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

for the Period April 1, 1999 to March 31, 2000

July 2000

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

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

# 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, Köln as well as from the PTB Braunschweig. The emphasis in the work reported here is on measurement, calculation, compilation and evaluation of nuclear data for applied science programmes, such as those relevant to reactor technology, transmutation concepts, accelerator shielding and development, astrophysics research, cosmogenic and meteoritic investigations, radiation therapy, production of medically important radioisotopes, 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 should therefore also serve as a background information to some areas of work of those committees.

Each contribution is presented under the laboratory heading from where the work is reported. The names of other participating laboratories are also mentioned. When the work is relevant to the World Request List for Nuclear Data, WRENDA 93/94 (INDC(SEC)-104/U+G), the corresponding identification numbers are given.

Jülich, July 2000 S.M. Qaim

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**APPENDIX**

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## 1. Neutron Capture of $^{26}$Mg at kT = 52 keV and the Resonance at $E_n$ = 68.7 keV*

P. Mohr[1], H. Beer, H. Oberhummer[2], W. Rochow[3], P.V. Sedyshev[4], S. Volz[1], A. Zilges[1]

The neutron capture cross section of $^{26}$Mg was measured relative to the known gold cross section at $kT = 52$ keV using the fast cyclic activation technique. The experiment was performed at the Van de Graaff accelerator, Universität Tübingen. From the new experimental result we deduce the resonance strength for the resonance at $E_{\rm n} = 68.7$ keV which is in good agreement with a direct measurement by Weigmann *et al.* Finally, we calculate the Maxwellian averaged capture cross section $< \sigma >$ in the astrophysically relevant energy region from the available experimental data, and discuss the discrepancies of $< \sigma >$ between our work and a previous calculation by Weigmann *et al.*

* Phys. Rev. C **60** (1999) 17603/1-4.

## 2. The Stellar Neutron Capture Rate of $^{34}$S - The Origin of $^{36}$S Challenged*

R. Reifarth, K. Schwarz, F. Käppeler

The (n,$\gamma$) cross section of $^{34}$S has been measured by means of the activation technique using a quasistellar neutron spectrum corresponding to a thermal energy of $kT = 25$ keV. In spite of the very small cross section of this neutron-magic nucleus and a number of additional experimental complications, the stellar value of $\langle \sigma \, v \rangle / v_T = 226 \pm 10$ $\mu$b could be reliably determined. Because of this small cross section, $^{34}$S acts as a bottle neck for the reaction flow to $^{36}$S. Consequently, the suggested interpretation of the $^{36}$S abundance as a neutron monitor for the *s* process in massive stars must be ruled out. This implies, however, that the origin of $^{36}$S remains an open problem.

* Astrophys. J. **528** (2000) 573.

## 3. Neutron Capture of $^{46}$Ca at Thermonuclear Energies*

P. Mohr[1], P.V. Sedyshev[4], H. Beer, W. Stadler[2], H. Oberhummer[2], Yu.P. Popov[4], W. Rochow[3]

The measurement of the reaction $^{46}$Ca(n,$\gamma$)$^{47}$Ca is of astrophysical interest, because $^{46}$Ca is bypassed by charged-particle reactions. The nucleus $^{46}$Ca is produced and destroyed by neutron-induced nucleosynthesis in hydrostatic helium, carbon and neon burning through

[1]Technical University Darmstadt, D-64289 Darmstadt, Germany
[2]Technical University Vienna, A-1040 Vienna, Austria
[3]University of Tübingen, D-72076 Tübingen, Germany
[4]Frank Laboratory of Neutron Physics, JINR, 141980 Dubna, Moscow Region, Russia

the reaction chain $^{45}$Ca(n,$\gamma$)$^{46}$Ca(n,$\gamma$)$^{47}$Ca. At the Karlsruhe and Tübingen 3.75 MV Van de Graaff accelerators the thermonuclear $^{46}$Ca(n,$\gamma$)$^{47}$Ca(4.54 d) cross section was measured by the activation technique via the 1297.09 keV $\gamma$-ray line of the $^{47}$Ca decay. Samples of $CaCO_3$ enriched in $^{46}$Ca by 5 % were irradiated between two gold foils which served as capture standards using the $^{7}$Li(p,n) and T(p,n) reactions. The capture cross section was measured at the mean neutron energies of 30, 104, 149, 180 and 215. Maxwellian averaged capture cross sections were measured at the quasi-thermal neutron energies kT=25 keV and 52 keV. It was found that the $^{46}$Ca(n,$\gamma$)$^{47}$Ca cross section in the thermonuclear energy region and at thermal energy is dominated by an s-wave resonance at 28.4 keV with a neutron width $\Gamma_n = \left(17.4^{+3.6}_{-2.8}\right)$ keV and a radiation width $\Gamma_\gamma$=(2.4±0.3) eV. The stellar reaction rate is determined in the temperature range from kT=1 to 250 keV and is compared with previous investigations using Hauser-Feshbach calculations or experimental cross section data. The astrophysical consequences of the new stellar reaction rate with respect to the nucleosynthetic abundance of $^{46}$Ca are discussed.

* Phys. Rev. C **59** (1999) 3410.

### 4. Measurement of Neutron Capture on $^{50}$Ti at Thermonuclear Energies*

P.V. Sedyshev[4], P. Mohr[1], H. Beer, H. Oberhummer[2], Yu.P. Popov[4], W. Rochow[3]

At the Karlsruhe and Tübingen 3.75 MV Van de Graaff accelerators the thermonuclear cross section of the reaction $^{50}$Ti(n,$\gamma$)$^{51}$Ti(5.8 min) was measured by the fast cyclic activation technique via the 320.852 and 928.65 keV $\gamma$-ray lines of the $^{51}$Ti decay. Metallic Ti samples of natural isotopic composition and samples of $TiO_2$ enriched in $^{50}$Ti by 67.53% were irradiated between two gold foils which served as capture standards. The capture cross section was measured at the neutron energies of 25, 30, 52 and 145 keV. The direct capture cross section was determined to be 0.387±0.011 mb at 30 keV. We found evidence for a bound state *s*-wave resonance with an estimated radiative width of 0.34 eV which destructively interferes with direct capture. The strength of a suggested *s*-wave resonance at 146.8 keV was determined. The present data served to calculate, in addition to the directly measured Maxwellian averaged capture cross sections at 25 and 52 keV, an improved stellar $^{50}$Ti(n,$\gamma$)$^{51}$Ti rate in the thermonuclear energy region from 1 to 250 keV. The new stellar rate leads at low temperatures to much higher values than the previously recommended rate; e.g., at $kT = 8$ keV the increase amounts to about 50%. The new reaction rate therefore reduces the abundance of $^{50}$Ti due to *s* processing in asymptotic giant branch stars.

* Phys. Rev. C **60** (1999) 054613/1-8.

### 5. Stellar Neutron Capture Cross Section of the Yb Isotopes*

K. Wisshak, F. Voss, F. Käppeler, L. Kazakov[5]

The neutron capture cross sections of $^{170}$Yb, $^{171}$Yb, $^{172}$Yb, $^{173}$Yb, $^{174}$Yb and $^{176}$Yb have been measured in the energy range from 3 to 225 keV at the Karlsruhe 3.75 MV Van de

[5]Institute for Physics and Power Engineering, Obninsk, Kaluga Region, Russia

Graaff accelerator. Neutrons were produced via the $^7$Li$(p,n)^7$Be reaction by bombarding metallic Li targets with a pulsed proton beam. Capture events were registered with the Karlsruhe $4\pi$ Barium Fluoride Detector. The cross sections were determined relative to the gold standard. Neutron capture in the even ytterbium isotopes is characterized by a strong population of isomeric states, resulting in severe systematic uncertainties in previous experiments. In the present work, the partial cross sections to the ground– and isomeric states in $^{172}$Yb, $^{174}$Yb, and $^{176}$Yb could be experimentally separated for the first time, yielding cross section ratios with an overall uncertainty of 1–1.5%. Compared to previous measurements, this corresponds to an improvement by factors of 4 to 10. Maxwellian averaged neutron capture cross sections were calculated for thermal energies between kT = 8 keV and 100 keV. The results of four isotopes differ by more than 15% from recent evaluations.

* Report FZKA-6194 (1998).

## 6. Photoactivation of $^{180m}$Ta and its Implications for the Nucleosynthesis of Nature's Rarest Stable Isotope*

D. Belic[6], C. Arlandini, J. Besserer[7], J. de Boer[7], J.J. Carroll[8], J. Enders[1], T. Hartmann[1], F. Käppeler, H. Kaiser[1], U. Kneissl[6], M. Loewe[7], H.J. Maier[7], H. Maser[6], P. Mohr[1], P. von Neumann-Cosel[1], A. Nord[6], H.H. Pitz[6], A. Richter[1,9], M. Schumann, S. Volz[1], A. Zilges[1]

The depopulation of the quasi stable isomer in $^{180}$Ta with $J^\pi = 9^-$ at $E_x$ =75 keV by resonant photoabsorption was investigated with intense bremsstrahlung. The results indicate a dramatic acceleration of the isomer decay to the short-lived $J^\pi = 1^+$ ground state under stellar $s$-process conditions. The consequences for a possible nucleosynthesis of nature's rarest stable isotope $^{180}$Ta within the $s$ process is discussed.

* Phys. Rev. Lett. **83** (1999) 5242.

## 7. Neutron Capture Cross Section of $^{232}$Th*

K. Wisshak, F. Voss, F. Käppeler

The neutron capture cross section of $^{232}$Th has been measured in the energy range from 5 to 225 keV at the Karlsruhe 3.75 MV Van de Graaff accelerator relative to the gold standard. Neutrons were produced via the $^7$Li$(p,n)^7$Be reaction by bombarding metallic Li targets with a pulsed proton beam and capture events were registered with the Karlsruhe $4\pi$ Barium Fluoride Detector. The main difficulty in this experiment is the detection of true capture events characterized by a comparably low binding energy of 4.78 MeV in the presence of the high-energy $\gamma$-background (up to 3.96 MeV) associated with the decay chain of the natural thorium sample. With the high efficiency and the good energy

[6]Institut für Strahlenphysik, Universität Stuttgart, D-70569 Stuttgart, Germany
[7]Sektion Physik, Universität München, D-85748 Garching, Germany
[8]Department of Physics and Astronomy, Youngstown State University, Youngstown, Ohio 44555, USA
[9]Wissenschaftskolleg zu Berlin, D-14193 Berlin, Germany

resolution of the $4\pi$ detector the sum energy peak of the capture cascades could be reliably separated from the background over the full range of the neutron spectrum, yielding cross section uncertainties of about 2% above 20 keV and of 4% at 5 keV. The clear identification of the various background components represents a significant improvement compared to existing data for which sometimes high accuracy was claimed, but which were found to be severely discrepant. A comparison with the evaluated files shows reasonable agreement in the energy range above 15 keV, but also severe discrepancies of up to 40% at lower neutron energies.

* Report FZKA-6275 (1999).

## 8. The Origin of the Heavy Elements: the *s* Process*

F. Käppeler

The heavy elements with Z≥30 are made in about equal quantities by neutron capture reactions during stellar He burning and presumably in supernovae. This contribution deals mainly with the slow neutron capture (*s*) process which is responsible for about one half of the abundances in the mass region between Fe and Bi. The slow time scale implies that the reaction path of this process involves mostly stable isotopes which can be studied in detail in laboratory experiments. Based on these data, the quantitative interpretation of the natural abundances provides an exciting possibility for exploring a variety of problems related to stellar and Galactic evolution. The *p* process, which provides a very small but significant admixture to many of the *s* abundances, has recently attracted increasing interest as a possibility for supernova studies.

* Progress in Particle and Nuclear Physics **43** (1999) 419.

## 9. Neutron Capture in Low-Mass Asymptotic Giant Branch Stars: Cross Sections and Abundance Signatures*

C. Arlandini, F. Käppeler, K. Wisshak, R. Gallino[10], M. Lugaro[11], M. Busso[12], O. Straniero[13]

Recently improved information on the stellar (n,$\gamma$) cross sections of neutron magic nuclei at N=82, and in particular of $^{142}$Nd, turn out to represent a sensitive test for models of *s*-process nucleosynthesis. While these data were found to be incompatible with the classical approach based on an exponential distribution of neutron exposures, they provide significantly better agreement between the solar abundance distribution of *s* nuclei and the predictions of models for low-mass asymptotic giant branch (AGB) stars. The origin of this phenomenon is identified as lying in the high neutron exposures at low neutron density obtained between thermal pulses when $^{13}$C burns radiatively in a narrow layer of a few $10^{-4}$ $M_\odot$. This effect is studied in some detail, and the influence of the currently

[10]Dipartimento di Fisica Generale, Universitá di Torino, I-10125 Torino, Italy
[11]Department of Mathematics, Monash University, Clayton, Victoria 3168, Austalia
[12]Osservatorio Astronomico di Torino, I-10025 Torino, Italy
[13]Osservatorio Astronomico di Collurania, I-64100 Teramo, Italy

available nuclear physics data is discussed with respect to specific further questions. In this context, particular attention is paid to a consistent description of $s$-process branchings in the region of the rare earth elements. It is shown that, in certain cases, the nuclear data are sufficiently accurate that the resulting abundance uncertainties can be completely attributed to stellar modeling. Thus, the $s$-process becomes important for testing the role of different stellar masses and metallicities as well as for constraining the assumptions used in describing the low neutron density provided by the $^{13}C$ source.

* Astrophys. J. **525** (1999) 886.

**FORSCHUNGSZENTRUM KARLSRUHE**
**INSTITUT FÜR KERN- UND ENERGIETECHNIK**

## Integral Data Tests, Sensitivity and Uncertainty Analyses of the EFF-3 $^{58}$Ni Data Evaluation

U. Fischer, Ch. Konno[1], R. Perel[2]

In the framework of the European Fusion File (EFF) Project, benchmark, sensitivity and uncertainty analyses are being performed as part of the QA (quality assurance) procedure for new and updated EFF data evaluations. The applied data testing methodology includes three-dimensional Monte Carlo calculations with the MCNP code to calculate the neutron leakage flux spectra as measured in the considered integral experiments. Sensitivity and uncertainty analyses include three-dimensional Monte Carlo calculations with a local update to MCNP4A [1] for calculating sensitivities of point detector tallies. With these sensitivity profiles and the covariance matrices generated with the NJOY processing code from the EFF data files, uncertainties of the calculated point detector responses due to uncertainties of the cross-section data are being assessed.

Over the reporting period, the focus of the data test analyses has been on the new EFF-3.0 $^{58}$Ni evaluation. Two integral nickel experiments have been considered: the OKTAVIAN [2] and the IPPE Obninsk [3] time-of-flight experiments on spherical nickel assemblies with a central 14 MeV neutron source. The effective thickness of the nickel shell was 16 and 7.5 cm in the OKTAVIAN and the IPPE experiment, respectively.

The two experiments show consistent results in the data test analyses. For the measured neutron leakage flux spectra, good agreement has been obtained with the new EFF-3.0 $^{58}$Ni evaluation and the FENDL-1 (ENDF/B-VI) nickel data as well, see Fig. 1 for the neutron leakage spectrum in the IPPE experiment. For FENDL-1 this applies to the whole measured energy range.

*[1] Permanent address: Centre for Neutron Science, JAERI, Tokai-mura, Ibaraki-ken 319-1195, Japan*
*[2] Permanent address: Racah Institute of Physics, Hebrew University of Jerusalem, 91904 Jerusalem, Israel*

For EFF-3.0, this does not apply to the energy range 5 – 10 MeV where the neutron flux integral is overestimated by 20 – 30 %. In accordance with the results of the sensitivity analyses, this suggests to reduce the inelastic cross-section around 14 MeV which is responsible for the neutron population of the energy range 5 – 10 MeV. A major progress has been achieved with the new covariance data of EFF-3.0 resulting in significantly reduced uncertainties of the calculated neutron flux integrals, see Tables 1 - 2.

**References**


[1] R.L. Perel, J.J. Wagschal, and Y. Yeivin, "Monte Carlo calculation of point-detector sensitivities to material parameters", Nucl. Sci. Eng., **124** (1996) 197-209.

[2] T. Kasahara et al., "Measurements of Neutron Leakage Spectra from 16 cm Nickel Sphere", Report OKTAVIAN A-84-04, Osaka University 1984.

[3] S. Simakov et al., "Benchmarking of evaluated neutron data for nickel by 14 MeV spherical shell transmission experiments", Fusion Technology **36,** (1999) 173 – 180.


Table 1 Calculated uncertainties (in %) of the neutron leakage due to cross-section uncertainties in the OKTAVIAN experiment.

| Energy [MeV] | < 0.05 | 0.05 – 0.1 | 0.1 – 0.5 | 0.5 - 1 | 1 – 5 | 5 – 10 | > 10 | Total |
|---|---|---|---|---|---|---|---|---|
| FENDL-1 | 32.9 | 6.5 | 4.6 | 4.1 | 5.1 | 8.4 | 8.6 | 4.56 |
| EFF-3.0 | 47.2 | 11.7 | 6.75 | 3.4 | 1.35 | 2.5 | 1.3 | 2.5 |

Table 2 Calculated uncertainties (in %) of the neutron leakage due to cross-section uncertainties in the IPPE Obninsk experiment.

| Energy [MeV] | < 0.05 | 0.05 – 0.1 | 0.1 – 0.5 | 0.5 - 1 | 1 – 5 | 5 – 10 | > 10 | Total |
|---|---|---|---|---|---|---|---|---|
| FENDL-1 | 12.5 | 4.1 | 3.0 | 2.8 | 3.4 | 5.4 | 4.5 | 2.9 |
| EFF-3.0 | 21.4 | 6.8 | 4.0 | 2.2 | 0.9 | 1.7 | 0.6 | 0.9 |

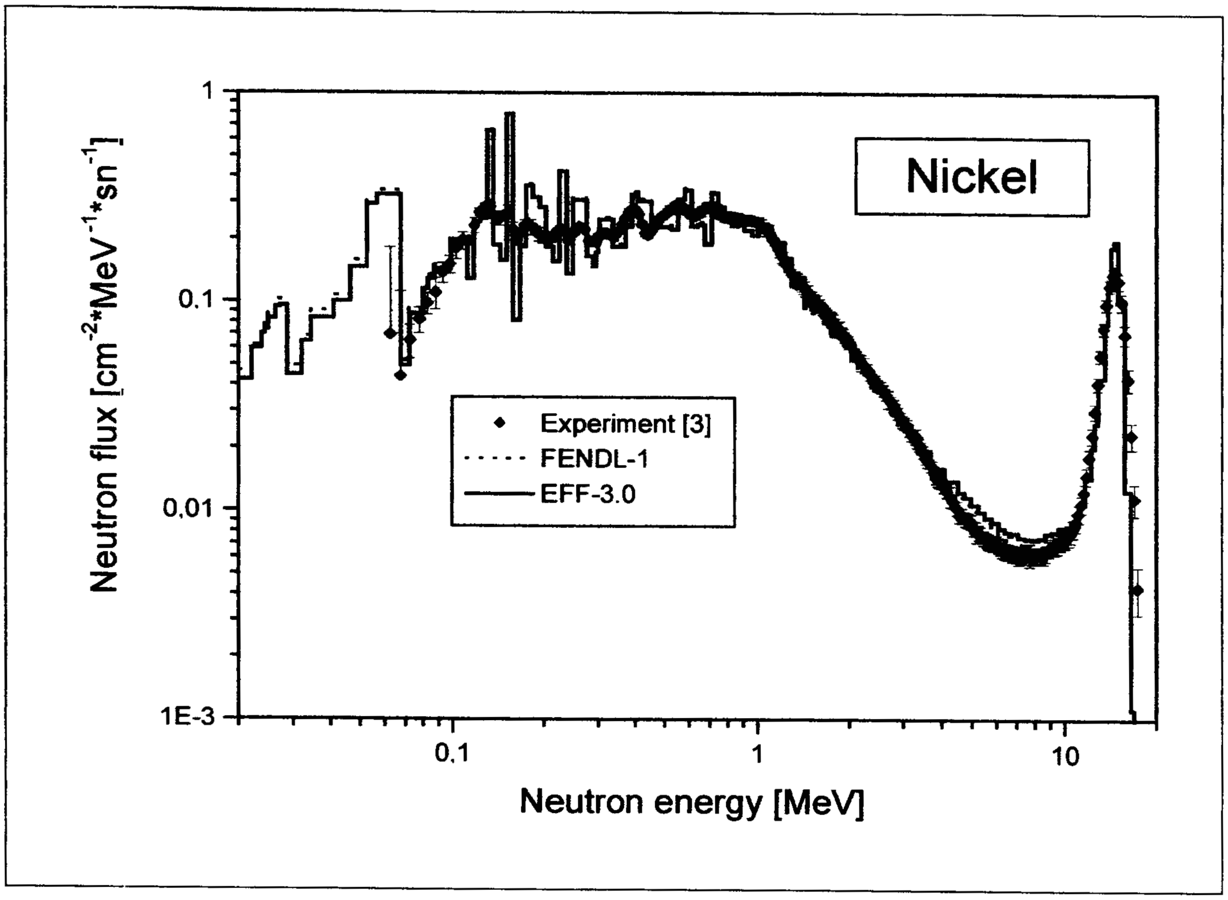


**Fig. 1** Neutron leakage flux spectra in the IPPE Obninsk spherical shell (t=7.5 cm) experiment on nickel.

# INSTITUT FÜR NUKLEARCHEMIE
# FORSCHUNGSZENTRUM JÜLICH

## 1. Fundamental Studies on Nuclear Reactions

A.Hohn, S.Kastleiner, F.Cserpák*, S.Sudár*, N.Shubin†, J.Csikai*, S.M.Qaim

Experimental and theoretical studies on nuclear reactions, especially from mechanistic point of view, have been under way for several years. Recent work consisted of measurement of excitation functions of (p,xn) reactions on $^{68}$Zn, $^{85}$Rb, $^{120}$Te and $^{122}$Te over proton energies up to 70 MeV and of (n,2n) and (n,$\alpha$) reactions on $^{107}$Ag and $^{109}$Ag over neutron energies of 5 to 12 MeV. Besides measurement of total cross section for a particular reaction channel, partial cross sections for the population of isomeric states were also determined. To interpret the experimental data, nuclear model calculations were performed. In the low energy region up to 30 MeV the statistical model incorporating precompound effects was used. For this purpose the Code STAPRE was employed. In the higher energy region up to 70 MeV the Code ALICE-IPPE, based on the hybrid model, was used.

It was found that processes like $^{68}$Zn(p,xn)$^{66-68}$Ga, $^{120}$Te(p,xn)$^{119,120}$I, $^{122}$Te(p,xn)$^{119-122}$I, $^{107}$Ag(n,2n)$^{106}$Ag and $^{107}$Ag(n,p)$^{107}$Pd can be described well by the nuclear model calculations. However, discrepancies between experiment and theory were observed when emission of complex particles or formation of isomeric states was involved. The excitation functions of the $^{107}$Ag(n,$\alpha$)$^{104}$Rh and $^{109}$Ag(n,$\alpha$)$^{106}$Rh reactions, for example, could not be described by the STAPRE calculations so well as the (n,p) and (n,2n) excitation functions. The isomer formation is discussed below in more detail.
The isomeric cross section ratios for the isomeric pair $^{120m,g}$I were determined in three reactions, viz.$^{122}$Te(p,3n), $^{120}$Te(p,n) and $^{120}$Te(d,2n). The experimental data and the results of nuclear model calculations for the $^{122}$Te(p,3n) reaction are shown in Fig.1. The ground state spin is known ($2^-$) but there is uncertaintry both about the excitation energy and the spin of the isomeric state. Calculations were therefore performed for spin values

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* Institute of Experimental Physics, Debrecen University, Debrecen, Hungary
† Institute of Physics and Power Engineering (IPPE), Obninsk, Russia

between 4 and 8 and parity (+) and (–), assuming the excitation energy of $^{120m}I$ as 900 keV. From Fig.1 it is evident that only the $4^+$,$4^-$ and $5^+$ spin/parity assignments give results near to the experimental data. A calculation with spin $4^+$ and excitation energy 550 keV gave the best result, not only for the $^{122}Te(p,3n)$ reaction shown in Fig.1 but also for the $^{120}Te(p,n)$ and $^{120}Te(d,2n)$ processes. Studies on isomeric cross section ratios can thus shed some light on the level structure of the product isotope. The isomeric cross section ratio is strongly dependent on the spins of the isomers concerned. The statistical model is generally adequate to describe the experimental data; however, a careful choice of the input parameters is absolutely essential.

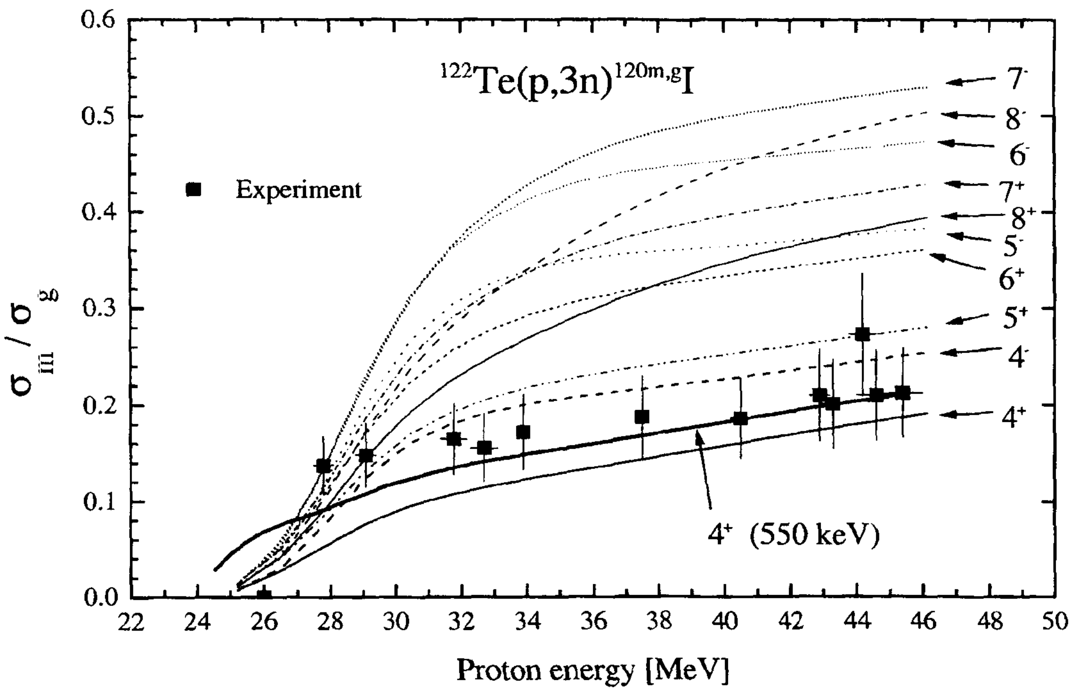


Fig.1 Isomeric cross-section ratio ($\sigma_m/\sigma_g$) for the isomeric pair $^{120m,g}I$ in $^{122}Te(p,3n)$-reaction as a function of proton energy. Results of calculations using an excitation energy of $^{120m}I$ as 900 keV, various spin values and change in parity, are shown as curves. The best result (thick solid line) appears to be obtained with a calculation assuming spin $4^+$ and an excitation energy of 550 keV for $^{120m}I$.

## 2. Neutron Activation Cross Sections

F.Cserpak*, P.Reimer†, S.Sudár*, S.Spellerberg, M.Ibn Majah**, J.Csikai*, S.M.Qaim

*(Relevant to request identification numbers: 861129 F, 861142 F, 861182 F, 924008 R)*

During the period of this report, the emphasis has been in four directions :

(1) Formation of long-lived activities

(2) Cross sections for radioactive target nuclei

(3) Study of (n,n′γ) reactions

(4) Cross section measurement in the 5 to 12 MeV energy region and integral tests of differential data.

Work on the first two topics is being performed mainly in collaboration with the IRMM Geel (A.Plompen). Regarding the formation of long-lived activities, the reactions of interest include $^{94}Mo(n,p)^{94}Nb$ ($T_{½}=2\times10^4$ a) and $^{204}Pb(n,p)^{204}Tl$ ($T_{½}=3.78$ a). Long irradiations covering several neutron energy points between 12 and 20 MeV were done and clean radiochemical separations have been performed. Measurement of radioactivity via low-level γ- and β-counting is in progress. Regarding measurements on radioactive target nuclei, detailed studies on $^{99}Tc$ ($T_{½}=2.1\times10^5$ a) are under way. The excitation functions of the $^{99}Tc(n,p)^{99}Mo$ and $^{99}Tc(n,\alpha)^{96}Nb$ reactions over the neutron energy range of 8 to 18 MeV have been determined and work on the $^{99}Tc(n,n'\gamma)^{99m}Tc$ process over the energy region of 5 to 12 MeV is continuing.

As is well known, the cross section data base for the (n,n′γ) process in the neutron energy region above 5 MeV is rather weak. This is mainly due to the large uncertainties in correcting the contributions of low-energy background neutrons to the formation of the activation product. A programme of measurement has been initiated at Jülich in collaboration with the University of Debrecen, Hungary, using a relatively low-

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* Institute of Experimental Physics, Debrecen University, Debrecen, Hungary

† Mainly at IRMM, Geel, Belgium

** CNESTEN, Rabat, Morocco

background $^{2}H(d,n)^{3}He$ neutron source at the compact cyclotron CV28. The neutron energy range covered extends from 5 to 12 MeV. After completing measurements on $^{89}Y(n,n'\gamma)^{89m}Y$ and $^{99}Tc(n,n'\gamma)^{99m}Tc$ processes, detailed studies are now under way on $^{107}Ag(n,n'\gamma)^{107m}Ag$ and $^{109}Ag(n,n'\gamma)^{109m}Ag$ reactions. Use of thin target samples and soft $\gamma$-ray spectrometry was found to be mandatory.

In continuation of our on-going studies on neutron induced reactions over the energy range of 5 to 12 MeV, we completed cross section measurements on the reactions $^{107}Ag(n,\alpha)^{104m,g}Rh$ and $^{109}Ag(n,\alpha)^{106m,g}Rh$. Furthermore, integral cross sections were measured for (n,p) and (n,$\alpha$) reactions on several Zr isotopes using a 14 MeV d(Be) breakup neutron field. A comparsion of the experimental integral data with the integrated data obtained from the excitation functions and the neutron spectral distribution served to validate the previously measured differential data of several reactions. In addition to our own results on the excitation functions, the evaluated values available in two data files (ENDF/B-VI and JEF-2) for a few reactions were also considered. The results are given in Table1. In general, our two data sets agree within 12 %; adding confidence to our both differential and integral measurements. These results also depict that, for the reactions considered, new evaluations of excitation functions are needed, taking into account the recently published data.

Table 1. Measured and calculated 14 MeV d(Be)-breakup neutron spectrum-averaged cross sections

| Reaction | Measured <br> <σ> <br> (mb) | Calculated[a] <σ> (mb) | | |
|---|---|---|---|---|
| | | Jülich data | ENDF/BV1 | JEF-2 |
| $^{90}Zr(n,p)^{90m}Y$ | 1.54 ± 0.12 | 1.63 | | |
| $^{90}Zr(n,\alpha)^{87m}Sr$ | 0.41 ± 0.03 | 0.45 | | |
| $^{91}Zr(n,p)^{91m}Y$ | 2.39 ± 0.15 | 2.67 | | |
| $^{92}Zr(n,p)^{92}Y$ | 1.88 ± 0.16 | 2.14 | 2.16 | 1.92 |
| $^{94}Zr(n,p)^{94}Y$ | 0.61 ± 0.05 | 0.59 | 0.83 | 0.81 |
| $^{94}Zr(n,\alpha)^{91}Sr$ | 0.57 ± 0.05 | 0.53 | 0.35 | 0.27 |
| $^{96}Zr(n,2n)^{95}Zr$ | 322 ± 25 | 275 | 226 | 278 |

[a] From reported excitation functions

## 3. Charged Particle Induced Reaction Cross Sections

S.Kastleiner, E.Heß, A.Hohn, T.Bastian, B.Scholten, S.Spellerberg, F.M.Nortier*, T.N. van der Walt*, F.Tárkányi†, Z.Kovács†, S.Sudár**, M.R.Zaman††, H.H.Coenen, S.M.Qaim

In continuation of our systematic studies on charged particle induced reaction cross section data for medical applications, during the present report following activities were pursued.

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* NAC, Faure, South Africa
† ATOMKI, Debrecen, Hungary
** Institute of Experimental Physics, Debrecen University, Debrecen, Hungary
†† Department of Applied Chemistry, Rajshahi University, Rajshahi, Bangladesh

a) *Measurement of excitation functions relevant to the production of the positron emitters $^{11}C$, $^{18}F$, $^{55}Co$, $^{83}Sr$ and $^{124}I$*

The radioisotope **$^{11}$C** ($T_{½}$=20 min) is generally produced via the $^{14}N(p,\alpha)^{11}C$ reaction on an $N_2$ gas target.Simultaneously the radioisotope $^{14}O$ ($T_{½}$=70 s) is formed as impurity via the $^{14}N(p,n)^{14}O$ reaction. The excitation function of the latter process is not known well. We performed measurements from the reaction threshold up to 20 MeV using the stacked gas cell technique commonly employed in this laboratory. Presently data analysis is in progress.

In connection with the **$^{18}$F** ($T_{½}$=110 min) production via the $^{18}O(p,n)^{18}F$ reaction, the on-going work on excitation function measurement was extended. Besides the use of the stacked gas cell technique, stacked thin solid samples were employed. For this purpose, on the one hand thin $Si^{18}O_2$ samples on Ti-backing and on the other, ultra thin $Al_2{}^{18}O_3$ on Al-backing were used. Measurements near the threshold of the reaction (3-4 MeV) were done at the Debrecen Van de Graaff, between 4 and 7 MeV at the Debrecen cyclotron and between 20 and 30 MeV at the Injector of COSY at Jülich. Data analysis is presently in progress.

The radioisotope **$^{55}$Co** ($T_{½}$=17.5 h) is produced via several routes, the purest product being obtained via the $^{54}Fe(d,n)^{55}Co$ reaction on highly enriched $^{54}Fe$ as target material. The excitation function of this reaction was measured some time ago. However, the data for the competing $^{54}Fe(d,\alpha)^{52m,g}Mn$ process are not known with the required accuracy. In the present work thin $^{54}Fe$ samples were prepared via electrolytic deposition and the excitation function of the above mentioned $(d,\alpha)$ process was determined.

As mentioned in the last Progress Report the radioisotope **$^{83}$Sr** ($T_{½}$=32.4 h) is a positron emitting analogue of the therapy radioisotope $^{89}Sr$ and appears to be well-suited for dosimetry and therapy planning via PET. Measurements on the potentially important production reaction $^{85}Rb(p,3n)^{83}Sr$ were completed and the results are shown in Fig 2. The proton energy range $E_p$=45→25 MeV appears to be most suitable for production purposes: the thick target yield of $^{83}Sr$ amounts to 350 MBq/μAh and the level of the $^{85}Sr$ impurity to 0.21 %.

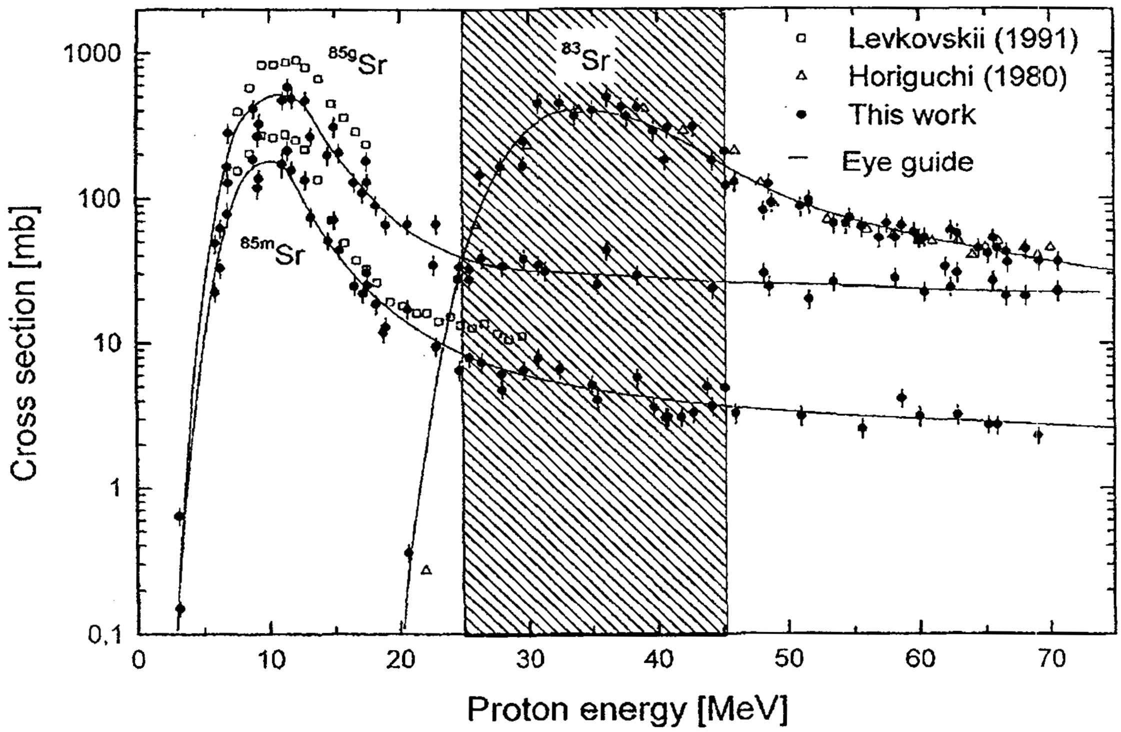


Fig. 2 Excitation functions of $^{85}Rb(p,n)^{85m,g}Sr$ and $^{85}Rb(p,3n)^{83}Sr$ processes

After having completed measurements on the production of the positron emitting $^{120}I$ ($T_{½}$=81 min), we devoted more efforts to the data relevant to the production of the longer-lived $\beta^+$ emitting radioisotope $^{124}$**I** ($T_{½}$=4.18 d). The excitation function of the commonly used $^{124}Te(d,2n)^{124}I$ reaction has not been accurately determined. We initiated studies on this reaction. A comparison of this process with the low-energy reaction $^{124}Te(p,n)^{124}I$ is planned. Furthermore, we measured excitation functions of the $^{125}Te(p,2n)^{124}I$ and several competing reactions like $^{125}Te(p,n)^{125}I$, $^{125}Te(p,3n)^{123}I$, etc. on highly enriched $^{125}Te$ from threshold up to 100 MeV. The energy range $E_p$=22→14 MeV appears to be optimum for the production of $^{124}I$: its thick target yield amounts to 111 MBq/μAh and the level of $^{125}I$-impurity to < 0.9 %. The $^{125}Te(p,2n)^{124}I$ reaction thus gives about four times higher yield of $^{124}I$ than the $^{124}Te(p,n)^{124}I$ and $^{124}Te(d,2n)^{124}I$ processes.

*b) Cross sections and yields of therapy related radioisotopes $^{67}Cu$, $^{103}Pd$ and $^{140}Nd$*

In continuation of our studies on the production of endotherapy related radioisotopes, yields of $^{67}$**Cu** ($T_{½}$=61.9 h) and $^{140}$**Nd** ($T_{½}$=3.4 d) were measured in $^{68}Zn(p,2p)$ and $^{140}Ce(^3He,3n)$ reactions, respectively, the latter in collaboration with the Institute of Nuclear Chemistry, University of Mainz (F.Rösch).Both routes are feasible for production of the respective radioisotpe.

The radioisotope $^{103}$**Pd** ($T_{½}$=16.96 d) is gaining considerable significance in endoradiotherapy. It is produced routinely in sufficient quantities via the $^{103}Rh(p,n)$ reaction. The cross sections of the process, however, are unknown. We measured the excitation function of this reaction up to 30 MeV using the stacked-foil technique in combination with X-ray spectrometry. The competing nuclear processes $^{103}Rh(p,3n)^{101}Pd$ and $^{103}Rh(p,pn)^{102m,g}Rh$ were also investigated. The data analysis is in progress.

*c) Compilation of data and dissemination of knowledge on nuclear data for medical applications*

Under the auspices of the IAEA, a "Co-ordinated Research Programme (CRP) on the Standardisation of Nuclear Data for the Production of Medically Important Radioisotopes" has been under way for about 4 years and has now been completed. S.M.Qaim.chaired this CRP. As a participating institute, we carried the responsibility of compiling the data on the short-lived positron emitters. An IAEA-TECDOC Report has been written by the participating groups. It has been edited and is now in press for publication.

An autumn college on "Nuclear Data for Medical Applications" was organised for the first time under the auspices of the IAEA and UNESCO. It was held at the Abdus Salam ICTP Trieste, Italy. The institute contributed appreciably to the dissemination of knowledge since S.M.Qaim worked as a Course Director and also delivered a series of lectures. The condensed manuscripts are expected to be published in a peer-reviewed journal.

## 4. Decay Data

A.Hohn, H.H.Coenen, S.M.Qaim

The $\beta^+$/EC ratio in the decay of $^{120g}I$ ($T_{½}$=1.35 h), i.e. the $\beta^+$ emission intensity ($I_{\beta^+}$), is not known well: values between 39 and 81 % have been reported in the literature. We determined the intensity for the first time using a high purity $^{120g}I$ source, which was produced by irradiation of 99 % enriched $^{120}Te$ with 15 MeV protons, followed by a chemical separation of radioiodine. Both $\gamma$-ray and X-ray spectrometry were applied. In the former case a comparison of the intensity of the annihilation radiation with that of the 1523 keV $\gamma$-ray of $^{120g}I$ was done and in the latter with that of the $K_\alpha$ or $K_\beta$ X-ray of the daughter tellurium. Corrections for very small contributions of $^{121}I$ and $^{120m}I$ ipurities were evaluated. Our measurements lead to a value of 56±3 % for the $I_{\beta^+}$ of $^{120g}I$.

### Publications (During the Period of the Progress Report)


[1] C. Nesaraja, K.-H. Linse, S. Spellerberg, S. Sudár, A. Suhaimi and S.M. Qaim
Excitation functions of neutron induced reactions on some isotopes of zinc, gallium and germanium in the energy range of 6.2 to 12.4 MeV
Radiochimica Acta **86** (1999) 1

[2] A.Hohn, H.H. Coenen and S.M. Qaim
Excitation functions of $^{120}Te(d,xn)^{121,120m,g}I$ reactions from threshold up to 13.5 MeV: comparative studies on the production of $^{120g}I$
Appl.Radiat. Isot. **52** (2000) 923

[3] S. Sudár, A. Hohn and S.M. Qaim
Nuclear model calculations on proton and deuteron induced reactions on $^{122}Te$ and $^{120}Te$ with particular reference to the formation of the isomeric states $^{120m,g}I$
Appl. Radiat. Isot. **52** (2000) 937

[4] R. Dóczi, S. Takács, F. Tárkányi, B. Scholten and S.M. Qaim
Possibility of production of $^{81}Rb$ via the $^{80}Kr(d,n)$ reaction
Radiochimica Acta, **88** (2000) 135

[5] A. Hohn, H.H. Coenen and S.M. Qaim
Positron emission intensity in the decay of $^{120g}I$
Radiochimica Acta, **88** (2000) 139

[6] A.T.J. Klein, F. Rösch and S.M. Qaim
Investigation of $^{50}Cr(d,n)^{51}Mn$ and $^{nat}Cr(p,x)^{51}Mn$ processes with respect to the production of the positron emitter $^{51}Mn$
Radiochimica Acta, in press

[7] M. Ibn Majah, A. Chiadli, S. Sudár and S.M. Qaim
Cross sections of (n,p), (n,α) and (n,2n) reactions on some isotopes of zirconium in the neutron energy range of 10 to 12 MeV and integral tests of differential cross section data using a 14 MeV d (Be) neutron spectrum
Appl. Radiat. Isot., in press

# INSTITUT FÜR KERN- UND TEILCHENPHYSIK TECHNISCHE UNIVERSITÄT DRESDEN

## 1. Radioactivities induced in the Fusion Power Plant Structural Material SiC and in the Breeder Material $Li_4SiO_4$ by 14 MeV Neutrons*

R. A. Forrest[1], H. Freiesleben, V. D. Kovalchuk[2], D. V. Markovskij[3], D. Richter, K. Seidel, V. I. Tereshkin[2], S. Unholzer

In continuation of our experimental studies on radioactivities induced in fusion power plant structural materials by 14 MeV neutrons (steels SS316, MANET, F82H [1,3] and vanadium alloys V3Ti1Si, V4Cr4Ti, V5Ti2Cr [2,3]) we investigated SiC and $Li_4SiO_4$. SiC is under discussion as an advanced structural material with high thermodynamic efficiency and with inherently low induced radioactivity. $Li_4SiO_4$ is used as breeding material in the European Helium Cooled Pebble Bed Blanket design [4].

Small pieces of the materials were irradiated at the high-intensity d-t neutron generator SNEG-13 at Sergiev Posad, each in two separate runs, a short one (30 min) with fluence of the order of $10^{11}$ neutrons/cm² and a longer one (17 h) with fluence of the order of $10^{14}$ neutrons/cm². In this way radioactivities with half-lives of a few minutes mainly produced on Si, and long-lived radionuclides from impurities could be investigated. Gamma-spectra of the samples were measured with a Ge(Li)-spectrometer several times during decay.

For each of the measured values the activity was calculated with EASY, and calculated-to-experimental values (C/E) were determined. EASY, the European Activation System [5], represents the reference system for the design of the

* Work supported by the European Fusion Development Activity Programme and by the German Bundesministerium für Bildung und Forschung

[1] EURATOM/UKAEA Fusion Association, Culham Science Centre, Abingdon, United Kingdom
[2] Coordination Centre "Atomsafety", Sergiev Posad, Russia
[3] Russian Research Centre "Kurchatov Institute", Moscow, Russia

International Thermonuclear Experimental Reactor (ITER). Its main components are the European Activation File (EAF) and the inventory code FISPACT developed at Culham.

The results from the short irradiation are presented in Table 1 for both materials. The short-lived radionuclides $^{27}Mg$, $^{28}Al$ and $^{29}Al$ are mainly produced by nuclear reactions on silicon isotopes. The next step, the investigation of the long-lived radionuclides from minor elements and impurities is in progress.

Table 1 Radionuclides produced in **SiC** and **$Li_4SiO_4$**, their half-lives, the γ-rays used to determine the activity, the relative contributions of various nuclear reactions and the ratio of calculated-to-experimental activity (C/E) obtained for several decay times, as well as the percentage of the activity (Rel. C) to the total γ–dose rate at these times.

| **Radionuclide** | | | **$^{27}Mg$** | | **$^{28}Al$** | | **$^{29}Al$** | |
|---|---|---|---|---|---|---|---|---|
| Half-life / min | | | 9.46 | | 2.25 | | 6.57 | |
| γ-ray / keV | | | 844 and 1014 | | 1779 | | 1273 | |
| **Reaction contribution / %** | | | **$^{30}Si(n,\alpha)$ 92.5**<br>**$^{27}Al(n,p)$ 7.5** | | **$^{28}Si(n,p)$ 99.5**<br>**$^{29}Si(n,d)$ 0.5** | | **$^{29}Si(n,p)$ 97.5**<br>**$^{30}Si(n,d)$ 2.5** | |
| | | **Material** | **C/E** | **Rel. C** | **C/E** | **Rel. C** | **C/E** | **Rel. C** |
| **Decay time / min** | 2.97 | SiC | 0.99 | 1.03 | 0.85 | 95.0 | 0.96 | 3.90 |
| | 4.43 | $Li_4SiO_4$ | 0.87 | 1.36 | 0.83 | 93.4 | 0.99 | 5.17 |
| | 7.17 | SiC | 1.02 | 2.59 | 0.91 | 88.8 | 1.00 | 8.57 |
| | 10.5 | $Li_4SiO_4$ | 0.83 | 4.87 | 0.81 | 79.8 | 0.99 | 15.2 |
| | 12.8 | SiC | 1.02 | 7.79 | 0.92 | 70.5 | 0.96 | 21.5 |
| | 20.9 | $Li_4SiO_4$ | 0.83 | 21.3 | | | 0.99 | 47.5 |
| | 24.1 | SiC | 0.99 | 27.6 | | | 1.05 | 52.9 |

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## 2. The Dresden EBIT: A Room Temperature Ion Trap for Studying Atomic Properties of Highly Charged Ions

V. P. Ovsyannikov[1,2], G. Zschornack, F. Ullmann[2], T. Werner, F. Grossmann[2], S. Landgraf

Extensive basic atomic data sets in the X-ray, VUV- and visible light region have been hitherto determined for neutral and low ionized atoms. In a growing manner information on highly-charged heavy ions up to the highest ionization stages is requested in fields like basic atomic physics, fusion diagnostics, plasma physics, astrophysics, metrology and also for technological applications. To meet these requirements, besides atomic structure calculations on different approximation levels, ion sources are needed that are able to produce very highly ionized ions.

[1] Joint Institute for Nuclear Research, Dubna, Russia
[2] LEYBOLD Systems + Service GmbH, Dresden

For the production and investigation of slow highly-charged ions the most common sources are EBIS [1-3] and EBIT [4-6]. Powerful sources are equipped with cryogenic techniques and superconducting magnet coils forming magnetic fields for the generation of highly compressed electron beams, where the atoms are effectively ionized by electron impact.

A novel room temperature EBIT (Dresden EBIT), which produces highly charged ions, has been developed. Working without any cryogenic techniques, it represents a compact, cheap and stable device. A detailed description of this EBIT is given in [7]. The basic idea is to form a highly dense electron beam (by a specially designed electron gun with a highly emissive metal alloy cathode) compressed in the magnetic field of two $SmCo_5$ permanent magnet rings. The ion trap, where the ionization process occurs, is formed by a drift tube structure consisting of three sections. The high voltage of the central section accelerates the electrons to their final kinetic energy where the adjacent sections have electric potentials forming an electrostatic ion trap. The principal scheme of the EBIT is shown in Fig. 1.

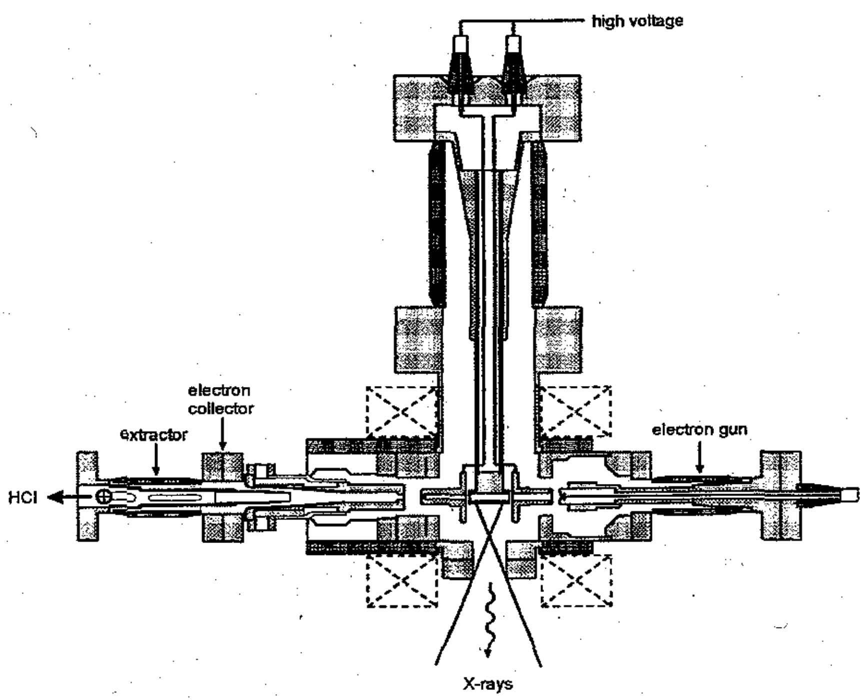


Fig. 1
Sketch of the room temperature Dresden EBIT. Shown are all important parts of the EBIT including the planned ion extraction system.

The main **ion trap parameters** are:

| | |
|---|---|
| electron energy: | up to 15 keV |
| electron beam current: | up to 50 mA |
| electron beam current density: | (200 - 250) A $cm^{-2}$ |
| vacuum without electron beam: | up to $5*10^{-11}$ Torr |
| vacuum with electron beam: | up to $2*10^{-10}$ Torr |
| ionization factor: | (2 - 5)$*10^{21}$ $cm^{-2}$ |
| trap length: | 20 mm |
| EBIT length: | 30 cm |
| EBIT weight: | less than 10 kg |

The Dresden EBIT is equipped with at least one spectroscopic window (optionally two windows are available) consisting of a 75µm thick Be foil. Thus, direct observation of x-rays emitted by different atomic processes (ionization, direct excitation (DE), radiative recombination (RR), dielectric recombination (DR)) is possible. Using a differential-pumped collimator-channel, VUV and visible light can be measured.

A typical x-ray spectrum measured is shown in Fig. 2 with mainly neon-like xenon ions. As the EBIT is able to run for days and weeks without any service, excellent possibilities exist to study the atomic structure of highly-charged heavy ions. The possibility to select the electron energy very precisely allows to prepare different ionic configurations and provides measurements above or below the energy that is characteristic of ionization or direct excitation of different ionic substates and ionization states, as well as for excitation functions and other atomic properties.

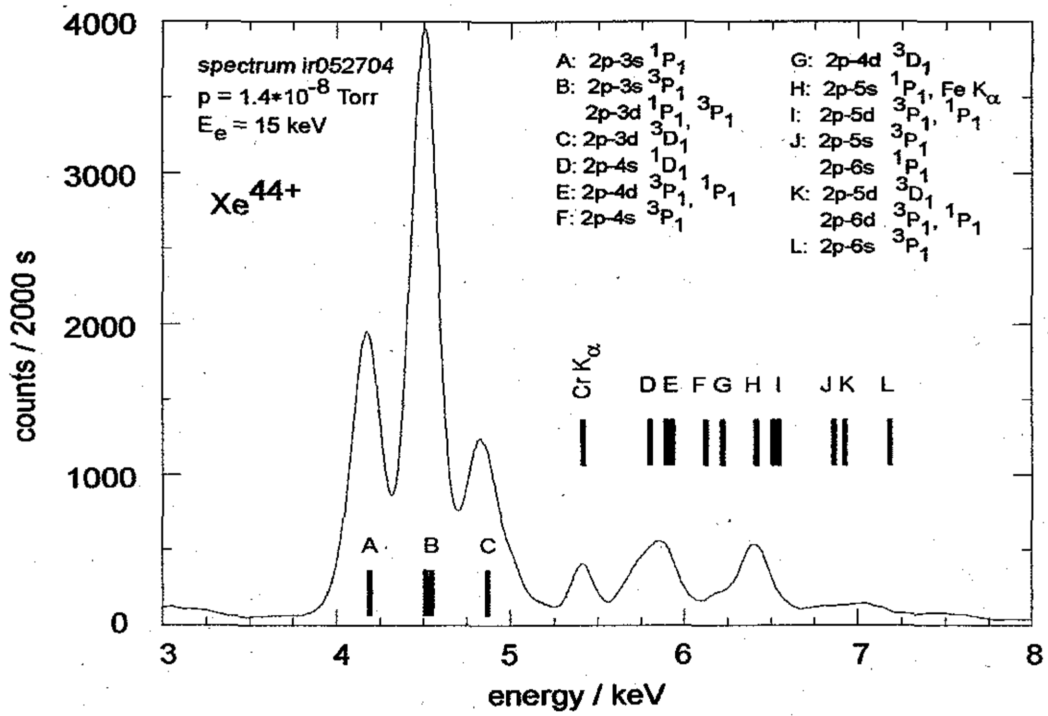


Fig. 2 Xenon x-ray spectrum at an electron energy of 15 keV and a pressure of $1.4 \cdot 10^{-8}$ Torr inside the trap (pressure measurement was done at a point about 15 cm away from the trap region) measured with a Si(Li) solid state detector. DE lines are shown, mainly from $Xe^{44+}$ ions with a blend of contributions from ions at neighbouring ionization stages.

The trap capacity of the described EBIT can be estimated to about $8 \cdot 10^7$ elementary charges in the trap at an electron energy of 15 keV and an electron beam current of 50 mA. The effective solid angle in the spectroscopic measurements is limited by the minimum distance between the Be window and the electron beam to 37 mm. Thus measurements with comparatively high count statistics needed in experiments for a precise determination of ionic quantities for metrology and atomic structure investigations are possible.

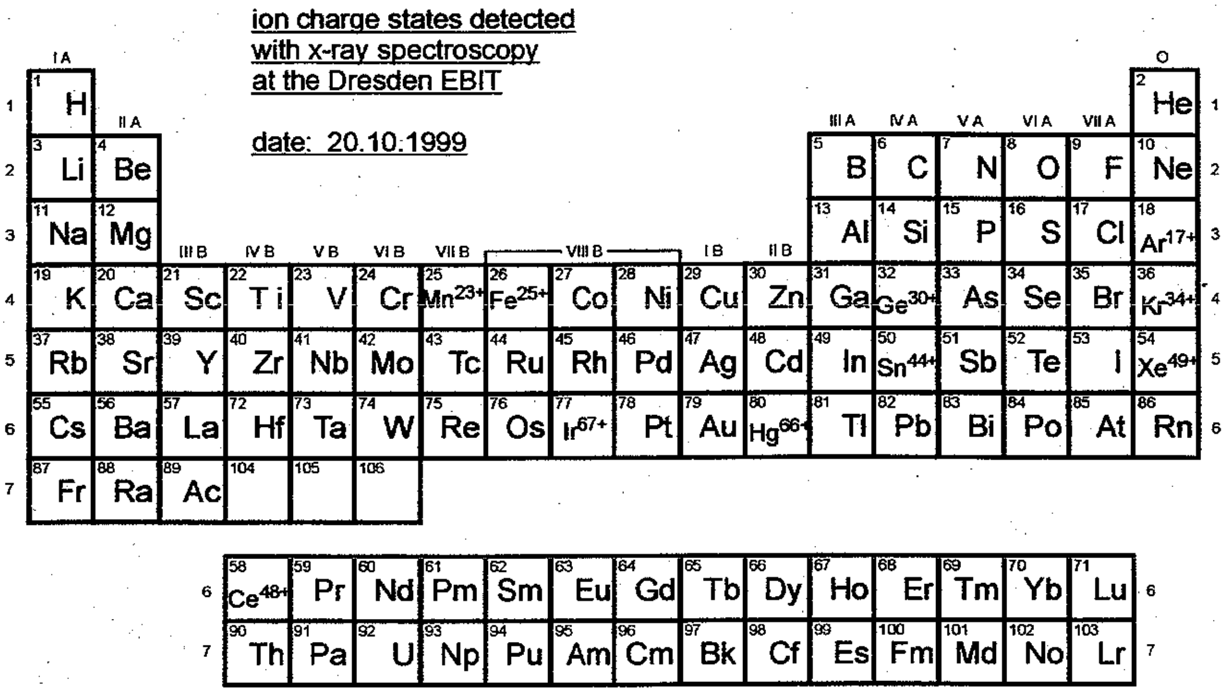


Fig. 3 Ion charge states detected with x-ray spectroscopy at the Dresden EBIT. The highest detected ion charge states of all elements introduced in the trap are inserted. The identification of the ion charge state was deduced from lines of DE and from RR processes.

The work was supported by the EFRE fund of the EU and by Freistaat Sachsen (Projects 5352/826 and 5353/826) and partially by DFG (Project Zs 14/7-1).

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# ABTEILUNG NUKLEARCHEMIE, UNIVERSITÄT ZU KÖLN, AND ZENTRUM FÜR STRAHLENSCHUTZ UND RADIOÖKOLOGIE, UNIVERSITÄT HANNOVER

## 1. Cross Sections for Radionuclide Production Relevant to Accelerator Driven Systems

J. Kuhnhenn[1], U. Herpers[1], W. Glasser[2], R. Michel[2], P.W. Kubik[3], M. Suter[4]

Investigations on residual nuclide production by proton-induced reactions on heavy target elements ($73 \leq Z \leq 83$) were continued with irradiation experiments up to 70 MeV at the injector cyclotron of PSI. These experiments are part of a project to measure integral cross sections for the production of residual nuclides by proton-induced reactions from thresholds up to 2.6 GeV for a wide variety of target elements. To obtain complete excitation functions, we combined experiments at various accelerators: LNS/Saclay ($E_p$ = 200 - 2600 MeV), PSI/Villigen ($E_p \leq$ 72 MeV), and TSL/Uppsala ($E_p$ = 70 - 180 MeV). The goals of this project are to provide a reliable data set for the modelling of cosmic ray interactions with matter and to satisfy some of the data needs of accelerator driven transmutation of nuclear waste (ADTW) [1] and accelerator driven energy amplification (ADEA) [2]. The experiments at PSI complete the previously measured excitation functions [3] in the lower energy range. Examples are given in Fig. 1.

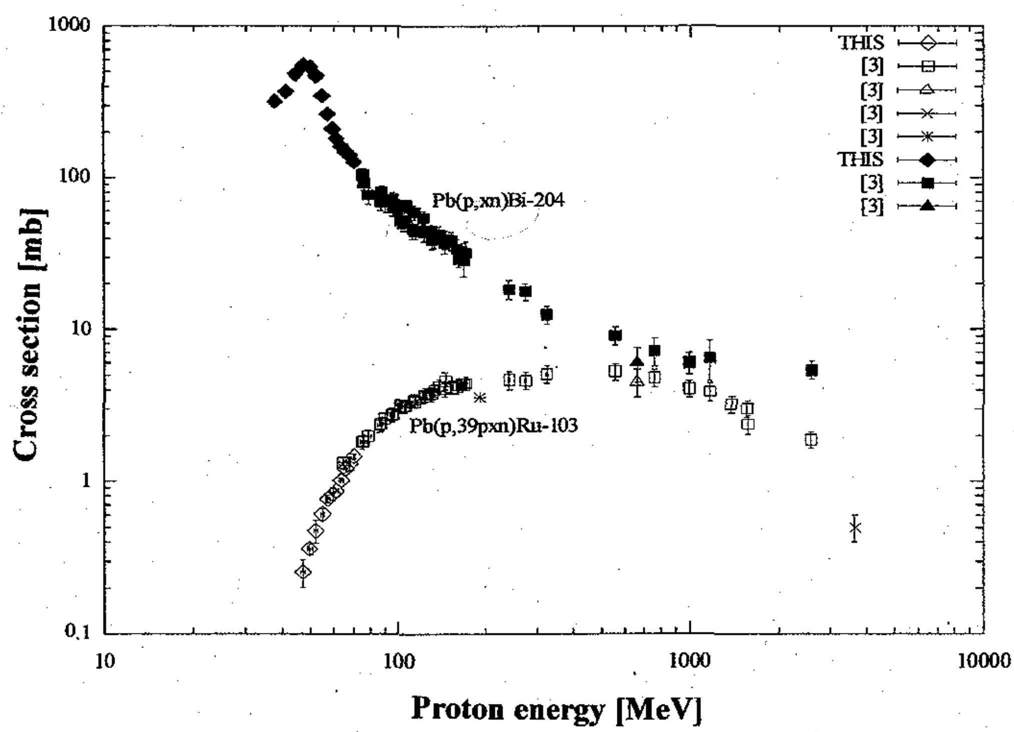


Fig. 1: Excitation functions for the formation of $^{103}$Ru and $^{204}$Bi from lead. For reference see Gloris et al. [3] and citations therein

The results obtained through our collaboration over more than twenty years represent worldwide the largest consistent data set of cross sections for residual nuclide production by proton-induced reactions. The entire data set for the target elements C, N, O, Mg, Al, Si, Ti, V, Mn, Fe, Co, Ni, Cu, Rb, Sr, Y, Zr, Nb, Mo, Rh, Ag, In, Te, Ba, Ta, W, Re, Ir, Au, Pb, and Bi covers nearly 1000 target/product combinations and contains about 22,000 cross sections. For a comprehensive survey on earlier work see [4]. With respect to the investigations relevant to ADTW and ADEA final results are available only for target element lead [3]. For the other target elements only preliminary data were reported so far [5,6].

In spite of these efforts, it will not be possible to measure all relevant nuclear reaction data needed to evaluate the feasibility of ADTW and ADEA technologies. Consequently, one will have to rely widely on theoretical model calculations. The quality of current nuclear reaction models and codes is, however, not sufficient to predict residual nuclide production with sufficient accuracy [7]. To improve this situation further experimental and theoretical work is needed covering radionuclide production and all other aspects of medium-energy nuclear reactions. A first step to such an approach is the ongoing EC Concerted Action *Physical Aspects of Lead as a Neutron Producing Target for Transmutation Devices*. It will be extended by an upcoming 5$^{th}$ Framework EC project HINDAS: *High and Intermediate Energy Nuclear Data for Accelerator Driven Systems.* In this European collaboration a complete and reliable experimental data base of all relevant nuclear reaction data for the target element lead is being established.

## 2. Cross Sections and Production Rates for the Production of Cosmogenic Nuclides

S. Merchel [5,1], T. Faestermann [6], U. Herpers [1], K. Knie [6], G. Korschinek [6], I. Leya [7], R. Michel[2], G. Rugel [6], C. Wallner [6]

About twenty years ago, we started systematic measurements of cross sections which are needed to understand the production by solar and galactic cosmic ray particles of the so-called cosmogenic nuclides in extraterrestrial matter. The cosmogenic nuclides measured in lunar samples and meteorites cover the radionuclides $^{48}$V, $^{51}$Cr, $^{37}$Ar, $^{7}$Be, $^{58}$Co, $^{56}$Co, $^{46}$Sc, $^{57}$Co, $^{54}$Mn, $^{22}$Na, $^{55}$Fe, $^{60}$Co, $^{3}$H, $^{44}$Ti, $^{32}$Si, $^{39}$Ar, $^{14}$C, $^{59}$Ni, $^{60}$Fe, $^{41}$Ca, $^{81}$Kr, $^{36}$Cl, $^{26}$Al, $^{10}$Be, $^{53}$Mn, $^{129}$I, and $^{40}$K. Moreover, stable cosmogenic nuclides can be observed as positive isotopic abundance anomalies of all rare gases He, Ne, Ar, Kr, and Xe. The relevance of target elements with respect to their contribution to the production of a cosmogenic nuclide depends on the relative abundances of the elements in extraterrestrial matter. Therefore, cosmochemically relevant target elements are restricted to atomic number $Z \leq 28$, with the exception of a few heavier elements such as Rb, Sr, Y, Zr, Nb, Te, and Ba.

All these elements were covered by our investigations; see [4] for a survey up to 1997 and [5,6] for more recent references. Residual nuclides with half-lives above a few hours were measured by γ-ray spectrometry, long-lived radionuclides ranging from $^{14}C$ to $^{129}I$ by accelerator mass spectrometry (AMS), and stable rare gas isotopes by conventional mass spectrometry.

Today, the data set of cross sections for the production of cosmogenic nuclides by proton-induced reactions is fairly complete [4]. Further progress is, however, manifested in highly sophisticated measurements of particular residual nuclides which were not investigated previously, either because of lack of analytical technology or because they became of interest only recently.

Among the new measurements is that on the excitation function for the production of $^{44}Ti$ ($T_{1/2}$ = 59.2 a) (Fig. 2) which provides a basis to investigate the solar modulation on a time scale of a few hundred years [8].

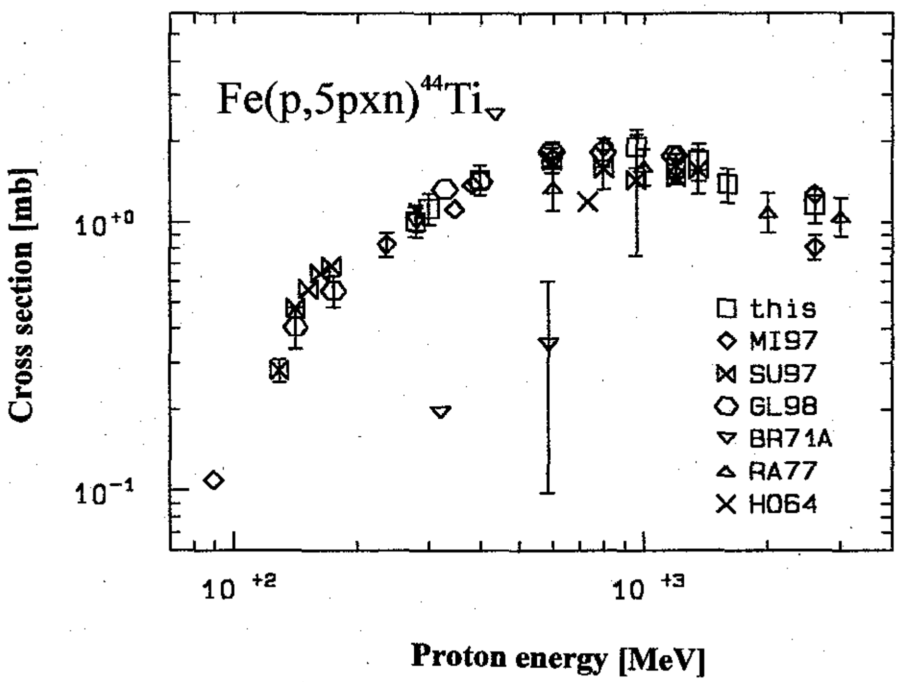


Fig. 2: Cross sections for the production of $^{44}Ti$ from iron. For references see [8].

AMS techniques were also improved and they allowed to measure radionuclides not accessible earlier. For $^{53}Mn$ ($T_{1/2}$ = 3.7 Ma) now AMS measurements are available (Fig. 3) [9] which finally settle the problem of modelling this cosmogenic radionuclide which, because of its long half-life, is of particular importance for the investigation of long-term constancy of the solar and galactic radiation [10]. AMS measurements of $^{60}Fe$ became feasible and first measurements of production cross sections (Fig. 4) and thick-target production rates [9,11] provided the basis for model calculations on its production rates in extraterrestrial matter. Since $^{60}Fe$ in meteorites is exclusively produced from nickel, it allows for an unambiguous interpretation of its abundances independent of the chemical composition of the object analysed [11].

Also thick-target production rates for $^{44}Ti$ from Ti, Fe and Ni, for $^{53}Mn$ from iron and nickel and for $^{60}Fe$ from nickel were measured [8,9,11] in targets from experiments in which artificial stony and iron meteoroids were irradiated with 1.6 GeV protons [12] in order to simulate the interaction of galactic protons with meteoroids. On the basis of the experimental thin-target cross sections and the thick-target production rates excitation functions on the neutron-induced production of these cosmogenic nuclides could be obtained by unfolding techniques [13]. The total information for the proton- and neutron-induced production provides the basis for a reliable modelling of the production rates in extraterrestrial matter of all these cosmogenic nuclides.

Further investigations are going on. First cross sections for the production of $^{41}Ca$ and $^{129}I$ were determined [17], but further measurements are underway. Procedures to measure $^{59}Ni$ have still to be worked out. Summarizing, our understanding of cosmogenic nuclide production in extra-terrestrial matter is excellent now for nearly all nuclides [18]. There still remain problems with the understanding of cosmic ray interactions with the terrestrial atmosphere and surface rocks. Here, further cross sections are needed.

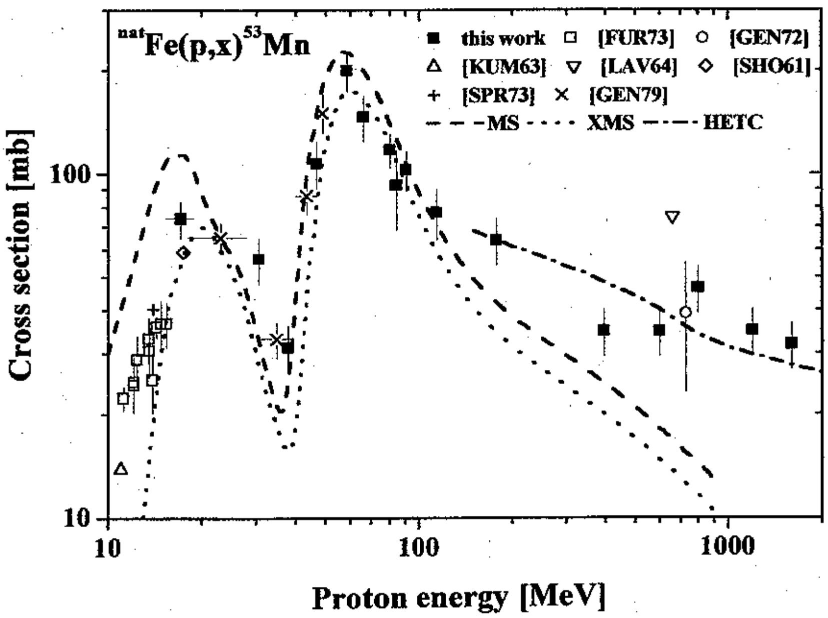


Fig. 3: Experimental cross sections from this and earlier work of the nuclear reaction $^{nat}Fe(p,x)^{53}Mn$. For references see [9].The theoretical calculations are based on the ALICE-IPPE (MS/XMS) [14] and HET-KFA2 [15] codes.

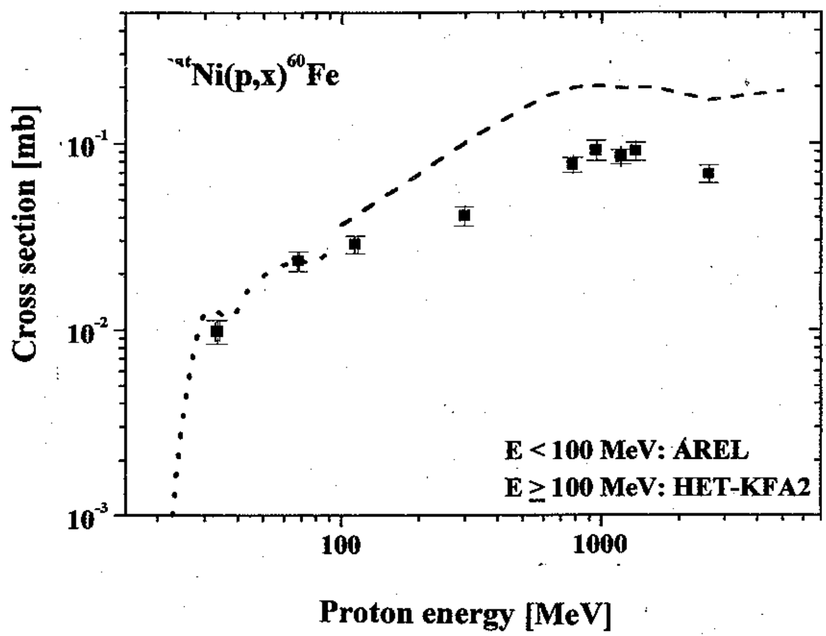


Fig. 4: Experimental cross sections of the nuclear reaction $^{nat}Ni(p,x)^{60}Fe$. The theoretical calculations are based on the AREL (MS/XMS) [16] and HET-KFA2 [15] codes.

## 3. Thin-Target Cross Sections for the Production of Residual Nuclides by Neutron-Induced Reactions

### 3.1 Neutron-Induced Production of I-129 and Other Radionuclides from Te at 14.7 MeV

C. Schnabel[1,4], I. Leya[7], R. Michel[2], J. Csikai[8], Z. Dezso[9], J.M. Lopez-Gutierrez[10], H.-A. Synal[3]

$^{129}I$ is a long-lived ($T_{1/2}$= 15.7 x $10^6$ a) cosmogenic radionuclide that can be used for example to determine exposure ages of iron meteorites above 10 Ma or to check the constancy of the galactic cosmic radiation on that time scale. Neutron (and proton) induced production cross sections have to be determined to model the cosmogenic production of $^{129}I$. Whereas a preliminary report on the determination of proton-induced production cross sections and thick target production rates was given elsewhere [17], this work represents the first step in the determination of neutron-induced production cross sections of $^{129}I$. Since in normal sized meteoroids the production of $^{129}I$ is dominated by the reaction $^{130}Te(n,2n)^{129}Te(\beta^-)$ $^{129}I$ and because the excitation function of that reaction has its maximum close to 15 MeV, a neutron generator using the D-T reaction is well suited to obtain important information on the production of $^{129}I$ from its main target element in meteoroids, tellurium. Furthermore, cross sections of twelve neutron-induced reactions on tellurium at 14.7 MeV have been determined from targets irradiated with 14.7 MeV neutrons at the Institute of Experimental Physics, Debrecen University, Debrecen.

The precursors of $^{129}I$ formed directly by the (n,2n) reaction on $^{130}Te$ are $^{129m}Te$ ($T_{1/2}$=33.6 d) and $^{129g}Te$ ($T_{1/2}$=69.6 min) which can be measured non-destructively by γ-ray spectrometry to determine the cross sections that lead to both precursors of $^{129}I$. The sum of the cross sections for these reactions, $^{130}Te(n,2n)^{129m}Te$ and $^{130}Te(n,2n)^{129g}Te$, is equal to the cross section for the production of $^{129}I$ from Te for neutrons (when other reaction channels can be neglected, which is the case at 14.7 MeV). After more than about six half-lives of $^{129m}Te$ a radiochemical separation of $^{129}I$ with addition of $^{127}I$ carrier combined with a determination of the $^{129}I/^{127}I$ ratio by accelerator mass spectrometry can also be used to determine the production cross section of $^{129}I$ from $^{130}Te$ for 14.7 MeV neutrons. A comparison between the results obtained by instrumental γ-ray spectrometry of $^{129m}Te$ and $^{129g}Te$ on the one hand and by radiochemical separation combined with accelerator mass spectrometry on the other hand can be employed to check both methods. The results for the production cross section of $^{129}I$ determined by the two methods agree within 5% (Fig. 5).

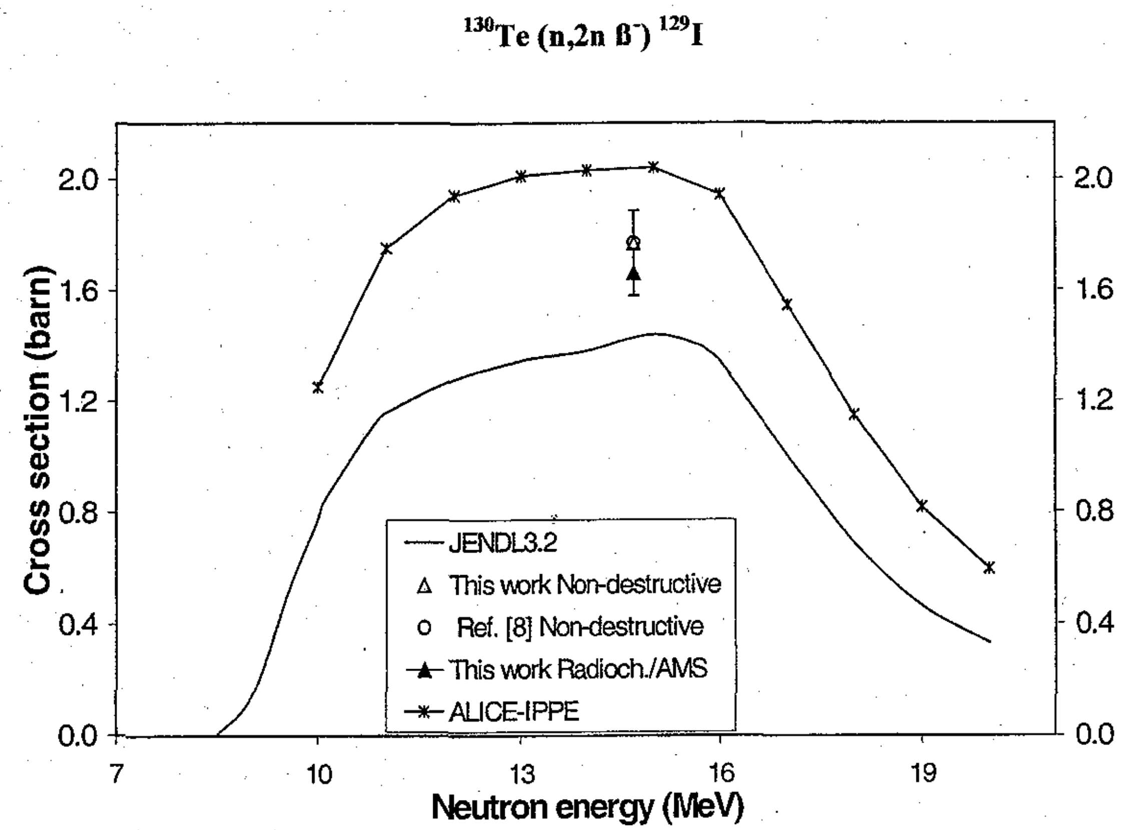


Fig. 5: The excitation function for $^{130}Te(n,2n)^{129}Te$ from JENDL3.2 [19] and the calculated value using ALICE-IPPE [14] compared with recent instrumental and radiochemical data. The previous measurement "[8] instrum." is from reference [20].

### 3.2 Activation Experiments with Medium-Energy Neutrons up to 180 MeV

R. Michel[2], W. Glasser[2], S. Neumann[2,11], U. Herpers[1], A.N. Smirnov[12], A.V. Prokofiev[12], P. Malmborg[13], O. Jonsson[13], H.Schuhmacher[11], H.J.Brede[11], V.Dangendorf[11], R. Nolte[11], J.P.Meulders[14]

Despite their importance for a wide variety of applications, the availability of respective neutron cross sections above 30 MeV is marginal and theoretical model calculations do not fulfil the accuracy requirements when predicting cross sections for residual nuclide production. A special solution for the problem of missing neutron cross sections was found for the case of the interactions of galactic cosmic ray protons with meteorites by unfolding neutron cross sections from experimental nuclide production rates measured in thick-target experiments [12]. A general solution for the problem, however, is to perform irradiation experiments with quasi mono-energetic neutrons produced by the $^{7}Li(p,n)^{7}Be$ reaction. Such quasi-monoenergetic neutron beams became available with sufficiently high beam currents to allow for activation experiments and for the determination of production cross sections at UCL [21] and TSL [22]. The UCL cyclotron facility provides such neutron beams in the energy range from about 25 MeV to 70 MeV and well-characterized neutron reference fields have been previously established [21]. At, TSL, a special irradiation chamber was installed [23] allowing for activation experiments for energies between 50 MeV and 180 MeV. The experiments performed at UCL and TSL are part of a research project to determine cross sections for the production of residual nuclides for neutron energies up to 180 MeV. The project was supported by UCL and TSL within the EC LIFE programme. Irradiation experiments were performed to activate target stacks containing the target elements C, N, O, Mg, Al, Si, Fe, Co, Ni, Cu, Ag, Te, and Pb.

At UCL, proton beams with energies of 36.4 MeV, 48.5 MeV, 62.9 MeV and beam currents up to 6 μA were used to irradiate 5 mm thick Li-targets. Special emphasis was laid on the establishment of monitoring cross sections for medium-energy neutrons. The target elements copper and silver are promising candidates for this purpose. For all experiments at UCL, the absolute neutron fluence was determined with three independent methods: a proton recoil telescope, a $^{238}U$ fission ionization chamber, and an NE213 liquid scintillation detector. The spectral fluence was measured with the TOF method, employed with the latter two systems. A specially developed multi-wire proportional counter yielded the transverse intensity profile of the collimated neutron beam. Measurements with and without the target stacks in the beam allowed to determine the attenuation of the neutrons in the stacks and to validate respective calculations of this attenuation.

At TSL, activation experiments with proton energies of 69.1 MeV, 78.1 MeV, 98.5 MeV, 136.7 MeV and 162.7 MeV were performed at special irradiation facility allowing for parasitic activation experiments. Further irradiation experiments with energies of 150 MeV and 180 MeV will be performed until the end of 2000 completing the experimental part of this project. Neutron fluences inside the target stack are measured using thin-film breakdown detectors. Measurements of the neutron peak fluence were carried out by means of thin-film breakdown counters [24].

Residual radionuclides with half-lives between 20 min and 5 a are investigated using off-line γ-ray spectrometry. Though the irradiations provide neutron fluences of about $10^{10}$ $cm^{-2}$ the activities obtained are low and low-level techniques have to be used over nearly one year to measure long-lived radionuclides. Also the evaluation of the experiments takes a long time and painstaking corrections for self-absorption and for systematic coincidences are necessary as consequences of relatively thick targets and close-to-detector counting geometries, respectively. First experimental response integrals of γ-emitting radionuclides are available and the feasibility of measurements by accelerator mass spectrometry of long-lived radionuclides, such as $^{10}Be$, $^{14}C$, $^{26}Al$, $^{36}Cl$, and $^{129}I$ has been demonstrated. However, the thin-target neutron cross sections cannot be directly calculated from these responses since the neutrons used are just "quasi-monoenergetic". Actually the targets are irradiated with a spectral neutron flux density with a significant contribution of low-energy neutrons. Only about 30% to 50% of the neutrons are observed in the high-energy peak with a width of a few MeV. The neutron cross sections have to be determined by unfolding from the experimental response integrals determined in a series of irradiation experiments with different neutron energies.

After completion of this series of experiments covering the energies from 30 MeV to 180 MeV, a set of complete neutron excitation functions from thresholds up to 180 MeV will be evaluated for more than 100 target-product combinations. It will provide a consistent set of neutron cross sections urgently needed for a wide variety of applications. These experiments contribute also to the EC Concerted Action "Physical Aspects of Lead as a Neutron Producing Target for Transmutation Devices" and will be continued in the upcoming HINDAS project.

Acknowledgement: The authors thank the authorities of PSI, LNS, TSL and UCL for the beam-time and the accelerator staffs for their cooperation and assistance. This work was supported partially by the Deutsche Forschungsgemeinschaft, by the Swiss National Science Foundation, by the European Union in the Human Capital and Mobility Program, in the LIFE and TEMPUS Programs, and in the Concerted Action LEAD FOR TRANSMUTATION.

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*Affiliations*

[1] Abteilung Nuklearchemie, Universität zu Köln, Köln, Germany
[2] Zentrum für Strahlenschutz und Radioökologie, Universität Hannover, Hannover, Germany
[3] Paul Scherrer Institut, c/o Institut für Teilchenphysik, ETH Hönggerberg, Zürich, Switzerland
[4] Institut für Teilchenphysik, ETH Hönggerberg, Zürich, Switzerland
[5] Max-Planck-Institut für Chemie, Mainz, Germany
[6] Fakultät für Physik, TU München, Garching, Germany
[7] Institut für Isotopengeologie und Mineralische Rohstoffe, ETH Zürich, Zürich, Switzerland
[8] Institute of Experimental Physics, University of Debrecen, Debrecen, Hungary
[9] Isotope Laboratory, University of Debrecen, Debrecen, Hungary
[10] Depto. Fisica Atomica, Molecular y Nuclear, Universidad de Sevilla, Sevilla, Spain
[11] Physikalisch Technische Bundesanstalt, Braunschweig, Germany
[12] Khlopin Radium Institute, St. Petersburg, Russia
[13] The Svedberg Laboratory, University Uppsala, Sweden
[14] Institut de Physique Nucléaire, Université Catholique de Louvain, Louvain La Neuve, Belgium

# PHYSIKALISCH-TECHNISCHE BUNDESANSTALT BRAUNSCHWEIG

## 1. Measurement of the Ti(n,x)$^{47}$Sc Cross Section between 7.5 and 14.3 MeV

W. Mannhart, D. Schmidt

Using a D(d,n) neutron source and samples of elemental titanium, the cross section leading to the production of the radionuclide $^{47}$Sc was determined in the energy range between 7.5 and 14.3 MeV. The neutron fluence was monitored with the $^{238}$U(n,f) reaction based on cross section data taken from ENDF/B-VI. Corrections for interfering neutrons from the D(d,np) breakup reaction were applied and were based on experimentally measured neutron source spectra [1].

The given cross section of Ti(n,x)$^{47}$Sc is defined as follows:

$$\sigma_{Ti(n,x)Sc\text{-}47} = \sigma_{Ti\text{-}47(n,p)Sc\text{-}47} + (h_{48}/h_{47}) \cdot \sigma_{Ti\text{-}48(n,np+d)Sc\text{-}47} \qquad (1)$$

with $h_x$ being the abundance of isotope x. Below 11.7 MeV the cross section is identical with that of the $^{47}$Ti(n,p)$^{47}$Sc reaction. Our data, according to eq. (1), are given in Fig. 1 and compared with the ENDF/B-VI evaluation and the International Reactor Dosimetry File (IRDF-90). Between 8 and 13.5 MeV our data form a detailed experimental set, describing the shape of the excitation function. Below 11 MeV, our data confirm the IRDF-90 evaluation. Around 14 MeV our data point to a substantial underestimation of the cross section in the ENDF/B-VI and the IRDF-90 evaluation. It seems that both evaluations have deficits as regards the proper description of the $^{48}$Ti(n,np+d)$^{47}$Sc cross section.

Our data are also compared with the data obtained in other experiments and taken from the EXFOR database. With a single exception, all these experiments were performed with samples of elemental titanium. Some of the experiments dealt with the cross section of one of the two components of eq. (1), assuming numerical values of the remaining component. These data were restored to the values originally measured. The data of all experiments were updated to the most recent values of the monitor cross section and of the radioactive decay parameters used.

The experiment of Firkin (1983) [2] covers measurements with the D(d,n) and T(d,n) neutron sources. The data below 12 MeV, obtained with the D(d,n) source, are systematically higher than our data by about 15%. The experiment of Ikeda (1991) [3] is of special interest as it uses another neutron source, viz. the $^{1}H(^{11}B,n)$ reaction. Both data of this experiment agree with our measurement within the uncertainties. Nevertheless, the data of ref. [3] are higher by 10% than our result. A possible explanation for this is an incomplete correction for low-energy neutrons in the $^{1}H(^{11}B,n)$ neutron field.

In the experiment of Ikeda (1988) [4] enriched isotopic samples were used and the cross sections of $^{47}Ti(n,p)^{47}Sc$ and $^{48}Ti(n,np+d)^{47}Sc$ were determined separately. According to eq. (1), the data of both reactions were combined in the comparison shown in Fig. 1. In the region of overlap, fair agreement is observed between the data of ref. [4] and our recent data.

## 2. Double Differential Neutron Emission Cross Sections of $^{51}V$ for Incident Neutron Energies between 8 and 14 MeV

D. Schmidt, W. Mannhart

After measurements on lead and chromium, the determination of double-differential neutron emission cross sections (DDX) was continued for $^{51}V$. In the incident neutron energy range of 8 to 14 MeV, monoenergetic neutron sources are not available. The D(d,n) neutron source used emits monoenergetic neutrons (DD neutrons) and a broad continuum stemming from the D(d,np) deuteron breakup reaction, both separated by an energy gap of about 6.5 MeV.

The neutron scattering spectra of both source fractions are partly superimposed; thus experimental separation is impossible. A complete simulation of the PTB's scattering experiment by the Monte Carlo (MC) technique makes it possible to calculate the

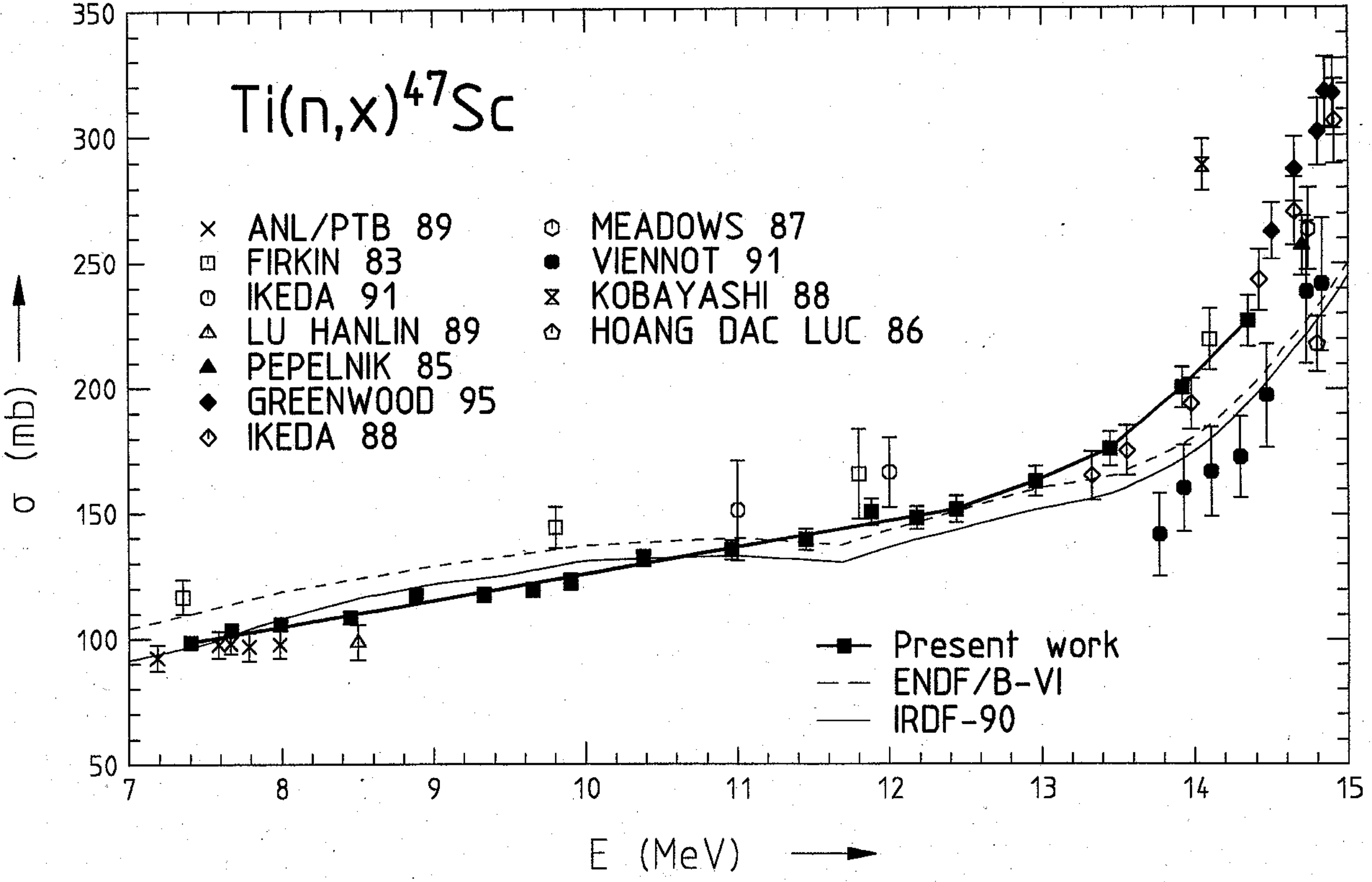


Fig. 1 Database of the $Ti(n,x)^{47}Sc$ reaction

scattered breakup neutrons separately. After subtraction, the DDX of the monoenergetic DD neutrons can be determined.

The database for the MC simulation is derived from the ENDF/B-VI evaluation, but the differential data are not sufficiently exact to simulate reliably the breakup scattering [5]. The data must, therefore, be adjusted. This was done on the basis of additional scattering measurements with the pure breakup reaction $^4$He(d,np). If such adjustment is carried out at different incident energies, the elastic and inelastic data needed for the simulation of the scattered breakup can be adjusted independently [6]. This is an essential improvement in comparison with former analyses.

Fig. 2 shows the angle-integrated neutron emission spectrum of $^{51}$V at 13.90 MeV. The measured spectrum, including the scattered breakup, has also been plotted demonstrating the large corrections. Both neutron emission spectra and angular distributions for the emission energy intervals selected were determined at 10 incident neutron energies between 7.99 MeV and 13.90 MeV.

The discrepancies between our results and the ENDF/B-VI evaluation are rather large, (see Fig. 2). Our data are also compared with measurements performed with the T(d,n) reaction [7], *i.e.* a pure monoenergetic neutron source. Differences in the shapes of the emission spectra at energies below 4 MeV may arise from an incorrect shape of the efficiency curve used in that measurement. It should be noted that our measurement at 14 MeV incident neutron energy was not made in competition to measurements with the T(d,n) neutron source. However, it offers the possibility to check the reliability of our correction procedure, even if the correction is large, and validates our corrections applied at neutron energies below 14 MeV.

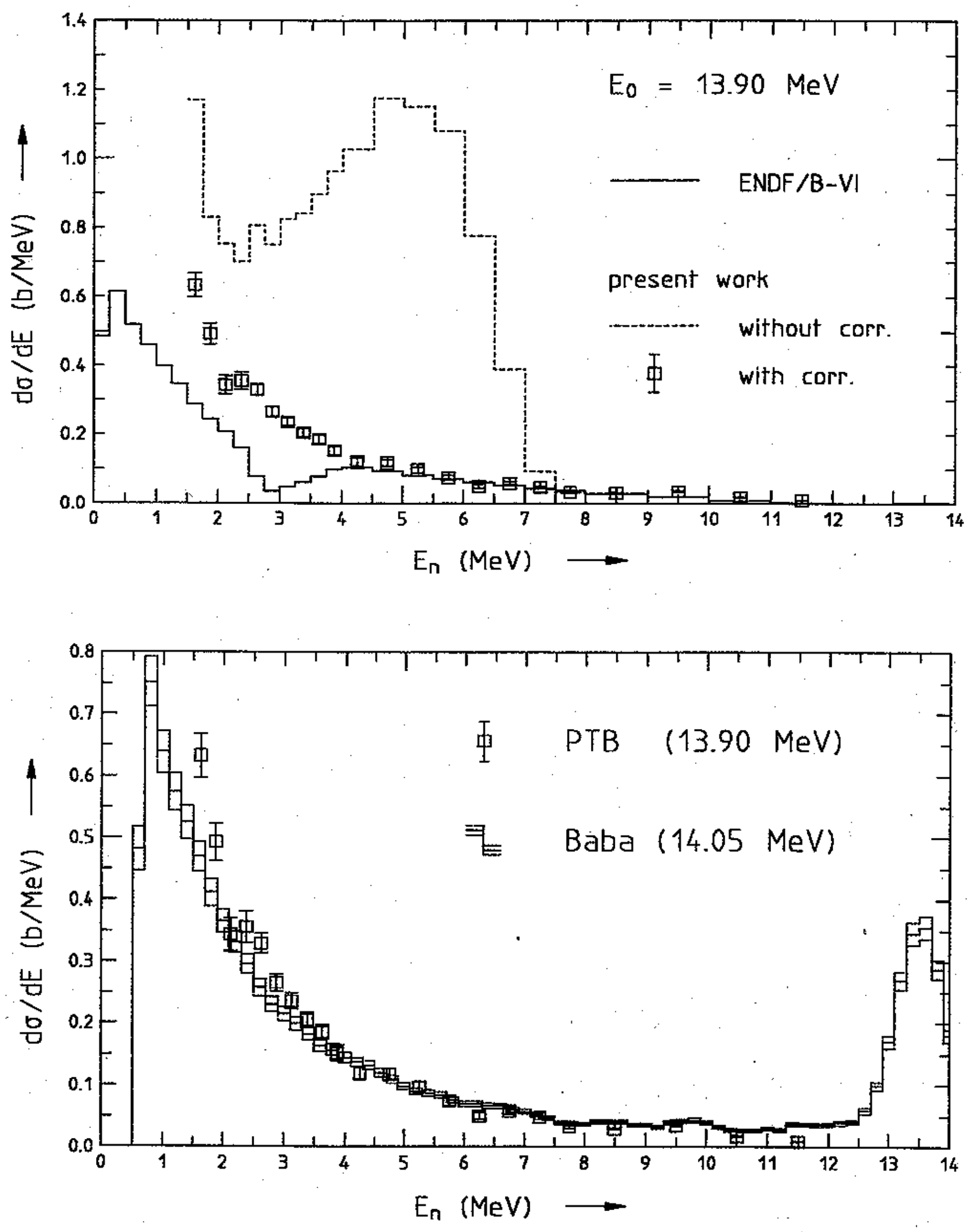


Fig. 2 Upper part: Measured neutron emission spectrum of $^{51}V$ at $E_0 = 13.90$ MeV incident neutron energy (squares); the experimental data without correction for scattered breakup neutrons (dashed line) and the ENDF/B-VI evaluation (solid line) are drawn for comparison.

Lower part: Comparison of measured neutron emission cross sections (PTB: present work) with those obtained using a 14 MeV monoenergetic neutron source (Baba and Ishikawa: [7]).

## 3. X-ray and Gamma-ray Emission Probabilities in the Decay of $^{237}$Np

U. Schötzig, E. Schönfeld, H. Janszen

X-ray and gamma-ray emission probabilities have been derived from emission rates of several sources, measured with efficiency-calibrated high-purity Ge and Si(Li) detectors, and from the activity, which was determined by $4\pi\alpha$ counting, by α-particle spectroscopy with defined solid angle and by liquid scintillation counting. Results are summarized in Table 1. Standard uncertainties are given in parentheses in terms of the last digit. For details see reference [8].

Table 1 Photon-emission probabilities *P* in the decay of $^{237}$Np → $^{233}$Pa → $^{233}$U.

| Energy in keV | Radiation | *P* (this work) |
|---|---|---|
| 3.15 | Pa-M (?) | 0.048(25) |
| 3.25 | U-M (?) | 0.015(8) |
| 8.22 | Np-γ | 0.0012(5) |
| 11.37 | Pa-$L_\lambda$ | 0.0155(8) |
| 11.62 | U-$L_\lambda$ | 0.0118(7) |
| 13.27 | Pa-$L_\alpha$ | 0.260(28) |
| 13.60 | U-$L_\alpha$ | 0.157(17) |
| 14.95 | Pa-$L_\eta$ | 0.0064(6) |
| 15.4 | Pa-$L_{\beta6}$; U-$L_{\beta6}$, U-$L_\eta$ | 0.0066(5) |
| 16.0 | Pa-$L_{\beta2,15}$, Pa-$L_{\beta4}$ | 0.0791(29) |
| 16.6 | Pa-$L_{\beta1}$, $L_{\beta5}$; U-$L_{\beta2,15}$, $L_{\beta4}$ | 0.274(11) |
| 17.2 | Pa-$L_{\beta3}$; U-$L_{\beta1}$, $L_{\beta3}$, $L_{\beta5}$ | 0.115(5) |
| 19.50 | Pa-$L_{\gamma1}$ | 0.0465(17) |
| 20.16 | Pa-$L_{\gamma2}$, $L_{\gamma3}$, $L_{\gamma6}$; U-$L_{\gamma1}$ | 0.0349(13) |
| 20.49 | U-$L_{\gamma2}$ | 0.00806(29) |
| 20.7 | U-$L_{\gamma3}$; U-$L_{\gamma6}$ | 0.0103(4) |
| 21.5 | | 0.00356(13) |
| 27.7 | | 0.0084(7) |
| 29.37 | Np-γ | 0.141(15) |
| 46.53 | Np-γ | 0.00104(4) |

| Energy in keV | Radiation | $P$ (this work) |
|---|---|---|
| 57.1 | Np-γ, Pa-γ | 0.00354(8) |
| 75.35 | Pa-γ | 0.0138(4) |
| 86.48 | Np-γ; Pa-γ | 0.141(3) |
| 86.81 | Pa-γ | |
| 92.28 | Pa-$K_{\alpha 2}$ | 0.0182(5) |
| 94.65 | U-$K_{\alpha 2}$; Np-γ | 0.097(3) |
| 95.86 | Pa-$K_{\alpha 1}$ | 0.0298(7) |
| 98.43 | U-$K_{\alpha 1}$ | 0.144(4) |
| 103.97 | Pa-γ | 0.00844(17) |
| 107.6 | Pa-$K_{\beta 3}$ | 0.00249(8) |
| 108.2 | Pa-$K_{\beta 1}$; Np-γ | 0.00864(19) |
| 108.7 | Np-γ | |
| 109.1 | Pa-$K_{\beta 5}$; Np-γ | |
| 110.4 | U-$K_{\beta 3}$ | 0.0190(5) |
| 111.4 | Pa-$K_{\beta 2}$; Pa-$K_{O;P2,3}$; U-$K_{\beta 1}$ | 0.0384(9) |
| 114.9 | U-$K_{\beta 2}$; U-$K_{\beta 4}$ | 0.0159(9) |
| 115.19 | Np-γ; U-$K_{O;P2,3}$ | 0.00332(10) |
| 117.70 | Np-γ | 0.00169(4) |
| 131.10 | Np-γ | 0.000857(22) |
| 134.28 | Np-γ | 0.000670(28) |
| 143.25 | Np-γ | 0.00443(8) |
| 151.41 | Np-γ | 0.00232(24) |
| 155.24 | Np-γ | 0.000889(18) |
| 162.41 | Np-γ | 0.000327(12) |
| 169.19 | Np-γ | 0.000633(19) |
| 176.12 | Np-γ | 0.00012(4) |
| 180.81 | Np-γ | 0.000158(10) |
| 191.46 | Np-γ | 0.000192(12) |
| 193.26 | Np-γ | 0.000437(10) |
| 194.95 | Np-γ | 0.00177(5) |
| 196.86 | Np-γ | 0.000208(12) |
| 201.6 | Np-γ | 0.000393(9) |
| 202.9 | Np-γ | |
| 209.19 | Np-γ | 0.000142(9) |
| 212.29 | Np-γ | 0.00151(3) |
| 214.01 | Np-γ | 0.000362(8) |
| 237.86 | Np-γ | 0.000569(6) |
| 248.5 | Pa-γ | 0.000618(11) |

| Energy in keV | Radiation | $P$ (this work) |
|---|---|---|
| 248.90 | Pa-γ | |
| 258.46 | Pa-γ | 0.000274(6) |
| 262.44 | Np-γ | 0.0000471(18) |
| 271.48 | Pa-γ | 0.00323(4) |
| 279.65 | Np-γ | 0.000109(4) |
| 288.3 | | 0.000164(5) |
| 300.34 | Pa-γ | 0.0655(7) |
| 312.17 | Pa-γ | 0.385(4) |
| 340.81 | Pa-γ | 0.0450(5) |
| 375.45 | Pa-γ | 0.00686(7) |
| 398.62 | Pa-γ | 0.01406(15) |
| 415.76 | Pa-γ | 0.01765(18) |

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# APPENDIX

## Addresses of Contributing Laboratories

Institut für Kernphysik III
Director: Prof. Dr. G. Schatz
Reporter: Dr. F. Käppeler
Forschungszentrum Karlsruhe
Postfach 36 40
**76021 Karlsruhe**

Institut für Kern- und Energietechnik
Director: Prof. Dr. U. Müller
Reporter: Dr. U. Fischer
Forschungszentrum Karlsruhe
Postfach 36 40
**76021 Karlsruhe**

Institut für Nuklearchemie
Director: Prof. Dr. H.H. Coenen
Reporter: Prof. Dr. Dr. h.c. S.M. Qaim
Forschungszentrum Jülich
Postfach 1913
**52425 Jülich**

Institut für Kern- und Teilchenphysik
Director: Prof. Dr. B. Spaan
Reporter: Prof. Dr. K. Seidel
Technische Universität Dresden
Mommsenstr. 13
**01062 Dresden**

Zentrum für Strahlenschutz und Radioökologie
Head and reporter: Prof. Dr. R. Michel
Universität Hannover
Am Kleinen Felde 30
**30167 Hannover**

Abteilung Nuklearchemie
Head: Prof. Dr. H.H. Coenen
Reporter: Dr. U. Herpers
Universität zu Köln
Otto-Fischer-Straße 12 - 14
**50674 Köln**

Physikalisch-Technische Bundesanstalt
Abteilung Ionisierende Strahlung
Director: Prof. Dr. G. Dietze
Reporter: Dr. W. Mannhart
Bundesallee 100
**38116 Braunschweig**