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PROBLEMS RELATED TO THE RESONANCE ANALYSIS OF ^{235}U

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I- HISTORY

Since 1985 the resonances parameters of ^{233}U[1], ^{235}U[2], ^{239}Pu[3,4] and ^{241}Pu[5,6] were extensively studied by using the computer code SAMMY in a sequential Bayesian analysis of a large experimental data base including old data and recent measurements of neutron transmission, fission and capture cross sections and yields, and eta. The analyses were performed at ORNL and JAERI. The results were incorporated into the main international and national evaluated data files and checked against integral measurements and various integral benchmarks. In particular,the results of the first ^{235}U SAMMY analysis of Leal and De Saussure[2] used in an early version of ENDF/B-VI were not able to reproduce the ratio of the capture to fission obtained from the integral measurements. Lubitz[7] proposed a solution to match the calculated and the measured ratio by modifying slightly the partial widths of the resonances,mainly increasing the capture widths, resulting in some degradations of the representation of the high resolution experimental data. Lubitz results were used in the next version of ENDF/B-VI. But a new evaluation using SAMMY was recommended by the international nuclear data committees. This new evaluation was performed at ORNL in 1994-1995 by Leal et al.[8], and resulted in a new single set of resonance parameters for the energy range up to 2250 eV. The new set removed all the inconveniences of using 11 separated sets and gave calculated cross sections in excellent agreement with the integral data from various benchmark calculations.

For several reasons, the new ORNL evaluation is not yet considered by the international body as the current evaluation to be used in ENDF/B-VI and JEF next versions. However, the new evaluation is already included in a new preliminary version of JENDL. The reasons for the reluctance of CSEWG and JEF committees to immediately consider the ORNL evaluation were mainly due to the criticisms[9] as explained in the next part of this note.

II-THE PROBLEMS CONCERNING THE ORNL EVALUATION-

Some of the problems could be related to the possible differences between REFIT[10] and SAMMY[11] computer codes. The computer code REFIT is used for a simultaneous least square fit of various sets of experimental data. SAMMY uses the Bayes method in a sequential fit of the experimental data. Like SAMMY, REFIT calculates the cross sections using the Reich-Moore formalism or the multilevel Breit-Wigner formalism. The covariance matrix for all the fitted parameters are calculated by both REFIT and SAMMY. In addition SAMMY calculates the covariance matrix for the averaged cross sections. Several tests have shown that SAMMY and REFIT calculate the same cross sections when used with the same parameters, to within a factor of 1 in 10^6 for the nuclear cross-sections and to within a factor of 1 in 10^4 for the Doppler broadened cross sections. In particular both codes are reasonably consistent with NJOY and other reactor processing codes. SAMMY uses the free gas model for the Doppler broadening of the cross sections. REFIT can also use an unproven model to take into account the solid state effect in the samples.

The input to REFIT consists of transmission, capture, fission and scattering yield as functions of neutron time of flight . SAMMY uses the measured data as functions of neutron energy . In both codes the theoretical calculations that are compared with the experimental data include the effects of temperature, finite energy resolution, finite sample thickness, multiple scattering and the effect of the backing for the fission foils. The scattering correction in SAMMY, at present, only includes the corrections for incident neutron scattering. In REFIT the resolution function is calculated by convoluting all the known contributions, using measured parameters and the assumed known shape of each contribution. In SAMMY, this analysis uses a Gaussian function whose variance is the sum of the variance of each contribution and an exponential tail for the description of the asymmetric part of the resolution function. The resolution and Doppler broadening parameters can be adjusted by both codes in order to provide better fits to the experimental data.

The criticisms concerning the last ORNL evaluation are mainly the following:

1/ The ORNL resonance parameters do not reproduce the cross-section in the valleys between the resonances of the fission yield data measured by Moxon et al. at ORELA[12]. In the ORNL evaluation, the effect of the aluminum backing in the fission fragment data was assumed to be negligible. This is true in the regions of the resonance peaks and at neutron energies above a few of 10's of eV, however this is not true in the minima between resonances below ~10 eV

2/ The fluctuations in the ORNL set of radiative capture widths are much larger than expected from the present theory. A spread of only a few percent in the capture widths from resonance to resonance is expected in the mass region of uranium and for the observed spacing of less than 1 eV. Using the program REFIT in the neutron energy region below 100eV a constant radiation width has been found that fits the data to within the experimental statistical uncertainty.

3/ In higher energy range, i.e. above ~250 eV ,where the resonances are called "pseudo-resonances" by the ORNL evaluators, an increasing number of peaks due to more than one resonance are assumed to be single ones. This can lead to large values of neutron and radiation widths, creating problems in the observed distribution of the partial widths, i.e. the distribution of the neutron widths do not agree with a Porter-Thomas distribution. The parameters give only an accurate representation of the cross sections at temperature equal to those of the measurements used in the evaluation.

III- ANSWER TO THE ABOVE FORMULATED CRITICISMS

A-General considerations

The calculation of the Doppler broadening in REFIT using the equation that take into account the solid state effect may give a better fit to the data in the low energy region (<5 eV) than the free gas model. This effect is probably negligible above neutron energies of about 10 eV. At present, this type of Doppler broadening is not incorporated in any of the reactor codes. It may be better to use the nuclear parameters obtained from fits using the free gas model in reactor calculations. An assessment of the impact of the Doppler effect on the cross sections using a more rigorous approach in the prediction of the multiplication factor of the thermal systems has to be made.

In the past, the experimentalist analyzed their own experimental data, often this was a measurement of only one type of cross-section, using various methods of analysis in order to obtain the individual partial widths of the resonances. The observed distributions of the radiation widths, with frequently only a small number of resonances, had a number of degrees of freedom ranging from about 50 to 100 . This observed large spread came mainly from the uncertainty in the experimental data and from the method of analysis (area analysis or shape analysis with Breit-Wigner formalism). These observed resonance parameters obtained from independent analysis of various experimental data in different laboratories were collected by the data centers. These values were then extracted by the evaluators, and, after variety of averaging techniques, used as input for the evaluated data files. It is unlikely that the cross sections calculated from those "recommended resonance parameters" give an accurate representation of any measured cross sections. The situation has been improved by the use of fitting codes such as REFIT or SAMMY to carry out simultaneously or sequentially analysis of large sets of experimental data, old or new, and of different nature. Using these codes it is possible to check the consistency of all the data. However it must be remembered that not all the experimental effects may be taken into account in the analysis. In the case of the fissile nuclei large fluctuations in the fitted capture widths may still exist. These observed fluctuations may be due to the difficulties in measuring the capture cross section or genuine, due the competition between the fission channels and the capture channels. The question is, does the present data warrant the use of values of the capture widths constrained to a narrow distribution, or allow more freedom in the variation of the capture widths? There are many accurate measurements of the total and fission cross-sections for ^{235}U, but there is some doubt about the accuracy of the capture measurements. This doubt is due to the fact that the gamma-ray detector used in the measurements not only detects capture events but also fission events. The correction for the detection of the fission events can be large as the fission cross-section is often an order of magnitude greater than the capture cross-section.

B- Specific problems

1/ The results of the calculations carried out with SAMMY to compare the fission yields of Moxon et al.[12] with the fission yields calculated with ORNL resonance parameters in the energy range up to 50 eV are shown in Figure 1 to 4. As can be seen the calculation is in good agreement with the data around the peaks. The only problems are seen in the regions of low cross-sections between some of the low energy resonances. There are problems in fitting the low cross-section regions both using

SAMMY and REFIT, eg. at 2.7 eV and 4.0 eV where the interference effects in the fission channels create a dip in the observed cross sections that are not reproduced very accurately in the evaluations. Some of the difficulties may be resolved by including a larger correction for the effects of the aluminum backing foils or the self screening effects than were include by the original measurers of the fission fragment data. In the publications the authors state that the corrections for self-screening, multiple scattering, aluminum backing effects were performed on the raw data or were negligible. The samples used in the fission and capture measurements are thin and the use of simple formula to calculate the corrections were thought to be adequate. The problems associated with the determination of the background corrections will be much greater in the regions of low cross-sections. In most cases the normalization can be adjusted in the fits to the data and is generally within the limits given by the experimenters. In the region below about 20 eV where the resonances are well resolved, the effect of the experimental resolution is negligible, the problems of fitting the data may be due to the inadequacy in the use of the formalism that has only two fission channels. There are certainly more than two open or partially open fission channels for each spin state of the s-wave resonances. It is only in this low energy region that it may be possible to separate out the fission widths for the individual channels from the observed interference effects where the experimental effects are thought to be small. In some of the fits to the more accurate data there may also be some evidence of the presence of more than two fission channels. However, it could be difficult to separate the interference effects from other physical effects (in particular hidden small resonances) or unknown experimental effects.

2/ In the energy range from 4 to 100 eV the capture widths smaller than 30 meV or greater than 60 meV were set to the average value of 40 meV and a re-fit to the experimental data carried out using SAMMY, for the corresponding resonances. The fits to the data were good in the region of most peaks in the capture data. But for 7 of resonances the fits were poor.An example is given in fig.5.The poor fit for these resonances were also noted in the evaluation using REFIT.These resonances have small values of the capture cross section at the peak and small errors in the correction for the fission contribution will have a large effect on the capture width. The alternative explanation is that these few resonances have much smaller radiation widths than the average.Another example is given in fig.6 in the energy range from 125 eV to 150 eV where the SAMMY fits were carried out by starting with a capture width of 40 meV with a constraint of 2% for all the resonances;the large peaks are quite well reproduced with capture widths ranging from 37 meV to 43 meV;but the capture widths obtained for the small resonances of the energy range from 131 eV to 135 eV are between 32 meV and 35 meV and the peaks are not well reproduced, smaller values of the capture width are needed.

3/ In the energy from 100 eV to 500 eV, statistical methods ,mainly Delta3 statistics, were used by Leal et al. to put in missing levels so that the average spacing of the resonances was about the same as that observed in the energy below 100 eV. This technique does not give a unique set of resonances but gives a better representation of the cross-section and conserves some of the statistical properties of the parameters, i.e. the Wigner distribution of the resonance spacings and the Porter-Thomas distribution of the reduced neutron widths. Fits were carried out using SAMMY on the transmission data of Harvey et al.[13] and on the fission data of Weston et al,[14,15]to find the neutron and fission widths for all of the resonances, and radiation widths for most of them. The average values of the capture cross section calculated from the fitted parameters were compared to the average values of the experimental capture data. The experimental data are well represented and the average calculated capture data are in fairly good agreement with the experimental data of Perez et al.[16].

In the energy region between 500 eV and 2 keV,the average level spacing of the pseudo-resonances varies from 0.55 eV to about 1 eV, which means that an increasing number of unresolved multiplets are present in the calculated cross sections. Due to a lack of time it was not possible for Leal et al.[2]. to increase the number of pseudo-resonances for the calculation of the cross sections. It is obvious that in this energy range the statistical properties of the resonance parameters are not conserved. In particular,the unresolved multiplets give large values of the corresponding reduced neutron widths, not consistent with the Porter-Thomas distribution. The cross-sections calculated from these parameters are in good agreement with the experimental transmission and fission data. The energy range between 2 keV and 2.25 keV was a test case in which Leal succeeded in representing the experimental data with a number of pseudo-resonances with an average spacing close to the one in the well resolved energy range. However this work was time consuming and for this reason could not be performed in the energy range 500 eV to 2 keV.

The idea of extending the "resolved resonance region" as far as possible in energy came from G. de Saussure who estimated that a set of pseudo-resonances ,which could represent with good accuracy the structure of the measured cross sections, will allow better calculation of the self-shielding factors, compared to the methods used in the so-called unresolved range (statistical method of sampling with average resonance parameters, etc...), even at temperatures different of the temperature of the experimental data. The number of pseudo-resonances used for the description of the experimental data and the parameters of these resonances could play a role in the accuracy achieved on the self-shielding factors. Calculations should be performed to check the accuracy of the different methods.

IV- CONCLUSIONS

It is valuable to ask different qualified people in different laboratories using different computing codes to check for possible errors or inconsistencies in the results of an evaluation, both on microscopic and integral point of view. For these reasons Leal et al. made a wide distribution of the new ORNL ^{235}U evaluation in the resonance region. The feedback concerning the consistency with integral data show excellent results. Only one review of the resonance parameters was available[9]. This review concerns both the accuracy of the cross section calculations and the individual values of the resonance parameters. Search on the accuracy of the cross section calculations shows that REFIT and SAMMY give exactly the same results for the unbroadened cross sections. For the Doppler broadened cross section, differences of less than 0.01% in the peaks and less than 0.1% in the low cross section regions between the peak of the resonances are observed.

The comments on the values of the ORNL resonance parameters and on the quality of the experimental data which were analyzed could have been misunderstood in different national or international meetings on evaluation problems. One example is the discrepancies between the fission yields in the valleys of the resonances. These discrepancies appear only in a small energy range at low energy and the origin of the discrepancy is not clear. The problem of the large spread of the capture widths from resonance to resonance in the evaluation will be reconsidered at ORNL by constraining the capture widths at values close to an average values. New SAMMY calculations are in progress in the energy range up to 500 eV. About 10% of the resonances can not be fitted in the experimental capture data by using capture widths close to the average value. At energy above 500 eV we should,again,point out that the parameters are pseudo parameters which should be used for accurate calculation of the self-shielding factors. They give an accurate representation of the measured cross section at the temperature of the measurements. Up to 500 eV the statistical properties of the parameters agree with the Wigner distribution of the resonance spacings and with the Porter-Thomas distribution of the reduced neutron widths.

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FIGURE CAPTIONS

Fig.1 to 4

The fission yields are given versus the energy in eV.The experimental data are given with the statistical errors.The solid lines are the data calculated from the ORNL-SAMMY resonance parameters.

Fig.5

The capture cross sections are given versus the neutron energy in eV.The experimental data are given with the experimental errors.The solid line is the capture cross sections calculated by SAMMY.The figure shows very bad fits for 3 resonances whose capture width was fixed at 40 meV in the SAMMY fit.

Fig.6

The capture cross sections are given versus the neutron energy in eV.The crosses represent the experimental data points.The solid line is the capture cross section calculated by SAMMY from a fit with a constraint of 2% on all the capture widths.

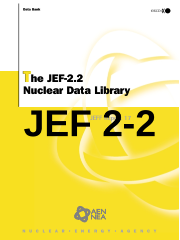
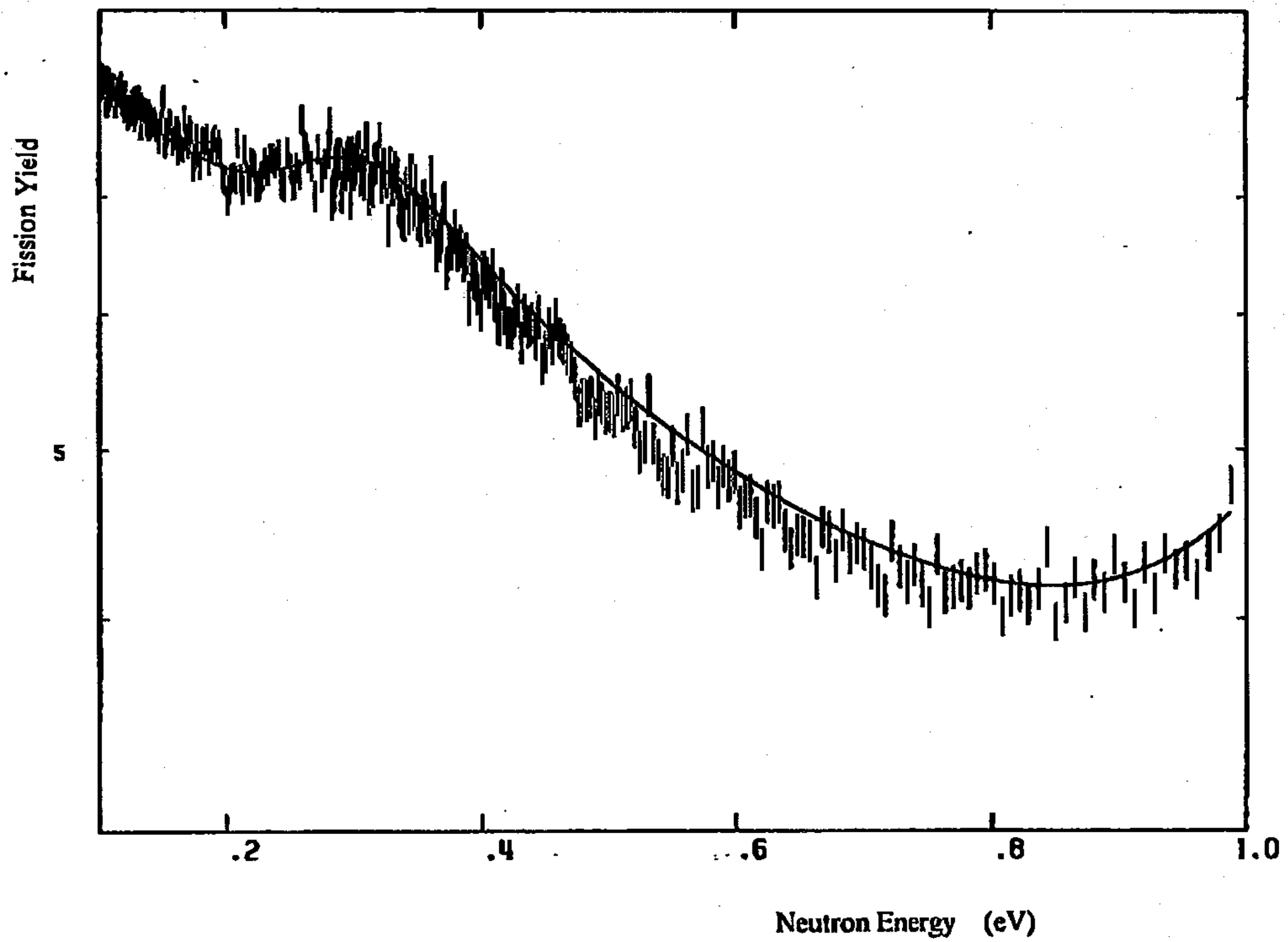


Fig. 1



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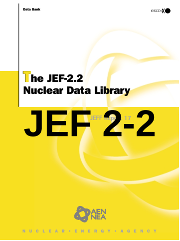

Fig.2

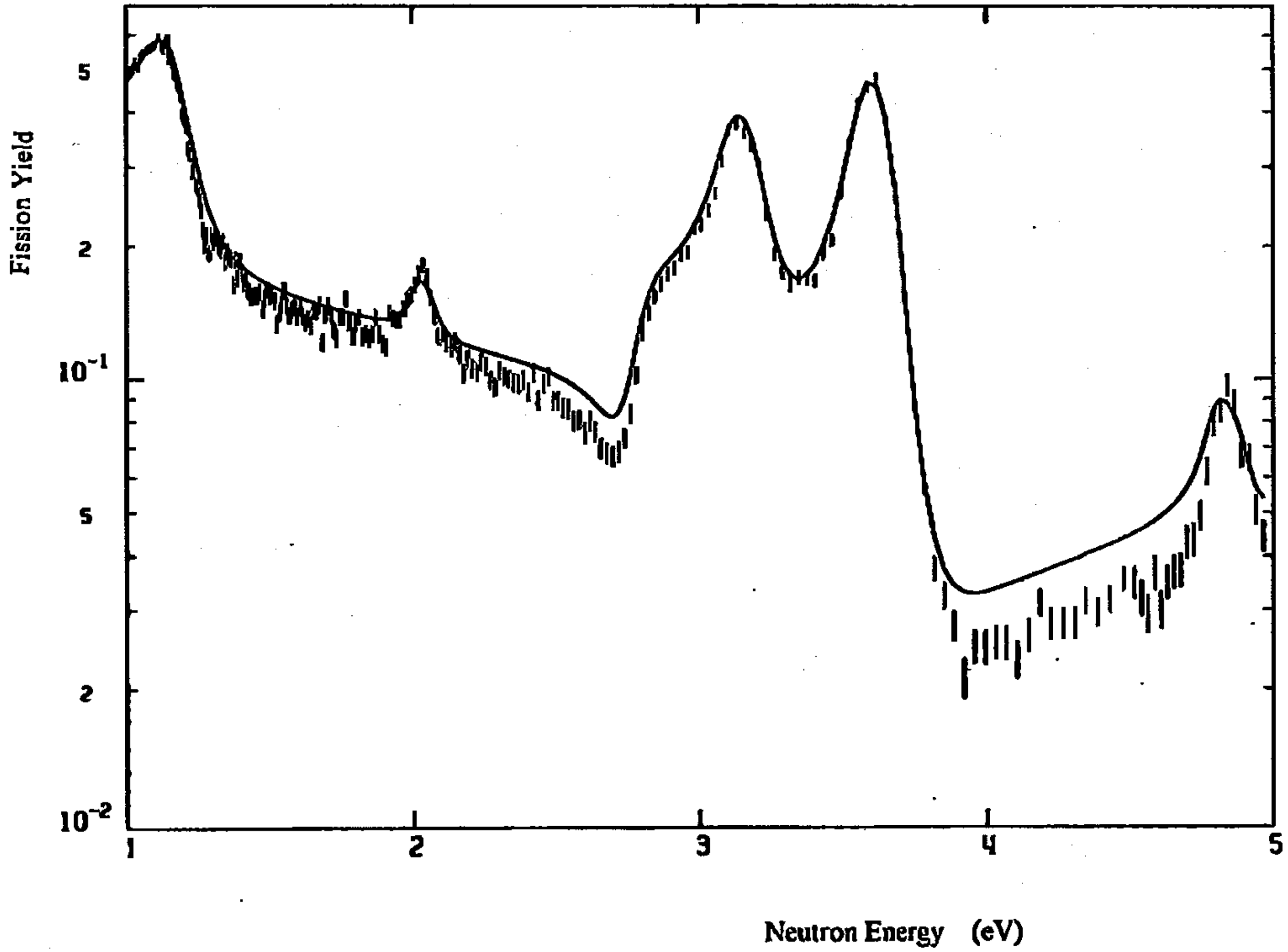

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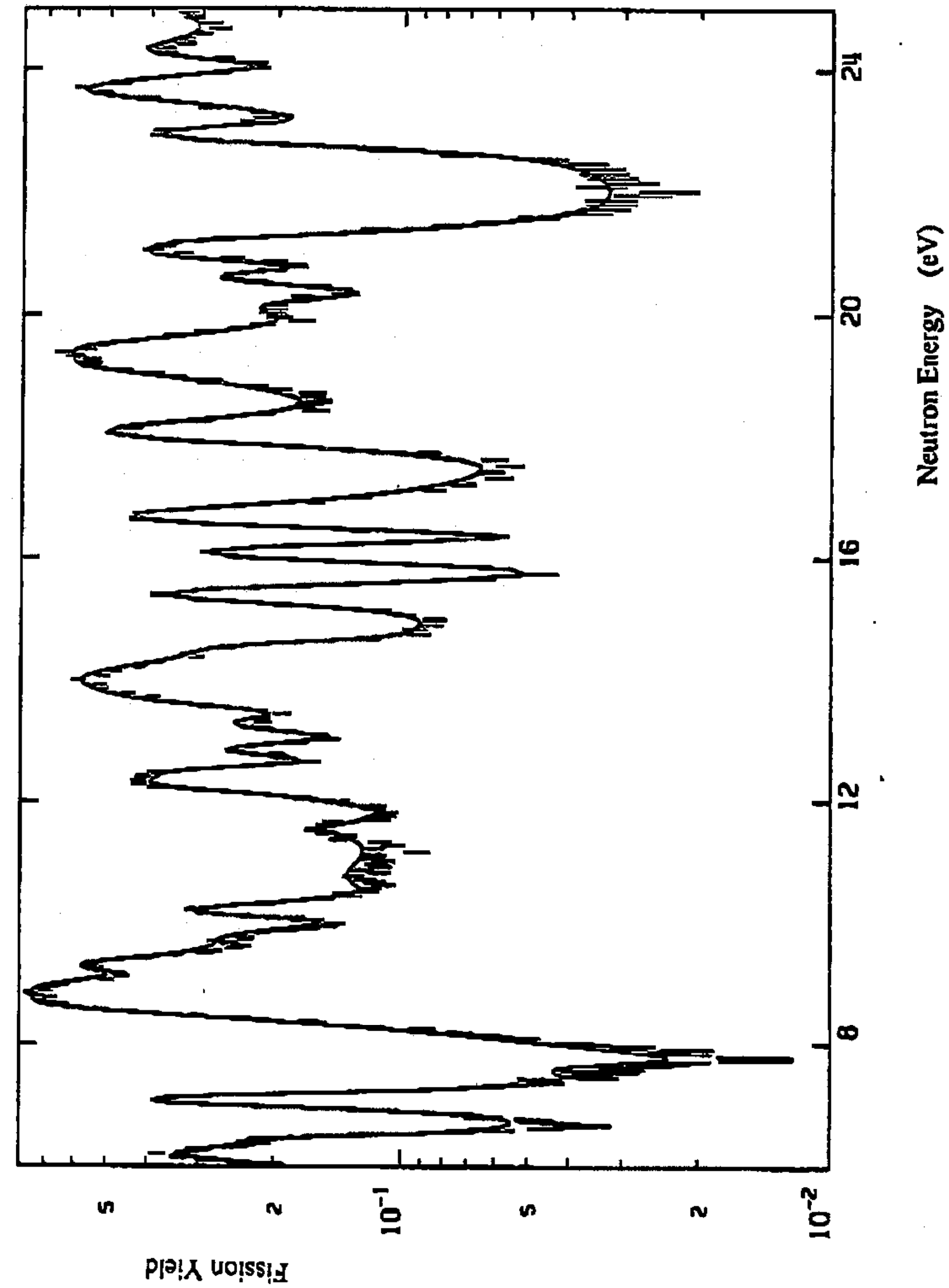
Fig.3



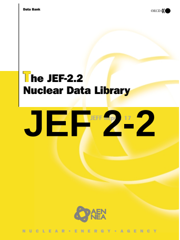

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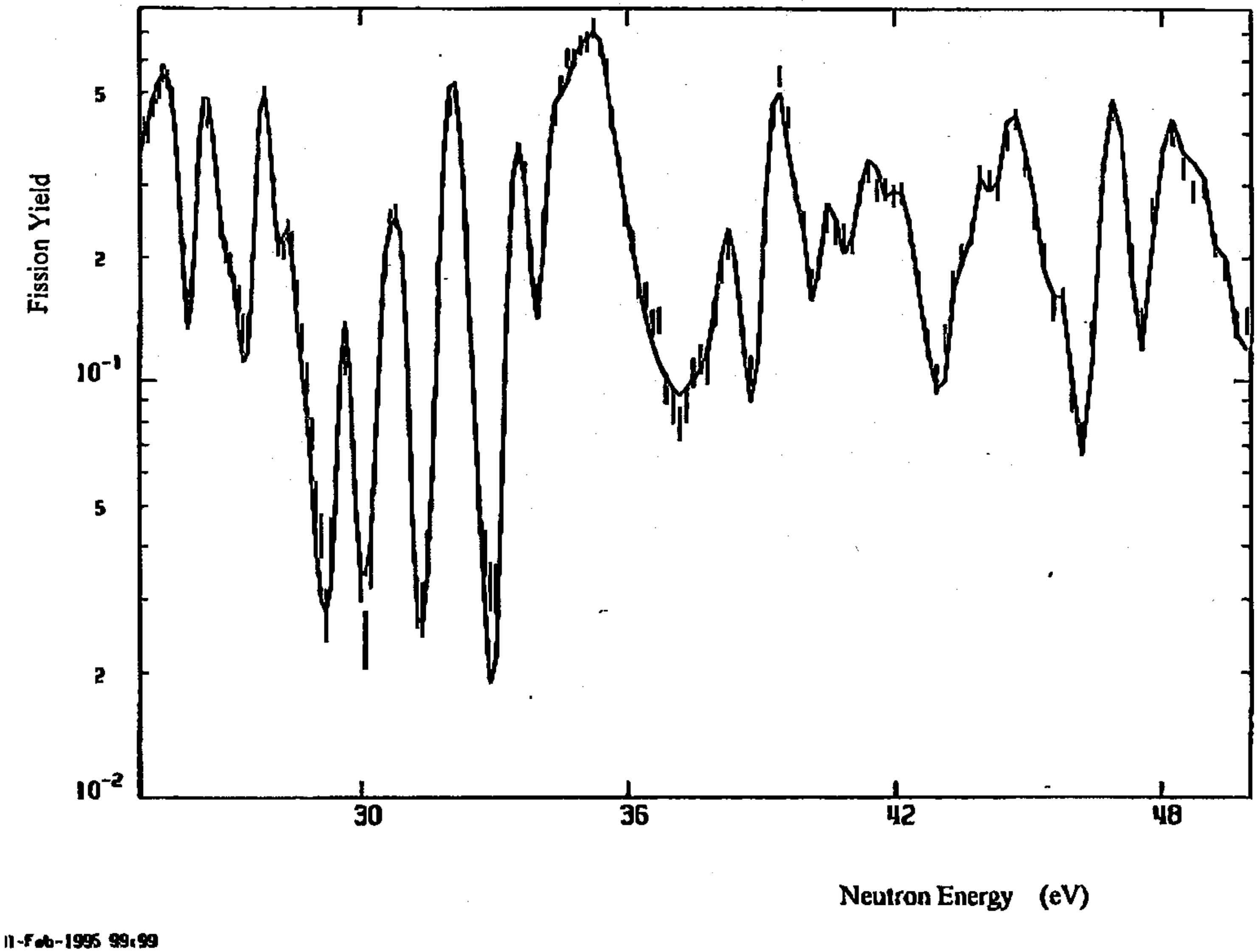
Fig.4


Fission Yield
5
2
10^{-1}
5
2
10^{-2}
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36
42
48
Neutron Energy (eV)
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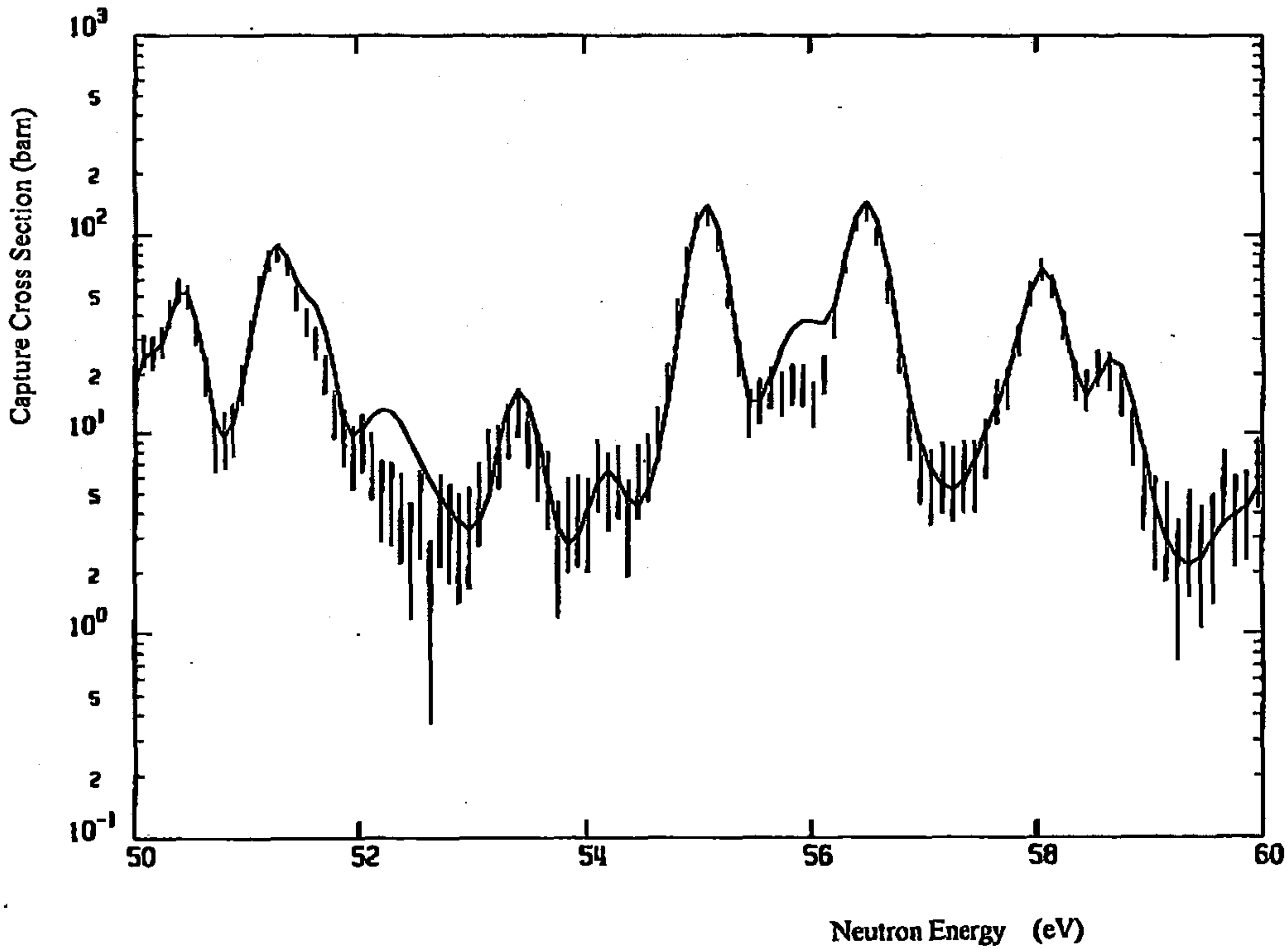
Fig.5



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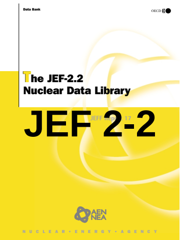
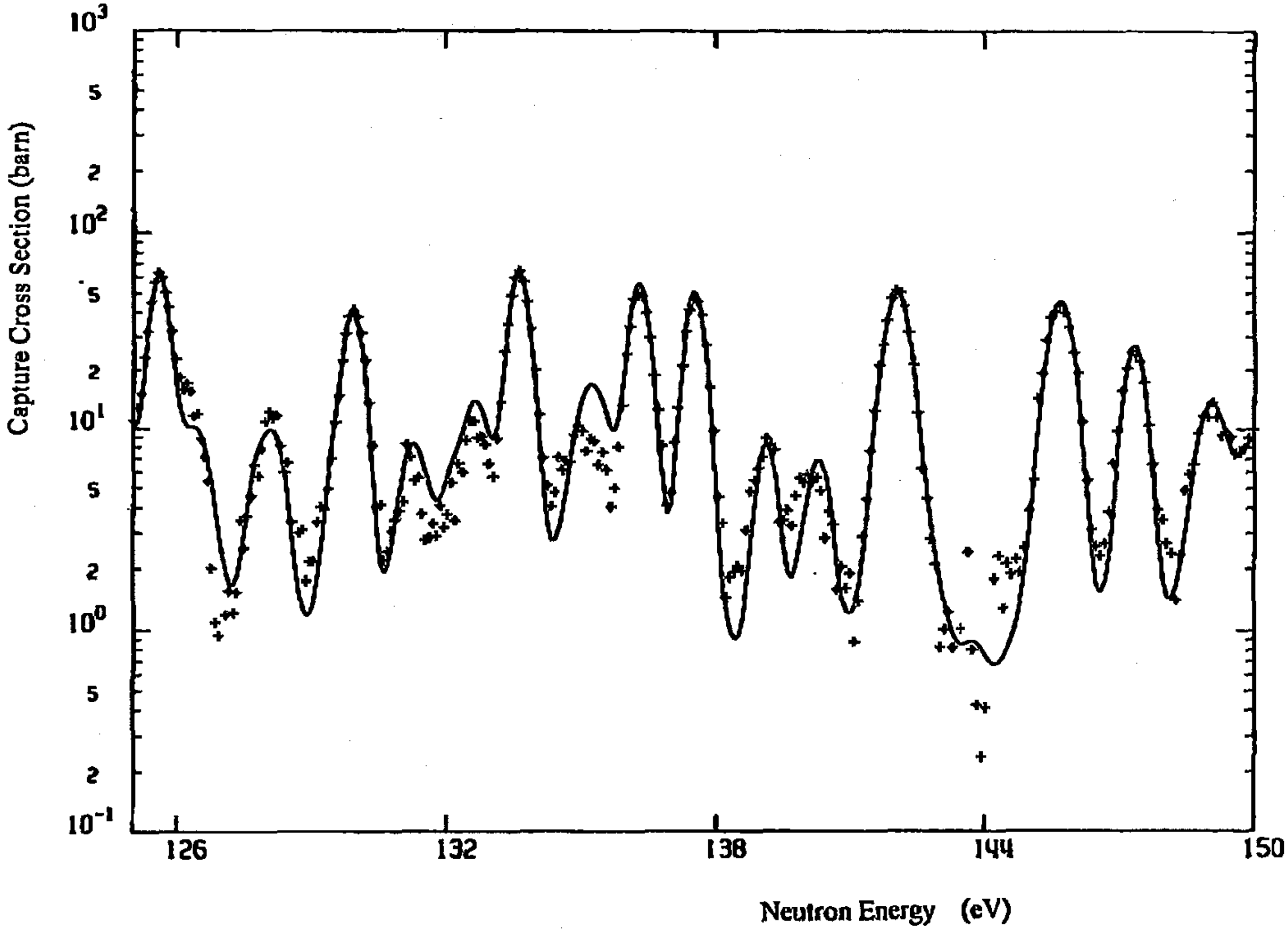


Fig.6



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