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SOME NEW MEASUREMENTS AND RENORMALIZATIONS OF NEUTRON CAPTURE CROSS SECTION DATA IN THE KEV - ENERGY RANGE

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Federal Republic of Germany

I. Introduction

Radiative Neutron Capture is one of the most important nuclear processes relevant to the neutron balance in fast reactors. Consequently, there has been a major effort in many laboratories in order to measure capture cross sections, particularly in the keV energy range. Nevertheless, the situation is still far from satisfactory. The main regions where data are incomplete, as discussed recently within international data committees, are the following:

a) Although cross section ratios can be measured with sufficient accuracy in many cases, the deduction of absolute cross section values has not been carried out consistently in most cases due to the lack of a proper and internationally recognized standard cross section[1]. This holds particularly in the energy range 10 - 100 keV where flux measurements (and thus standard cross section determinations) are difficult to carry out.

b) For some of the most important reactor materials, especially the fertile and fissile nuclei, a very high accuracy - in some cases down to $\pm$ 1 % - is required. Even if standard cross sections were known to this accuracy, these requests could not always be fulfilled since the cross section ratio measurement methods are not that accurate.

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1) This is although true for fission cross sections.

c) Cross section ratio measurements on very small or on highly radioactive materials are difficult to perform and require the development of special techniques. Similarly, accurate measurements on nuclei with small capture cross sections, e.g. lighter structural materials like Fe, cannot be carried out easily.

d) There is finally a number of nuclei for which adequate measuring methods as well as samples are available but which hitherto have just escaped attention. To these belong many stable fission products and control materials.

It is obvious that these problems, whose solution might require a total effort of several 100 man-years, can only be attacked in a close international cooperation between many laboratories. Some first steps in this direction have recently been taken by the European-American Nuclear Data Committee.

In order to contribute to some of the above problems, a programme has been launched at the Karlsruhe 3 MeV pulsed Van de Graaff accelerator which will be briefly reviewed in this paper. So far, a large part of our effort has been spent on the standardization problem where we have remeasured the absolute capture cross section of gold between 25 and 500 keV to an accuracy of about $\pm$ 5 %. We propose to use this cross section curve as a standard for further renormalizations of capture and fission cross sections. The results of this experiment are described in section 2 of this paper. Our method to determine the gold cross section is rather tedious and time-consuming, nevertheless we believe that some cross sections like U^{238} capture and U^{235} fission are so fundamental to fast reactor calculations that they warrant a direct determination by essentially the same method in order to avoid additional errors due to a cross section ratio measurement. As a first step along this line, we have redetermined the U^{238} capture cross section in the energy range 25 - 500 keV (section 3). The standard time-of-flight technique using a large liquid scintillator tank for capture cross section measurements has also been further developed in our laboratory, some results on heavy structural materials are presented in section 4.

In conclusion, section 5 contains some implications of these new data on reactor calculations and some general comments on future capture cross section work.

II. The Absolute Capture Cross Section of Gold

This experiment was performed in two steps: First, the shape of the cross section curve between 25 and 500 keV was measured by a new technique. Then, this realtive cross section curve was normalized at 30 keV neutron energy, using results from several independent absolute measurements.

The experimental setup for the shape measurement is shown in fig. 1. A thin gold foil was irradiated by a collimated beam of nearly monoenergetic neutrons, obtained at an angle of 80° from the $Li^7(p,n)Be^7$ reaction (target thickness: 12-18 keV for low energies, 12-40 keV in the middle energy region and 40-100 keV for high energies). The gold sample was located within a 1.1 m - diameter large liquid scintillator tank [1,2] which was used to determine the relative capture rate. The pulsed beam - time-of-flight method was used for background discrimination. The neutrons transmitted through the gold foil were thermalized and totally absorbed in a "grey neutron detector" [3]. This is a paraffin pile, size 60 x 60 x 60 cm, with a 10 x 10 x 30 cm beam entrance hole, located at the end of the neutron path. Capture of thermalized neutrons by hydrogen leads to the well-known 2.2 MeV γ-ray; since γ-ray absorption in paraffin at this energy is small, the photopeak counting rate of a NaI (Tl) detector located near the pile is very nearly proportional to the capture rate and thus to the impigning neutron current. This detector has to a first approximation an efficiency curve which is independent of neutron energy; to a higher approximation, there exist small deviations from the flat efficiency curve which can however be calculated [3].

The relative capture cross section follows directly from the counting rate of the large liquid scintillator tank and from the corrected counting rate of the grey detector. However, some slight corrections have to be applied in addition: Relation between average cross section and cross section at an average energy; resonance selfshielding and multiple scattering within the gold foil; efficiency change of the large liquid scintillator whose bias was set at about 3,6 MeV photon energy with neutron energy; air scattering of neutrons between the gold foil and the grey detector; appearance of the second neutron group from the $Li^7(p,n)Be^7$ reaction above ≈ 380 keV neutron energy. These corrections, together with a detailed discussion of the experimental approach, will be published elsewhere [4].

The absolute normalization of the relative cross section curve was performed at 30 keV neutron energy. Table 1 lists the results of several

more recent determinations at this energy from five independent methods. The table includes data taken with Sb-Be sources which were transformed to 30 keV assuming E_n = 22,8 keV [5] for Sb-Be and a cross section ratio of 0,915 ± 0,040 [2]. The fitted best value from all these results is

$$\sigma_{n,\gamma}^{(Au)} \ (E_n = 30 \text{ keV}) = 0.596 \pm 0.012 \text{ barn.}$$

The gold capture cross section as a function of energy, normalized with the above value, is shown in fig. 2.

We have carried out a further independent determination of the gold capture cross section shape in the energy range 10 - 150 keV. A time-of-flight method with neutrons from a thick Li^7 target was used. The capture rate in a gold foil was measured with the large liquid scintillator tank, while the relative neutron flux was simultaneously determined with a thin boron 10 slab viewed by NaI (Tl) scintillators or, in a different run, by a Li^6 glass detector. Both detectors were placed at the exit of the large liquid scintillator tank. The Boron 10 n,$\alpha\gamma$ cross section as recommended by Spaepen [6] and the Li^6 n,α cross section from the Breit-Wigner fit of Bergström et al. [7] were used for the calculation of the neutron detector efficiencies. In these calculations, corrections for multiple scattering of neutrons within the detector were applied. The measurement yielded however only the relative shape of the gold cross section since no effort was made to determine the absolute efficiency of the neutron detectors. The shape curves were again normalized to a value of 596 millibarn at 30 keV. The resulting curves are compared to the grey detector results in fig. 3. Note that the values found relative to boron and to lithium agree very well among themselves. Since they were taken with higher resolution than the grey detector data they show some structure. On the average, they agree very well with the grey detector data, at least below 80 keV. The deviations above 80 keV are small and well within the limits of experimental error.

A detailed comparison of our new standard cross section as shown in fig. 2 with the results of previous experiments will be given in ref. [4]. Briefly speaking, there is good agreement of the shape with most of the recent determinations by various methods. There is also good agreement in the absolute values over the whole energy range with a recent experiment by Harris et al. [8] which was performed by an absolute method and is, like ours, independent of reference cross sections. On the other hand, there is a striking disagreement in the absolute cross section values above 100 keV with several other experiments which are based essentially on a measure-

ment of the ratio $\sigma_{n,\gamma}$ (Au) : $\sigma_{n,f}$ (U^{235}). This holds in particular for the recent experiments of Barry [9] and of Grench et al. [10] which are in themselves consistent but yield, when evaluated with White's [11] U^{235} fission cross section, a gold capture cross section which is always about 15 % higher than our results.

III. The U^{238} Capture Cross Section

The U^{238} n,γ cross section could be determined by measuring the value relative to gold and subsequent normalization, using the data of fig. 2. Since this would introduce several errors - i.e. the errors in the standard cross section and in the ratio measurement would combine - we have determined it by the same method as was described for gold in the previous section. The normalization of the shape curve which had been found by the grey detector - large scintillator tank method was again done at 30 keV. Therefore, an absolute cross section measurement was performed at the Li^7 (p,n) Be^7 threshold [12] , using an activation method and a neutron flux determination by the associated Be^7 activity method. The induced Np^{239} activity ($T_{1/2}$ = 2.346 d) was determined by two independent methods (γ-ray counting with a 30 cm^3 Ge (Li) detector; 106 keV γ-X-ray coincidence counting method), the detectors being calibrated utilizing an absolutely calibrated Am^{243} source. The result at 30 keV (i.e. for a beam with an average energy of 30 keV and a distribution with a half-width of 15 keV) is $\sigma_{n,\gamma}^{(U\ 238)}$ (E_n = 30 keV) = 0.479 t 0 014 barn. The U^{238} capture cross section as normalized with this value is shown in fig. 4. The error is about 5 % at the lower energy limit and increases to 9 % at the upper end.

In fig. 5, we compare our results with evaluated data as taken from recent compilations of Parker [13], Schmidt [14] and Stehn [15]. The agreement with Stehn's data below 100 keV is quite satisfactory; our data are distinctly lower than those of all three compilers in the energy region above 100 keV. The implication of this deviation on some reactor calculations will be discussed in section 5.

IV. Some Renormalizations of Capture Cross Section Data

Using a time-of-flight method and the large liquid scintillator tank, we have measured the capture cross section, relative to gold, for a number of medium weight and heavy nuclides in the energy range 10 keV - 150 keV.

Preliminary data for Ta, W, Mo, In, Re, Hf, Cd, Ag, Pd, Nb and Cs were presented at the 1966 IAEA nuclear data conference [2]. These data have now been reevaluated, using the new gold capture cross section data[2]. Reevaluated data for those nuclei which may be of some interest for reactor calculations are shown in fig. 6. The data are corrected for resonance selfshielding and multiple scattering. Their accuracy (including the standard cross section error) is about 10 % over the whole energy range.

Further measurements of relative cross sections, especially for stable fission products and for lighter structural materials like iron, are presently prepared at our laboratory.

V. Some Implications of the New Data on Fast Reactor Calculations.

In order to check the influence of the new U^{238} capture cross section on reactor parameters, the new data were converted into group constants and included on a trial basis into the KFK-SNEAK-26-group cross section set [16]. This set is based on Schmidt's cross section curves [14]. As can be seen from fig. 5, the Schmidt data are distinctly higher than ours above 100 keV, there is a corresponding decrease of the group cross sections in this energy range.

The modified set was used for calculations of the SNEAK 3-A 1 critical assembly [17], a 670 l - steam cooled U^{235}/U^{238} system. Whereas the unmodified KFK-SNEAK-set slightly underestimates reactivity ($K_{eff} = 0.997$ for a system of critical size), the modified data increase the multiplication factor by about 1 %. At the same time, they lead to a reduction of the conversion ratio by about 5 %. It is possible that the increase in reactivity would be partly compensated if the U^{235} fission cross section was reduced by normalizing measured gold capture to U^{235} fission cross section ratios using our new gold cross section standard. Such investigations are presently underway but it seems premature to draw any conclusions.

In any case, we feel that it is highly desirable to discuss the problems of keV capture and fission cross section normalizations in close connection

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2) Strictly speaking, the measured cross section ratios were evaluated using a gold cross section curve intermediate between the two sets of curves in fig. 3, the difference between this and the grey detector result of fig.2 is, however, unimportant.

with criticality calculations. In particular, interpretations of measurements on pulsed subcritical systems where the ratio of absorption to leakage can be arbitrarily shifted, may be helpful.

Acknowledgement

We would like to thank Dr. H. Küsters for performing the calculations mentioned in section 5 and for helpful discussions.

Tab. 1 Gold Capture Cross Section Values at 30 keV

Method		Reference	Average Value
Flux Det.	Capture Rate Det.		
Associated Activity	Activity	8, 18	598 ± 30 mb
Integral ($MnSO_4$-bath)	Activity	18, 19, 20	596 ± 20 mb
Relative (U^{235}-fission)	Activity	21	608 ± 40 mb
Relative (B^{10}- n,α)	Prompt Capture γ - rays	22, 23, 24	587 ± 21 mb
Shell Transmission		25, 26, 27, 28	614 ± 37 mb

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Figure Captions

Fig. 1 Experimental Arrangement for Cross Section Shape Determination

Fig. 2 Gold Capture Cross Section as a Function of Neutron Energy

Fig. 3 Comparison of Gold Cross Section Shape as Measured with Grey Detector (———), Li^6 Glass Scintillator (o) and B^{10} slab (Δ)

Fig. 4 U^{238} Capture Cross Section as a Function of Neutron Energy

Fig. 5 Comparison of Experimental Values of U^{238} Capture Cross Section with Compiled Values

Fig. 6 Capture Cross Sections for Nb, Hf, Mo, Ta, Cs, W and Re

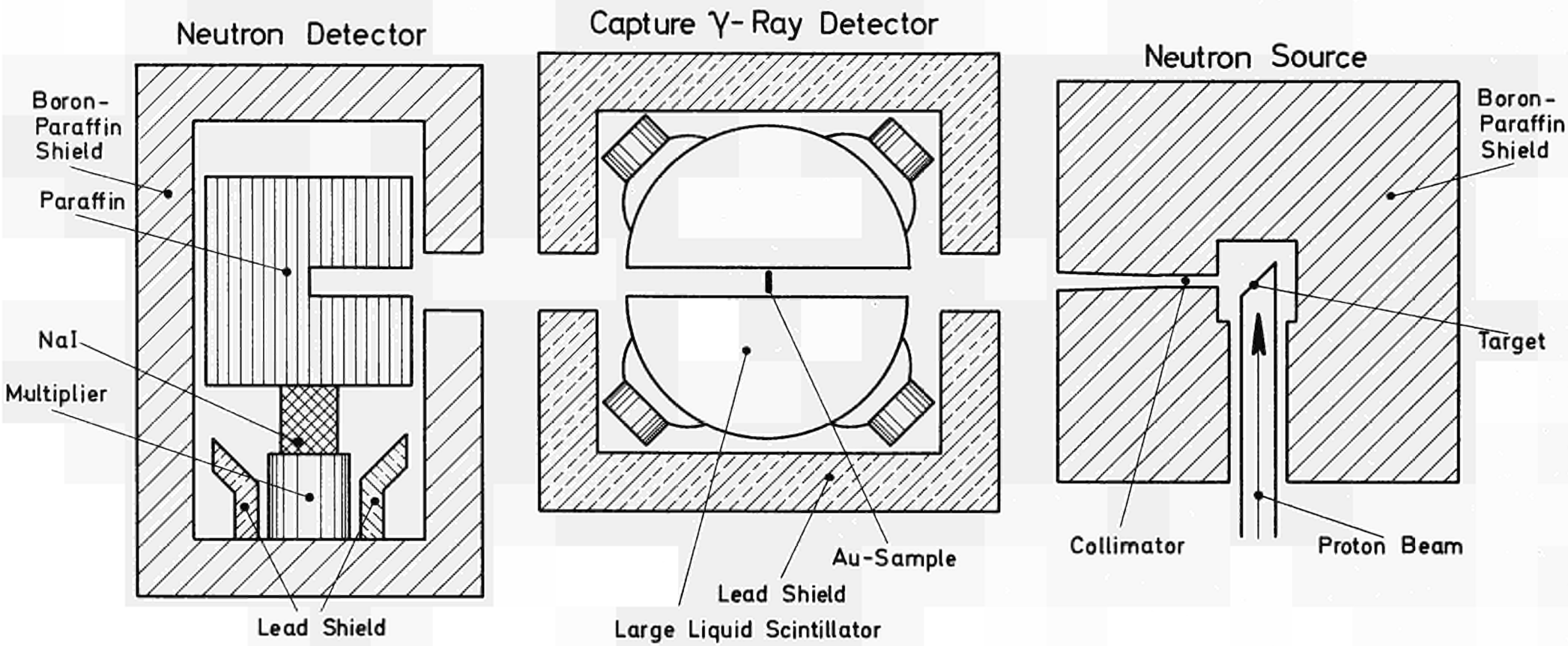

Neutron Detector
Boron-
Paraffin
Shield
Paraffin
NaI
Multiplier
Lead Shield
Capture γ-Ray Detector
Au-Sample
Lead Shield
Large Liquid Scintillator
Neutron Source
Boron-
Paraffin
Shield
Target
Collimator
Proton Beam

Fig. 1

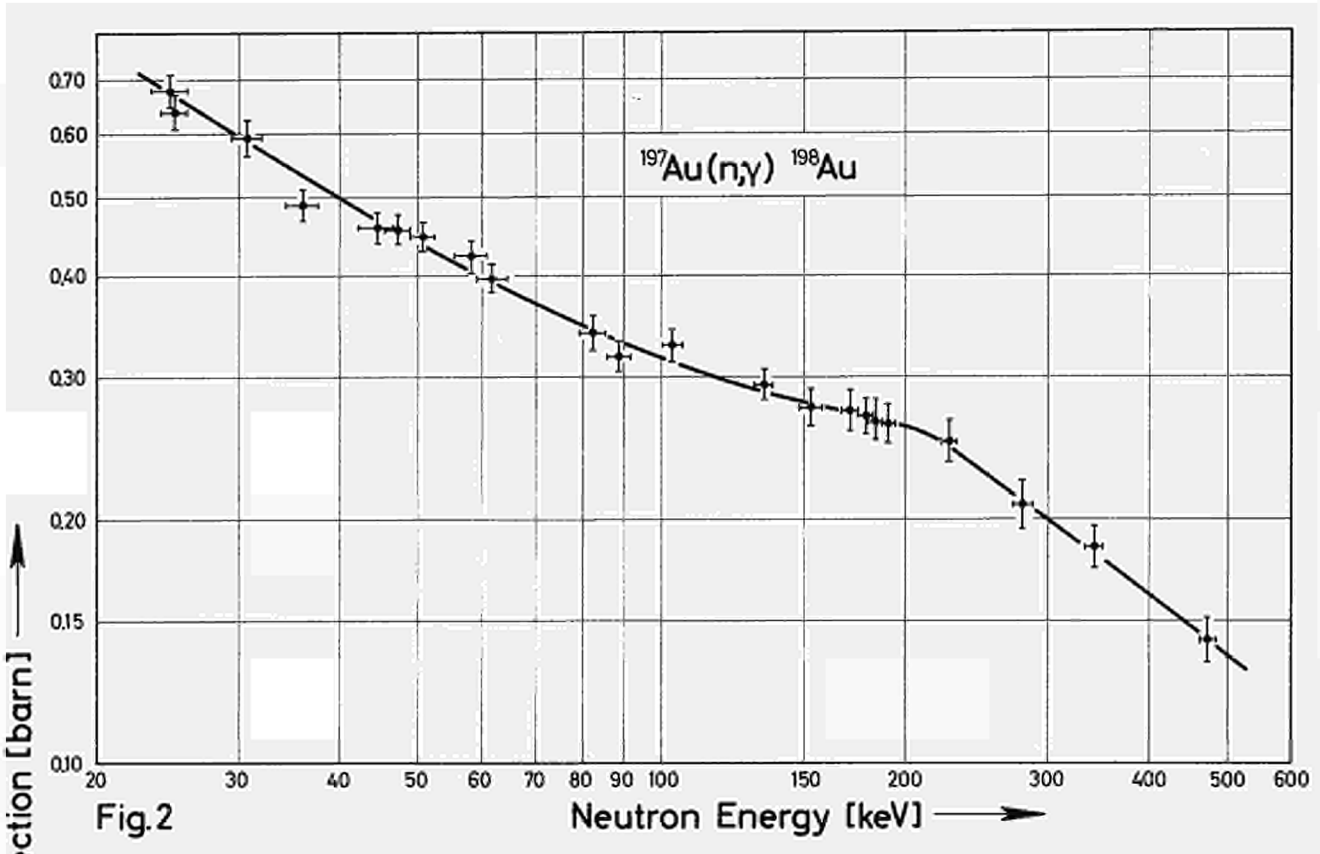

$^{197}Au(n,\gamma)\ ^{198}Au$
Cross Section [barn]
0.70
0.60
0.50
0.40
0.30
0.20
0.15
0.10
20
30
40
50
60
70
80
90
100
150
200
300
400
500
600
Neutron Energy [keV]

Fig. 2

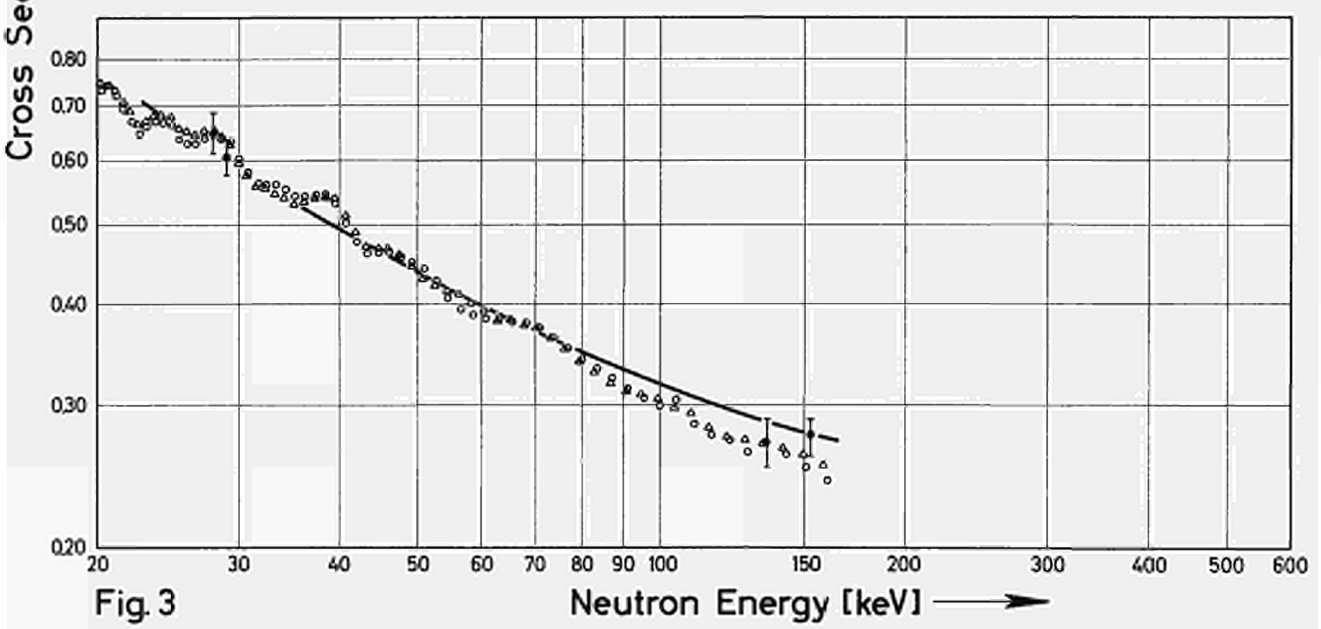

Cross Section [barn]
0.80
0.70
0.60
0.50
0.40
0.30
0.20
20
30
40
50
60
70
80
90
100
150
200
300
400
500
600
Neutron Energy [keV]

Fig. 3

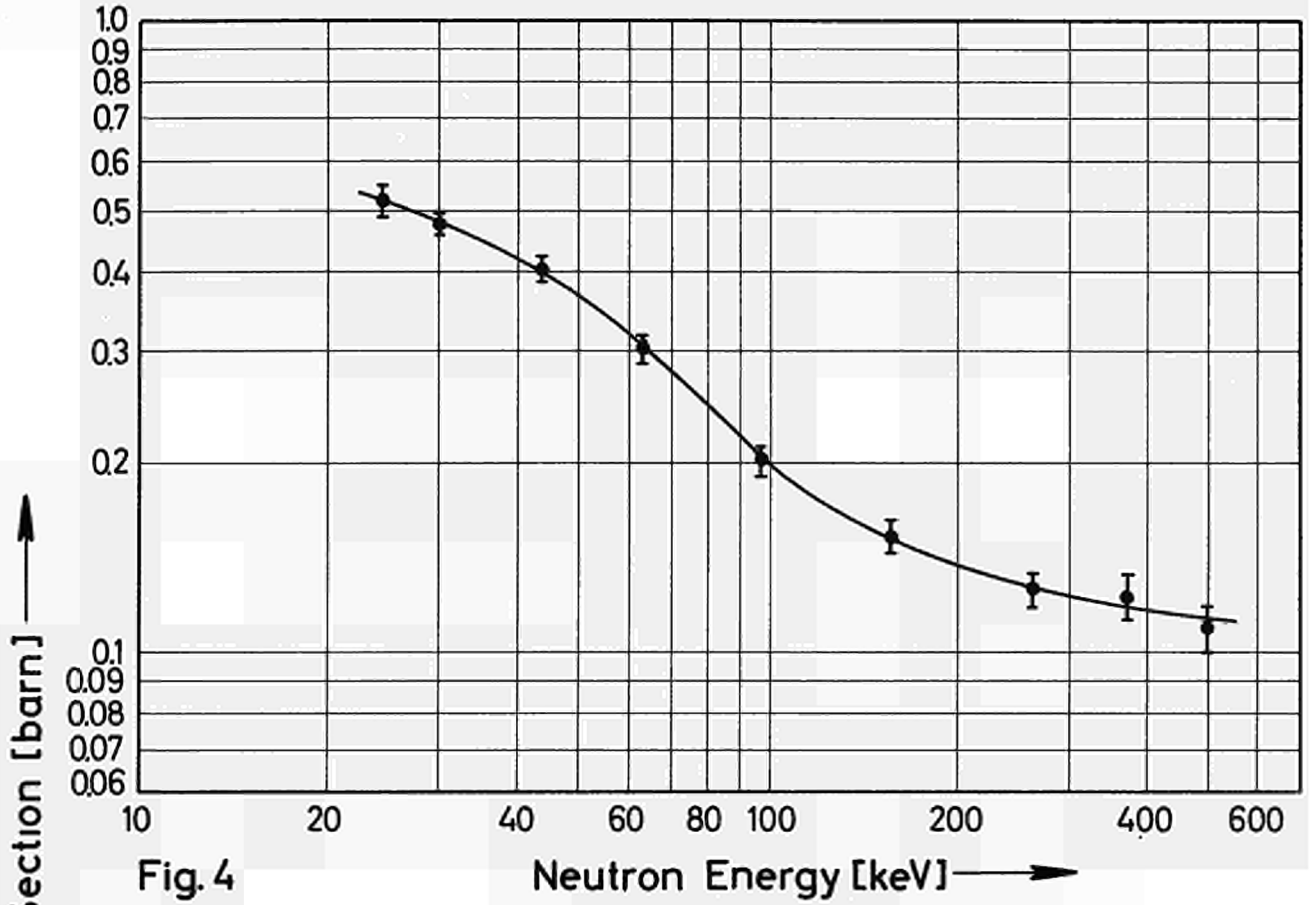

Cross Section [barn]
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0.09
0.08
0.07
0.06
10
20
40
60
80
100
200
400
600
Neutron Energy [keV]

Fig. 4

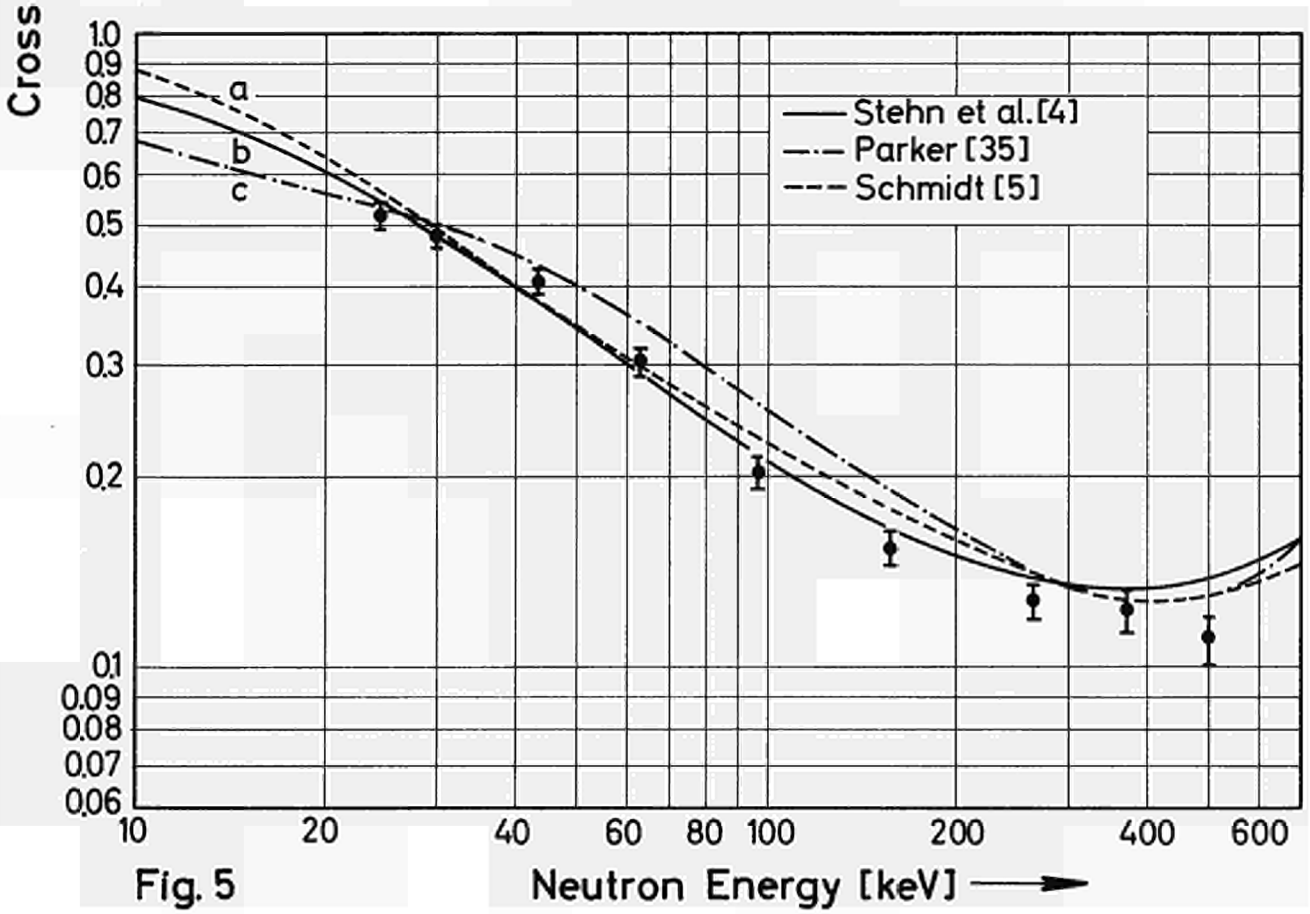
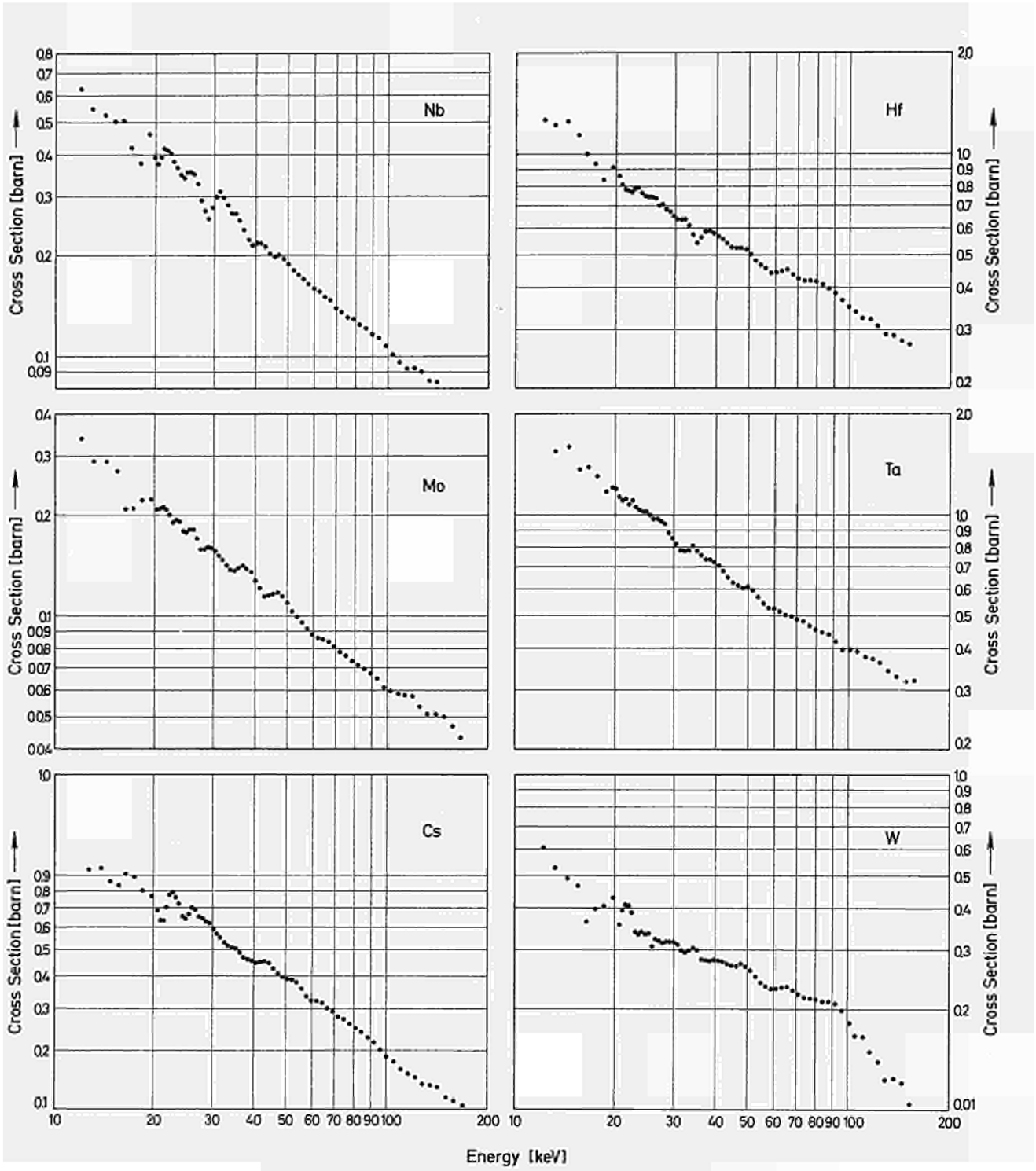

Cross Section [barn]
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0.09
0.08
0.07
0.06
a
b
c
Stehn et al.[4]
Parker [35]
Schmidt [5]
10
20
40
60
80
100
200
400
600
Neutron Energy [keV]

Fig. 5


Nb
Hf
Mo
Ta
Cs
W
Cross Section [barn]
Energy [keV]

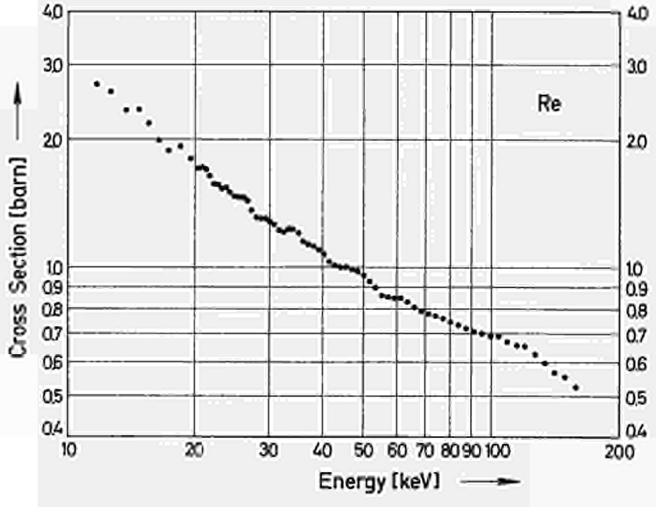

Re
Cross Section [barn]
Energy [keV]

Fig. 6