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FAST NEUTRON CAPTURE CROSS SECTION MEASUREMENTS, EVALUATIONS AND MODEL CALCULATIONS OF FISSION PRODUCT NUCLEI

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FAST NEUTRON CAPTURE CROSS SECTION MEASUREMENTS, EVALUATIONS AND MODEL CALCULATIONS OF FISSION PRODUCT NUCLEI*

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ABSTRACT

The fast neutron capture cross sections of elemental rhodium, palladium, neodymium, and samarium were measured in the energy range 0.5 - 4.0 MeV relative to the standard capture cross section of gold. A large liquid scintillator and the time-of-flight technique were used in these measurements. Experimental data are rare or non-existent in this energy range and evaluations differ substantially, with a factor of 5 being common. The present data were used together with other experimental data and nuclear model calculations in order to provide a consistent set of isotopic and elemental capture cross sections.

INTRODUCTION

The fast neutron capture cross sections of the more frequently occuring fission product nuclei play a significant role in the calculation of fast reactor reactivity, after-heat and sodium void effects, and are important for spent fuel-handling considerations. A 1% uncertainty of the calculated reactivity would result from a 30% uncertainty of these cross sections./1/ This led to uncertainty requests of ±10% for fast neutron capture cross sections of fission product nuclei between 1 keV and 10 MeV./2,3/ Whereas such requests may be modified to allow for larger uncertainties in the higher keV and MeV range, it is obvious from the consideration of existing (or nonexisting) data and evaluations that uncertainties are largest at higher energies. A recent comparison of evaluated neutron capture cross sections of 27 fission product nuclei at 2 MeV shows differences of a factor of 5 to be common while factors of 10 or more are encountered./4/

The fast neutron capture cross sections of fission product nuclei are a prime example for the proposition that nuclear model parameters can be determined with experimental data of some nuclei and cross sections for other nuclei can be calculated subsequently with these parameters. It has been observed that substantial differences exist between such calculated cross sections for nuclei where experimental data do not exist./5/ A similar

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observation can be derived from a table containing 30 - keV capture cross sections for 27 fission product nuclei./4/ This table compares five different evaluations with experimental values where such are available.

Some remarkable improvements of the evaluated capture cross sections have been obtained by using integral measurements as a constraint./6,7/ However, such integral data are more likely to help in the normalization of cross sections in the lower-keV range where evaluation differences are less pronounced than for the MeV range.

Present measurements were carried out in the 0.5 - 4.0 MeV range for Zr, Mo, Sb, Nb, Y, In, Rh, Pd, Ag, Cd, Ho, La, Yb, Sm, Nd, Gd, Dy, Eu, Tb, Er, Hf, W, Re, Ta, Au, Th, and U. Data for Ho, Nb, Ta, Au, Th and U were reported previously and the data for Rh, Pd, Nd, and Sm are presented here. Isotopes of the latter elements are among the 20 most important fission product nuclei./1,8/ The present data should provide a useful constraint for the evaluation of these cross sections.

MEASUREMENTS

The $^{7}Li(p,n)^{7}Be$ reaction was used as a neutron source with the primary proton beam accelerated with the Argonne National Laboratory Fast Neutron Generator (FNG). The proton beam was pulsed with a repetition rate of 2 MHz and bunched to ~1 nsec. Metallic lithium was evaporated onto a 0.025 cm thick tantalum backing with a thickness appropriate to provide neutrons with energy spreads betwen 50 keV and 100 keV in the energy range from 0.5 to 4.0 MeV. The neutron energy was determined from the known kinematic relations, the primary proton energy (calibrated with $^{7}Li(p,n)$- and $^{10}B(p,n)$-thresholds), the stopping cross sections and the target thickness as measured near the $^{7}Li(p,n)$-threshold.

The neutron source was surrounded with a 4π-lithium-baron-polyethylene shield in order to reduce γ-ray and neutron background for the capture and neutron detectors. A conical opening in the 4π-shield provided a well collimated neutron beam which penetrated the capture sample after a flight-path of 250 cm. The neutron beam was totally captured in a neutron monitor at a flight-path of 380 cm.

The capture samples were metallic discs with a diameter of 8.0 cm and they ranged in thickness from ~0.012 at/b (Rh, Pd) through ~0.006 at/b (Au) to ~0.005 at/b (Nd, Sm) and were at least 99.9% chemically pure. They were placed in the center of a 1300 liter large liquid scintillator tank which approximated a sphere in shape and was well shielded by low-activity iron, 5 - 15 cm of lead and 60 cm of concrete. The scintillator was a mixture of pseudo-cumene, p-terphenyl, POPOV, and baron-methyl. The scintillation light was detected with 12 8"-photo multipliers which were uniformly spaced over the surface of the tank. The timing of all multipliers was carefully matched and a rise-time correction was applied with an on-line computer resulting in a time-resolution of ~3 nsec. The gamma-ray energy resolution was 26% for the ^{60}Co decay gamma rays. The time-resolution and flight-path of the present experiment permitted the separation, by time-of-flight, of capture events produced by the second neutron group of the $^{7}Li(p,n)$-reaction up to ~3 MeV. The electronic threshold for the detection of capture events was usually set at ~2 MeV and $(n,n'\gamma)$ events were rejected for higher primary neutron energies with digital selection in the on-line computer system.

A grey neutron detector/9/ was used as a neutron monitor. Specific features of this detector are not of great importance for the present experiment, because the measurements were made relative to the capture cross section of gold.

Measurements were carried out between 500 keV and 4 keV with irradiation times in the range of 1 to 2 hours. With exception of palladium two sets of measurements were made. These yielded consistency which was usually better than 5%.

The time-of-flight spectrum and the energy pulse height spectra for the capture events corresponding to the neutron capture peak in the time-of-flight spectrum and for an equally spaced adjacent time interval were stored with the on-line computer./10/ Background was subtracted from the time-of-flight spectrum and additional measurements were made with carbon-samples, without a sample, and with a plugged collimator hole. These measurements showed that below 2 MeV all background was ambient, and non-ambient background above 2 MeV lead to only a small correction. The measured energy spectra were extrapolated to zero pulse-height. The neutron energy dependence of this extrapolation could be described very well by a simple analytical representation of the spectra with a weighted neutron binding energy for the elemental samples (within 1-2%). Inelastic scattering events were eliminated by setting appropriately higher integration limits in the energy pulse-height spectra. The capture detection efficiency was in the range of 65 - 85%. This is the major limiting factor of measurements with a large liquid scintillator because the exact shape of the energy spectra below 2 MeV cannot be determined due to high background.

The major correction besides the detection efficiency is attributed to capture events produced by neutrons which scatter once or more within the sample. Such corrections may be large due to the small size of the capture cross section relative to scattering cross sections, to the energy loss due to inelastic scattering which increases the capture probability due to the substantially higher capture cross sections at lower energies, and to the increase of the average path through the sample due to both elastic and inelastic scattering. This correction was calculated with a Monte Carlo Code which is exact at lower energies but uses approximate solutions at higher energies where details of inelastic level structure are not known or the scattering data is unavailable, and the complexity due to the many isotopes involved is enormous. Other corrections were applied for the flux attenuation in the samples and the secondary components of the neutron beam.

The general experimental procedure and the corrections have been discussed in more detail at previous occasions./11-14/ The results from the present measurements are shown in Fig. 1. The ENDF/B-V standard capture cross section of gold was used as reference. The present data for palladium, neodymium and samarium provide the sole source of experimental cross section information for these nuclei since no previous data exist in this energy range. Previous data are available for rhodium and the more recent data are compared with the present results in Fig. 2. All data shown in Fig. 2 were obtained with the prompt detection technique. The data by LeRigoleur et al. /18/, Macklin/15/, and by Drake et al./16/ were obtained with the spectra weighting technique, the data by Knox et al./17/ were, as were the present data, obtained with a large liquid scintillator. The present data agree reasonably with the recent results by Macklin but indicate, as do all other data shown in Fig. 2, lower cross sections (~10 - 15%).

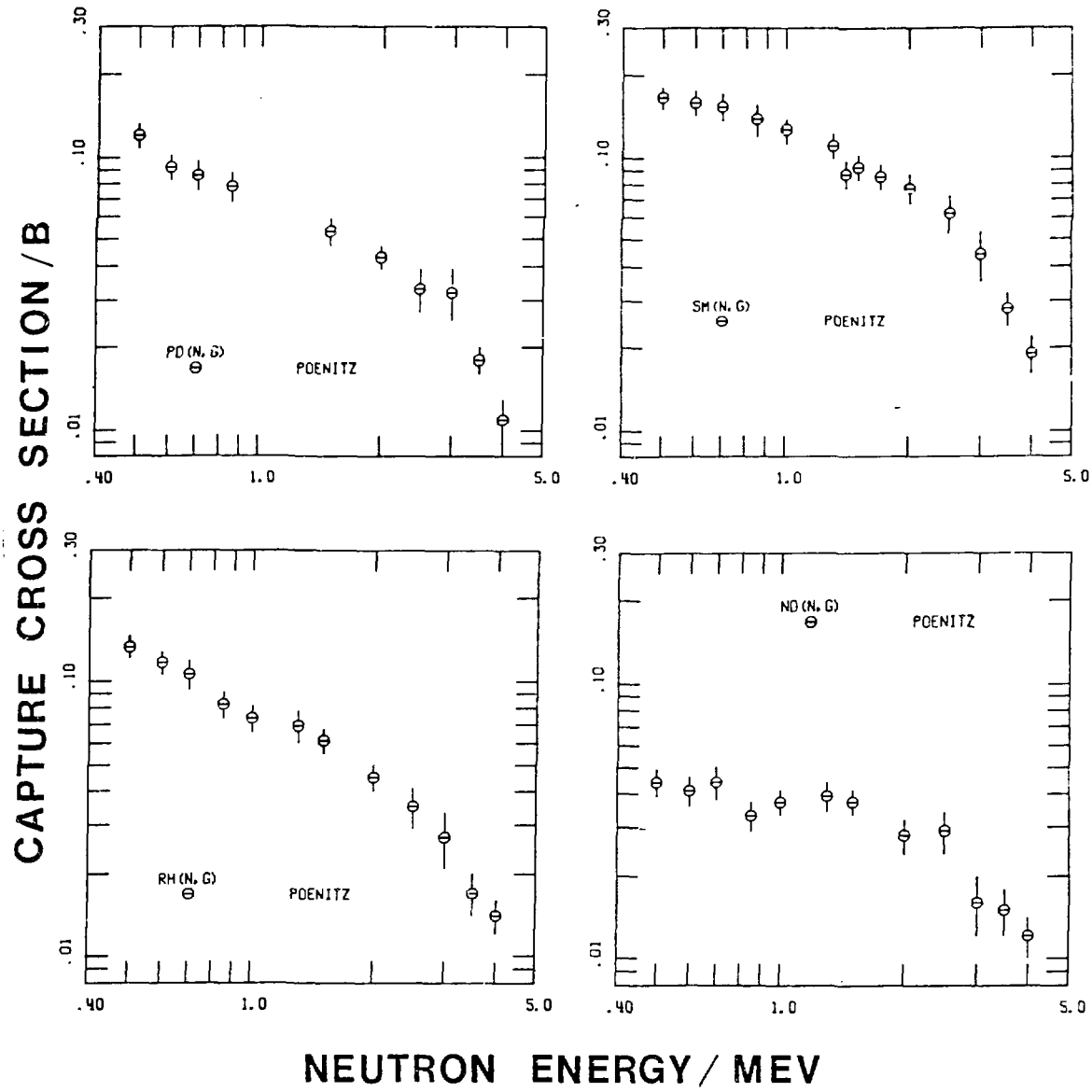

Fig. 1. Experimental Results from the Present Measurements.

DATA INTERPRETATION

The average capture cross sections of medium and heavy nuclei can be calculated in terms of the statistical model and the optical model. Since the bulk reactivity effects of fission product nuclei are of major interest, one might expect that satisfactory results from such calculations can be obtained if experimental elemental capture cross sections are used as a constraint in such calculations. Recent model calculations of capture cross sections for heavy actinides have yielded good results and a similar approach was used in the present calculations. The statistical and optical model code ABAREX/19/ was used in the present calculations. A major concern is the large number of parameters available to adjust the calculated cross section used to represent the experimental data. It cannot be expected that a parameter set derived only from fitting some experimental capture cross sections will reasonably describe capture cross sections of other nuclei for which experimental data are unavailable. Additional experi-

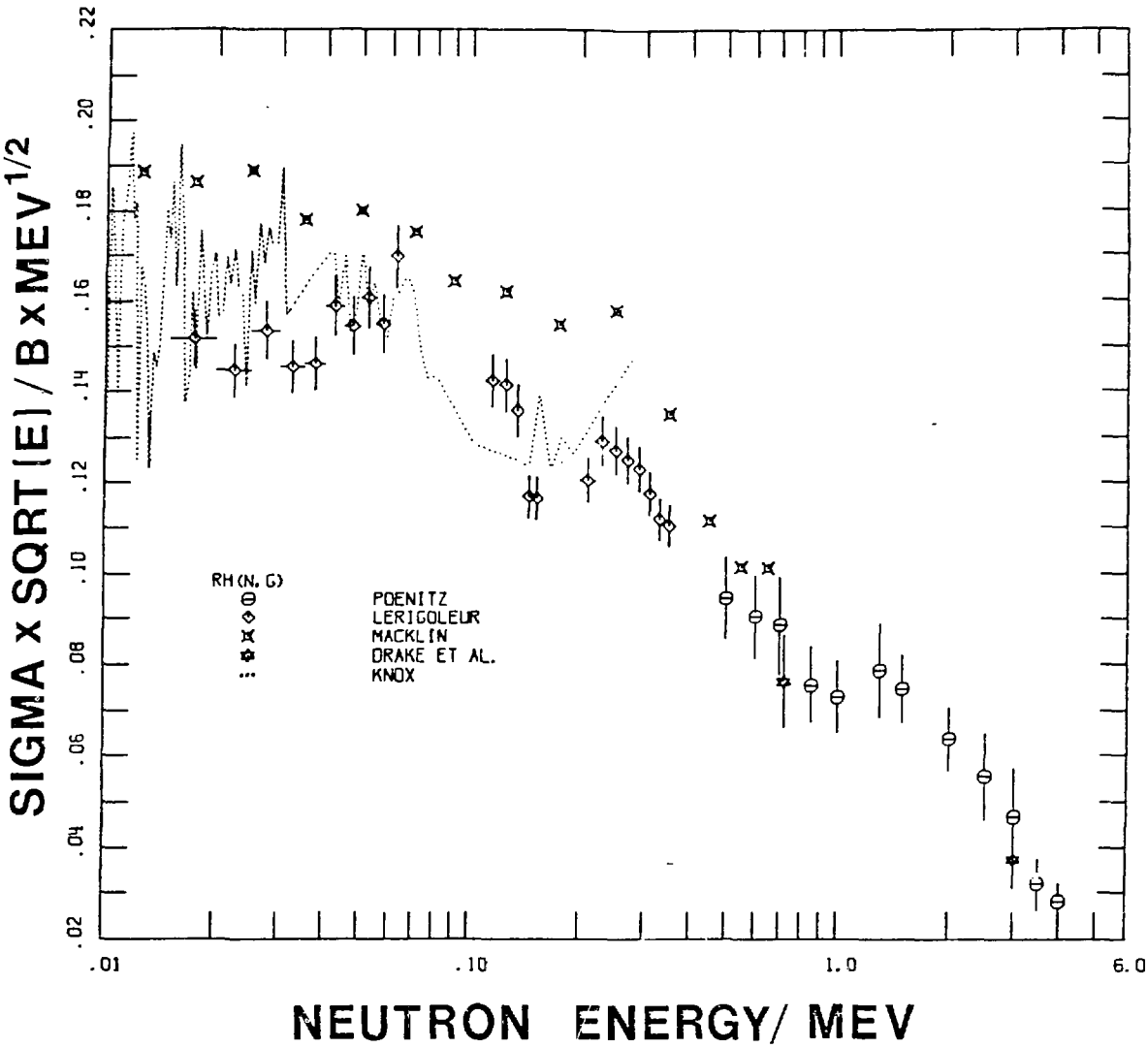

Fig. 2. Comparison of the Present Results with Other Recent Data of Rh(n,γ).

mental information will have to be used to establish some of the parameters. Total, and elastic and inelastic scattering cross sections were used in the previous calculations to establish the optical model parameters./5/ The major parameter sets to be determined are:

Optical Model Parameters

These can be determined by fitting total cross sections and angular distributions of elastically and inelastically scattered neutrons, polarization data etc. ABAREX uses a spherical optical model which was shown to represent well also deformed nuclei if the deformation of these nuclei is similar. However, parameter sets cannot be derived which would have global validity and ultimately a deformed nuclear model should be used. There is a substantial lack of neutron scattering and total cross section data in the range of the fission product nuclei (e.g. Pd, Nd, considered here) which hampers the determination of optical model parameter sets.

Low Energy Levels of the Target Nucleus

Inelastic scattering has the most pronounced effect on the calculated capture cross section. In the range of major importance of these cross sections (< 1 MeV), the levels structure is now fairly well known and the information given in Ref. 20 was used in the present calculations.

Target Level Density

Most level density formulas, for example the one derived from the Fermi-gas model, assume high excitation energies of the nucleus and cannot be expected to represent very well the true level density at low excitation energies./25/ Thus, the level density of the target nucleus is one of the more uncertain quantities of such calculations and can be adjusted to represent the measured data well. ABAREX used the Gilbert and Cameron/21/ formalism and the level density of the target nucleus is represented by $\rho \sim \exp((E-E_0)/T)/T$.

Compound Nucleus Level Density

The level density of the compound nucleus is represented in ABAREX, following Gilbert and Cameron, by a Fermi-gas model. The parameters, a and σ, may be adjusted to yield the experimental average level spacing measured for s-wave resonances at low neutron energies. However, the available data needed for this purpose are sparse and uncertain.

Average Radiation Width

The average radiation width can be calculated with an appropriate level density formula and a giant-dipole resonance for the - transition probability. The parameters for the γ-transition probability can be determined from (γ,n)-cross section measurements, however, it is common to utilize Γ_γ/D values determined in the low eV energy range for resolved resonances. Such data are sparse and substantial uncertainty exists. Γ_γ/D values are, for example, only available for one of the palladium isotopes (105). The high resolution data from RPI/17/ for Rh (Fig. 2) show substantial fluctuation of the cross section. Such fluctuations are now well known and were discussed for ^{238}U and ^{232}Th in a recent review./5/ The implications of these fluctuations are the existence of local values of Γ_γ/D and the neutron strength function and they cast doubt at the use of resolved resonance parameters for the calculation of cross sections at higher energies./22/

The parameter sensitivity of the capture cross section of ^{103}Rh is investigated in Figs. 3 and 4. Only the experimental data by LeRigoleur et al. and by Macklin are shown at lower energies. The calculated cross section labeled H results in a reasonable description of the data. This curve was calculated with optical model parameters obtained in a fit of the total cross section (unpublished data by Whalen/23/ below 700 keV and data given in BNL 325 at higher energies). The curve C was obtained with the global optical model parameters by Moldauer/24/ and the black-nucleus approximation was used to calculate neutron transmission coefficients to the continuum of the target nucleus. Curve A used the same optical model parameters as H, but also the black-nucleus approximation for the continuum. For curve B the width fluctuation correction was omitted. All other parameters were identical for the calculated cross sections shown in Fig. 3.

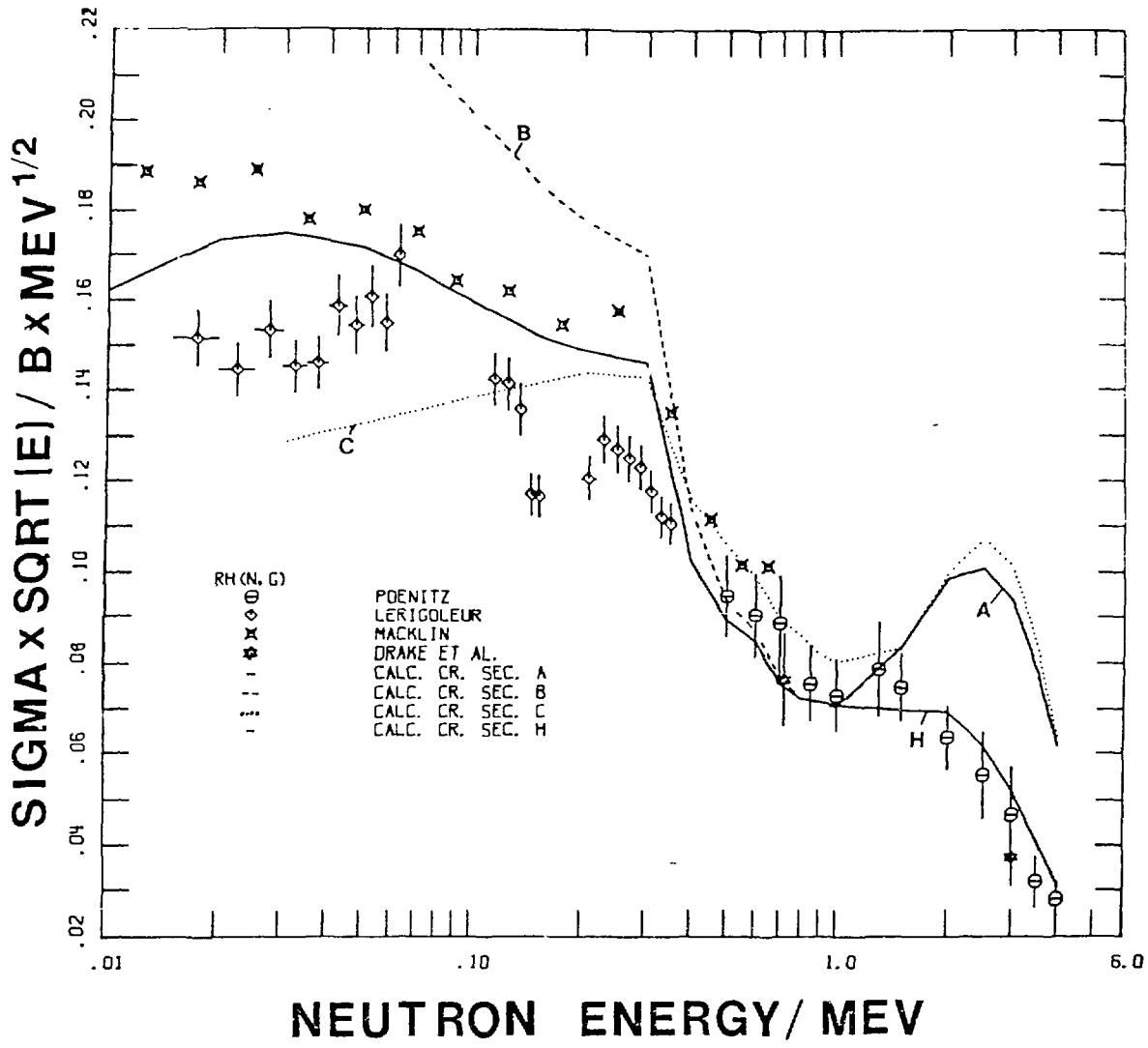

Fig. 3. Comparison of Nuclear Model Calculations with Experimental Data of Rh(n,γ). (See Text.)

All calculated cross sections shown in Fig. 4 used the optical model parameters obtained in the fit of the total cross section but the Γ_γ/D normalization was changed by 10% for curve I, the compound nucleus level density parameter a was lowered for curve D, and σ was increased for curve E. The target nuclear level density was increased for curve F. In all calculations for Fig. 4 the black-nucleus approximation was used for the transmission coefficients to the target nuclear continuum.

It is obvious that optical model calculations more realistically describe nuclear behavior and Figs. 3 and 4 demonstrate that the black nucleus approximation should not be used for the calculation of capture cross sections. With a well established normalization (Γ_γ/D) the cross section is insensitive to level density formula parameters at low energies and only sensitive to optical model parameters and the fluctuation correction. The range between ~500 keV and the beginning of the target nucleus continuum (~1 - 2 MeV) can be used to adjust the compound nucleus level density parameters. Agreement with experimental data at higher energies can then be achieved by adjusting target nucleus level density parameters.

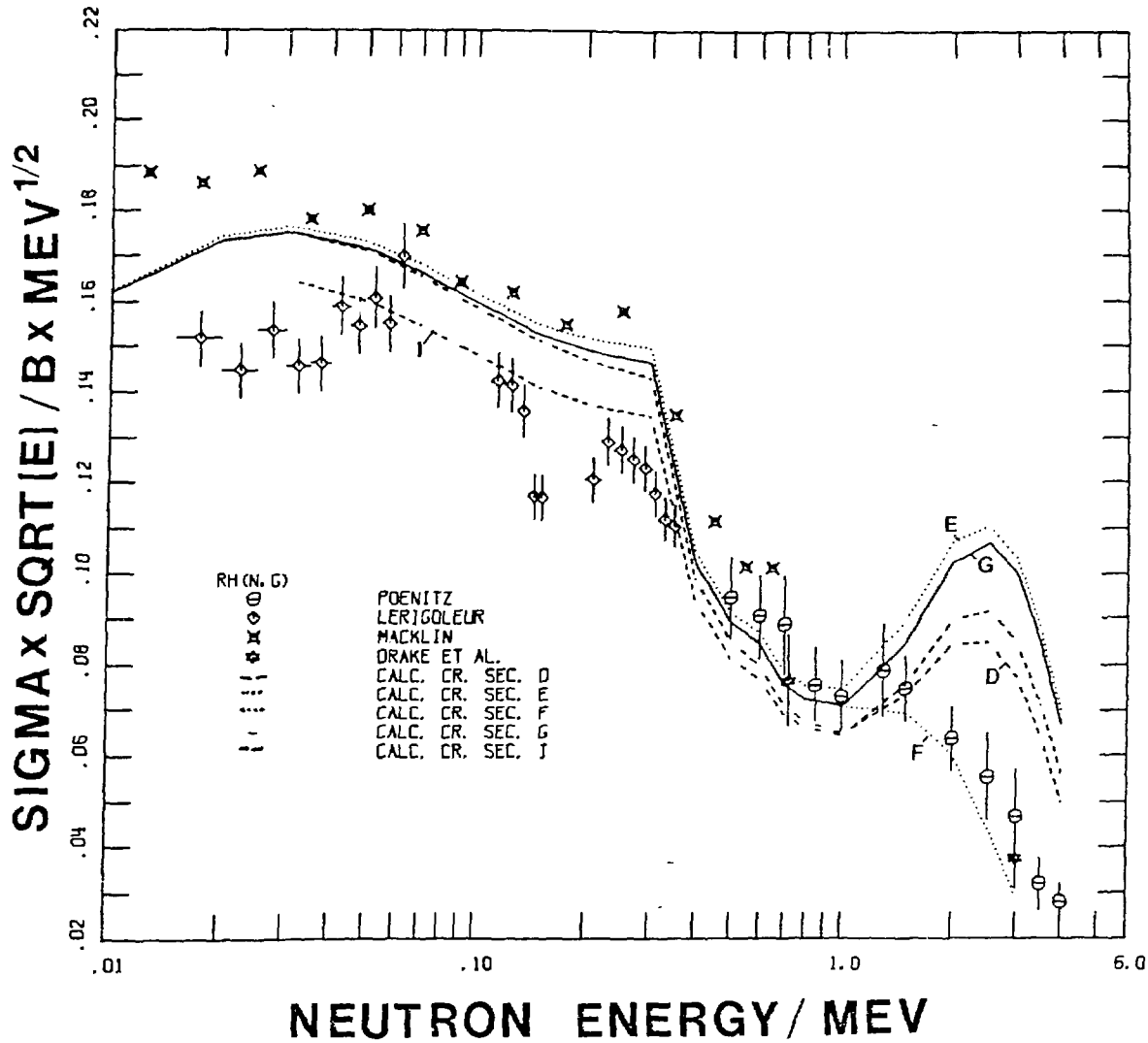

Fig. 4. Comparison of Nuclear Model Calculations with Experimental Data of Rh(n,γ). (See Text.)

Calculations were also carried out for the six stable isotopes of Pd. The optical model parameters obtained for Rh were slightly adjusted to give a good representation of the total cross section of Pd. The result for the elemental cross section is compared in Fig. 5 with the present experimental values and the recent data by Macklin.

CONCLUSIONS

Nuclear model calculations can represent the capture cross sections of fission product nuclei reasonably well, however, the uncertainties due to a large number of parameters is large if experimental data do not exist. This specifically applys to the higher energy range where different calculations resulted in large differences with a factor of 5 being common. The present measurements of the elemental cross sections above 500 keV provide a valuable constraint for such calculations, and should considerably reduce the uncertainties, specifically for bulk reactivity effects. Calculations of capture cross sections for which experimental data do not

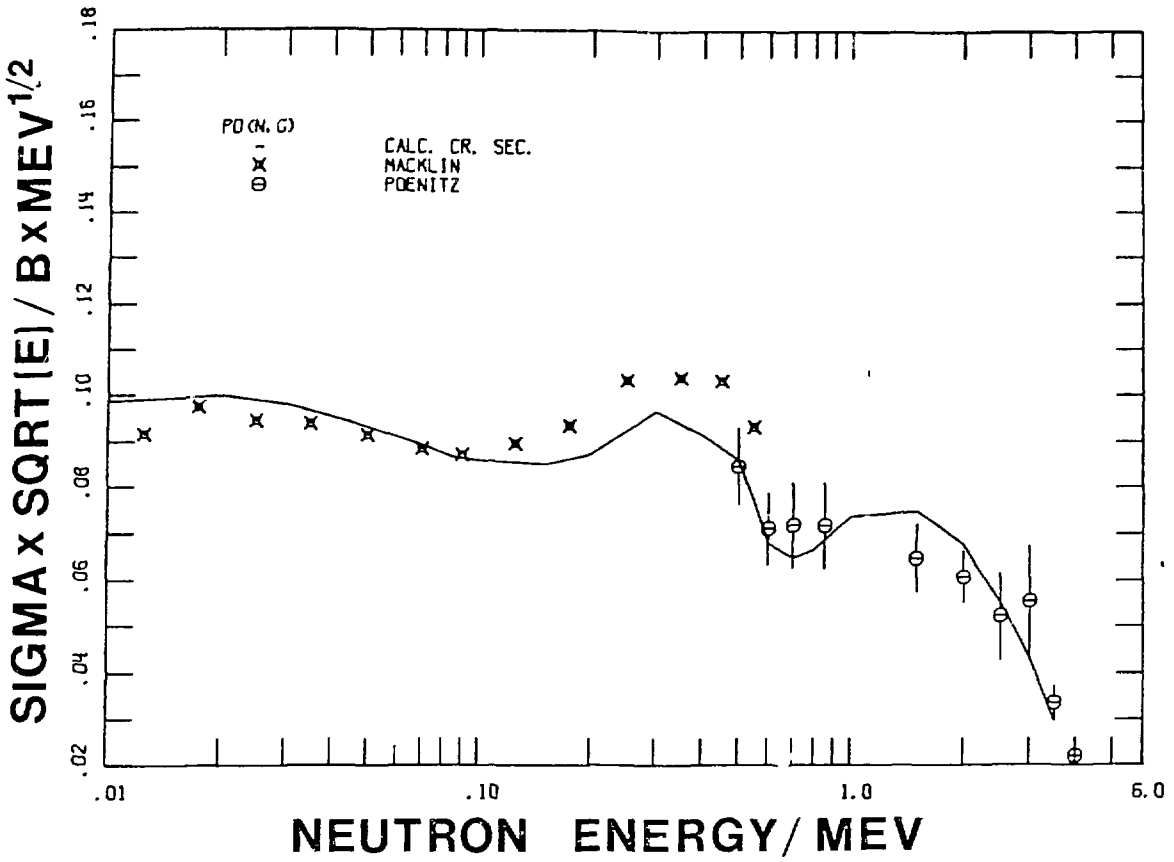

Fig. 5. Comparison of Nuclear Model Calculations with Experimental Data of Pd(n,γ).

exist cannot be expected to yield satisfactory values until optical model parameters in the range of the fission product nuclei are well established. This seems to require a better and more complete data base than presently available.

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