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## United Kingdom Atomic Energy Authority

REACTOR GROUP

### CHAPTER 4

NEUTRON CROSS-SECTIONS AND FISSION PARAMETERS OF U233, U235 AND PU239 AT 2200 M/SEC

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### CHAPTER 4

NEUTRON CROSS-SECTIONS AND FISSION PARAMETERS OF U<sup>233</sup>, U<sup>235</sup> AND Pu<sup>239</sup> AT 2200 m/sec

by

N-G SJÖSTRAND and

### J. S. STORY

### Abstract

This memorandum is to be published as one chapter (4) of a book entitled "Nuclear Data for Reactor Design, Vol. I" by N-G. Sjöstrand and J. S. Story, (pub. Pergammon Press) and is a critical assessment of experimental data relating to the 2200 m/sec. cross-sections of the three main fissile materials.

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### 4.1 INTRODUCTION

In this chapter we collect and discuss the experimental information on the reaction parameters

$$\sigma_{\rm S}$$
,  $\tau_{\rm A}$ ,  $\sigma_{\rm F}$ ,  $\alpha$ ,  $\nu$ , and  $\eta$ ,

for the three most important fissile nuclides under bombardment with neutrons of 2200 m/sec velocity. The notation is described in the next sub-section. We have given only very cursory discussions of the experimental techniques, but have attempted to outline some of the difficulties and uncertainties encountered, and have noted some of the special features of the individual experiments. More detailed accounts of the experimental methods have been given by others, by HARVEY & SANDERS (1956) for example, and in the references which are cited in the tables.

In sections 4.3 to 4.8 we collect and analyse the many experimental data. Weighted mean values for the various parameters are determined, from the most direct measurements of each. However the parameters are linked by the relationships

$$(1 + \alpha) \sigma_{\mathbf{F}} = \sigma_{\mathbf{A}} \tag{1+1}$$

$$(1 + \alpha) \eta = \overline{\nu} \tag{4.2}$$

and the measurements form an over-determined set. Consistent values of the various parameters have therefore been calculated by a least-squares analysis of the weighted means, and the results are given in the <u>table of recommended values</u> in section 4.9.

In weighting the various data we have tended to reject those derived from measurements made in the broad neutron spectra of reactor lattices. Knowledge of reactor spectra is still very limited and, in correcting measurements to 2200 m/sec it is all too easy to bias the estimates towards the preferred value. Moreover, to appraise the reliability of reactor theories it is desirable to have data available which are relatively independent of those theories

### 4.11 Notation

We use the notations of GOLDSTEIN (1957) and of WESTCOTT (1958, 1960). The definitions are given in chapter 1, but we list here some of those which are used repeatedly.

- E energy of incident neutron in a stationary frame of reference
- v its velocity
- T effective temperature of the thermal component of the neutron spectrum from a reactor or moderator
- T the moderator temperature
- d(E) energy distribution of the neutron flux
- o(E) cross-section per atom for neutrons of energy E
- $\sigma_A$  absorption cross-section,  $\sigma_A = \sigma_T \sigma_S$
- op fission cross-section

scattering cross-section S total cross-section Om radiative capture cross-section or v  $\overline{g}(E)$  average of  $\sigma_{\overline{g}}$  for neutron energies near E ô effective cross-section in the experimental spectrum  $\hat{\sigma} = \int \phi(E) \cdot \sigma(E) \cdot dE / \int \phi(E) \sqrt{(E0/E)} dE$ =  $(g + rs)\sigma^{0}$  in the notation of WESTCOTT (1958, 1960) = of /on the capture to fission probability ratio 12 = 0,/2, â the average number of neutrons evolved per fission v n(E) the average number of neutrons evolved per neutron absorbed in the fissile material, with incident neutrons of energy E =  $\int \phi(E) \cdot \eta(E) \cdot \sigma_{A}(E) dE / \int \phi(E) \cdot \sigma_{A}(E) \cdot dE$ ĥ = 2200 m/sec, standard velocity v = 0.025297 eV, the corresponding neutron energy E = E /k, k being Boltzmann's constant T  $= 297.59^{\circ} K = 20.44^{\circ} C$ 

 $\sigma^{\circ}, \alpha_{\circ}, \eta_{\circ}$ , values of  $\sigma, \alpha, \eta$  for monokinetic neutrons of velocity v

g, r, s, symbols used in WESTCOTT's (1958, 1960) formalism; see chapter 1.

Various other symbols are used, but they are defined in the particular sections where they occur.

### 4.2 CORRECTION OF EXPERIMENTAL DATA

It may be noticed that many of the data listed in this chapter differ slightly from the values given in the references. Usually these small changes have resulted from revision of the standards used in interpreting the measurements, or from correction of measurements in broad spectra to obtain values for monokinetic incident neutrons of 2200 m/sec. Usually we have listed the experimental results for the broad spectra also.

### 4.21 Revision of standards

Many of the data were obtained by comparative measurements and we have revised the reported results, using for reference cross-sections the values recommended in chapter 3, Table 3. Assay of thin samples of fissile materials has often been made by absolute alpha counting. We have revised the reported results using the half-lives and specific disintegration rates recommended in chapter 2, Table 2.

The standards we have used are collected, for convenience, in Table 4.1.

### TABLE 4.1

Element or nuclide	Parameter
Lithium Boron ANL standard Harwell standard Unspecified Sodium Manganese Gold	$\frac{\text{CROSS-SECTIONS AT 2200(m/sec})}{\sigma_{A} = 71.0 \pm 1.5 \text{ barns}}$ $\sigma_{A} = 757.7 \pm 3.0 \text{ barns}$ $\sigma_{A} = 767.2 \pm 3.5 \text{ barns}$ $\sigma_{A} = 757.7 \pm 7 \text{ barns, for measurements}$ in America, Canada, or Russia $\sigma_{act} = 0.534 \pm 0.007 \text{ barns}$ $\sigma_{act} = 13.17 \pm 0.1 \text{ barns}$ $\sigma_{act} = 98.4 \pm 0.52 \text{ barns}$
Uranium (natural) U <sup>233</sup> Pu <sup>239</sup> Pu <sup>235</sup>	SPECIFIC ALPHA ACTIVITIES: 1504.6 ± 2.5 alphas/min.mg 21050 ± 120 alphas/min.µg 136100 ± 190 alphas/min.µg ATOMIC ABUNDANCE IN NATURAL URANIUM: (0.7200 ± 0.00052)%

### Reference standards used in re-evaluations

### 4.22 Correction to 2200 m/sec

Many measurements have been made in broad neutron spectra, in reactors and thermal columns, or in neutron beams from these sources. For convenience and generality it is necessary to correct the results to an incident neutron velocity of 2200m/sec. If the measurements were made with thin foils or <u>dilute</u> samples the correction factors may be estimated with the help of Westcott's formulae and tables; WESTCOTT (1958, 1960). It is usually necessary to guess the neutron spectra involved, and in assessing the uncertainties it should be borne in mind that:

(i) Westcott's g and s coefficients may be in error;

 (ii) the effective temperature of the thermal component of the neutron spectrum may be uncertain; Sub-cadmium spectra. For many experiments cadmium-difference techniques were used. To make some allowance for the rumps of the epithermal neutron spectra we assumed these components to have 1/E distributions between 0.11 and 0.47 eV, and used the absorption integrals listed in Table 4.2.

### TABLE 4.2

	$\int_{0.11 \text{ eV}}^{0.47 \text{ eV}} \sigma_{A}(E) \frac{dE}{E}$	$\int_{0.11 \text{ eV}}^{0.47 \text{ eV}} \sigma_{\mathrm{F}}(\mathrm{E}) \frac{\mathrm{dE}}{\mathrm{E}}$
1/v absorber U <sup>238</sup> U <sup>235</sup> 239 Pu	0.4950 <sup>0</sup> 284 barns 290 " 2250 "	255 barns 240 " 1320 "

Sub-cadmium epithermal absorption integrals

This treatment is obviously extremely crude, but it is probably good enough for our purpose. An alternative approach is to use the formula

$$\frac{\partial_{sub-Cd}}{\sigma^0} = \frac{g + r(s-t\pi/\sigma^0)}{1-0.4742rt} \text{ with } t = \sqrt{\frac{\pi T_0}{4T}} \sqrt{\frac{4T}{11}}$$

in which, for consistency, all quantities used on the right hand side are to be those tabulated by WESTCOTT (1960), and T, To are in the absolute temperature scale.  $\Sigma$  is the epi-cedmium resonance integral.

#### 4.3 SCATTERING CROSS-SECTIONS

### 4.31 Introduction

The absorption cross-sections of the fissile nuclei are usually determined from the total cross-sections by subtracting the scattering cross-sections. The total cross-sections have been measured with neutron spectrometers by the transmission method, and are about 600 to 1000 barns in the thermal region for  $U^{239}$ ,  $U^{239}$  and  $Pu^{239}$ . To determine their absorption to about  $\pm 1$  percent it is necessary to know the scattering cross-sections to within about  $\pm 5$  barns.

The following phenomena affect the low energy neutron scattering in the harvy isotopes:

(1) Potential scattering, for which the cross-section is about 11 barns.

(2) Resonance scattering, from neighbouring resonances at positive and negative resonance energies.

- (3) Interference between resonance and potential scattering.
- (4) Interference between neighbouring resonances of like spin.
- (5) Coherence effects, which depend on the chemical environment and configuration,
- and are not usually very noticeable above about 0.1 eV.
- (6) Thermal motion of the scattering atoms.

The data on the scattering cross-sections of  $U^{233}$ ,  $U^{238}$  and  $Pu^{239}$  are meagre and it is prudent to compare these data with those for other heavy nuclides, specifically Th<sup>232</sup> and  $U^{238}$ .

### 4.32 Potential scattering

In most applications the potential scattering is taken to include the scattering effects of the more distant resonances. Small variations in  $\sigma_{pot}$  result, over a broad energy scale, if there is a group of strong resonances of the same parity and total angular momentum. Each of the three nuclides U<sup>233</sup>, U<sup>235</sup> and Pu<sup>239</sup> has non-zero spin, so that two states of the total angular momentum are excited by interaction with slow neutrons, and somewhat different values of the potential scattering may be associated with each state. These comments suffice to explain that  $\sigma_{pot}$  is neither uniquely defined nor uniquely determined.

The potential scattering is not expected to vary much between neighbouring heavy elements. SETH et al. (1958) calculated that of Th<sup>232</sup> and U<sup>238</sup> from their total cross-sections in the energy range 1 to 80 eV, and also from transmission measurements in the keV region. The results were:

 $\sigma_{\text{pot}}$  [Th<sup>232</sup>] = 12.0 ± 0.3 barns  $\sigma_{\text{pot}}$  [U<sup>238</sup>] = 10.7 ± 0.3 barns

A somewhat lower value for U238

 $\sigma_{\rm pot} = 9.2 \pm 0.3$  barns

may be estimated from the total cross-section data in the region 0.2 to 5 eV, which may be found in BNL325. This reduction is probably caused by destructive interference with the strong resonances at 21, 37, 66 and 103 eV.

Multi-level analyses of the low energy cross-sections of  $U^{233}$  and  $U^{235}$  are cited in the next section and are consistent with the free-atom scattering data listed there. If the potential scattering is adjusted to give the best consistency below 1 eV one finds:

> σ<sub>pot</sub> [U<sup>233</sup>] = 13.1 barns, for VOGT's (1960) analysis σ<sub>pot</sub> [U<sup>235</sup>] = 9.5 barns, SHORE & SAILOR (1958) = 11.3 barns, for VOGT's (1958, 1960) analyses.

The U<sup>233</sup> analysis was somewhat incomplete. Negative energy resonances were not taken into account explicitly and this is probably the reason for the anomalously high value needed for the potential scattering at low energies. BOLLINGER et al. (1958) estimated the potential scattering of Pu<sup>239</sup> from

 $\sigma_{\rm S} = \sigma_{\rm T} - (1 + \alpha)\sigma_{\rm F}$  in the range 29 to 38 eV, where the resonances are particularly weak so that between resonances  $\sigma_{\rm S} \approx \sigma_{\rm pot}$ . Although  $(1 + \alpha)$  is not known very reliably  $\sigma_{\rm F}$  is very small between the resonances, so it was possible to estimate  $\sigma_{\rm S}$  with fair accuracy. The data indicated that

$$\sigma_{\rm pot} [Pu^{239}] = 10.5 \pm 0.5 \text{ barns}$$

The data cited above illustrate that the potential scattering is not uniquely defined and that it varies slowly with neutron energy. As a typical value for the heavy nuclides one might assume

$$\sigma_{\rm not} = 10.9 \pm 1.3 \, \rm barns$$

with the corresponding "nucleon radius"

$$r = (1.50 + 0.09) \times 10^{-13} \text{ cm}$$

defined by

$$\sigma_{\text{pot}} = 4\pi r^2 (\Lambda + 1)^{2/3}$$

where A is the mass number of the target nucleus

### 4.33 Free-atom scattering

The scattering cross-sections of  $U^{233}$  and  $U^{235}$  have been measured at epithermal energies by OLEKSA (1958) and by FOOTE (1958). Their results are given in Table 4.3. Earlier measurements of  $\sigma_{\rm S}$  [ $U^{235}$ ], by MELKONIAN, were reported by SAILOR (1956) at Geneva, but appear to have been superseded by the work of FOOTE.

#### TABLE 4.3

υ <sup>233</sup> ,	OLEKSA (1958)	U <sup>235</sup> , FOOTE (1958)			
E ev	o <sub>S</sub> barns	E eV o <sub>S</sub> barns			
0.27	13.4 + (105)	0.271 14.7			
.35	12.3 "	0.756 14.1 + 0.4			
.45	13.0 "	1.044 13.4			
.55	12.2 "	1.81 12.4			
.65	12.1 "	2.51 12.9			
. 74	12.8 "	4.31 11.5			
.85	11.9 "	7.74 11.0 + 0.7			
.95	12.5 "	Uncertainties are for counting			
2.09	11.6 + (15%)	statistics only. Gross uncer-			
2.71	14.2 "	tainties probably as for U233			
3.31	12.6 "	•			

Epithermal scattering cross-sections of U233 and U235

MOORE & REICH (1960) and VOGT (1958, 1960) have fitted multi-level resonance formulae to the low energy total and fission cross-sections of  $U^{233}$  and  $U^{235}$ . Their analyses are consistent with the observed epithermal scattering data and may be used for prediction of the "free-atom" scattering cross-sections in the thermal region. First however, notice that the calculated scattering can be brought into much closer agreement with the measurements by small adjustments of the assumed potential scattering. The "free-atom" scattering at thermal energies is then predicted to be:

> σ<sub>fa</sub> [U<sup>233</sup>] = 13.3 barns, from MOORE & REICH (1957) 12.8 barns, from VOGT (1960) σ<sub>fo</sub> [U<sup>235</sup>] = 16.3 barns, from SHORE & SAILOR (1958) using the

> of [0""] = 16.3 barns, from SHORE & SALLOR (1958) using the formulae of MOORE & REICH

> > 16.0 barns, from VOGT (1958, 1960)

It will be noticed that  $\sigma_{ra} [U^{235}]$  increases at low energies. This is caused mainly by a very strong negative-energy resonance. However the uncertainty of the extrapolation cannot well be assumed less than  $\pm 1$  barn.

No direct measurements of the scattering by Pu<sup>239</sup> have been reported. VOGT (1960) made . multi-level analysis of the low energy total and fission cross-sections, but this analysis is very far from being unique. The main features in the thermal region arise from the well-known resonance at 0.3 eV and another in the range -1.2 to +0.02 eV approximately. Probably these two levels are of different spins and do not interfere. Allowing for the effects of these two resonances on the potential scattering the "free-atom" scattering in the thermal region may be estimated:

$$\sigma_{P_0} [Pu^{239}] = 12.7 \pm 1.7$$
 barns

Contributions of the more distant resonances at positive and negative energies must cancel to some extent and cannot be calculated from the data available.

### 4.34 Molecular binding, and Doppler effect

The effects of chemical binding in an inchoate configuration, and of the thermal motion of the atoms, can be evaluated by the formulae of MARSHALL & STUART (1959); see particularly equation (70) of their paper. The effects are small for thermal neutron scattering by heavy atoms at normal temperatures, so that the scattering differs negligibly from the "free-atom" cross-section discussed in the previous subsection.

### 4.35 Coherence

No measurement has been reported of the low energy neutron scattering,  $E \leq 0.2$ eV, from U<sup>233</sup>, U<sup>235</sup>, or Pu<sup>239</sup>. The scattering from pure metallic U<sup>238</sup> must be wholly coherent, because the nuclide has zero spin. From powder diffraction studies at 0.074 eV SHULL & WOLLAN (1951) estimated the coherent scattering cross-section of U<sup>238</sup> to be 9.0 ± 0.5 barns. This is indistinguishable from the "free-atom" scattering cross-section of 8.8 ± 0.3 barns estimated at the same energy from  $\sigma_{\text{pot}}$  [U<sup>238</sup>] = 9.2 barns, see sub-section 4.32, and the parameters of the 6.7 eV resonance. In contrast, U<sup>233</sup>, U<sup>236</sup>, and Pu<sup>239</sup> have non-zero spins, which may give rise to some "spin incoherence". However, the elementary theories available suggest that the incoherence should be small, probably 0.1 barns. Evidence that the scattering from  $U^{233}$ , and  $U^{235}$ , is largely coherent may be gleaned from the measurements at long wavelengths by SAFFORD et al. (1959 and 1960).

The coherent elastic scattering from polycrystalline samples exhibits sharp discontinuities at low neutron energies. However, in analysing the slow neutron transmission data for  $U^{233}$ ,  $U^{235}$ , or  $Pu^{239}$ , it is quite customary to treat  $\sigma_T(E)$  as a smooth function of the neutron energy. To determine  $\sigma_A(E)$  from such an analysis we must subtract a smoothed scattering cross-section. This locally averaged  $\bar{\sigma}_S(E)$  is the main objective of the discussion that follows.

With an ideal polycrystalline sample  $\tilde{\sigma}_{S}(E)$  would probably be sufficiently well represented at thermal energies by the "incoherent approximation", PLACZEK et al. (1951), sketched in the preceding sub-section. In practice the coherent scattering may vary considerably from one sample to another in consequence of extinction effects, or because the crystallites are not randomly oriented. Extinction results from large grain size with small mosaic spread and has the effect of reducing the effective coherent cross-section; WIWTAKER & BEYER (1939), WEISS (1952). Preferential orientation of grains is common in rolled or extruded metal samples. The angular distribution of the coherent scattering from a single crystallite is highly anisotropic, so the integral cross-section can be strongly affected by preferred orientation of the grains. In transmission measurements with rolled gold foils CARTER et al. (1953) found that the coherent scattering at thermal energies was reduced as much as 50 percent by extinction and orientation effects. They also measured the transmission of a powdered sample and it may be inferred that the coherent scattering was about 25 percent low in the thermal region; HARVEY (1956) attributed this to extinction effects.

The total cross-section of  $U^{238}$  is given in BNL325 as a function of the neutron energy, from transmission measurements with thick metal samples. By subtracting the absorption cross-section it can be shown that  $\bar{\sigma}_{\rm S}$   $[U^{238}]$  is about  $(10 \pm 6)$  percent below the "free-atom" scattering, for neutrons of around 0.025 eV. This reduction may be due to extinction or to grain orientation. The chemical binding would be little affected if these samples could be transmuted into  $U^{238}$  (or  $U^{234}$ , or  $U^{235}$ , or  $U^{238}$ ), so we should expect  $\sigma_{\rm S}/\sigma_{\rm Fa}$  to remain unchanged. In practice, of course, much thinner samples are used for slow neutron transmission measurements with  $U^{233}$  or  $U^{235}$ , so the extinction and orientation effects may be different. Considering the magnitude of the effects observed with gold samples it is reasonable to assume generally

$$\overline{\sigma}_{s} = (0.90 \pm 0.23) \sigma_{r_{0}} \text{ at } \sim 0.025 \text{ eV},$$

for metal samples of U233. or U235. The same assumption may be applied to Pu239 also, with a small increase in the uncertainty.

### 4.36 Recommended values for thermal neutron scattering

From the considerations of the preceding sub-sections we have estimated the values listed in Table 4.4 for the scattering cross-sections of U<sup>233</sup>, U<sup>235</sup> U<sup>238</sup>, and Pu<sup>239</sup> at about 0.025 eV.

### TABLE 4.4

Nuclide	Nature of sample	σ <sub>S</sub> , barns
	Free atoms, and liquid compounds	13•0 <u>+</u> 1•4
U <sup>233</sup>	Polycrystalline powders	12.4 ± 1.4
	Sintered " "	12.4 + 2.0
	Metal (rolled, or extruded)	11.7 <u>+</u> 3.2
	Free atoms, and liquid compounds	16.0 <u>+</u> 2.3
U <sup>235</sup>	Polycrystalline powders	15.2 ± 2.3
	Sintered " "	15•2 <u>+</u> 2•9
	Metal (rolled, or extruded)	14•4 <u>+</u> 4•2
	Free atoms, and liquid compounds	8.8 ± 0.3
U 298	Polycrystalline powders	8.4 <u>+</u> 0.5
	Sintered " "	8.4 <u>+</u> 1.2
	Metal (rolled, or extruded)	8.1 ± 2.1
	Free atoms, and liquid compounds	12•7 ± 1•7
Pu <sup>239</sup>	Polycrystalline powders	12.1 ± 1.7
	Sintered " "	12.1 ± 2.3
	Metal (rolled, or extruded)	11.4 ± 3.7

Recommended scattering cross-sections for U<sup>233</sup>, U<sup>235</sup>, U<sup>238</sup> and Pu<sup>239</sup>, with neutrons of about 0.025 eV

The samples are assumed to be at normal temperatures, around 20°C. Even with mixtures of uranium isotopes, or U-Pu mixtures the incoherent scattering is likely to be rather small, and the tabulated cross-sections should remain valid.

### 4.4 ABSORPTION CROSS-SECTIONS

### 4.41 Methods of measurement

The absorption of slow neutrons by U<sup>233</sup>, U<sup>235</sup>, or Pu<sup>239</sup> gives rise to radiative capture or to fission. As there are two distinct reaction processes the gross absorption cross-section cannot be directly measured by observing the product nuclei or the reaction products. It can be measured only by observing the destruction of the absorbing nuclei by a sufficiently large neutron irradiation, or by observing the attenuation of the neutrons by a sufficiently large mass of the absorbing material.

Direct measurements in broad neutron spectra. A few direct measurements of  $\sigma_A$  have been made by comparative methods in broad neutron spectra, and they are listed in Table 4.5. KUKAVADSE et al. (1956) used alpha counting and mass spectrometry to compare the destruction of U<sup>233</sup> and of Li<sup>6</sup> caused by intense irradiation of small samples in a reactor. SPIVAK & YEROZOLIMSKY (1956) made transmission measurements in wide beam geometry, with neutrons from a shallow hole in a graphite thermal column. Special devices were used to annul the effects of neutron scattering. The samples were relatively thin, the thicknesses being chosen so that each had approximately the same transmission. All the samples were assumed to be 1/v absorbers, and boron was used as a standard to eliminate the uncertainties and mathematical difficulties associated with the complex geometry, the spectrum and angular distribution of the incident neutrons, the transmission hardening, and the variations of detector efficiency with neutron energy and direction. GREEN et al. (1957) made comparative measurements of  $\sigma_A$  [U<sup>233</sup>] using a local pile oscillator in the reflector of BEPO. The fission neutrons evolved have a relatively small effect, as they migrate away from the detector during moderation, and this effect was measured with the help of a calibrated neutron source.

Transmission measurements of the total cross-sections. The absorption cross-sections of  $U^{233}$ ,  $U^{235}$ , and  $Pu^{239}$ , have most often been determined from the total cross-sections by subtraction of the scattering. The total cross-sections have been measured by the transmission method in good geometry. The results are listed in Tables 4.6, 4.7 and 4.8.

Several of the early transmission measurements were made using neutron beams from thermal columns; FERMI et al. (1944), ANDERSON & MAY (1944), ZINN & KANNER (1945). Similar measurements with calibrated pyrex and gold plates, were used to determine the effective temperature of the incident neutron spectrum. The calibrations were made with monokinetic neutrons from neutron spectrometers.

The most reliable measurements of  $\sigma_{\rm T}$  are those made with monokinetic neutrons by using one or another of the various forms of velocity selector. The excellent consistency of recent American measurements may be seen from the data listed in Tables 4.6 and 4.7. Different spectrometers were used, and samples of different origins. For typical accounts of the problems and techniques of this method the reader is referred to the papers of MELKONIAN (1949 and 1951), LEONARD et al. (1953), SAFFORD et al. (1959), and BLOCK et al. (1960).

To evaluate  $\sigma_T(2200 \text{ m/sec})$  as accurately as possible it is a common practice to fit a simple smooth function of the neutron energy to the transmission data at neighbouring energies. In this way the counting statistics are improved, but at the same time small discontinuities in the coherent scattering in the same energy range are smoothed away. The scattering cross-sections are discussed rather fully in the previous section, and preferred values of the smoothed cross-sections,  $\bar{\sigma}_S$ , for neutrons of about 0.025 eV are given in Table 4.4.

### 4.42 Correction of broad-spectrum data to 2200 m/sec Hardening factors. Self-screening factors

A transmission measurement in good geometry with a broad neutron spectrum

determined the transmission

 $t = \frac{\left[\phi(E), \varepsilon(E), \exp\left[-x, \sigma_{T}(E)\right]\right]dE}{\left[\phi(E), \varepsilon(E), dE\right]}$ (4.3)

where  $\phi(E)$  is the distribution of the incident neutron flux,

 $\varepsilon(E)$  is the detector efficiency for neutrons of energy E,

x is the thickness of the absorbing sample in atoms/barn.

If the energy dependence of  $\phi(\mathbf{E})_{\cdot} \epsilon(\mathbf{E})$  is known, together with that of the total crosssection  $\sigma_{\mathbf{T}}(\mathbf{E})/\sigma_{\mathbf{T}}^{\mathbf{O}}$ , one can compute  $h_{\mathbf{X}}\sigma_{\mathbf{T}}^{\mathbf{O}}$  as a function of  $\sigma_{\mathbf{T}}^{\mathbf{O}}$  from difinition

$$exp(-hr\sigma_{0}^{\circ}) = right-hand side of equation (4.3). (4.4)$$

Then  $\sigma_m^{o}$  can be calculated from the measured transmission, using

$$hr\sigma_{\rm T}^{0} = \ln^{1}/t_{\bullet}$$
 (4.5)

A transmission measurement necessarily demands a sample which is not thin so that in general the "hardening factor" h cannot be expressed in terms of the coefficient g + rs) of WESTCOTT's (1958, 1960) formalism. Hardening factors for 1/v absorbers in Maxwellian spectra have been evaluated by ZAHN (1937), LAPORTE (1937) and WESTCOTT (1959), and can be used whenever  $\sigma_T(E)$  can be written in the form a + b/v, with a and b constant.

If the sample is fairly thin the exponent in equation (4.3) can be expanded giving

$$1-t \approx \frac{S(x)_{\bullet} x_{f\phi}(E)_{\bullet \mathcal{E}}(E)_{\bullet} \sigma_{m}(E) dE}{f\phi(E)_{\bullet \mathcal{E}}(E)_{\bullet} dE}, \qquad (4.6)$$

where

$$S(x) = 1 - \frac{1}{2} x \bar{\sigma}_{m} + \cdots ,$$
 (4.7)

with a suitably chosen value of  $\sigma_{T}$ , may be called the "self-shielding factor". In comparative measurements the self-shielding factors will cancel, very nearly, if the samples are thin and about equally transmissive.

If also  $\varepsilon(E)$  is constant and if the variations of  $\sigma_{S}(E)$  are very small compared with  $\sigma_{A}(E)$ 

$$1-t \approx S(x) \cdot x \{ \overline{\sigma}_{S} + \sigma_{A}^{\circ} \cdot (g + rs) \sqrt{(\pi T_{o}/4T)} \}$$
(4.8)

### 4.43 The experimental data

Direct measurements of  $\sigma_A$  for U<sup>233</sup>, U<sup>235</sup> and Pu<sup>239</sup>, from comparative experiments in broad spectra are listed in Table  $l_{+}.5$ . We give no weight to any of these data, except to that of GREEN et al. (1957) which, for convenience, is listed again in Table 4.6. Total cross-sections from transmission measurements are given in Tables 4.6, 4.7 and 4.8, together with the values of  $\sigma_A$  derived by subtraction of the scattering. We used the scattering cross-sections given in Table 4.4. From the discussion in subsection 4.35 it will be appreciated that the coherent scattering can vary from one sample to another, and that a large part of the uncertainty in  $\sigma_S$  stems from this variation. In consequence  $\sigma_T$  also may be expected to vary slightly from one sample to another. However in calculating the uncertainty in-the-mean of  $\sigma_A$  it is necessary to bear in mind the small systematic element in the uncertainty of  $\sigma_S$ .

The experimental methods have been outlined in sub-section 4.41; the following notes discuss some of the corrections needed:

- A. KUKAVADSE et al. (1956) used samples about 0.1, mean-free-path thick, so there was appreciable self-shielding. Corrections were made assuming that the incident neutrons were isotropic with a Maxwellian spectrum; 1/v absorption was assumed. To secure a sufficiently large burn-up the irradiation was made in the reactor core. It seems very unlikely that the epithermal neutron flux was negligible although the sample was irradiated "in a water filled channel". We have roughly corrected the result to 2200 m/sec by guessing the reactor spectrum and using WESTCOTT's (1960) tables. However we prefer to give no weight to the result because of the uncertainties in the neutron spectrum and the resonance self-shielding.
- B. SPIVAK & YEROZOLIMSKY (1956) used samples only about 0.04 mean-free-path thick, and almost equally absorptive. All samples were treated as 1/v absorbers. It is not practicable to correct the results for deviations from the 1/v law, because the detector efficiency varied with neutron energy and the geometry was very complex. The angular distribution and spectrum of the incident neutrons may have been distorted by the proximity of the moderator surface and of a cadmium disc in the beam well. For these reasons we give no weight to the results.
- C. GREEN et al. (1957) used small dilute samples in D<sub>2</sub>O solutions. They were about equally absorptive, so that the self-shielding effects should cancel, approximately. The local oscillator has a 1/v neutron detector; the epithermal component of the neutron spectrum was very weak with  $r \sim 0.0001$ ; none of the sample nuclides has a resonance in the thermal region. Under these conditions the measurements give relative values of  $\sigma_A(kT)$ , where T is the neutron temperature and k is Boltzmann's constant. The moderator temperature was probably about 20  $\pm 10^{\circ}$ C, but no estimate is available of the distortion of the spectrum by the material of the oscillator and sample. Fortunately each of the materials compared has nearly 1/v absorption, so that the results are insensitive to the uncertainties in the neutron spectrum.
- D. FERMI, ANDERSON, ZINN, and colleagues made several transmission measurements in good geometry, using neutrons from thermal columns. The effective neutron temperature was calculated from the transmission through a pyrex plate, the plate having been first calibrated with monokinetic neutrons from a slow-neutron chopper. ZINN & KANNER (1945) also used a gold plate which had been calibrated with the chopper and with a crystal spectrometer. The observations were interpreted by treating all samples as 1/v absorbers with constant scattering cross-sections. We have revised the results for U<sup>233</sup>, assuming that v.oT(E) is constant, but it has not seemed worth while to calculate revised hardening corrections for U<sup>235</sup> and Pu<sup>239</sup>. The method of measurement is very indirect and we give no weight to the results.

### TABLE 4.5

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Absorption cross-sections of [	1 333 U 335	and Pu 1	or neutrons	of 2200 m	sec.	from comparative	measurements i	n broad	neutron spectra	
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Reference	U233 barns	U235 barns	Pu239 barns	Weight	Method and comments
KUKAVADSE et al. (1956)	595 ± 38			o	Destruction of U <sup>233</sup> and Li <sup>6</sup> through reactor irradiation compared, by mass- spectrometry and alpha counting, giving $\sigma_{A}$ [U <sup>233</sup> ]/ $\sigma_{A}$ [Li <sup>6</sup> ] = 0.66 ± 0.03. We assume $\sigma_{A}^{\circ}$ = 950 ± 20 barns for Li <sup>6</sup> and have corrected for non-1/v absorption by U <sup>233</sup> . See § 4.43, Note A
SPIVAK & YEROZOLIMSKY (1956)	584 <u>+</u> 13	652 <u>+</u> 14	1001 ± 22	o	Transmission measurements in wide-beam geometry with neutrons from a thermal column. Scattering effects annulled. Thin samples used, with boron as standard. We assume $\sigma_A^{\circ} = 758 \pm 7$ barns for boron. See § 4.43, Note B
BURGOV (1956)	594 <u>+</u> 8	692	1149	0	Comparisons in thermal spectra gave the ratios to $\sigma_A$ [B] of 0.784 ± 0.008, 0.914 and 1.517, respectively. We do not know the method of measurement and so cannot correct to 2200 m/sec
GREEN et al. (1957)	571 <u>+</u> 15			See Table 4.6	Comparisons with boron and gold using a local oscillator in EEPO gave $\sigma_A [U^{0.53}]/\sigma_A$ [B]= 0.738 ± (3%), and $\sigma_A$ [B]/ $\sigma_A$ [Au] = 7.94 ± 0.09. We assume alternatively that $\sigma_A^{-0}$ = 767.2 ± 3.5 barns for Harwell standard boron, or 98.4 ± 0.5 barns for gold. The uncertainty given in the reference appears to have been over-estimated. See also § 4.43, Note C

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Reference	barns	barns	Weight	Method and comments			
ANDERSON & MAY (1944)	562 ± 9	550 ± 9	C	Transmission of thermal neutron beam through $U_3O_8$ samples, relative to that of a pyrex plate which had been calibrated with a slow-neutron chopper. Revised using isotopic analysis by RALL & DELPSTER (1946) and assuming $v \ge \sigma_T(E)$ is constant. See § 4.43, Note D.			
ZINN & KANNER (1945)	576 ± 15	564 ± 15	o	Method as above. Original data corrected assuming that v x $\sigma_{\rm T}(E)$ is constant in the thermal region. See also § 4.43, Note D.			
PALEVSKY & MUETHER (1954)	597 <u>+</u> 10	585 ± 11	0.2	BNL slow-neutron chopper, with highly enriched metal samples,			
NIKITIN et al. (1956)	580 <u>±</u> 20	568 <u>+</u> 20	0.05	Time-of-flight, using a pulsed cyclotron. Tabular data are listed by EVELSTAFF (1957); averaging v x $\sigma_{\rm T}$ near 0.025 eV gives a result 5 barns lower.			
FATTENDEN (1956b)	600 <u>+</u> 15	588 ± 15	0	Harwell slow-neutron chopper and crystal spectrometer, with highly enriched samples of $UO_2$ and of uranium metal. See § 4.43, Note E.			
GREEN et al. (1957)		571 ± 15	0.09	From Table 4.5. A pile oscillator measurements.			
SAFFORD et al. (1960)	586 ± 2 587 ± 5	574,± 3.8 574 ± 5.2	1.0 0.6	Columbia University crystal spectrometer, using metal and liquid samples respectively. See § 4.43, Note F.			
SIMPSON et al. (1960)	587 ± 6	575 ± 6.8	0.4	MTR fast chopper, with rolled metal sample prepared at Los Alamos.			
BLOCK et al. (1960)	587 ± 3	575 ± 4.4	c.8	ORNL fast chopper, with rolled metal samples. See § 4.43, Note F.			
Weighted mean value		575 ± 3.1	The un	certainty quoted allows for a systematic component associated with $\sigma_S$			

Total cross-section of U<sup>333</sup> for neutrons of 2200 m/sec, from transmission measurements, and derived values of the absorption cross-section

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# Total cross-section of U<sup>235</sup> for neutrons of 2200 m/sec, from transmission measurements, and derived values of the absorption cross-section

TABLE 4.7

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Reference	or T barns	σ A barns	Weight	Method and comments
ANDERSON et al. (1944)	662 ± 16	648 ± 17	0.02	Time-of-flight, using a pulsed cyclotron.
FERMI et al. (1944)	648 ± 13	633 ± 14	0	Transmission with thermal neutron beam; relative to that of a pyrex plate which had been calibrated with a slow-neutron chopper. See §4.43, Note D. Revises data of FERMI et al. (1943)
ZINN & KANNER (1945)	650 ± 15	635 <u>+</u> 16	0	Method as above. See also § 4.43, Note D
Columbia University unpublished work (1948) cited by MELKONIAN et al. (1953)	670.5	656 ± 20	0.03	Time-of-flight, using a pulsed cyclotron and a highly enriched metal sample. Result possibly too low, by $\lesssim 3\%$ , because of an unexplained variation of detector efficiency with counting rate
MEIKONIAN et al. (1953)	693 ± 5	679 ± 6.5	0.2	Time-of-flight, using a pulsed cyclotron and rolled metal samples $\sim 20\%$ enrichment. $\sigma_T$ calculated from smooth curve fitted to the data near 2200 m/sec
EGELSTAFF (1954)	729 <u>+</u> 15	715 ± 15	0.06	Harwell slow-neutron chopper, with metal samples of 81. 7% enrichment. The original value, 739 barns, was later revised for an error in the velocity calibration; see LYNN & PATTENDEN (1956), EGELSTAFF (1956)
LEONARD (1954)	701 •4 ± 4	687 ± 6	0.3	Hanford crystal spectrometer. This is the result of a single obser- vation at 0.0253 eV. A value 6 barns higher is obtained from a smooth curve fitted to the data in the range 0.02 to 0.03 eV
PALEVSKY et al. (1954)	700 ± 5	686 ± 6.5	0.3	BNL slow-neutron chopper, with highly enriched metal samples
NIXITIN et al. (1956)	710 ± 20	696 <u>+</u> 20	0.03	Tunic-of-flight, using a pulsed cyclotron. Result calculated from data in the range 0.02 to 0.03 eV
MOSTOVOI et al. (1957)	683	669	0.3	RFT slow-neutron chopper, from smooth curve fitting the data in the range 0.02 to 0.03 eV
BOLLINGER (1957)	694 ± 2	680 ± 4.7	0.4	ANL fast chopper. No details available
SAFFORD et al. (1959)	698.7 ± 4.8 695.3 ± 1.8	684.3 ± 6.4 679.3 ± 2.9	0.3	Columbia University crystal spectrometer, using metal and liquid samples respectively
SIMPSON et al. (1960)	690 ± 10	676 ± 11	0.1	MTR fast chopper, with metal foils
BLOCK et al. (1960)	693 ± 5	679 ± 6.5	0.3	ORNL fast chopper, with rolled metal samples
Weighted mean values 680.6 ± 3.6			The ur	certainty quoted allows for a systematic component associated with og

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TAB	LE .	4-8
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TA om, Weight Mathod and comments Reference barns barns Details not available. Method probably as described in § 4.4.3 Note D. Result cited by HARVEY & HURHES (1953) may be some amendment of the ANDERSON et al. (1944) 2 original value ANDERSON et al. (1945) 1056 + 25 1045 + 25 0.1 Time-of-flight, using a pulsed cyclotron. No details available Time-of-flight, using a pulsed cyclotron. Value given here is from an 1059 + 20 1047 + 20 analytic formula fitted to the date. SCHWARTZ et al. (1957) cited a HAVENS et al. (1951) 0.2 value about 7 barns higher ZIMMERMAN & PALEVSKY 1025 + 10 1014 + 11 1.0 BNL slow-neutron chopper. No details available (1955)ABOV (1955) 1050 + 13 1039 + 14 0.3 Bent crystal spectrometer NIKITIN et al. (1956) 1040 + 30 1029 + 30 Time-of-flight, using a pulsed cyclotron 0.2 Hanford crystal spectromater, using same sample as HAVENS et al. (1951). 1048 + 13 1037 + 14 Value given here is from smooth curve fitting the data near 0.025 eV. LEONARD et al. (1956) 0.4 Measured value at 2200 m/sec is 8 barns higher Harwell slow-neutron chopper, using samples of powdered PuO2. Result PATTENDEN (1956a) 1050 + 30 1038 + 30 0.2 corrected for Pu<sup>240</sup> and oxygen present RFT slow-neutron chopper. Value given is from a smooth curve fitting 0.6 MOSTOVOI et al. (1957) 1044 1033 the data in the range 0.02 to 0.03 eV BOLLINGER et al. (1958) 1015 + 10 1004 + 11 ANL fast chopper 1.0 The uncertainty quoted allows for a small systematic component associated with 1024 + 8.1 Weighted mean value OS

Total cross-section of Pu<sup>239</sup> for neutrons of 2200 m/sec, from transmission measurements, and derived values of the absorption cross-section

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- PATTENDEN (1956b) reported  $\sigma_{T}[U^{233}] = 590 \pm 15$  barns at 2200 m/sec, from E. measurements with a slow-neutron chopper and a crystal spectrometer. However the data show a step near 0.025 eV which is not found in any other measurements and is probably spurious. Smoothing the data locally gives the somewhat higher value of om listed in Table 4.6, but we prefer to give no weight to the result.
- It can be seen from Table 4.6 that SAFFORD et al. (1960) and BLOCK et al. (1960) made some of the most precise measurements for U<sup>233</sup>. However the samples were F. all prepared at Oak Ridge from a common stock. To avoid bias we have somewhat increased the weights given to other measurements.

#### 4.5 FISSION CROSS-SECTIONS

#### 4.51 Methods of measurement

The direct absolute measurement of a fission cross-section involves the following elements :-

- (1) Determination of the fission rate during neutron bombardment of a fissile sample.
- (2) Assay of the fissile material.
   (3) Measurement of the neutron flux through the sample.

The fission rate may be measured by observing the ionisation produced by the fission-fragments, by use of photographic emulsions or of ionisation chambers (fission chambers). Usually ionisation chambers have been used, with 2 x counting geometry. In ideal conditions this should permit all the fissions to be detected because the two fragments fly apart in opposite directions.

Usually the fissile material has been used in the form of a thin foil. To minimise absorption of the fission fragments the foil is made as an almost weightless deposit on a smooth flat support. The fissile materials are alpha active and it is necessary to bias the fission counter so that it will discriminate against the alpha particles. This results in the loss of some weak fission pulses also, for which a correction must be made. The amount of fissile material which can be used is limited by its alpha activity. If the rate of alpha emission is too high the bias must also be high, to prevent saturation of the counter by a pile-up of alpha pulses during the resolving time. The correction for loss of fission pulses is then correspondingly large and uncertain.

A closely related problem, which has received little attention, is that of the effective counting geometry. A proportion of the fission fragments emerges from the foil at almost grazing angle, and it may be imagined that the efficiency of their detection is influenced by the uniformity and flatness of the foil and the nature of the backing material, as well as the bias voltage. Recently DERUYTTER (1960a) has developed a  $4\pi$  fission chamber. This requires the use of a nearly transparent backing for the fission foil, but improves the discrimination against alpha pile-up. However it cannot be said that the problem of the effective counting geometry is completely removed.

Assay of the fissile sample must ultimately be based on weighing, with isotopic analysis by mass-spectrometry and alpha spectrometry, but the problem is complicated by the meagreness of the fissile deposit on the foil, and the assay measurements are often very indirect. For typical examples of the methods used the reader is referred to the papers of ALLEN & FERGUSON (1957), BIGHAM et al. (1958), and RAFFLE (1959). The fundamental advantage of the novel technique described by SAPLAKOGLU (1958) is that the sample assay is by a fairly direct route. With enriched  $U^{235}$  the alpha activity stems mainly from the small proportion of  $U^{234}$  present, so that assay by absolute alpha counting is not very precise. Some authors have overcome this difficulty by comparative fission counting against a foil of natural uranium, which can be assayed by alpha counting. However it looks as if this technique throws away much of the advantage of using enriched uranium.

Measurement of the neutron flux may be by activation, of gold or manganese for example, followed by absolute  $\beta$ -counting, or by counting the alpha particles which result from neutron capture in Li<sup>o</sup> or B<sup>10</sup>. To minimise scattering corrections it seems desirable that the flux monitor should be sited as nearly as possible in the same position and environment as the fissile sample. To illustrate this remark, note that in the early measurements of TUNNICLIFFE (1951) the neutrons scattered in the foil backing would have caused about  $6\frac{1}{270}$  of the fissions. These scattered neutrons, however, being isotropically distributed, would mostly have missed the flux monitor which was some inches away. On the other hand, the albedo of the walls and end of the BF3 chamber used as flux monitor would have enhanced the neutron count rather than the fission count.

### 4.52 The experimental date

Absolute measurements of the fission cross-sections are collected in Table 4.9, and a number of relative measurements are given in Table 4.10. A few of the earliest measurements have been omitted, and for some of the unpublished data we give references only, if the information available at second-hand is incomplete or inconsistent. The values in the tables mostly differ a little from those given in the references as a result of corrections which are discussed in section 4.2.

Some of the more recent experiments have been re-examined by HANNA (1960). However he was unable to discover why the results of the careful work on U<sup>235</sup> by FRIESEN et al. (1956) and by SAPLAKOGLU (1958) are so far from the preferred value.

In most experiments fission counting and alpha counting were done with ionisation chambers; however POPOVIC and his colleagues (1953, 1955) used photographic emulsions for these purposes.

IEONARD et al. (1956) measured the  $U^{235}$  fission cross-section, and also compared that of  $Pu^{239}$  with it. However we prefer to interpret the latter measurement as an absolute measurement of the plutonium fission cross-section in which " $U^{235}$ " foil played only an intermediary role. In this way the uncertainties in assay of the  $U^{235}$ foil are climinated. Similar arguments may be used in relation to the measurements by BIGHAM et al. (1959). These however, in view of their quality and self-consistency, we have presented in both tables, so they are given more weight than is at first apparent. SAFFORD & MELKONIAN (1959) measured 1 +  $\alpha$  for  $U^{235}$  at 0.00291 eV. By using additional data we have derived values for both op and 1 +  $\alpha$  at 0.0253 eV. The original datum appears in two roles, and this was taken into consideration in the weighting.

In weighting the data we have preferred to ignore the measurements in the illknown reactor spectra, and have generally given reduced weight to those using neutrons from thermal columns.

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Reference	σ <sub>F</sub> a U <sup>233</sup> barns	t 2200 m/ U <sup>235</sup> barns	Pu <sup>239</sup> barns	Weight	ôp in broad spectrum	Method and comments
BAILEY et al. (1944)		?				Unpublished. Comparisons of U <sup>235</sup> rith boron and lithium
DEUTSCH & LINENBERGER (1944)		?				Unpublished. Comparisons of U <sup>235</sup> with ectivations of Au and Mn
MARSHALL (1944)		592 ± 25		0.1	577	Comparison with lithium in thermal column spectrum
BISWAS & PATRO (1949)		557 ± 13		o	539 <u>±</u> 12	Comparison of natural U with boron, in peraffin block with Ra- Be source and Cd differences. The Cd ratio for U was very low, suggesting appreciable fast-fission of U 208. Result not corrected for finite thickness of fission foil, so may be 5% low
FACCINI & GATI (1950 with revision cited by COHEN et al. (1952b)		611 ± 21		0	584 ± 18	Similar to above with lithium as standard. The Cd ratios were rather low
TUNNICLIFFE (1951)	512 <u>+</u> 30		657 <u>+</u> 60	0		Measurements at 0.050 eV with neutrons from a crystal spectro- meter which was also used to measure the energy dependence of $\sigma_{\rm P}$ . Assay by absolute alpha counting, with BF3 chamber as flux moni- tor. Scattering of neutrons by foil backing and BF3 counter give rise to large uncertainties; see comment in § 4.51
BARLOUTAUD & LÉVÊQUE (1952)		621 ± 14		0	599 ± 12	Comparison of natural U with activation of Mn in a reactor neutron beam, with Cd differences
POPOVIC & GRIMELAND (1953)		584 ± 15 587 ± 17		0.3	569 ± 14 564 ± 14	Measurements with natural U in thermal column and reactor core respectively. U assayed by alpha counting. Fission and alpha counting by use of photographic emulsions. Sodium activation used to monitor neutron flux
POPOVIC & SAELAND (1955)	525 ± 17			0.4	526 ± 17	Method as above. Measurements in thermal column only. Alpha count may not have been corrected for U <sup>232</sup> content of sample. so result may be slightly too low
GERASIMOV (1956)			625	0		An old unpublished USSR value, by comparison with lithium
GERASIMOV (1956)	525	570	720	000		USSR recommended values. Method unpublished. These data may have been derived from $\sigma_A$ by using data on $\alpha$ , or $\bar{\nu}$ and $\eta$ , so we do not use them when averaging

TABLE 4.9 Fission cross-sections of U<sup>233</sup>, U<sup>235</sup>, and Pu<sup>239</sup> for neutrons of 2200 m/sec

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Reference	07 U <sup>233</sup> barns	at 2200 m U <sup>235</sup> barns	/sec Pu <sup>239</sup> barns	Weight	ôp in broad spectrum	Method and comments
FRIESEN et al. (1956)		549 <u>+</u> 6		o		Comparison with Au activation using monokinetic neutrons of 0.10 eV, from a crystal spectrometer which was also used to measure the energy dependence of op. Assay by comparative alpha counting against a standard which had been foil assayed by isotope dilution
LEONARD et al. (1956)			676 ± 21	0.6		Measured as above by comparison with the U <sup>235</sup> foil. Evidence reported by RAFFLE (1959) throws some doubt on the assay of the U <sup>235</sup> , which is therefore best treated as a purely nominal inter- mediary in determination of the plutonium cross-section. Samples were of several different isotopic compositions, none apparently containing more than 30% Pu <sup>239</sup>
SAPLAROGLU (1958)		605 ± 6		0.5		Novel technique permitting rather direct assay of the fissile sample. Flux determination independent of any standard cross- section, but the monitor was remote from the fissile sample
BIGHAM et al. (1958)	514 ± 6	570 ± 6	740 ± 9	1 0.5 1	514 <u>+</u> 4.8 555.7 <u>+</u> 4.5 780.1 <u>+</u> 7.4	Irradiations in a large $D_20$ thermal column at ~27°C. $\sigma_{\rm p}[U^{233}]$ $U^{233}$ measured absolutely in comparison with gold activation, the gold foils being disposed around the periphery of the fission foil, and in the same plane. $U^{e_{33}}$ and $Pu^{233}$ compared with $U^{233}$ and with one another. $Pu^{239}$ assayed by absolute alpha counting. $U^{233}$ and $U^{235}$ by isotope dilution, with comparative alpha and fission counting. In revising $U^{235}$ and $Pu^{239}$ data we treated the $U^{233}$ as a nominal intermediary
RAFFLE (1959)	506 ± 17 513 ± 13 514 ± 14	584 ± 18 595 ± 11 563 ± 12	700 ± 20 686 ± 14 645 ± 29	0.4 0.3 0.3 0.2 0 0.6 0.5 0	$514 \pm 13515 \pm 14580 \pm 11538 \pm 10724 \pm 14768 \pm 25$	Recalculated from the data given in RAFFLE's paper. The three data for each nuclide are respectively from measurements with monokinetic neutrons from a slow chooper, neutrons from a ther- mal column, and sub-Cd neutrons from a reactor core. Assay of the fission foils was by various methods and the recalculation makes use of some measurements which RAFFLE recorded but did not use. The weighting of companion measurements is also revised. The neutron flux was monitored with a EF3 chamber, calibrated by gold activation in a thermal beam. The gold sample had the same environment as the fission foils

TABLE 4.9, continued Fission cross-sections of  $U^{233}$ ,  $U^{235}$ , and  $Pu^{238}$  for neutrons of 2200 m/sec

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### TABLE 4.9 continued

Fission cross-sections of  $U^{233}$ ,  $U^{235}$ , and  $Pu^{239}$  for nuetrons of 2200 m/sec

1.4

Reference	OF U233 barns	at 2200 m U <sup>235</sup> barns	/sec Pu <sup>285</sup> barns	Weight	or in broad spec trum	Method and comments
SAFFORD & MELKONIAN (1959)		538 ± 6		0.7		1 + $\alpha$ measured at 0.00291 eV. To derive $\sigma_{\rm F}$ at 0.0253 eV we used data on $\sigma_{\rm T}$ and on the energy dependence of $\sigma_{\rm F}$ by SEPPI et al. (1958) and by SAFFORD et al. (1959). See also section 4.6
DERUYTTER (1950b)		585 <u>+</u> 6		1		Counting rates of $4_{\pi}$ fission-chamber and $B^{10}$ chamber compared, using neutrons from a slow chopper. $B^{10}$ chamber calibrated by gold activation in sub-Cd spectrum from thermal column; the spec- trum was analysed with the chopper
Weighted means	514•4 ± 7•9	587 ± 5.0	707 ± 18			

Reference	or ratios :	at 2200 m/sec	- 239 - 233	Weight	Ubserved ratio in broad	Method and comments
	0 /0	Pu 70	Pu - / 0		spectrum	
ANDERSON & MAGLE (1944)		?				Unpublished
De WIRE (1944)		?				Unpublished
ANDERSON & MAY (1944)	0.883 ± 0.018			0.1	0.907 <u>+</u> 0.017	Measurements in thermal column of CP2. U <sup>238</sup> assay by absolute alpha counting; U <sup>285</sup> by weighing and volumetric dilution
ZINN & KANNER (1945)	0.903 ± 0.018			0	0.927 ± 0.014	As above. Methods of assay not reported
COHEN et al. (1952a, 1952b)		1.231 <u>+</u> 0.051		0	-	Comparisons of Pu with natural U in a reactor neutron beam. Assay by comparative alpha counting
COCKROFT (1952)		1.288 ± 0.018		0.6	1.397 ± 0.007	Comparisons of Pu with natural U in beams from top thermal column and reactor core respectively. Assay from comparative alpha counting and alpha
		1.209 ± 0.055		0	1.051 ± 0.015	spec trome try
AUCLAIR et al. (1956)		-	1.440 ± 0.034	0.2	1.587 ± 0.016	Comparisons in reactor neutron beam, with Cd differences. Beam spectrum and energy dependence of cross-sections observed with a slow chopper
MoMILLAN et al. (1955)	0.919 ± 0.027	1.293 <u>+</u> 0.035		0	0.948 1.515	Comparisons of gross fission-produce activities after irradiation in central moderator of TTR. Assumes that activity per fission is independent of target nuclide
SELLARS et al. (1955)	0.921 ± 0.008	1.328 ± 0.021		0.6 0.4	C.946 1.440	Comparisons in thermal column of CP3. Assay by weighting and volumetric dilution, and for Pu <sup>239</sup> by absolute alpha counting also. Isotopic analy- sis not discussed
PRATT et al. (1956)		1.25 ± 0.06		0	1.725 ± 0.027	Comparisons in reactor neutron beam, with Cd dif- ferences. Neutron temperature said to be 222°C
BIGHAM et al. (1958)	0.9089 ± 0.0066	1.297 ± 0.014	1.429 ± 0.015	1 1 1	0.9333 1.4038 1.5045	See comments in Table 4.9
Weighted mean values	0.9107 ± 0.0077	1.301 ± 0.016	1.431 ± 0.017	Note	that all the m	easurements listed above were made in broad neutron

TABLE 4.10 Relative measurements of the fission cross-sections of U<sup>233</sup>, U<sup>235</sup>, and Pu<sup>239</sup> for neutrons of 2200 m/sec

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### 4.6 CAPTURE TO FISSION RATIOS

### 4.61 Methods of measurement

For a direct measurement of the ratio of capture and fission cross-sections,

$$\alpha = \sigma / \sigma_{\rm F},$$

a fissile sample is irradiated and the changes in its isotopic composition are then determined by mass-spectrometry or by alpha spectrometry. The number of fissions may be determined from the destruction of the fissile isotope, or from fission-product activities, or otherwise. In order to obtain measurable changes in isotopic composition comparatively long irradiations are necessary at high flux. In practice every such irradiation has been made in a reactor core, and little effort has been made to characterise the irradiation spectrum. The high resolving-power of the multi-stage spectrometers now available will permit the use of the less intense neutron fluxes available in thermal columns so that the uncertainties of the irradiation spectra can be greatly reduced.

The next step is to estimate  $\alpha_0$ , for monokinetic neutrons of 2200 m/sec, from the values of  $\hat{\alpha}$  obtained with broad neutron spectra. The fissile samples have usually been fairly thin so we were able to use WESTCOTT's (1960) tables, after guessing the neutron spectra; see § 4.22. Available evidence shows that  $\bar{\nu}$ , the neutron yield per fission, is sensibly constant over the slow neutron region, and therefore the energy dependence of  $(1 + \alpha)$  is the same as that of  $1/\eta_0$ 

Measurements of  $1 + \alpha$  have been made by a different technique with neutrons of long wavelengths; COCKING (1958), SAFFORD & MELKONIAN (1959). Essentially the transmission and the fission rate are compared. Calculation of  $\alpha_0$  from such a measurement can at present only be done by using data on the energy dependencies of fission and absorption cross-sections.

### 4.62 Experimental data for U233

Only three measurements of the capture to fission ratio have been reported, and they are given in Table 4.11. There is an error of sign in one of the genetic relationships given by INGHRAM et al. (1956). For this reason the result was recalculated by CHAPMAN (1959) and was found to be somewhat sensitive to the neutron spectrum, through the value assumed for  $\hat{\sigma}_{\rm A}[U^{238}]$  in allowing for  $U^{238}$  burn-up. The uncertainty is enhanced by our ignorance of the extent to which resonance absorption in the dilute  $U^{236}$  in the sample is shielded by the  $U^{238}$  in the reactor fuel. KUKAVADSE et al. (1956) avoided this difficulty by isotope dilution after the irradiation. This technique is more complex, and the samples used were rather thick, see note A of § 4.43, so in Table 4.11 we have increased the uncertainty quoted by the authors. The value of  $\alpha$  is not much changed on going from a reactor spectrum to 2200 m/sec, because  $\alpha$  is fairly constant in the thermal region, and indeed over almost the whole energy range except at the 2.3 eV resonance. The evaluation of the correction and its uncertainty have been discussed in some detail by EVANS & FLUHARTY (1959). We have used their estimate of the uncertainty, but have preferred to re-evaluate the corrections themselves from the tables of WESTCOTT (1960).

### TABLE 4.11

Reference	Value in experimental spectrum	Corrected to 2200 m/sec.	Conments
INGHRAM et al. (1956)	0.096 <u>+</u> 0.007	0.095 <u>+</u> 0.009	Mass-spectrometer analysis before and after irradiation in NRX reactor. Original value 0.0976 + 0.0018 recalculated with revised cross- section data.
KUKAVADSE et al. (1956)	0.0968 <u>+</u> 0.0046	0.0961 ± 0.0068	Absolute alpha counting, and mass- spectrometry after reactor irradiation. Reported value $\alpha/(1+\alpha)=0.0870 \pm 0.003$ increased by 1.4% to allow for U <sup>234</sup> burn-up during irradiation. EVANS and FLUHARTY (1959).
COCKING (1958)	0.113 ± 0.018		Measurement of 1 + a with cold neutrons of about 0.0011 eV
Weighted mean val	ue	0.0958 ± 0.0063	

Capture to fission ratio, a, for U<sup>233</sup> with slow neutrons

The close agreement of the first two data in Table 4.11 is probably somewhat fortuitous. COCKING's (1958) measurement was made with effectively monokinetic neutrons of long wavelength. Unfortunately the variation of  $\alpha(E)$  at very low neutron energies is not known so that his result gives no information on  $\alpha_0 [U^{233}]$ .

### 4.63 Experimental data for U235

The data on the capture to fission ratio for U<sup>235</sup> are given in Table 4.12. Most of the measurements were by mass-spectrometry after irradiations in reactors, with little or no attempt to characterise the neutron spectra. Corrections to 2200 m/sec have been evaluated by using WESTCOTT's (1960) tables, with plausible assumptions about the neutron temperatures and the epithermal fluxes. It is difficult to make any convincing assessment of the uncertainties involved, and measurements in thermal spectra are much needed.

### TABLE 4.12

## Capture to fission ratio, $\alpha$ , for U<sup>238</sup> with slow neutrons

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Reference	Value in experimental spectrum	Corrected to 2200 m/sec	Weight	Comments
DEUTSCH et al. (1946)	0.188 <u>+</u> 0.008 (probable error)	0.173 ± 0.012	0.02	Mass-spectrometry, with radio- chemical determination of no. of fissions. Irradiation in Oak Ridge reactor gave very low yield U <sup>236</sup> /U <sup>235</sup> ~0.03%
- ditto -	0.188 ± 0.011 (probable error)	0•172 ± 0•017		As above. Irradiation in Hanford reactor gave rather low yield $U^{236}/U^{235} \sim 0.2\%$
WILLIAMS (1946)	0.177 <u>+</u> 0.006	0.165 <u>+</u> 0.007	1.0	Mass-spectrometric analyses of $U^{236}$ production and $U^{235}$ burn- up. Irradiation in Hanford reactor give $U^{236}/U^{235} \sim 2\%$
KANNE et al. (1956)	0.174. <u>+</u> (10%) (probable error)	0.165 <u>+</u> 0.026	0.1	Irradiation in NRX. Two-stage mass-spectrometer for U <sup>236</sup> assay, but no. of fissions crudely estimated, from radio- chemical yields of Cs <sup>137</sup> and Ce <sup>144</sup>
TINGEY & VANCE (1955)	0.184 <u>±</u> 0.012	0.177 <u>+</u> 0.012	0.3	Irradiation in MTR
CRAIG et al. (1958)	0.194 <u>+</u> 0.002	0.184 <u>+</u> 0.006	0.7	Sections of fuel element highly irradiated in NRX. Analyses by mass-spectrometry and by alpha spectrometry, respectively
(1959)	± 0.003			
COCKING (1958)	0.172 <u>+</u> 0.022			Measurement of $1 + \alpha$ with nearly monokinetic cold neutrons of about 0.0011 eV
SAFFORD AND MELKONIAN (1959)	0.171 ± 0.009	0.159 ± 0.014	0.9	Measurement of 1 + α at 0.00291 eV. Transformation to 2200 m/sec discussed in text
CORNISH (1960)	0.200 <u>±</u> 0.008	0.189 ± 0.008	0.5	Irrediation in DIDO. $U^{ess}$ burn- up measured by mass-spectrometer relative to the $U^{234}$ present in low abundance, and from gross fission-product $\gamma$ radiation
Weighted	mean	0.1714 ± 0.0077		See § 4.63, Note A

The measurements by COCKING (1958) and by SAFFORD & MELKONIAN (1959) were made with essentially monokinetic neutrons at long wavelengths. Estimation of  $\alpha_0$  from these data depends at present on the accuracy of relative measurements of  $\sigma_A$  and  $\sigma_P$ at the energies of interest. The most suitable data appear to be those of SEPPI et al. (1958) and of SAFFORD et al. (1959), who measured  $\sigma_A$  at long wavelengths and in the thermal region, using crystal spectrometers. Relative measurements of  $\sigma_P$  at 0.00291 and 0.0253 eV have been made by the same two groups, and the results are cited by SAFFORD and MELKONIAN (1959). Using the values 1.171 for  $(1 + \alpha)$  at 0.00291 eV, which was reported by the authors last named, the following estimates are obtained for  $\alpha$  at 2200 m/sec:

Origin of cross-section data	α <sub>0</sub> [Ü <sup>235</sup> ]	
SEPPI et al. (1958)	metal samples	0.179 ± 0.013
SAFFORD et al. (1959)	metal samples	0.164 ± 0.014
	liquid samples	0.152 ± 0.007

The weighted mean value is  $\alpha_0 = 0.159 \pm 0.014$ , wherein the uncertainty of the initial datum is included.

The dispersion of the data presented in Table 4.12 is not inconsistent with the uncertainties quoted there. However, in weighting the data the following comments should be considered:

- A. The majority of the data are from samples irradiated in nuclear reactors, and the uncertainties given in the table for these data should be increased by  $\pm$  0.005 to allow for systematic errors in the corrections to 2200 m/sec.
- B. For the first two measurements in Table 4.12, reported by DEUT3CH et al. (1946) the atomic ratios of  $U^{236}/U^{235}$  after irradiation were very low. It is difficult to believe the absolute accuracy of the mass-spectrometric analyses could have been better than  $\pm 30.5$  and  $\pm 5.5$  respectively.
- C. Reduced weight is given to the result by CORNISH (1960) for similar reasons. He irradiated very small samples of highly enriched U<sup>235</sup> on aluminium foils. Because of contamination from natural uranium present in the aluminium, the mass-spectrometric assessments of U<sup>235</sup> burn-up depend on comparisons with the U<sup>234</sup> which was present in low abundance only. There is likely to be a strong systematic element of uncertainty in the measurements.
- D. KANNE et al. (1956) used a mass-spectrometer of high resolution, but made only a crude measurement of the number of fissions. They were more interacted to study the relative variations of  $\alpha$  over a broad energy range.
- E. The measurements reported by CRAIG et al. (1958) and by BIGHAM et al. (1959) followed a prolonged irradiation which gave a good yield of  $U^{238}$ . However the samples were sliced from a thick fuel element. Elaborate calculations by KUSHNERIUK (1959) have been used to infer  $a_0$  from the observed results, but the

\*We have assumed  $\sigma_s = 16.0 \pm 0.7$  barns per  $0^{235}$  atom in the liquid samples both at long wavelengths and in the thermal region. For the metal samples  $\sigma_s = 1 \pm 1$  barn at 0.00291 eV, and 14.4  $\pm$  3.7 barns at 0.0253 eV.

complexity of the environment detracts very much from the reliability of the result.

It may reasonably be argued that greater weight should be given to the measurement by SAFFORD & MELKONIAN (1959) since the other data are from integral measurements whose correction to 2200 m/sec is based on guess-work. However the datum in question has also been used, in § 4.52 and Table 4.9, for direct determination of  $\sigma_{\rm F}^{0}$  [U<sup>235</sup>] so that in fact it has been given double weight.

### 4.64 Experimental data for Pu239

Only two measurements of the capture to fission ratio in Pu have been reported; they are given in Table 4.13 and it may be seen that the measurements were only of low precision.

The measurements reported by KANNE et al. (1956) appear to have been made about 1947. The uncertainty in the result arises mainly from the fission yield data which were used. Correction to 2200 m/sec is quite sensitive to the neutron spectrum. For a rough estimate we have used the parameters of WESTCOTT (1960) assuming T = 80 to 220 °C for the effective temperature of the thermal neutrons, and r = 0.03 to 0.09 for the epithermal intensity in the Hanford irradiation. Then

$$1 + \alpha_{0} = f_{*} (1 + \hat{a})$$
 (4.9)

with f = 0.093 + (1.8%).

### TABLE 4.13

Reference	Value in experimental spectrum	Corrected to 2200 m/sec	Comments
KANNE et al. (1956)	0.42 <u>+</u> (15%) (probable	0.310 <u>+</u> 0.090	Irradiation in Hanford pile. No. of fissions from fission-product analy- sis. Pu <sup>240</sup> assayed by spontaneous fission rate before and after irradia- tion
COCKING (1958)	0.300 <u>±</u> 0.01+0	0.300 <u>+</u> 0.10	Measurement of $1 + \alpha$ with cold neu- trons of about 0.0011 eV
Mean	valuə	0.305 <u>+</u> 0.10	

Capture to fission ratio, a, for Pu<sup>239</sup> with slow neutrons

To calculate  $\alpha(2200 \text{ m/sec})$  from COCKING's (1958) measurement with cold neutrons one must have recourse to available data on the energy dependence of  $\sigma_F$  and  $\sigma_A$  in the energy range 0.0011 to 0.0253 eV. These are very inexact, giving at best

### for the correction factor of equation (4.9).

### 4.7 THE AVERACE NUMBER OF NEUTRONS EMITTED PER FISSION

### 4.71 Methods of measurement

To measure  $\bar{\nu}$ , the average number of neutrons evolved per fission, one must be able to count fission events and the neutrons emitted. In order that the fissions may be counted it is necessary to use a very thin fissile sample and a reasonably low fission rate. The intensity of the fission neutrons is consequently very low, often lower than that of the fast neutron background. Various methods have been used to overcome this difficulty; for examples the reader is referred to the papers by SNYDER & WILLIAMS (1944), KALASHNIKOVA et al. (1955), and SANDERS (1956). For most measurements of  $\bar{\nu}$  the neutrons have been counted in coincidence with fission events. This greatly alloviates the problem of distinguishing the fission neutrons from the neutron background, and also eliminates the need to know the absolute efficiency of the fission counter. However it should be noted that the yield of neutrons from fission is correlated with the mass ratio and energy of the fission fragments, and there is a directional correlation also. In measuring  $\bar{\nu}$  with coincidence techniques care must be taken to ensure the random sampling of fission events, and this may be done, for instance, by trying to count nearly all the fissions.

To measure  $\overline{\nu}$  absolutely the efficiency of the neutron counter must be determined absolutely. Usually this has been done indirectly, by using a standard neutron source for example; (the reliability of the source calibrations is briefly examined in the next section). In recent work the efficiency of the neutron counter has been measured by observation of "associated charged particles"; DIVEN et al. (1956), COLVIN & SOWERBY (1958). This method is not only more direct, but also permits calibration of the counter efficiency as a function of neutron energy.

For relative measurements of  $\bar{\nu}$ , with different fissile nuclei for example, absolute calibration of the neutron counter is unnecessary. It is important nonetheless, to ensure that the response is insensitive to the small differences between the spectra of emitted neutrons.

### 4.72 Corrections to published data

### Neutron-source calibrations

Calibrations and comparisons of neutron sources have been discussed in detail by LARSSON (1958), RICHMOND (1958), and GEIGER (1960). They showed that there is good agreement between source strength measurements by various techniques in different laboratories. None of these authors has made a "least squares" analysis of all the available data, and a complete review is much needed. We do not attempt that here, but have critically re-examined the data collected by LARSSON (1958) and RICHMOND (1958). Recommended values of the source strengths which are of interest in the present context are given in Table 4.14, and a few general comments follow:

### TABLE 4.14

Sources	Output at given date. Neutron/sec.	Reference and comments
Los Alamos No. 44 Ra- α -Be	$(6.06 \times 10^6) \pm (5\%)$ AugDec., 1944	WALKER (1944); revised as noted in § 4.71, note B
Los Alamos No. 43 Ra- α -Be	(9.19 x 10 <sup>6</sup> ) <u>+</u> (5%)	WALKER (1944); by comparison with LA source No. 44 above
USSR N.22 Ra- $\alpha$ -Be	(5.96 x 10 <sup>6</sup> ) <u>+</u> (3%) June, 1951	EROZOLIMSKY & SPIVAK (1957)
Oxford Rd Th- Y -D <sub>2</sub> 0	$(6.20 \times 10^4 \pm (1.6\%))$ 25th Jan., 1955	RICHMOND (1958); revised as noted in § 4.71, note C
Harwell Pu <sup>240</sup> spont. fiss.	$(2.036 \times 10^4) \pm (1.7\%)$ 1956-7	RICHMOND (1958); by comparison with the strength of the Oxford source about 1956-7
Natural U spont.fiss. sources	15.6 <u>+</u> (4%) per K3.	Typical value. See § 4.71, note E

Output of some standard neutron sources; recommended values

- A. There is inconclusive evidence suggesting that the strength of the Harwell Ra-α-Be source may have fallen by 6.6% about 1951-2. Evidence for a 3% loss of intensity from the Los Altmos source no. 40 has been cited also, by COON (1955). Such changes might result from mechanical vibration for example, or from radar leakage. Because of these possible changes the early measurements on the Harwell and Los Alamos sources can have no weight in evaluating calculations for the post 1955 era.
- B. Conversely, the intensities of the Los Alamos sources in 1944 should be inferred from the contemporanous studies by WALKER (1944). His calibrations have been increased in Table 4.14, by 2.2% to allow for absorption of fast neutrons by the 016 ( $\eta$ ,  $\alpha$ ) reaction, and by 0.2% for absorption by the source itself of neutrons thermalised in the water bath.
- C. Three independent calibrations of the Oxford photo-neutron source are listed by RICHMOND (1958). In correcting the later measurements to the date of the first calibration a half-life of 1.90 years was assumed. However both the available date. MEITNER (1918) and KIRBY et al. (1956), indicate a half-life of 1.910

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years, and this leads to the value given in Table 4-14 for the weighted mean of the three direct calibrations. The strength of the Harwell Pu spontaneous fission source was obtained by comparison with the Oxford source about the end of 1956, and we have revised the result correspondingly.

- D. Several similar small corrections may be made to the other calibration data reviewed by LARSSON (1958) and by RICHMOND (1958). By chance these data together then exactly corroborate the value proposed in Table 4.14 for the strength of the Oxford source, and they were taken into account when evaluating the uncertainty. The Russian source calibrations by EROZOLIMSKY & SPIVAK (1957) appear by comparison to be 0.5% too low, but it has not been worth while to modify the original value.
- E. Spontaneous fission sources of natural uranium have also been used. Recent reports, WALTNER & LEONARD (1959), LITTLER (1959), suggest that the spontaneous neutron output from a small sample is (15.3 ± 0.6) neutrons/Kg sec. A slightly higher value is proposed in Table 4.14 to allow for the fast neutron multiplication which must occur in sources of practical size.

### Delayed neutrons

In tables 4.15 and 4.16 below,  $\bar{\nu}$  has been used to denote the average number of neutrons evolved per fission, <u>including both prompt and delayed neutrons</u>. In many of the experiments coincidence techniques were used so that only the prompt neutrons were observed. The delayed-neutron yields which we have added are, from KEEPIN et al. (1957) and COX et al. (1958):

Fissile nucleus	No. of delayed neutrons per fission		
U233 + thermal neutron	$0.0066 \pm 0.0004$		
U235 + " "	$0.0158 \pm 0.0007$		
Pu239 + " "	$0.0061 \pm 0.0004$		
Cf252 spontaneous fission	$0.0086 \pm 0.0010$		

This correction is very small and its contribution to the uncertainty of  $\bar{\nu}$  is quite negligible.

### 4.73 The experimental data on $\overline{v}$

In Tables 4.15 and 4.16 are collected the results of absolute and relative measurements of  $\bar{\nu}$  for fission of  $U^{233}$ ,  $U^{235}$ , and  $Pu^{239}$  by slow neutrons. Absolute measurements of  $\bar{\nu}$  for spontaneous fission of  $Cf^{252}$  have also been made, and  $\bar{\nu} [U^{235}]$  can be deduced by the use of comparative data, so these results have been included also in the tables.

For most of the measurements the fissions were induced by bombardment with thermal neutrons or with neutrons from thermal reactors. However the results can equally well be interpreted as 2200 m/sec. values, because with slow neutrons  $\bar{\nu}$  is virtually independent of the bombarding energy, and even on a broader scale the rate of increase of  $\bar{\nu}$  is only about 0.12 (neutrons/fission) per MeV; see for instance BONDARENKO et al. (1958). The insensitivity of  $\bar{\nu}$  to small changes of the incident energy is an expected consequence of the fact that  $\nu$  is averaged over a very broad

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population. Measurements using monochromatic neutrons confirm this expectation for both  $U^{235}$  and  $Pu^{239}$  over the range 0.025 to 0.4 eV, within the experimental uncertainty of  $\frac{1}{2}$  to 1 percent; see for example LEONARD et al. (1955), and AUCLAIR et al. (1956).

In many of the experiments the fission neutrons were detected with "wax castle" counter assemblies, consisting of a ring of BF3 chambers embedded in a matrix of paraffin wax. Inevitably such devices are quite sensitive to variations of the neutron spectrum, and this is confirmed by the observations of KENWARD et al. (1958). Protonrecoil counters also have been used for fast neutron detection, and suffer even more strongly from the same defect. The large liquid scintillation counter used by DIVEN et al. (1956) has a much more uniform response, and so too do the boron-pile counter of COLVIN & SOWERBY (1958) and the water-bath technique used by BOWMAN & THOMPSON (1958).

The following considerations affect the weighting of the data:

- (a) KENWARD et al. (1958), MOAT et al. (1959), and COLVIN & SOWERBY (1959) all used the same standard source for calibration purposes, so there is a large systematic component in the uncertainties of their results in Table 4.15. In the same table the data of JOHNSTONE (1954) and of SANDERS (1956) are similarly correlated.
- (b) The spectra of the various spontaneous fission and "mock-fission" sources used for calibration purposes are all rather uncertain; see TERRELL (1959) and BONNER (1959). These uncertainties affect the reliability of the measurements obtained with wax castle and proton-recoil counters.
- (c) The absolute measurements of  $\bar{\nu}$  for  $U^{233}$  and Cf<sup>252</sup> are of low weight when (a) and (b) have been taken into account. To simplify the analysis we treated them as estimators of  $\bar{\nu}[U^{235}]$ , by using the weighted mean ratios from Table 4.16. The loss of accuracy is negligible.
- (d) The fission-rate comparisons used by McMILLAN et al. (1955) are based on the unproven assumption that the gross activity of fission products 20 to 130 minutes after fission is independent of the fissioning nuclide. These measurements are better interpreted as giving relative values of  $\overline{\nu} \sigma_{\overline{p}}$ ; see Table 4.18.
- (e) Values for v of Cf<sup>252</sup> were published by CRANE et al. (1955 and 1956), but are now considered to be erroneous; CRANE (1960). A revised value is given by BOWMAN & THOMPSON (1958), and is listed in Table 4.15, but few details are available.

### TABLE 4.15

v, the average number of prompt and delayed neutrons emitted per fission under slow-neutron bomberdment

Reference	U233	U 235	Cf <sup>252</sup> spontaneous fission	Weight	Method and comments
SNYDER & WILLIAMS (1944)		2.55 ± 0.13		0.03	Sample at centre of large graphite moderator. Fission neutrons detected by In foils in Od at various distances from the sample, and compared with neutron field from Los Alamos Ra- $\alpha$ -Be source No. 43. Source strength taken from table 4.14, but reduced by $(1.5 \pm 1)$ % to allow for absorption by threshold reactions in the graphite and for self- absorption of moderated neutrons
KALASHNIKOVA et al. (1955)	2.63 ± 0.08			0.05	Coincidence technique. Wax-castle neutron counter, calibtated with a neutron source whose spectrum was "very similar" to that of fission neutrons. Original result recalculated and delayed neutron contribu- tion added. Source calibration probably by method of EROZOLIMSKY & SPIVAK (1957)
JOHNSTONE (1954)		2.51 ± 0.11		0.02	Apparatus and method sensibly the same as used by SANDERS (1956); see below. Result was corrected to include delayed neutrons
DIVEN et al. (1956)		2.428 <u>+</u> 0.060		0.16	Coincidence technique using incident neutrons of 80 keV. Neutrons counted in large Cd loaded liquid scintallator. Direct calibration by associated particle method. Corrected to thermal energy and to include delayed neutrons
SANDERS (1956)		2.42 ± 0.11		0.02	Coincidence technique. Wax-castle neutron counter calibrated with natural uranium spontaneous fission source. Original result corrected for source strength as in Table 1, and to include delayed neutrons
BOUTMAN & THOMPSON (1958)			3.8 ± 0.16	0.05	Apparently revision of earlier work by CRANE et al. (1955, 1956). They compared neutron output of sample with that of calibrated Ra-Be source by activation of Mn in a water bath. Fission rate observed separately.
KENWARD et al. (1958)		2.421 ± 0.041		0.19	Coincidence technique. Wax-castle neutron counter calibrated with Harwell Pu <sup>240</sup> spontaneous fission source. Delayed neutron yield added to original datum
MOAT et al. (1959, 1960)			3.69 ± 0.07	0.02	Comparison with Harwell Pu <sup>240</sup> spontaneous fission source, using a wax- castle counter. Neutron spectra were thought to be the same. Fission rate determined separately
COLVIN & SOWERBY (1959)		2.418 ± 0.043		0.18	Coincidence method. Boron-pile neutron counter calibrated with Harwell Pu240 spontaneous fission source. Delayed neutron yield added
COLVIN & SOWERBY (1960)		2.435 ± 0.024		1	As above. Boron-pile calibrated directly by associated particle method. Preliminary result, so we treat uncertainty as $\pm 1\%$
Weighted mean values	-	2.438 ± 0.020	-		

Relative values of V, for slow-neutron induced fission of U233, U235, and Pu239, and for spontaneous fission of Cf

Reference	U233 U235	Pu 239 U 235	<u>Pu</u> 239 <u>U</u> 235	Cf 252 U 235	Weight	Method and comments
SNYDER & WILLIAMS (1944)		1.17 <u>+</u> 0.021			0.1	Sample at centre of large graphite moderator. Fission neutrons detected by activation of In foils in Cd at various distances from sample. Fission rates observed separately
DeWire et al. (1944)	1.029 ± 0.010	1.177 ± 0.009			0.2	Coincidence technique. Fission neutrons detected with proton-recoil counter. Original data 1.033 and 1.182 corrected to include delayed neutrons
ANDERSON & MAY (1944)	1.079 <u>+</u> 0.02				0	Fission rate comparisons in thermal column of CP2 reactor Neutron outputs compared in that of CP3, by activation of In foils in Cd at various distances from sample. Result suggests neutron spectrum in CP3 thermal column is harder
JOHNSTONE (1954)		1.159 <u>+</u> 0.033			0	Apparatus and method essentially the same as used by SANDERS (1956) below, who obtained almost identical results which he amended subsequently. Presumably similar amendments are needed here. Revised to include delayed neutrons
McMILLAN et al. (1955) GAERTINER et al. (1958)	1.027 <u>+</u> 0.034	1.247 ± 0.031			0	Fission rates compared from gross fission-product activ- ities, assumed independent of fissile nuclide. Neutron outputs compared by reactivity changes both inside and outside a Cd sandwich. Results recalculated, and "probable errors" converted to standard errors. We could not reproduce original values 1.017 and 1.251 from the data reported
KALASHNIKOVA et al.	1.035 ± 0.010	1.188 ± 0.012			0.3 0.3	Fissions and neutrons counted simultaneously but not in coincidence. Cd filter in incident beam for background correction. Coincidence measurements using a wax-castle neutron counter gave results which agreed within experimental uncertainties. Effects due to small differ- ences of fission neutron spectra were thought negligible
DIVEN et al. (1956)	1.042 ± 0.022	1.229 ± 0.028		1.566 ± 0.025	0.07 0.06 0.6	Coincidence technique with incident neutrons of 80 keV, using large Cd loaded liquid scintillator as neutron counter. Results corrected to thermal energy and to include delayed neutrons

Continues next page

### TABLE 4.16, continued

il a

Relative values of  $\overline{\nu}$ , for slow-neutron induced fission of U<sup>235</sup>, U<sup>235</sup>, and Pu <sup>239</sup>, and for spontaneous fission of Cf <sup>252</sup>

Reference	U 238 U 235	Pu239 U235	Pu 239 U 233	Cf252 U235	Weight	Method and comments
SANDERS (1956)	1.006 ± 0.017	1.179 ± 0.033			0.2 0.05	Coincidence method, with wax-castle neutron counter. Original results revised, SANDERS (1960), to 1.010 and 1.184 for prompt neutrons, and are amended here to include delayed neutrons. Uncertainties quoted allow $\pm$ 1% for possible effects of small differences in the spectra of fission neutrons
JACOB (1958)			1.160 <u>+</u> 0.020		0.1	Coincidence method, with wax-castle neutron counter. No allowance for possible effects of small differences in fission-neutron spectra. Result 1.165 amended to include delayed neutrons.
DeSAUSSURE & SILVER (1959)	1.020 ± 0.010	1.222 <u>+</u> 0.010			0.3	Coincidence method, with proton-recoil counter as fast neutron detector. Uncertainty of only + 2% allowed for possible effects of small differences in spectra of fissi neutrons. Results amended to include delayed neutrons
MOAT et al. (1959)				1.542 ± 0.019	1	Coincidence method, with large liquid scintillator as neutron detector. Improved dead-time corrections, MOAT (1960) give 1.546 with 75 keV neutrons on U <sup>235</sup> . Amended here to thermal energy and to include delayed neutrons
COLVIN & SOWERBY (1959)	1.025 ± 0.006	1.191 ± 0.007	1.163 ± 0.008		1 1 1	Coincidence method, with boron-pile neutron counter. Results corrected to include delayed neutrons
SOWERBY (1959)				1.511 ± 0.011	0	A tentative result from boron-pile measurements, believed to contain errors resulting from high fission rate
Weighted mean values	1.025 + 0.006	1.194 ± 0.008	1.163 ± 0.009	1.551 ± 0.021		

### 4.8 THE NEUTRON REGENERATION FACTOR, ETA

### 4.81 Introduction

We define  $\eta$  as the average number of primary fission neutrons emitted per neutron absorbed in the fissile material;

$$\eta = \bar{\nu} \sigma_{\rm F} / \sigma_{\rm A}, \qquad (4.10)$$

where prompt and delayed neutrons are included.

The only direct absolute measurements of  $\eta$  are those recently reported by MACKLIN et al. (1960), but considerable number of indirect determinations have been attempted.

#### 4.82 Direct measurements

 $\eta$  is a dimensionless parameter so that, in principle, it can be determined by comparative measurements. With an incident beam of slow neutrons:

- i) The sample can be made thick enough to absorb almost the whole incident flux, yet without serious self-absorption and multiplication of the fast neutrons produced.
- Only fission neutrons are evolved, and they can be readily distinguished from the bombarding neutrons.

So, apart from corrections,

$$\eta = \frac{No. of neutrons emitted per unit time}{No. of neutrons incident per unit time}$$

The numerator and denominator of this expression can be compared directly if the neutron counting system has a response which is insensitive to variations of neutron energy from the thermal region to several MeV. For example, MACKLIN et al. (1960) measured the activation of manganese in a large water bath.

Relative values of  $\eta$  for different fissile nuclides can be measured by less exacting techniques, using separate counters to observe the incident thermal neutrons and the emitted fission neutrons. Measurements of this kind have been reported only by RICHMOND (1955), who used a wax-castle counter assembly to detect the fission neutrons. Unfortunately this kind of detector is rather sensitive to variations of neutron energy, so the results may be in error by about  $\pm$  0.3 percent because of small differences between the spectra of the fission neutrons from the different fissile nuclides.

By using thin fissile sample in a similar arrangement JAFFEY et al. (1955 and 1959) made comparative measurements of  $\eta \sigma_A$ , which may also be expressed as  $\overline{\nu}$  or. The same criticism applies.

### 4.83 Indirect measurements

Estimates of  $\eta$  may be derived from measurements of various kinds in critical and sub-critical assemblies. Many of the estimates are best regarded as test of the theoretical approximations used. However two classes of experiment merit closer

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attention and are discussed in the following sub-sections. Apart from small correction terms these measurements usually determine relative or absolute values of the parameter.

$$(\eta - 1) \sigma_{\Lambda} = x$$
, sey.

Although  $\eta$  is only determined indirectly from this expression it is important that, because  $\eta \approx 2$ , uncertainties of 1% in x or in  $\sigma_A$  yield only about  $\frac{1}{2}$ % uncertainty in  $\eta_{\bullet}$ 

#### REACTIVITY EXPERIMENTS

Many estimates of  $\eta$  have been derived from reactivity perturbation experiments, from pile period determinations, control-rod movement, or by oscillator techniques. The measurements are comparative, determining

$$\hat{X} = \frac{\int \phi(E) \{W_{F^*}, \eta(E) - W(E)\} \sigma_A(E), dE}{\int \phi(E) W(E), \sigma_o(E), dE}$$
(4.11)

In this expression:

- $\phi(E)$  is the distribution of the neutron flux
- W(E) is the worth or importance of a neutron of energy E at the sample position relative to that of a thermal neutron.
- $W_F$  is the average worth of a fission neutron, at the sample position.
- $\sigma_{\rho}$  is the absorption cross-section of a non-fissile absorber used as a standard for calibration of the reactivity scale.

The other symbols are defined in sub-section 4.11. Usually nearly all the neutron absorption in the samples occurs below about 1 eV, and W(E) is not expected to vary significantly from unity in this energy region, so that equation (4.11) can be written

$$\hat{\mathbf{X}} = (\mathbf{W}_{\mathbf{p}}\hat{\boldsymbol{\eta}} - 1) \hat{\boldsymbol{\sigma}}_{\mathbf{A}}/\hat{\boldsymbol{\sigma}}_{\boldsymbol{\rho}}$$
(4.12)

Correction to 2200 m/sec gives

$$X_{o} = (W_{F}\eta_{o} - 1) \sigma_{A}^{o} \sigma_{p}^{o} \qquad (4.13)$$

$$= \frac{G_{\rho}}{G_{\rm F}} \hat{\mathbf{X}} + \left(\frac{G_{\rm A}}{G_{\rm F}} - 1\right) \frac{\sigma_{\rm A}^{0}}{\sigma_{\rho}^{0}} \tag{4.14}$$

where G denoted the g+rs of Westcott's symbolism: WESTCOTT (1958, 1960). In equation (4.14) the major uncertainty is that arising from the first term on the right hand side; the second is only a small correction term.

W<sub>F</sub> varies from one location to another, but is usually about unity in thermal reactor systems. It differs from unity because of the following possibilities:

- a) Fast neutron multiplication in the fuel or in the sample.
- b) Neutron migration to positions of higher or lower worth during moderation. This includes the possibility of leakage from the assembly.

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- c) Resonance capture during moderation.
- d) Differences of detector efficiency for neutrons of different energies. Usually there is enough moderator between sample and monitor that this effect may be ignored.

We should mention also that in equation (4.11) it has been implicitly assumed that the neutron importance at the sample position is sufficiently <u>isotropic</u> that sample scattering effects may be ignored.

If  $W_F$  can be estimated from theoretical considerations  $\eta_0$  can be evaluated absolutely, and many authors have made use of this possibility. However no detailed investigation has been reported of the accuracy of such theoretical estimates of  $W_F$ . We prefer to take the view that in general the reactivity measurements are <u>primarily</u> measurements of  $W_F$ . However, if several different fissile nuclides are compared, the relative values of  $(\eta_0 - 1)\sigma_0$  can be determined also. From equation (4.13)

$$(\tau_{0} - 1)\sigma_{A}^{\circ} = \frac{\sigma_{\rho}^{\circ}}{W_{F}} \left[ X_{o} + (1 - W_{F}) \frac{\sigma_{A}^{\circ}}{\sigma_{\rho}^{\circ}} \right]$$
(4.15)

On taking ratios the factor  $\sigma_{\rho} \circ / W_F$  drops out, and in the correction term small errors in  $W_F$  and in  $\sigma_{\rho} \circ$  cancel in part and may be ignored.  $W_F$  can be evaluated well enough by using equations (4.13) and (4.14), together with the preliminary data from Table 4.17.

### TABLE 4.17

Preliminary data used for approximate evaluation of Wp and

Parameter	Value assumed, at 2200 m/sec	Origin		
$\sigma_{A}[U^{238}]$ $\sigma_{A}[U^{235}]$ $\sigma_{A}[Pu^{238}]$	575 ± 3 barns 681 ± 4 barns 1024 ± 8 barns	Weighted mean values from Tables 4.6, 4.7 and 4.8		
η[U <sup>233</sup> ] η[ <b>U<sup>235</sup>]</b> σ <sub>A</sub> [B]	2.296 <u>+</u> 0.010 2.077 <u>+</u> 0.010 See Table 4.1	MACKLIN et al. (1960)		

of small correction terms in equations (4.14) and (4.15)

GWIN & MAGNUSON (1960a) made reactivity measurements with small samples at the centre of a large internal thermal column ("flux trap") of  $H_2O$ . In this assembly  $W_F$  had an unusually large value, about 2.3 apparently. Therefore  $W_F\eta >> 1$ , and instead of equation (4.15) it is more appropriate to write

$$\vec{v} \sigma_{\mathbf{F}}^{\circ} = \frac{\sigma_{\rho}^{\circ}}{W_{\mathbf{F}}} \left\{ \frac{G_{\rho}}{W_{\mathbf{F}}} \hat{\mathbf{x}} + \frac{G_{A} \sigma_{A}^{\circ}}{G_{\mathbf{F}} \sigma_{\rho}^{\circ}} \right\}$$
(4.16)

On taking ratios the factor  $\sigma_{\rho}^{\circ}/W_{\rm F}$  drops out. The second term in the braces is relatively small and may be estimated from the preliminary data in Table 4.17 without adding seriously to the uncertainties.

#### CRITICAL EXPERIMENTS IN SIMPLE GEOMETRIES

A very detailed series of experiments, with homogeneous aqueous solutions of fissile materials in large spherical and cylindrical containers, have been made at the Oak Ridge national laboratory, THOMAS et al. (1956)\*, MAGNUSON & GWIN (1959), and GWIN & MAGNUSON (1960b).

For criticality with fissile material of high isotopic purity

$$(\hat{\eta} f q) = 1$$
 (4.17)

averaging over the neutron spectrum throughout the system. In this expression q is the non-leakage probability, which is close to unity for a large system; f is the neutron absorption rate, in the fissile material as a fraction of the total absorption rate in the medium, and it is calculated from the observed critical dilution and the absorption cross-sections of the constituent atoms.

From equation (4.17) it can be seen that the measurements determine

$$(q \hat{\eta} - 1)\hat{\sigma}_{A}/\hat{\sigma}_{\rho} = R \qquad (4.18)$$

where  $\hat{\sigma}_A$  is the absorption cross-section of the fissile material, R is the ratio of diluent atoms to fissile atoms, and  $\hat{\sigma}_\rho$  is the average absorption cross-section of the diluent atoms. The spectrum averages involved are similar to those of the preceding sub-section, and equation (4.18) is entirely similar to (4.12). Since qc1 the measurements permit the valuation of

$$(\eta_0 - 1)\sigma_A^{0}$$
 (4.19)

exactly as in equation (4.15)

The experimental study of homogeneous systems of simple geometric configurations should stimulate relatively detailed mathematical studies. The use of large well-moderated systems reduces the uncertainties in determining q and in the neutron spectrum. The use of dilute fissile material of high isotopic purity simplifies the calculation of absorption effects during moderation. Thus the measurements permit the absolute evaluation of  $(\eta_0 - 1)\sigma_A^{\circ}$  with good accuracy.

\*The data of THOMAS et al. (1956) have been revised by FRANCIS et al. (1957) and further revisions are cited by MAGNUSON & GWIN (1959).

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### 4.84 Correction of experimental data to 2200 m/sec

Absolute measurements of eta, and comparative measurements between different fissile nuclides, have not yet been made with monokinetic bombarding neutrons. Only broad neutron spectra have been used, so that corrections are needed to derive data for neutrons of 2200 m/sec.

Thick sample measurements. In the direct measurements of eta the whole incident neutron flux is absorbed in the thick sample, so the measurements give

$$\bar{\eta} = \phi(E) \cdot \eta(E) \cdot dE / \phi(E) \cdot dE$$
(4.20)

Notice that the flux is not weighted by an absorption cross-section, so that to obtain a measurement of n in the low energy region it is essential to curtail the epithermal component of the neutron spectrum. RICHEMOND (1955) and MACKLIN et al. (1960 used Cd-difference techniques.

The latter authors made a rather detailed study of all the corrections needed for their measurements of  $\eta$  and of the uncertainties involved; corrections to 2200 m/sec. were included. RICHMOND (1955) corrected his results for the effects of the rump of the epithermal spectrum below the Cd cut-off, so that his data refer to a Maxwellian neutron spectrum at a temperature of perhaps  $(70 \pm 40)^{\circ}$ C. To transform them to 2200 m/sec we have assumed

	U <sup>233</sup>	U <sup>235</sup>	Pu <sup>239</sup>
n/no	1.000 <u>+</u> (0.6%)	0.995 <u>+</u> (0.4%)	0.958 <u>+</u> (0.8%)

Thin sample measurements. The other measurements have all been made with relatively thin or dilute samples. We have converted the results to 2200 m/sec by using the formalism and tables of WESTCOTT (1960), and making allowances for the uncertainties of the neutron spectra and of the g and s coefficients. The conversion is based on equation (4.10), and the fact that  $\overline{\nu}$  is virtually independent of the bombarding spectrum in a thermal reactor. For examples, see equations (4.14), (4.15) and (4.16). It is worthwhile to comment that for the reactivity experiments this treatment is only approximately correct. To bring about a measurable change of reactivity one must use a macroscopic amount of the absorbing material, so that small spectral distortion and self-shielding effects are inevitable. However these effects are no doubt much reduced by using samples in the form of dilute solutions in D<sub>2</sub>O, and in comparative measurements the effects must balance to some extent if the samples are reasonably well-matched. See for comparison the discussion in § 4.42.

### 4.85 Experimental data for eta

The experimental methods outlined above, and various modifications of them have been used to obtain values of

$$\eta$$
,  $(\eta - 1)\sigma_A$ ,  $\eta\sigma_A$ 

The experimental data are collected in Tables 4.18, 4.19, 4.20 and 4.21. For the most part the tables should be self-explanatory. Table 4.20 can be most easily understood by referring to sub-section 4.83. The measured values of

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 $(W_{F\eta-1})\sigma_A\sigma_p = 1$  are given in the third column. These results were transformed to 2200 m/sec by using equation (4.14) and WESTCOTT's (1960) tables, with reasonable guesses at the neutron spectrum. Thence WF was evaluated sufficiently accurately, by using the preliminary data of Table 4.17. The values of WF obtained in this way are given in column 4, and the values which were used in the references are given there also in brackets, for comparison. Finally relative values of  $(\eta_0 - 1)\sigma_A^{\circ}$  were calculated by using equation (4.15) and are given in columns 5 and 6 of Table 4.20.

In weighting the data we have preferred to reject those derived from measurements in heterogeneous lattices, both on general grounds and because the neutron spectra involved are necessarily very uncertain.

To put the data on a consistent basis, to correct to 2200 m/sec, and to give each result in the most appropriate form, it has been necessary to recalculate the majority of the results. In some instances we were unable to reproduce exactly the original values from the data reported. A variety of techniques has been used for reactivity perturbation measurements and in the following notes we draw attention to some of the special features of the different measurements, and to some of the uncertainties incurred.

- The measurements of SPIVAK & YEROZOLIMSKY (1956) were similar to typical reactivity A. pertubation measurements, see § 4.83, but were made in a large graphite thermal column. Care was taken to obviate the effects of neutron scattering by the samples. The neutron absorption rates in the samples were compared by a broad beam transmission experiment, eliminating the need for sample assay. This latter experiment is triefly discussed in § 4.43, note B. Corrections are needed because the detector response was not independent of neutron energy, and because the incident neutron spectrum may have been affected by the proximity of the noderator surface and of a cadmium disc. We have only been able to make rather crude estimates of these corrections. The weight factor WF was determined by "poisoning" the graphite. The result depends on a somewhat uncertain extrapolation and we have preferred to treat the measurements as yielding only relative values of  $\eta_{\bullet}$  We could not reproduce exactly the reported value for  $\eta[U^{235}]$  from the data in the reference. Unfortunately the absorption cross-sections derived from the transmission experiment are not very plausible, see Table 4.5, and greatly reduce confidence in the results for n.
- B. McMILLAN et al. (1955), later reference GAERITINER et al. (1958), made an extensive series of measurements, including reactivity perturbations with novel cadmium difference techniques. Additional reactivity and activation measurements were made to analyse the neutron spectrum. Several fission parameters may be evaluated more or less independently from the observations. The results suggest rather strongly that introduction of the cadmium caused a significant change in the reactivity scale. This is not surprising since the reactor used is small and highly inhomogeneous. We have recalculated the results using the standards from Table 4.1 and subsidiary data from Table 4.17, and have transformed them to 2200 m/sec. However we give no weight to these results because of the uncertainties of interpretation mentioned above.
- C. An extensive series of measurements was reported by ZINN & KANNER (1945). Reactivity measurements were made in the CP2 reactor using a pyrex standard of unspecified composition. It was calibrated by measurements with neutron spectrometers, but the calibration is unreliable and should be ignored. Transmission experiments with a thermal neutron beam relate the pyrex standard to  $\sigma_{A}[U^{233}]$ ,

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but similar measurements with U<sup>235</sup> cannot be used until the correct "hardening factor" is computed for this non-1/v absorber. The residual data may be interpreted as in Table 4.20. The samples were not withdrawn to a position of zero flux gradient in the swing measurements, so very small scattering corrections should be made.

- D. CRUIKSHANK et al. (1948) have given a detailed account of a series of pile oscillator experiments with  $U^{233}$ , which we interpret as a measurement of  $W_{\rm F}$ , only. Sample 6 appears to have leaked during the course of the work, and for an unbiased evaluation of the date we felt it necessary to assign a large uncertainty to the assay of this sample. In recalculation we also used a revised value of (2.105  $\pm$ 0.012) x 10<sup>7</sup> disints/min. mg. for the decay rate of  $U^{233}$ .
- E. MUEHLHAUSE (1952 and 1959) and HARRIS & ROSE (1953) made pile oscillator measurements in the D<sub>2</sub>O moderated reactor CP3: The reactor was large, with highly enriched fuel and, as HUGHES (1956) has remarked, these factors permitted an unusually satisfactory theoretical estimate for W<sub>F</sub>. Comparison with the other data in Table 4.20, columns 5 and 6, suggest that the reactivity of U<sup>235</sup> has been overestimated by  $(4 \pm 2)\%$  from MUEHLHAUSE's measurements.
- F. ALICHANOV et al. (1956) made reactivity measurements at the centre of a D<sub>2</sub>O moderated reactor, both with completed lattice, and with fuel elements removed from the central zone to make a thermal well of 70 cm diameter. Dilute samples were used in D<sub>2</sub>O solutions. There was a slight uncertainty in the control rod calibration for the larger reactivity perturbations. It was felt that more accurate results might be obtained by first measuring the small effect of a sample of U<sup>233</sup> with boron. Then the U<sup>235</sup> and Pu<sup>239</sup> samples were compared with a pure U<sup>233</sup> sample of about the same reactivity.

We were unable to reproduce the reported results from the reactivity data in the reference. Consequently our recalculations have a somewhat uncertain basis, but fortunately this has not seriously augmented the uncertainties of the derived ratios of  $(\eta_0 - 1)\sigma_A^o$  given in Table 4.20, columns 5 and 6.

G. An extensive series of measurements with pile oscillators in DIMPLE and GLEEP have been reported by CABELL et al. (1960). Several of the samples were analysed both at Harwell and at Oak Ridge, and there was good agreement as to their assay and isotopic composition. However there remains an unexplained systematic discrepancy between the reactivities of  $U^{233}$  samples of American and British origins. The American material appears to be  $(3.3 \pm 0.8)$  percent more reactive. The data given in Table 4.20 are the mean values taking all samples into account. Because the oscillation periods were short some of the dealyed neutrons were not at saturation intensity throughout the pulse. The data in Table 4.20 have been corrected for this effect.

	Ab	solute value	Relativ	e values				
Reference	U <sup>233</sup>	U <sup>235</sup>	Pu <sup>239</sup>	U <sup>233</sup> /U <sup>235</sup>	Pu239/U235	Weight	Method and comments	
ZINN & KANNER (1945)				1.114 ± 0.006		o	Absorption of thermal neutrons in "grey" samples; 54% transmission. The analysis given is extremely crude and many correc- tions ought to be made	
RICHWOND (1955)				1.102 ± 0.021	1.006 ± 0.023	0.2	Absorption of reactor neutrons in thick samples, with Cd-differences. Fission neutrons detected with wax-castle counter. Original data 1.108, 0.969, corrected to 2200 m/sec. See sections 4.82 and 4.84	
SPIVAK & YEROZOLIMSKY (1955)				1.101 ± 0.015	0.986 ± 0.028	<b>0</b>	Neutron yield in graphite thermal column compared with absorption by boron. We have corrected results to 2200 m/sec. See section 4.85 note A	
McMILIAN et al. (1955) CAERTINEE et al. (1958)				1.067 ± 0.049	0.969 <u>+</u> 0.036	0 0	Reactivity measurements with and without a Cd sandwich. Results recalculated using standard from Table 4.1, but derivation is extremely complex See section 4.85 note B	
MACKLIN et al. (1960)	2.296 <u>+</u> 0.010	2.077 ± 0.010	1.105 ± 0.005	->		1 1 1 1	Direct measurement with thick samples, by activation of Mn in water bath. Incident neutrons from D <sub>2</sub> O tank next to reactor core, with Cd-differences. The 3 data are not independent. See sections 4.82 and 4.84	
. Weighted mean values	2.296 ± 0.012	2.077 <u>+</u> 0.012		1.104 ± 0.0078	1.006 <u>+</u> 0.023	We hav the th treat	e enlarged the uncertainties assigned to ree data of Macklin et al. so that we may them as independent	

### Table 4.18

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Eta for U233, U235 and Pu239, with incident neutrons of 2200 m/sec

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TABLE 4.19

Absolute values of  $(\eta - 1)\sigma_A$  for U<sup>233</sup> and U<sup>235</sup> with incident neutrons of 2200 m/sec

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Reference	(n-1) o <sub>A</sub> at U <sup>233</sup>	at 2200 m/sec Values U <sup>235</sup> Spect compon		Method and comments
GWIN & MAGNUSON (1960 b)	737.6 <u>+</u> 8.0	731.4 <u>+</u> 10.2	739 ± 7 711•4 ± 9•3	Criticality measurements with dilute homogeneous aqueous solutions of fissile material in spheres and cylinders. We have recalculated the results with revised standards, using for $\eta$ only the data from systems with small neutron leakage. See section 4.83

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### Table 4.20

Ratios of  $(\gamma - 1) \sigma_A$  for U233, U235 and Pu239 with incident neutrons of 2200 m/sec.

1

10

1	2	3	4	5	6	7	В
Reference	ce $\frac{\text{Pissile}}{\text{nuclide}} \begin{pmatrix} \text{Observed} \\ (W_F \eta - I) \hat{\theta}_{\lambda} \hat{\phi}_{\rho} \\ \text{in broad} \\ \text{in broad} \\ \text{Spectrum} \end{pmatrix} \begin{pmatrix} W_F \\ \text{from} \\ \text{experiments} \\ (W_F, \text{theor.}) \end{pmatrix} = \frac{\text{Relative values of } (\eta - I) \hat{\theta}_{\lambda}}{235} \frac{1}{9} \frac{1}{2} \frac{1}{$		$Pu^{239}/U^{235}$	Weight	Lethod and comments		
FERMI et al. (1944)	U235	?					Reactivity measurement with boron as standard. Too few details are available
ZINN & KANNER (1945)	0233 · 0235	0,983 0.860	0.867 ± 0.008 (0.851)	1.007 <u>+</u> 0.021		0	Resctivity measurements in CP2, with $r_0 = 0.050$ . Based on $O_A[U233]$ as standard. See section 4.85 note C
-ditto-	U233 U235	0.815 0.734	0.790	1.010 <u>+</u> 0.030		0	Reactivity measurements in CP2 in a sub-Cd spectrum. Based on $\mathcal{O}_{A}[U233]$ as standard
CRUIKSHANK et al. (1948)	U233	0.834 <u>+</u> 0.063	0.892 <u>+</u> 0.035 (0.829)				Pile cscillator measurements in ZEEP relative to boron of unspecified origin. Assay of U233 by absolute alpha counting. Recalculated: see section 4.85 note D
MUEHLHAUSE (1952 and 1959)	U233 U235 Pu239	0.842 ± 0.010 0.791 ± 0.010 1.252 ± 0.016	0.914 ± 0.013 (0.915 ± 0.007)	0.977 <u>+</u> 0.019	1.408 <u>+</u> 0.048	0	Pile oscillator measurements in CP3 relative to ANL standard boron. Results recalculated assuming r = 0.024 ± 0.01, from Cd-ratio data. See section 4.85 note E
HARPIS & ROSE (1953)	Pu239	ş					Continuation of work of MUEHLHAUSE (1952) above. Too few details available
ALICHANOV et al. (1955)	U233 U235 Pu239	1.070 1.028 1.602	$1.049 \pm 0.016$ (1.013 $\pm 0.005$ )	1.014 ± 0.018	1.456 <u>+</u> 0.049	0.5 0.3	Reactivity measurements in a thermal well at centre of a D20 moderated reactor, with boron as standard. See 4.85 note F
-ditto-	U233 U235 Pu239	0.760 0.668 1.083	0.856 <u>+</u> 0.013 (0.818 <u>+</u> 0.004)	1.031 ± 0.026	1.486 ± 0.062	0	As above, but in spectrum obtained by completing the lattice

continues next page

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### Table 4.20, continued

1	2	3	4	5	6	7	8
Reference	Fissile nuclide	Observed (WFN-I) OA/Op in broad spectrum	W <sub>F</sub> from experiments (W <sub>F</sub> , theor.)	$\frac{\text{Relative valu}}{\text{at } 2200 \text{ m/s}}$ $\frac{U^{233}}{U^{235}}$	es of Pu <sup>239</sup> /U <sup>235</sup>	Weight	Method and comments
McMILLAN et al. (1955) GAERTINER et al. (1958)	U233 U235 Pu239	0.751 ± 0.010 0.704 ± 0.009 1.195 ± 0.016	0.868	1.004 <u>+</u> 0.022	1.521 <u>+</u> 0.036	0 0	Reactivity measurements in TTR with Cd-differences, using boron as standard Results revised and recalculated. See section 4.85 note B
McMILLAN et al. (1955)	U233 U235 Pu239	0.826 ± 0.007 0.719 ± 0.008 1.235 ± 0.014	0.881	1.010 <u>+</u> 0.016	1.499 <u>+</u> 0.027	0 0	As above, but derived from data in complete spectrum, with/Od-difference
CABELL et al. (1960)	U233 U235 Pu239	1.103 ± 0.011 1.065 ± 0.012 1.687 ± 0.023	1.075 <u>+</u> 1.010 (1.02 <u>+</u> 0.06)*	1.017 <u>+</u> 0.017	1.499 <u>+</u> 0.030	1	Pile oscillator measurements in a thermal well in DILPLE, relative to boron. Neutron spectrum assumed Maxwellian at tempersture of moder- ator, 20°C. In recalculation the data in column 3 were corrected for loss of some delayed neutrons. See section 4.851 note G
- ditto -	U233 U235 Pu239	0.828 ± 0.011 0.720 ± 0.008 1.240 ± 0.039	0.890 <u>+</u> 0.009 (0.877) <sup>*</sup>	1.019 <u>+</u> 0.019	1.467 <u>+</u> 0.042	0 0	As above, but measured in reactor spectrum: $T = 66^{\circ}C$ , $r \approx 0.042$
GWIN & MAGNUSCN (1960b)	U233 U235	various various		1.009 ± 0.015		0	Criticality experiments with dilute homogeneous aqueous solutions of fissile material in spheres and cylin- ders. Cnly preliminary data are available. We have revised the standards and corrected to 2200 m/sec but the ratio here is not independent of absolute values given in table 4.21
	W	eighted mean va	lues	1.016 ± 0.015	1.489 ± 0.033		

Ratios of  $(\eta - I) \mathcal{O}_A$  for U233, U235 and Pu239 with incident neutrons of 2200 m/sec.

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### TABLE 4.21

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## Ratios of $\eta \sigma_A (= \overline{\gamma} \sigma_F)$ for U233, U235 and Pu239 with incident neutrons of 2200 m/sec

C.Fr

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	Reference	$\eta \sigma_A \text{ ratios}$ $U^{2,33}/U^{2,35}$	Pu <sup>23</sup> 9/U <sup>235</sup>	Value in experimental spectrum	Weight	Method and comments		
	ANDERSON & MAY (1944)	ANDERSON & MAY (1944) 0.950 ± 0.016		0.979 ± 0.015	0	Measured in thermal column of CP3 reactor. Fission neutrons detected with In foils in Cd sheaths. U233 assayed by alpha-counting. Original value corrected assuming 21050 disints./min. pg of U233		
	ANDERSON & NAGLE (1944)		?			No details available. Possibly similar to the measurement by ANDERSON & MAY above		
-47-	NcLILLAN et al. (1955) GAERTINER et al. (1958)	0.935 <u>+</u> 0.029	1.617 ± 0.052	$0.964 \pm 0.029$ $1.895 \pm 0.059$	0	Reactivity measurements with thin samples placed inside or outside a Cd box. Fecalculated with revised standards. We could not reproduce exactly the original results from the data reported. See section 4-85, note B		
	JAFFEY et al. (1955 and 1959)	0.914 <u>+</u> 0.019	1.501 ± 0.032	0.942 <u>+</u> 0.018 1.670 <u>+</u> 0.015	1 0.6	Thin samples irradiated in a pile neutron beam with Cd- differences. Fission neutrons detected with annular boron counter embedded in paraffin wax and shielded with Cd. Moderator temperature 35°C. Recalculated		
	GWIN & MAGNUSON (1960a)	0.948 <u>+</u> 0.019	1.510 ± 0.025		1	Reactivity measurements in internal thermal column, with $r_{0}\approx0.015$ and moderator temperature 25°C. Corrected to 2200 m/sec, with revised standards. Results are preliminary. See section 4.83		
	Weighted mean values	0.931 <u>+</u> 0.022	1.506 <u>+</u> 0.024					

### TABLE 4.22

Recommended values	for	reaction	parameters o	f U233, U235
and Pu239	for	neutrons	of 2200 m/se	C

Item	Source of input datum. Table No.	Paramet for neutr 2200 m/	er, mons of sec.	Input datum	Recommended value
1 2 3	4.6 4.7 4.8	$\sigma_{A}$ , barns	U233 U235 Pu239	575 ± 3.1 680.6 ± 3.6 1024 ± 8.1	573.7 ± 2.5 680.5 ± 2.9 1026.7 ± 7.5
456	4.9	$\sigma_{\rm F, \ barns}$	U233 U235 Pu239	514•4± 7•9 587 ± 5•0 707 ± 18	$524_{+}.5 \pm 2.7$ $579.9 \pm 2.7$ $740.6 \pm 5.5$
7 8 9	4.10	$\sigma_{\rm F}$ ratios	U233/U235 Pu239/I235 Pu239/U233	0.9107 ± 0.0077 1.301 ± 0.016 1.431 ± 0.017	$\begin{array}{r} 0.9045 \pm 0.0045 \\ 1.277 \pm 0.009 \\ 1.412 \pm 0.010 \end{array}$
10 11 12	4.11 4.12 4.13	$\alpha = \sigma_{\rm y}/\sigma_{\rm F}$	U233 U235 Pu239	0.0958 ± 0.006 0.1714 ± 0.007 0.305 ± 0.10	$\begin{array}{c} 0.0938 \pm 0.0047 \\ 0.1734 \pm 0.0050 \\ 0.386 \pm 0.013 \end{array}$
13 14 15	4.15	♥, neuts./f	iss U233 U235 Pu239	2.438 ± 0.020	$\begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$
16 17 18	4.16	<b>v</b> ratios	U233/U235 Pu239/U235 Pu239/U233	1.025 ± 0.006 1.194 ± 0.008 1.163 ± 0.009	$\begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$
19 20 21	4.18	η	U233 U235 Pu239	$2.296 \pm 0.012 \\ 2.077 \pm 0.012 \\ -$	$\begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$
22 23 24	4.18	$\eta$ ratios	U233/U235 Pu239/U235 Pu239/U233	$\begin{array}{r} 1.104 \pm 0.0078 \\ 1.006 \pm 0.023 \end{array}$	$\begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$
25 26 27	4.19	(η-1)0 <sub>A</sub> , bar	ns U233 U235 Pu239	737.6± 8.0 731.4± 10.2	$\begin{array}{rrrr} 74.0.1 & \pm 4.6 \\ 733.4 & \pm 5.0 \\ 1123 & \pm 16 \end{array}$
28 29 30	4.20	(n-1) $\sigma_A$ ratios	U233/U235 Pu239/U235 Pu239/U233	1.016 ± 0.015 1.489 ± 0.033	$\begin{array}{rrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrrr$
31 32 33	4.21	ησ <sub>A</sub> =Jo <sub>F</sub> ratios	U233/U235 Pu239/U235 Pu239/U233	$\begin{array}{r} 0.931 \pm 0.022 \\ 1.506 \pm 0.024 \end{array}$	0.929 ± 0.005 1.520 ± 0.010 1.635 ± 0.01°
		σ <sub>S</sub> , barns		Sec § 4.3 ar	nd Table 4.4

### 4.9 RECOMMENDED VALUES

In sections 4.3 to 4.8 we have collected and discussed the many experimental measurements of the reaction parameters of  $U^{\pm 33}$ ,  $U^{\pm 35}$  and  $Pu^{\pm 39}$ , with neutrons of 2200 m/sec. No fewer than 26 weighted mean values are given from the absolute and relative measurements of

$$\sigma_{\Lambda}$$
,  $\sigma_{\rm F}$ ,  $\alpha$ ,  $\overline{\nu}$ ,  $\eta$ ,  $(\eta - 1)\sigma_{\Lambda}$ , and  $\eta \sigma_{\Lambda}$ 

for the three nuclides. However only 9 of the parameters are independent, so that a set of consistent values must now be derived. In addition to the obvious relationship between the parameters the equations

$$\sigma_{A} = (1+\alpha)\sigma_{F}$$
$$\bar{\nu} = (1+\alpha)\eta$$

must also be satisfied.

The 26 input data and the recommended values obtained from them by a leastsquares adjustment are given in Table 4.22. The analysis was done by FOSSEY (1961). Recommended values for the scattering cross-sections are given in Table 4.4 on page 16.

A  $\chi^2$  test of the data shows that the deviations of the input data from the recommended values have 55 percent probability. The differences between the input data and the recommended values all lie within the uncertainties assigned. In relation to the uncertainties the largest shifts are for

$$\sigma_{\rm F}[{\rm Pu}^{238}], \sigma_{\rm F}[{\rm Pu}^{238}]/\sigma_{\rm F}[{\rm U}^{238}], \sigma_{\rm F}[{\rm U}^{238}], \sigma_{\rm F}[{\rm U}^{238}]$$
  
 $(\eta - 1)\sigma_{\rm A}[{\rm Pu}^{238}]/(\eta - 1)\sigma_{\rm A}[{\rm U}^{238}]$ 

and

#### 4.10 RECOMMENDATIONS FOR FURTHER WORK

In attempting to assess the reliability of the "recommended values" which are given in Table 4.22 the following features appear to be the most questionable and to require further study experimentally.

- 1) Many of the measurements were made with thermal neutron spectra, and have been corrected to 2200 m/sec. In particular, all the measurements relating to eta were made with thermal neutrons, or with reactor neutrons. Better data are needed on the variation of  $\eta$  with energy in the range 0.005 to 0.5 eV, for U<sup>23</sup>, and U<sup>235</sup>.
- 2) Very high accuracy has been claimed for the direct absolute measurements of  $\eta_{i}$  although only a single pair of measurements has been reported; MACKLIN et al. (1960). Better absolute data on  $\overline{\nu}$  and on  $\alpha$  would help to confirm their results.
- 3) The very extensive and important series of measurements by CABELL et al. (1960), see Table 4.20 and sub-section 4,85 Note G, appear to show a difference of about 3% in the reactivities of U<sup>223</sup> of American and British origins. The resolution of this improbable result is very desirable.
- 4) Most of the measurements of the capture to fission ratios, α, were made with ramples which had been irradiated in reactor lattice spectra. In general we have given no weight to measurements in lattice spectra, giving preference to data obtained with monokinetic neutrons or with thermal neutrons. However it was not practicable to maintain this policy towards the data for α. New measurements with well-moderated neutrons are needed. This recommendation re-emphasises the need for reliable data on the energy dependence of η.

To improve the precision of the results attention should be given also to :-

5) Better measurements of the parameters

 $\sigma_A$ ,  $\alpha$ ,  $\sigma_F$ , and  $\eta$ 

for Pu 239. For on and n improved comparisons with U 233 or U 235 would be helpful.

6) Better measurements of the fission cross-sections generally.

7) New measurements of og in the range 0.01 to 0.5 eV for all three nuclides.

#### References

ABOV Yu. G. (1955) Conf. Acad. Sci. USSR on the peaceful uses of atomic energy. U.S. Consultants Bureau translation; division of phys. and math. sciences, p. 209. ALICHANOV A.J., VLADIMIRSKI V.V. & NIKITIN S.J. (1956) Proc. Geneve I, 4, 303. ALLEN W.D. & FERGUSON A.T.G. (1957) Proc. Phys. Soc. 70A, 573. ANDERSON H.L. (1944) CF 2161 unpublished. ANDERSON E.E., LAVATELLI L.S., MeDANIEL B.D. & SUTTON R.D. (1944) unpublished. Quoted by HALVEY & HUGHES (1953) and by SAILOR (1957). ANDERSON H.L. & MAY A.N. (1944) TID 5223 part 1, paper 2.6, (1952). ANDERSON H.L. & NAGLE D.E. (1944) unpublished. Cited by HARVEY & SANDERS (1956). ANDERSON E.E., MODANIEL B.D. & SUTTON R.B. (1945) LA 266. AUCLAIR J.M., LANDON H.H. & JACOB M. (1955) Comptes rendus 241, 1935. AUCLAIR J.M., GALULA M., HUBERT P., JACROT B., JOLY R., NETTER F. & VENDRYES G. (1956) Proc. Geneva I, 4, 235. BAILEY C., BLAIR J., & RUSSELL H. (1944) unpublished. See MAY (1944) and ANDERSON & MAY (1944). BARLOUTAUD R. & LEVEQUE A. (1952) J. Phys. Rad. 13, 412. BIGHAM C.B., HANNA G.C. & TUNNICLIFFE P.R. (1959) ALCL 924. BIGHAM C.B., HA MA G.C., TUNNICLIFFE P.R., CAMPICN P.J., LOUNSBURY M. & MACKENZIE D.R. (1958) Proc. Geneva II, 16, 125. BISWAS S. & PATRO A.F. (1949) Indian J. Phys. 23, 97: BLOCK R.C., SLAUGHTER G.G. & HARVEY J.A. (1960) ORNL 2910, 41. BOLLINGER L.M. (1957) unpublished. Quoted by SAFFORD & HAVENS (1959). BOLLINGER L.M., COTE R.E. & THOMAS G.E. (1958) Proc. Geneva II, 15, 127. BONDARENKO I.I., KUZMINOV B.D., KUTSAYEVA L.S., PROKHOROVA L.I. & SMIRKNKIN G.N. (1958) Proc. Geneva II, 15, 353. BONNER T.W. (1959) WASH 1026. BOWMAN H.R. & THOMPSON S.G. (1958) UCRL 5038 revised. BURGOV N.A. (1955) unpublished. Quoted by ALICHANOV et al. (1956). See also BURGOV (1956) and NIKITIN et al. (1955).

BURGOV N.A. (1956) Proc. Geneva I, 4, 305.

- 51 -

CABELL M.J., ROSE H. & TATTERSALL R.B. (1960), unpublished.

CARTER R.S., PALEVSKY H., MYERS V.W. & HUGHES D.J. (1953) Phys. Rev. 92, 716.

CHAPMAN F.G. (1959), private communication.

COCKIN S.J. (1958) J. Nucl. En. 6, 285.

COCKROFT H.S. (1952) AERE N/R 890.

COHEN R., COTTON E. & LEVEQUE A. (1952a) Comptes rendus 234, 2355, and (1952b) Comptes rendus 235. 159.

COLVIN D.W. & SOWERBY M.G. (1958) Proc. Geneva II, 16, 121.

COLVIN D.W. & SOWERBY M.G. (1959) THCC (UK) 43.

COLVIN D.W. & SOWERBY M.G. (1960) EANDC (UK) 3, and private communications.

CCON J.H. (1955). Verbal communication recorded by LITTLER (1955).

CORNISH F.W. (1960) unpublished.

COX S., FIELDS P., FRIEDMAN A., SJOBLOM R. & SMITH A. (1958) Phys, Rev. 112, 960.

- CRAIG D.S., HANVIA G.C., HURST D.G., KUSHNERIUK S.A., LEWIS W.E. & WARD A.G. (1958) Proc. Geneva II, <u>16</u>, 83.
- CRANE W.W.T., HIGGINS G.H. & THOMPSON S.G. (1955) Phys. Rev. 97, 242.
- CRANE W.W.T., HIGGINS G.H. & BOWMAN H.R. (1956) Phys. Rev. 101, 1805.

CRANE W.W.T (1960). Private communication to MOAT (1960).

CRUIKSHANK A.J., LITTLER D.J. & WARD A.G. (1948) CRP 378.

DERUYTTER A.J. (1960a) Nucl. Instr. and Methods 7, 145, and (1960b) CEN-R.1914 (to be published in Reactor Science).

De SAUSSURE G. (1959a,b,c) ANL 6122, 256, 259, 261, respectively.

De SAUSSURE G. & SILVER E.G. (1959) Nucl.Sci.Eng. 5, 49.

DEUTSCH M. & LINENBERGER A. (1944) LA 100 unpublished. Cited by HARVEY & SANDERS (1956).

DEUTSCH M., KAHN M. and MISKEL J.A. (1946) LA 511. Some corrections are due to NEUMANN H. (1955), unpublished.

De WIRE J.W. (1944) unpublished.

De WIRE J.W., WILSON R.R. & WOODWARD W.M. (1944) unpublished. Quoted by HARVEY & SANDERS (1956).

DIVEN B.C., MARTIN H.C., TASCHEK R.F. & TERRELL J. (1956) Phys. Rev. 101, 1012.

- 52 -

EGELSTAFF P.A. (1954) J. Nucl. En. 1, 92.

EGELSTAFF P.A. (1956) NRDC 81.

EGELSTAFF P.A. (1957) unpublished.

EROZOLIMSKY B.G. & SPIVAK P.E. (1957) Atomnaya Energiya 2, 327; J. Nuc. En. 6, 243 (1958).

EVANS J.E. & FLUHARTY R.G. (1957) TID 7547, 98.

EVANS J.E. & FLUHARTY R.G. (1959) unpublished.

EVANS J.E. & FLUHARTY R.G. (1960), to be published in Nucl. Sci. Eng.

- FACCINI U. & GATTI E. (1950) Nuovo Gim. 7, 589. A correction, privately communicated, is cited by COHEN et al. (1952b).
- FERMI E., MARSHALL J. & MARSHALL L. (1943) CP <u>1186</u> unpublished. Quoted by HARVEY & HUGHES (1953), KATZIN (1954), SAILOR (1957), and others.
- FERMI E., ALIISON S.K. & COMPTON A.H. (1944) CP <u>1389</u> unpublished. Quoted by SAILOR (1957); see also MAY (1944).
- FOOTE H.L. (1958) Phys. Rev. 109, 1641.
- FOSSEY E.B. (1961), private communication.

FRANCIS N.C., HURWITZ H. & ZWEIFEL P.F. (1957) Nucl. Sci. Eng. 2, 253.

- FRIESEN W.J., LEONARD B.R. & SEPPI E.J. (1956) HW 47012, 50. See also PATTENDEN (1958).
- FURSOV V.S. (1955) Conf. Acad. Sci. USSR on the peaceful uses of atomic energy. U.S. Consultants Bureau translation; division of phys. and math. sciences p. 1.
- GAERTTNER E.R., JONES M.E., MCMILLAN D.E., SAMPSON J.B. & SNYDER T.M. (1958) Nucl. Sci. Eng. 3, 758. See also McMILLAN et al. (1955) for a more detailed account.

GEIGER K.W. (1960) Can. J. Phys. 38, 569.

- GERASIMOV V.F. (1956) Proc. Geneva I, 4, 287, verbal communications.
- GOLDSTEIN H. (1957) NDA 2-33, TNCC(US) 9.
- GREEN T.S., SMALL V.G. & GLANVILLE D.E. (1957) J. Nucl. En. 4, 409.
- GWIN R. & MAGNUSON D.W. (1960a) ORNL 60-4-11. See also MAGNUSON & GWIN (1958, 1959) and De SAUSSURE (1959a).
- GWIN R. & MAGNUSON D.W. (1960b) ORNL 60-4-12. See also MAGNUSON (1958), MAGNUSON & GWIN (1959) and De SAUSSURE (1959b and c).

- 53 -

HANNA G.C. (1960) AECL 873.

HARRIS S. & ROSE D. (1953) unpublished. Cited by MUEHLHAUSE (1959).

HARVEY J.A. & HUCHES D.J. (1953) BNL 221.

HARVEY J.A. (1956) Proc. Geneva I, 4, 147.

HARVEY J.A. & SANDERS J.E. (1956) Progr. Nucl. En., series I, vol. 1.

- HAVENS W.W., MELKONIAN E., RAINWATER L.J. & LEVIN M. (1951) CUD <u>92</u>. See also LEONARD (1956).
- HUGHES D.J. (1956) quoted in TNCC 2, 6 unpublished.

INGHRAM M.G., HESS D.C., HAYDEN R.J. & STEVENS C.M. (1956) Proc. Geneva, I, 4, 105.

JACOB M. (1958) CEA 652.

JAFFEY A.H., HIBDON C.T. & SJOBLOM R. (1955) ANL <u>5396</u>, and (1959) J. Nucl.En. (A) <u>11</u>, 21.

JOHNSTONE I. (1954), unpublished, and private communications.

JOWITT D., PATTENDEN S.K., ROSE H., SMALL V.G. & TATTERSALL R.B. (1958) AERE R/R 2516.

KALASHNIKOVA V.I., LEBEDEV V.I., MIKAELYAN L.A., SPIVAK P.E. & ZAKHAROVA V.P. (1955) Proc. conf. Acad. Sci. USSR on the peaceful uses of atomic energy. Division of phys. and math. sciences, p. 123 in Consultants' Bureau translation.

KANNE W.R., STEWART H.B., and WHITE F.A. (1956) Proc. Geneva I, 4, 315.

KATZIN L.I. (1954) National Nuclear Energy Series, division IV, <u>14A</u>, Chap. 3, McGRAW HILL, New York.

KEEPIN G.R., WINETT T.R. & ZEIGLER R.K. (1957) J. Nucl. En. 6, 1.

KENWARD C.J., RICHMOND R. & SAND J.E. (1958) AERE R/R 2212 revised.

KIRBY H.W., GROVE G.R. & TIMMA D.L. (1956) Phys. Rev. 102, 1140.

- KUKAVADSE G.M., GOLDIN L.L., ANIKINA M.P. and ERSHLER B.W. (1956) Proc. Geneva I 4, 230.
- KUSHNERIUK S.A. (1959) unpublished; also AECL 497 (1957).
- LAPORTE 0. (1937) Phys. Rev. 52, 72.
- LARSSON K-E. (1958) J. Nucl. En. 6, 322.
- LEONARD B.R., HAUSER S.M. & SEPPI E.J. (1953) HW 30128.
- LEONARD B.R. (1954) HW 33384 unpublished. Quoted by SAFFORD & HAVENS (1959); tabular data are listed by EGELSTAFF (1957).

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