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INTEGRAL EXPERIMENTS ON FAST SYSTEMS OF PLUTONIUM, URANIUM AND THORIUM*

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Abstract — Résumé — Аннотация — Resumen

Integral experiments on fast systems of plutonium, uranium and thorium. The paper describes two types of integral experiment which have been undertaken to provide experimental checks on nuclear data and methods of calculation.

The first consists of a cylindrical assembly of material with a point source of DD or DT neutrons at its centre. The neutron flux and various reaction rates in the cylinder material are measured as a function of position. Results obtained are compared with calculation. Experimental results for natural uranium and thorium are presented.

The second type of experiment is a low-power experimental reactor of simple regular geometry and approximately homogeneous composition. Holes can be provided as required for the insertion of counters or perturbation samples. The core composition is adjusted by the addition of a moderator to provide a neutron spectrum which tests the nuclear data in the energy region of interest. Measurements are made of the neutron-energy spectrum and various reaction rates and perturbation cross-sections as a function of position in the reactor and the results are compared with computer calculations. Results are presented for a system consisting of a core of of U^{235} and graphite surrounded by a natural-uranium reflector.

Expériences intégrales sur réacteurs à neutrons rapides de plutonium, d'uranium et de thorium. Les auteurs décrivent deux types d'expériences intégrales visant à contrôler expérimentalement des données nucléaires et des méthodes de calcul.

Le premier dispositif expérimental est un empilement cylindrique de matériau au centre duquel est placée une source ponctuelle de neutrons DD ou DT. On mesure le flux de neutrons et l'intensité de diverses réactions dans le matériau, en fonction de la position. On compare les résultats des mesures à ceux des calculs. Le mémoire donne les résultats des expériences effectuées avec de l'uranium naturel et du thorium.

Le deuxième dispositif est un réacteur expérimental de faible puissance, ayant une géométrie régulière simple et une composition sensiblement homogène. On peut le munir, selon les besoins, de canaux pour l'introduction de compteurs ou d'échantillons de matériaux perturbateurs. On règle la composition du cœur par l'adjonction d'un ralentisseur, afin d'obtenir un spectre de neutrons tel que le contrôle des données nucléaires s'effectue dans la zone d'énergie voulue. On mesure le spectre d'énergie des neutrons, ainsi que l'intensité de diverses réactions et les sections efficaces de perturbation, en fonction de la position dans le réacteur. On compare les résultats à ceux qui sont donnés par des calculatrices. Le mémoire donne les résultats obtenus avec un réacteur ayant un cœur composé de ^{235}U et de graphite, entouré d'un réflecteur en uranium naturel.

Интегральные эксперименты с системами на быстрых нейтронах с плутонием, ураном и торием. В докладе дается описание двух экспериментальных установок, созданных для экспериментальной проверки ядерных данных и вычислительных методов.

Первая установка состоит из сборки материала цилиндрической формы с точечным источником нейтронов DD или DT расположенным в ее центре. Поток нейтронов и различные скорости реакции в цилиндре измеряются как функция положения. Полученные

* Including work by L. R. Day, J. R. Dominey, H. Goodfellow, M. H. McTaggart, H. Shieff, A. F. Thomas, K. A. Wallace.

результаты сравниваются с расчетом. Даются экспериментальные результаты для природного урана и тория.

Вторым типом установки является экспериментальный реактор низкой мощности с простой обычной геометрией и приблизительно гомогенным составом. Реактор может иметь каналы, необходимые для введения счетчиков или возмущающих образцов. Состав активной зоны регулируется добавлением замедлителя с тем, чтобы иметь спектр нейтронов, необходимый для проверки ядерных данных в интересующей области энергии. Измеряются энергетический спектр нейтронов, различные скорости реакции и изменение поперечного сечения как функции положения в реакторе, и результаты сравниваются с расчетами на вычислительных устройствах. Даются результаты для системы, состоящей из активной зоны из урана-235 и графита, окруженной отражателем из природного урана.

Experimentos integrales realizados con reactores rápidos de plutonio, uranio y torio. Los autores describen dos tipos de experimentos integrales que se han llevado a cabo con el propósito de comprobar experimentalmente determinados datos nucleares, así como la exactitud de los métodos de cálculo empleados.

El primer dispositivo experimental consiste en un montaje cilíndrico en cuyo centro se halla una fuente puntual de neutrones DD o DT. El flujo neutrónico y las diversas velocidades de reacción en el material del cilindro se miden en función de la posición de la fuente y los resultados así obtenidos se comparan con los calculados. La memoria presenta los resultados de experimentos efectuados con uranio natural y con torio.

Para el segundo tipo de determinaciones, se utiliza un reactor experimental de baja potencia, con una geometría regular sencilla y una composición sensiblemente homogénea. Los orificios practicados en el mismo pueden utilizarse para introducir, conforme sea necesario, contadores o muestras perturbadoras. La composición del cuerpo se regula mediante la adición de un moderador, a fin de disponer de un espectro neutrónico que permita comprobar los datos nucleares en la región energética que interesa. Se realizan mediciones del espectro de energías neutrónicas y de diversas velocidades de reacción y secciones eficaces de perturbación en función de la posición en el reactor y los resultados se comparan con los obtenidos con calculadoras. La memoria da los resultados correspondientes a un sistema que consiste en un cuerpo de ^{235}U y grafito, rodeado de un reflector de uranio natural.

Introduction

A broad programme of research is in progress at Aldermaston to improve our understanding of the neutron physics of fast systems. Its primary aim is to reduce the errors and uncertainties which arise in the present calculations of nuclear characteristics for fast-reactor systems. These errors arise mainly from the inadequacy of the basic nuclear data, and the work therefore centres on an attempt to improve these data and to establish a single set of data for fast-reactor materials which is sufficiently accurate over the whole energy range of interest. The overall programme comprises three interrelated areas of work:

(*a*) Measurements of important nuclear cross-sections and parameters;

(*b*) Compilation of nuclear-data sets using results of (*a*) and other available data, and the development of methods of theoretical analysis of the nuclear characteristics of fast systems; and

(*c*) Checks of the accuracy of the nuclear data sets and system analysis against integral experiments.

Aspects of the data measurements and of the theoretical analysis are described in other Aldermaston papers presented at this Seminar. The present paper will describe some relevant integral experiments.

First of all, however, it is instructive to indicate, by reference to some of our problems at Aldermaston during the past two years, how the three programmes relate one to another and how they help to establish which microscopic

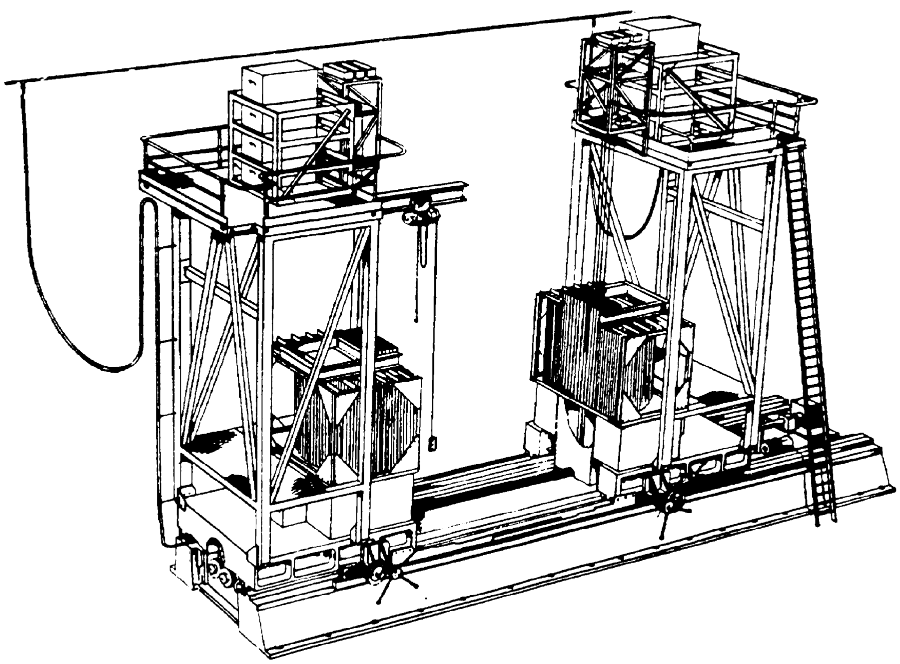

Fig. 1
Versatile experimental reactor essembly (VERA).

data need to be checked and which integral experiments are essential. During 1959 a literature review of nuclear data was made and a set of nuclear data established by selecting "best values" from the published microscopic data. These data were checked by calculating critical masses for a number of Pu- and highly-enriched U-systems, both bare and reflected by various thicknesses of natural U. The calculated critical masses were in all cases about 20% smaller than the experimental values. About this time a direct experimental measurement of ν for U^{235} suggested a downward revision of this quantity. A low ν value was therefore adopted for the U^{235} calculations and a corresponding reduction was applied for Pu^{239}. Good agreement with experimental critical masses for fast concentrated systems was then obtained and this set of nuclear data was therefore adopted for general use [1]. Nevertheless, the need for further measurements of ν and further checks against integral experiments was recognized and new work was put in hand. Later it became clear that the low ν value was incorrect and that the adopted nuclear-data set was less accurate for U-systems of lower enrichment. New experimental data had also accumulated and it was decided to make a complete revision of data for U^{235}, U^{238} and Pu^{239}. As an interim measure an attempt was made to adjust the nuclear data empirically to fit integral results over a wider range of U concentration. This was done [2] by making plausible adjustments to the values of ν and σ_f for U^{235} and to ν and σ (n, γ) for U^{238}. The procedure was successful, reducing the critical-mass errors to $\pm 3\%$ over the concentration range 29% to 93% U^{235}. It is not satisfactory, however, to rely permanently on this expedient. New microscopic measurements and integral checks for the revised data are evidently required and these are progressing in parallel with the detailed revision of the basic nuclear-data set. Initial results for ν of U^{235} are reported by MOAT [3], and initial results of a new U^{235} integral experiment are reported as part of the present paper.

Our integral experiments are of three types. The first is the determination of critical mass by extrapolation of multiplication measurements made on a critical assembly machine; the second is the analysis of neutronic processes in large blocks of material in which a strong compact neutron source is located; the third is the detailed study of neutron reactions in a simple system operated as a zero-energy reactor. Some of the critical-mass measurements were done primarily to provide data for safety assessment; the other experiments were designed specifically for testing nuclear data. All the experiments have simple regular geometry and uniform composition, so that corrections relating the actual experimental system to the calculated model are minimized.

1. Critical mass measurements for fast systems

A number of critical-mass measurements have been made for plutonium metal in spherical geometry and for 45.5% enriched U metal in slab geometry.

The Pu measurements are for a bare sphere and for spheres reflected by natural uranium, pile-quality graphite, steel or water. The results are based on central-source multiplication measurements using Pu spheres of 4.5, 6, 7.5 and 9 kg and various thicknesses of each reflecting material. Each Pu sphere had a 2.16-cm-diameter spherical cavity at the centre to accommodate the neutron source used for multiplication measurements. The plutonium metal density was 15.64 g/cm^3 and the metal in the form of two hemispheres was clad in copper foil. 0.010 in thick on the curved surfaces and 0.005 in thick on the flat surface.

Calculations for these systems have been made using the nuclear data of reference [1] in a multi-group Carlson S_8 code. Seven energy groups from 10 keV to 11 MeV were used for the Fe- and U-reflected systems and 13 energy groups from thermal to 11 MeV for the graphite-reflected system. The results are compared with experimental values for a number of reflector thicknesses in Table I.

TABLE I

COMPARISON OF CALCULATED AND EXPERIMENTAL CRITICAL MASSES FOR Pu SYSTEMS

Reflector	Reflector thickness (in)	Experimental crit. mass (kg Pu)	Calculated crit. mass (kg Pu)
None	—	16.8	17.0
Natural U (18.7 g/cm^3)	2	8.5 ± 0.1	8.10
	3	7.4 ± 0.1	7.19
	6	6.3 ± 0.1	6.31
Fe (7.87 g/cm^3)	2	10.9	10.3
	4	9.6 ± 0.1	8.80
	6	9.1 ± 0.1	8.11
C (1.61 g/cm^3)	2	11.2	11.0
	4	9.2 ± 0.1	9.39
	6	8.2 ± 0.1	8.62

The experimental critical masses are derived from graphical extrapolations of graphs relating central-source multiplication to reflector thickness and the errors indicated are estimates of the probable errors of the extrapolations. In three cases the extrapolation was rather long (the largest available sphere being 9 kg) and no error is quoted for these. The result for the bare sphere was obtained by analytical extrapolation of a least-squares fit to a diffusion-theory approximation.

It should be noted that data used in the calculations are "best values" except for ν, for which the low value of ν (Pu^{239})$=2.82+0.105\,E$ was chosen in order to obtain agreement between calculation and experiment for the bare and thick-U-reflected systems. For graphite the agreement is reasonably good but for Fe the calculated values are too small by amounts ranging from 6% to 11%. Taken with the fact that the ν value should probably be higher, the latter result suggests that the Fe scattering data need checking.

The U-slab experiments were done specifically for criticality safety assessment, to provide data against which calculations of fast-reactor melt-down configurations could be checked. Measurements of critical size were made for bare slabs and for slabs reflected by natural uranium, graphite, steel and aluminium respectively. The reflector covered both faces and edges of the slabs. The slabs were constructed of machined blocks in the form of hexagons $2\frac{3}{4}$-in across the flats and of various thicknesses. These were assembled into pseudo-cylindrical assemblies with a neutron source located at the centre, and measurements of the central-source multiplication were made as a function of thickness of the enriched-U slab. Critical thicknesses were obtained by graphical extrapolation. The results are listed in Table II.

TABLE II

RESULTS OF U SLAB EXPERIMENTS

Reflector	Critical thickness of enriched U (in)		
	11.6 in diam.	16.9 in diam.	22.1 in diam.
None	7.26 ± 0.03	5.47 ± 0.04	—
Nat. U (6-in thick, 16.3 g/cm³)	3.27 ± 0.02	2.36 ± 0.02	2.01 ± 0.02
Graphite (6-in thick, 1.74 g/cm³)	3.00 ± 0.02	2.14 ± 0.02	1.78 ± 0.02
Steel (6-in thick, 7.24 g/cm³)	4.12 ± 0.03	2.98 ± 0.02	2.57 ± 0.02
Aluminium (6-in thick, 2.58 g/cm³)	4.40 ± 0.02	3.21 ± 0.02	2.72 ± 0.02

The diameter indicated is that of the enriched-U slab. The natural-U reflector was solid for the first inch on the flat faces of the slab; the remainder of the reflector was a 5.7-in layer of $1\frac{1}{8}$-in rods at a packing density of 0.85. The effective density of the enriched-U slabs was 18.44 g/cm³.

These experimental results have been used to check the adjusted set of nuclear data [2] for U^{235} and U^{238}. Calculations were carried out using a two-dimensional Carlson code (S_4) with 13 energy groups from thermal to 11 MeV. The results are listed in Table III.

TABLE III

COMPARISON OF CALCULATED AND EXPERIMENTAL CRITICAL THICKNESSES FOR 45.5% U SLABS (inches)

Reflector / Slab diameter	None		Graphite 6-in thick all round	
	Exp.	Calc.	Exp.	Calc.
11.6	7.26	6.99	3.00	3.18
16.9	5.47	5.43	2.14	2.31
22.1	—	—	1.78	1.95

Agreement is good for the bare systems but for the graphite-reflected systems errors range from 6% to 10%, the calculated results being too large. This discrepancy is ascribed to errors in U^{238} data at the lower neutron energies produced by the graphite reflector.

2. Accelerator-driven integral experiments with uranium and thorium

The neutron balance in a large cylindrical pile of material with a 14-MeV neutron source at its centre has been studied experimentally for natural uranium metal and for thorium metal. (A full account of the U experiment is given in a recent paper [4]). The neutron source was a Zr-H^3 target located at the centre of the cylinder and bombarded by a beam of deuterons brought in through a drift-tube from a 200-kV Cockcroft-Walton accelerator. Each pile was made of vertical rods packed closely together, and it was possible to remove a rod from any position to provide access for a counter to measure reaction rates and neutron spectra as a function of position in the pile. Copper cylinders were inserted and irradiated to measure the 14-MeV neutron flux by the Cu^{63} (n, 2n) threshold reaction; fission rates were measured by inserting fission chambers loaded with the appropriate material; and neutron-capture rates were measured by irradiating small pellets of U or Th metal and separating and counting the Pa^{233} or Np^{239} activity. All the measurements were related to the strength of the central 14-MeV neutron source which was continuously monitored by counting the associated α-particles emitted into a known solid angle at 177° to the deuteron beam. The total number of neutrons of all energies escaping from the pile for each central-source neutron was measured by means of a "long" counter mounted outside the system, its position being varied to take account of the variation of the emergent flux with angle. The fission and capture rate results were integrated to determine the total number of fissions and captures in the pile per central-source neutron.

The way in which these results can be used to draw up a detailed neutron balance is described (for the U pile) in reference [4]. Recently the results have been used to check the accuracy of nuclear data for natural U in the fast-neutron energy range. Two Monte Carlo calculations have been made by PARKER and MERCER [5] using first the nuclear data of reference [1] and secondly the same data with the original (unadjusted) values for ν. The results of these calculations are compared with the experimental results in Table IV.

TABLE IV

EXPERIMENTAL AND CALCULATED VALUES FOR THE NUMBER OF REACTIONS PER 14-MeV CENTRAL-SOURCE NEUTRON IN THE U PILE*

Reaction	Experiment	Calculation (A)	Calculation (B)
U^{238} fission	1.18 ± 0.06	0.78	0.86
U^{235} fission	0.28 ± 0.02	0.20	0.23
U^{238} capture.........	4.08 ± 0.24	3.45	3.96
Leakage.............	0.41 ± 0.02	0.53	0.52

* The effective pile dimensions were: height 112 cm, diameter 99 cm; the effective density was 16.3 g/cm³.

Calculation (A) (Table IV) used the data of reference [1], in which ν $(U^{235}) = 2.25 + 0.132\ E$ and ν $(U^{238}) = 2.32 + 0.129\ E$.

Calculation (B) (Table IV) used the same data except that the values were taken as ν $(U^{235}) = 2.46 + 0.144\ E$ and ν $(U^{238}) = 2.53 + 0.141\ E$.

Evidently the higher values of ν give better agreement with experiment, but there is a large error in the U^{238} fission calculation. This must be due to an error in the fission cross-section or in the inelastic scattering cross-section or energy distribution.

The Th pile results have not yet been compared with calculations but the experimental integral results are quoted (in Table V), as they provide useful data against which nuclear constants for this material can be checked.

TABLE V

EXPERIMENTAL INTEGRALS FOR THE Th PILE*

Reaction	Number of reactions per 14-MeV central-source neutron
Th (n,f) ...	0.174 ± 0.009
Th (n,γ) ...	1.51 ± 0.08
Leakage ..	0.69 ± 0.02

* The pile was 110 cm high, 96.5 cm in diameter and contained 9.70 g/cm³ Th and 0.13 g/cm³ Fe.

3. Experiments on fast critical systems

We have recently commissioned a new Versatile Experimental Reactor Assembly (VERA) and it is being used for studies of the physics of fast critical systems. The measurements are primarily intended to provide information for a systematic check of nuclear data in the energy range 1 keV to 1 MeV. For each assembly the neutron flux, neutron spectrum, fission and capture rates and perturbation cross-sections are measured as a function of position, and the fast chain-decay constant (Rossi-α) is measured in the region of delayed critical. Multi-group Carlson calculations are compared in detail with the experimental results and discrepancies are interpreted in terms of corresponding errors in group cross-sections. The first assembly, which is now under study, has a core of U^{235} and C surrounded by a reflector of natural U. The properties of this system provide a sensitive check of nuclear data for U^{235} and will be used to select (within experimental cross-section errors) a set of group constants for this nuclide.

VERA is constructed of square-section fuel boxes mounted vertically in a steel lattice plate. The core and the axial portions of the reflector are composed of small square plates or bricks of material and are loaded in the fuel boxes. The radial portion of the reflector consists of closely-packed round rods which surround the square fuel boxes and are located by holes drilled in the lattice plate. The assemblies are arranged as pseudo-cylindrical cores surrounded by cylindrical reflectors and the compositions of the axial and radial portions of the reflector are adjusted to be the same. Accurate calculations are thus possible by means of the two-dimensional Carlson codes. In these calculations the core is taken as a homogeneous mixture and the experimental results, which apply to mixtures of $\frac{1}{8}$-in-thick plates, therefore require a slight correction for the fuel "bunching" effect.

To ensure safety during loading and core modifications and to provide a second shut-down mechanism, the lattice plate is divided into two portions and the reactor is correspondingly divided in half. One half is mounted on a moveable table which rests on rails and can be driven along these rails by an electrically-driven leadscrew. Loading and core modifications are only carried out when the two halves of the reactor are separated by a distance of 5 ft. A number of safety and control elements are provided. These are normal fuel boxes loaded with core and blanket material and suspended by cables from winches on the reactor superstructure. They can be dropped below their normal positions in the reactor through holes provided in the lattice plate. The safety elements are fully raised into the core before the reactor halves are driven together; the control elements are raised as the last step in the approach to criticality. The reactor is not provided with local shielding but is located in a large room with thick concrete walls, and this room is sealed during reactor operation, the reactor then being controlled remotely. No cooling system is provided as it is not intended to operate the reactor at powers above 100 W.

The dimensions and composition of the U^{235}/graphite assembly (referred to as VERA Assembly 2) are as follows:

Core height (cm)		27.15
Core diameter (cm)		38.10
Core volume (l)		30.8
Reflector radial thickness (cm)		37.4
Reflector axial thickness (cm)		39.4
U^{235} content of core (kg)		88.96
U^{238} content of core (kg)		6.80
Stainless-steel content of core (kg)		23.44
Graphite content of core (kg)		35.55
Hydrogen content of core (g)		3.0 (this is the protective lacquer)
Composition of reflector:	U-natural (g/cm^3)	14.39
	Stainless steel (g/cm^3)	0.832

With all safety and control elements fully raised, the above system has an excess reactivity of 0.3%, i.e. 0.9% in core mass, assuming $\Delta K/K = \frac{1}{3}(\Delta M/M)$. Holes provided to accommodate counters at the core-reflector interface make the reactivity 0.67% less than it would be in ideal geometry. The bunching of the fuel into $\frac{1}{8}$-in plates makes the critical mass 2% smaller than it would be for a homogeneous system (reference [6]).

With these corrections the critical parameters for a homogeneous system of the above composition are:

Critical volume: 30.4 l
Critical mass: 87.8 kg U^{235}.

A two-dimensional Carlson calculation using the nuclear data of reference [1] in 13 energy groups predicts a critical mass of 92.1 kg U^{235} for this system.

Measurements of fission rates, perturbation cross-sections and the prompt chain-decay constant for this system are now in progress. Preliminary results of measurements at the core centre are listed in Table VI.

TABLE VI

MEASURED CROSS-SECTION RATIOS AT THE CENTRE OF THE VERA CORE

Fission ratios	Experimental results*	Calculated results**
$\frac{U^{238}}{U^{235}}$	0.077	0.074
$\frac{U^{236}}{U^{235}}$	0.162	0.153
$\frac{Pu^{239}}{U^{235}}$	1.18	1.12
$\frac{Np^{237}}{U^{235}}$	0.488	0.365
$\frac{U^{238}}{U^{235}}$ (capture)	0.121	0.138
Perturbation ratio $\frac{Pu^{239}}{U^{235}}$	1.72	1.53

* These results have a probable error of about 6%.

** The calculated values are from the two-dimensional Carlson calculations using data from reference [1].

Future work

During the next two years work will be mainly concentrated on the VERA programme. The next step after completion of the U^{235}/C studies will be to add U^{238} to the core, adjusting the C content so that the spectrum is not appreciably changed, and repeat the measurements and calculations, thus checking the nuclear data for U^{238}. Subsequent systems will include Pu^{239} and various diluent materials such as Fe, Al and Na, and small quantities of a moderating material will be added so that the neutron-energy spectrum is varied over the desired range. Some diluent materials of particular interest will be studied separately in accelerator-driven integral experiments of the type described for U and Th. A Na assembly is planned as the next in this series. When the revised nuclear-data sets are available, detailed analysis of the U and Th pile results will be carried out.

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