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The ^{99}Tc(n,γ)^{100}Tc cross section, ^{99}Tc(d,p)^{100}Tc and the ^{100}Tc decay scheme and neutron binding energy

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Abstract. Analysis of measurements on the ^{99}Tc(n,γ)^{100} and the Tc ^{99}Tc(d,p)^{100}Tc reactions leads to the conclusion that the determination of the inferred total thermal neutron capture cross section from partial capture cross sections necessarily has a large uncertainty at this time due to lack of accurate knowledge about the ^{100}Tc level structure and decay scheme. A more precise value of the neutron binding energy of ^{100}Tc of 6764.85 ± 0.25 keV was determined from the energies of the primary γ-rays.

1 Introduction

The isotope ^{99}Tc constitutes about 45 percent of the long-lived (>500 a) nuclear waste from pressurised water moderated reactors. Its ability to migrate and the efficient intake by living organisms make ^{99}Tc a hazardous radiotoxic material. Furthermore, the extremely long half-life (2.1×10^5 a), the low beta-decay endpoint energy (294 keV), and the single weak (6.5×10^{-4}%) low-energy (89.5 keV) γ-ray [1], which is superimposed on a high bremsstrahlung background, make the non-destructive assay of ^{99}Tc with passive nuclear methods difficult.

Fortunately, ^{99}Tc can be transformed by neutron capture into ^{100}Tc, which decays with a 15 s half-life to stable ^{100}Ru. This is the basis of transmutation of technetium contained in the nuclear waste [2]. The prompt γ-radiation produced can in principle be used for assaying technetium by prompt gamma activation analysis (PGAA). It is, however, not easy to use during the bulk transmutation process. Thus the off line, neutron activation analysis (NAA) technique has been applied in the Transmutation by Adiabatic Resonance Crossing (TARC) experiment performed at the Spallation Neutron Source at CERN [2] for controlling the transmutation process. The calculation of the yield of ^{100}Tc from NAA measurements requires a knowledge of the absolute gamma decay probabilities, P_γ of the states of ^{100}Ru which are populated in the ^{100}Tc(β^-)^{100}Ru beta decay.

It is the aim of our work to improve the precision of the gamma-ray partial capture cross sections for use in the assay of ^{99}Tc and ^{100}Tc, and also the total thermal neutron capture cross section for ^{99}Tc. The latter is needed for the design of an efficient transmutation system for the disposal of ^{99}Tc and other hazardous nuclear waste materials.

2 Summary of previous results

The total thermal neutron capture cross section has been measured many times over the years by various methods, and varying degrees of precision. The results are collected below from the EXFOR database [3], and many values are inconsistent, as can be seen in table 1. The latest evaluation of Mughabghab [4] is also shown.

Table 1. Values of ^{99}Tc total thermal neutron capture cross sections obtained from various experiments.

First author and year		Cross section	Method
H. Pomerance	1975	19 ± 2 b	pile oscillator
R.B. Tattersall	1960	16 ± 7 b	pile oscillator
N.J. Pattenden	1958	25 ± 2 b	transmission
M. Lucas	1977	20 ± 2 b	mass spectrometer
V.V. Ovechkin	1973	24 ± 4 b	activation
H. Harada	1995	22.9 ± 2.6 b	activation
Mughabgab	2003	20 ± 1 b	evaluation [4]

Table 2. Partial gamma ray cross sections of the most intense gamma rays from the ^{99}Tc(n,γ) reactions.

E_γ (keV)	Origin	σ_γ (b)
172.1	^{99}Tc(n, γ)^{100}Tc	16.61 ± 0.15
223.4	^{99}Tc(n, γ)^{100}Tc	1.472 ± 0.013
263.5	^{99}Tc(n, γ)^{100}Tc	1.425 ± 0.012
539.5	^{100}Tc(β^-)^{100}Ru	1.604 ± 0.014
590.7	^{100}Tc(β^-)^{100}Ru	1.296 ± 0.011

Several measurements of the singles spectrum from the ^{99}Tc(n,γ)^{100}Tc capture reaction have been made at the Budapest Research Reactor to determine the partial γ-ray capture cross sections, as well as the total thermal neutron capture cross section. For details on the procedures used, see refs. [5–7]. The results for the partial gamma ray capture cross sections are given in table 2, and those for the inferred total thermal-neutron-capture cross section are given in table 3.

3 Recent results

Since the above results were measured, additional measurements have been made to further refine the determination

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Table 3. Inferred total thermal-neutron-capture cross section of ^{99}Tc.

Method	Basis	scap (b)	Comment
^{100}Tc(β^-)^{100}Ru	539 γ	24.7 ± 2.3	with P_γ from [5]
	591 γ	23.9 ± 1.8	
	Average	24.3 ± 2.2	unweighted average
^{99}Tc(n, γ)^{100}Tc	$\sum \sigma_\gamma$ g.s.	21.21 ± 0.17	lower limit

of the ^{100}Tc decay scheme since a very precise and detailed knowledge of the gamma decay placements is needed in order to calculate the inferred total thermal neutron capture cross section from the individual partial γ-ray cross sections. As mentioned in refs. [5,7], the total thermal neutron capture cross section can be calculated using the formula:

$$\sigma_{cap} = \sum_{\gamma} \sigma_{\gamma g.s.}(1 + \alpha_\gamma)$$

where $\sigma_{\gamma g.s.}$ are the partial γ-ray cross sections which feed the ground state and α_γ is the internal conversion coefficient. Actually the equation is equally true for all the γ-rays which end on a particular level plus those that cascade past the level. Hence, the need for a very complete decay scheme can be seen for this method to give an accurate total cross section value. Any missing decay or misplaced decay can lead to a value which is too low or too high.

Two types of measurements have been made. The first was a γ-γ coincidence measurement on the ^{99}Tc(n,$\gamma\gamma$)^{100}Tc reaction to improve our knowledge of the placement of the γ-rays in the ^{100}Tc decay scheme. In order to have a correct decay scheme, it is absolutely necessary to have a complete and correct level scheme for the region under study. One way of determining the existence and positions of levels in the final nucleus is with a non-selective particle transfer experiment such as (d,p). The second type of experiment performed was measurements of proton spectra from the ^{99}Tc(d,p)^{100}Tc reaction to locate the low-lying levels of ^{100}Tc.

3.1 ^{99}Tc(n,$\gamma\gamma$)^{100}Tc coincidence experiment

The previous knowledge about the decay scheme of the low-lying levels of ^{100}Tc, taken from the work of Pinston et al. [8], is shown in figure 1.

To clarify and confirm the placement of the gamma rays in this decay scheme, we have performed a ^{99}Tc(n,$\gamma\gamma$)^{100}Tc coincidence experiment on a 15 mg Tc-target with a planar HPGe and a small volume HPGe detector. We used leading edge timing in order to be able to study very low energy gamma transitions. An example of a gated prompt coincidence spectrum can be seen in figure 2.

The gate was set on the very strong 299 keV γ-ray, which is shown to decay from a level near 500 keV in the Pinston decay scheme. Many of the coincident γ-rays shown there are either inconsistent with the placements in the adopted decay scheme [8], or are not placed in the decay scheme at all. In particular, the γ-rays with energies of 31, 43, 75, 99, 105, 179 and 181 keV are all shown on the Pinston decay scheme as not being in coincidence with the 299 keV γ-ray. In fact many of them decay to or from levels which the 299 keV γ-ray crosses over. Two of the strongest γ-rays in the coincidence spectrum, 64 and 90 keV, are not included in the Pinston decay scheme at all, and have unknown placements. It can be concluded that the strong 299 keV γ-ray is either misplaced in the Pinston decay scheme, or else is a composite of two or more different γ-rays, with only one of them possibly having the present placement. This sort of problem is seen in very many of the coincidence-gated spectra, indicating that there are many mistakes in the Pinston decay scheme which remain to be corrected. Not only are some γ-rays misplaced, but some of them are unresolved multiplets having different placements. In addition, there are very many of the stronger γ-rays which have no placement yet in the decay scheme.

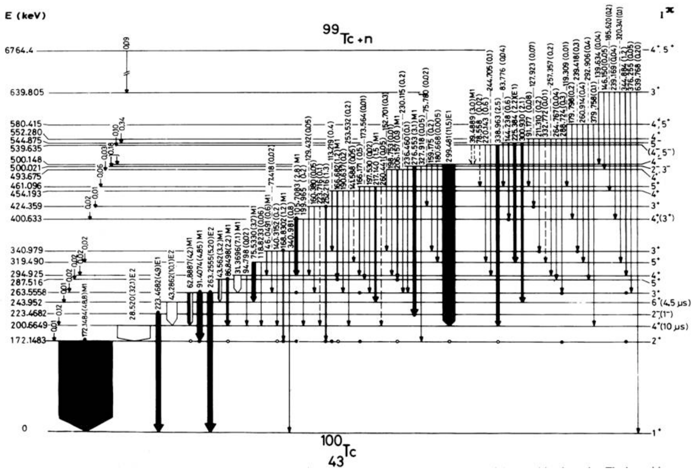

Fig. 1. Decay scheme of ^{100}Tc, based on the work of Pinston et al. [8].

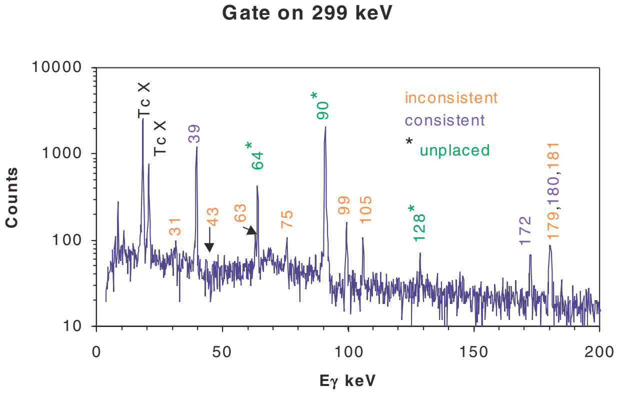

Fig. 2. ^{99}Tc(n,$\gamma\gamma$)^{100}Tc prompt-coincidence spectrum. A gate was set on the very strong 299 keV gamma ray shown in the Pinston decay scheme.

3.2 ^{99}Tc(d,p)^{100}Tc measurements

The (d,p) particle transfer reaction is relatively non-selective, populating all states in the final nucleus up to some maximum angular momentum. Measurements were made on the ^{99}Tc(d,p)^{100}Tc reaction to identify and locate all the low-lying

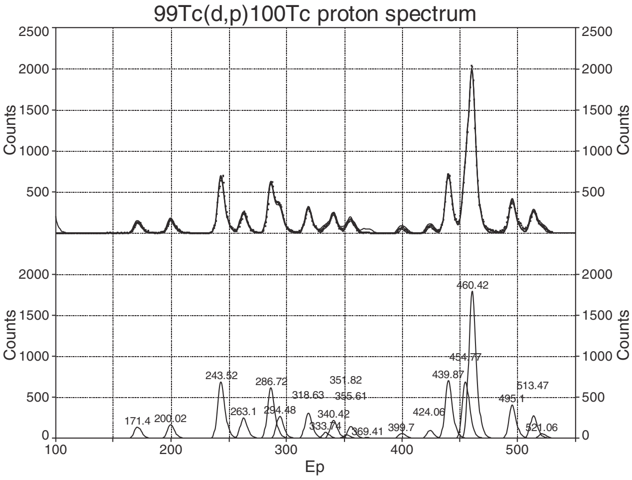

Fig. 3. Proton spectrum measured at emission angle of 30 degrees on the ^{99}Tc(d,p)^{100}Tc reaction.

levels of ^{100}Tc for use in the decay scheme. The measurements were made with the Q3D magnetic spectrometer in the Munich Tandem Laboratory. A 120 microgram/cm^2 target of ^{99}Tc was bombarded with a 22 MeV deuteron beam with observation angles of 30 and 60 degrees. The spectrometer had an acceptance angle of 5.5 msr and the proton energy resolution was 7–8 keV. The spectra were accumulated for 1–2 hours using a 950 nA beam. The proton spectra covered an energy range corresponding to excitation energies in ^{100}Tc of 0 to over 1.5 MeV. The spectrum measured at 30 degrees is shown in figure 3.

E_p is the excitation energy of the final level in ^{100}Tc. The top panel is the measured spectrum with the calculated fit (which is almost indistinguishable from the data points in the figure), while the bottom panel shows the constituent proton peaks used in the fit. The experimental spectrum was reduced from the raw data using the program GASPAN. The fit was done using the program Spectrum-Fit4. Prominent groups are observed that correspond to the first and second excited states of ^{100}Tc at 172 and 200 keV, but there is absolutely no sign of a proton group corresponding to the third excited state shown in figure 1 at 223 keV. This group is not observed in the 60 degree spectrum of the present work either, nor in the earlier (d,p) measurements of Slater and Booth [9], though that work has worse resolution and background conditions. The reputed level at 223 keV in ^{100}Tc is thought to have a spin of 1 or 2, and so should have been easily populated in the (d,p) experiment. In fact there is no unequivocal evidence in the literature to date for a level at 223 keV. The 223 keV γ-ray that is alleged to depopulate the state has only been observed in the (n,γ), (p,nγ) and (d,2nγ) reactions, but with no particle-gamma coincidence or corresponding primary gamma ray to verify the emitting level. It is therefore possible that there is no level at 223 keV, and that the 223 keV γ-ray needs to be placed elsewhere in the decay scheme.

3.3 Neutron binding energy of ^{100}Tc

The neutron capture measurements on ^{99}Tc not only give the γ-rays decaying from the low-lying levels of ^{100}Tc, but the spectra also contain many primary γ-rays from the capturing state to the various low-lying levels of ^{100}Tc. These primaries can be seen in the upper inset panel of figure 4.

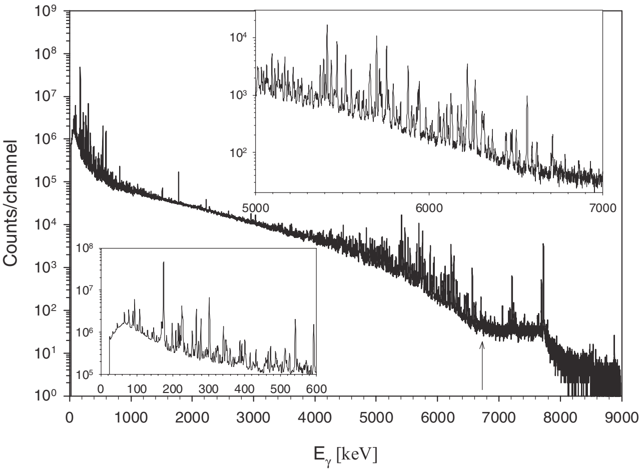

Fig. 4. γ-ray spectrum for ^{99}Tc(n,γ)^{100}Tc reaction. The primary γ-rays form the thick forest in the energy range from 5.3 to 6.8 MeV.

The energies of the primary γ-ray decays in ^{100}Tc were calibrated with a two-point calibration based on the ^{27}Al(n,γ)^{28}Si γ-rays with energies of 1778.969 ± 0.012 and 7724.034 ± 0.007 keV, taken from ENSDF. The energies were corrected for non-linearity in the detection system and also for nuclear recoil. The nonlinearity correction was based on the energies of the γ-rays from the ^{35}Cl (n,γ) and ^{52}Cr(n,γ) reactions. Each fully corrected primary γ-ray energy was then added to the corresponding level energy of the final state for the γ-decay. The level energies were the very accurate values determined with a crystal diffraction spectrometer by Pinston et al. [8]. Each sum gives a value of the neutron binding energy of ^{100}Tc. The average of the sums for the 10 lowest laying levels fed by primary gamma rays of ^{100}Tc was calculated to give a new value of the neutron binding energy of:

$$E_b = 6764.85 \pm 0.25\,\text{keV}$$

which is 0.45 keV higher and has four times smaller uncertainty than the previously accepted value [8] in TOI.

4 Conclusions and summary

Further analysis of the data on the ^{99}Tc(n,$\gamma\gamma$)^{100}Tc reaction and the ^{99}Tc(d,p)^{100}Tc reaction have lead to the conclusion that there are difficulties in determining the total thermal neutron capture cross section of ^{99}Tc by the method of summing the strengths of all the γ-transitions to the ground state.

The lack of any observable proton group for the 223 keV third excited state of ^{100}Tc in the proton spectrum of the ^{99}Tc(d,p)^{100}Tc reaction makes the existence of a level at 223 keV excitation energy questionable, since there is no independent evidence for the existence of this level. If the level does not exist, then the strong 223 keV γ-ray cannot be part of the sum of transition strengths to the ground state, and this will have the effect of reducing the inferred capture cross section by 6–8%. On the other hand, it is hoped that further analysis of the (d,p) spectra will determine many new higher lying levels to which presently unplaced γ-rays can be assigned, and this may open the possibility of using other sums of γ-ray strength to infer a total capture cross section.

The prompt coincidence experiment results cast doubt on the present knowledge of the placements of the gamma rays in the decay scheme of all the low-lying levels, not to mention the decay scheme of all the higher lying levels whose existence is not yet determined. The presently adopted placements of the second and third strongest gamma rays in the spectrum, namely 223 keV and 299 keV, are suspected from the coincidence results to be incorrect for at least part of their strength, since both are suspected to be unresolved multiplets.

As a result of these new uncertainties, and the wide spread of the previous results given in table 1, we currently prefer to quote a larger uncertainty in the inferred total thermal neutron capture cross section of ^{99}Tc of:

$$\sum_{g.s.} \sigma_\gamma = 21 \pm 3\, b.$$

Based on an accurate energy calibration of the spectrum of primary γ-rays for the ^{99}Tc(n,γ)^{100}Tc reaction, a new value of the neutron binding energy for ^{100}Tc has been determined to be:

$$\mathrm{E}_b = 6764.85 \pm 0.25\,\mathrm{keV}.$$

It is planned to try to improve the knowledge of the ^{100}Tc level and decay schemes by particle-gamma coincidence measurements on the ^{99}Tc(d,pγ)^{100}Tc reaction.

References

1. L.K. Peker, Nucl. Data Sheets. **73**, 1 (1994).
2. A. Abánades et al., Nucl. Instrum. Meth. Phys. Res. A **478**, 577 (2002).
3. EXFOR Experimental Nuclear Reaction Data Library (in USA usually called CSISRS). This data base contains data compiled as the result of the co-operative efforts of the world-wide Nuclear Reaction Data Centre Network, co-ordinated by the IAEA Nuclear Data Section, 2002.
4. S.F. Mughaghab, INDC-NDS-440 (2003).
5. G.L. Molnár, T.Belgya, Zs. Révay, S.M. Qaim, Radiochim. Acta **90**, 479 (2002)
6. T. Belgya, J. Weil, G.L. Molnár, in *Proceedings of the 11th International Symposium on Capture Gamma-Ray Spectroscopy and Related Topics,* Pruhonice, near Prague, Sept. 2002, p. 462.
7. T. Belgya, J. Weil, G.L. Molnár, H.-F. Wirth, *Proceedings of NEMEA Workshop,* Budapest, 2004.
8. J.A. Pinston, W. Mampe, R. Roussille, K. Schreckenbach, D. Heck, H.G. Börner, H.R. Koch, S. Andre, D. Barneoud, Nucl. Phys. A **321**, 25 (1979); *Table of Isotopes*, CD-ROM, 8th edn., edited by Richard B. Firestone, Virginia S. Shirley, S.Y. Frank Chu. (Wiley-Interscience, 1996).
9. D.N. Slater, W. Booth, Nucl. Phys. A **267**, 1 (1976).