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Neutron Flux Measurements
in Uranium Metal Exponential Columns
of 6.53% and 9.12% ^{235}U

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NEUTRON FLUX MEASUREMENTS IN URANIUM METAL EXPONENTIAL COLUMNS

OF 6.53% AND 9.12% ^{235}U

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R. G. Steinke

ABSTRACT

Material bucklings measured in the equilibrium neutron spectra of heterogeneous uranium metal exponential columns containing 9.12% and 6.53% ^{235}U are reported. These results of exponential-column measurements are compared with values computed by means of Hansen-Roach cross sections. Through application of a fine-structure insert in the equilibrium spectral region of each column, fission cross-section ratios are structure-averaged. The measured spectral indices are given.

INTRODUCTION

This report presents the results of neutron flux measurements performed in cylindrical, uranium metal exponential columns with average enrichments of 9.12% and 6.53% ^{235}U . The material buckling and the equilibrium spectral indices, $\sigma_f(^{235}\text{U})/\sigma_f(^{238}\text{U})$, $\sigma_f(^{237}\text{Np})/\sigma_f(^{238}\text{U})$, and $\sigma_f(^{239}\text{Pu})/\sigma_f(^{235}\text{U})$, were determined for each column. They supplement results reported by Steinke ⁽¹⁾ for a column averaging 4.29% ^{235}U .

The work was performed in an environment that eliminated some of the problems which adversely affected the accuracy of previously measured parameters. ⁽²⁾ The earlier values were determined by experiments performed in 15-in.-diam columns housed inside a steel-reinforced concrete building. Experimental difficulties encountered in the early work were ultimate failure of the original fast neutron source assembly and a large amount of neutron back-scattering from the building structure. The back-scattering was so intense that experimental deter-

minations of the radial buckling parameters, α , were inconclusive.

The exponential column setup was moved outdoors some 11 ft above ground level; a new fast neutron source, "Hydro", was employed; ⁽³⁾ and sufficient materials were fabricated to make 21-in.-diam columns possible.

EQUIPMENT

Complete descriptions of the fast reactor source, a typical exponential column, and the supporting structure are found in the literature. ^(3,4) The 21-in.-diam columns are axially heterogeneous structures consisting of one 93.3% ^{235}U plate of 0.118-in. (3 mm) thickness for every two or three normal uranium plates of 0.590-in. (15 mm) thickness. The plate structure containing one enriched plate for every two normal plates has an average ^{235}U content of 9.12%; for the plate structure with one enriched plate for every three normal plates

Fig. 1. Material buckling versus ^{235}U concentration of uranium metal.

the average ^{235}U content is 6.53%.

In an attempt to diminish the effect of the axial heterogeneous structure for improved spectral index measurements, several 1.5-in.-diam coarse structure units in the center of the column were replaced with units having a finer structure by a factor of 10 or 15. This was accomplished by sandwiching natural uranium discs of 0.118-in. thick-

ness and U(93.3) foils having thicknesses ranging from 0.006 to 0.012 in. Figure 2 shows the fine-structure insert used in the 6.53% column.

Cadmium sheets, 0.030-in. thick, covered the top and cylindrical surfaces of the column as a means of reducing the effect of low-energy back-scattered neutrons.

Fig. 2. The fine-structure stack, coarse-structure axial glory-hole filler, and the 6.53% ^{235}U exponential column.

Foils of ^{235}U and depleted ^{238}U (4235/1), about 0.5-in. diameter and various thicknesses, were used for traverses and $\sigma_f(^{235}\text{U})/\sigma_f(^{238}\text{U})$ measurements. A ^{237}Np foil of 0.003-in. thickness and two ^{239}Pu foils of 0.001 and 0.003-in. thickness (each canned in nickel with a diameter of 0.5 in.) were used for measurements of $\sigma_f(^{237}\text{Np})/\sigma_f(^{238}\text{U})$ and $\sigma_f(^{239}\text{Pu})/\sigma_f(^{235}\text{U})$. The well-known spectra of the ^{235}U and ^{238}U cores of the Flattop critical assembly provide the factors for converting the measured beta-activity ratios of fission cross-section ratios.

PROCEDURE

Foil Activation

For traverses, in which 20 to 40 foils were activated, the foils were first counted for background beta activity. They were then sandwiched between aluminum foils of 0.001-in. thickness and of equal or larger diameter, and positioned in notches of an axial or radial foil holder. The aluminum foils prevent fission fragment deposition on the foil from the surrounding column material. For the special spectral index measurements in the fine-structure unit or in the center of the Flattop critical assembly core, each foil was wrapped in aluminum of 0.001-in. thickness.

The foils, positioned in the exponential column, were then irradiated for 20 to 30 min at a source power level of from 2 to 5 kW. The foils were removed from the column one hour after source reactor shutdown.

The Flattop activation involved positioning of five foils [^{238}U (0.010-in.-thick), ^{239}Pu (0.001-in.), ^{239}Pu (0.003-in.), ^{235}U (0.001-in.), ^{237}Np 0.003-in.)] at the center of the ^{235}U core and activating them with a 20-min irradiation. The foils were removed from the assembly 15 min after shutdown. Three of the foils [^{238}U (0.010-in.), ^{235}U (0.001-in.), ^{237}Np (0.003-in.)] were also activated in the ^{238}U core of Flattop.

Beta Activity Detection

The Jukebox ⁽⁵⁾ and methane-flow proportional

beta counter of Grundl ⁽⁶⁾ were used in the course of this experiment. Because of the large number of foils involved, the fully automated Jukebox counting system is best suited for traverse measurements. This detection system can be programmed to measure, in any predetermined order, the activities of as many as 100 foils.

A ^{235}U and a ^{238}U foil were selected from each traverse as standards for normalizing all measured foil activities to 180 min after activation shutdown. Starting with the traverse foil pair of lowest activation and by alternating between the standard foil pair and the traverse foil pair, beta activities were measured in order of increasing value. This counting process was repeated until 4 to 5 h after shutdown, providing two to four activity measurements for each pair.

Determining the ^{238}U (n,f) activity involved taking into account both the ^{238}U (n, γ) and ^{235}U (n,f) activities. The ^{238}U (n, γ) contribution was reduced to a negligible level by placing a 0.020-in.-thick brass shield between the foil and detector and by waiting for the 23.5-min ^{238}U (n, γ) activity to decay for several half-lives. Figure 3 presents a plot of the ^{235}U to ^{238}U beta activity ratio versus time after shutdown for a Flattop irradiation and various brass-shield thicknesses. The sharp drop in the beta-activity ratio for short times was interpreted as being due to the ^{238}U (n, γ) contribution. The 0.020-in. brass shields result in negligible ^{238}U (n, γ) activity for times greater than 100 min after activation shutdown. These same measurements with unshielded foils after activation in the 9.12% exponential column show no ^{238}U (n, γ) contribution after one hour.

Digital-Computer Data Analysis

Calculations were programmed in FORTRAN IV language for the "STRETCH" IBM 7030 computer. Foil activities were corrected for background activity, counting system deadtime losses, radioactive decay, axial-end correction, and competing activities. The resulting activities are in counts per minute normalized to 180 min after source shutdown.

Fig. 3. Beta-activity ratio versus time after irradiation in a Flattop with enriched uranium core.

RESULTS

Material Buckling

The buckling equation, $\nabla^2 \phi + B_M^2 \phi = 0$, can be derived from the application of neutron diffusion theory.⁽⁷⁾ When solving this equation for the conditions of a cylindrical geometry in free space having a point source centered at one end, the eigenvalue B_M^2 , the material buckling, is represented by $(\alpha^2 - \gamma^2)$, where α and γ are parameters in the fundamental solution:

$$\phi = A J_0(\alpha r) \sinh[\gamma(H-z)].$$

In a heterogeneous assembly, this solution will only approximate the overall neutron flux distribution.

By fitting the functions $J_0(\alpha r)$ and $\sinh[\gamma(H-z)]$ to activity distributions in the column, values of α and γ are obtained. Figure 4 represents typical axial traverses of ^{235}U activity for the 9.12% and 6.53% columns, and Fig. 5 gives a typical

radial ^{235}U distribution for the 6.53% column. The radial traverses in the 9.12% and 6.53% columns are at the 9.1- and 10.4-in. levels and the 11.3- and 13.4-in. levels, respectively, where spectral equilibrium had been indicated.

The ^{238}U activities are influenced by spectral changes near the column boundaries caused by fast neutron backscattering and neutron leakage spectrum hardening. Because of the relatively constant fission cross section of ^{235}U over the energy range of the neutron spectrum, enriched uranium foils provide the principle traverse data from which bucklings were established.

Axial and radial buckling parameters determined by averaging the results of least-squares curve fitting are as follows.

For the 9.12% column:

$$\gamma = 0.1050 \pm 0.0027 \text{ in.}^{-1}$$

$$\alpha = 0.2211 \pm 0.0062 \text{ in.}^{-1}$$

where the measured radial extrapolation distance is $0.38 \pm 0.30 \text{ in.}$

For the 6.53% column:

$$\gamma = 0.1776 \pm 0.0054 \text{ in.}^{-1}$$

$$\alpha = 0.2052 \pm 0.0073 \text{ in.}^{-1}$$

where the measured radial extrapolation distance is $1.22 \pm 0.40 \text{ in.}$

Fig. 4. Axial activation ^{235}U traverses in 9.12% and 6.53% ^{235}U exponential columns.

From the listed experimental extrapolation distances, it is apparent that the attempt to measure the radial buckling parameter leaves much to be desired. The radial buckling is primarily dependent on the column radius and only weakly dependent on the column uranium composition. A better value of α than that measured directly can be determined from $\alpha = 2.405/\sqrt{r_{\text{column}} + 0.710 \lambda_{\text{tr}}}$ if a crude value of λ_{tr} is known.

The Carlson S_4 and asymptotic diffusion theory calculations (8,9) represented in Fig. 1 made use of the Hansen-Roach sixteen-group cross sections. (10)

The S_4 calculations for the 9.13% and 6.53% enrichments established the critical radii of bare spheres. Further, the flux-averaged macroscopic transport cross section, $\Sigma_{\text{tr}}(R)$, at the critical radius, R , was used to determine the extrapolation distances, $0.710 \lambda_{\text{tr}}$, which with the critical radii led to buckling values.

Fig. 5. Radial activation ^{235}U traverses in 6.53% ^{235}U exponential column.

The best values for material buckling, B_M^2 , determined by using the radial extrapolation distances from Hansen-Roach cross sections with an absolute error of $\pm 0.21 \text{ in.}$ and the measured values of γ are as follows.

For 9.12% ^{235}U :

$$\gamma = 0.1050 \pm 0.0027 \text{ in.}^{-1}$$

$$\alpha = 0.2136 \pm 0.0028 \text{ in.}^{-1}$$

$$B_M^2 = \alpha^2 - \gamma^2 = 0.03460 \pm 0.00132 \text{ in.}^{-2}$$

For 6.53% ^{235}U :

$$\gamma = 0.1776 \pm 0.0054 \text{ in.}^{-1}$$

$$\alpha = 0.2141 \pm 0.0029 \text{ in.}^{-1}$$

$$B_M^2 = \alpha^2 - \gamma^2 = 0.01430 \pm 0.00228 \text{ in.}^{-2}.$$

Spectral Indices

The fission cross-section ratios for ^{235}U and ^{238}U , ^{237}Np and ^{238}U , and ^{239}Pu and ^{235}U were measured in the fine-structure stack of the 9.12% and 6.53% columns. These measurements involve activating five foils ^{235}U (0.001 in.), ^{238}U (0.010 in.), ^{237}Np (0.003 in.), ^{239}Pu (0.001 in. and 0.003 in.) at the center of the exponential column and also at the center of the Flattop core (where the fission cross-section ratios are known*). The ratio of the known fission cross-section ratio to the measured

	Flattop Core	Fission Cross-Section Ratio
$\sigma_f(^{235}\text{U})/\sigma_f(^{238}\text{U})$	^{235}U	$6.78 \pm 2\%$
	^{233}U	$5.30 \pm 2\%$
$\sigma_f(^{237}\text{Np})/\sigma_f(^{238}\text{U})$	^{235}U	$5.23 \pm 2.5\%$
	^{233}U	$4.76 \pm 2.5\%$
$\sigma_f(^{239}\text{Pu})/\sigma_f(^{235}\text{U})$	^{235}U	$1.44 \pm 3\%$

beta-activity ratio in the Flattop core is a calibration factor that converts a beta-activity ratio for the column to a fission cross-section ratio.

Determination of the fission cross-section ratios representative of the average column composition involves some difficulty due to the axial heterogeneity. For both columns, the coarse structure dictated two measurement positions, each having different spectral characteristics, where the foils could be placed. At one position, between an oralloy and tuballoy slug, the spectrum is harder than average and at the other position, near the center of a unit between two tuballoy slugs, the spectrum is softer.

The fine-structure insert, mentioned earlier, was used in an attempt to overcome this difficulty. As is shown in Figs. 6 and 7, the finer-structure stack used in the 9.12% column does reduce the effect of spectrum variation on $\sigma_f(^{235}\text{U})/\sigma_f(^{238}\text{U})$ to less than 3% as compared to the ~30% variation that occurs within a coarse-structure unit.

Fig. 6. Results of $\sigma_f(^{235}\text{U})/\sigma_f(^{238}\text{U})$ measurements in coarse-structure unit; 9.12% column.

Fig. 7. Results of $\sigma_f(^{235}\text{U})/\sigma_f(^{238}\text{U})$ measurements in fine-structure stack compared with results across coarse-structure unit, 9.12% column.

Fig. 8. Results of $\sigma_f(^{235}\text{U})/\sigma_f(^{238}\text{U})$ measurement in fine-structure stack compared with coarse-structure results, 6.53% column.

Results from the 6.53% column are displayed in Fig. 8. In this case, the value considered most reliable (measured with the same foil used for Flat-top normalization) falls above the average throughout the fine-structure stack. The other cross-section ratios were also derived from measurements within the fine-structure stack. Resulting experimental values of the three spectral indices compare with values from S_n calculations as follows:

	MEASURED	CALCULATED
<u>For the 9.12% Column</u>		
$\sigma_f(^{235}\text{U})/\sigma_f(^{238}\text{U})$	$27.0 \pm 3\%$	28.28
$\sigma_f(^{237}\text{Np})/\sigma_f(^{238}\text{U})$	$8.86 \pm 4\%$	
$\sigma_f(^{239}\text{Pu})/\sigma_f(^{235}\text{U})$	$1.31 \pm 4\%$	1.19
<u>For the 6.53% Column</u>		
$\sigma_f(^{235}\text{U})/\sigma_f(^{238}\text{U})$	$39.0 \pm 4\%$	36.72
$\sigma_f(^{237}\text{Np})/\sigma_f(^{238}\text{U})$	$9.40 \pm 4\%$	
$\sigma_f(^{239}\text{Pu})/\sigma_f(^{235}\text{U})$	$1.27 \pm 4\%$	1.16

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