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AVERAGE EFFECTIVE CROSS SECTIONS FOR THE FAST PLUTONIUM REACTOR SPECTRUM

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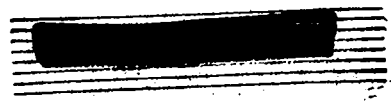
Average Effective Cross Sections for the Fast Plutonium Reactor Spectrum

The change of reactivity when foreign material is introduced into the center of the reactor has been used as a measure of the effective cross sections of that material for the neutron spectrum of the reactor. The present report covers the first of a planned series of measurements which will attempt to explore the dependence of absorption and scattering processes upon the energy spectrum and upon the geometry and dilution of the active material.

I. Experimental Basis

Material introduced into the active region of a nuclear reactor will affect the reactivity (effective k) of the reactor by reason of additional absorption, altering the quality of neutrons sustaining the reaction, or by changing the fraction of neutrons escaping by leakage. The leakage factor can be minimized by restricting the samples to a small volume in the center of the active region. Then, since isotropy exists throughout the central region, scattering processes which do not alter the neutron energy can be neglected.

Absorption of neutrons by elements introduced can be considered as either productive or destructive according to whether or not a neutron emerges from the reaction. Under this definition, productive absorption will include (n,f), (n,2n), (n,n), while destructive absorption will include (n,α),(n,p)



(n,γ) processes. The degree of productivity can be described by a number ν .

Scattering of neutrons (n,n), both elastic and inelastic, can further be considered as either increasing or decreasing the pile reactivity depending on whether $\nu \gtrless 1$. For example, elements of low atomic mass, may, through elastic scattering, cause reduction of neutron energy to regions where σ_f of plutonium is increasing rapidly (below 100 kev) and thereby increase the reactivity. Inelastic scattering in elements of higher atomic weight, however, may easily reduce the neutron energy to the extent that fission in U-238 is no longer possible, but not far enough to gain by increased fission cross section in plutonium, thereby causing a net decrease in reactivity.

Now, if the change in reactivity produced by an element is compared to the change caused by Pu^{239} or U^{235}, a ratio of the various cross sections of the element under test to the well-established values for the fissile element is obtained.

$$\frac{[\Delta k]_x}{[\Delta k]_f} = \frac{\left[(\nu_p - 1)\sigma_p - \sigma_a\right]_x}{\left[(\nu_p - 1)\sigma_p - \sigma_a\right]_f} \qquad (1)$$

where

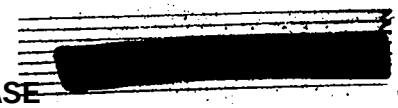
σ_p = productive absorption = $\sigma_{n,2n}, \sigma_f, \sigma_{e.s.}, \sigma_{in.s.}$

σ_a = destructive absorption = $\sigma_{(n,\alpha)}, \sigma_{(n,p)}, \sigma_{(n,\gamma)}$

ν_p = effective number of neutrons emerging from process p

x = element under test

f = fissile material



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For Pu^{239} and U^{235} $\sigma_p = \sigma_{fission}$ and $\nu = \nu_f$, then

$$\frac{[\Delta k]_x}{[\Delta k]_f} = \frac{[(\nu_p - 1)\sigma_p - \sigma_a]_x}{[\nu_f - 1 - \alpha]_f}$$

or

$$[\sigma_a - (\nu_p - 1)\sigma_p]_x = -[\nu_f - 1 - \alpha]\sigma_f \frac{(\Delta k)_x}{(\Delta k)_f} \quad (2)$$

ν_p is used to measure the relative effectiveness of neutrons produced by a reaction compared to the neutron absorbed. For (n,2n) reactions $\nu_p \backsim 2$, for $(n,n)_{e.s.}$ $\nu_p \backsim 1$ or > 1, depending upon the atomic mass and for $(n,n)_{in.s.}$ $\nu_p \backsim 1$ or < 1, again depending upon the atomic mass.

Equation (2) can be re-written

$$[\sigma_a - (\sigma_1(n,2n) + (\nu_{e.s.} - 1)\sigma_{e.s.} + (\nu_{in.s.} - 1)\sigma_{in.s.})]_x = -\left[(\nu_f - 1 - \alpha)\sigma_f \frac{(\Delta k)_x}{(\Delta k)_f}\right] \quad (3)$$

If this general relation is considered in terms of atomic mass of the element the relation can be somewhat simplified.

Low Atomic Mass

Elements of low atomic mass will have no $\sigma_{in.s.}$ term ($\nu_{in.s.} \backsim 1$) and only two elements are known to have low enough (n,2n) thresholds to be considered: Be (1.63 Mev threshold) and D (2.18 Mev). Other elements have known thresholds which range

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from 5 to 10 Mev but because the neutron energy spectrum of the reactor has only 2 percent of the total neutrons above 2.5 Mev the (n,2n) contribution from other elements has been ignored.

For elements from A = 1 to A ~ 15 where $\sigma_{e.s.}$ is important because relatively large energy losses occur in a single collision, $\nu_{e.s.} > 1$ because of the increase in ($\sigma_f \nu$) of plutonium at the reduced energies. Eq. (3) becomes (with the exception of Be and D) for these elements

$$\left[\sigma_a - (\nu_{e.s.}-1)\sigma_{e.s.}\right]_x = -\left[(\nu_f - 1 - \alpha)\sigma_f \frac{(\Delta k)_x}{(\Delta k)_f}\right] \quad (4)$$

If $\sigma_a > (\nu_{e.s.}-1)\sigma_{e.s.}$ the total effect will be to decrease the reactivity and conversely, if $\sigma_a < (\nu_{e.s.}-1)\sigma_{e.s.}$ the total effect will be to increase the reactivity.

For Be and D

$$\left[\sigma_a - (\nu_{e.s.}-1)\sigma_{e.s.} - \sigma_{(n,2n)}\right]_{\substack{Be \\ D}} = -\left[(\nu_f - 1 - \alpha)\sigma_f \frac{(\Delta k)_x}{(\Delta k)_f}\right] \quad (5)$$

Medium Atomic Mass

Elements in the middle of the periodic table can be considered to have $\sigma_{(n,2n)} \cong 0$, $\nu_{e.s.} \leq 1$ and $\nu_{in.s.} \leq 1$, depending upon the fissile material affected by the degraded neutrons. Little evidence exists for inelastic scattering in which the final neutron energy lies below 200 kev and in this region the value of ($\sigma_f \nu$) for plutonium does not change rapidly. Hence, for the reactivity contributed by the inelastically scattered neutrons producing fission in plutonium $\nu_{in.s.} \sim 1$.

For the reactivity contributed in 28 fission $\nu_{in.s.} < 1$ because the neutrons can be degraded below the 28 threshold.

High Atomic Mass

The same arguments may be used here as for elements of medium atomic mass.

Equation 3 can be written for medium and high atomic mass elements

$$\left[\bar{\sigma}_a + (1 - \nu_{in.s.})\,\sigma_{in.s.}\right]_x = -\left[(\nu_f - 1 - \alpha)\,\sigma_f \frac{(\Delta k)_x}{(\Delta k)_f}\right] \quad (6)$$

Here, all effects are to decrease the reactivity since $\nu_{in.s.} < 1$ upon the consideration of the effect on 28 fission.

II. Experimental Method

An experimental thimble of thin walled steel tubing was installed in the center of the active region of the fast plutonium reactor. Sufficient uranium and plutonium fuel rods for operation were inserted in the configuration shown in Fig. 1. The carrying case for the samples to be measured is shown in Fig. 2. Solid samples which could be machined were shaped to just fill the cavity of 2.5 cm by 1.15 cm diameter. Samples prepared in the form of powders and liquids were enclosed in spun aluminum cups which fitted the cavity.

The procedure was first to obtain a careful calibration for one control rod of excess reactivity in terms of pile period as a function of position. The units used are cents (1/100 k - space between critical and prompt critical conditions; in the

plutonium fast reactor this has been measures as $1¢ = 2.5 \times 10^{-5}$ in k.) The calibration and all measurements were done with pile operating at 1 watt. With the sample case in place and the cavity empty, the critical position of the calibrated control rod was determined for a base. The sample was then inserted and the new critical position measured. From the calibration curve, the change in pile reactivity produced by the sample was then interpreted in cents. A base point with empty cavity was taken following two successive sample measurements to correct for any changes in reactivity due to temperature.

In order to estimate the relative accuracy of the method, several determinations were made on selected elements such as Fe, Cu and C. The spread of data indicated that differences in reactivity could be measured with a probable uncertainty of .05¢.

III. Experimental Results

Table I lists the elements measured together with the reactivity changes observed. The values of $\bar{\sigma}$ listed in Table I and shown graphically in Fig. 3 are obtained from an average of the constants for U^{235} and Pu^{239}. The values of the fission constants used are

Element	(σ_f)	$(\nu - 1 - \alpha)$
25	1.37	1.36
49	1.95	1.86

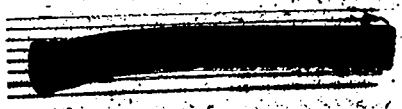

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Table I

Element	Mass(gms)	Δc/mol	$\bar{\sigma}$ (milli-barns)
H_2O	4.27	+5.6 ±0.2	+48.5 ±1.7
CH_2	4.87	+3.8 ±0.1	+33 ±1
Be^9	7.52	+2.70±0.07	+23 ±1
B^{10}	5.48	-97.5 ±0.05	-843 ±5
B	1.83	-16.8 ±0.3	-145 ±3
C	7.98	+0.81±0.08	+7 ±1
Na^{23}	1.27	0 ±0.09	- 0 ±0.8
Al^{27}	13.37	- 1.67±0.11	- 14 ±1.0
V^{51}	13.1	- 1.27±0.2	- 11 ±2
Cr	17.38	- 3.35±0.15	- 29 ±1.3
Mn^{55}	20.18	- 2.25±0.14	- 19.5 ±1.2
Fe	38.68	- 3.74±0.07	- 32.3 ±0.6
Co	39.12	- 3.24±0.07	- 28.0 ±0.6
Ni	43.56	- 5.32±0.06	- 46.0 ±0.5
Cu	44.13	- 4.83±0.07	- 41.8 ±0.6
Ga	7.44	- 8.1 ±0.5	- 70 ±4
$Y_2O_3(Y^{89})$	2.79	- 5.7 ±1.6	- 49 ±14
Zr	8.04	- 4.3 ±0.5	- 37 ±4
Cb^{93}	19.29	-11.8 ±0.2	-102 ±2
Mo	49.45	- 8.1 ±0.1	- 70 ±1
Ag	51.86	-31.3 ±0.1	-271 ±1
Cd	41.74	-14.5 ±0.1	-125 ±1
In	35.35	-33.6 ±0.2	-291 ±2
Sm	8.95	-19.8 ±0.8	-171 ±7
Ta	80.99	-29.9 ±0.1	-259 ±1
W	91.9	-15.1 ±0.1	-131 ±1
Au^{197}	87.22	-24.1 ±0.1	-208 ±1
Hg	32.02	-11.9 ±0.3	-103 ±3
Pb	55.92	- 1.86±0.2	- 16.1 ±1.7
Bi^{209}	47.0	- 2.28±0.2	- 19.7 ±1.7
Th^{232}	57.28	-19.6 ±0.2	-170 ±2
U	89.94	+ 3.2 ±0.1	+ 28 ±1
U^{235}	21.0	218	
Pu^{239}	18.23	417	

Elements listed with atomic number are single isotopes.

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Referred to 25, $\bar{\sigma} / \Delta \not\ell = (1.36)(1.37)/(218) = 8.57 \times 10^{-3}$

Referred to 49, $\bar{\sigma} / \Delta \not\ell = (1.86)(1.95)/(417) = 8.72 \times 10^{-3}$

Average $= 8.65 \times 10^{-3}$

The agreement is better than the accuracy assigned to the constants. Note that $\bar{\sigma}$ is the left-hand side of Equations (4), (5), (6) and therefore includes σ_a and σ_s of the element measured, and reactivity effects due to degradation.

III. Interpretation

The values of $\bar{\sigma}$ obtained in this investigation include scattering and absorption cross sections in a manner previously described. Until more information is obtained about the nature of these specific processes, it is difficult to separate the individual factors which make up the gross observable. It is interesting, however, to examine the results of a few elements in the light of the previous discussion.

A. Boron Absorption

Using the spectral distribution of neutron energy as determined in LAMS-727, and the absorption curve for boron reported by Bailey et. al. (Wall Paper Book) or RMP 19, 265, (1947) it is possible to calculate an effective absorption cross section for the reactor.

$$\bar{\sigma}_a = \frac{\int nv\,\sigma\,dE}{\int nv\,dE} = 156 \text{ mb}$$

The experimental value, measured as B-10 is 843 millibarns, which, corrected for isotopic abundance, becomes 155 millibarns.

This excellent agreement is certainly fortuitous but it probably reflects the fact that σ_a is large and represents the primary interaction with neutrons.

B. Gold Absorption

The same calculation and comparison can be made in the case of gold.

$$\bar{\sigma}_r = \frac{\int nv\sigma \, dE}{\int nv \, dE} = 157 \text{ mb.}$$

The experimental value is 204 mb. The discrepancy of 47 mb possibly measures the inelastic degradation of neutrons by gold.

C. Positive Reactivity Changes with Light Elements

Several of the light elements produced positive reactivity changes which demonstrate the predominance of the scattering term over absorption. To examine these cases, one can assume absorption to be negligible and the $\sigma_p = \sigma_t$. Substituting the proper constants in Equation (4), one obtains:

Element	$\bar{\sigma}$ mb	σ_t(0.65 Mev)	ν
H(in CH_2)	13	5b	1.0026
C	7	3b	1.0023
B-11(in B)	9.5	2.5b	1.0038
		Average $\nu_{e.s.}$ =	1.003

In the case of beryllium, Equation (5) must be used. Here there are three terms to separate:

n,n; assume similar to H, C, B-11 ($\sigma_t = 3.3$ b, $\nu = 1.003$)

n, ; calculate according to B-10

n,2n; credit with balance to effect.

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$$\left[\sigma_a - (\nu-1)\sigma_{e.s.} - \sigma_{n,2n}\right] = -23 \text{ mb}$$

$$\sigma_{n,2n} = 23 - (0.003)(3.3) + \frac{\int nv\,\sigma\,dE}{\int nv\,dE}$$

$$= 23 - 10 + 6$$

$$\sigma_{n,2n} = 19 \text{ mb.}$$

Further interpretation of the results in terms of the component factors will be postponed until additional information is obtained with other fuel rod configurations and activation cross section measurements.

Conversations with Harvey Brooks of the Knolls Atomic Power Laboratory, S. Untermeyer of the Argonne, and John Menke of Clinton, at the April Information Meeting at Brookhaven, were very helpful in the preliminary interpretation of these data. A private communication from Harvey Brooks pointed out the importance of inelastic scattering in connection with fast fission in U^{238}.

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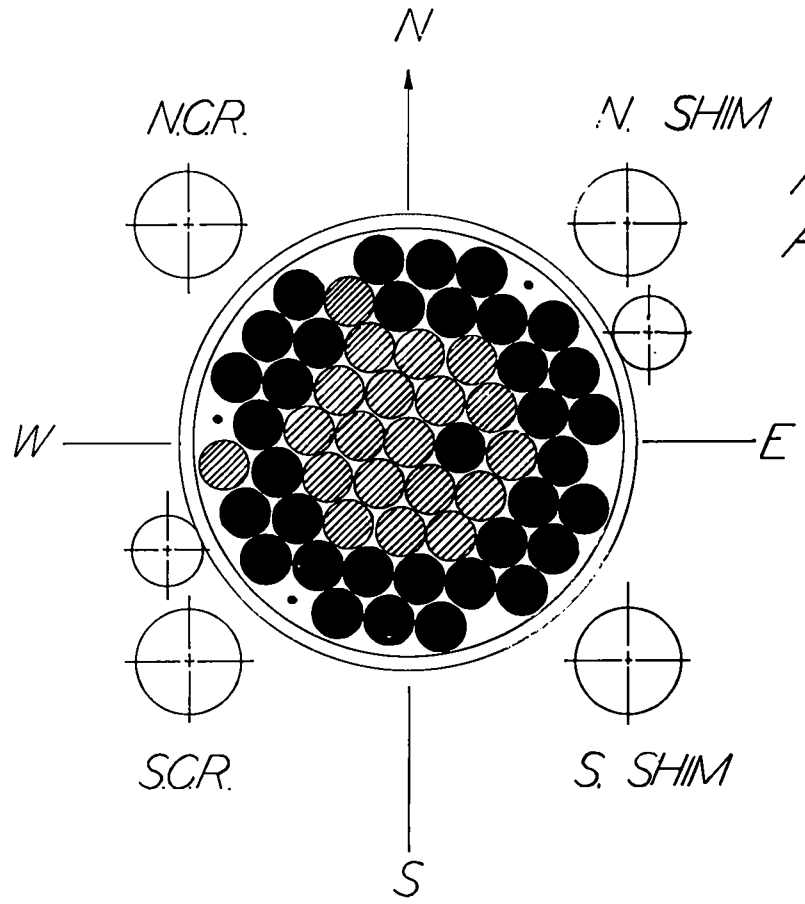
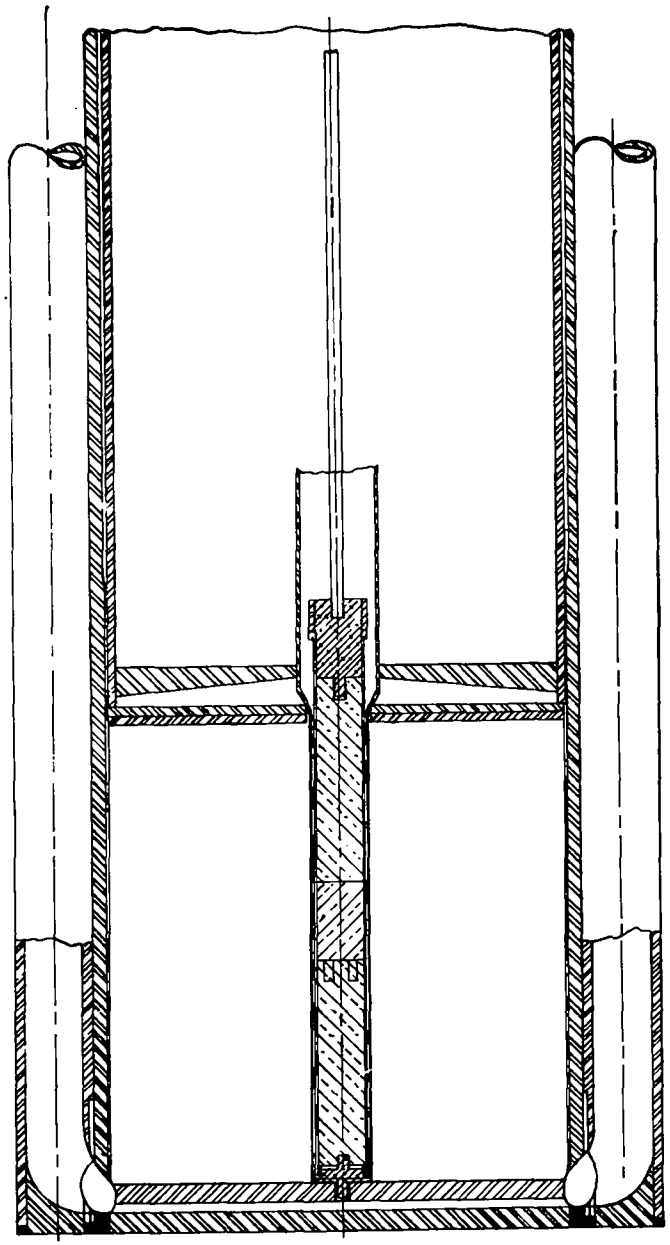


REACTOR LOADING FOR ABSORPTION CROSS-SECTION AND NEUTRON SPECTRUM MEASUREMENTS

PLUTONIUM

URANIUM

NOTE: THE PLUTONIUM ROD SHOWN DISPLACED FROM THE CENTER IS NORMALLY IN THE CENTER.



DANGER COEFFICIENT

SAMPLE POSITION

STEEL

URANIUM

DURAL

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