JRC TECHNICAL REPORT

JRC data for the Ti-48 standard

Summary report on the JRC contributions to the establishment of a new γ ray standard with the ^{48}Ti(n,n'γ) reaction

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The picture on the front cover is the Titan Atlas carrying the world. It alludes to the element titanium that is the subject of this work and is technologically renowned for the strength it conveys to constructions.. Here it serves as the potential basis of a new nuclear data standard.

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Abstract

A measurement of the ^{48}Ti(n,n'γ)^{48}Ti reaction was performed at the GELINA neutron source of EC-JRC-IRMM using the GAINS γ spectrometer with the purpose of establishing a new γ-ray standard for neutron induced cross section measurements. A natural target was used and the γ-production cross section was measured for 10 transitions in the neutron energy range 0-18 MeV. The lowest achieved uncertainty was 4.8%..

Executive Summary

Neutron data standards are accurately determined cross sections for neutron induced reactions that are readily employed to measure the neutron fluence rate at a well-defined position using a method that is tailored to the prevailing conditions.

Updates to the standard cross sections are made to reflect new experimental results aiming at improving the accuracy of the existing standards and for extending their range of application. The last update was made in 2007 [1]. This report is a contribution to the IAEA standards evaluation that will be released in 2016/17. Here, we summarize recent results obtained at the JRC accelerator based neutron source GELINA concerning a potential new standard reaction suitable for fluence measurement in gamma-ray emission reactions induced by fast neutrons. The particular measurement of the ^{48}Ti(n,nγ)^{48}Ti reaction was performed at the GELINA neutron source of EC-JRC-IRMM using the GAINS gamma spectrometer with the purpose of establishing a new γ-ray standard for neutron induced cross section measurements. A natural target was used and the gamma-production cross section was measured for 10 transitions in the neutron energy range 0-18 MeV. The achieved uncertainty was 4.8% for the main γ-ray of interest and in the incident neutron energy range where this potential standard is most suitable. The data were delivered to the standards evaluation working group of the IAEA.

1 JRC data for the Ti-48 standard

The investigations of neutron-induced reactions on ^{48}Ti are very important for various reasons. The first motivation to perform the experiment is the presence of ^{nat}Ti in nuclear reactors. Other important motivation is the fact that the first transition in ^{48}Ti with E_γ=983.5 keV is a candidate for a reference cross section [1]. Many investigations were performed during the last years in order to establish a recognized γ-ray reference cross section for neutron-induced reactions. Often used reference transitions were from ^{56}Fe (E_γ=847 keV) and ^{52}Cr (E_γ=1434 keV) but both of them have some important issues. The main disadvantages of the measurements of ^{56}Fe(n,n'γ_{847}) are: the contribution from the (n,p) reaction on the sample that creates ^{56}Mn which through β^- decay populates the 847-keV level in ^{56}Fe, the background contribution coming from the iron present in the components of the experimental setup, and also the non-isotropic angular distribution of the γ rays. The disadvantages for ^{52}Cr(n,n'γ_{1434}) are similar as for ^{56}Fe(n,n'γ_{847}) but the main difficulty is to produce the sample [1].

One of the advantages of using ^{48}Ti (the most abundant isotope of ^{nat}Ti) is the fairly constant large cross section over a broad neutron energy range. Other advantages are the relatively low price of the isotope and also the simple preparation of the sample. The disadvantages in this case are the low number of experimental results, some of them discrepant, the angular anisotropy and the contribution from the ^{48}Ti(n,p)^{48}Sc at $E_n > 5$ MeV. This reaction creates ^{48}Sc which through β^- decay emits γ rays with E_γ=983.5 keV, 1037.5 keV, 1212.9 keV and 1312.1 keV. We performed simple investigations in order to see if we find a sub threshold contribution in the cross sections of the corresponding γ from the inelastic scattering. We did not observe any contributions and this is in a good agreement with the TALYS 1.6 code which predicts small values of the (n,p) cross sections in comparison with the (n,n') cross sections.

In the following a short overview of the neutron inelastic cross section measurements on ^{48}Ti will be presented. In 1969, M. W. Pasechnik *et al.* were the first to report an experimental value of the 984-keV level cross section in ^{48}Ti. They used neutrons with incident energies of 2.9 MeV in order to perform an experiment of inelastic scattering on titanium and chrome nuclei at a Van de Graaff accelerator. For the measurement they used the time of flight method and neutron detection using organic scintillators [2]. Soon after, W. Breunlich and G. Stengel determined the (n,n'γ) cross section for neutrons with E_n=14.4 MeV in the mass range A=46-88. The experiment was performed at a pulsed Cockroft-Walton accelerator using natural samples. For the γ-ray detection they used a coaxial Ge(Li) detector. The γ-production cross sections for the first transitions in ^{46}Ti, ^{48}Ti and ^{50}Ti were reported [3]. In 1973 E. S. Konobeevskij *et al.* performed a neutron inelastic scattering experiment at a Van de Graaff accelerator using a metallic sample with natural isotopic composition and determined the level population cross section for the first excited level in ^{46}Ti and ^{48}Ti in the neutron energy range near the threshold. For the γ-ray detection they used Ge(Li) detectors [4]. In the same year, W. E. Kinney and F. G. Perey performed a neutron inelastic scattering experiment at the Van de Graaff accelerator of the Oak Ridge National Laboratory. Using the time of flight method they determined the cross sections for the first four excited states in ^{48}Ti in the energy range from 4.07 to 8.56 MeV [5]. In 1975, I. A. Korzh *et al.* reported the cross sections of the first three excited states in ^{48}Ti [6]. Later, in 1994, A. I. Lashuk *et al.* performed an experiment at a Van de Graaff accelerator in order to determine the cross sections for several excited states of ^{48}Ti. For the γ-ray detection they used a Ge(Li) detector [7]. All these experiments resulted in a rather limited number of data points.

In 2007 D. Dashdorj *et al.* performed a neutron inelastic scattering experiment using a 99.8% enriched sample of ^{48}Ti and reported the γ-production cross sections for neutron energies in the range from 1 to 200 MeV. The experiment was performed at the Weapons Neutron Research (WNR) facility at the Los Alamos Neutron Science Center (LANSCE) using a 800 MeV proton beam inducing spallation reactions on a tungsten target as a neutron source. The γ rays were detected using the Gamma Array for Neutron Induced Excitations (GEANIE) Spectrometer. The neutron flux was monitored with a ^{235,238}U fission chamber [8]. This experiment was the first to cover a wider energy range with a better but still limited energy resolution. Our experimental results will extensively be compared with those reported by D. Dashdorj *et al.* in Ref. [8].

1.1 Experimental setup

The present neutron inelastic scattering experiment was performed at the time of flight facility GELINA (Geel Electron Linear Accelerator), on flight path 3 at a distance of 198.684 m from the neutron source. For γ-ray detection we used the GAINS spectrometer (Gamma Array for Inelastic Neutron Scattering), with 12 HPGe detectors (10 were operational during the experiment) placed at 110°, 150° and 125° (see Fig. 1). The detectors are located about 17 cm from the center of the sample. The neutron flux measurement was performed using a ^{235}U fission chamber and the total neutron flux in the measurement station was about 500 n/cm^2/s [9]. The experimental setup is the same as the one described in Ref. [9] with one exception: the U filter on the beam line used to suppress the γ flash was missing during the measurements. The sample used in this experiment was ^{nat}Ti with a diameter of 8.000(1) cm, 0.45 cm thickness and 99.995% purity. ^{nat}Ti has five stable isotopes and ^{48}Ti is the most abundant. The areal density of the sample was 2.136(71) g/cm^2 deduced from the measured mass and area. The mass of the sample was 107.520(1) g. Table 1 displays the abundance of each isotope and the corresponding areal density. The sample was irradiated for 429 hours. The γ spectrum from one of the detectors is displayed in figure 2. In order to calculate the γ-production cross sections, the level cross sections and the total inelastic cross section, the primary data were analyzed as described in Ref [9]. The procedure used to calculate the efficiency of the detectors is described in Ref [11] and consist of a method combining calibration measurements and MCNP5 [12] simulations. The calibration measurements were performed using a ^{152}Eu point-like source with an activity of 18.6(2) kBq. The efficiency of the fission chamber was 85.5(4)% and was calculated as described in Refs [13, 14]. The yields of the fission chamber were added for all the weeks of the experiment and a double-smooth procedure (each smooth consisting in a 61 channels second-order polynomial fit) was applied.

Table 1: The isotopic abundance of each stable isotope of ^{nat}Ti [10] and the corresponding areal density.

Isotopes	^{46}Ti	^{47}Ti	^{48}Ti	^{49}Ti	^{50}Ti
Isotopic composition (%)	8.25(3)	7.44(2)	73.72(3)	5.41(2)	5.18(2)
Areal density (g/cm^2)	0.176(1)	0.159(1)	1.576(1)	0.115(1)	0.110(1)

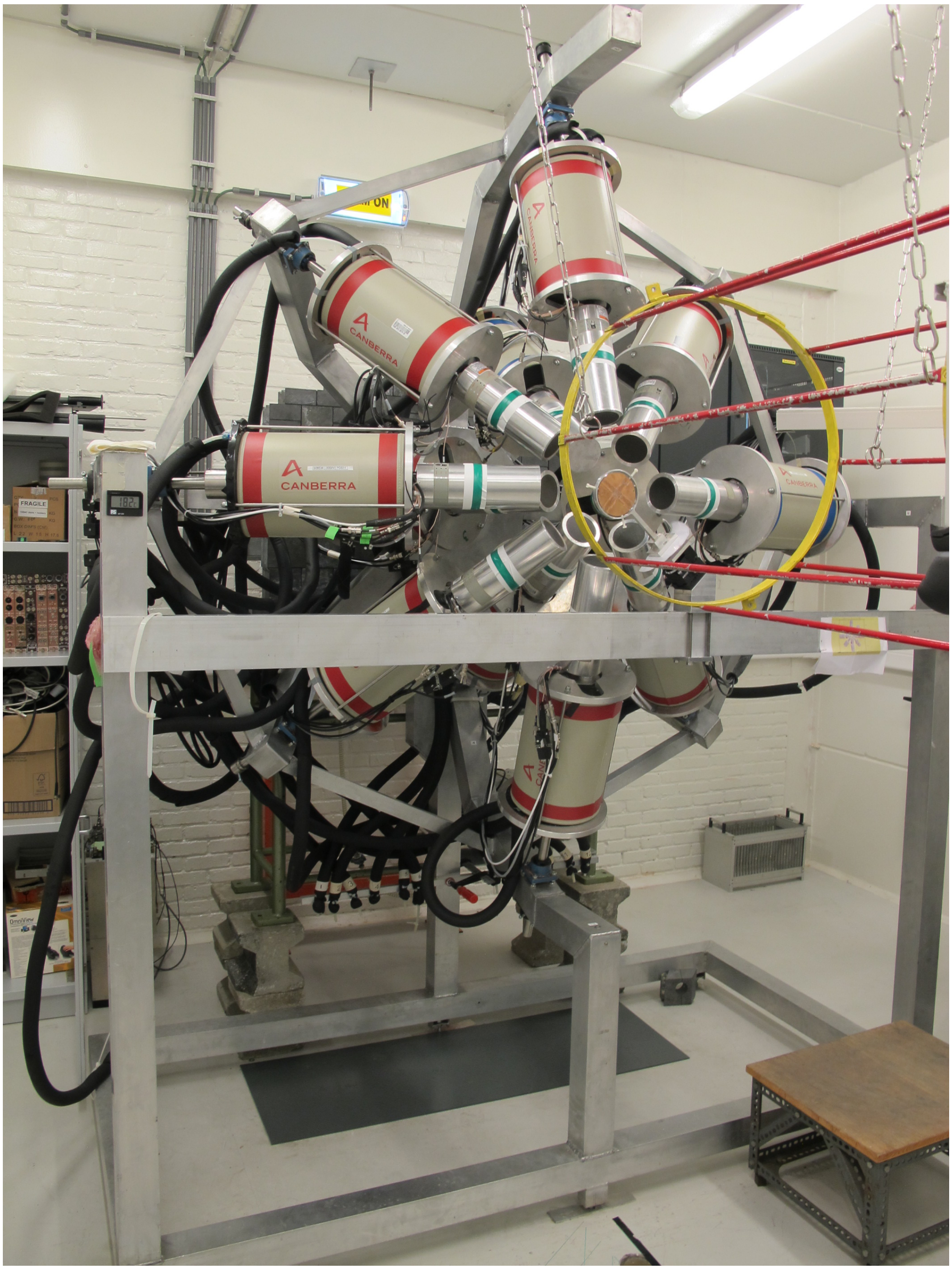

Figure 1: The GAINS spectrometer.

The experimental cross sections were corrected for the multiple scattering of the neutrons in the sample. If a neutron suffers multiple scattering in the sample, the effective flux is altered while the time of the (n,n'γ) event will not correspond to the energy of the incident neutron anymore. The correction factor for this effect is calculated through Monte Carlo simulations (MCNP5) as the ratio between the reaction rate in the sample for the full geometry and the reaction rate when the materials in the beam following the fission chamber are absent.

Because ^{nat}Ti has five stable isotopes with mass numbers from A=46 up to A=50 and the incident neutron spectrum included energies higher that the threshold energy for the

$^{47-50}$Ti(n,2n) reaction channel, we observed in the inelastic scattering peaks the contribution from the corresponding (n,2n) channel.

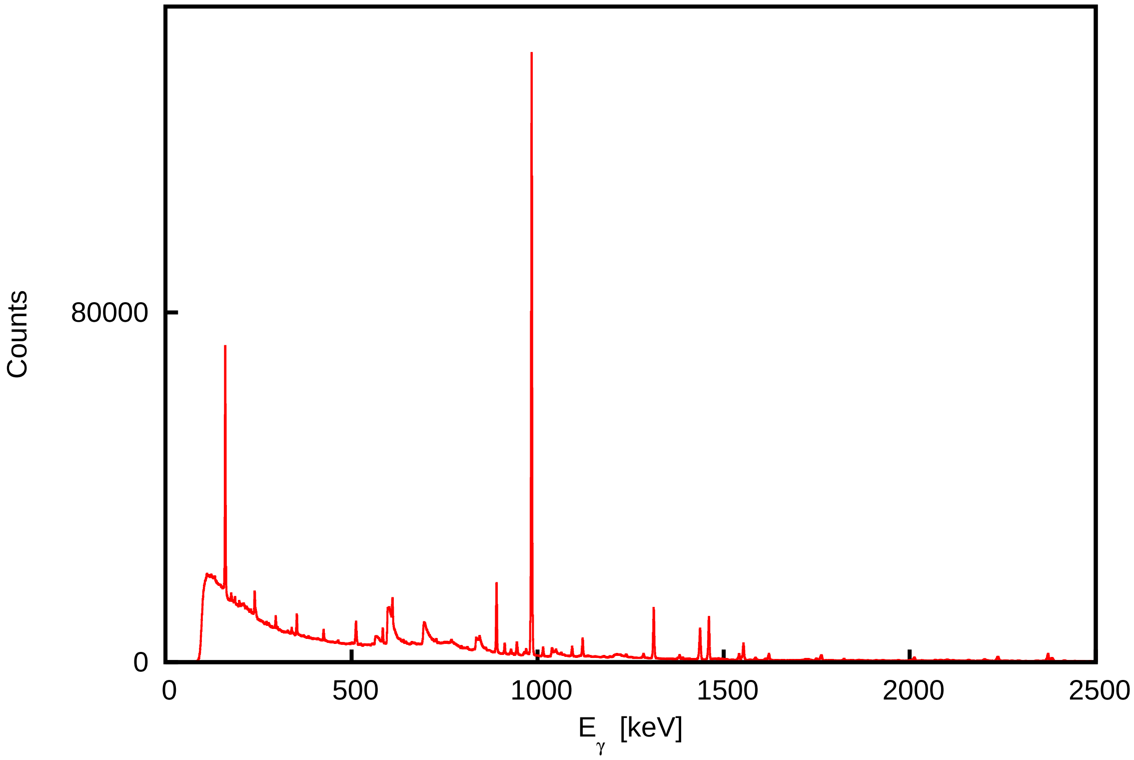

Figure 2: The γ-ray spectrum from one of the HPGe detectors.

1.2 Theoretical calculations performed with the TALYS 1.6 code

A good knowledge of the reaction mechanisms and decay possibilities of the excited nuclei is needed in order to model the different reactions. Several codes are used for the modeling such as EMPIRE [15] and TALYS [16]. In this work the experimental results are compared with TALYS 1.6 calculations using two features: the "default" calculations and the "microscopical" calculations. Both features will be shortly presented in the following.

TALYS is a computer code system used for the simulation of nuclear reactions. It can be used in the incident energy range from keV to 200 MeV and for targets with the mass number larger than 12. The projectile and ejectiles can be n, γ, p, d, t, h and α. It uses modern nuclear models such as the optical model, direct reactions, compound nucleus reactions, pre-equilibrium reactions, fission reactions and level densities. The total and partial cross sections, residual production cross sections and recoils, energy spectrum angular distributions of γ rays and double-differential spectra can be calculated due to its large nuclear structure database. The goal of TALYS is to use theoretical codes in order to predict nuclear reaction cross sections for cases where no or partial experimental data are available. In order to perform the theoretical calculations several parameters can be selected that were obtained from global optimization of semi empiric and microscopic description. As we pointed out before two features of the TALYS code were used: the "default" and the "microscopical" calculations.

The "TALYS default" calculations involve the semi-empirical model with parameters obtained from global optimizations. The optical model potentials are the local and global parametrization of Koning and Delaroche [17] while the level densities are calculated in the Gilbert and Cameron approach [18]. The back shifted Fermi gas model with an energy-dependent level-density parameter a that accounts for the damped shell effect proposed by Ignatyuk *et al.* [19] is used for high energies. For low energies it uses the constant temperature model. The gamma-ray strength functions are described using the Brink-Axel option for all transition types other than E1 while for the E1 radiation the generalized Lorentzian form of Kopecky and Uhl [20] is used. TALYS also relies on a nuclear structure and decay table derived from the Reference Input Parameter Library [21] in order to describe the de-excitation of the nuclei for the first 20 excited levels in the target and residual nuclei.

The calculations labeled "TALYS microscopic" were performed with the semi-microscopic nucleon-nucleus spherical optical model potential as described in Ref. [22]. It uses the microscopic optical model of E. Bauge *et al.* [23] obtained from nuclear densities and Jeukenne-Lejeune-Mahaux optical model potential for nuclear matter. Usually this model has good predictions for nuclei with $A > 30$ and for incident energies ranging from 10 keV up to 200 MeV. The level densities are calculated based on the microscopic combinatorial model proposed by Hilaire and Goriely [24]. Using the nuclear structure properties determined within the deformed Skyrme-Hartree-Fock-Bogolyubov framework the model includes a detailed microscopic calculation of the collective enhancement and the intrinsic state density. The gamma-ray strength functions were calculated in the Hartree-Fock-Bogolyubov approach.

1.3 Results and discussions

As we already pointed out ^{48}Ti is the most abundant isotope in ^{nat}Ti representing 72.73% in the isotopic composition. Ten γ-ray transitions were observed from the inelastic scattering of the neutrons on ^{48}Ti (see figure 3). For each of them we calculated the differential γ production cross section (see figure 4) and the integral γ-production cross section (see figure 5). From figure 4 one can observe the small angular anisotropy that was pointed out at the beginning of the report. The contribution from the ^{49}Ti(n,2n) reaction is insignificant because the ratio between the isotopic composition of ^{49}Ti and ^{48}Ti is small. We compared the experimental results with theoretical calculations performed with the TALYS 1.6 code using the default input parameters and the microscopical model and also with previously reported results. We also calculated the γ production cross section for the sum of the two contributions ((n,n) and (n,2n)) from the TALYS "microscopical" calculations and we compared the theoretical values with the experimental results. The uncertainties related to the experimental cross sections will be discussed at the end.

This section presents the results of the integral γ-production cross section for each transition observed from the ^{48}Ti(n,n'γ)^{48}Ti reaction. The results are compared with TALYS calculations and with previous results reported by D. Dashdorj *et al.* [8], E. S. Konobeevskij *et al.* [4].

The 983.5-keV γ-production cross section The first observed transition in ^{48}Ti has E_γ=983.5 keV and E2 multipolarity. It represents the major contribution to the total inelastic cross section. The comparison of our experimental results with the TALYS calculations (performed with default and microscopical input parameters) and previous experimental re-

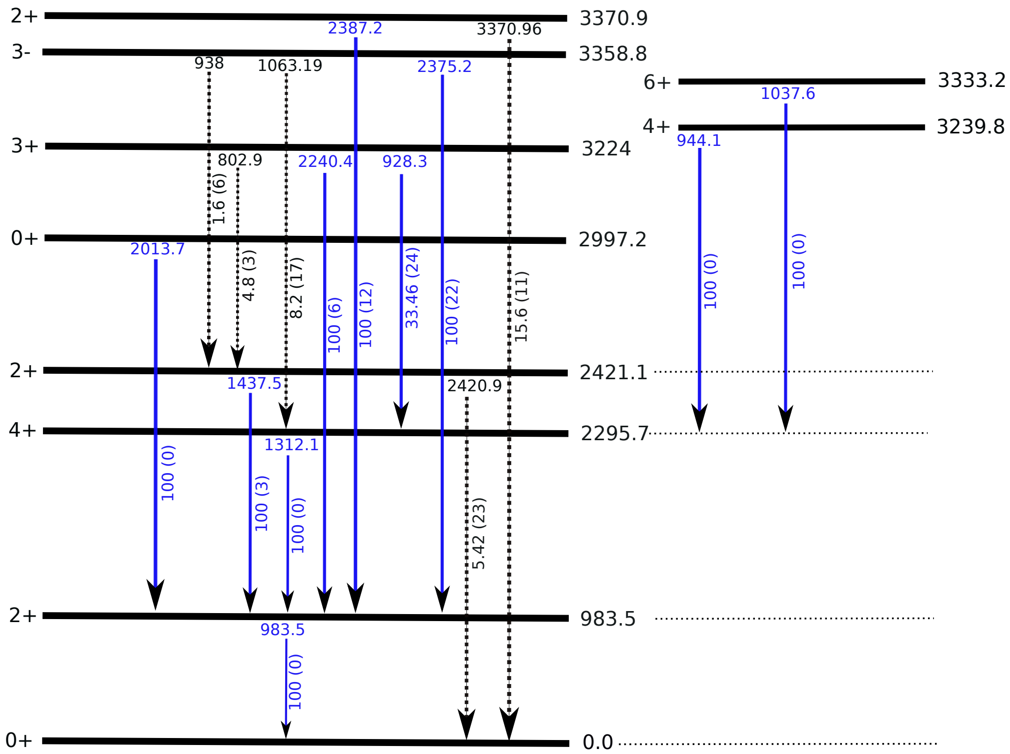

Figure 3: The low excitation energy level scheme [25] for ^{48}Ti. The transitions displayed with a continuous blue line were examined in the present experiment.

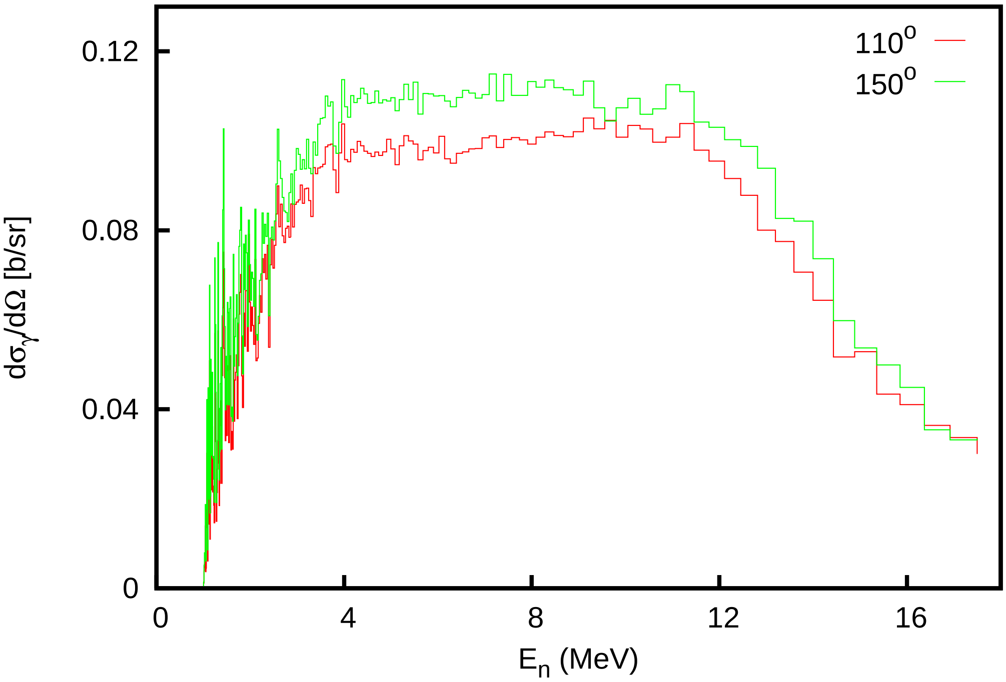

Figure 4: The differential production cross section for the transition with E_γ=983.5 keV.

sults reported by D. Dashdorj *et al.* [8], E. S. Konobeevskij *et al.* [4] and W. Breunlich *et al.* [3] is displayed in figure 5 panel a. Our experimental values are in a good agreement with the results reported by D. Dashdorj *et al.* and by E. S. Konobeevskij *et al.*. The value

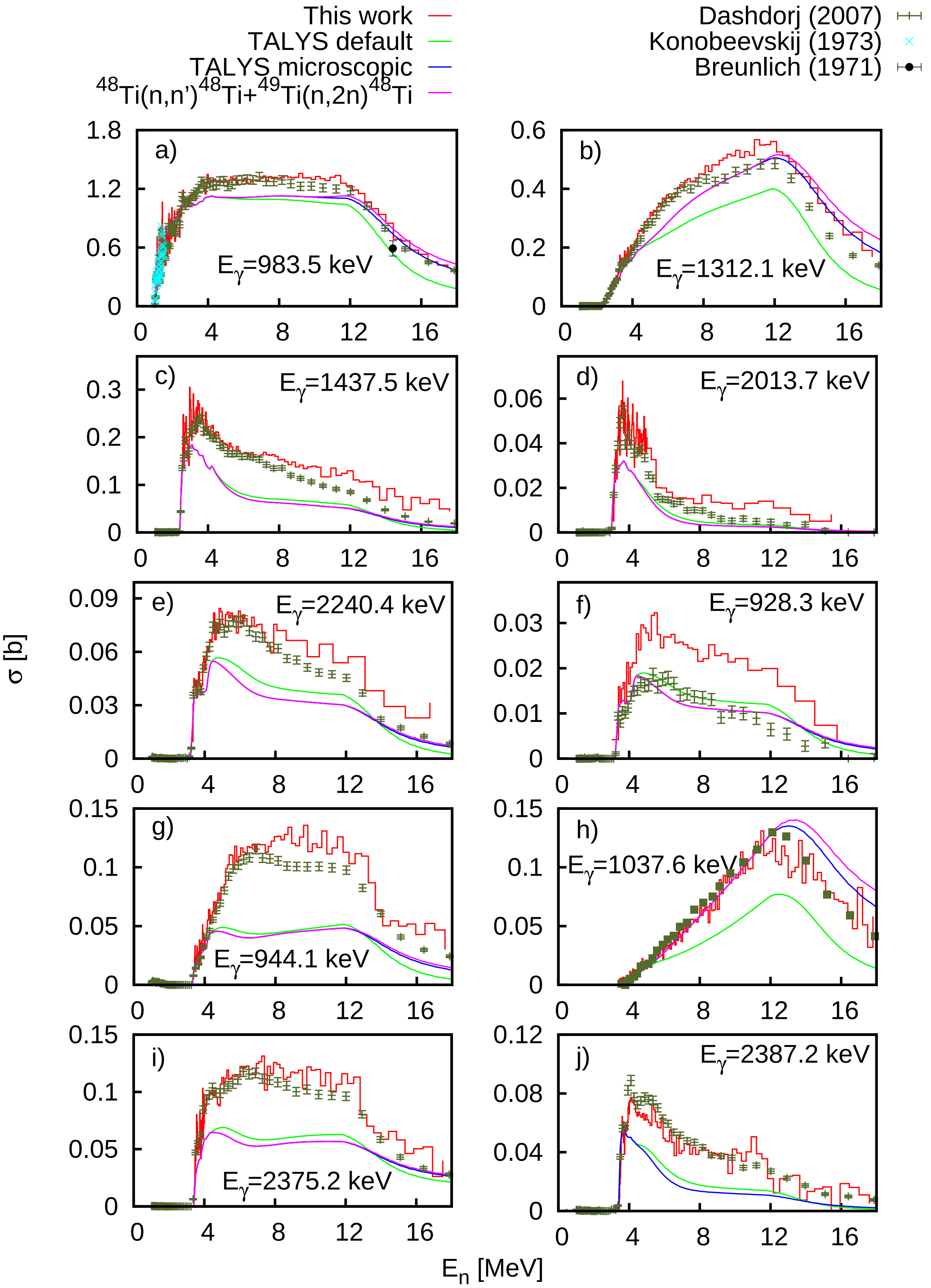

Figure 5: The experimental γ-production cross sections of the transitions observed from the inelastic scattering of the neutrons on ^{48}Ti compared with the theoretical calculations performed with TALYS 1.6 code and previous reported results.

reported by W. Breunlich *et al.* for the cross section at 14.4 MeV is lower that our experimental value. The theoretical calculations using the microscopical model describe fairly well our experimental results up to 3 MeV and beyond 12 MeV. Between this 3-12 MeV range the theoretical values underestimate the experimental ones. The theoretical calculations using the default input parameters are in a good agreement with our results up to 3 MeV but above this energy our values are clearly higher.

The 1312.1-keV γ-production cross section The second excited state decays to the first excited state through a transition with E_γ=1312.1 keV. Figure 5 panel b displays the comparison between our results, the TALYS calculations and the experimental results reported by D. Dashdorj *et al.*. Up to 4 MeV our experimental values are in a good agreement with the theoretical calculations and previous experimental values. Between 3 and 8 MeV the two sets of experimental results are not well described by the TALYS calculations. In the 8-12 MeV range the experimental results of D. Dashdorj *et al.* are well described by the TALYS microscopical calculations while our results are clearly higher. Beyond 12 MeV the TALYS microscopical calculations describes very well our experimental results, while the other set of data and the TALYS default calculations are lower.

The 1437.5-keV γ-production cross section The third excited state decays through a transition with E_γ=1437.5 keV to the first excited state. Figure 5 panel c displays the comparison between our experimental results, other experimental results reported by D. Dashdorj *et al.* and the TALYS calculations. It can be observed that up to around 7 MeV the two experimental sets are in a good agreement, while above that energy the experimental values obtained by us are higher. The TALYS theoretical calculations are underestimating both experimental results.

The 2013.7-keV γ-production cross section The next observed transition has E_γ=2013.7 keV and it populates the first excited state. In figure 5 panel d is displayed the comparison between our experimental results, the theoretical calculations performed with TALYS and previous results reported by D. Dashdorj *et al.*. The TALYS calculations underestimate the experimental results for all energies. The results of D. Dashdorj *et al.* are in a fairly good agreement with our results up to around 5 MeV, while above that energy our results are higher.

The 2240.4-keV and 928.3-keV γ-production cross sections The 2240.4-keV transition occurs between the sixth excited state and the first excited state, while the 928.3-keV transition is to the second excited state. The two γ rays are from the same level so the threshold energy and the shape of the cross section are the same. The ratio between the gamma production cross sections of each transition represents the emission probability. We performed a simple calculation in order to determine the emission probability for the 928.3-keV transition in the neutron energy range from E_{th}=3.3 MeV up to 18 MeV. The average emission probability resulted to be 36.2(9)%, while in the adopted level scheme the value is 33.46(24)% [25]. Performing the same calculation for the results reported by D. Dashdorj *et al.* the value is significantly different. This makes us confident that our results are correct. Because both γ-production cross sections were measured, none of the values of the emission probability had to be used in the analysis. Figure 5 panel e displays the comparison between our experimental results for the γ-production cross section of the 2240.4-keV transition, previously results reported by D. Dashdorj *et al.* and theoretical calculations performed with TALYS. While TALYS calculations underestimate the experimental results for the entire energy range, the agreement between the two experimental sets is very good up to around 8 MeV, where our results start to be higher. Panel f of figure 5 displays the comparison between our experimental results of the γ-production cross section for the 928.3-keV transition, the TALYS calculations and the results of D. Dashdorj *et al.*. The theoretical calculations describe well the experimental results of D. Dashdorj *et al.*, while our results are clearly higher. Taking into consideration that the calculated value of the branching ratio is close to the evaluated one, and that the results of D. Dashdorj *et al.* describe well our results for 2240.4-keV γ-production cross section but not the 928.3-keV γ-production cross section we conclude that our results are consistent, while the results of D. Dashdorj *et al.* are not.

The 944.1-keV γ-production cross section Another observed transition has E_γ=944.1 keV and M1+E2 multipolarity and populates the second excited state. Figure 5 panel g displays the comparison between our results for the γ-production cross section, the TALYS calculations and the results of D. Dashdorj *et al.*. TALYS calculations are underestimating both sets of experimental results. The results of D. Dashdorj *et al.* are in a very good agreement with our results up to around 8 MeV.

The 1037.6-keV γ-production cross section The γ-production cross section for the 1037.6-keV transition is compared in figure 5 panel h with the theoretical calculations and with previous experimental results. This transition has a E2 multipolarity and populates the second excited state. The agreement between our results and the results of D. Dashdorj *et al.* is very good. The TALYS microscopical calculations describe well our experimental results up to around 10 MeV and overestimate the results beyond that energy while the TALYS default calculations underestimate our experimental results.

The 2375.2-keV γ-production cross section The 2375.2-keV transition populates the first excited state. Figure 5 panel i displays the comparison between our results, the results of D. Dashdorj *et al.* and the TALYS calculations. While the TALYS calculations underestimate both experimental results, there is a good agreement between the two experimental γ-production cross sections.

The 2387.2-keV γ-production cross section The last observed transition in ^{48}Ti has E_γ=2387.2 keV. Figure 5 panel j displays the comparison between our experimental results, the results of D. Dashdorj *et al.* and the theoretical calculations. Once again, TALYS calculations underestimates the experimental values, but the results of D. Dashdorj *et al.* are in good agreement with ours.

Uncertainties In the following we present a short discussion on the statistical and systematic uncertainties in the data analysis procedure. Statistical uncertainties can be reduced by a proper binning of the data or by increasing the measurement time. The systematic uncertainties are the most difficult to handle. Analyzing the initial HPGe yields we observed a statistical uncertainty of 2% for the strongest channels. As we pointed out, in order to reduce the statistical uncertainty we bin the data. This is based on the fact that in the region above 2 MeV the cross section is more or less structureless. The experimental fission chamber counting rate is very low (corresponding to an unacceptable 7%) so we added and smoothed the spectra from several experiments. This is possible because there is no structure in the fission chamber data. We used a double smoothing procedure, each smooth consisting in a 61 channels second-order polynomial fit. Therefore, the resulted yield corresponding to the fission chamber has a relative uncertainty of 3%. The areal densities calculated using the geometrical details of the sample have an uncertainty of less than 0.1%. The HPGe detectors efficiency was calculated with an uncertainty of 2% for all γ rays below 1.4 MeV and up to 4% above this energy. This uncertainty is mainly from the activity and the position of the calibration source. The uncertainties coming from the MCNP5 simulations are less than 1%. For the strongest transition, the total resulted uncertainty was typically below 5% (see figure 6 panel b). For the other γ production cross sections the total resulted uncertainties are in the 6-10% range.

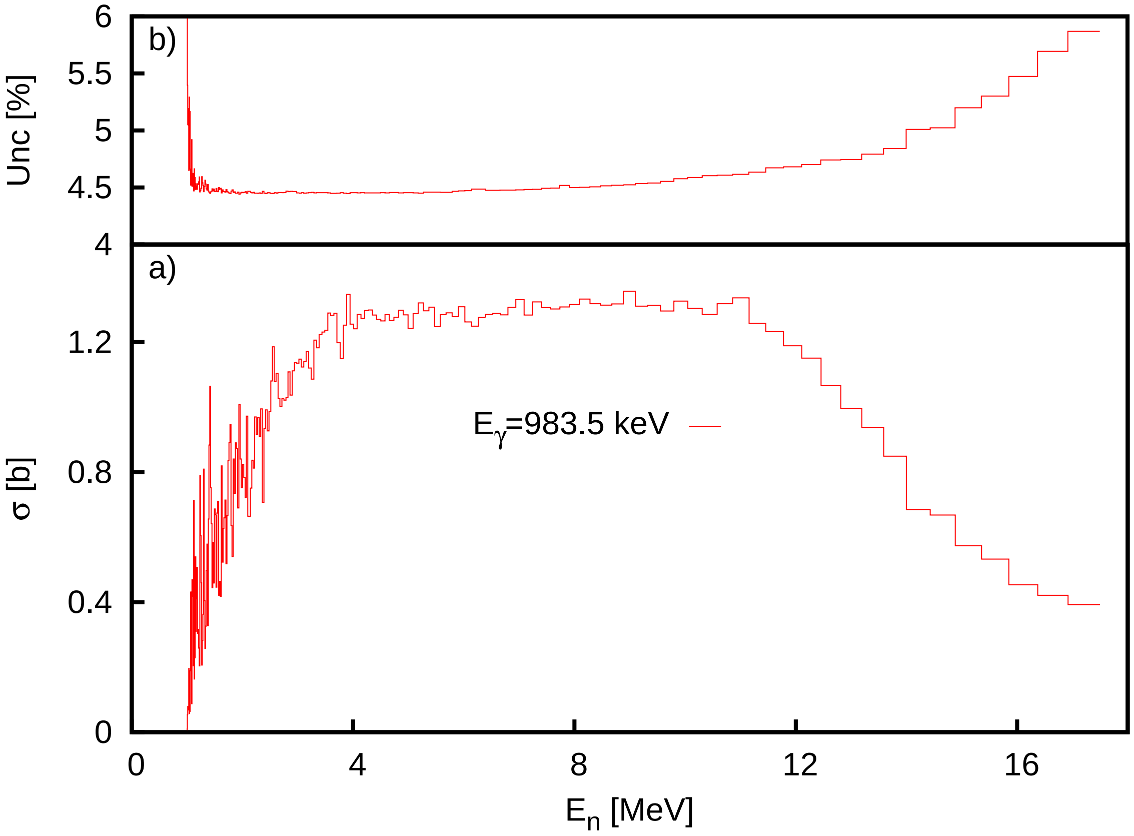

Figure 6: a) The experimental γ-production cross section of the 983.5 keV transition. b) The relative total uncertainty of the cross section.

1.4 Conclusions

During this measurement we observed ten γ transitions from the (n,n'γ) reaction on the ^{48}Ti. The contributions from the (n,2n) reaction was insignificant because of the small ratio between the isotopic abundances of the ^{49}Ti and ^{48}Ti. We calculated and reported the γ-production cross section of each transition. The experimental results were compared with theoretical calculations performed with TALYS 1.6 code and also with previously reported experimental results. For most of the transitions we observed a fairly good agreement between our results and other experimental results. The agreement seems typically better at low energies (E_n<8 MeV). The TALYS calculations usually underestimate the experimental results. It can be observed that when our data do not agree with TALYS and other experimental results, our values are higher. The total resulted uncertainty was typically below 5% for the strongest transition.

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