Cross sections and neutron yields for U^{233}, U^{235} and Pu^{239} at 2200 m/sec.

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This is a preliminary report written in February 1959 and not intended for wide distribution. No corrections or additions have been made for this new edition.

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Summary:

The experimental information on the 2200 m/sec values for σ_{abs}, σ_f, α, $\bar{\nu}$, and η for U^{233}, U^{235} and Pu^{239} has been collected and discussed. The values will later be used in an evaluation of a "best" set of data. In an appendix the isotopic abundances of the uranium isotopes are discussed and also the alpha activities of the uranium isotopes and Pu^{239}.

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In this report we discuss the experimental information on σ_{abs}, σ_f, α, $\bar{\nu}$ and η for the three most important fissionable nuclides under bombardment with neutrons of 2200 m/sec. As the experimental techniques used in the measurements have been described thoroughly by others (see for instance HARVEY and SANDERS, 1956) we will limit ourselves to such comments as may be necessary.

Unfortunately, many of the earlier measurements have not yet been described in the open literature, and we must still use some of them. In such cases we have often had to rely on secondhand information and may have misinterpreted it.

In order to reduce all the measurements to 2200 m/sec values some assumptions must be made about the variation of the different parameters with energy and also about the neutron spectrum. We have adopted the scheme of WESTCOTT (1958) who gives the g-factors, listed in Table 1, for Maxwellian spectra at different neutron temperatures.

Table 1.

g-factors according to WESTCOTT (1958).

T, °C		20	40	60	80
U^{233}	g_f	1.0112	1.0112	1.0112	1.0114
	g_{abs}	1.0078	1.0073	1.0069	1.0068
	g_η	1.0034	1.0039	1.0043	1.0046
U^{235}	g_f	0.9754	0.9702	0.9653	0.9607
	g_{abs}	0.9749	0.9698	0.9649	0.9603
	g_η	1.0005	1.0005	1.0005	1.0004
Pu^{239}	g_f	1.0499	1.0629	1.0777	1.0946
	g_{abs}	1.0737	1.0920	1.1128	1.1362
	g_η	0.9778	0.9734	0.9685	0.9634

For measurements done inside reactors or in reactor beams we have assumed the neutron temperature was effectively 60°C except when a thermal column was used,for which we assumed 30°C ,or when a different temperature has been stated in the reference. When experimental details have been inadequately reported, we have assumed that corrections were made for the contributions of epithermal neutrons, by cadmium difference methods for example.

In recalculating relative measurements the following standard cross sections and g-factors have been used (according to BNL-325, second ed. and WESTCOTT, 1958).

Table 2.

	σ_{abs} barns	g-factor
Li	71.0	1.000
Li^6	945	1.000
B	755	1.000
Na	0.536	1.000
Mn	13.3	1.000
Au	98.8	1.006 (at T = 300°C)

The absorption cross-sections.

Most of the measurements of the absorption cross-sections, σ_{abs}, have been made by the transmission method, which measures total cross-sections. The most reliable values of the scattering cross-sections for subtraction seem to be the following:

U^{233}	12.5 ± 1.0 b	OLEKSA (1958)
U^{235}	15 ± 2.5 b	FOOTE (1958)
Pu^{239}	10 b	Calculated potential scattering.

However, as HAVENS and MELKONIAN (1958) have observed, these values may be somewhat high at 2200 m/sec. as a result of molecular and crystallic binding effects in the coherent scattering.

The measured values of absorption cross-sections are listed in Table 3.

Table 3.

σ_{abs} at 2200 m/sec.

Reference	U^{233}	U^{235}	Pu^{239}	Method	Remarks
MAY (1944)		687 ± 15 695 ± 20		Beam experiment, rel. to boron Slow chopper	Original datum unknown. See note 1 below.
ANDERSON et al. (1944)			1057	Time-of-flight measurement with cyclotron	
PALEVSKY & MUETHER (1954)	585 ± 10			Slow chopper	
PALEVSKY et al. (1954)		685 ± 5		Slow chopper	
EGELSTAFF (1954) (Revised)		709 ± 15		Slow chopper	
ZIMMERMAN & PALEVSKY (1955)			1020 ± 10	Slow chopper	
PATTENDEN (1956)	595 ± 15		1005 ± 30	Slow chopper	See note 2 below.
NIKITIN et al. (1956)	570 ± 20	695 ± 20	1030 ± 30	Time-of-flight measurements with cyclotron	
KUKAVADSE et al. (1956)	618 ± 30			Mass spectrometric, relative to Li^6, in a thermal spectrum	Original datum $\frac{\sigma_{abs}(U)}{\sigma_{Li^6}} = 0.66 \pm 0.03$
GREEN et al. (1957)	578 ± 17			Oscillator, relative to gold	Original datum same. Corrections to 2200 m/sec. and new standard cross section does not change the result.
SCHWARTZ (1958)		683 ± 6 681 ± 6		Crystal spectrometer Chopper	Unpublished work at Columbia and Argonne
BOLLINGER et al. (1958)			988 ± 10	Chopper	

Notes on data in Table 3.

(1) MAY's (1944) measurement was made with the Argonne slow chopper. At that time rather low values were obtained with the same instruments for other cross-sections:

	σ_T at 2200 m/sec.	
	ANL 1944-47	1959 data
B	705	759
Au	100	106
U^{235}	660 $\pm$ 16	

It is plausible to assume that the energy calibration of the instrument was erroneous and that values quoted for neutrons of 2200 m/sec. were really measured at 2350 $\pm$ 20 m/sec. Then May's datum would imply $\sigma_T(U^{235})$ = 710 $\pm$ 18 at 2200 m/sec or σ_{abs} = 695 $\pm$ 18 b.

(2) PATTENDEN's (1956) data shows a discontinuity in $\sigma_T(U^{233})$ at 0.025 eV. This might be due to a very weak level in that region but it has not been observed by others and does not appear in measurements of the functions σ_f and α. So we have smoothed Pattenden's data locally to get σ_T = 608 b or σ_{abs} = 595 b. Alternatively if one should accept Pattenden's data in detail it would be necessary to revise the corresponding g-factors for reactor calculations.

The fission cross section.

Direct measurement of fission cross sections, σ_f, usually involves:

1) Calibration of the neutron flux incident on the fissile sample. This is usually done by using a BF_3 chamber or by an activation of gold or manganese foils, and the fission cross section measurement is then only comparative.

2) Observation of the fission rate in a fission chamber or with photographic emulsions.

3) Assay of the thickness of the fission foil.

4) Assessment of counter efficiencies.

Various methods have been used to assay the fission foil. They rely basically on

a) absolute alpha-counting

b) weighing

but the procedure is usually rather complex. We refer the reader to the various papers cited below, and especially to FLEMING (1952).

When the measured cross section is that of U^{235} in natural uranium we give the result for pure U^{235}, assuming that the isotopic abundance of U^{235} in natural uranium is 0.7200 ± 0.0005 per cent (see the Appendix).

The data are listed in Table 4.

Table 4.

σ_f at 2200 m/sec.

Reference	U^{233}	U^{235}	Pu^{239}	$\frac{U^{233}}{U^{235}}$	$\frac{Pu^{239}}{U^{235}}$	Method	Remarks
DeWIRE et al. (1944)					1.27 ± 0.05		Original datum unknown. Corrected by EGELSTAFF et al. (1955).
DEUTSCH & LINENBERGER (1944)		540 ± 30 575 ± 30				Relative to gold and manganese.	Original data unknown. Corrected by HARVEY & SANDERS (1956)
MAY (1944 and 1945)		587 ± 13		0.83 ± 0.03		U^{235} relative to boron	Original data unknown. Corrected to $\sigma_B = 755$.
BISWAS & PATRO (1949)		561 ± 15				Fission counter and boron counter in paraffin block. RaBe source, and Cd difference measurements.	Original datum 526 ± 10, corrected for boron cross section, U^{235} abundance, and g-factor
FACCINI & GATTI (1950)		598 ± 15				Ion chambers in paraffin. Relative to Li. RaBe source and Cd difference measurements.	Original datum corrected according to COHEN et al. (1952) and to 2200 m/sec.
TUNNICLIFFE (1951)	536 ± 16		648 ± 20			Fission counter and boron counter with monocromatic neutrons from a crystal spectrometer.	Original data unknown. Boron cross section corrected.
COHEN et al. (1952)					1.31 ± 0.04	Double fission counter in reactor neutron beam.	Original datum $\frac{\sigma(Pu)}{\sigma(u)} = 203,3 \pm 4.0$ corrected for g-factors.

σ_f (continued)

Reference	U^{233}	U^{235}	Pu^{239}	$\frac{U^{233}}{U^{235}}$	$\frac{Pu^{339}}{U^{235}}$	Method	Remarks
BARLOUTAUD & LÉVÊQUE (1952)		626 ±20				Relative to manganese, double ionisation counter in reactor neutron beam. Corrected for epithermal neutrons.	Original datum $\sigma_{Mn}/\sigma_U = 3.057 \pm 0.055$. Corrected with g-factor.
COCKROFT (1952)					1.26 ±0.05	Thermal column beam.	Original datum 1.405 ± 0.009 g-corrected.
RAFFLE (1953) revised 1958)	513 ±15	590 ± 12	702 ±20			Slow chopper, fission counter relative to gold.	U^{235} value provisional.
POPOVIĆ & GRIMELAND (1953)		591 ±30				Photographic emulsions for detection of fission fragments, NaI crystal for neutrons.	Original datum $\frac{\sigma(\text{Nat. U})}{\sigma(\text{Na})} = 7.66 \pm 0.20$ g-corrected.
POPOVIĆ & SAELAND (1955)	522 ±25					D:o	Original datum 492 ± 25 corrected for cross section standard and g-factor.
AUCLAIR et al. (1956)				0.91 ±0.06	1.32 ±0.07	Double fission chamber in neutron beam with known spectrum.	Originally given as $\frac{\sigma(233)}{\sigma(239)} = 0.626 \pm 0.010$. U^{235} ratios and corrections from EGELSTAFF et al. (1955).
SAPLAKOGLU (1958)		605 ± 6				Chopper, simultaneous measurement of fissions, flux, and efficiencies.	

σ_f (continued)

Reference	U^{233}	U^{235}	Pu^{239}	$\frac{U^{233}}{U^{235}}$	$\frac{Pu^{239}}{U^{235}}$	Method	Remarks
BIGHAM et al. (1958)	513 ± 5			0.911 ±0.009	1.302 ±0.013	Double fission counters in large D_2O thermal column, U^{233} relative to gold.	

The capture to fission ratio.

The ratio between the capture and the fission cross sections, α , is usually determined by measuring with a mass spectrometer the changes in isotopic composition of a sample resulting from irradiation in a reactor. Some uncertainty may arise if the neutron spectrum is not well known. Also, in case of U^{235} it has been found by PALMER (1958) that potassium ions can distort the analysis of U^{236}.

As $\overline{\nu}$ is constant in the thermal region, the g-factor to use for $1 + \alpha$ is the inverse of the g-factor for η : it is g_{abs}/g_f where g_{abs} and g_f are the g-factors for the indicated cross-sections.

The data are given in Table 5.

Table 5.

α at 2200 m/sec.

Reference	U^{233}	U^{235}	Pu^{239}	Method	Remarks
WILLIAMS & YUSTEF (1946)		0.183 $\pm$0.006		Mass spectrometry.	According to Schwartz (1958) later investigations have shown that no spectrum corrections are necessary. This follows from $g_\eta \approx 1$. In the resonance region $\alpha \sim 0.6$ on the average, but the contribution from resonance neutrons is small in a thermal reactor.
DEUTSCH et al. (1946)		0.186 $\pm$0.008		Mass spectrometry and γ activity measurement.	
HANNA (1955)		0.18 $\pm$0.02			Original datum unknown. Quoted by EGELSTAFF et al. (1955)
KANNE et al. (1956)		0.175 $\pm$0.017	0.40 $\pm$0.06	Fission product yield, mass spectrometry of U^{236}, spontaneous fission rate of Pu^{240}. Reactor spectrum .	Error assumed to be 10% for U and 15% for Pu. Pu value corrected to 2200 m/sec. from 0.42, and U value from 0.174.
INGHRAM et al. (1956)	0.102 $\pm$0.002			Mass spectrometry, reactor spectrum.	Original datum 0.098 corrected to 2200 m/sec.
KUKAYADSE et al. (1956)	0.099 $\pm$0.003			Mass spectrometry, α counting and fission product yield. Thermal spectrum.	Original datum 0.095 corrected to 2200 m/sec.
CRAIG et al (1958)		0.197 $\pm$0.004		Mass spectrometry.	Original data 0.196 and 0.186 corrected to 2200 m/sec.
		0.187 $\pm$0.004		α pulse height analysis.	

The number of neutrons per fission.

At present all but one of the absolute measurements of $\bar{\nu}$, the average number of neutrons emitted per fission, have been made indirectly, through comparisons with standard neutron sources. The accuracy of the measurements depends very largely on the accuracy of calibration of the sources; consistency of independent measurements depends on the consistency of the relative calibrations of the various sources.

Calibration of neutron sources has been discussed in detail by RICHMOND (1958) and by LARSSON (1958), and they show that there is now good agreement between the source strengths measured with different techniques in various laboratories throughout the world. Neither of the authors cited has attempted a "least squares" calibration from all available absolute and relative measurements, nor shall we. In compiling the various absolute measurements of $\bar{\nu}$ we have, when possible, revised the data to accord with the "world average" source strength defined by LARSSON (1958). In doing this we have assumed that the neutron sources which were actually used in the experiments were themselves calibrated against those which were used in the network of international comparisons. The following correction factors have been applied:

Los Alamos source	1.008	WALKER (1946) via NBS value given by LARSSON (1959)
Russian source	1.008	LARSSON (1958)
Harwell source	1.014	LARSSON (1958)

Because of the recent improvements in the accuracy of source calibrations the uncertainties originally cited for $\bar{\nu}$ have been decreased somewhat.

The increase of $\bar{\nu}$ with energy of the incident neutrons is very small (about 0,13 per MeV according to LEACHMAN, 1958), so that measurements done with a thermal neutron spectrum can equally well be interpreted as 2200 m/sec values.

To be precise we shall define $\bar{\nu}$ as including both prompt and delayed neutrons. However, in some of the experiments only the prompt neutrons were measured, because a coincidence technique was used. The delayed neutron yields which have been added in these cases are (KEEPIN 1957)

for U^{233} 0.0066 ± 0.0003 per fission
for U^{235} 0.0158 ± 0.0005 " "
for Pu^{239} 0.0061 ± 0.0003 " "

The data are listed in Table 6.

Table 6.

$\bar{\nu}$ at thermal energy.

Reference	U^{233}	U^{235}	$\frac{U^{233}}{U^{235}}$	$\frac{Pu^{239}}{U^{235}}$	Method	Remarks
SNYDER & WILLIAMS (1944)		$^{+2.46}_{-0.06}$		$^{+1.17}_{-0.02}$	Fission counting and comparison with standard source, using In-foils in large graphite moderator.	Original datum 2.44 ± 0.12, corrected for source strength.
DEWIRE et al. (1944)			$^{+1.029}_{-0.010}$	$^{+1.178}_{-0.009}$	Coincidences between fissions and proton recoils.	Original data 1.033 and 1.182, corrected for delayed neutrons.
KALASHNIKOVA et al. (1955)	$^{+2.64}_{-0.06}$		$^{+1.035}_{-0.010}$	$^{+1.188}_{-0.012}$	Relative measurements with boron and fission counters. Absolute calibration by a coincidence method together with a standard source.	Original datum 2.62 ± 0.10 corrected for source strength and delayed neutrons.
DIVEN (1955)		$^{+2.428}_{-0.060}$	$^{+1.043}_{-0.022}$	$^{+1.230}_{-0.028}$	Liquid scintillator, calibrated with recoil protons.	Original data given for 80 keV neutrons; corrected to thermal energy and for delayed neutrons
SANDERS (1956)		$^{+2.47}_{-0.15}$	$^{+1.001}_{-0.016}$	$^{+1.164}_{-0.022}$	Neutron and fission counters in coincidence. Absolute value is relative to spantaneous fission rate of natural U.	Original data 2.45, 1.005 and 1.168 corrected for delayed neutrons. JOHNSTONE (1955) obtained closely similar results.
KENWARD et al. (1957)		$^{+2.455}_{-0.030}$			Neutron and fission counters in coincidence. Absolute value from Pu^{240} spontaneous fission source.	Original datum 2.405 ± 0.037 corrected for delayed neutrons and source strength.
COLVIN & SOWERBY (1958)			$^{+1.025}_{-0.006}$	$^{+1.191}_{-0.007}$	Coincidences between fission counter and boron pile detector.	

The number of fission neutrons per absorbed neutron.

The average number of fission neutrons emitted per neutron absorbed in a fissile nuclide, η , is defined as

$$\eta = \bar{\nu}\, \sigma_f / \sigma_{abs}.$$

No direct measurements of η have been reported, but relative measurements have been made by LICHTENBERGER and ZINN (1946), and by RICHMOND (1955) and are given in Table 7 below. Essentially, a thermal neutron beam is completely absorbed in a thick block of U or Pu. Measurement is made of incident flux and of the fast neutron emission. The flux calibration is not required for relative measurements.

By using thin fissile samples in a similar arrangement JAFFEY et al. (1955) made relative measurements of $\eta\, \sigma_a$ (which may also be written $\bar{\nu}\sigma_f$): see Table 8.

The great majority of measurements on η have been made by reactivity measurements in a reactor (critical size, danger coefficient, pile oscillator, etc.). Such an experiment measures

$$(W\eta - 1)\, \sigma_{abs} / \sigma_\rho ,$$

where W is the average importance of a fission neutron compared with that of a reactor neutron, σ_{abs} is the absorption cross-section of the fissile material and σ_ρ is that of the absorber used to calibrate the reactivity scale (usually boron has been used).

W varies from one location to another, but is usually about unity in thermal systems. Thus to give a simple interpretation of the experiment we may say that it permits evaluation of

$$(\eta - 1)\, \sigma_{abs} / \sigma_\rho$$

in which, of course, numerator and denominator are averaged separately over the neutron spectrum. In heterogeneous regions, the value of W may be somewhat uncertain, and it is then better to interpret the measurements as giving relative values of $(\eta - 1)\, \sigma_{abs} / \sigma_\rho$.

Available data are listed in Table 9 below. Two exceptions are made: SPIVAK and YEROZOLIMSKY (1956) and GAERTTNER et al. (1958) determined the absorption cross section ratios in the very experiment.

It was felt that these two experiments are more reliably interpreted as measurements of η, and the results are therefore included in Table 7.

Table 7.

η at 2200 m/sec.

Reference	U^{233}	U^{235}	Pu^{239}	$\frac{U^{233}}{U^{235}}$	Natural U	Method	Remarks
LICHTENBERGER & ZINN (1946)				$^{+1.110}_{-0.015}$		Pile neutron beam.	Original datum unknown. Value taken from EGELSTAFF (1955) Corrected to 2200 m/sec.
RICHMOND (1955)				$^{+1.107}_{-0.020}$	$^{+1.019}_{-0.030}$	D:o	D:o
SPINAK & YEROZOLIMSKY (1956)	$^{+2.27}_{-0.028}$	$^{+2.064}_{-0.025}$	$^{+2.081}_{-0.025}$		$^{+1.337}_{-0.017}$	Oscillator in graphite thermal column.	Original data 2.28, 2.065 and 2.035 corrected to 2200 m/sec.
GAERTTNER et al. (1958)				$^{+1.075}_{-0.012}$	$^{+0.978}_{-0.010}$	Reactivity measurement with Cd difference.	Original U^{235} datum 1.078 corrected to 2200 m/sec. (T = 42°C according to measurements.)

Table 8.

$\bar{\nu}\sigma_f$ ratios at 2200 m/sec.

Reference	$\frac{U^{233}}{U^{235}}$	$\frac{Pu^{239}}{U^{235}}$	Method	Remarks
JAFFEY et al. (1955)	$\pm^{0.902}_{0.011}$	$\pm^{1.496}_{0.020}$	Pile neutron beam, fission neutrons detected by annular boron counter, Cd difference.	Original data 0.945 and 1.670 corrected to 2200 m/sec.

Table 9.

$(\eta - 1)\ \sigma_{abs}/\sigma_B$ at 2200 m/sec.

References	U^{233}	U^{235}	Pu^{239}	$\frac{U^{233}}{U^{235}}$	$\frac{Pu^{239}}{U^{235}}$	Natural U	Method	Remarks
FERMI & MARSHALL (1944)							Reactivity measurement relative to boron	$\eta(U^{235}) = 2.10 \pm 0.04$. Assumed cross sections are not known.
ANDERSON & NAGLE (1944)							Reactivity measurement in reactor core.	$\eta\ (Pu^{239})\ \eta\ (U^{235}) = 1.00 \pm 0.03$ at 2200 m/sec. Assumed cross sections are not known.
LICHTENBERGER & ZINN (1946)							Pile oscillator	$\eta\ (U^{233}) = 2.28 \pm 0.03$; $\eta\ (U^{235}) = 2.07 \pm 0.03$ at 2200 m/sec. Assumed cross sections are not known.
CRUIKSHANK et al. (1948)	$^{+1.09}_{-0.07}$						Pile oscillator	See note (1) below
MUELHAUSE (1952 and 1958)	$^{+0.95}_{-0.03}$	$^{+0.98(5)}_{-0.03}$	$^{+1.43}_{-0.07}$				Pile oscillator, thermal spectrum	
HARRIS & ROSE (1953)							Pile oscillator, thermal spectrum	$\eta\ (Pu^{239}) = 2.05 \pm 0.04$. Assumed cross sections are not known.
THOMAS et al. (1955)				$^{+1.02}_{-0.03}$			Criticality experiments	Corrected by FRANCIS et al. (1957)
ALICHANOV et al. (1956)	$^{+1.05}_{-0.04}$	$^{+1.05}_{-0.05}$	$^{+1.5}_{-0.08}$				Reactivity measurement in internal thermal column.	

Table 9 continued.

References	U^{233}	U^{235}	Pu^{239}	$\frac{U^{233}}{U^{235}}$	$\frac{Pu^{239}}{Pu^{235}}$	Natural	Method	Remarks
BURGOV (1956)						$^{+0.00350}_{-0.00006}$	Reactivity measurement in internal thermal column.	
ROSE et al. (1958)				$^{+0.961}_{-0.013}$	1.46 0.02		Pile oscillator in internal thermal column.	
MAGNUSSON & GWIN (1958)							Reactivity measurements in internal thermal column.	$\eta(U^{233}) = 2.31 \pm 0.06$ and $\eta(Pu^{239}) = 2.03 \pm 0.08$ in the experimental spectrum, but cross sections used are not known.
MAGNUSSON (1958)							Criticality experiments.	$\eta(U^{233}) = 2.268 \pm 0.042$ in the experimental spectrum, but cross sections used are not known.

APPENDIX

Half-lives and atomic abundances of the isotopes in natural uranium.

Relative isotopic abundances of U^{234}, U^{235}, U^{238} in natural uranium have been measured with mass spectrometers. Specific alpha activities of these isotopes have been measured with ionisation chambers of well-defined geometrical efficiency, both with natural and enriched samples. The data are super-abundant, but it proves easy to select a set of recommended values.

A previous summary and discussion of the data was given by FLEMING (1952) in an admirable report. We include some newer data and make minor modifications.

Notation. All the isotopes referred to decay by alpha emission only, the spontaneous fission rates being relatively quite negligible. We write:

A for the atomic abundance of an isotope in natural uranium
M " its atomic weight in AMU (physical scale)
λ " " decay constant
$\tau = \lambda^{-1}$. ln 2 is its half-life
$\alpha = N_o \lambda / M$ " " specific alpha activity
$N_o = 6.0250_2 \times 10^{23}$/g.mole (physical scale)
Suffixes 4, 5, 8, n denote U^{234}, U^{235}, U^{238} and natural uranium respectively.

Experimental Results.

Mass spectrometry.

Measurements of the relative abundance by mass spectrometry are collected in Table A 1 below.

The relative abundance of U^{234} is so low that it cannot be measured very accurately with a mass spectrometer.

No variation of the U^{235} abundance has been found in natural uranium from various sources of widely differing geological ages. SENFTLE et al. (1957) examined eleven samples of uranium ore from the Colorado plateau, one from Joachimstal, and one from Great Bear Lake, and could find no significant variation in the ratio A_8/A_5. FOX and RUSTAD (1946) found this ratio constant to 0.03% for two African

ores, one from Canada, and a Colorado carnotite. NIER (1939) examined Swedish, Canadian, and American ores and found no variation to 0.07% (well within the experimental uncertainty).

Table A 1

Relative U abundances by mass spectrometry.

Item	Authors	A_8/A_5	A_8/A_4	A_5/A_4
1.	SENFTLE et al. (1957)	$137.7 \pm (0.3)$		
2.	LOUNSBURY (1956)	137.80 ± 0.14		
3.	WHITE et al. (1956)	138.3 ± 1.0	17730 ± 320	
4.	GREENE et al. (1955)	137.96 ± 0.11		
5.	BOARDMAN and MESERVEY (1948)			
6.	FOX and RUSTAD (1946)	137.0 ± 0.7		
7.	" " "	138.0 ± 0.3		
8.	INGHRAM (1946)	$137.8 \pm (1)$		
9.	CHAMBERLAIN et al. (1946)	$139 \pm (1.4)$		$141.5 \pm (6\%)$
10.	NIER (1939)	138.9 ± 1.4	$16760 \pm (10\%)$	
	Weighted mean of above	137.89 ± 0.10		

Alpha Counting Experiments.

The data cited below are collected for convenience in Table A2 on page 6.

To determine absolutely the specific alpha activity of a radioactive sample it is necessary to use a thin sample, so that essentially all the alphas will emerge, and to determine its mass accurately. An ionisation chamber of well-defined geometry is used, which will count all the alpha particles entering it, so that the fraction of alphas counted is determined by the solid angle subtended at the source.

In some experiments aluminium or nickel mounts have been used: These oxidise when heated so that the weight of the radioactive

sample is overestimated. Often a counter with 2π solid angle has been used: then corrections are necessary for alphas scattered back into the counter by the mount. FLEMING (1952) showed that, because many of these back-scattered particles have low energies, the counting rate varies with the gain setting.

FLEMING (1952) himself measured α_4 and α_5 with great care. Highly enriched samples were used and their chemical purity examined. The isotopic compositions of these samples were determined by mass spectroscopy. With a sample containing 99.9% U^{235} the small U^{234} content contributes about 1/3 of the alpha emanation: this fraction was more accurately assessed by pulse analysis of the alpha spectrum. With the sample enriched in the U^{234} isotope competing activities were negligible.

The alpha particles were counted with a medium geometry counter, which had been calibrated against a low geometry counter with help of a more active source. The results are:

$$\alpha_n = 1.370 \times 10^7 \pm (0.64\%) \text{ alphas/min.mg.}$$
$$\alpha_5 = 4742 \pm 106 \text{ alphas/min.mg.}$$

KNIGHT (1950) measured the specific activity of a portion of the same highly enriched U^{235} sample as Fleming used. He found 7279 ± 23 α's/min.mg. but made only a rather inaccurate pulse analysis to find out how much of this activity was due to U^{235} count. If we use Fleming's values, viz.

$$\left.\begin{array}{l}(65.3 \pm 1.3)\ \%\ \text{of disintegrations} \\ (99.94_n \pm 0.01_o)\ \%\ \text{of the mass}\end{array}\right\} \text{ due to } U^{235} \text{ content}$$

we deduce

$$\alpha_5 = 4756 \pm 69 \text{ alphas/min.mg.}$$

Since Knight used a 2π counter, subject to the uncertainties mentioned above, the precision claimed is probably too optimistic.

KIENBERGER (1952) measured the activity of a sample of highly enriched U^{234} in a 2π chamber. The backscattering correction (1.19 ± 0.08) % was measured experimentally and is in reasonable accord with theoretical estimates. The final result is given as

$$\alpha_n = (1.348 \pm 0.004) \times 10^7 \text{ alphas/min.mg.}$$

Recalling Fleming's findings with 2π chambers the precision claimed seems optimistic.

In an earlier paper KIENBERGER (1949) used a 2π chamber in measuring the specific activities of natural and enriched samples of uranium. The backscattering correction was taken as $(1.26 \pm 0.06)\%$ from Crawford's rough estimates. Probably the experimental value $(1.19 \pm 0.08)\%$ cited above is more accurate. With this value we correct Kienberger's results to

$$\alpha_n = 1503.0 \pm 1.5 \quad \text{alphas/min.mg.}$$
$$\alpha_8 = 743.2 \pm 1.6 \quad " \quad " \quad "$$
$$\alpha_4 = (1.346 \pm 0.004) \times 10^7 \quad \text{alphas/min.mg.}$$

Again, the precision seems optimistic. In both these experiments nickel mounts were used. As these oxidize on heating, the sample weight may have been overestimated slightly. Thus Kienberger's results are likely to underestimate the specific activities.

KOVARIK and ADAMS (1955) measured the specific activity of natural uranium to be

$$\alpha_n = 1503 \quad \text{alphas/min.mg.}$$

The precision of this result is not stated.

This experiment is presumably a modification of an earlier one, KOVARIK and ADAMS (1941) in which the sample was covered by a thick brass grid to eliminate the backscattering. The particles emerge through holes in this grid or collimator: uncertainties may arise from the possibility that alpha particles are scattered from the walls of these holes.

The result of the earlier measurement was

$$\alpha_n = 1501 \pm 6 \quad \text{alphas/min.mg.}$$

the uncertainty being that cited by FLEMING (1952).

CURTISS et al. (1941) measured the specific alpha activity of natural uranium with a 2π cahmber and give the result

$$\alpha_n = 1501.2 \pm 3.0 \quad \text{alphas/min.mg.}$$

The samples were so thick that the backscattering correction is negligible; various sample thicknesses were used and the results extrapolated to zero thickness. The extrapolation is not very reliable because the thinnest samples, which affect it most strongly, will contain backscattered components.

CHAMBERLAIN et al. (1946) by alpha counting of natural and enriched uranium samples with a 2π counter measured the ratio:

$$\frac{\alpha_4}{\alpha_n} = 0.962 \times 10^4 \pm (6\%).$$

GOLDIN et al. (1949) measured the product $\alpha_4 \alpha_8$ by an extremely complex genetic study. Th^{234} formed by alpha decay of U^{238} was separated chemically from its parent and allowed to decay into U^{234}. The weight of U^{234} was calculated from the weight of parent U^{238} and the decay constants of U^{238} and the intermediate isotopes involved. If we assume a back-scattering correction of 1.19% for the alpha counting the result is

$$\alpha_4 \alpha_8 = 9.45 \times 10^9 \pm (1.33\%) \quad (\text{disint./min.mg.})^2$$

The precision claimed seems improbably high in view of the complexity of the technique.

SAYAG (1953) measured activity ratios in natural uranium by pulse analysis of the alpha spectrum. The resolution of their analyser was rather poor so that weak branches of high and low energies from the U^{235} alpha spectrum were concealed in the "U^{234}" and "U^{238}" alpha peaks. Correcting for these weak branches, the results reported lead to:

$$\frac{A_4 \lambda_5}{A_4 \lambda_4} = (4.79 \pm 0.18) \times 10^{-2}$$

$$\frac{A_4 \lambda_4}{A_8 \lambda_8} = 0.9869 \pm 0.0012$$

The latter ratio should exceed unity and the reason for the low value observed is not understood.

CLARK et al. (1944) made a similar measurement. The same comments and corrections apply, and lead to

$$\frac{A_4 \lambda_4}{A_8 \lambda_8} = 1.0012 \pm 0.0073$$

$$\frac{A_5 \lambda_5}{A_8 \lambda_8} = (4.230 \pm 0.053) \times 10^{-2}$$

Table A 2

Experimental data and best values.

Constant	Value	Weight	Reference
A_8/A_5	137.88 ± 0.10		Weighted mean from table A 1
α_5	4742 ± 106	1	FLEMING (1952)
	4756 ± 69	1	KNIGHT (1952)
	4749 ± 90 alphas/min.mg.		Mean value
α_n	1503.0 ± 1.5	1	KIENBERGER (1949)
	1503	1	KOVARIK & ADAMS (1955)
	1501 ± 6	1/3	" " " (1941)
	1501.2 ± 3.0	1	CURTISS et al. (1941)
	1509.6 ± 3.4	1	from KIENBERGER's value for α_8
	1504.6 ± 2.5 alphas/min.mg.		Weighted mean
α_8	743.2 ± 1.6		KIENBERGER (1949)
	740.7 ± 1.3 alphas/min.mg.		Best value, from α_n
$\alpha_4 \times 10^{-7}$	1.370 ± 0.009	1	FLEMING (1952)
	1.348 ± 0.004	1	KIENBERGER (1952)
	1.346 ± 0.004	1/2	" (1949)
	1.276 ± 0.017	0	GOLDIN et al. (1949) using best value for α_8
	1.482 ± 0.090	0	mass sp: CHAMBERLAIN et al.
	1.447 ± 0.087	0	α counting (1946)
	1.26 ± 0.13	0	NIER (1939), mass sp.
	1.336 ± 0.024		WHITE et al. (1956)
	1.356 ± 0.010		Recommended value
$\frac{A_5 \lambda_5}{A_4 \lambda_4}$	(4.79 ± 0.18) %	0	SAYAG (1953)
	(4.23 ± 0.05) %	0	CLARK et al. (1944)
	(4.59 ± 0.10) %		Recommended value

A_4 and α_4 are rather accurately tied by α_n and equation (1) giving

$$A_4 \alpha_4 = 748.0 \pm 1.6 \text{ alphas/min.mg.}$$

So we do not present separately the data for A_4.

The latter value is very low in comparison with other data: the reason is not understood.

These two measurements of activity ratios, by pulse analysis, might be interpreted, in conjunction with other data, as measurements of α_5. However, it would not be reasonable to think they can compare with the similar measurement by Fleming using highly enriched U^{235}.

Assessment of Recommended Values.

We shall not attempt a "least squares" fit to all the data, but are content to pick out those results which seem sufficiently reliable: the weighting which we show in table A2 above is purely subjective.

Since U^{238} and U^{234} are in radioactive equilibrium in natural uranium we have

$$\frac{A_4 \lambda_4}{A_8 \lambda_8} = \frac{\lambda_4}{\lambda_4 - \lambda_8} = 1.00005556 \pm 0.00000043 \qquad \text{(A1)}$$

Since A_4 is only $\sim 5.5 \times 10^{-5}$

$$A_5 + A_8 = 0.99994484 \pm 0.00000041 \qquad \text{(A2)}$$

In both these results the uncertainty is quite trivial.

The only experiments giving reliable information on the ratio A_8/A_5 are the mass spectroscopic measurements. Weighting the data of table A1 according to the errors we infer

$$\frac{A_8}{A_5} = 137.88 \pm 0.10$$

and using equation (2)

$$A_5 = (0.7200 \pm 0.0052)\ \% \qquad \text{(A3)}$$
$$A_8 = (99.2745 \pm 0.0052)\ \%$$

There are two direct measurements of α_5 to which we give equal weight. A third value can be inferred from α and α_8 by making use of equations (A1) and (A2): however the result is too inaccurate to carry any weight. We infer then that

$$\alpha_5 = 4749 \pm 90 \quad \text{alphas/min.mg. } U^{235}$$

Then the half-life is

$$\tau_5 = (7.11 \pm 0.13) \times 10^8 \text{ yr.}$$

and the contribution of U^{235} to the activity of natural uranium is

$$A_5\alpha_5 \, M_5/M_n = 33.76 \pm 0.71 \quad \text{alphas/min.mg.} \qquad \text{(A4)}$$

Herein we have assumed the atomic weights

$$M_4 = 234.1143 \pm 0.0007$$
$$M_5 = 235.1174 \pm \text{ "}$$
$$M_8 = 238.1254 \pm \text{ "}$$

and infer

$$M_n = 238.1035 \pm 0.0007$$

from the atomic abundances derived in this section: dependence on the abundance of U^{234} is very weak.

There are four direct measurements of the disintegration rate in natural uranium. A fourth value can be inferred from KIENBERGER's measurement of α_8 by using equations (A1), (A3) and (A4). That gives the rather high value

$$\alpha_n = 1509.6 \quad \text{alphas/min.mg.}$$

With the weighting given in table A2 we infer as best value

$$\alpha_n = 1504.6 \pm 2.5 \quad \text{alphas/min.mg.} \qquad \text{(A5)}$$

Subtracting equation (A4) from (A5) and using (A1)

$$\alpha_8 = 740.7 \pm 1.5 \quad \text{alphas/min.mg. } U^{238}$$
$$A_4\alpha_4 = 748.0 \pm 1.6 \quad \text{alphas/min.mg.} \qquad \text{(A6)}$$

The latter precise relationship permits us to collect together all the mass spectroscopic and alpha counting data on U^{234}.

Weighting the data as indicated in table A2:

$$\alpha_4 = (1.356 \pm 0.010) \times 10^7 \quad \text{alphas/min.mg. } U^{234}$$

Then equation (A6) gives

$$A_4 = (5.516 \pm 0.041) \times 10^{-5}$$

Recommended values are collected, finally, in table A 3.

Table A 3.

Recommended values of decay rates and isotopic abundances.

SEPARATED ISOTOPES			
Specific activities:			
U^{234}	$\alpha_4 = (1.356 \pm 0.010) \times 10^7$	disints./min.mg.	U^{234}
U^{235}	$\alpha_5 = 4749 \pm 90$	-----	------ U^{235}
U^{238}	$\alpha_8 = 740.7 \pm 1.5$	-----	----- U^{238}
Half-lives:			
U^{234}	$\tau_4 = (2.501 \pm 0.018) \times 10^5$ years		
U^{235}	$\tau_5 = (7.11 \pm 0.13) \times 10^8$ "		
U^{238}	$\tau_8 = (4.502 \pm 0.009) \times 10^9$ "		
NATURAL URANIUM			
	Abundance, atoms %	Disints./min.mg.nat. U	
U^{234}	$(5.516 \pm 0.041) \times 10^{-3}$	$735.4_4 \pm 1.5$	
U^{235}	0.7200 ± 0.0052	33.76 ± 0.71	
U^{238}	99.2745 ± 0.0052	$735.4_0 \pm 1.5$	
		Total 1504.6 ± 2.5	
Relative alpha activities in natural uranium:			
$\frac{U^{234}}{U^{238}}$	$\frac{A_4 \lambda_4}{A_8 \lambda_8} = 1.00005556 \pm 0.000\,000\,43$		
$\frac{U^{235}}{U^{234}}$	$\frac{A_5 \lambda_5}{A_4 \lambda_4} = 0.04591 \pm 0.00097$		

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