, Italy, <sup>3</sup>INFN Bari, Italy, <sup>4</sup>CEA Saclay, France, <sup>5</sup>CERN, Switzerland, <sup>6</sup>CIEMAT, Spain, <sup>7</sup>University of Giessen and GSI, Germany, <sup>8</sup>Karlsruhe Institute of Technology, Germany, <sup>9</sup>Embassy of Italy, Japan, <sup>10</sup>Goethe University of Frankfurt, German, <sup>11</sup>PSI, Switzerland, <sup>12</sup>IFIC, Spain, <sup>13</sup>University of Vienna, Austria and ANSTO, Australia

About half of the overall abundances of elements heavier than Fe are produced by the slow neutron capture process (s-process). This process occurs during He burning stages of stellar evolution and is characterized by small reaction rates, thus the reaction flow takes place close to the valley of stability - if a radioactive nuclide is produced, its decay happens faster than a subsequent neutron capture.

An exception are long-lived radionuclides, where neutron capture competes with radioactive decay and the reaction path at these so-called branching points is very sensitive to the neutron capture cross section. Despite the importance of neutron capture cross sections at branching points for investigating physical conditions of s-process sites and calculating s-abundances, few experimental data are available since samples are hard to obtain and measurements are often complicated by additional background due to sample activity.

$^{63}\text{Ni}$ , with a laboratory half-life of 100.1 years, represents such an example. It is produced in the weak s-process taking place in massive stars during two different burning stages: during He Core burning (at temperatures of  $kT=26$  keV), where neutron densities are too small to bypass the decay channel and  $^{63}\text{Ni}$  entirely decays to  $^{63}\text{Cu}$ , and later during C shell burning ( $kT=91$  keV) where subsequent neutron capture on  $^{63}\text{Ni}$  becomes the dominant reaction. Network calculations showed that e.g. the final  $^{63/65}\text{Cu}$  abundances strongly depend on the  $^{63}\text{Ni}(n,\gamma)$  cross section [1]. However, no experimental data are available above thermal neutron energies and all estimates for stellar environments are based on either calculations or extrapolations of the thermal cross section. Therefore, the n\_TOF collaboration proposed to measure the neutron capture cross section of  $^{63}\text{Ni}$  at the n\_TOF facility from thermal up to energies of hundreds of keV [2].

The neutron time-of-flight facility n\_TOF at CERN is dedicated to neutron cross section measurements of interest for astrophysics and nuclear technologies [3]. Neutron capture cross sections are determined via measuring the neutron flight time and detecting the prompt  $\gamma$  rays following a capture event. n\_TOF provides a highly intense neutron beam produced by spallation reactions of pulsed 20 GeV/c PS protons on a massive Pb target and a high energy resolution due to its long flight path of 185 m. Two detector systems are available, a  $4\pi$  BaF<sub>2</sub> array and a pair of C<sub>6</sub>D<sub>6</sub>-liquid scintillation detectors, optimized for low neutron sensitivity. The measurement, scheduled for spring 2011 will be performed using the C<sub>6</sub>D<sub>6</sub> detector setup, since we expect a sizable neutron scattering background. The background will be measured using an empty sample holder and dummy samples, revealing the energy dependence of possible sample-dependent backgrounds. The sample activity itself will not add an additional component, since  $^{63}\text{Ni}$  is a pure  $\beta$ -emitter, which directly decays into the ground state of  $^{63}\text{Cu}$ .

The  $^{63}\text{Ni}$  sample available was produced via breeding an enriched  $^{62}\text{Ni}$  sample, about 1 g in mass, in a thermal reactor, yielding a final enrichment of approximately 12%. Since already a significant amount of the decay product  $^{63}\text{Cu}$  was present in the sample, this contaminant was removed chemically at PSI.

The measurement, data analysis and first results will be presented.

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## ATHENA – A NETWORKING ACTIVITY WITHIN ENSAR

*K. Sonnabend*

*Goethe-University Frankfurt, Germany*

### OBJECTIVES OF ATHENA

ATHENA [1] is short for Advanced Theory and Experiments in Nuclear Astrophysics and is a networking activity within ENSAR [2] (European Nuclear Science and Applications Research) that is funded within the Seventh Framework Programme (FP7) of the European Commission under the specific programme “Capacities” [3]. Partners from so far 40 institutions in 15 European countries are contributing to the aim of stimulating, collecting, and streamlining the European research in theoretical and experimental nuclear astrophysics.

The tasks of nuclear astrophysics research are not restricted to, e.g., a single energy range or a special experimental method. In contrast, the diversity of stellar scenarios forces nuclear astrophysicists to work at the frontiers of different experimental techniques related to radioactive ion beams as well as high-intensity stable beams, to recoil mass separators as well as particle and photon detection, to activation as well as ultra-low background experiments. Concerning theoretical aspects, the latest techniques and developments have to be used to handle, on the one hand, the computation of stellar evolution and nucleosynthesis and to calculate, on the other hand, the most reliable nuclear physics input needed for these simulations.

The main goals of ATHENA are to consolidate the efforts on a steady improvement of the relevant nuclear physics input by the combined expertise of experimentalists and theoreticians, to enhance the synergetic effects and to allocate an environment of mutual stimulation. As a networking activity ATHENA focuses on the organisation of workshops and meetings to reach these goals.

Three main themes in the field, each representing a key problem in nuclear astrophysics research and being of equal priority, are the topics of ATHENA. Task 1 is dedicated to „Explosive nuclear astrophysics“ and is closely related to experimental physics with radioactive ion beams. Task 2 deals with „Reactions at low temperatures and on broad mass ranges“. The experimental research is in that case mostly performed at so-called small-scale accelerating facilities. The „Nucleosynthesis in neutron capture processes“ is topic of Task 3 and is mainly supported by the n\_TOF collaboration.

### EXPLOSIVE NUCLEAR ASTROPHYSICS

Explosive stellar environments such as novae, supernovae and X-ray bursts are currently among the most exciting topics in nuclear astrophysics. Reactions on unstable nuclei play a crucial role in energy generation and nucleosynthesis due to the high temperatures and short reaction time scale in these events. Knowledge of these reactions is essential to interpret remarkable new observations from ground and space based telescopes of elemental abundances, light curves, and cosmic  $\gamma$ -ray emitters. Successful

measurements of these reactions require intense, low energy, high-purity radioactive beams and the development of suitable detector and spectrometer systems. This programme has to be closely allied with theoretical models of the explosive processes for maximum scientific impact.

The aim of this task is to establish working groups discussing about the following points:

- *Key reactions:* Discussion to identify the reactions with major impact in the above mentioned scenarios including specification of energy range and precision required for future experimental studies.
- *Key technical developments:* Exchange of technical ideas to produce the highest priority radioactive beams to measure key reactions and coordination of how and where developments should be best achieved.
- *Key characteristics of novel detection systems:* Exchange of ideas to design and construct novel detector and spectrometer systems to measure key reactions and coordination of single activities.
- *Key features of nuclear theory:* Discussion of experimental and/or theoretical verification of nuclear properties, nuclear reactions, and the nuclear equation of state for astrophysical simulations.

Further information including the contact persons of this task can be found at Ref. [4].

### REACTIONS AT LOW TEMPERATURES AND ON BROAD MASS RANGES

Most of their life many stars are in phases of quiescent stellar burning. The reactions providing the power to stabilize the stars and additionally producing new elemental species out of the basic components hydrogen and helium take place at low temperatures so that the cross sections have to be known down to the so-called Gamow peak. In many cases, this energy region is still not accessible experimentally, thus, the results provided to theoretical models rely on extrapolations from the existing data.

In contrast, the access to the data needed to predict the abundances of the heavy nuclei above the iron peak is not hampered by the energy region to be observed but by the vast number of information being required for the modelling. In that case, theory has to provide extrapolations of cross sections and reaction rates based on the existing data to cover the huge number of isotopes and reactions being involved.

Therefore, this task deals with the following problems:

- *Increase of the synergy of the existing laboratories and equipments:*

Coordination of the capacities provided by small-scale accelerator facilities and review of the existing variety of different beams and detection systems as well as of the diversity of supported experimental methods.

– *Major upgrades for long-term experimental programmes:*

Discussion about the possibility of major upgrades at existing laboratories to establish a long-term experimental programme concerning the measurements of key reactions in various non-explosive scenarios of quiescent stellar burning and of crucial reaction rates of heavy-element nucleosynthesis.

– *Reaction rates of heavy-element nucleosynthesis:*

Exchange of ideas between theoreticians and experimentalists to improve the reliability of predicted reaction rates in astrophysical scenarios of heavy-element nucleosynthesis.

Further information including the contact persons of this task can be found at Ref. [4].

## NUCLEOSYNTHESIS IN NEUTRON CAPTURE PROCESSES

Concerning nucleosynthesis in the slow neutron capture process the observed abundances are directly correlated to the neutron capture cross section,  $\sigma(n,\gamma)$ , so that their measurement along the valley of stability is an ongoing process since the early days of nuclear astrophysics. The astrophysical site of the production of the main component ( $90 < A < 209$ ) are the helium layers of asymptotic giant branch stars while the weak component ( $A < 90$ ) is thought to be synthesized in massive stars.

In any case, the capture cross sections have to be known with accuracies at the level of a few percent to find a quantitative description of s-process nucleosynthesis. The n\_TOF facility at CERN can contribute these data besides the study of capture cross sections for nuclear waste transmutation, advanced nuclear technologies and basic nuclear physics.

– *Support during experimental campaigns at CERN:*

Coordination of teams formed by young researchers to measure the neutron capture cross sections of the stable Fe and Ni isotopes at the n\_TOF facility, CERN.

– *Support of data analysis and transfer to astrophysically relevant data:*

Coordination of teams to analyse the data and derive the Maxwellian averaged cross sections after the experimental campaigns at CERN.

– *Preparation of future neutron sources:*

Establish teams to prepare experiments at future neutron sources available across Europe.

Further information including the contact persons of this task can be found at Ref. [4].

## PROGRESS AND OUTLOOK

Since the start of ENSAR in September 2010, ATHENA has supported a workshop on “p-process: present status and outlook” [5] held in Istanbul, Turkey, in May 2011 by reimbursing the travel costs of some of the invited speakers. In addition, the participation of some young students at the Helmholtz International Summer School “Nuclear Theory and Astrophysical Applications” [6] held in Dubna, Russia, in July and August 2011 was enabled in reimbursing their travel costs.

To advertise the existence and the aims of ATHENA the networking activity was presented at several workshops and meetings of other European activities such as the First EuroGENESIS Workshop (November 2010, Dubrovnik, Croatia) and the ERINDA Kick-Off-Meeting (January 2011, Dresden, Germany).

For the upcoming three years, it is planned to organize a workshop on s-process nucleosynthesis by task 2 and 3 and on neutron-induced reactions by task 3 in collaboration with DANCE@LANSCE (Los Alamos National Laboratory, USA) [7] besides the support of otherwise organized workshops, schools, and meetings. In addition, ATHENA joined an initiative of the currently running European scale projects that include astrophysics as a topic, such as e.g. EuroGENESIS [8], CompSTAR [9], and ERINDA [10], to organize a common event to exchange the knowledge between the experts of the different communities and develop interdisciplinary links between them.

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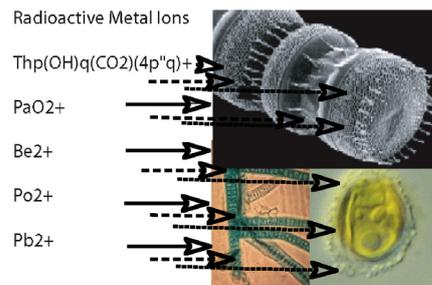
## BIOPOLYMERS AS CARRIERS OF NATURAL (Th, Pa, Pb, Po, Be) RADIONUCLIDES IN AQUATIC SYSTEMS

Peter H. Santschi, Chia-Ying Chuang, and Kathleen A. Schwehr

Texas A&M University at Galveston, USA

Th(IV), Pa(IV,V), Po(IV, II,-II), Pb(II) and Be(II) radioisotopes are important proxies in oceanographic investigations, e.g., for tracing particle dynamics and particulate organic matter (POC) fluxes out of the euphotic zone, and for studying boundary scavenging, paleo-productivity and ocean circulation. Even though considered routine, these approaches rely on often poorly constrained, empirically determined and variable isotope ratios or ratios to POC. Previously published laboratory and field investigations suggest that a number of biopolymers, potentially produced by both phytoplankton and bacteria, are carrier molecules for most of these isotopes, rather than purely inorganic surfaces[e.g., 1-6]. We hypothesize that specific binding and redox processes control marine scavenging and the most efficient binding would occur to acid polysaccharide- and protein-containing biomolecules. Ongoing experiments attempt to separate, identify and characterize radioisotope carriers that are hypothesized to be effective binding ligands. Our interdisciplinary research project requires instrumental approaches for characterization studies, in combination with controlled laboratory and field experimentation. Laboratory studies consist of comparative uptake experiments of a suite of naturally occurring radionuclides to a number of substrates, including model organic and inorganic compounds, marine colloidal and particulate organic matter, and biopolymers harvested from cultures. The field program includes collections and characterization of diverse types of suspended and sinking organic matter from different parts of the ocean. 10 s of mg of colloidal and particulate organic matter samples were collected in 2009 in the Pacific Ocean using ships from our collaborating institutions in Taiwan. Preliminary experiments carried out so far indicate that when organic matter is present in diatom frustules grown in the lab, and cleaned to different degrees to selectively remove organic matter, the presence of diatom-associated

organic matter greatly enhances particle-water partitioning of all radionuclides, including  $^7\text{Be}$  (tracer obtained from PSI).



**Fig. 1:** Schematic of interactions between (radioactive) metal ions and marine particle assemblages.

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## A PROPOSAL FOR RE-DETERMINATION OF $^{146}\text{Sm}$ HALF-LIFE

M. Paul

Racah Institute of Physics, Hebrew University, Jerusalem, Israel

The radioactive half-life of the p-process nuclide  $^{146}\text{Sm}$  ( $t_{1/2}=103\pm 5$  Myr) plays an important role in the chronology of the Early-Solar System formation and of geochemical fractionation processes in planetary bodies. We performed a new measurement of this half-life by determining both alpha-activity ratio and atom ratio of  $^{146}\text{Sm}$  to naturally-occurring  $^{147}\text{Sm}$  ( $t_{1/2}=107$  Gyr) in activated  $^{147}\text{Sm}$  material.  $^{146}\text{Sm}$  was produced via the  $^{147}\text{Sm}(\gamma, n)$ ,  $^{147}\text{Sm}(n, 2n)$  and

$^{147}\text{Sm}(p, 2n\text{EC})$  reactions and the  $^{146}\text{Sm}/^{147}\text{Sm}$  atom ratio was measured by accelerator mass spectrometry. The value obtained for the  $^{146}\text{Sm}$  half-life  $68\pm 7$  Myr is significantly shorter than the presently adopted value. We propose to prepare a Sm sample highly enriched in  $^{146}\text{Sm}$  using the upcoming ERAWAST capabilities and to confirm the value of  $^{146}\text{Sm}$  half-life using an independent experimental method.

## MEASUREMENT OF THE CROSS SECTION OF THE ${}^7\text{Be}(n,\alpha)\alpha$ REACTION AND THE PROBLEM OF PRIMORDIAL ${}^7\text{Li}$

M. Gai<sup>1</sup>, L. Weissman<sup>2</sup>

<sup>1</sup>University of Connecticut and Yale University, USA, <sup>2</sup>Soreq National Research Centre, Israel

The disagreement of the predicted abundance of primordial  ${}^7\text{Li}$  with the observed abundance is a longstanding problem in Big Bang Nucleosynthesis (BBN) theory [1]. While BBN theory correctly predicts the abundances of  ${}^1\text{H}$ ,  ${}^2\text{H}$ ,  ${}^3\text{He}$  and  ${}^4\text{He}$  (that vary over five orders of magnitudes), it over predicts the abundance of primordial  ${}^7\text{Li}$  by a factor of approximately 2.5-4.5 (approximately 4-5  $\sigma$  discrepancy). Primordial  ${}^7\text{Li}$  is copiously produced directly (e.g. via the  ${}^6\text{Li}(n,\gamma)$  reaction etc.) but later during the first 4-15 minutes approximately 99% of the so produced  ${}^7\text{Li}$  is destroyed primarily via the  ${}^7\text{Li}(p,\alpha)$  reaction. Hence most of the predicted primordial  ${}^7\text{Li}$  is predicted to be produced via the (electron capture beta) decay of the primordial  ${}^7\text{Be}$  that is produced primarily in the  ${}^3\text{He}(\alpha,\gamma)$  reaction. We propose to investigate the destruction of  ${}^7\text{Be}$  during (the first 10-15 minutes of) BBN via the  ${}^7\text{Be}(n,\alpha)$  reaction. If during that time the majority of the primordial  ${}^7\text{Be}$  is destroyed (before decaying to  ${}^7\text{Li}$ ) it will lead to a reduction of approximately 3 of the predicted abundance of the primordial  ${}^7\text{Li}$ , hence a resolution of the long standing disagreement. The rate of the  ${}^7\text{Be}(n,\alpha)$  reaction [2,3] relies on cross section of thermal neutron (only) measured in the 60's and tabulated for the

first and last time by Wagoneer et al. in the 60's [4]. We propose to measure the cross section of the  ${}^7\text{Be}(n,\alpha)$  reaction with neutron beams that mimic a quasi Maxwellian flux at 50 keV and a  ${}^7\text{Be}$  produced at PSI [5]. A prototype experiment and the proposed final experiment could be performed at Phase I of SARAF [6] using the LILIT target [7].

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## THE ${}^{40}\text{Ca}(\alpha,\gamma){}^{44}\text{Ti}$ REACTION STUDIED BY ACTIVATION

Konrad Schmidt<sup>1,2</sup>, Chavkat Akhmadaliev<sup>1</sup>, Michael Anders<sup>1</sup>, Daniel Bemmerer<sup>1</sup>, Konstanze Boretzky<sup>3</sup>, Antonio Cacioli<sup>4</sup>, Zoltán Elekes<sup>1</sup>, Zsolt Fülöp<sup>5</sup>, György Gyürky<sup>5</sup>, Roland Hannaske<sup>1</sup>, Arnd Junghans<sup>1</sup>, Michele Marta<sup>1,3</sup>, Ronald Schwengner<sup>1</sup>, Tamás Szücs<sup>5</sup>, Andreas Wagner<sup>1</sup>, and Kai Zuber<sup>2</sup>

<sup>1</sup>Helmholtz-Zentrum Dresden-Rossendorf (HZDR), <sup>2</sup>TU Dresden, <sup>3</sup>GSI Darmstadt, <sup>4</sup>INFN Padua, Italy, <sup>5</sup>ATOMKI Debrecen, Hungary

The radioactive nuclide  ${}^{44}\text{Ti}$  is believed to be produced in the  $\alpha$ -rich freezeout preceding supernova explosions. The  $\gamma$ -lines from its decay have been observed in space-based  $\gamma$ -observatories for the Cassiopeia A supernova remnant. The rates of the nuclear reactions governing the production and destruction of  ${}^{44}\text{Ti}$  should therefore be known with precision. Using the  $\alpha$ -beam of the 3.3 MV Tandatron of

Helmholtz-Zentrum Dresden-Rossendorf, the strengths of the  ${}^{40}\text{Ca}(\alpha,\gamma){}^{44}\text{Ti}$  resonance triplet at 4.5 MeV  $\alpha$ -energy have been re-studied by activation. The samples have been analyzed in the Felsenkeller underground  $\gamma$ -counting facility. Preliminary data on lower-lying resonances will be presented, as well.

## STELLAR PRODUCTION AND DESTRUCTION RATES OF $^{60}\text{Fe}$

Tanja Heftrich<sup>1</sup>, Ethan Uberseder<sup>2</sup>, Tatsuya Adachi<sup>3</sup>, Thomas Aumann<sup>4</sup>, Saul Beceiro<sup>5</sup>, Konstanze Boretzky<sup>6</sup>, Christoph Caesar<sup>4</sup>, Santosh Chakraborty<sup>7</sup>, Olga Ershova<sup>1</sup>, Alfredo Estrade<sup>6</sup>, Michael Heil<sup>6</sup>, Marcel Heine<sup>4</sup>, Matthias Holl<sup>4</sup>, Alexander Ignatov<sup>6</sup>, Hakan Johansson<sup>8</sup>, Aleksandra Kelic<sup>6</sup>, Christoph Langer<sup>1</sup>, Tudi LeBleis<sup>9</sup>, Yuri Litvinov<sup>6</sup>, Justyna Marganiec<sup>10</sup>, Alina Movsesyan<sup>4</sup>, Joerg Neuhausen<sup>11</sup>, Ali Najafi<sup>3</sup>, Valerii Panin<sup>4</sup>, Ralf Plag<sup>1</sup>, Anisur Rahaman<sup>7</sup>, Ganna Rastrepina<sup>1</sup>, Rene Reifarh<sup>1</sup>, Valentina Ricciardi<sup>6</sup>, Catherine Rigollet<sup>3</sup>, Dominic Rossi<sup>12</sup>, Dorothea Schumann<sup>11</sup>, Deniz Savran<sup>10</sup>, Haik Simon<sup>6</sup>, Kerstin Sonnabend<sup>4</sup>, Branislav Streicher<sup>3</sup>, Vasily Volkov<sup>6</sup>, Felix Wamers<sup>4</sup>, Helmut Weick<sup>6</sup>, Mario Weigand<sup>1</sup>, Michael Wiescher<sup>2</sup>, Christine Wimmer<sup>1</sup>, Nikolas Winckler<sup>6</sup>, and Philip J. Woods<sup>13</sup>

<sup>1</sup>University of Frankfurt, Germany - <sup>2</sup>University of Notre Dame, USA - <sup>3</sup>KVI Groningen, Netherlands - <sup>4</sup>TU Darmstadt, Germany - <sup>5</sup>University of Santiago de Compostela, Spain - <sup>6</sup>GSI Darmstadt, Germany - <sup>7</sup>SINP Kolkata, India - <sup>8</sup>Chalmers University of Technology, Sweden - <sup>9</sup>TU Munich, Germany - <sup>10</sup>ExtreMe Matter Institute EMMI, GSI - <sup>11</sup>Paul Scherrer Institute, Switzerland - <sup>12</sup>Johannes Gutenberg University of Mainz, Germany - <sup>13</sup>University of Edinburgh, United Kingdom

### ASTROPHYSICAL MOTIVATION

One of the fundamental signatures for active nucleosynthesis in our Universe is the observation of long-lived radioactive elements in our galaxy. Using high-resolution  $\gamma$ -ray observatories, such as COMPTEL, RHESSI, and INTEGRAL, the reaction rates of long-lived radioactive isotopes, for instance  $^{18}\text{F}$ ,  $^{22}\text{Na}$ ,  $^{26}\text{Al}$ ,  $^{44}\text{Ti}$  and  $^{60}\text{Fe}$ , can be determined [1]. A detailed analysis of the two long-lived radioactive isotopes  $^{26}\text{Al}$  and  $^{60}\text{Fe}$  will give important information about stellar nucleosynthesis. The production of  $^{26}\text{Al}$  and  $^{60}\text{Fe}$  is thought to be associated with the nucleosynthesis in hot carbon or oxygen shell burning in massive pre supernova stars and in the subsequent shock-front driven explosive nucleosynthesis of type II supernovae [2,3]. While the reactions associated with  $^{26}\text{Al}$  have been studied extensively in the past, very little is known about the reactions associated with the production of  $^{60}\text{Fe}$ . RHESSI and INTEGRAL [4,5] observed the characteristic smooth  $^{60}\text{Fe}$  distribution of  $\gamma$ -radioactivity along the galactic plane [6]. Model predictions show that the  $^{60}\text{Fe}$  is produced in the supernova shock front as well as in a hot s-process environment during carbon shell burning in the massive supernova progenitor star. Based on simulations of hot carbon shell burning the radioactive isotope  $^{60}\text{Fe}$  with a half-life of  $t_{1/2}=2.6$  My is associated with the neutron capture process of stable iron isotopes for instance  $^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}(n,\gamma)^{60}\text{Fe}$ . Because of its short half-life ( $t_{1/2}=44.5$  d), neutroncapture on  $^{59}\text{Fe}$  is very difficult to measure directly. Therefore, the coulomb dissociation cross section of the inverse reaction  $^{60}\text{Fe}(\gamma,n)^{59}\text{Fe}$  provides important information via the principle via detailed balance. In order to prove this method,  $^{59}\text{Fe}(\gamma,n)^{58}\text{Fe}$  was studied additionally to determine the already directly measured  $^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$  cross section [7].

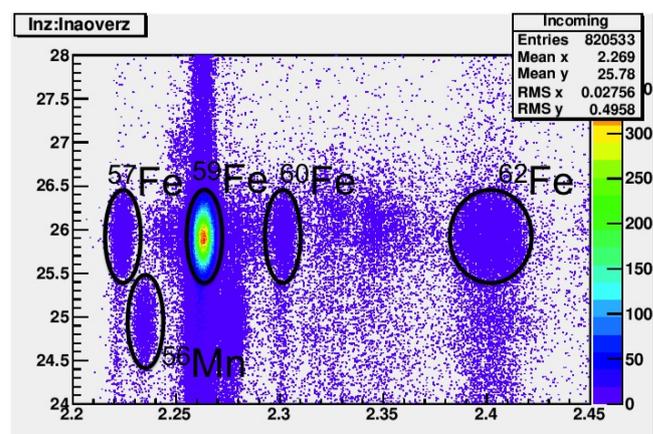
### MEASUREMENTS

Coulomb dissociation measurements of  $^{59}\text{Fe}$  and  $^{60}\text{Fe}$  were performed at the R3B/LAND Setup at GSI. The unstable iron isotopes were produced by fragmentation of a 660 AMeV primary beam of  $^{64}\text{Ni}$  on a 4 g/cm<sup>2</sup> Be target at FRS. The beam energy at the lead target was 535 AMeV. With intensities of 2-5 $\cdot$ 10<sup>7</sup> particles/s, the  $^{60}\text{Fe}$  beam on a 500 mg/cm<sup>2</sup> lead target produce a virtual photons with an energy of astrophysical interest. Figure 1 shows the incom-

ing particle identification from the experimental setup. It is possible to identify all reaction products with R<sup>3</sup>B/LAND setup. The fast neutrons were detected with the LAND detector. The heavy fragments were detected with scintillator walls and fiber detectors.

An activation experiment was performed at the Karlsruhe Van de Graaf accelerator to determine the stellar  $^{60}\text{Fe}(n,\gamma)^{61}\text{Fe}$  average cross section at kT=25 keV of 9.0(2.7) mbarn [8]. The  $^{60}\text{Fe}$  sample for this experiment was prepared at Paul Scherrer Institute, Switzerland and the collaboration was a direct outcome of the first ERAWAST workshop.

Another activation experiment at the University of Mainz TRIGA reactor was performed in order to measure the direct component of  $^{59}\text{Fe}(n,\gamma)^{60}\text{Fe}$  reaction. Furthermore, the experiment at R<sup>3</sup>B/LAND setup generates information about the time-reversal reaction  $^{59}\text{Fe}(\gamma,n)^{58}\text{Fe}$ . Together with experimental results for the direct measurement of  $^{58}\text{Fe}(n,\gamma)^{59}\text{Fe}$ , we will be able to justify the use of this methods.



**Fig. 1:** The incoming particle identification from the experiment. The x-axis shows the relation of mass and charge which is 2.27 for  $^{59}\text{Fe}$ . The y-axis shows the particle charge which is 26 for iron.

### OUTLOOK

The experiment was successfully performed at the

R3B/LAND Setup and is currently under analysis. The next steps of the analysis are the identification of the outgoing particles, tracking of all reaction products and eventually energy-dependent information about the dissociation cross section  ${}^{60}\text{Fe}(\gamma, n){}^{59}\text{Fe}$ . Once the  ${}^{59}\text{Fe}(n, \gamma){}^{60}\text{Fe}$  is determined, nucleosynthesis simulations will be performed, leading to a better understanding of the late stages of massive stars. Furthermore, the Frankfurt Neutron source at the Stern-Gerlach Center (FRANZ) is currently under construction and is designed to produce the highest neutron flux in the keV-regime for astrophysics experiments. The time-of-flight setup will have a time resolution of about 1 ns and a flight path of about 1 m. At the sample position,  $10^7$  neutrons/s/cm<sup>2</sup> are expected. During activations, where the sample can be placed very close to the neutron-producing Li-layer neutrons fluxes of up to  $10^{12}$

neutrons/s/cm<sup>2</sup> are anticipated. We plan to extend the direct measurements performed with  ${}^{60}\text{Fe}$  by activations with higher energies at FRANZ and, if the sample mass permits, by time-of-flight measurements [9].

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## THE STELLAR NEUTRON CAPTURE OF $^{60}\text{Fe}$

R. Reifarth<sup>1</sup>, L.P. Chau<sup>1</sup>, I. Dillmann<sup>4</sup>, C. Domingo Pardo<sup>4</sup>, J. Görres<sup>2</sup>, M. Heil<sup>4</sup>, F. Käppeler<sup>5</sup>, J. Marganiec<sup>4</sup>, O. Meusel<sup>1</sup>, J. Neuhausen<sup>3</sup>, R. Plag<sup>1</sup>, M. Pignatarì<sup>6</sup>, S. Schmidt<sup>1</sup>, D. Schumann<sup>3</sup>, K. Sonnabend<sup>1</sup>, E. Überseder<sup>2</sup>, F. Voss<sup>5</sup>, S. Walter<sup>5</sup>, C. Wiesner<sup>1</sup>, M. Wiescher<sup>2</sup>

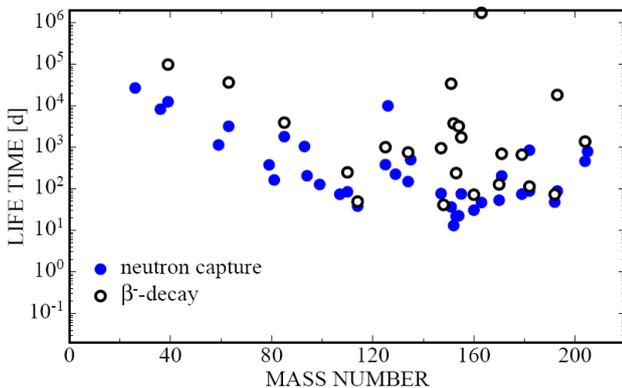
<sup>1</sup>Goethe University Frankfurt, Germany - <sup>2</sup>University of Notre Dame, USA - <sup>3</sup>Paul Scherrer Institut, Switzerland - <sup>4</sup>GSI Helmholtzzentrum f. Schwerionenforschung, Germany - <sup>5</sup>Karlsruhe Institute of Technology, Germany - <sup>6</sup>University Basel, Switzerland

### INTRODUCTION

About 50% of the element abundances beyond iron are produced via slow neutron capture nucleosynthesis (s-process). Starting at iron-peak seed, the s-process mass flow follows the neutron rich side of the valley of stability. If different reaction rates are comparable, the s-process path branches, and the branching ratio reflects the physical conditions in the interior of the star. Such nuclei are most interesting, because they provide the tools to effectively constrain modern models of the stars where the nucleosynthesis occurs. As soon as the  $\beta^-$ -decay is faster than the typically competing neutron capture, no branching will take place.

Therefore experimental neutron capture data for the s-process are only needed, if the respective neutron capture time under stellar conditions is similar or smaller than the  $\beta^-$ -decay time, which includes all stable isotopes. Depending on the actual neutron density during the s-process, the "line of interest" is closer to or farther away from the valley of stability. Within the classical s-process model [1] the neutron density was recently estimated to be  $n_n = 4.94 \cdot 10^8 \text{ cm}^{-3}$  [2].

Fig. 2 shows a summary of the neutron capture and  $\beta^-$ -decay times for radioactive isotopes on the neutron-rich side of the valley of stability, under the condition that the classical neutron capture occurs faster than the terrestrial  $\beta^-$ -decay. Obviously the vast majority of isotopes, where an experimental neutron capture cross section is desirable, has  $\beta^-$ -half-lives of at least hundreds of days.

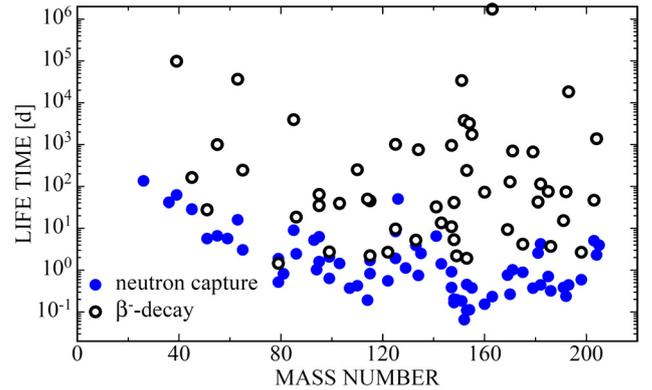


**Fig. 2:** Neutron capture life times (filled circles) for a neutron density of  $5 \cdot 10^8 \text{ cm}^{-3}$  and  $\beta^-$ -life times (open circles) for unstable isotopes on the s-process path as a function of mass number. Shown are only isotopes where

the neutron capture is faster than the (terrestrial)  $\beta^-$ -decay. The neutron capture cross sections are taken from [3].

The modern picture of the main s-process component refers to the He shell burning phase in AGB stars [4]. The highest neutron densities in this model occur during the  $^{22}\text{Ne}(\alpha, n)$  phase and are up to  $10^{11} \text{ cm}^{-3}$ . Fig. 3 shows the same as Fig. 2, but for the higher neutron density. Now isotopes with half-lives down to a few days can be of interest for the s-process reaction network.

Improved experimental techniques, especially as far as the neutron source and sample preparation are concerned, are necessary to perform direct neutron capture measurements on such isotopes [5].



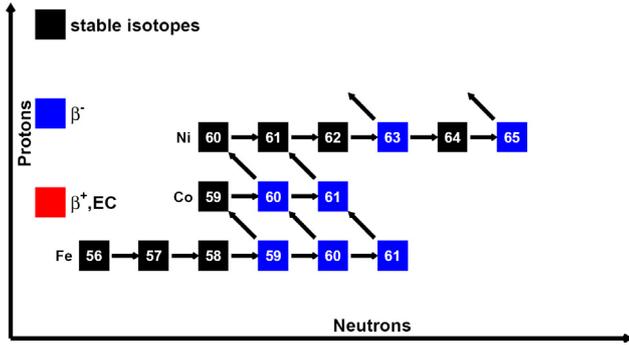
**Fig. 3:** Neutron capture life times (filled circles) for a neutron density of  $10^{11} \text{ cm}^{-3}$  and  $\beta^-$ -life times (open circles) for unstable isotopes on the s-process path as a function of mass number. Shown are only isotopes where the neutron capture is faster than the (terrestrial)  $\beta^-$ -decay. The neutron capture cross sections are taken from [3].

### THE STELLAR PRODUCTION OF $^{60}\text{Fe}$

A significant contribution to the interstellar  $^{60}\text{Fe}$  abundance is provided by the slow neutron capture (s) process in massive stars. The s-process operates in two major evolutionary stages, first during convective core He-burning and, subsequently, during convective shell C-burning. Neutrons are mainly produced by the  $^{22}\text{Ne}(\alpha, n)$  reaction in both cases, but at rather different temperatures and neutron densities [6-8].

As illustrated in Fig. 4, the s-process path to  $^{60}\text{Fe}$ , which starts from the most abundant seed nucleus  $^{56}\text{Fe}$ , is determined by the branching at  $^{59}\text{Fe}$  ( $t_{1/2}=44.5 \text{ d}$ ). At the low neutron densities during convective core He burning,  $^{60}\text{Fe}$  is shielded from the s-process chain, because the  $\beta^-$ -decay rate of  $^{59}\text{Fe}$  dominates over the  $(n, \gamma)$  rate by orders

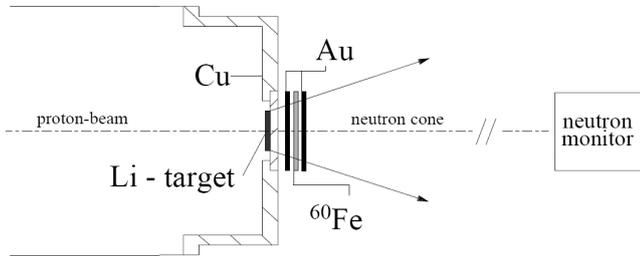
of magnitude. On the other hand, the production of  $^{60}\text{Fe}$  becomes efficient during the shell C-burning phase, where higher temperatures of  $T_9=1.0-1.4$  K give rise to the neutron densities in excess of  $10^{11}\text{ cm}^{-3}$  necessary for bridging the instability gap at  $^{59}\text{Fe}$ . The interpretation of all the above observations depends critically on the reliability of the stellar models as well as on the reaction rates for neutron capture relevant to the production and depletion of  $^{60}\text{Fe}$  [9]. These rates can only be determined reliably in laboratory experiments, because theoretical calculations are too uncertain. Especially in case of  $^{60}\text{Fe}$ , the calculated values may vary by an order of magnitude [10].



**Fig. 4:** The s-process reaction path to  $^{60}\text{Fe}$  via the branching at  $^{59}\text{Fe}$ .

#### NEUTRON CAPTURE ON $^{60}\text{Fe}$

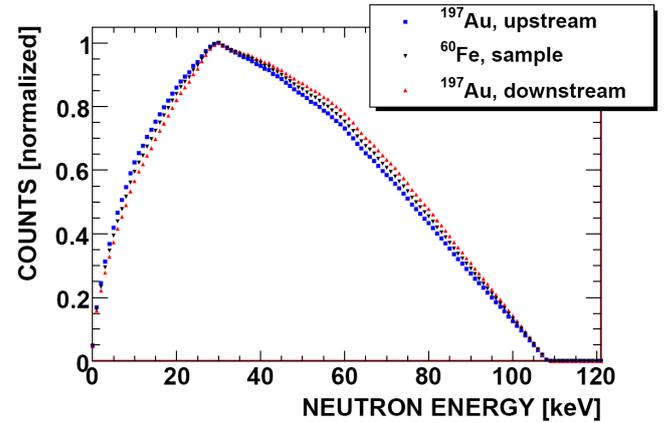
A direct measurement of the  $^{60}\text{Fe}(n,\gamma)$  cross section was performed via the activation technique. Because of the short half life of  $^{61}\text{Fe}$  ( $t_{1/2}=5.98$  min), the activation was carried out as a sequence of cycles. Each cycle consisted of an activation time of  $t_{\text{beam}}=15$  min, a transfer time of about 60 s from the irradiation position to the counting station, the  $\gamma$ -ray detection time  $t_{\text{detector}}=10$  min (during which the proton beam was switched off), and an additional waiting time before the next activation of about 15 min. The sample was sandwiched between two 0.03 mm thin gold foils (see Fig. 5), allowing a measurement relative to the well known  $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$  cross section [11]. At the end the activity of the gold foils was determined via the 412 keV  $\gamma$ -ray from the  $^{198}\text{Au}$  decay ( $t_{1/2}=2.69$ d). Fig. 6 shows a simulation of the neutron spectrum used during the activation.



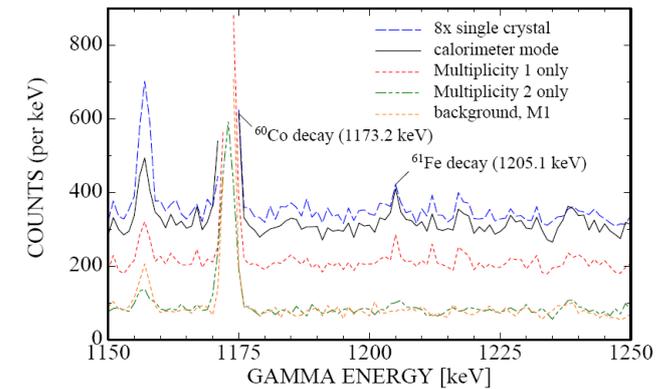
**Fig. 5:** Sketch of the activation setup at the Van de Graaff accelerator.

The required  $^{60}\text{Fe}$  atoms were extracted from a Cu beam dump previously irradiated with high energy protons at PSI. Besides the desired iron isotope, the copper sample contained also about 150 MBq  $^{60}\text{Co}$  and 2 MBq  $^{44}\text{Ti}$  as the main contaminants, which would have impeded the envisaged neutron activation of  $^{60}\text{Fe}$ . Together with the

macro-amount of copper, these contaminants have been carefully separated by liquid-liquid extraction, resulting in decontamination factors of  $\sim 3 \cdot 10^8$  for  $^{60}\text{Co}$  and  $\sim 6 \cdot 10^6$  for  $^{44}\text{Ti}$ . Details of the chemical separation can be found in [12]. In addition to the isotope of interest, the sample contained 100 MBq of  $^{55}\text{Fe}$ , which is co-produced along with  $^{60}\text{Fe}$ , as well as traces of stable iron isotopes, originating partly from nuclear reactions and partly from the drilling process. The  $^{60}\text{Fe}$  contained in the final diluted HCl solution was dried on a graphite backing 6 mm in diameter, which served as the sample for the neutron capture experiment. The number of  $^{60}\text{Fe}$  nuclei was determined from the ingrowth of the daughter activity of  $^{60}\text{Co}$ . Using the latest recommended value for the half life of  $2.62 \pm 0.04$  Myr [13] the amount of  $^{60}\text{Fe}$  was determined to be  $(1.37 \pm 0.07) \cdot 10^{16}$  atoms.



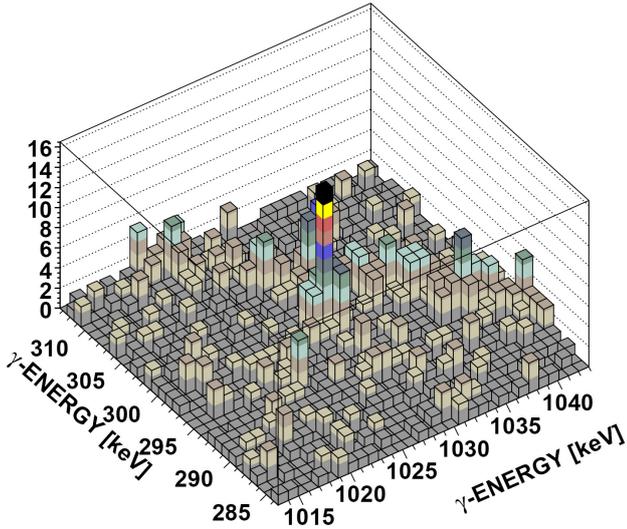
**Fig. 6:** Simulated spectra for the activation at the Van de Graaff accelerator. The three curves correspond to the spectra seen by the sample and the sandwiching gold foils. The simulations were performed with the code PINO [14].



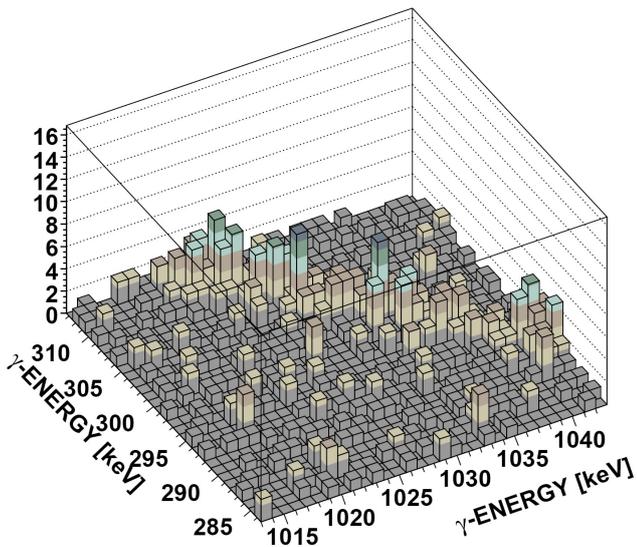
**Fig. 7:** The strongest expected transition from the decay of  $^{61}\text{Fe}$  results in a single  $\gamma$ -ray of 1205.1 keV. Different cuts were applied to the data to improve the signal-to-background ratio. See corresponding text for a detailed discussion.

The induced activity was measured with two Clover type HPGe detectors facing each other in close geometry [15]. The strongest expected transition from the decay of  $^{61}\text{Fe}$  results in a single  $\gamma$ -ray of 1205.1 keV. Fig. 7 shows the result of the search for this line assuming different cuts to increase the signal-to-background ratio. The first approach is treating the clover system as an array of eight

independent detectors. Under this assumption the desired line is barely visible. A slightly improved result is gained under the assumption that the clover system acts as a calorimeter, where the energies of all crystals of one event are added up. The best result is gained, if only events are considered, where exactly one out of eight crystals had fired within the coincidence window. This requirement drastically reduces events resulting from Compton-scattering of the 1332 keV line from the decay of  $^{60}\text{Co}$ . Pure multiplicity-2 events as well as events from a non-activated sample do not show any hint of the 1205 keV line.



**Fig. 8:**  $\gamma$ -ray spectrum of coincident events from the activated  $^{60}\text{Fe}$  sample. The spectrum was obtained by off-line analysis of the data taken with the Ge Clover array. Events due to cascades from the decay of  $^{61}\text{Fe}$  consisting of the 298 and 1027 keV transitions are concentrated in the center, clearly separated from the overall background and from Compton-scattered events of the 1332 keV line from the intrinsic  $^{60}\text{Co}$ , which appear as the diagonal band in the upper right part.



**Fig. 9:** Same plot as Fig. 8, but without the irradiation of the  $^{60}\text{Fe}$  sample. It is obvious that the background as

described in the caption of Fig. 8 is still present, while the signal (events in the center of the plot) vanished.

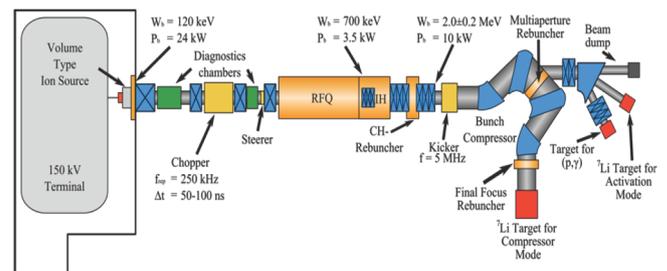
A much clearer picture could be achieved, if only events characterized by two-step cascades were investigated. The strongest two-step cascade consists of the coincident emission of  $\gamma$ -rays with 298 and 1027 keV. Fig. 8 shows the result of such a coincidence requirement, while Fig. 9 shows the same plot for the non-irradiated sample. Especially those two Figures gave us confidence that the observed activity indeed resulted from the freshly produced  $^{61}\text{Fe}$ .

The  $^{60}\text{Fe}(n,\gamma)$  cross section for the experimental neutron spectrum in Fig. 6 is  $5.8 \pm 1.3^{\text{sys}} \pm 0.8^{\text{stat}}$  mb. The extracted Maxwellian averaged cross section at  $kT=25$  keV is then  $5.6 \pm 1.2^{\text{sys}} \pm 0.8^{\text{stat}}$  mb (renormalized from the value given in [16]).

## OUTLOOK

As already discussed in the introductory sections, it is desirable to improve the currently available experimental possibilities for neutron capture experiments. Spallation or photo-neutron sources require large accelerators, but a small accelerator as used for the  $^{60}\text{Fe}$  activation at FZK (see previous section) is best suited for neutron experiments in a university environment. This solution has the additional advantage that the neutron spectrum can be tailored to the specific energy range of interest.

The Stern-Gerlach-Zentrum SGZ, which was recently founded at the University of Frankfurt, intends to build and operate larger experiments in accelerator physics, astrophysics, and material science research. It was decided to develop an intense neutron generator within the next years. The proton driver LINAC currently under construction consists of a high voltage terminal to provide primary proton beam energies of up to  $\sim 150$  keV. A volume type ion source will deliver a DC beam current of 100–250 mA at a proton fraction of 90%. A low energy beam transport using four solenoids will inject the proton beam into an RFQ/IH structure while a chopper at the entrance of the RFQ will create pulse lengths in the range of 100 ns at a repetition rate of up to 250 kHz. A drift tube cavity, which delivers variable end energies between 1.8 and 2.2 MeV, will be installed downstream of the RFQ/IH structure. Finally a bunch compressor of the Mobley type forms a proton pulse length of 1 ns at the Li target. The maximum energies of the neutrons will be adjustable between 200 keV and about 500 keV by the primary proton beam energy (Fig. 10).



**Fig. 10:** Schematic layout of the Frankfurter Neutron Source FRANZ.

This facility will offer the possibility to investigate the neutron capture cross section of  $^{60}\text{Fe}$  also at higher energies via activation with the currently available sample. This is important, because the production of  $^{60}\text{Fe}$  happens most likely at temperatures around 90 keV. In principle also a time-of-flight measurement is possible, but the sample requirements are much more stringent in that case. The amount of other iron isotopes would have to be reduced to less than 10% in order to make a successful measurement.

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