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Report

MASS SPECTROMETRIC MEASUREMENTS OF THE RATIO OF THERMAL NEUTRON CAPTURE TO FISSION FOR ^{241}Pu

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by

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ABSTRACT

Highly-enriched samples of ^{241}Pu have been irradiated in five different positions in a flux scanning tube of the PLUTO reactor so that alpha (the ratio of neutron capture to fission) could be measured for this nuclide. Highly-enriched samples of ^{239}Pu and ^{240}Pu were also irradiated at the same time so that allowance could be made for the effects of their destruction.

Experimental values of $\hat{\alpha}$ for ^{241}Pu varied from 0.352 to 0.395, with a mean value of 0.371 ± 0.016. Correcting all results to 2200 m/sec neutrons gave a final value for α_o of 0.390 ± 0.023. Where a comparison has been possible of the results reported here with previously published data, good agreement has been found, but the scarcity and imprecision of the latter renders such a comparison of little value.

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CONTENTS

TABLES

ILLUSTRATIONS

INTRODUCTION

1. The large and ever-increasing amounts of plutonium which are resulting from the United Kingdom's nuclear power programme have added impetus to the desire to use this element as fuel in future reactor systems. Thus, not only is plutonium now suggested as the main fuel for fast reactors, but the possibility of using it in thermal reactors is also being considered[1]. Indeed, even at the present time with thermal reactors originally fuelled with natural uranium, by the end of its life one third of the heat generated in the fuel will have arisen from the fission of plutonium.

2. It is easy to see therefore that the nuclear properties of the plutonium isotopes are of great interest to reactor designers and, as ESHBACH and GOLDSMITH[2] have pointed out, of key importance in this respect is the value of α, the relative probability of an absorbed neutron being captured rather than causing fission.

3. Yet the precise measurement of α in the thermal region for the fissile plutonium isotopes ^{239}Pu and ^{241}Pu has received little attention, and this fact is particularly noticeable in the case of the latter nuclide. WESTCOTT et al[3], at the instigation of the Organising Committee of the 1964 Geneva Conference, have made an exhaustive study of the 2200 m/sec data available for the primary fissile nuclides, but have been unable to find a single measurement of α_o for ^{241}Pu worthy of note*. The only data we have found ourselves is indirect in nature. FIELDS et al[4] have measured the changes in isotopic composition produced in a plutonium sample (originally largely ^{239}Pu) by irradiations of varying duration in the Chalk River reactor NRX, and have concluded that the effective capture and fission cross sections of ^{241}Pu for the reactor neutrons were 390 ± 80 barns and 1060 ± 210 barns respectively i.e. that $\hat{\alpha} = 0.37 \pm 0.10$. However this result is not very precise and the neutron spectrum conditions under which it was obtained are uncertain.

4. The purpose of this present investigation was to measure $\hat{\alpha}$ for ^{241}Pu under defined and varying reactor spectrum conditions and to derive a more precise value for α_o.

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*We use the symbols α_o and $\hat{\alpha}$ to imply values of alpha for 2200 m/sec neutrons and reactor neutrons respectively.

METHOD

5. The mass spectrometric method of measuring $\hat{\alpha}$ has been employed since it is the simplest and the least dependent on a knowledge of subsidiary nuclear or other data; it is not necessary, for example, to know fission yields or to employ absolute counting methods. Examples of the method applied to the nuclides ^{233}U, ^{235}U and ^{239}Pu have already been published[5,6,7,8]. In the case of ^{241}Pu the aim is to use a mass spectrometer to measure both the amount of ^{242}Pu formed by neutron capture by ^{241}Pu during an irradiation, and the amount of ^{241}Pu lost from the mixture at the same time. Since the former quantity is proportional to the reactor neutron capture cross section of ^{241}Pu (which we denote as $\hat{\sigma}_1$) and the latter quantity, suitably corrected for radioactive decay, is proportional to the reactor neutron absorption cross section of ^{241}Pu (which we denote as $\hat{S}_1$), the ratio of the two can be obtained and related to $\hat{\alpha}$ by the expression:-

$$\hat{S}_1/\hat{\sigma}_1 = (1 + 1/\hat{\alpha}) \qquad (1)$$

6. However, a mass spectrometer does not measure numbers of atoms but atomic ratios, consequently the amounts of ^{241}Pu and ^{242}Pu present in the mixture must be measured relative to another plutonium isotope. At the time these experiments were conceived ^{238}Pu was not available in macro quantities leaving the choice of reference isotope between ^{239}Pu and ^{240}Pu. Neither of these alternatives is very satisfactory. In Table I we have collected the current 'best' values for the neutron cross sections of the chief plutonium isotopes in the thermal and epithermal regions and in Fig. I we show diagrammatically the most important reactions occurring when a mixture of plutonium isotopes is irradiated with neutrons. It will be seen that both ^{239}Pu and ^{240}Pu will be destroyed fairly rapidly by neutrons in either energy group.

7. We have chosen ^{239}Pu as the reference isotope because the immediate product of neutron capture i.e. ^{240}Pu, does not interfere in the main measurements. Nevertheless, under the irradiation conditions we have employed, an appreciable proportion of the ^{239}Pu is converted into ^{240}Pu and some of this in turn also undergoes neutron capture and is transmuted

into ^{241}Pu. In addition further quantities of ^{239}Pu are destroyed by neutron fission. Corrections terms are necessary for these effects. These could have been calculated entirely from the nuclear data given in Table I and the known irradiation conditions, but we have preferred the more certain method of actually measuring them in subsidiary experiments. (In these cases the less neutron sensitive ^{242}Pu is used as reference isotope).

8. The experiment therefore consists in irradiating three different mixtures of plutonium isotopes under known spectrum conditions, and measuring the isotopic changes which take place. The first mixture consists largely of ^{239}Pu and ^{241}Pu so that the rate of formation of ^{242}Pu from ^{241}Pu and the rate of destruction of ^{241}Pu can be measured relative to the ^{239}Pu content. The second mixture consists largely of ^{239}Pu and ^{242}Pu so that the rate of destruction of ^{239}Pu and the rate of formation of ^{240}Pu from ^{239}Pu can both be measured relative to the comparitively unchanged ^{242}Pu. The third mixture consists mainly of ^{240}Pu and ^{242}Pu so that the rate of conversion of ^{240}Pu to ^{241}Pu can be measured relative to the ^{242}Pu content.

9. In this way the rates of all the relevant nuclear transformations occurring in the system can be measured except that at which ^{242}Pu is destroyed by neutron capture. This is the slowest reaction of all and its magnitude must be determined by calculation.

10. Another requirement for the success of the experiments is that, having obtained $\hat{\alpha}$, α_o may be deduced from the expression:-

$$(1 + \alpha_o) = (1 + \hat{\alpha})(g_f + rs_f)/(g_a + rs_a) \qquad (2)$$

in which the g and s terms for neutron fission and absorption and the epithermal index, r, are defined by WESTCOTT, WALKER and ALEXANDER[13].

11. WESTCOTT has recently calculated the relevant g and s factors for ^{241}Pu and we have used these[14]. Epithermal indices have been determined in two quite different ways using either (a) a combination of silver and cobalt monitors or (b) the ^{240}Pu - ^{242}Pu mixture as an epithermal monitor. A full description of these measurements is given elsewhere[15].

EXPERIMENTAL DETAILS

A. Mass Spectrometric Measurements

12. All mass spectrometric measurements were made using an A.E.I. Type M.S.5 mass spectrometer. For each isotopic analysis the plutonium was mounted on one of the side filaments of a tungsten triple-filament surface ionization source and examined with centre and side filament currents of about 6A and 3.5A respectively. Whenever possible at least twenty consecutive measurements were made of each isotopic ratio of interest.

B. Target Materials and Treatment

13. Nitric acid solutions of the four highly enriched plutonium isotopes - ^{239}Pu (99.9 per cent), ^{240}Pu (99.3 per cent), ^{241}Pu (97.3 per cent) and ^{242}Pu (98.0 per cent) - were supplied by the Electromagnetic Separation Group of Chemistry Division, A.E.R.E. Aliquots were mixed together to give the three compositions required and fractions from each mixture were mounted on precipitated silica in the manner described by CABELL and SLEE[7]. Sample tubes were then made up by sealing about 10 mg silica containing about 10μg plutonium in a number of small silica phials under reduced pressure.

14. The isotopic compositions of the plutonium samples, both before and after irradiation, were determined by treating irradiated and un-irradiated samples alike, except that it was necessary to handle irradiated samples remotely inside a shielded cell whilst the unirradiated samples could be treated in a dry box.

15. Each sample tube was broken open by percussion, the contents were transferred to a platinum crucible, then treated with hydrofluoric and perchloric acids to remove the silica. Final traces of hydrofluoric acid were then removed by evaporation of the solution to small bulk and repeated treatments of the residue with nitric acid followed by evaporation to small bulk once more. In order to ensure that all the plutonium was eventually in the Pu^{4+} state before it was fed to the anion exchange column, the plutonium was first reduced to Pu^{3+} by evaporating the nitric acid solution almost to dryness, adding 1 ml water and a few crystals of hydroxylamine, then warming gently to complete the reduction. After cooling, an equal volume of concentrated nitric acid was added dropwise

and very carefully in order to destroy excess hydroxylamine and oxidise the plutonium to Pu^{4+}. The resultant solution was then evaporated almost to dryness once more and finally diluted with 2 ml 8M nitric acid.

16. This solution was now fed to a glass column containing 100 mg Deacidite FF which had previously been conditioned with 8M nitric acid. On passage of the solution through the resin the plutonium was absorbed whilst the bulk of the fission products, ^{241}Am and ^{243}Am (daughters of ^{241}Pu and ^{243}Pu), passed through. Decontamination of the plutonium was then completed by elution of the column with 5 ml 8M nitric acid, before it was finally recovered from the column by elution with 3 ml 1M hydrochloric acid.

17. The eluate was next evaporated almost to dryness and a few drops of 8M nitric acid were added. Suitable aliquots of this solution were then loaded onto source beads for the mass spectrometer such that the plutonium was distributed between five or more beads. Mass spectrometric analyses were then carried out as soon as possible.

C. Cobalt and Silver Monitors

18. For the cobalt monitors approximately 1 cm lengths (weighing about 1 mg) of 0.005 in diameter pure metal wire were used. The silver monitors were approximately 1.5 cm lengths (weighing about 12 mg) of a 1 per cent by weight silver in aluminium alloy. The method employed to count these monitors, their purity and the method of interpreting measurements made with them are discussed elsewhere[15].

D. Irradiations

19. These were carried out in thirteen small aluminium cans[15] strung end to end to form a flexible chain such that their centres were 5.75 cm apart. Irradiation was for 48.35 days in the flux scanning tube of the B2 fuel element of the reactor PLUTO.

20. Numbering the cans from 1 to 13, can 13 being the lowest in the reactor, each can contained a cobalt and a silver-aluminium monitor. In addition, cans 1, 5, 9 and 13 contained the ^{240}Pu-^{242}Pu mixture ampoules, cans 3, 7 and 11 contained the ^{239}Pu-^{242}Pu mixture ampoules, whilst the remaining cans (i.e. 2, 4, 6, 8, 10 and 12) contained the ^{239}Pu-^{241}Pu

mixture ampoules.

21. After irradiation the cans were allowed to 'cool' for six months then, with the exception of can 7 which was damaged, they were opened for monitor counting and sample treatment.

CALCULATIONS AND RESULTS

22. In the following we use the first subscript 9, 0, 1 or 2 to imply ^{239}Pu, ^{240}Pu, ^{241}Pu or ^{242}Pu respectively, and the second subscript 0 to imply a sample which has <u>not</u> been irradiated; where the second subscript is missing we imply the sample has been irradiated. For example, $N_{1,0}$ and N_1 represent the number of atoms of ^{241}Pu in a sample before and after irradiation.

23. Isotopic ratios are defined as follows:-

$R_1 = N_{0,0}/N_{9,0}$, $R_2 = N_0/N_9$, $R_3 = N_{1,0}/N_{9,0}$, $R_4 = N_1/N_9$, $R_5 = N_{2,0}/N_{9,0}$,

$R_6 = N_2/N_9$, $R_7 = N_{0,0}/N_{2,0}$, $R_8 = N_0/N_2$, $R_9 = N_{9,0}/N_{2,0}$ and $R_{10} = N_{1,0}/N_{2,0}$.

For example, R_7 and R_8 are the ratios of the number of atoms of ^{240}Pu to the number of atoms of ^{242}Pu in a mixture, before and after irradiation respectively.

24. Cross sections are either written as σ or S, implying neutron capture or absorption. When surmounted by a circumflex the cross section applies to reactor and not to mono-energetic neutrons.

25. Based on measurements made with manganese and lutetium monitors in a similar flux scanning tube in PLUTO to the one employed here, the neutron temperature is assumed to be $90 \pm 10^{\circ}$ for all irradiated samples.

26. All measured atomic ratios involving ^{241}Pu were corrected for the radioactive decay of this nuclide from the time of measurement to the beginning of the irradiation (for unirradiated samples) or the end of the irradiation (for irradiated samples). For this purpose we have used a radioactive disintegration constant of $\lambda_1 = (1.428 \pm 0.006) \times 10^{-4}$ days^{-1}, which corresponds to a half life of 13.29 ± 0.06 years. This last figure was obtained as a mean of published values (see Table II).

^{240}Pu-^{242}Pu mixture

27. As a result of a large number of mass spectrometric analyses, R_7, R_9 and R_{10} for the ^{240}Pu-^{242}Pu samples were determined to be 1.3780 ± 0.0004, 0.01614 ± 0.00004 and 0.01065 ± 0.00002 respectively. Analyses of the irradiated samples were then used to calculate $\hat{\sigma}_o$ from the expression:-

$$\exp\left[(\hat{\sigma}_2 - \hat{\sigma}_o)i\right] = \frac{R_8}{R_7}\left[1 + \frac{R_8 p - q}{R_8 \exp(-\hat{\sigma}_2 i)}\right] \qquad (3)$$

in which p and q are correction factors involving R_7, R_9 and R_{10}, which make allowance for the production of ^{242}Pu and ^{240}Pu respectively by neutron capture from the other plutonium isotopes in the mixture; p and q were obtained by interpolation from measurements of these rates in adjacent tubes, after taking into account spectrum differences between them. The symbol i in equation (3) represents the neutron dose (i.e. $i = (nv_o)t$, the product of the mean neutron flux and the time of irradiation) received by the sample. The effective capture cross section of ^{242}Pu was calculated from the expression:-

$$\hat{\sigma}_2 \text{ (barns)} = 19.8 + 1520r \qquad (4)$$

which uses the data of Table I.

28. Having determined the effective capture cross section of ^{240}Pu at the four positions 1, 5, 9 and 13 in the chain of samples, the same quantity in the other positions can be determined by interpolation, after making allowances for changes in neutron spectrum. Reference to Table I will show however that ^{240}Pu has a very large resonance capture integral with the result that $\hat{\sigma}_o$ is very sensitive to changes in r. The derived value of $\hat{\sigma}_o$ can therefore also be used to deduce the epithermal index by means of the expression:-

$$\hat{\sigma}_o \text{ (barns)} = 281(1.0488 + 37.62r) \qquad (5)$$

which is based on data given in Table I and the g value derived by WESTCOTT[20].

29. Details of the results for the ^{240}Pu-^{242}Pu samples are summarised in Table III. In Table IV we compare r values obtained using cobalt monitors and either (a) silver monitors or (b) the ^{240}Pu-^{242}Pu mixture. It will be

seen that the two sets are in agreement within the limits of uncertainty but that the results using silver monitors are always the smaller of the two. We have adjusted values of r obtained for other positions in the chain of samples, where (due to considerations of space) it has not been possible to include ^{240}Pu-^{242}Pu samples as well as silver monitors, to allow for this difference.

^{239}Pu-^{242}Pu mixture

30. For this mixture R_1, R_3 and R_5 were determined to be 0.01734 ± 0.00002, 0.007151 ± 0.000014 and 1.0357 ± 0.0007 respectively. From the results of the samples irradiated in positions 3 and 11 of the string, the quantity $\exp[(\hat{\sigma}_2 - \hat{S}_9)i]$ is obtainable directly from the expression:-

$$\exp[(\hat{\sigma}_2 - \hat{S}_9)i] = R_5/R_6 + (a+b+c)/R_6\exp(-\hat{\sigma}_2 i) \qquad (6)$$

in which a, b and c are terms involving R_1 and R_3 which allow for the production of ^{242}Pu from ^{241}Pu, ^{240}Pu and ^{239}Pu respectively during the irradiation. Details of the calculations are given in Table V.

31. The quantity $\exp[(\hat{\sigma}_2 - \hat{S}_9)i]$ has been obtained for other positions, where it has not been measured directly, by the following procedure. Values for all positions have been calculated from the measured neutron doses (and epithermal indices) and values of $\hat{\sigma}_2$ and $\hat{S}_9$ calculated from equation (4) and the expression

$$\hat{S}_9(\text{barns}) = 1157.9 + 2809r \qquad (7)$$

which is based on the data of Table I and CRITOPH'S tables[21]. The resulting values for positions 3 and 11 are given at the bottom of Table V where it will be observed that the values obtained by experiment are 1.06 per cent and 4.58 per cent respectively higher than the calculated values. Calculated values for other positions have therefore been corrected by the mean difference, i.e. by +(2.82 ± 1.76) per cent, to take this discrepancy into account. Having deduced $\exp[(\hat{\sigma}_2 - \hat{S}_9)i]$ for the other positions in the chain, $\exp(-\hat{S}_9 i)$ and $\hat{S}_9$ can be deduced also when required.

32. Results for the ^{239}Pu-^{242}Pu mixture can also be used to determine $\hat{\sigma}_9$ by means of the expression

$$\hat{S}_9/\hat{\sigma}_9 = (1-\exp(-\hat{S}_9 i))/[(1+B)(R_2\exp(-\hat{S}_9 i) - R_1\exp(-\hat{\sigma}_o i))] \qquad (8)$$

in which[7]

$$B = \hat{\sigma}_o i \frac{[\frac{1}{2} - (\hat{S}_9 i + \hat{\sigma}_o i)/6 + ((\hat{S}_9 i)^2 + (\hat{\sigma}_o i)^2 + (\hat{S}_9 i)(\hat{\sigma}_o i))/24 \ldots]}{[1 - (\hat{S}_9 i + \hat{\sigma}_o i)/2 + ((\hat{S}_9 i)^2 + (\hat{\sigma}_o i)^2 + (\hat{S}_9 i)(\hat{\sigma}_o i))/6 \ldots]} \quad (9)$$

Details are given in Table VI.

33. The data of Table I and CRITOPH'S tables[21] give

$$\hat{\sigma}_9(\text{barns}) = 334.8 + 1215r \quad (10)$$

Values calculated from this expression for positions 3 and 11 are given at the bottom of Table VI where it will be observed that the experimental values differ from the calculated values by + 1.8 per cent and -2.1 per cent respectively. For the other positions in the chain $\hat{\sigma}_9$ has been obtained by calculating this quantity by means of equation (10) then applying a correction of -(0.2 ± 2.0) per cent to allow for these discrepancies.

<u>^{239}Pu-^{241}Pu mixture</u>

34. For these we determined R_1, R_3 and R_5 to be 0.0463 ± 0.0010, 2.9307 ± 0.0046 and 0.02275 ± 0.00004 respectively. From the results for the irradiated samples we determined $\exp(-\hat{S}_1 i - \lambda_1 t)$ by means of the expression:-

$$R_3 \exp(-\hat{S}_1 i - \lambda_1 t) = R_4 . \exp[(\hat{\sigma}_2 - \hat{S}_9) i] . \exp(-\hat{\sigma}_2 i) - d - h \quad (11)$$

in which $\exp[(\hat{\sigma}_2 - \hat{S}_9) i]$ was obtained as described above, and d and h are terms which correct for the formation of ^{241}Pu from ^{240}Pu and ^{239}Pu and are given by

$$d = R_1 \hat{\sigma}_o [\exp(-\hat{\sigma}_o i) - \exp(-\hat{S}_1 i - \lambda_1 t)] / (\hat{S}_1 + \lambda_1/(nv_o) - \hat{\sigma}_o) \quad (12)$$

and

$$h = \hat{\sigma}_9 \hat{\sigma}_o \left[\begin{array}{l} \exp(-\hat{S}_9 i)/(\hat{S}_9 - \hat{\sigma}_o)(\hat{S}_9 - \hat{S}_1 - \lambda_1/(nv_o)) \\ + \exp(-\hat{\sigma}_o i)/(\hat{S}_1 + \lambda_1/(nv_o) - \hat{\sigma}_o)(\hat{S}_9 - \hat{\sigma}_o) \\ - \exp(-\hat{S}_1 i - \lambda_1 t)/(\hat{S}_9 - \hat{S}_1 - \lambda_1/(nv_o))(\hat{S}_1 + \lambda_1/(nv_o) - \hat{\sigma}_o) \end{array} \right] \quad (13)$$

The calculations are summarised in Table VII, those for position 10 are not included as the sample was inadvertently contaminated after opening.

Finally $\hat{S}_1/\hat{\sigma}_1$ was determined by means of the expression

$$\hat{S}_1/\hat{\sigma}_1 = \frac{R_3(1 - \exp(\lambda_1 t) . \exp(-\hat{S}_1 i - \lambda_1 t))}{(1+E)(R_6 \exp(-\hat{S}_9 i) - R_5 \exp(-\hat{\sigma}_2 i) - j - k)} \quad (14)$$

in which E is given by

$$E = \hat{\sigma}_{2i} \frac{[\frac{1}{2} - ((\hat{S}_1 i+\lambda_1 t) + \hat{\sigma}_2 i)/6 + ((\hat{S}_1 i+\lambda_1 t)^2 + (\hat{\sigma}_2 i)^2 + (\hat{\sigma}_2 i)(\hat{S}_1 i+\lambda_1 t))/24 \ .....]}{[1 - ((\hat{S}_1 i+\lambda_1 t) + \hat{\sigma}_2 i)/2 + ((\hat{S}_1 i+\lambda_1 t)^2 + (\hat{\sigma}_2 i)^2 + (\hat{\sigma}_2 i)(\hat{S}_1 i+\lambda_1 t))/6 \ .....]} \quad (15)$$

and j and k are terms which correct for the formation of ^{242}Pu during the irradiation from the original ^{240}Pu and ^{239}Pu contents of the mixture, and are given by

$$j = R_1 \hat{\sigma}_o \hat{\sigma}_1 \left[\begin{array}{l} \exp(-\hat{\sigma}_o i)/(\hat{\sigma}_2-\hat{\sigma}_o)(\hat{S}_1+\lambda_1/(n v_o) - \hat{\sigma}_o) \\ + \exp(-\hat{\sigma}_2 i)/(\hat{\sigma}_o-\hat{\sigma}_2)(\hat{S}_1+\lambda_1/(n v_o) - \hat{\sigma}_2) \\ + \exp(-\hat{S}_1 i-\lambda_1 t)/(\hat{S}_1+\lambda_1/(n v_o) - \hat{\sigma}_2)(\hat{S}_1+\lambda_1/(n v_o)-\hat{\sigma}_o) \end{array} \right] \quad (16)$$

$$\text{and } K = \hat{\sigma}_9 \hat{\sigma}_o \hat{\sigma}_1 \left[\begin{array}{l} \exp(-\hat{S}_9 i)/(\hat{S}_1+\lambda_1/(n v_o) - \hat{S}_9)(\hat{S}_9-\hat{\sigma}_o)(\hat{S}_9-\hat{\sigma}_2) \\ -\exp(-\hat{\sigma}_o i)/(\hat{S}_1+\lambda_1/(n v_o)-\hat{\sigma}_o)(\hat{S}_9-\hat{\sigma}_o)(\hat{\sigma}_o-\hat{\sigma}_2) \\ -\exp(-\hat{S}_1 i-\lambda_1 t)/(\hat{S}_1+\lambda_1/(n v_o)-\hat{\sigma}_2)(\hat{S}_1+\lambda_1/(n v_o)-\hat{S}_9)(\hat{S}_1+\lambda/(n v_o)-\sigma_o) \\ +\exp(-\hat{\sigma}_2 i)/(\hat{S}_9-\hat{\sigma}_2)(\hat{\sigma}_o-\hat{\sigma}_2)(\hat{S}_1+\lambda_1/(n v_o)-\hat{\sigma}_2) \end{array} \right] \quad (17)$$

DERIVATION OF α_o AND ERRORS

36. To correct the values of $\hat{\alpha}$ for ^{241}Pu given in Table VIII to α_o, by means of expression (2), we have taken WESTCOTT'S published values for s for this nuclide[20], and have adjusted them to the more recent resonance integral data given in Table I, and a neutron temperature of 90° (using a mean of s_2 and s_4). This gives s values of 0.8797 and 0.8540 respectively for neutron fission and absorption.

37. WESTCOTT[14] has also caloulated g values based on the latest differential cross section data available; his values for fission and absorption appropriate to 90° are 1.080 ± 0.006 and 1.0658 ± 0.0030 respectively. Using these data we have derived the values of α_o given in Table IX; they give a mean value of 0.390 ± 0.007 (standard error).

38. The most important source of possible systematic error in deducing $\hat{S}_1/\hat{\sigma}_1$ arises from uncertainty in the term $\exp[(\hat{\sigma}_2-\hat{S}_9)i]$, which we take to be ±1.8 per cent (see above); other possible errors are estimated to be

± 14 per cent in $\hat{\sigma}_2$, ± 10 per cent in r, ± 5 per cent in i and ± 0.2 per cent in isotopic ratios. Taking these into account we estimate a total possible error of ± 4 per cent in $\hat{S}_1/\hat{\sigma}_1$, which is equivalent to an uncertainty of ± 5.5 per cent in $\hat{\alpha}$.

39. To summarise, we consider possible sources of error in α_o may arise as follows:-

from the determination of $\hat{\alpha}$	± 5.5 per cent
from the random error in α_o	± 1.9 per cent
from uncertainty in g_f	± 0.5 per cent
from uncertainty in s_a and s_f	± 0.4 per cent
from uncertainty in g_a	± 0.3 per cent
from mass discrimination in the mass spectrometer	± 0.1 per cent
from uncertainties in T, r and the epithermal cut-off function	< 0.1 per cent

giving a quadrature sum of ± 5.9 per cent. Our final value of α_o is therefore 0.390 ± 0.023.

DISCUSSION

40. As was stated in the preamble to this report, the only published data which leads directly to alpha for ^{241}Pu (by FIELDS et al[4]) results in a value for $\hat{\alpha}$ of 0.37 ± 0.10; the mean value of 0.371 ± 0.016 which we have obtained for this quantity (Table IX), is in excellent agreement with this figure.

41. No measurements of α_o have yet been published. WESTCOTT et al[3] have obtained this, and other nuclear parameters for ^{241}Pu, indirectly, by making a least squares fit of all the relevant nuclear data available for this nuclide, and all the relationships existing between them. In this computation they concluded that absolute measurements of the individual parameters were so meagre and imprecise that they had no significant effect on the final results; only comparisons (e.g. of the fission cross sections and $\bar{\nu}$) with the values for other fissile nuclides had any appreciable weight. The final value obtained by WESTCOTT et al for α_o was 0.359 ± 0.025 (the error of which should probably now be increased to about ± 0.037[9]) a result which is even at present in agreement with our value of 0.390 ± 0.023.

It is clear however that when our results are taken into account also, the discrepancy between the two figures will be considerably reduced.

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TABLE I

2200 m/sec NEUTRON CROSS SECTIONS AND RESONANCE INTEGRALS (ABOVE 0.45 eV) OF PLUTONIUM ISOTOPES

	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
2200 m/sec cross section, in barns				
for absorption[9]	1008.4 ± 4.9	-	1389.1 ± 20.0	-
for fission[9]	742.7 ± 3.6	<0.1	1006.1 ± 7.2	<0.2
for capture[9,10,11]	265.7 ± 3.7	281 ± 7	382.9 ± 18.8	19.8 ± 1.1
Resonance integral (above 0.45eV), in barns				
for absorption[13]	519 ± 15	-	707	-
for fission[12]	324 ± 9	negligible	541 ± 14	negligible
for capture[10,13]	195 ± 12	8453 ± 600	166	1220 ± 200

TABLE II

THE HALF-LIFE OF ^{241}Pu

Reference	Value*
SMITH (1961)[16]	13.3 ± 0.3 years
BROWN et al (1960)[17]	13.33 ± 0.24 years
MACKENZIE et al (1953)[18]	13.32 ± 0.12 years
ROSE et al (1956)[19]	13.04 ± 0.28 years
Mean	13.29 ± 0.06 years

*All values corrected to a half-life of 458.1 ± 0.5 years for ^{241}Am.

TABLE III

RESULTS FOR ^{240}Pu-^{242}Pu MIXTURES

(EQUATION 3)

	POSITION			
	1	5	9	13
R_8	1.0708	0.8098	0.7800	0.9677
p	0.0185	0.0461	0.0502	0.0377
q	0.0016	0.0018	0.0019	0.0021
$\hat{\sigma}_2$(barns)	75	140	136	69
i(n/cm^2)	3.62×10^{20}	4.51×10^{20}	4.96×10^{20}	5.58×10^{20}
$\hat{\sigma}_o$(barns)	723	1217	1182	637
r (from $\hat{\sigma}_o$)	0.041	0.087	0.084	0.032

TABLE IV

A COMPARISON OF EPITHERMAL INDEX DETERMINED USING COBALT AND (A) SILVER MONITORS OR (B) ^{240}Pu MONITORS

Position in flux scanning tube	EPITHERMAL INDEX	
	(A) Using Silver Monitors	(B) Using ^{240}Pu Monitors
1	0.031 ± 0.004	0.041 ± 0.006
5	0.073 ± 0.009	0.087 ± 0.010
9	0.071 ± 0.009	0.084 ± 0.010
13	0.030 ± 0.004	0.032 ± 0.005

TABLE V

DETERMINATION OF $Exp[(\hat{\sigma}_2-\hat{S}_9)i]$ (EQUATION 6)

	POSITION	
	3	11
R_6	1.6210	1.7501
a	0.0007	0.0008
b	0.0004	0.0005
c	0.0011	0.0018
$\hat{\sigma}_2$(barns)	122	119
$i(n/cm^2)$	3.73×10^{20}	4.63×10^{20}
$exp[(\hat{\sigma}_2-\hat{S}_9)i]$ by experiment	0.6404	0.5937
$exp[(\hat{\sigma}_2-\hat{S}_9)i]$ by calculation	0.6337	0.5677

TABLE VI

DETERMINATION OF $\hat{\sigma}_9$ (EQUATION 8)

	POSITION	
	3	11
R_2	0.1841	0.2154
$exp(-\hat{S}_9 i)$	0.6119	0.5618
$exp(-\hat{\sigma}_o i)$	0.6692	0.6162
B	0.234	0.291
$\hat{S}_9/\hat{\sigma}_9$	3.111	3.076
$\hat{\sigma}_9$(barns) by experiment	423.7	404.6
$\hat{\sigma}_9$(barns) by calculation	416.2	413.7

TABLE VII

DETERMINATION OF $Exp(-\hat{S}_1 i-\lambda_1 t)$ (equation 11)

	POSITION				
	2	4	6	8	12
R_4	2.799	2.814	2.868	2.814	2.706
$\exp[(\hat{\sigma}_2-\hat{S}_9)i]$	0.6553	0.6169	0.5676	0.5512	0.5679
$\hat{\sigma}_2$(barns)	97	138	141	141	90
$\hat{\sigma}_o$(barns)	894	1205	1228	1227	811
$i(n/cm^2)$	3.74×10^{20}	4.12×10^{20}	4.79×10^{20}	5.02×10^{20}	4.96×10^{20}
d	0.010	0.013	0.015	0.015	0.011
h	0.016	0.026	0.033	0.035	0.022
$\exp(-\hat{S}_1 i-\lambda_1 t)$	0.5946	0.5462	0.5029	0.4761	0.4902

TABLE VIII

DETERMINATION OF $\hat{S}_1/\hat{\sigma}_1$ AND $\hat{\alpha}$ (equations 14 and 1)

	POSITION				
	2	4	6	8	12
$\exp(-\hat{S}_1 i-\lambda_1 t)$	0.5946	0.5462	0.5029	0.4761	0.4902
$\exp(\lambda_1 t)$	1.00693	1.00693	1.00693	1.00693	1.00693
E	0.0199	0.0318	0.0382	0.0405	0.0251
$R_6 \exp(-\hat{S}_9 i)$	0.3301	0.3708	0.4196	0.4070	0.4182
$R_5 \exp(-\hat{\sigma}_2 i)$	0.0219	0.0215	0.0213	0.0212	0.0218
j	0.0008	0.0013	0.0017	0.0017	0.0012
k	0.0008	0.0016	0.0025	0.0026	0.0016
$\hat{S}_1/\hat{\sigma}_1$	3.761	3.690	3.535	3.843	3.679
$\hat{\alpha}$	0.362	0.372	0.395	0.352	0.373

TABLE IX

DERIVATION OF a_o (equation 2)

Position	r	$\hat{a}$	(g_f+rs_f)	(g_a+rs_a)	a_o
2	0.051	0.362	1.125	1.1094	0.381
4	0.078	0.372	1.149	1.1324	0.391
6	0.080	0.395	1.150	1.1341	0.414
8	0.080	0.352	1.150	1.1341	0.371
12	0.046	0.373	1.121	1.1051	0.393

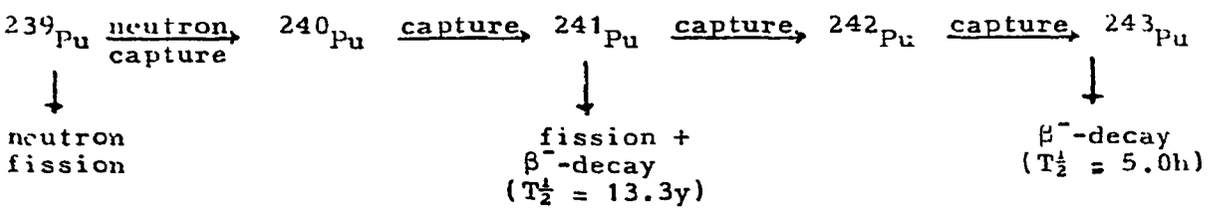

FIG. 1. THE CHIEF REACTIONS OCCURRING WHEN A MIXTURE OF PLUTONIUM ISOTOPES IS IRRADIATED WITH NEUTRONS