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PROGRESS REPORT ON NUCLEAR DATA RESEARCH IN THE FEDERAL REPUBLIC OF GERMANY

for the Period April 1, 2004 to September 30, 2005

December 2005

Edited by
S. M. Qaim
Forschungszentrum Jülich GmbH
Institut für Nuklearchemie
Jülich, Federal Republic of Germany

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FOREWORD

As in previous years, this report has been prepared to promote exchange of nuclear data research information between the Federal Republic of Germany and other member states of OECD/NEA and IAEA. It covers progress reports from the research centres at Karlsruhe and Jülich, the universities of Dresden, Hannover and Köln, as well as from the PTB Braunschweig. Each contribution is presented under the laboratory heading from where the work is reported. The names of other participating laboratories are also mentioned. The emphasis in the work reported here is on nuclear data for applied science programmes, such as those relevant to reactor technology and safety, transmutation concepts, accelerator shielding and development, astrophysical research, production of medically important radioisotopes, radiation therapy, etc.

The coordination of nuclear data activities at the international level is done by two committees: the NEA-Nuclear Science Committee (NEA-NSC) and the IAEA-International Nuclear Data Committee (INDC). The present Editor has the privilege and the responsibility of representing Germany in both the committees. This report therefore also serves as a background information to some areas of work of those committees.

Jülich, December 2005

S.M. Qaim

This document contains information of a preliminary nature. Its contents should be used with discretion.

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FORSCHUNGSZENTRUM KARLSRUHE INSTITUT FÜR KERNPHYSIK

1. Measurement of Neutron Capture Cross Sections for ADS-Related Studies*

P.M. Milazzo¹, G. Aerts², E. Berthoumieux², N. Bustreo³, D. Cano-Ott⁴, P. Cennini⁵, N. Colonna⁶, C. Domingo⁷, M. Embid⁴, L. Ferrant⁸, E. Gonzales⁴, F. Gunsing², M. Heil⁹, F. Käppeler⁹, S. Marrone⁶, P. F. Mastinu³, A. Mengoni^{5,10}, C. Moreau⁸, J. Pancin¹¹, T. Papaevangelou¹², C. Paradela¹³, P. Pavlopoulos¹², R. Plag⁹, R. Reifarh⁹, C. Stephan⁸, L. Tassan-Got⁸, G. Tagliente⁶, J. L. Tain⁷, R. Terlizzi⁶, V. Vlachoudis⁵

Capture cross sections on several isotopes relevant to accelerator driven systems for energy production and nuclear waste transmutation, and to stellar nucleosynthesis can be studied at the innovative neutron time of flight facility (n_TOF) at CERN. The experimental apparatus is based on a low-mass Si-based flux monitor and a set of C₆D₆ liquid scintillator detectors. The accurate reconstruction of the cross sections relies on the pulse height weighting function technique. The set-up used in the measurements is described here. The first results on reference isotopes, Au, Ag and Fe, used to verify the accuracy of the method are presented.

* Nucl. Instr. Meth. Phys. Res. B, 213 (2004) 36

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¹² Astroparticle Consortium, Greece

¹³ Universidad de Santiago de Compostela, Spain

2. Neutron Capture Studies on ¹³⁵Cs for Nucleosynthesis and Transmutation*

N. Patronis¹, P.A. Assimakopoulos¹, D. Karamanis¹, S. Dababneh^{2,3}, M. Heil³, F. Käppeler³, R. Plag³, P.E. Koehler⁴, A. Mengoni⁵, R. Gallino⁶, and the n_TOF collaboration⁷

The neutron capture cross section of the unstable isotope ¹³⁵Cs was measured relative to that of gold by means of the activation method. The sample was produced by ion implantation in a high resolution mass separator and irradiated with quasi-monoenergetic neutrons at 30 keV and 500 keV, using the ⁷Li(p,n)⁷Be reaction. An additional irradiation with thermal neutrons has been carried out for defining the sample mass and for measuring the half-life of ¹³⁶Cs. The neutron capture cross sections were determined as 164 ± 10 mbarn and 34.8 ± 3.0 mbarn at 30 keV and 500 keV, respectively, and were used to normalize the theoretically derived cross section shape. Based

on these data, refined statistical model calculations were performed to obtain the (n, γ) cross sections of the short-lived isotopes ^{134}Cs and ^{136}Cs . Updated Maxwellian averaged capture cross sections of all unstable Cs isotopes were calculated for a range of thermal energies characteristic of helium burning scenarios for an improved s-process analysis of the Xe-Cs-Ba region.

* *Phys. Rev. C*, 69 (2004) 025803

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⁷ The n_TOF collaboration, see <http://pceet075.cern.ch/>

3. Neutron Capture Measurements at a RIA-Type Facility*

R. Reifarh¹, R.C. Haight¹, M. Heil², F. Käppeler², D.J. Vieira¹

Neutron capture cross sections of unstable isotopes are important for neutron induced nucleosynthesis as well as for technological applications. The Rare Isotope Accelerator (RIA) or comparable facilities will be able to produce radioactive ion beams up to 10^{12} particles/s and would, therefore, be a suitable place for (n, γ) studies on radioactive isotopes with half-lives between days and months. We propose a facility for measurements of (n, γ) cross sections of unstable isotopes in the keV range suited for minimal sample masses down to 10^{15} atoms, corresponding to minimum half-lives of only 10 d.

* *Nucl. Instr. Meth. Phys. Res. A*, 524 (2004) 215

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² Forschungszentrum Karlsruhe, Institut für Kernphysik, 76021 Karlsruhe, Germany

4. Stellar Neutron Capture on $^{180}\text{Ta}^m$. I. Cross Section Measurement Between 10 and 100 keV*

K. Wisshak¹, F. Voss¹, C. Arlandini¹, F. Käppeler¹, M. Heil¹, R. Reifarh², M. Krťicka³, F. Becvar³

The neutron capture cross section of $^{180}\text{Ta}^m$ has been measured at energies between 10 keV and 100 keV in a time-of-flight experiment at the Karlsruhe 3.7 MV Van de Graaff accelerator. Neutrons were produced via the $^7\text{Li}(p, n)^7\text{Be}$ reaction by bombarding metallic Li targets with a pulsed proton beam, and gold was used as a cross section standard. Though the world supply of enriched $^{180}\text{Ta}^m$ was available for this experiment, the sample consisted of only 150 mg Ta_2O_5 with a $^{180}\text{Ta}^m$ content of only 5.5%. The difficult separation of the comparably few capture events in $^{180}\text{Ta}^m$ from the much larger background due to captures in ^{181}Ta could be achieved by means of the Karlsruhe 4π Barium Fluoride Detector, taking advantage of its combination of high efficiency, good energy resolution, and high granularity. The cross section was determined with an overall uncertainty of better than 10% in the energy range from 30 keV to 100 keV and could be used for renormalizing statistical model calculations in the entire energy range of astrophysical interest, which had predicted about two times larger values. Based on these first experimental data, Maxwellian

averaged neutron capture cross sections were calculated for thermal energies between $kT = 8$ keV and 100 keV.

* Phys. Rev. C, 69 (2004) 55801

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5. **Stellar Neutron Capture on $^{180}\text{Ta}^m$. II. Defining the s -Process Contribution of Nature's Rarest Isotope***

F. Käppeler¹, C. Arlandini¹, M. Heil¹, F. Voss¹, K. Wisshak¹, R. Reifarth², O. Straniero³, R. Gallino⁴, S. Masera⁴, C. Travaglio⁵

The contribution of the slow neutron capture process (s process) to the solar $^{180}\text{Ta}^m$ abundance has been investigated on the basis of new experimental information. Measured neutron capture cross sections of $^{180}\text{Ta}^m$ and the corresponding Maxwellian averaged (n, γ) rates were important for defining the s abundance of $^{180}\text{Ta}^m$, and the result of a recent photoactivation experiment was provided an estimate of its half-life at the temperatures of the s -process site. Following the s -process network with stellar evolutionary models from the pre-main sequence through the asymptotic giant branch (AGB) phase, it was found that the produced $^{180}\text{Ta}^m$ survives the high temperatures during He shell flashes because of the fast convective mixing, which provides an efficient means for transporting freshly synthesized matter into cooler, outer zones. Accordingly, $^{180}\text{Ta}^m$ appears to be predominantly of s -process origin.

* Phys. Rev. C, 69 (2004) 55802

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6. **^{128}Xe and ^{130}Xe : Testing He Shell Burning in AGB Stars***

R. Reifarth¹, F. Käppeler², F. Voss², K. Wisshak², R. Gallino³, M. Pignatari³, O. Straniero⁴

The s -process branching at ^{128}I has been investigated on the basis of new, precise experimental (n, γ) cross sections for the s -only isotopes ^{128}Xe and ^{130}Xe . This branching is unique since it is essentially determined by the temperature- and density-sensitive stellar decay rates of ^{128}I , whereas it is only marginally affected by the specific stellar neutron flux. For this reason it represents an important test for He-shell burning in AGB stars. The description of the branching by means of the complex stellar scenario reveals a significant sensitivity to the time scales for convection during He shell flashes, thus providing model-free constraints for this phenomenon. The s -process ratio $^{128}\text{Xe}/^{130}\text{Xe}$ deduced from stellar models allows for a $9 \pm 3\%$ p -process contribution to solar ^{128}Xe , in agreement with the Xe-S component found in meteoritic presolar SiC grains.

* Ap. J., 614 (2004) 363

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7. Neutron Capture Cross Section Measurement of ^{151}Sm at n_TOF*

U. Abbondanno¹ et al.²

The $^{151}\text{Sm}(n, \gamma)^{152}\text{Sm}$ cross section has been measured at the spallation neutron facility n_TOF at CERN in the energy range from 1 eV to 1 MeV. The new facility combines excellent resolution in neutron time-of-flight, low repetition rates, and an unsurpassed instantaneous luminosity, resulting in rather favourable signal/background ratios. The ^{151}Sm cross section is of importance for characterizing neutron capture nucleosynthesis in Asymptotic Giant Branch stars. At a thermal energy of $kT=30$ keV the Maxwellian averaged cross section of this unstable isotope ($t_{1/2} = 93$ yr) was determined to be 3100 ± 160 mb, significantly larger than theoretical predictions.

* Phys. Rev. Lett., 93 (2004) 161103

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² The n_TOF collaboration, see <http://pceet075.cern.ch/>

8. Nucleosynthesis at the Termination Point of the s Process*

U. Ratzel¹, C. Arlandini¹, F. Käppeler¹, A. Couture², M. Wiescher², R. Reifarth³, R. Gallino⁴, A. Mengoni⁵, C. Travaglio⁶

Stellar cross sections of importance with respect to the termination of the s -process reaction chain have been determined for the two cases, $^{208}\text{Pb}(n, \gamma)^{209}\text{Pb}$ and $^{209}\text{Bi}(n, \gamma)^{210}\text{Bi}^g$, yielding $kT=30$ keV values of $\langle \sigma v \rangle / v_T = 0.30 \pm 0.02$ mb and 2.54 ± 0.14 mb, respectively. The measurements were carried out by activation of Pb and Bi samples in a quasi-stellar neutron spectrum using gold as a cross section standard. With this technique the uncertainties reported in previous works could be considerably reduced. The measurements are complemented by a discussion of the recycling at the termination point of the s -process neutron capture chain in a $3 M_{\text{sol}}$ and $[\text{Fe}/\text{H}] = -1.3$ asymptotic giant branch star. At this metallicity AGB stars give rise to the maximum production of s -process lead. The sensitivity of the isotopic lead abundances is discussed with respect to the remaining cross section uncertainties. The information obtained in this work is also of relevance for an assessment of the α -activity due to build-up of ^{210}Po in Pb/Bi cooled fast reactor systems.

* Phys. Rev. C, 70 (2004) 065803

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9. Time–Energy Relation of the n_TOF Neutron Beam: Energy Standards Revisited*

G. Lorusso¹ et al.²

The accurate determination of neutron cross-sections as a function of the neutron energy at a time-of-flight facility requires a precise knowledge of the time–energy relation for the neutron beam. For the n_TOF neutron beam at CERN, produced by spallation of high-energy protons on a Pb target, the time–energy relation is connected to the production mechanism and to the subsequent moderation process. A calibration of the neutron energy scale is proposed on the basis of detailed Monte Carlo simulations of the facility. This time–energy relation has been experimentally validated by means of dedicated measurements of standard energy resonances, from 1 eV to approximately 1 MeV. On the basis of the present measurements, it is proposed to correct the energy of the 1.3 eV resonance of ¹⁹³Ir, which is commonly considered as an energy standard.

* Nucl. Instr. Meth. Phys. Res. A, 532 (2004) 622

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10. The Data Acquisition System of the Neutron Time of Flight Facility n_TOF at CERN*

U. Abbondanno¹ et al.²

The n_TOF facility at CERN has been designed for the measurement of neutron capture, fission and (*n,xn*) cross sections with high accuracy. This requires a flexible and—due to the high instantaneous neutron flux—almost dead time free data acquisition system. A scalable and versatile data solution has been designed based on 8-bit flash-ADCs with sampling rates up to 2 GHz and 8 Mbyte memory buffer. The software is written in C and C++ and is running on PCs equipped with RedHat Linux.

* Nucl. Instr. Meth. Phys. Res. A, 538 (2005) 692

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11. Neutron Reactions in Astrophysics: The Role of New Facilities and Improved Laboratory Approaches*

F. Käppeler¹

The abundances of the elements between Fe and the actinides have essentially all been produced by neutron reactions, either by the slow neutron capture process during helium burning in Red Giant stars (*s* process) or by explosive nucleosynthesis, e.g. in supernovae (*r* and *p* process). Laboratory studies have so far concentrated on the *s* process, which operates in or near the valley of beta stability. Under these conditions laboratory results are of direct impact for the interpretation of the observed abundance patterns and their role as crucial tests for stellar evolution models. The high flux at spallation neutron sources allows one to investigate numerous difficult and hitherto inaccessible cases, including the cross sections of important radioactive nuclei.

* J. Neutron Res., 13 (2005) 33

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12. Quasistellar Spectrum for Neutron Activation Measurements at $kT = 5$ keV*

M. Heil¹, S. Dababneh^{1,2}, A. Juseviciute¹, F. Käppeler¹, R. Plag¹, R. Reifarth³, S. O'Brien⁴

We have measured the neutron energy spectrum of the $^{18}\text{O}(p,n)^{18}\text{F}$ reaction at a proton energy of 2582 keV, 8 keV above the reaction threshold. At this energy the resulting neutron spectrum resembles almost perfectly a Maxwellian distribution at a thermal energy of $kT = 5.1 \pm 0.1$ keV. Since all neutrons are emitted in a forward cone of 140 deg. opening angle, this reaction can be used for neutron activation measurements similar to the $^7\text{Li}(p,n)^7\text{Be}$ reaction, which is known for producing a thermal spectrum with $kT = 25$ keV. Measured neutron capture cross sections at $kT = 5.1$ keV and $kT = 25$ keV can be used to interpolate to $kT = 8$ keV, which characterizes the dominant neutron exposure during s -process nucleosynthesis in thermally pulsing low-mass AGB stars. In a first application of this new method the Maxwellian-averaged neutron capture cross section of ^{138}Ba was measured to be $\sigma v/v_T = 13.0 \pm 0.5$ mb at $kT = 5.1$ keV.

* Phys. Rev. C, 71 (2005) 025803

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13. s -Process Nucleosynthesis and the Interior of Red Giants*

F. Käppeler¹

Nucleosynthesis by the slow neutron capture process (s process) in the He burning zones of Red Giant stars contributes about half of the isotopic abundances in the mass region between Fe and Bi. Since these abundances are essentially determined by the respective neutron capture cross sections of stable and long-lived isotopes, the nuclear physics part of the s -process problem can be studied in laboratory experiments. The status of these data and the remaining experimental quests are briefly sketched in the light of recent achievements and future possibilities. Presently, s -process calculations are capable of providing a quantitative description of the observed abundances that can be attributed to an s -process origin. Moreover, these results can be interpreted as constraints for models of the red giant phase, an important feature for improving our understanding of persisting difficulties in stellar evolution.

* Nucl. Phys., A752 (2005) 500c

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FORSCHUNGSZENTRUM KARLSRUHE
INSTITUT FÜR REAKTORSICHERHEIT

1. Evaluation of W Cross Sections up to 150 MeV
P. Pereslavitsev, U. Fischer

In the frame of the European Fusion Technology Programme, general purpose data evaluations were performed for the cross sections of the reaction systems $n + {}^{182}\text{W}$, ${}^{183}\text{W}$, ${}^{184}\text{W}$, ${}^{186}\text{W}$ up to 150 MeV neutron energy employing the evaluation procedure outlined in Fig. 1.

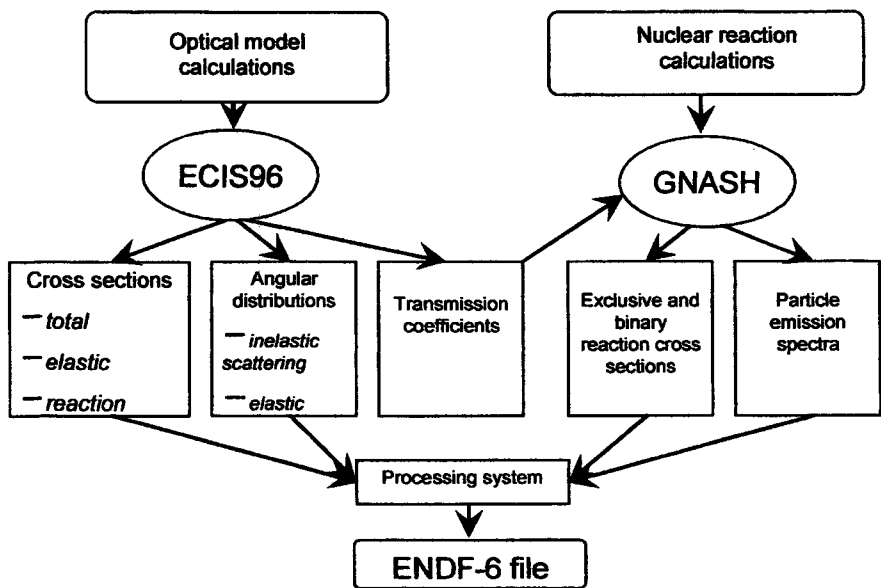


Fig. 1 Flowchart for the evaluation of neutron cross section data of the W isotopes up to 150 MeV neutron energy

Total, reaction, elastic and inelastic scattering cross sections were calculated with the ECIS96 code applying both spherical and deformed optical model potentials. Nuclear model calculations were performed with the GNASH code utilizing the Hauser-Feshbach theory for multiple equilibrium and the exciton model for pre-equilibrium particle emissions up to 150 MeV. Measurements of the total cross sections performed recently at Los Alamos for the tungsten isotopes were used to select the high energy neutron optical model potential. Evaluated and measured total

cross sections of the four W isotopes are compared in Fig. 2. Note the perfect agreement of the new evaluations (red curves) with the measured cross-sections over the entire energy range.

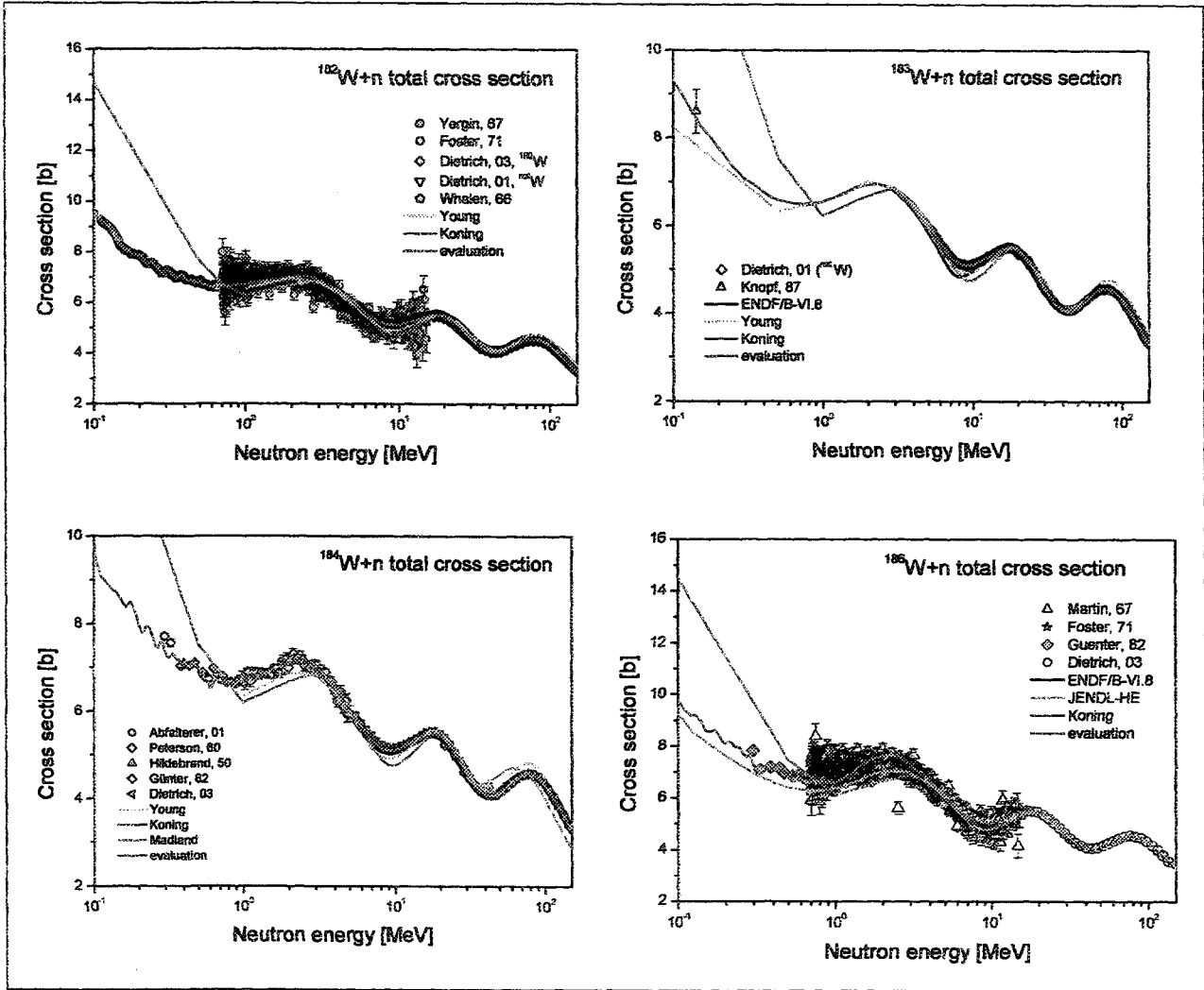


Fig. 2 Evaluated and measured total cross-sections for the W isotopes 182, 183, 184 and 186 up to 150 MeV neutron energy.

Complete nuclear data files were eventually prepared in the standard ENDF-6 data format. The data files were processed with the ACER module of the NJOY code and checked by means of MCNP Monte Carlo calculations for an integral benchmark experiment on tungsten. The tungsten data evaluations will be complemented by covariance data provided by the University of Vienna, and then will be integrated to the European Fusion File (EFF).

Reference

- [1] P. Pereslavl'tsev, U. Fischer, Evaluation of $n + W$ cross section data up to 150 MeV neutron energy, Int. Conf. on Nuclear Data for Science and Technology, Santa Fe, NM, September 26-October 1, 2004, AIP Conference Proceedings Vol. 769, Melville, New York 2005, pp 215-218.

2. Evaluation of D-Li Cross Section Data up to 50 MeV

P. Pereslavl'tsev, M. Avrigeanu, U. Fischer, S. P. Simakov

The McDeLicious approach [1] for the representation of the D-Li source term of the IFMIF neutron source requires the availability of a complete set of evaluated nuclear cross sections for the $d + {}^{6,7}\text{Li}$ reactions up to 50 MeV deuteron energy. A first set of such data was evaluated previously in a collaboration of FZK and INPE Obninsk [2].

Recent experimental results of the angle-energy distributions of the neutron yields and measurements of double-differential $d + {}^{6,7}\text{Li}$ cross sections initiated an effort to re-evaluate the $d + {}^{6,7}\text{Li}$ cross section data, applying a new methodology which takes into account compound nucleus reactions, pre-equilibrium processes, stripping and direct interactions. While the first two reactions are described by means of nuclear model calculations with the GNASH code, the stripping process is represented on the basis of the semi-empirical Serber model and the direct reactions with the DWUCK4 code. A microscopic optical model potential is used for incident deuterons and a modified Koning global optical model potential for neutrons and protons.

An example of the evaluated ${}^7\text{Li}(d,xn)$ double differential neutron emission cross section and its decomposition into reaction components is shown in the figure for 16.6 MeV deuterons. The new $d + {}^{6,7}\text{Li}$ data evaluation was shown to result in a significant better reproduction of measured thick and thin lithium target neutron yield spectra when applied with the McDeLicious approach.

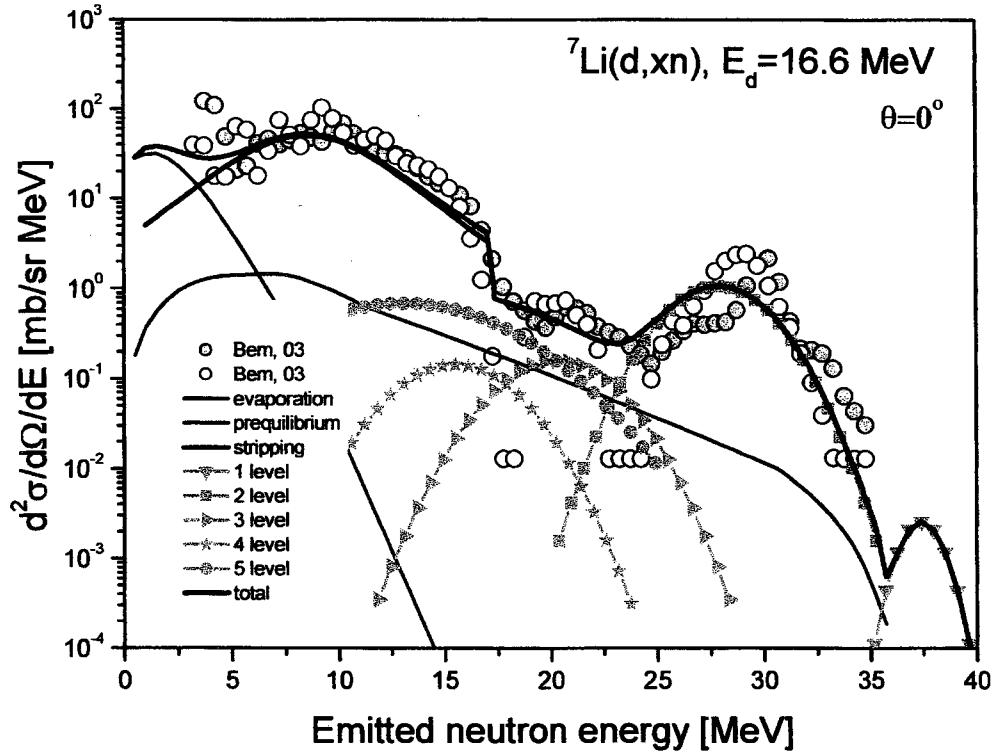


Fig. 3 ${}^7\text{Li}(d,xn)$ double differential neutron emission cross section at 16.6 MeV deuteron energy

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- [1] S.P. Simakov, U. Fischer, U. von Möllendorff, I. Schmuck, A.Yu. Konobeev, Yu. A. Korovin, P. Pereslavitsev, Advanced Monte Carlo procedure for the d-Li neutron source term based on evaluated cross section files, J. Nucl. Mat. **307-311** (2002), 1710-1714
- [2] A. Yu. Konobeyev, Yu. A. Korovin, P. E. Pereslavitsev, U. Fischer, U. von Möllendorff, Developments of methods for calculation of deuteron-lithium and neutron-lithium cross sections for energies up to 50 MeV, Nucl. Sci. Eng. **139** (2001) 1

INSTITUT FÜR NUKLEARCHEMIE FORSCHUNGSZENTRUM JÜLICH

1. Fundamental Studies on Isomeric Cross Sections

S. M. Qaim, S. Sudár*, M. Al-Abyad†

In continuation of our fundamental studies on the formation of isomeric states in a variety of nuclear reactions we completed investigations on the high-spin isomers ^{195m}Hg ($J^\pi = 13/2^+$), ^{197m}Hg ($J^\pi = 13/2^+$) and ^{196m}Au ($J^\pi = 12^-$). The formation of the isomeric pair $^{195m,g}\text{Hg}$ was investigated in $^{\text{nat}}\text{Pt}(^3\text{He},\text{xn})$ and $^{196}\text{Hg}(\text{n},2\text{n})$ reactions, that of $^{197m,g}\text{Hg}$ in $^{\text{nat}}\text{Pt}(^3\text{He},\text{xn})$, $^{\text{nat}}\text{Pt}(\alpha,\text{xn})$, $^{197}\text{Au}(\text{p},\text{n})$ and $^{198}\text{Hg}(\text{n},2\text{n})$ reactions, and of the pair $^{196m,g}\text{Au}$ in the $^{\text{nat}}\text{Pt}(^3\text{He},\text{x})$ reaction. The energy ranges covered extended from the respective reaction threshold up to 12.5 MeV in the case of neutrons (produced via the dd reaction using a deuterium gas target at the Compact Cyclotron CV 28), 20 MeV in the case of protons, 35 MeV for ^3He -particles and 26 MeV for α -particles. In each case the activation technique was used and the reaction products were identified via high-resolution HPGe detector γ -ray spectrometry. From the experimental data for the formation of the metastable and ground states, the isomeric cross-section ratios were calculated.

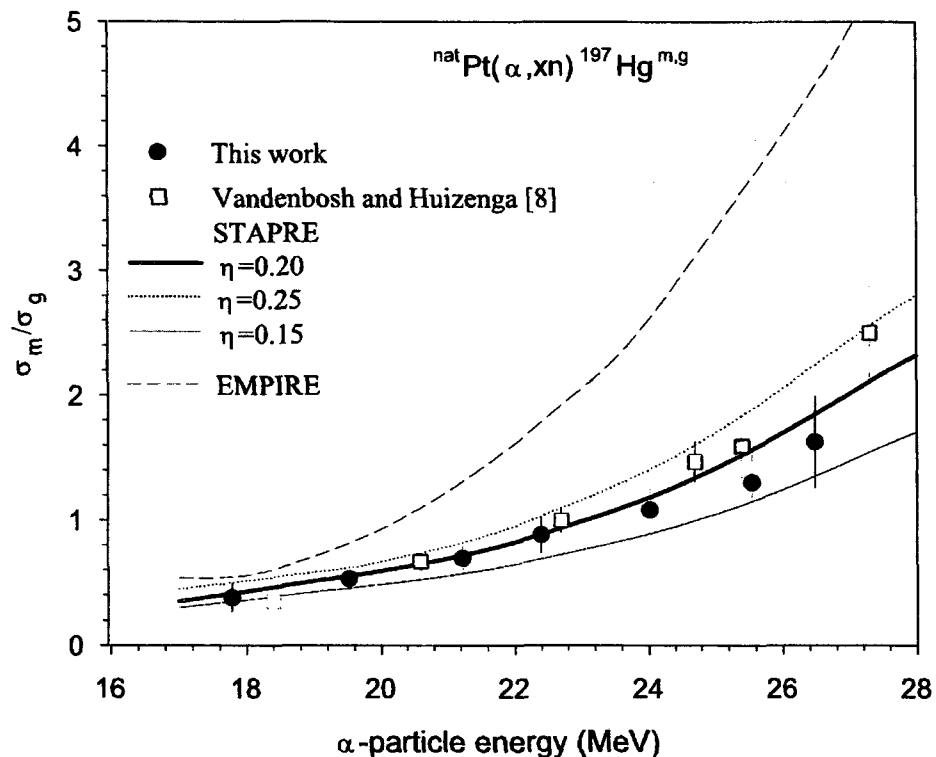
Nuclear model calculations using the code STAPRE, which employs the Hauser-Feshbach (statistical model) and exciton model (precompound effects) formalisms, were undertaken to describe the formation of both the isomeric and ground states of the products. The calculations were compared with the results of the EMPIRE-II code. The total cross section of each reaction is described fairly well by the model calculations, with STAPRE giving slightly better results. Regarding the isomeric cross sections, the agreement between the experiment and theory is only in approximate terms.

As an example, the experimental and theoretical isomeric cross-section ratios for the isomeric pair $^{197m,g}\text{Hg}$ in $^{\text{nat}}\text{Pt}(\alpha,\text{xn})$ and $^{198}\text{Hg}(\text{n},2\text{n})$ reactions are reproduced in Fig. 1. In each case the ratio increases with the increasing projectile energy, suggesting that

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(A)



(B)

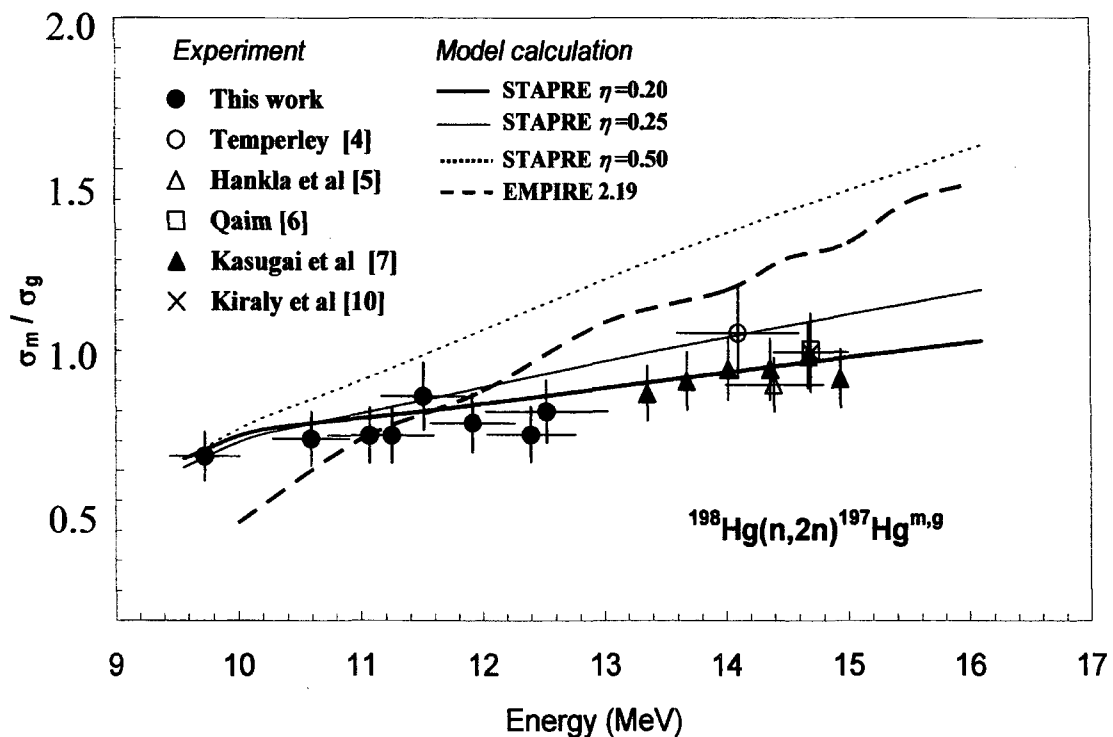


Fig. 1 Isomeric cross-section ratio for the isomeric pair $^{197m,g}\text{Hg}$ in (A) $^{nat}\text{Pt}(\alpha, xn)$, and (B) $^{198}\text{Hg}(n, 2n)$ reactions. The STAPRE calculations were done using different values of η .

the probability of formation of the high-spin metastable state increases with the increasing incident particle energy. This observation is in conformity with the earlier results on the isomeric pairs where, however, the spin difference between the metastable and ground states was not so high. An interesting feature of the present investigations is that the isomeric cross-section ratio is described well by the model calculation only when a relatively low value of η (about 0.20) is used. This result is in contrast to the η value of about 0.75 found appropriate in the light mass region and about 0.50 in the medium mass region. The present value of about 0.20 in the mass region 190 – 200 suggests that η is mass dependent. In physical terms this implies that the spin distribution of the level density is mass dependent. Based on the results of these investigations, two manuscripts have been prepared and submitted to Phys. Rev. C for publication.

2. Neutron Induced Reaction Cross Section Data

M. Al-Abyad[†], I. Spahn, S.Sudár^{*}, M. N. H. Comsan[†], S. M. Qaim

Excitation functions were measured for the reactions $^{196}\text{Hg}(n,2n)^{195\text{m,g}}\text{Hg}$, $^{198}\text{Hg}(n,2n)^{197\text{m,g}}\text{Hg}$, $^{204}\text{Hg}(n,2n)^{203}\text{Hg}$, $^{198}\text{Hg}(n,p)^{198}\text{Au}$ and $^{199}\text{Hg}(n,p)^{199}\text{Au}$, over the neutron energy range of 7.6 to 12.5 MeV. Quasi-monoenergetic neutrons were produced via the dd reaction on a deuterium gas target. Use was made of the activation technique in combination with high-resolution HPGe-detector γ -ray spectrometry. All the data were measured for the first time over the investigated energy range. The transition from the present low-energy data to the literature data around 14 MeV was found to be generally good. Nuclear model calculations using the codes STAPRE and EMPIRE-2.19, both of which employ the statistical and precompound model formalisms, were undertaken to describe the formation of the products. The total reaction cross section of a particular channel is reproduced fairly well by the model calculations, with STAPRE giving slightly better results. The results reported in this study should provide a good database for the (n,2n) and (n,p) reactions on several

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isotopes of mercury, especially with regard to calculations on neutron multiplication while using mercury as a target material in a spallation neutron source.

Integral cross sections of a few nuclear reactions were measured using a 14 MeV d(Be) neutron source. Of particular interest were the reactions $^{32}\text{S}(n,p)^{32}\text{P}$, $^{90}\text{Zr}(n,p)^{90}\text{Y}$ and $^{153}\text{Eu}(n,p)^{153}\text{Sm}$ which lead to the formation of the therapeutic radionuclides ^{32}P ($T_{1/2} = 14.3$ d), ^{90}Y ($T_{1/2} = 2.7$ d) and ^{153}Sm ($T_{1/2} = 1.9$ d). The measured cross sections are about three times larger than those with fission neutrons. The present data could be of interest in planning the production of these three important radionuclides using a fast neutron spectral source, e.g. a spallation source or a fusion reactor.

3. Charged Particle Induced Reaction Cross Section Data

As in previous years, systematic studies on charged particle induced reaction cross sections were continued. During the period of the present Progress Report following investigations were carried out.

a. *Cross sections of nuclear reactions relevant to the production of the positron emitters ^{72}As , ^{124}I and ^{82}Sr (^{82}Rb)*

K.F. Hassan[†], I. Spahn, B. Scholten, S. Spellerberg, D. Steyn[‡], N. van der Walt[‡], Z.A. Saleh[†], S. M. Qaim, H. H. Coenen

The positron emitting radionuclide ^{72}As ($T_{1/2} = 26.0$ h) is of considerable potential interest for use in Positron Emission Tomography. It can be produced either directly via proton and deuteron induced reactions on isotopes of germanium or can be obtained through the decay of ^{72}Se ($T_{1/2} = 8.5$ d) using a $^{72}\text{Se} - ^{72}\text{As}$ generator system. During the period of this Progress Report extensive measurements were carried out on proton induced reactions on ^{nat}Ge , with particular reference to the formation of ^{74}As ($T_{1/2} = 17.8$ d), ^{72}As ($T_{1/2} = 26.0$ h) and ^{71}As ($T_{1/2} = 65.3$ h). The investigated energy range extended up to 66 MeV. Irradiations were performed at the compact cyclotron CV 28 and injector of COSY (both at the Forschungszentrum Jülich), as well as at the Separate Sector

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Accelerator (at the iThemba LABS, Cape Town). Furthermore, a few irradiations were carried using highly enriched ^{72}Ge as target material. An analysis of the data is in progress.

The positron emitting radionuclide ^{124}I ($T_{1/2} = 4.18$ d) is both a diagnostic and a therapeutic nuclide and is attracting considerable attention for application in PET studies. Because of its increasing significance, a continuous search for its alternative methods of production is under way. The commonly used processes are $^{124}\text{Te}(p,n)^{124}\text{I}$ and $^{124}\text{Te}(d,2n)^{124}\text{I}$, whereby the (p,n) reaction leads to the highest purity product. In addition the $^{125}\text{Te}(p,2n)^{124}\text{I}$ and $^{126}\text{Te}(p,3n)^{124}\text{I}$ reactions have also been studied: the ^{124}I yield in both the reactions is high but the impurity level is also high. Now we investigated the ^3He - and α -particle induced reactions on $^{\text{nat}}\text{Sb}$ and ^{121}Sb . Thin samples of $^{\text{nat}}\text{Sb}$ and 99.45 % enriched ^{121}Sb , prepared via the sedimentation process, were stacked together and irradiated with 36 MeV ^3He -particles or 26.5 MeV α -particles at the compact cyclotron CV 28 in Jülich. The radioactivity of the samples was determined via high-resolution γ -ray spectrometry. The cross section data obtained for the formation of ^{124}I and ^{123}I in α -particle induced reactions on enriched ^{121}Sb are shown in Fig. 2. The data for the formation of ^{124}I in comparison to those for ^{123}I are low, but since ^{123}I is rather short-lived ($T_{1/2} = 13.2$ h), it may be possible to produce ^{124}I in a pure form. For further analysis of the production possibilities, the yields of ^{124}I and the radionuclidic impurities were calculated using the various excitation functions measured in this work. The results are given in Table 1. All values correspond to the separation of radioiodine 5 days after end of bombardment (EOB). Evidently only the α -particle induced reaction on highly enriched ^{121}Sb leads to high-purity ^{124}I ; the overall yield, however, is rather low. For general comparison it may be pointed out that, because of their low yields, the ^3He - and α -particle induced reactions on natural or enriched antimony cannot compete with the $^{124}\text{Te}(p,n)^{124}\text{I}$ process commonly used today. The detailed results of these investigations will be published in two papers in Appl. Radiat. Isotopes.

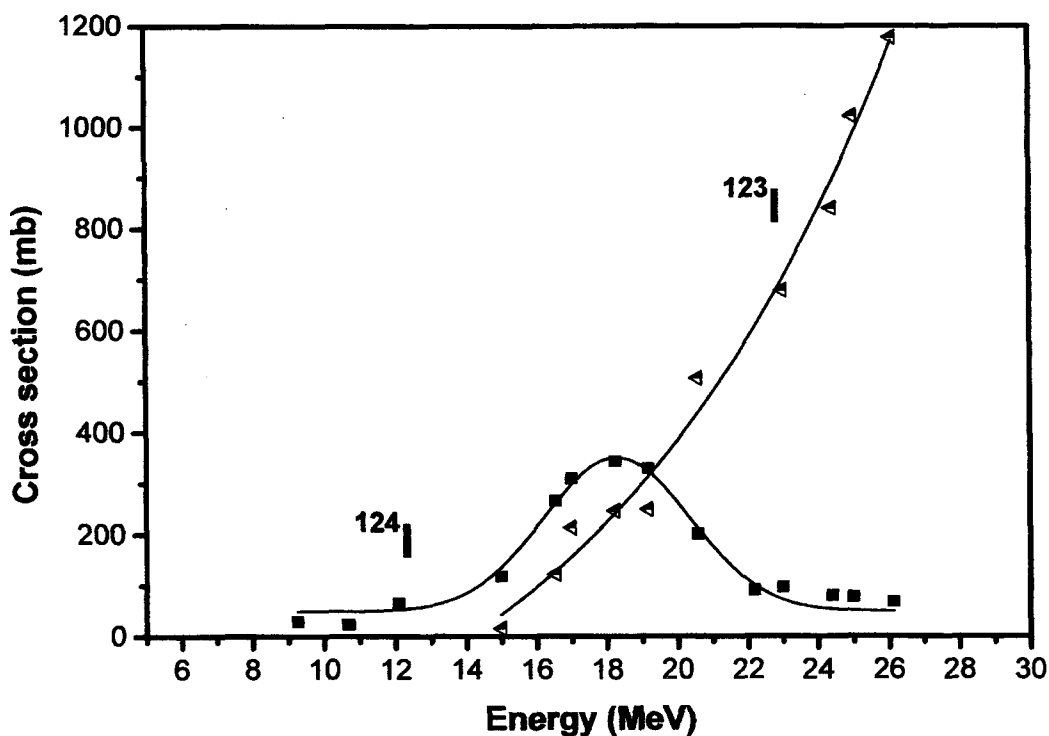


Fig. 2 Excitation functions of $^{121}\text{Sb}(\alpha, xn)^{124,123}\text{I}$ reactions.

Table 1 Comparison of ^3He - and α -particle induced reactions on antimony for production of ^{124}I

Nuclear reaction	Suitable energy range (MeV)	^{124}I yield* (MBq/ $\mu\text{A}\cdot\text{h}$)	Radionuclidic impurities (%)*		
			^{123}I	^{125}I	^{126}I
$^{\text{nat}}\text{Sb}(^3\text{He}, xn)^{124}\text{I}$	35 \rightarrow 13	0.42	14	1.3	1.2
$^{\text{nat}}\text{Sb}(\alpha, xn)^{124}\text{I}$	22 \rightarrow 13	0.45	4	27	27
$^{121}\text{Sb}(\alpha, n)^{124}\text{I}^\ddagger$	22 \rightarrow 13	0.92	< 4	< 0.2	< 0.2

* values at 5 days after EOB.

‡ using 99.45 % enriched ^{121}Sb as target material.

The radionuclide ^{82}Sr ($T_{1/2} = 25.3$ d) is the parent of the positron emitting short-lived ^{82}Rb ($T_{1/2} = 1.3$ min) which is widely used in PET studies. The parent

needed for preparing a generator can be produced via several routes. In recent years the $^{85}\text{Rb}(p,4n)^{82}\text{Sr}$ reaction has been commonly used; an evaluation by the IAEA, however, showed some discrepancies in the available data. Recently, two new detailed studies on this reaction were completed. We evaluated those data and obtained a reliable excitation curve. In order to validate those data, thick RbCl targets were irradiated under well-defined conditions and the yields of the desired radionuclide ^{82}Sr as well as of the undesired impurity ^{85}Sr ($T_{1/2} = 65.0$ d) were determined. As mentioned in the last Progress Report, the experimental and theoretical (deduced from the excitation function) yields of ^{82}Sr agreed well (within 10 %), thereby validating the evaluated excitation curve for the $^{\text{nat}}\text{Rb}(p,xn)^{82}\text{Sr}$ process. In the case of ^{85}Sr , however, the data were discrepant. We therefore decided to measure cross sections for the $^{\text{nat}}\text{Rb}(p,xn)^{85}\text{Sr}$ process as well, particularly in the energy range of 25 to 50 MeV. The work presented some difficulty since the characteristic γ -ray of ^{85}Sr at 514 keV needed special care to resolve it from the 511 keV annihilation peak of the various positron emitters. Data analysis has been completed. The results are expected to lead to a well-defined excitation function for the formation of ^{85}Sr in the interactions of protons with $^{\text{nat}}\text{Rb}$, and thereby to allow an estimate of the ^{85}Sr impurity in the radionuclide ^{82}Sr .

b. Cross sections for production of the therapeutic radionuclides ^{140}Nd , ^{153}Sm , ^{169}Yb and $^{193\text{m}},^{195\text{m}}\text{Pt}$

K. Hilgers, I. Spahn, F. Tárkányi^{**}, Yu. N. Shubin^{††}, S. M. Qaim, H. H. Coenen

The radionuclide ^{140}Nd ($T_{1/2} = 3.4$ d) decays almost 100 % via EC, and several Auger electrons are emitted per decay. It is thus a potentially useful therapeutic isotope. Its production via two routes, namely $^{\text{nat}}\text{Ce}(^3\text{He},xn)^{140}\text{Nd}$ and $^{141}\text{Pr}(p,2n)^{140}\text{Nd}$, has been under investigation for several years. During the period of this Progress Report detailed data analysis of the two production reactions as well as of several competing reactions was completed. Nuclear model calculations via the code ALICE-IPPE were also performed, applying the

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^{††} IPPE, Obninsk, Russia (deceased June 2005)

code for the first time to calculate excitation functions of ^3He -particle induced reactions. In general, good agreement was obtained between the experimental and theoretical data. For production of ^{140}Nd the $^{141}\text{Pr}(p,2n)$ reaction is superior to the $^{\text{nat}}\text{Ce}(^3\text{He},xn)$ reaction. The results of this completed study have been published in *Radiochimica Acta*.

The radionuclide ^{153}Sm ($T_{1/2} = 46.3$ h) is a very important β^- emitting therapeutic isotope. It is generally produced via the $^{152}\text{Sm}(n,\gamma)^{153}\text{Sm}$ reaction in a nuclear reactor, and consequently the achievable specific radioactivity is rather low. It was therefore thought worthwhile to investigate its production at a cyclotron. A possible route could be the $^{150}\text{Nd}(\alpha,n)^{153}\text{Sm}$ reaction. Thin samples of Nd_2O_3 were prepared via the sedimentation technique, and several such samples were stacked together and irradiated with 28 MeV α -particles. The radioactivity of the products was determined via high-resolution γ -ray spectrometry. The data analysis is in progress.

The radionuclide ^{169}Yb ($T_{1/2} = 32.0$ d) is an important Auger electron emitting therapeutic isotope. It is produced in a nuclear reactor via the $^{168}\text{Yb}(n,\gamma)^{169}\text{Yb}$ reaction. With a view to increasing the specific activity of the product, and under a Research Agreement with the IAEA, we studied the excitation function of the $^{169}\text{Tm}(p,n)^{169}\text{Yb}$ reaction from its threshold up to 40 MeV. The results show that from the viewpoint of yield the cyclotron production method cannot compete with the reactor production. However, if high specific activity is desired, the cyclotron method would be advantageous.

The radionuclides $^{193\text{m}}\text{Pt}$ ($T_{1/2} = 4.3$ d) and $^{195\text{m}}\text{Pt}$ ($T_{1/2} = 4.0$ d) are very interesting, almost pure Auger electron emitting, therapeutic radionuclides. They are produced in nuclear reactors via the (n,γ) process in low yield and with very low specific activity. We studied their formation in the $^{192}\text{Os}(\alpha,n)^{195\text{m}}\text{Pt}$ and $^{192}\text{Os}(\alpha,3n)^{193\text{m}}\text{Pt}$ processes using enriched ^{192}Os as target material. Very thin samples were prepared via electrolytic deposition of ^{192}Os on Ni foils and irradiations with 28 MeV α -particles were done using a stacked-foil geometry. The radioactivity of the products was determined via high-resolution X-ray spectrometry. From the measured excitation functions the integrated yields of the

two radionuclides were calculated. The results are shown in Fig. 3. Evidently the yields are rather low but the specific activity is high. So this method of production may gain considerable importance.

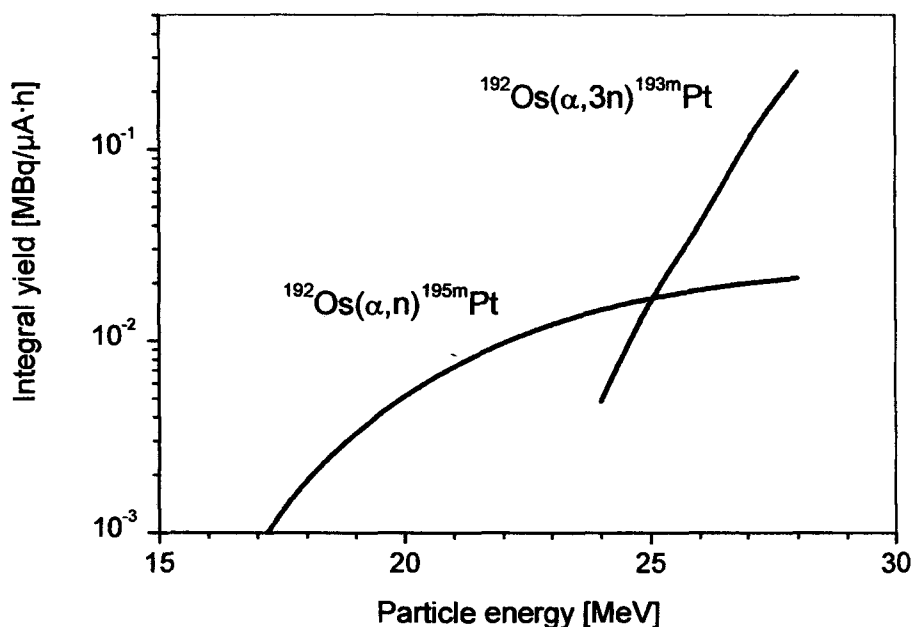


Fig. 3 Integral yields of ^{193m}Pt and ^{195m}Pt as a function of projectile energy in α -particle induced reactions on enriched ^{192}Os .

c. *Cross sections for production of the technologically important radionuclides ^{54}Mn , ^{55}Fe and ^{57}Co*

M. Al-Abyad[†], S. Spellerberg, S. M. Qaim, H. H. Coenen

The radionuclides ^{54}Mn ($T_{1/2} = 312.2$ d), ^{55}Fe ($T_{1/2} = 2.7$ a) and ^{57}Co ($T_{1/2} = 271.8$ d) are useful tracers. In order to investigate alternative methods of production of ^{54}Mn and ^{57}Co , excitation functions of the following two reactions were measured:

- $^{57}\text{Fe}(p,\alpha)^{54}\text{Mn}$ over the energy range of 10 to 30 MeV
- $^{57}\text{Fe}(p,n)^{57}\text{Co}$ over the energy range of 8 to 20 MeV

Thin samples of $^{\text{nat}}\text{Fe}$ and enriched ^{57}Fe were prepared by electrolytic deposition on Au foils. Irradiations were done at the compact cyclotron CV 28 and the

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injector of COSY. The radioactivity was determined via high resolution γ -ray spectrometry. Data analysis is in progress.

The possibility of production of ^{55}Fe via the $^{55}\text{Mn}(p,n)^{55}\text{Fe}$ reaction was also investigated. Thin samples of MnO_2 were prepared by sedimentation and irradiated at low currents at the CV 28. The assay of the 5.9 keV soft X-ray emitting ^{55}Fe was rather demanding. It was done after an elaborate radiochemical separation, using a high-resolution $\text{Si}(\text{Li})$ detector and applying correction for self-absorption of the radiation within the source. The data analysis is in progress.

4. Decay Data

S. M. Qaim, K. Hilgers, T. Bisinger, D. Nayak[♦], H. H. Coenen

The relatively long-lived positron emitters ^{64}Cu ($T_{1/2} = 12.7$ h), ^{76}Br ($T_{1/2} = 16.0$ h) and ^{124}I ($T_{1/2} = 4.18$ d) are gaining increasing significance in PET studies. Since there are some discrepancies in their positron emission intensities as well as in absolute intensities of some weak γ -rays, we investigated a few aspects of their decay properties using the following counting techniques:

- a) high-resolution HPGe detector γ -ray spectrometry (with careful analysis of the intensity of the annihilation radiation)
- b) $\gamma\gamma$ -coincidence counting (to check the accuracy of assay of the annihilation radiation)
- c) gas flow proportional beta counting (to determine β^- and β^+ decay)
- d) high-resolution $\text{Si}(\text{Li})$ detector X-ray spectrometry (to determine decay via EC).

For each investigated radionuclide it was absolutely necessary to produce it with the highest possible radionuclidic purity. ^{64}Cu was obtained via the $^{66}\text{Zn}(d,\alpha)$ -process, ^{76}Br via the $^{76}\text{Se}(p,n)$ -reaction and ^{124}I via the $^{124}\text{Te}(p,n)$ -process. In each case a highly enriched target was used. The positron branching ratios in ^{64}Cu and ^{124}I amount to 17.4 % and 22.0 %, respectively. Further analysis of the data is in progress.

[♦] Saha Institute of Nuclear Physics, Kolkata, India

5. Evaluation of Data and Integral Tests

M. Al-Abyad[†], I. Spahn, S. Sudár^{*}, M. N. H. Comsan[†], J. Csikai^{*}, B. Scholten,
S. M. Qaim, H. H. Coenen

Under a co-ordinated research project (CRP) of the IAEA on “Nuclear Data for Therapeutic Radionuclides” five (n,p) reactions, viz. $^{32}\text{S}(n,p)^{32}\text{P}$, $^{64}\text{Zn}(n,p)^{64}\text{Cu}$, $^{67}\text{Zn}(n,p)^{67}\text{Cu}$, $^{89}\text{Y}(n,p)^{89}\text{Sr}$ and $^{90}\text{Zr}(n,p)^{90}\text{Y}$, which are either commonly used, or are potentially useful, for production of the therapeutic radionuclides ^{32}P ($T_{1/2} = 14.3$ d), ^{64}Cu ($T_{1/2} = 12.7$ h), ^{67}Cu ($T_{1/2} = 61.9$ h), ^{89}Sr ($T_{1/2} = 50.5$ d) and ^{90}Y ($T_{1/2} = 64.8$ h) in a nuclear reactor, were critically evaluated. All the reaction cross section data compiled in EXFOR were taken into account and the ones showing large discrepancies were discarded. Through the concordant set of data the best curves were drawn; they were either based on the recommended curves given in the Dosimetry and/or Activation file of the IAEA, or relied on STAPRE or EMPIRE 2.19 calculation. Those curves were then adopted as standard excitation functions of the concerned reactions. A typical case is shown in Fig. 4. Due to light mass of the target nucleus (^{32}S), the nuclear model calculations are not very successful. In this particular case we adopted the IRDF-2002 curve as the standard excitation function.

For practical applications it is essential that the recommended data are also validated. We did the validation tests via measurement of integral cross sections of all the five reactions using a 14 MeV d(Be) neutron field. The integral measurements were then compared with the averaged cross sections deduced from the excitation function and the neutron spectrum under consideration. The measure and integrated cross sections agreed within about 15 %. It was therefore concluded that the recommended cross section data of all the above mentioned reactions are validated via the integral tests done in this work.

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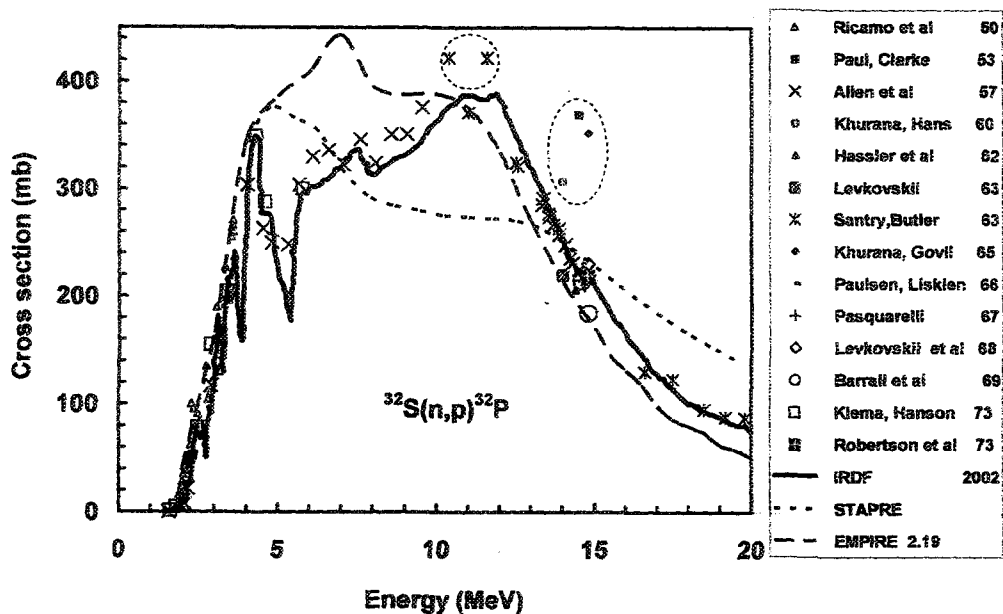


Fig. 4 Excitation function of the $^{32}\text{S}(n,p)^{32}\text{P}$ reaction. For references to experimental data cf. EXFOR. The discrepant data are encircled. The IRDF-2002 curve is recommended.

Some work on the evaluation of the reactions $^{76}\text{Se}(p,n)^{76}\text{Br}$ and $^{86}\text{Sr}(p,n)^{86}\text{Y}$ was also initiated. A preliminary screening of the data has been done. The final evaluation will be done by a theory oriented evaluator.

6. Other Activities

a) Ph. D. Theses

Two Ph. D. theses were completed during the period of the Progress Report. They were submitted to the Faculty of Natural Sciences of the University of Cologne and successfully defended. The details are:

Kerstin Kettern

Wirkungsquerschnitte zur Bildung von Aktivierungsprodukten in Wechselwirkung von Protonen mit biologisch relevanten Elementen und Kollimatormaterialien im Energiebereich bis 200 MeV, May 2004

Karsten Hilgers

Kernchemische Studien zur Entwicklung neuerer Produktionsverfahren für die therapierelevanten Radionuklide ^{140}Nd , ^{192}Ir , ^{191}Pt , $^{193\text{m}}\text{Pt}$ und $^{195\text{m}}\text{Pt}$, November 2005

b) *Chair of INDC*

Syed M. Qaim continued to act as Chairman of the International Nuclear Data Committee (INDC) of the IAEA. The term of office will cover the period up to the end of 2006.

c) *NRC-6*

The Institute organised the 6th International Conference on Nuclear and Radiochemistry (NRC-6) in Aachen, Germany, from 29 August to 03 September 2004. Nuclear data research constituted an integral part of the programme of the conference. A book of extended abstracts was published under the title: "Advances in Nuclear and Radiochemistry", S. M. Qaim and H. H. Coenen (Editors), Forschungszentrum Jülich, 2004, ISSN 1433-5565. Full texts of several of the submitted papers have also been published in a special issue of the journal *Radiochimica Acta*, Vol. **93**, No. 9 and 10, pages 497-651 (2005).

Publications (During the Period of the Progress Report)

- [1] K. Kettern, I. Spahn, S. Spellerberg, S.M. Qaim and H.H. Coenen
Nuclear reaction cross section measurements via characterisation of soft radiation emitting products
Proc. Int. Conf. Nuclear Data for Science and Technology, Santa Fe, USA, September/October 2004, (R.C. Haight et al, eds.), AIP Conference Proceedings, Vol. 769, Melville, New York, 2005, pp. 758-761
- [2] S.M. Qaim
New trends in nuclear data research for medical applications (**invited review**)
Proc. Int. Conf. Nuclear Data for Science and Technology, Santa Fe, USA, September/October 2004, (R.C. Haight et al, eds.), AIP Conference Proceedings, Vol. 769, Melville, New York, 2005, pp. 1600-1605
- [3] K. Hilgers, S.M. Qaim and H.H. Coenen
New cross section data for production of the therapeutic radionuclides ⁶⁴Cu, ¹⁴⁰Nd and ¹⁹²Ir
Proc. Int. Conf. Nuclear Data for Science and Technology, Santa Fe, USA, September/October 2004, (R.C. Haight et al, eds.), AIP Conference Proceedings, Vol. 769, Melville, New York, 2005, pp. 1631-1633
- [4] Ye. Skakun and S.M. Qaim
Excitation functions of helion induced nuclear reactions for the production of

the medical radioisotope ^{103}Pd

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INSTITUT FÜR KERN- UND TEILCHENPHYSIK TECHNISCHE UNIVERSITÄT DRESDEN

1. Validation Experiment of γ -Activities of Pb irradiated in Fusion Peak Neutron Field*

R. Eichin, R.A. Forrest¹, H. Freiesleben, K. Seidel, S. Unholzer

Lead is a major element of breeding blanket concepts for the DEMO reactor and for ITER Test Blanket Modules. The Helium Cooled Lithium Lead and the Water Cooled Lithium Lead blanket concepts are based on molten Pb-17Li alloys that act as coolant, breeding material and neutron multiplier [1]. The short-term radioactivity induced in the material by neutrons is of interest for the heat production and the shut-down dose rate, whereas the long-term radioactivity is interesting for waste management. It is expected from calculations that Pb-17Li used for the operation of a power plant can be recycled and used in the next reactor after about 50 years of cooling [2]. The aim of the present work is to contribute experimentally to the validation of the activation predictions based on calculations with codes and data libraries such as the European Activation System EASY [3]. The activation of Li with fusion peak neutrons was previously investigated [4]. Therefore, pure Pb was irradiated in the present work.

In a calculation with EASY, lead was assumed to be irradiated under power plant conditions; this means with a flux of the 14 MeV neutrons corresponding to a power density of 1.0 MW/m², for a period of one year. The results obtained for the contact dose rate as a function of the decay time after irradiation, are shown in Fig.1. The nuclides dominant in Pb in the first month after the irradiation are ^{204m}Pb and ²⁰³Pb. After that, up to the recycling-limit, reached after about 8 months, ²⁰²Tl and ²⁰³Hg are the dominant nuclides. The hands-on limit reached after about 40 years is determined by ²⁰⁷Bi. Considering the different half-lives, two irradiations were carried out: a short one with only 32 min of irradiation time followed by γ -ray acquisition in the time range between $2 \cdot 10^{-6}$ and 10^{-4} years (t_2 in Fig.1, right hand) and a long one of 8.8 hours irradiation time with spectral measurements in the range $2 \cdot 10^{-4} - 0.2$ years (t_1 in Fig. 1, right hand).

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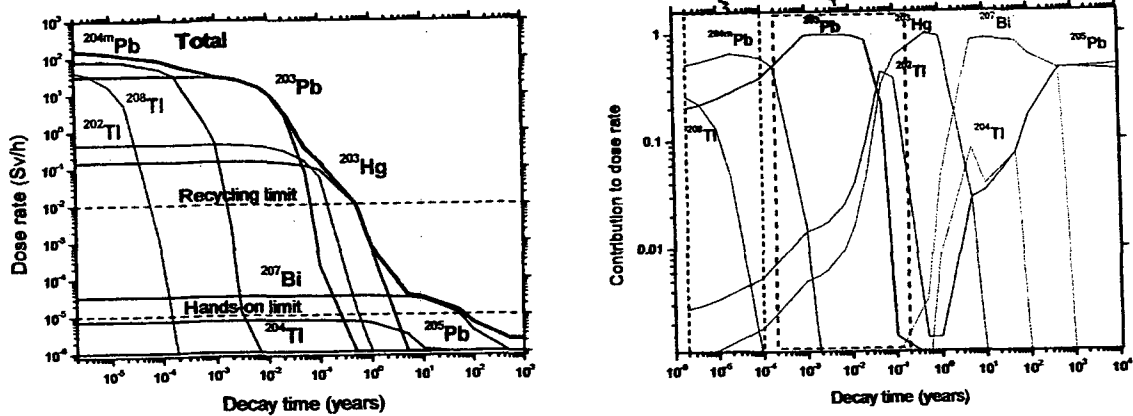


Fig. 1 Calculated contact dose rate (left hand) and contribution of the different radionuclides to the total dose rate (right hand) after irradiation of Pb with fusion peak neutrons of 1.0 MW/m^2 power density for one year as a function of the decay time.

The irradiation of the Pb samples (mass 5.553 g and 5.535 g) was performed at a D-T neutron generator of TUD. The neutron fluences applied were measured by simultaneous activation of niobium foils and evaluating the activity induced by the $^{93}\text{Nb}(n,2n)^{92m}\text{Nb}$ reaction. The attenuation of the neutron and γ -ray fluxes in the sample as well as the geometry relations source-sample and sample-detector were determined by 3D Monte Carlo calculations. γ -ray spectra were taken with an HPGe-spectrometer. The γ -activities identified by energy and half-life were used to determine the nuclide activities using γ -yield data from EASY.

The measured activities were analysed with the version EASY-2003. The results are presented in Table 1. The uncertainties of the calculated activities ($\Delta C/C$) include both cross section and half-life errors, as estimated by EASY. The uncertainties of the experimental values ($\Delta E/E$) take into account possible errors of the γ -activity measurements (statistical uncertainty of the γ -counting, the uncertainty of the efficiency calibration of the spectrometer including the geometry factor), sample mass, γ -yield data and of the neutron flux monitoring.

The activities given in Table 1 are produced each by one dominant reaction. For those reactions the EASY-2003 data used for the analyses of the measured activities are compared with differential cross section data measured during the last decade and with evaluated cross sections from the libraries ENDF/B-6.8, JENDL-3.3 and JEFF-3.0. They are discussed in detail with respect to the C/E of the present work [5].

Table 1 Results obtained for the activity of radionuclides; radionuclides identified, their half-lives and the γ -rays (energy and yield) used to determine the activity, the neutron reactions producing these nuclides, the ratios of calculated-to-experimental activity (C/E), and the uncertainties of both calculated ($\Delta C/C$) and experimental activity ($\Delta E/E$).

Nuclide	Half-life	E_γ (keV)	Y_γ (%)	Reaction Contribution (%)	C/E	$\Delta C/C$ (%)	$\Delta E/E$ (%)
^{203}Pb	2.17 d	279 401	81 3.4	$^{204}\text{Pb}(n,2n)$ 40 $^{204}\text{Pb}(n,2n)IT$ 60	0.97	18.5	8.6
^{204m}Pb	1.12 h	375 899	89 99	$^{204}\text{Pb}(n,n')$ 100	0.92	20.0	9.1
^{208}Tl	3.05 min	583	84.5	$^{208}\text{Pb}(n,p)$ 100	0.84	20.0	10.6
^{203}Hg	46.6 d	279	81	$^{206}\text{Pb}(n,\alpha)$ 93.1 $^{207}\text{Pb}(n,n'\alpha)$ 6.9	1.04	23.1	10.8

All the activities which are dominant up to the recycling limit of Pb after irradiation in a fusion peak neutron field were experimentally investigated. The measured activities agree with the corresponding calculated values for ^{203}Pb , ^{204m}Pb and ^{203}Hg within 8%. Hence the activation performance of Pb calculated with EASY for reactor conditions is validated on this level up to the recycling limit. Only the activity of ^{208}Tl which is of minor importance showed a deviation of the calculated from the measured value by 16%. The experimental uncertainties of the activities are smaller than the uncertainties of the EASY calculations. Hence, the measurements may contribute to a further reduction of the EASY uncertainties [6].

The radionuclide that determines the hands-on limit is ^{207}Bi . It is produced by the reaction $^{207}\text{Pb}(p,n)$ in a second step with protons from previous (n,p) reactions. This secondary charged particle reaction should further be investigated.

If Pb-17Li is irradiated at fusion power plant fluxes, a contact dose rate of about $8 \cdot 10^{-4}$ Sv/h is predicted to be produced by the sequential charged particle reaction $^7\text{Li}(p,n)^7\text{Be}$. In a previous irradiation of Li_4SiO_4 [4] a C/E = 0.9 was found for the ^7Be activity. Fortunately, the half-life of ^7Be (53 days) is sufficiently short not to significantly influence the total dose rate of Pb-17Li.

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2. Measurement and Analysis of Radioactivity induced in Ta by D-T Neutrons*

R. Eichin, R. A. Forrest¹, H. Freiesleben, K. Seidel, S. Unholzer

The activation induced by neutrons in the materials of fusion power plant projects represents a central safety related topic. Reduced activation structural materials, such as EUROFER, with acceptable radiological safety performance and low long-term radiation level have been developed for first wall and breeding blankets [1]. EUROFER was already previously irradiated with 14 MeV neutrons, and the activation originating from reactions on the main constituents have been investigated [2]. Contributions from Ta, that is an alloying constituent of EUROFER, could not be observed. Therefore, samples of pure Ta were irradiated with D-T neutrons in the present work, and the measured γ -activities were analysed with the European Activation System EASY [3].

In order to investigate the major activities up to the recycling limit of Ta after use under fusion power plant conditions, two irradiations were performed. A short irradiation with γ -spectra taken at decay times in the range of minutes up to two hours and a longer irradiation with spectra taken at decay times from 4 hours up to 2 days. The irradiation of the samples was performed at a neutron generator of TUD. The fluences applied were monitored by the $^{93}\text{Nb}(n,2n)^{92m}\text{Nb}$ activation.

The results obtained are summarized in Table 1. Each activity is produced by one dominant reaction. For these reactions the cross section data measured in the last decade are compared with the EASY data and with evaluated data from the libraries ENDF/B-6.8, JENDL-3.3 and JEFF-3.0 and are discussed in detail with respect to the C/E values of the present work in Ref. [4].

* Work supported by the European Fusion Development Agreement (Task TTMN-002)

¹ EURATOM/UKAEA Fusion Association, Culham Science Centre, Abingdon, United Kingdom

Table 1 Results of irradiation and analysis; radionuclides identified, their half-lives and γ -rays with yield data used to determine the activity, the neutron reactions producing the nuclide, the ratio of calculated-to-experimental activity (C/E) and the uncertainty of both calculated and experimental activity.

Nuclide	Half-life	E_γ (keV)	Y_γ (%)	Reaction Contribution (%)	C/E	$\Delta C/C$ (%)	$\Delta E/E$ (%)
^{178m}Lu	23.1 min	325.5 426.4	94.1 96.9	$^{181}\text{Ta}(n,\alpha)$ 100	0.92	20.0	10.3
^{180m}Hf	5.5 h	332.3 443.1	94.6 82.1	$^{181}\text{Ta}(n,d)$ 99.9	0.98	119.7	11.8
^{181}Hf	42.4 d	133 482	41.9 83.0	$^{181}\text{Ta}(n,p)$ 99.9	1.01	20.0	13.1
^{180}Ta	8.08 h	103.6	0.78	$^{181}\text{Ta}(n,2n)$ 100	1.17	15.0	25.7
^{182}Ta	114.7 d	1112.3 1189 1221.4	0.349 0.164 0.273	$^{181}\text{Ta}(n,\gamma)$ 93.7 $^{181}\text{Ta}(n,\gamma)$ IT 6.3	1.18	43.6	10.0

It has been concluded that for the dominant activities in the decay time range up to the recycling limit of Ta irradiated with D-T neutrons under fusion power plant conditions, the EASY-2003 data have a good quality. The recycling can be expected to be reached at decay times smaller than 10 years. As the experimental uncertainties ($\Delta E/E$ in Table 1) are for three of the activities smaller than the $\Delta C/C$, the measurements can contribute further to improve the EASY data base [5].

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**ZENTRUM FÜR STRAHLENSCHUTZ UND RADIOÖKOLOGIE,
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**Production of Residual Nuclides from Tungsten by Proton-Induced Reactions
up to 70 MeV**

R. Michel¹, A.S.M.F. Chowdhury¹, Z. Ullah¹, U. Herpers², U. Otto², E. Bolz²

Continuing our systematic investigations on the proton-induced production of residual nuclides from heavy target elements [1-4] irradiation experiments were performed at the injector-2 cyclotron of the Paul Scherrer Institute, Villigen, Switzerland. Integral cross sections for the production of residual nuclides were determined using the stacked-foil technique in irradiation experiments with classical kinematics and off-line gamma-ray spectrometry. Stacks consisting of tungsten foils of natural isotopic composition, separated by high purity copper, and aluminium foils for flux monitoring were irradiated for three to four hours by 71 MeV and 45 MeV protons at beam currents of about 30 nA.

The target materials were supplied by Goodfellow Metals Ltd., U.K., in the form of a rectangular foils of high purity (99.99%) in order to avoid additional production of residuals from the impurities. The stacks were designed to provide overlapping energy regions for the high- and low-energy irradiations, to avoid any recoil contamination or recoil loss of the produced nuclides by covering each measured foil with thin catcher-foils of the same element, and to allow for checking the accuracy of the calculated proton energies inside the stack by comparing the cross sections obtained from the aluminium and copper foils with previously measured values [5], and to detect possible interference caused by secondary particles. The energy degradation of the protons in the stack was calculated by a computer program based on the work of Andersen and Ziegler [6, 7].

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After irradiation the samples were transported to Cologne and Hanover where repeated gamma-ray spectroscopic measurements were done using Ge(Li)- and Ge-detectors at different geometries in order to minimize systematic influences. The spectra were evaluated with the code Gamma-W [8] using unfolding techniques. Detector efficiencies were determined with calibrated radionuclide sources. Half-lives, gamma-energies, and -intensities were taken from [9]. The proton flux densities were determined via standard monitor reaction $^{65}\text{Cu}(p,n)^{65}\text{Zn}$ and, for the 72 MeV irradiation experiments, additionally via the $^{27}\text{Al}(p,3p3n)^{22}\text{Na}$ reaction. For calculating the flux densities, the same cross sections as earlier [5] were considered.

In Figs. 1 and 2, we report cross sections for the production of ^{181}Re , ^{182}Re , ^{183}Re , ^{184}Re , and ^{177}Ta for energies between 10 and 70 MeV. The new experimental data are compared with earlier work [2, 4] and with model calculations using the code TALYS [10].

For ^{182}Re , ^{183}Re , and ^{184}Re , the agreement between the new data and the results of previous measurements is good in the overlapping energy regions. In the case of ^{181}Re , there is a slight discrepancy between the new data and the results obtained earlier [4]. Together with data for higher energies [2], we now have complete excitation functions up to 2.6 GeV for these four product nuclides. In the case of ^{177}Ta , there were no previously available data.

A comparison of the theoretical excitation functions with the experimental cross sections demonstrates good performance of the TALYS code for the $^{\text{nat}}\text{W}(p,xn)^{183}\text{Re}$ and $^{\text{nat}}\text{W}(p,xn)^{184}\text{Re}$ reactions, while the cross sections for the $^{\text{nat}}\text{W}(p,xn)^{181}\text{Re}$ and $^{\text{nat}}\text{W}(p,xn)^{182}\text{Re}$ reactions are under- and over-estimated, respectively, by the theory in the evaporation peaks. For the reaction $^{\text{nat}}\text{W}(p,2pxn)^{177}\text{Ta}$ the theory fails to describe the local maxima attributable to reaction channels involving emission of alpha-particles. There is still some necessity for further improvement of the models underlying the code.

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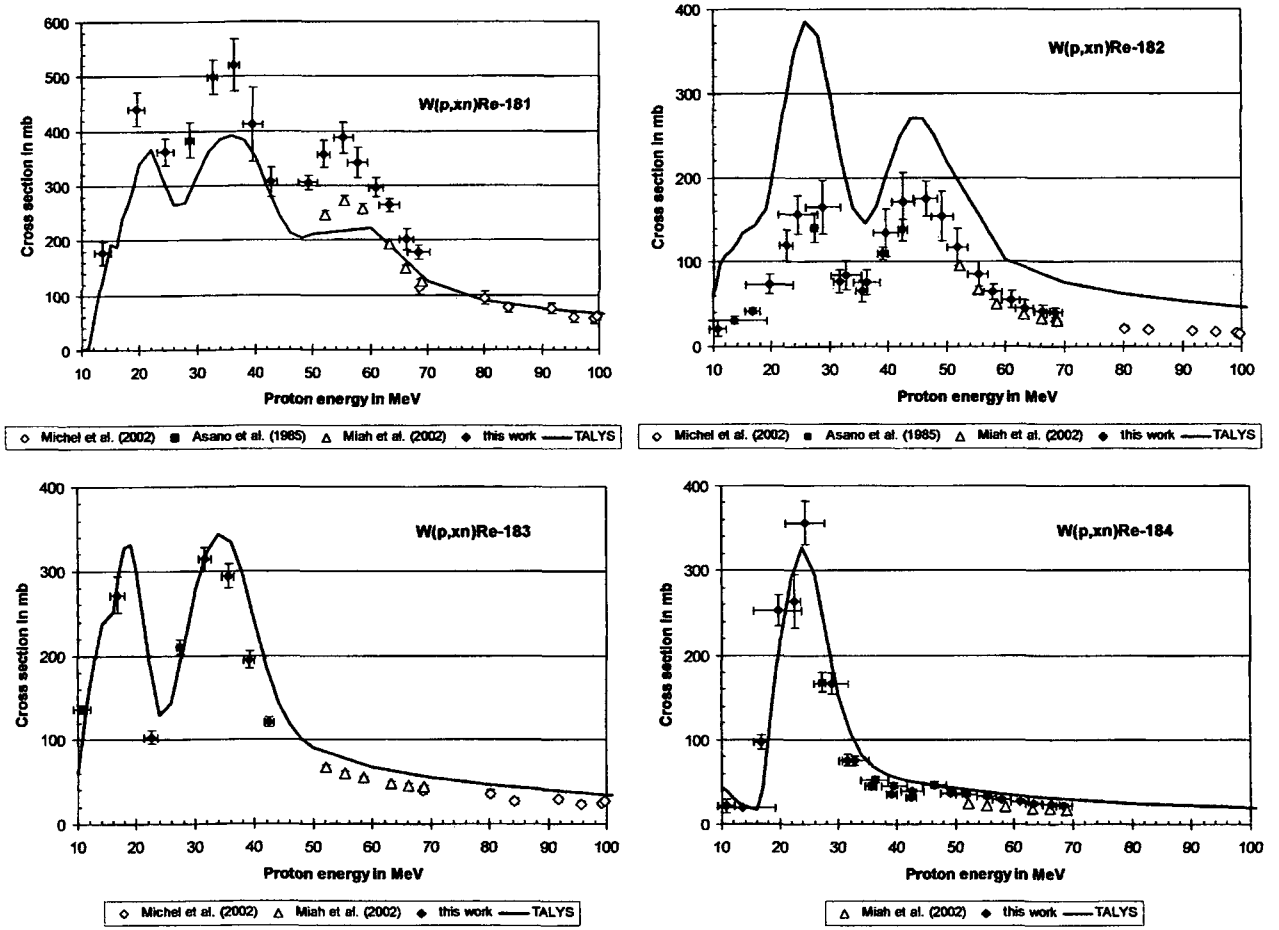


Fig. 1 Comparison of the experimentally obtained data for the reactions $^{nat}\text{W}(p,xn)^{181}\text{Re}$, $^{nat}\text{W}(p,xn)^{182}\text{Re}$, $^{nat}\text{W}(p,xn)^{183}\text{Re}$, and $^{nat}\text{W}(p,xn)^{184}\text{Re}$ with the results from previous work [2, 4] and with model calculations using the TALYS code [10].

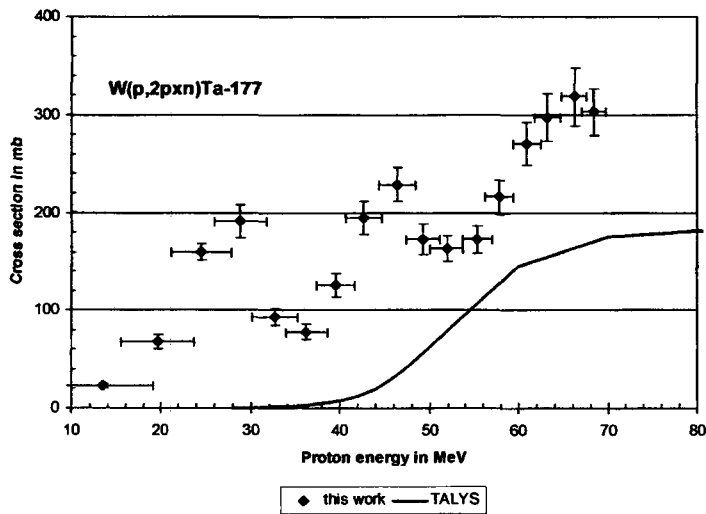


Fig. 2 Comparison of the experimentally obtained data for the reaction $^{nat}\text{W}(p,2pxn)^{177}\text{Ta}$ with model calculations using the TALYS code [10].

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**PHYSIKALISCH-TECHNISCHE BUNDESANSTALT
BRAUNSCHWEIG**

1. Overview of the Programme of Neutron Scattering Cross Section Measurements performed at PTB

D. Schmidt, W. Mannhart

For neutron energies between 7 MeV and 14 MeV, no monoenergetic neutron sources are available in practice. Therefore, there is a lack of neutron scattering data in this "energy gap". In order to overcome this deficiency, a long-term program has been started at PTB. Simultaneous to the installation of a multi-angle time-of-flight (TOF) spectrometer, principles and methods were elaborated to determine neutron scattering cross sections with the highest precision presently possible [1].

During measurement, careful and extended calibration and monitoring procedures are applied. The data analysis is based on a complete Monte Carlo simulation of all relevant processes, such as the description of the detector properties, including the detection efficiencies, the simulation of the neutron production in the gas target and the simulation of the scattering experiment. For that reason, further corrections of the cross sections determined are not necessary. In addition, a correct angle scaling is obtained in the course of the analysis [1-3]. In all scattering experiments, natural samples have been applied. The sample materials were selected with the focus on fusion neutronics and are summarized in Table 1. The advanced Monte Carlo technique allows mixed samples with different elements to be handled correctly, as was necessary for nitrogen and oxygen.

Table 1 Elements and incident neutron energy ranges in scattering cross section measurements at PTB in the last decades.

sample	incident energy		EXFOR entry		Reports	
	range (in MeV)	points	DX	DDX	DX	DDX
¹² C	13.33 - 15.82	4	22404	---		
¹⁴ N	7.89 - 13.85	7	22806	---	PTB-N-44	
¹⁶ O	6.42 - 14.89	11	22113	---	PTB-N-1	
^{nat} Si	7.89 - 13.85	10	22666	---	PTB-N-43	
^{nat} Ti	7.93 - 14.72	11	+	+		
⁵¹ V	7.99 - 14.37	11	22409	22412	PTB-N-36	PTB-N-46
^{nat} Cr	7.95 - 14.76	10	22408	22411	PTB-N-31	
^{nat} Fe	9.41 - 15.20	12	22403	---	PTB-N-20	
^{nat} Cu	6.95 - 14.18	8	+	+		
^{nat} Nb	(7 - 14)					
^{nat} W	7.19 - 14.10	8	+	+		
^{nat} Pb	7.93 - 14.23	7	22407	22410	PTB-N-27	PTB-N-38

+ data available, EXFOR entry in progress

The scattering experiment is based on TOF technique and simulation of the measured neutron TOF spectra. Elastic scattering and inelastic scattering with excitation of well-resolved low-lying states lead to pronounced peaks in the TOF spectra which are treated as partial cross sections (DX). The residual part of the TOF spectra is then analyzed as double-differential cross sections (DDX). But our neutron source - applying the $D(d,n)^3\text{He}$ reaction with a gas target, - is quasi-monoenergetic only. Thus, the low-energy part of the TOF spectra is contaminated with scattered parasitic neutrons stemming from the $D(d,np)D$ breakup reaction. This scattered breakup fraction will also be simulated and subtracted. As the differential neutron data taken from any evaluation (in most cases ENDF/B-VI) are often unrealistic, they have to be experimentally improved for the energy range of breakup neutrons, i.e. below about 8 MeV. Such improvement is realized applying an additional pure breakup source using the $^4\text{He}(d,np)^4\text{He}$ reaction. Details of that procedure are described elsewhere [4].

Fig. 1 shows as an example the elastic angle-integrated cross sections of some elements. The "energy gap", i.e. the lack of data between 10 MeV and 14 MeV, is clearly demonstrated. Furthermore, the ENDF/B-VI data sometimes show remarkable deviations from our precisely measured data. Thus, our data measurements are also a good basis for re-evaluations, which are recommended in some cases.

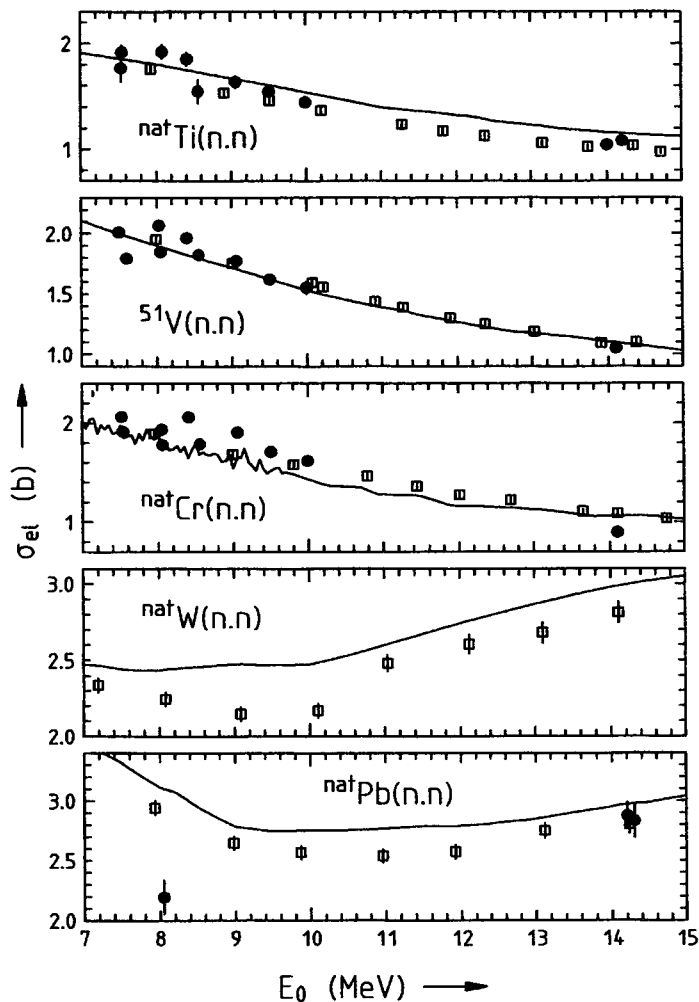


Fig. 1 Elastic angle-integrated cross sections of some elements; squares: data measured at PTB; full circles: data taken from the EXFOR database; curves: data taken from the ENDF/B-VI evaluation.

2. Neutron Scattering Cross Sections of Natural Tungsten at Energies between 7 and 14 MeV

D. Schmidt, W. Mannhart

Neutron scattering cross sections are determined at PTB using the time-of-flight (TOF) technique. In a first step, partial cross sections (DX) are obtained, belonging to low-lying states which are well resolved in the frame of the experimental energy resolution. The DX include in any case the elastic scattering which contributes with about 50% to the neutron interaction in the energy region between 7 and 14 MeV. In a second step, the remaining part of the neutron TOF spectra is treated as double-differential cross sections (DDX). The DDX include the cross sections of unresolved, overlapping levels as well as the physical continuum due to open particle channels.

Natural tungsten is a mixture of four stable isotopes (^{180}W with an abundance of 0.13% is negligible) which altogether have 13 low-lying levels with excitation energies below 400 keV, which is about the experimental energy resolution. Thus, in measurements at energies between 7 and 14 MeV, it is impossible to separate the elastic from the inelastic scattering. The elastic cross sections determined comprise therefore contributions from a number of low-lying levels. The technique of data reduction and cross section determination is described in detail elsewhere [1,4]. Except for the elastic scattering (including some inelastic components), the remaining neutron emission spectra are treated as DDX. It means that the determined cross sections completely describe the neutron emission for emission energies above the experimental threshold of 1.5 MeV.

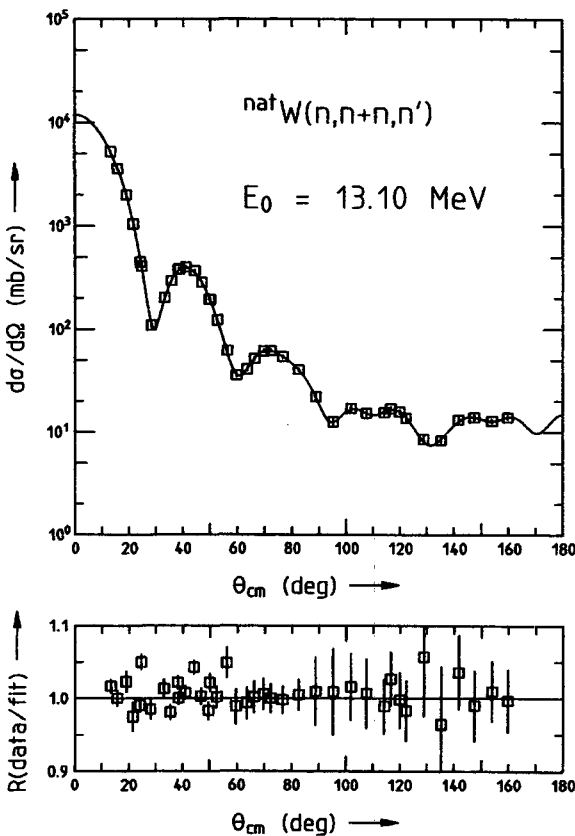


Fig. 2

Upper part:
Elastic angular distribution at incident neutron energy $E_0 = 13.10 \text{ MeV}$, including low-lying inelastic components.
Squares: measured cross sections; curve: fitted Legendre polynomial expansion.

Lower part:
Ratio of data to fitted Legendre polynomial expansion; the uncertainties given are the statistical standard uncertainties.

Cross sections were determined at eight incident neutron energies between 7.19 MeV and 14.10 MeV. Fig. 2 shows one of the elastic angular distributions measured. The quality of the data (uncertainties below about 5%) is also demonstrated. The maximum order of the fitted Legendre polynomial expansion is with $\ell_{\max} = 23$ rather large but necessary for the proper description of such deformed heavy nuclei.

DDX had been determined by subtracting the elastic scattering fraction from the neutron TOF spectra. For higher incident neutron energies, the measured neutron TOF spectra are contaminated by scattered parasitic neutrons stemming from the deuteron breakup continuum of the quasi-monoenergetic DD source. These parasitic fractions are simulated separately and then subtracted. The underlying data have to be improved using a second neutron source (^4He filling in the gas target instead of deuterium). This procedure is described in detail elsewhere [4]. The amplitude of the scattered breakup fraction depends on the incident neutron energy and neutron emission energy, respectively, and is illustrated in Fig. 3.

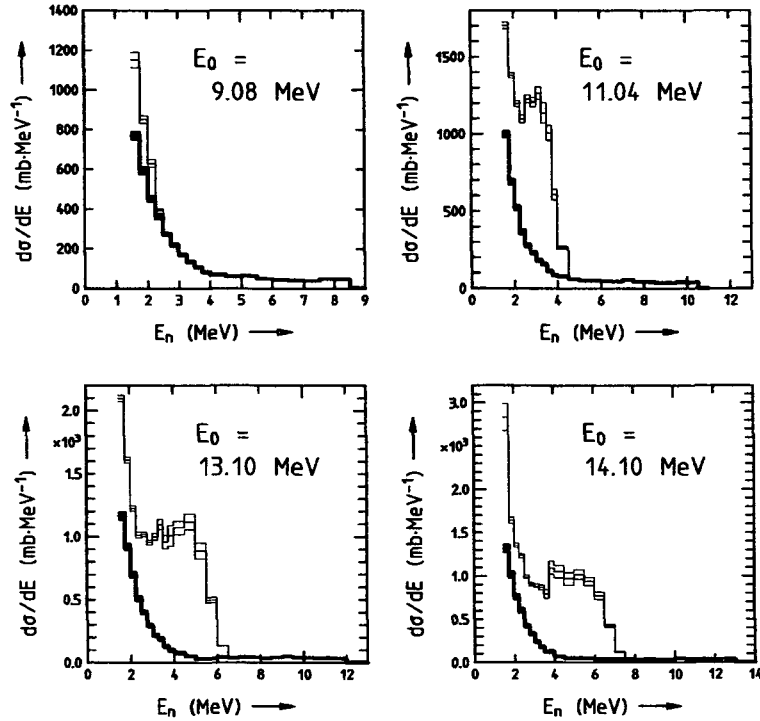


Fig. 3 Angle-integrated DDX (emission spectra) at selected incident neutron energies E_0 (thick histograms); the thin histograms represent pseudo-data without corrections of the parasitic breakup neutrons.

Although partly the corrections are rather large, the subtraction of the breakup fraction leads to reliable DDX data because the data for simulating the breakup fraction are improved experimentally. Fig. 4 shows our emission spectrum at $E_0 = 14.10$ MeV, compared with the results using a pure monoenergetic neutron source (14 MeV neutron generator). The differences between the two data sets are in the order of 20%, the integrals in the overlapping region (1.5 MeV - 6.0 MeV) differ by 7.4%. This comparison demonstrates the reliability of our correction procedure.

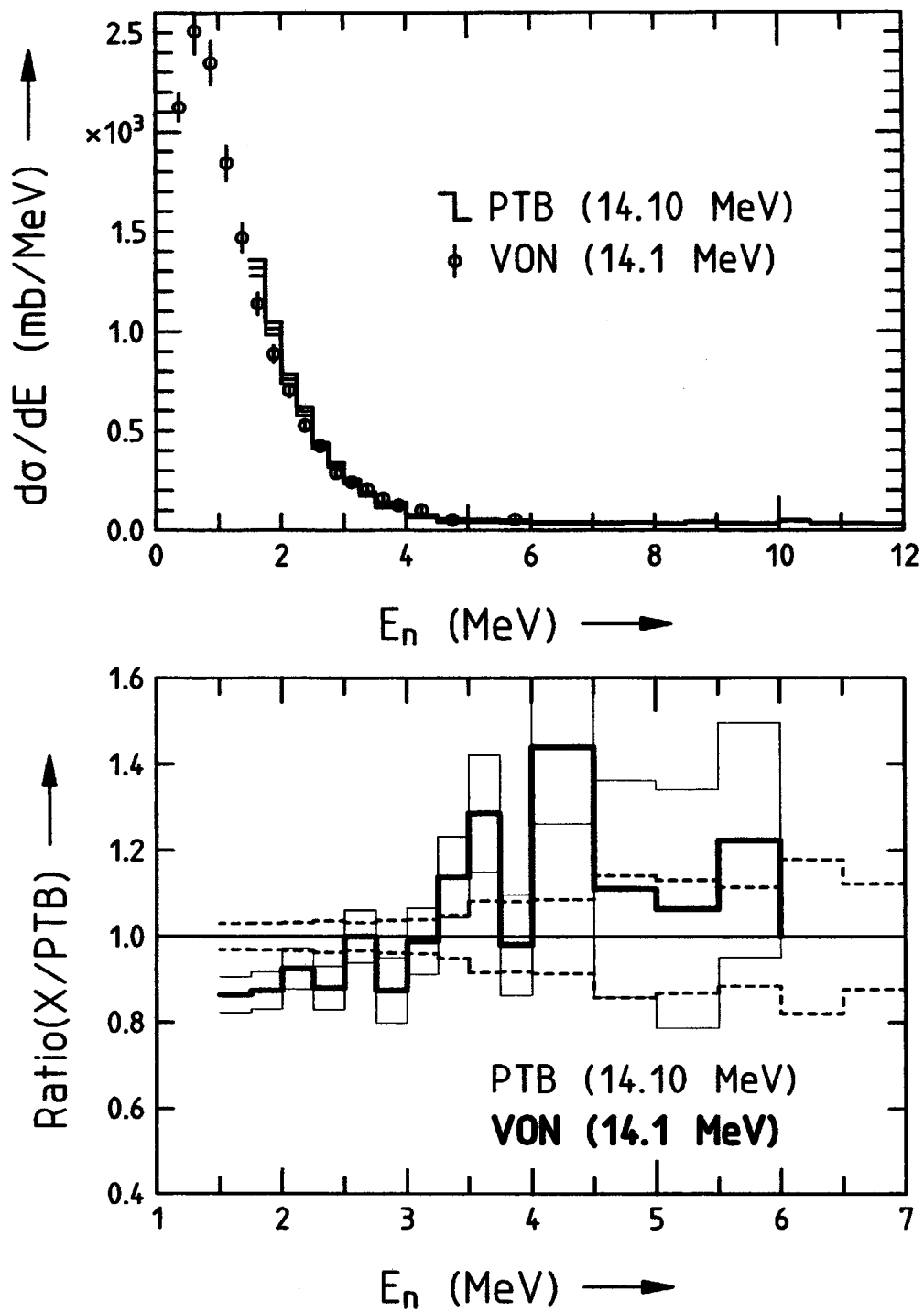


Fig. 4 Upper part: Neutron emission spectrum at $E_0 = 14.10$ MeV of the present work (histogram "PTB") and taken from Vonach et al. "VON" [5]; for better visibility, the group data "VON" are plotted like point data. Lower part: Ratio of the rebinned data "VON" to "PTB" (thick line: ratio, thin lines: uncertainties); the dashed lines represent the uncertainties of our data.

3. The $^{70}\text{Zn}(n,2n)^{69}\text{Zn}^m$ Cross Section between Threshold and 14 MeV

W. Mannhart, D. Schmidt

With a D(d,n) neutron source and elemental samples of zinc, the $^{70}\text{Zn}(n,2n)^{69}\text{Zn}^m$ cross section has been determined at 14 energies. The energy scale of the present work is uncertain within 20 keV and the energy resolution (FWHM) is in the order of 100 keV. The use of elemental samples required corrections for the competitive reaction $^{68}\text{Zn}(n,\gamma)^{69}\text{Zn}^m$, which is enhanced by a factor of 30 with the isotopic abundance of 18.75(51)% for ^{68}Zn and of 0.62(3)% for ^{70}Zn . The $^{68}\text{Zn}(n,\gamma)$ portion has been determined experimentally with measurements at 8.50, 8.97 and 9.53 MeV, i.e. below the threshold energy of $^{70}\text{Zn}(n,2n)$ of 9.79 MeV. After a careful correction for low energy neutrons generated in the wall and backing of the gas cell, performed with gas-out measurements, the remaining neutron field consists only of monoenergetic neutrons and a continuum of neutrons from the D(d,np) breakup component. The $^{70}\text{Zn}(n,2n)$ reaction is insensitive to the breakup neutrons, but the $^{68}\text{Zn}(n,\gamma)$ reaction with a strongly increasing cross section to low energies requires energy-dependent breakup corrections. These corrections were calculated based on the $^{68}\text{Zn}(n,\gamma)$ cross section taken from the ADL-3 [6] library and compared with our measurements. An examination of the original ADL-3 data of $^{68}\text{Zn}(n,\gamma)^{69}\text{Zn}^m$ with experimental data in EXFOR [7] available at 24 keV, 1 MeV and 2 MeV indicated a necessary scaling of the ADL-3 data by a constant factor of 3. With the re-scaled ADL-3 data and the calculated breakup corrections based on these data, a perfect agreement with our measurements of the $^{68}\text{Zn}(n,\gamma)$ component was obtained below 9.8 MeV neutron energy. This made it possible to calculate and subtract the $^{68}\text{Zn}(n,\gamma)$ contribution from the measured data above the threshold of the $^{70}\text{Zn}(n,2n)$ reaction. The correction varies between 20 mb at 9 MeV and 77 mb at 14 MeV. The increase is dominated by a strongly increasing breakup correction at higher neutron energies.

Our data of the $^{70}\text{Zn}(n,2n)^{69}\text{Zn}^m$ cross section and the correction function of the $^{68}\text{Zn}(n,\gamma)^{69}\text{Zn}^m$ component in a D(d,n) neutron field are given in Fig. 5. The radioactivity of the 13.76(2) h decay of $^{69}\text{Zn}^m$ was measured via the 438.6 keV line with $h\nu = 0.9477(20)$. The neutron fluence was measured with a fission chamber and is based on the $^{238}\text{U}(n,f)$ cross section taken from the ENDF/B-VI. The uncertainty comprises counting statistics, efficiency calibration, decay data, monitor cross section and all relevant corrections.

In Fig. 5 our data are compared with the ADL-3 library [6] and other experimental data taken from the EXFOR database [7]. If possible, the EXFOR data were updated to advanced monitor cross sections and decay parameters to obtain a mostly consistent set of data. On the average, our data exceed the ADL-3 evaluation over the whole energy range by 19% and confirm the shape of the evaluation. It is obvious that the data point labeled “Karolyi 68” was used to normalize the theoretical ADL-3 evaluation. Most of the experimental data were determined with elemental samples. Only the experiments “Nesaraja 03” [8] and “Hlavac 76” used enriched isotopic samples.

The D(d,n) neutron source was used in our own experiment and in the measurements “Nesaraja 03” and “Santry 72” [9]. In Ref. [9] elemental samples were used and the necessary correction of the $^{68}\text{Zn}(n,\gamma)$ component is quoted as 68 mb, based on a measured $^{68}\text{Zn}(n,\gamma)^{69}\text{Zn}^m$ cross section value of 2.3(2) mb at 1 MeV. Further details are not given. Even with the correction applied, the cross section data of “Santry 72” up to 12.6 MeV are relatively high and the existence of non-zero values below the reaction threshold is confusing.

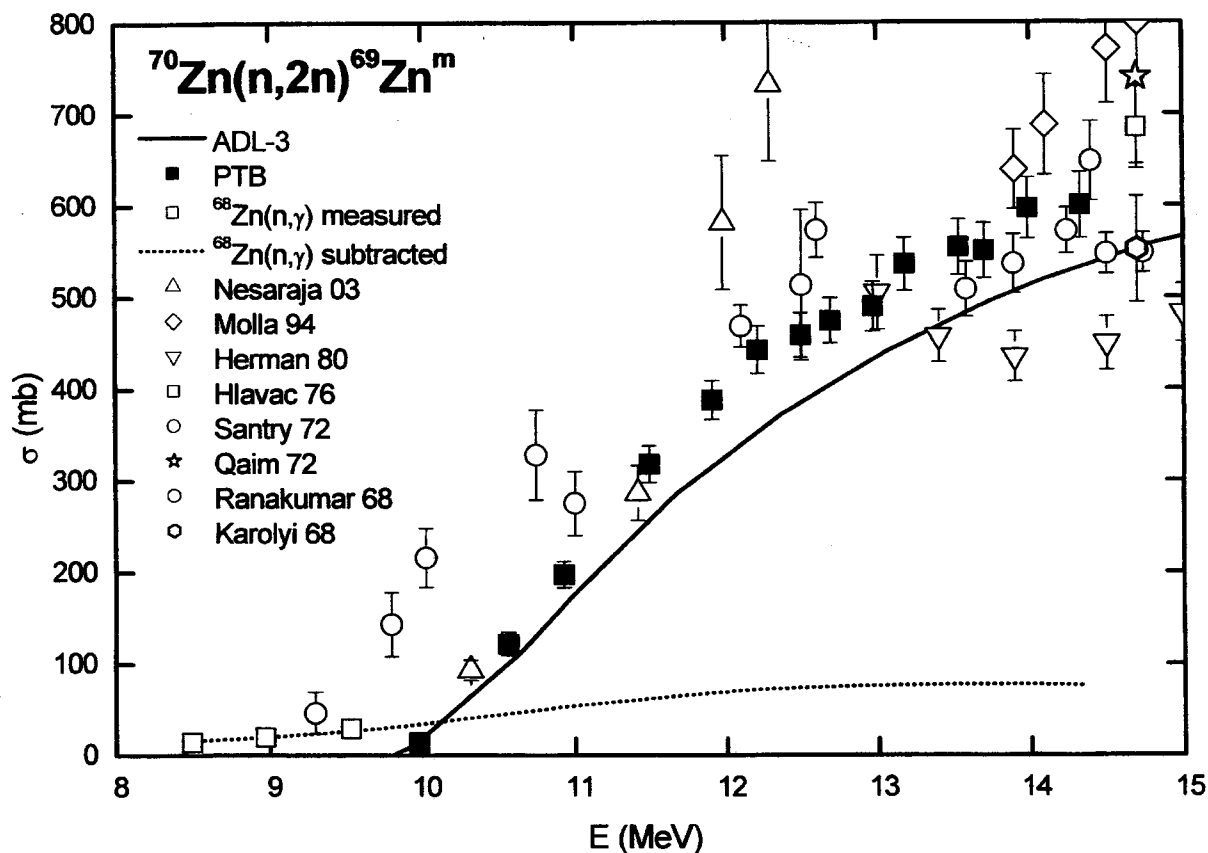


Fig. 5 Experimental database of the $^{70}\text{Zn}(n,2n)^{69}\text{Zn}^m$ cross section.

The data set “Santry 72” comprises additional measurements performed with the T(d,n) source at 12.5 MeV and at a series of energies above 13.58 MeV. At 13.58 MeV, measurements were done with both the T(d,n) and the D(d,n) source. The data obtained with the T(d,n) source show a clear jump and are about 100 mb lower than those determined with the D(d,n) source. The value quoted at 13.58 MeV seems to be based on the T(d,n) measurement. The experiment of Ref. [8] avoids the $^{68}\text{Zn}(n,\gamma)$ correction. But the data above 12 MeV are quite surprising. Even the quoted energy spread of the data of ± 0.6 MeV is not sufficient to explain the extremely high cross values. Above 11.4 MeV, the data indicate a change in the slope of the excitation function from 180 mb/MeV to 500 mb/MeV, which is very unlikely for an (n,2n) reaction.

The remaining data shown in Fig. 5 were determined with the 14 MeV neutron source of T(d,n). The (n, γ) correction is here in the order of 15 mb. The experiment “Herman 80” suggests a decrease in the (n,2n) cross section to higher energies which is not confirmed by other data. The data labeled “Ranakumar 68” and “Hlavac 76” describe fairly well the trend of the $^{70}\text{Zn}(n,2n)^{69}\text{Zn}^m$ cross section in the 14 MeV region.

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