# NEUTRON-INDUCED ACTIVATION CROSS SECTIONS: MEASUREMENTS AND MODEL SENSITIVITY

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## ABSTRACT

New measured values are presented for neutron induced reaction cross sections on the low-activation structural material $^{51}V$ and on the long-lived fission product $^{99}Tc$. In addition a method is illustrated that establishes the sensitivity of model parameters to measured reaction cross sections in the case of neutron-induced proton emission reactions on $^{56,57}Fe$.

Measurements have been made by the activation technique for the $^{51}V(n,n'\alpha)^{47}Sc$ reaction between 16.3 and 19.5 MeV to resolve large discrepancies in calculated over experimental values that resulted from benchmark tests of vanadium alloys. The data in this energy range are essentially unique.

The $^{99}Tc(n,p)^{99}Mo$ and $^{99}Tc(n,\alpha)^{96}Nb$ reaction cross sections have been measured from 8.5 to 12.3 and from 16 to 20.5 MeV. These measurements allow the first construction of an excitation curve for this candidate nuclide for transmutation.

Due to the large number of requested measurements for reaction cross sections the search for a reliable model description that allows extrapolation into the unknown is of particular importance. The sensititivity of model parameters to measured cross sections is a step towards quantitative statements about predictive power and validity of parameter systematics. In addition, it serves to establish priorities for new measurement campaigns.

## 1. INTRODUCTION

Neutron-induced reaction cross sections have been measured extensively between threshold and 15 MeV for fission and fusion reactor applications. Recent interest extends to the GeV region for the design of Accelerator Driven Systems (ADS, Ref. 1). Calculations of neutron transport, activity inventories and damage estimates complement and assist the interpretation of benchmark experiments. This requires a large body of good quality cross section data. The energy range below 20 MeV is important since most neutrons in an ADS originate from evaporation and fission (Ref 2).

At the IRMM 7 MV CN Van de Graaff accelerator a large number of neutron-induced reaction cross sections have been measured recently for incident energies between 16 and 20 MeV by the activation technique (Refs. 3-5). This work is of relevance to gas-production through emission of protons and alphas and neutron transport properties through (n,xn) reactions. The energy range of the measurements bridges the gap between the region from threshold to 15 MeV and the range currently addressed by others above 20 MeV. Measurements for the $^{51}$V(n,n'$\alpha$), $^{99}$Tc(n,p), and $^{99}$Tc(n,$\alpha$) reactions are presented as the most recent examples.

Vanadium alloys have long been considered by the fusion reactor community for their low-activation properties. Recent activation studies of vanadium alloys in various neutron fields of relevance to fusion reactors showed large discrepancies between measured activities of $^{47}$Sc and the activities calculated from EAF97 cross sections (Ref 6). Measured cross sections between 16.3 and 19.5 MeV will be presented for the reaction $^{51}$V(n,n'$\alpha$)$^{47}$Sc and a comparison will be made with existing evaluations.

Since $^{99}$Tc is considered for transmutation it is of interest to determine its properties in an intense hard neutron spectrum. For this isotope measured cross sections will be presented for the $^{99}$Tc(n,p)$^{99}$Mo and $^{99}$Tc(n,$\alpha$)$^{96}$Nb reactions. The data taken between 8.5 and 12.3 MeV incident energy at the Jülich cyclotron and between 16 and 20.5 MeV at IRMM are unique and complement a limited number of data points taken between 13 and 15 MeV.

Finally, a method is illustrated that determines the sensitivity of model parameters to the measured data for proton emission reactions on $^{56}$Fe and $^{57}$Fe. A systematic study of such sensitivities will serve as a guide to both evaluators and experimentalists and will allow quantitative statements about the predictive power of a model and its parameter systematics. Such a study will be the subject of a collaborative effort of a subgroup of the NEA Working Party on Evaluation Collaboration.

## 3. EXPERIMENT

The measurements above 15 MeV have been done with the activation technique at the IRMM 7 MV CN Van de Graaff accelerator. Neutrons were produced by the T(d,n) reaction using a deuterium beam of 1 to 4 MeV and 10-30 $\mu$A and a solid Ti-T target of 2 mg/cm$^2$. Samples sandwiched between monitor foils were irradiated for 4-8 hours at zero degrees and 1 cm from the neutron source in a low mass holder. For the irradiations of the vanadium samples Al-Co-Nb-V-Nb-Co-Al stacks were used with 0.166 g metallic vanadium samples. A similar stack was used for the technetium irradiations with the vanadium sample replaced with an 0.47 g $^{99}$Tc sample canned in an aluminum capsule. The use of these foils allow for the normalization of the cross section to the well-known $^{27}Al(n,\alpha)^{24}Na$ standard cross section, the determination and correction of flux gradients, and the correction for the neutron energy spectrum.

Activities were determined by gamma-ray counting using HPGe detectors. For the 3.345 d activity of $^{47}Sc$ the 159 keV transition (67.9% emission probability) was used. The 23.4 h activity of $^{96}Nb$ was determined via the 570 keV transition (58.0% emission probability) and that of the 65.9 h activity of $^{99}Mo$ followed from the 740 keV transition (12.1% emission probability). A Pb absorber was used to diminish the bremsstrahlung background.

Measurement and correction procedures are well described in Ref. 3, with differences only in the facts that no pneumatic transport system was used in the present work, and that neutron spectrum and multiple scattering corrections have not yet been applied. In this respect the data are still preliminary.

The measurements on $^{99}Tc$ between 8.5 and 12.3 MeV were carried out at the INC, FZ-Jülich. Irradiations were done with quasi-monoenergetic neutrons obtained from the D(d,n) reaction using the variable energy Compact Cyclotron CV 28 and a deuterium gas target. Details of the neutron source and the irradiation geometry are described in Ref. 7. Al-Fe-Tc-Fe-Al stacks were used for the irradiations with gas-in and gas-out. Neutron spectrum corrections were applied as described in Ref. 8. Gamma-ray activities were counted and corrected in the same way as described for the measurements done at IRMM and the final data were normalized to the $^{27}Al(n,\alpha)^{24}Na$ standard cross section.

## 4. RESULTS AND DISCUSSION

### 4.1 THE $^{51}V(n,n'\alpha)^{47}Sc$ REACTION

The results for the $^{51}V(n,n'\alpha)^{47}Sc$ reaction are compared to cross sections measured by others and to the ENDF/B-VI, EAF-97 and EAF-99 evaluations in Figure 1. The eye-guide clearly indicates that the trend of our data is consistent with the measurements by Grallert et al., Pepelnik et al. and Qaim et al. between 14 and 15 MeV (Refs 9-11). Good agreement is obtained with the single data point of Bormann et al. (1961, Ref 12) but large discrepancies are observed with the other measurements by Bormann et al. (1962, Ref 12). The new data are also discrepant with the result from the semi-integral measurement of Ref. 13.

The EAF-97 evaluation is seen to overpredict our new data by large factors (up to 20 times), whereas for EAF-99 the discrepancies are limited to a factor of 2.5 with underprediction above 17 MeV and moderate overprediction at the lower energy. In the energy range between 16 and 20 MeV ENDF/B-VI gives values intermediate between EAF-97 and EAF-99.

This reaction clearly illustrates the difficulties encountered by evaluators when insufficient good quality data are available to construct an excitation curve. Also it is clear that the contribution of the $^{51}V(n,n'\alpha)$ reaction to the production of the $^{47}Sc$ activity in vanadium alloys should be considerably below that estimated from calculations based on EAF-97. Although, this would make the contribution of this reaction marginal compared to the contributions of the proton emission reactions on $^{47,48}Ti$ it does not entirely resolve the discrepancies reported in Ref. 6.

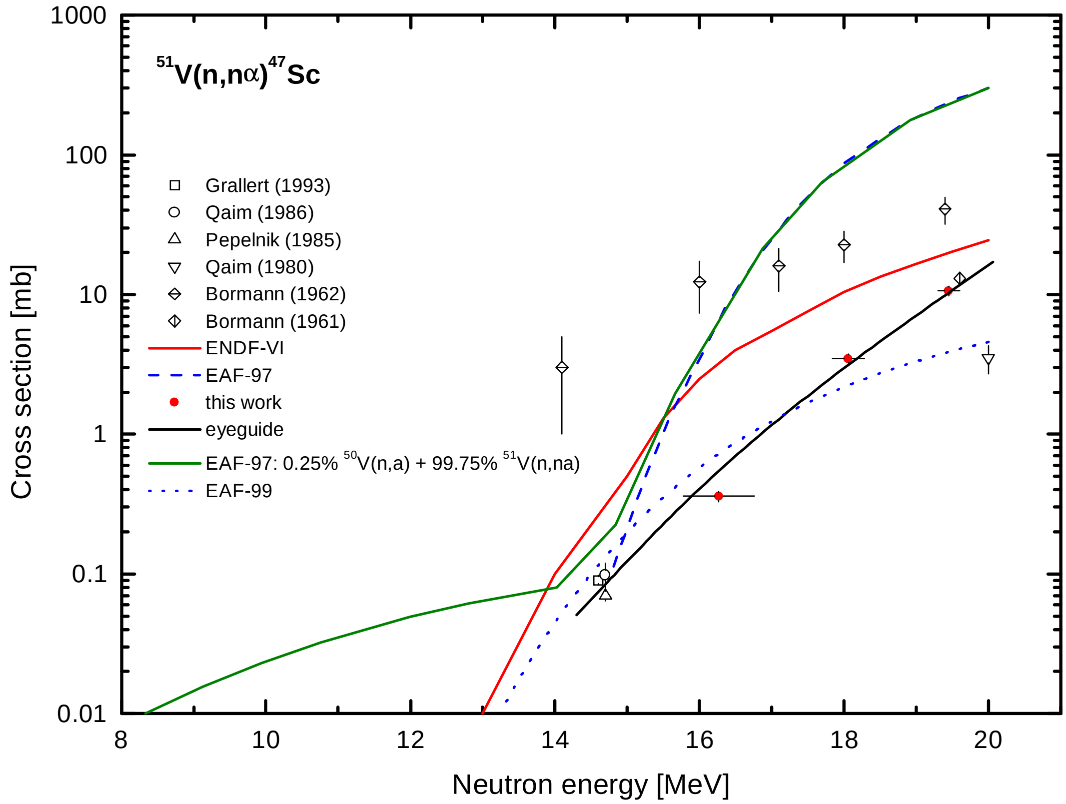


Figure 1. Experimental results and evaluated data for the $^{51}$V(n,n'α)$^{47}$Sc reaction

## 4.2 THE $^{99}$Tc(n,α)$^{96}$Nb AND $^{99}$Tc(n,p)$^{99}$Mo REACTIONS

In Figure 2 the new results obtained at FZ-Jülich and at IRMM are compared to the results of Ikeda et al., Qaim et al., and the early measurements by Goldstein et al. (Refs 14-16). In both cases the new data establish an excitation curve in good agreement with the data by Ikeda et al., Qaim et al. and Goldstein et al., however, for the (n,α) reactions the latter two authors give values that appear somewhat high. For the new data at 20.8 MeV a significant change may be expected from corrections for the neutron spectrum and for neutron multiple scattering.

The measured data are compared with evaluated data from the JENDL activation file and from JEF-2.2. Both files show excitation curves obtained from model calculations that are normalized to the data between 14 and 14.5 MeV. This works well in the case of the $^{99}$Tc(n,p) reaction and the JENDL activation file but fails for the other cases. In the case of the JEF-2.2 (n,p) reaction the shape of the curve is not consistent with that of the data. For the (n,α) reaction the JENDL and JEF-2.2 shapes are similar. Here the discrepancies with all recent data result from a normalization to the earlier values alone.

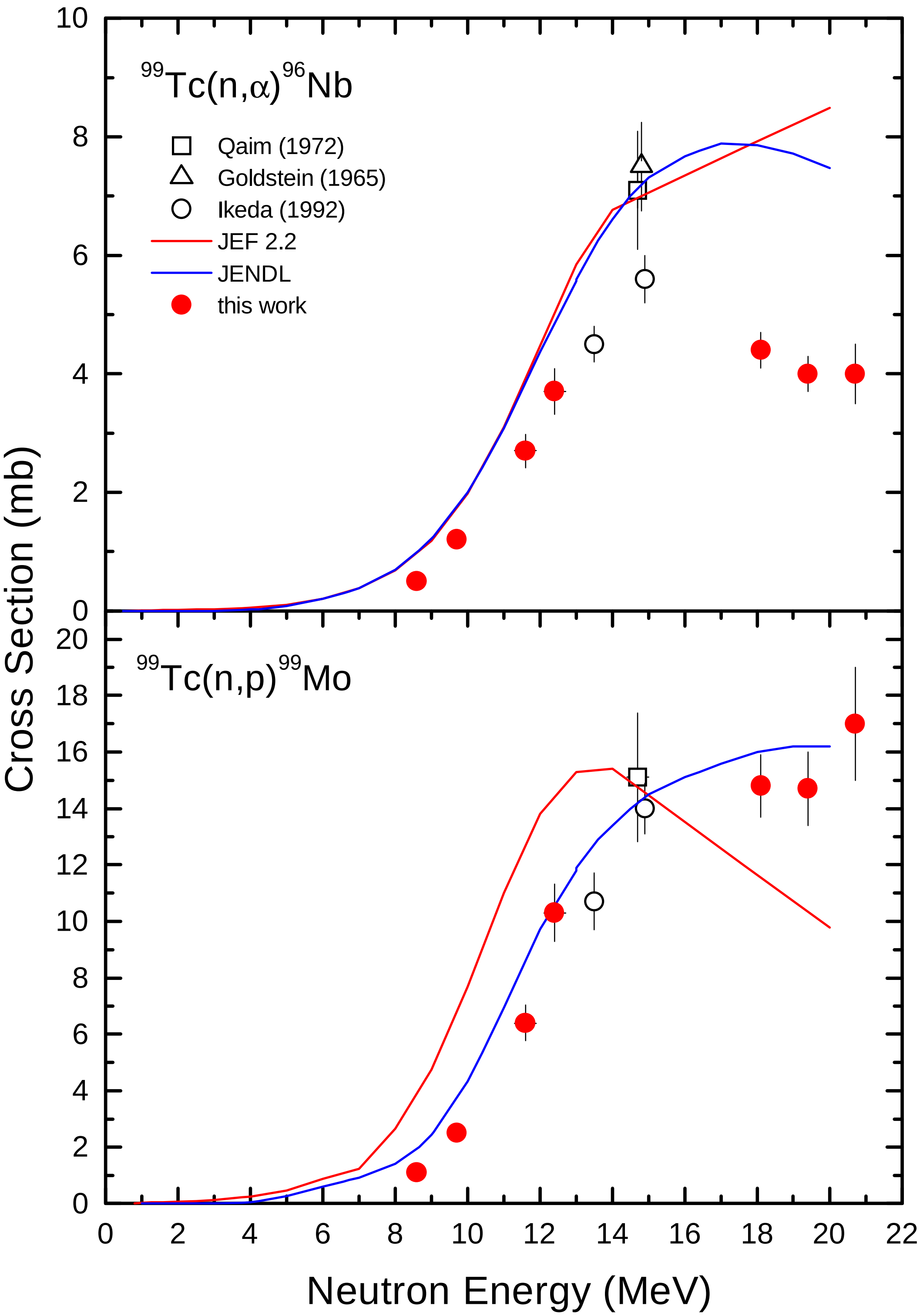


Figure 2. Results of the measurements for $^{99}$Tc compared to evaluations.

## 5. PARAMETER SENSITIVITIES TO CROSS SECTIONS

To demonstrate the usefulness of the concept of parameter sensitivities to cross sections (Ref. 17) and the status of parameter systematics (Ref. 18) a number of model calculations were performed with the STAPRE-H code (Ref. 19). Calculations were made for the $^{56}Fe$(n,p), $^{57}Fe$(n,p) and $^{57}Fe$(n,np) reactions for which new data were obtained recently (Ref. 3).

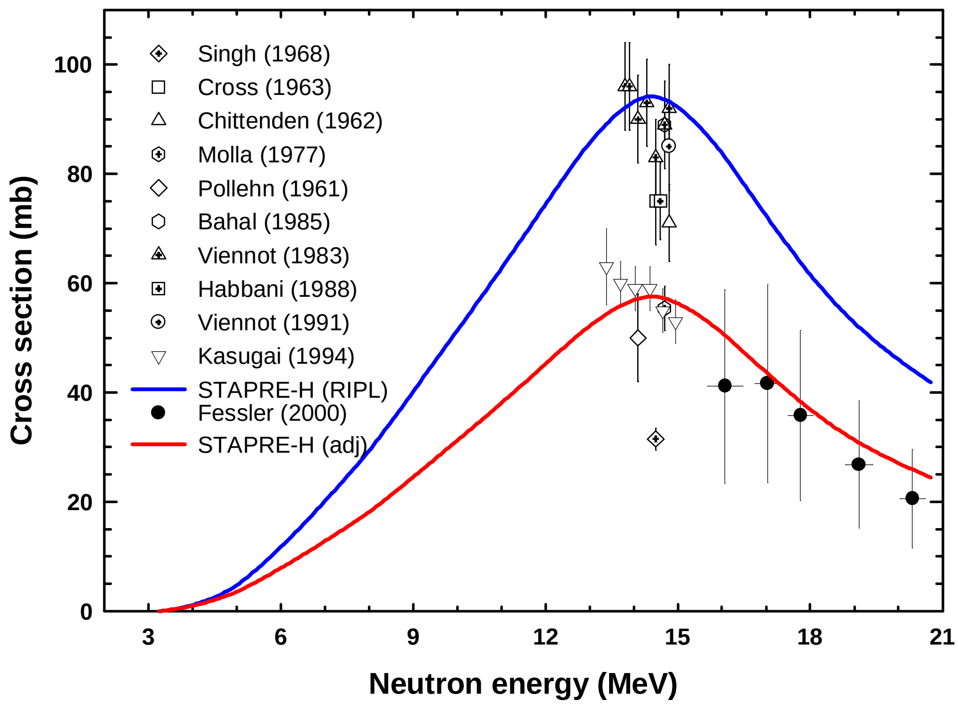


Figure 3. Statistical model calculations with estimates for pre-equilibrium contributions

In Figure 3 the results are shown for 2 model calculations of the $^{57}Fe$(n,p) reaction. The first calculation uses the Reference Input Parameter Library (RIPL, blue curve, Ref. 18) while the second is adjusted to describe the recent data (see the discussion of Ref. 3). Similar plots were made for the other two reactions with similar conclusions: 1) Blind calculations based on the RIPL parameter sets lead to large discrepancies. 2) To obtain the good agreement of the adjusted curve the Back-Shifted Fermi-Gas model level density parameters *a* for $^{56}Mn$ and $^{57}Fe$ had to be adjusted significantly to agree with the experimental data on the level density for these nuclei, as given in Ref. 18. Only minor adjustments of the level density parameters *a* sufficed in the cases of $^{56}Fe$ and $^{57}Mn$.

Table I presents the sensitivities of the cross sections to the model parameters for these calculations. As was detailed in Ref. 17, these sensitivities are dimensionless quantities defined by the relative change of a cross section or other measured value over the relative change in a model parameter's value. In the case of multiple cross sections and multiple model parameters they form a sensitivity matrix $S_{ij}$ where the row-index $i$ represents a measured value and the column-index $j$

a parameter of the model. Besides the sensitivities to the level density parameters we also give the values for the strength parameter *FM*. This parameter governs the estimates for the pre-equilibrium contributions according to the Exciton model.

Table I. Parameter sensitivities (dimensionless) of the reaction cross sections for $^{56}$Fe(n,p)$^{56}$Mn, $^{57}$Fe(n,p)$^{57}$Mn and $^{57}$Fe(n,np+pn+d)$^{56}$Mn

| Reaction | Energy (MeV) | a($^{56}$Fe) | a($^{56}$Mn) | a($^{57}$Fe) | a($^{57}$Mn) | FM |
|---|---|---|---|---|---|---|
| $^{56}$Fe(n,p)$^{56}$Mn | 14 | -4.6 | 5.4 | 0 | 0 | 0 |
| | 17 | -3.5 | 3.9 | 0 | 0 | -0.2 |
| | 20 | -1.8 | 1.9 | 0 | 0 | -0.4 |
| $^{57}$Fe(n,np+pn+d)$^{56}$Mn | 14 | -0.8 | 0 | -4.6 | 7.0 | 0.1 |
| | 17 | -1.3 | 0.2 | -4.2 | 6.2 | 0.1 |
| | 20 | -1.9 | 1.4 | -2.9 | 5.2 | 0.1 |
| $^{57}$Fe(n,p)$^{57}$Mn | 14 | 0 | 0 | -5.1 | 6.0 | -0.1 |
| | 17 | 0 | 0 | -4.1 | 4.8 | -0.3 |
| | 20 | 0 | 0 | -0.6 | 2.7 | -0.6 |

Table I clearly demonstrates the importance of the level density parameters of the residual nuclei as well as the strong anti-correlation of these parameters in the case of competing decay channels. It is also evident that this anti-correlation is not very energy dependent and that therefore an unambiguous determination of all the level-density parameters based on the reaction cross sections alone is not feasible. One has two options to determine these parameters unambiguously: 1) include measured cross sections for the competing reaction channels, that is (n,n') and (n,2n), or 2) include experimental data on the level-density for the nuclei involved. The latter approach was taken here. Of course a fully consistent approach would include both options given above.
Table I also demonstrates the very low sensitivity of the measured data to the *FM* strength parameter of the exciton model. A large change of this parameter is therefore required to affect the calculation appreciably. Although the trend as a function of energy differs from that for the level-density parameters it will be difficult to fix this parameter accurately from the present measurements until the values for the level-density parameters are fixed unambiguously. A change of the *FM* parameter is therefore meaningful, only if remaining discrepancies are significant and can be resolved with this parameter. As is suggested by the trend of the sensitivity coefficient as a function of energy and as is known from the theory of pre-equilibrium reactions, data at higher energies are required to fix this parameter reliably.

## 6. SUMMARY

New experimental data have been obtained for the $^{51}V(n,n'\alpha)^{47}Sc$ reaction between 16 and 20 MeV that show a trend consistent with good-quality data between 14 and 15 MeV. Large discrepancies were observed with older measurements and with the EAF-97, EAF-99 and ENDF/B-VI evaluations. The new data clearly indicate a reduced contribution of this reaction to the production of $^{47}Sc$ in vanadium alloys. As such the large C/E values for the production of $^{47}Sc$ in integral measurements on these alloys would at least partially be reduced. Further investigations to extend the measurements from 13.8 to 21 MeV are ongoing.

The new experimental data for the $^{99}Tc(n,p)^{99}Mo$ and $^{99}Tc(n,\alpha)^{96}Mo$ reactions between 8.5 and 20 MeV are the first that allow the construction of excitation curves for these reactions. Good agreement is obtained with measurements available between 13 and 15 MeV. The JENDL activation file describes the (n,p) data remarkably well, contrary to JEF-2.2. In the case of the $(n,\alpha)$ reaction both JEF-2.2 and the JENDL activation file need to be revised.

Finally, the validity of the RIPL level-density parameter set for proton emission reactions was discussed and the use of the concept of cross section sensitivities to model parameters was illustrated.

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