NOV 16 1965

CFSTI PRICES

H.C. \$ 3.00; MN 1.65

BNL 9607

MASTER

BNL NO.

RP No. 323

THERMAL SPECTRUM MEASUREMENTS IN SLIGHTLY ENRICHED URANIUM,
LIGHT WATER MODERATED LATTICES, BY THE LUTETIUM ACTIVATION METHOD. (*)

NOT APPROVED FOR PUBLIC RELEASE. AVAILABLE
TO THE AEC AND ITS CONTRACTORS ONLY.

S. Tassan (xx)

RELEASED FOR ANNOUNCEMENT
IN NUCLEAR SCIENCE ABSTRACTS

Brookhaven National Laboratory
Upton, New York, U.S.A.

September, 1965

(*) Work performed under the auspices of the USAEC

(xx) Present address: EURATOM, CCR ISPRA, ISPRA (Italy)

This Paper Contains: Fourteen Pages

One Table

Seven Graphs

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

THERMAL SPECTRUM MEASUREMENTS

Proofs should be sent to:

Stelvio Tassan
CCR Euratom
CP 191 Como
Ispra (Varese), Italy

ABSTRACT

Thermal spectrum measurements in 1% enriched uranium, 0.387 in. dia. rod lattices, light water moderated, have been performed by the lutetium activation method, in order to test the usefulness of this technique in not well thermalized lattices. Dy^{164} and Lu^{175} were used as reference thermal neutron detectors.

The experimental results are presented as the average values of the normalized (lattice/Maxwellian) activation ratios of $\text{Lu}^{176}/\text{Dy}$ and $\text{Lu}^{176}/\text{Lu}^{175}$, with the pertinent cadmium-ratios, in the fuel and in the moderator over the W/U range from 1:1 to 4:1. The agreement with the results of a calculation performed using the THERMOS code is satisfactory.

It is concluded that the $\text{Lu}^{176}/\text{Dy}$ set appears to be adequate for the thermal spectrum analysis of light-water-moderated lattices, while the $\text{Lu}^{175}/\text{Dy}$ set can provide rather accurate integral data for the epithermal component of the neutron spectrum.

THERMAL SPECTRUM MEASUREMENTS IN SLIGHTLY ENRICHED URANIUM,
LIGHT WATER MODERATED LATTICES, BY THE LUTETIUM ACTIVATION METHOD

S. Tassan

The lutetium activation technique for the determination of integral parameters representative of the thermal neutron spectrum, which is based on the existence of a resonance in the neutron activation cross section of Lu^{176} around 0.142 ev, has been described in detail by L. C. Schmid and W. P. Stinson ⁽¹⁾.

This paper reports on measurements which have been performed, using this technique, in slightly enriched uranium - light water moderated lattices, characterized by moderator-to-fuel volume ratios varying from 4:1 to 1:1. Purpose of the experiment was to explore the usefulness of the Lu-activation method in not well-thermalized lattices - i.e. whether this method is sensitive to thermal neutron spectrum variations in the fuel and in the moderator, as a function of the moderator-to-fuel ratio - and to provide a test for refined neutron spectrum calculation codes.

Briefly, the Lu-activation technique consists of comparing the thermal response of Lu^{176} to that of a thermal neutron detector, preferably of the $1/v$ type, irradiated in the neutron spectrum investigated. The data are normalized to the ratio of the activities of the same detectors, when irradiated in a Maxwellian neutron spectrum.

The measured activation ratios are compared to the corresponding ratios calculated by multigroup thermal-neutron codes, thus providing a test of the adequacy of both the theoretical model and the basic cross section data used for the codes. In particular, when the neutron spectrum can be approximated by the Westcott model ⁽²⁾, these activation ratios yield the "effective neutron temperatures" and "r factors", as defined by such formalism.

The quantities to be determined experimentally are then the "normalized" total activities and the Cd-ratios of Lu^{176} and of the thermal neutron detector, in the neutron spectrum of interest. For this experiment Dy^{164} and Lu^{175} were both used as thermal neutron detectors.

Although Dy is not exactly an $1/v$ detector, its cross section behaviour is sufficiently well known ⁽³⁾ for the correct interpretation of the data. The advantage in the use of Dy is that it has an epithermal activation considerably lower than

any of the usual $1/v$ type detectors, so that the Cd-ratio for dysprosium needs not to be accurately determined.

The obvious advantage in the use of Lu^{175} is that it is in isotopic mixture with the Lu^{176} , thus eliminating the need for cumbersome intercalibrations to determine the relative concentration of Lu^{176} and the thermal neutron sensitive isotope in the corresponding detectors (e.g. the Lu-Al and Dy-Al foils); these intercalibrations add appreciably to the total uncertainty of the experimental results. However, in some cases such advantage may be offset by the fact that Lu^{175} exhibits a considerably large resonance integral.

Measurements have been carried out in the fuel and in the moderator of light-water-moderated hexagonal lattices of 1% enriched uranium rods, 0.387 in. in diameter, canned by 0.028 in. of aluminum. The water-to-uranium volume ratios investigated were nominally 4:1, 3:1, 2:1, 1.5:1, and 1:1.

The facility used was a miniature assembly, measuring 12 in. in diameter by 18 in. high, contained in an aluminum tank surrounded on all sides, except the top, by 1/16 in. of cadmium sheet and 3 in. of paraffin. The whole assembly was irradiated in the tunnel under the Brookhaven National Laboratory graphite reactor, supplying a flux around 10^9 neutrons/cm² sec. Previous work had proved the feasibility of such small size assemblies for

measurements of microscopic lattice parameters ⁽⁴⁾.

The detectors were made from 0.005 in. thick sheets of Lu-Al and Dy-Al alloys, containing respectively about 10% Lu and 5% Dy by weight. The detectors in the fuel were circular foils, 0.387 in. in diameter, placed in a split rod of the first lattice hexagon; they were protected from fission product contamination by 0.001 in. thick Al buffers.

When irradiated Cd-clad, the detectors in the fuel were contained in a 1/4 in. high Cd-box, with 0.025 in. thick end pieces and a 0.020 in. thick wall embedded in the Al cladding. Two 0.050 in. thick fuel buttons were placed between the detector and the end pieces of the cadmium box, in order to minimize the streaming of epi-cadmium neutrons directly from the moderator into the detector.

The detectors in the moderator were sector-foils precision machined to reproduce an iterative portion of the lattice elementary cell. They were placed, sandwiched by two 0.020 in. thick lucite sheets, between three fuel rods, which were joined in a single unit and occupied the central position and two adjacent positions in the first hexagon of the lattice.

When irradiated Cd-clad, the detectors in the moderator were contained in a cadmium box, 0.020 in. thick, resting among three cadmium rings, 0.20 in. high and 0.020 in. thick, embedded

in the Al cladding of the three-rod unit. Thus the cadmium box completely surrounded the detector, without modifying the geometry of the lattice cell.

The irradiations in the Maxwellian flux, for the normalization of the activations in the lattice, were performed using the thermal column of the Brookhaven National Laboratory graphite reactor. Sandwiches of Lu-Al and Dy-Al detectors were irradiated, using arrangements of foils which also provided the experimental correction factors for the minor attenuation of the flux through the foils in the sandwich. Similar thermal flux irradiations were made for the circular foils and the sector-shaped foils corresponding to the W/U from 1 to 4. Thus the normalization of the data (lattice/Maxwellian) cancelled out the foil-geometry effect on the counter efficiency.

After the irradiation, the induced beta activity of Dy¹⁶⁵ was measured by an end-window gas-flow proportional counter; the induced gamma activity of Lu was measured by a NaI (Tl) crystal-single channel analyser unit, biased at 37 kev.

The Lu activity (from Lu¹⁷⁷ and Lu¹⁷⁶ decay) was measured about 3 hours after the end of the irradiations and again at least 3 days later, when the 3.71 hours Lu¹⁷⁶ decay activity had decreased to a negligible amount (note that the Lu¹⁷⁵ activation produces Lu¹⁷⁶ activity, and similarly for Lu¹⁷⁶).

The residual Lu^{177} activity was reduced to the time of the first counting and subtracted from it. From the Lu^{176} activity thus obtained, the measured Lu^{177} activity and the measured Dy^{165} activity, the corresponding "saturated activities" were obtained in the usual way.

Standard corrections for counter dead time, room background, Lu natural activity, irradiation length, were applied to the data, as well as foil intercalibration factors determined by the usual activation techniques. While the self-shielding of the Dy^{164} and Lu^{176} resonances was negligible for the high dilution of these isotopes in the detectors used, a minor correction was applied to the data to compensate for the self-shielding of the Lu^{175} resonance at 2.6 ev.

The stability of the counting equipment was tested before every use against the constant count rate of a standard Tl 205 beta source (for the gas-flow counter) and by gamma source calibration (for the scintillation counter), using the 0.0596 Mev gammas of Am^{241} and the 0.0875 Mev gammas of Cd^{109} .

The results of the experiment are shown in Table 1 and Figures 1 to 5, the quoted errors being the standard deviations of the mean from 4-6 independent measurements.

The data are presented as: the total activation ratios for Lu^{176} (lattice/Maxwellian) normalized to the corresponding total activation ratios for Dy (Figure 1), the cadmium-ratios

for Lu^{176} (Figure 2) and for Dy (Figure 3), the total activation ratios for Lu^{176} normalized to the corresponding total activation ratios for Lu^{175} (Figure 4), and the cadmium-ratios for Lu^{175} (Figure 5).

The curves of Figure 1 show the expected trend (i.e. the decrease of the total activation ratio (lattice/Maxwellian) for Lu^{176} normalized to that for Dy, with increasing moderator-to-fuel ratios), which corresponds to the higher thermalization of the neutron spectrum at larger lattice pitches. The gradient of the curves is larger at the smaller pitches, since there the spectrum variation in the lattice is more important.

Also shown in Figure 1 are some theoretical predications made by H. Honeck⁽⁵⁾ using the THERMOS code⁽⁶⁾ and the Nelkin kernel⁽⁷⁾ for water. These subcadmium results were corrected for epicadmium activation using the measured cadmium ratios for Dy and Lu^{176} . The theoretical values agree well with the measured values for the average ratio in the cell, but the computed values of the separation between the fuel and moderator ratios are about 3 times larger than measured.

The Cd-ratios for Dy and Lu^{176} are large both in the fuel and in the moderator; consequently, the error introduced in the thermal activation values by uncertainties in the above Cd-ratios is

generally very small. For instance, in the typical lattice with $W/U = 2$, a $\pm 10\%$ error in the cadmium ratios for Dy and Lu^{176} introduces an error around $\pm 0.3\%$ and $\pm 0.5\%$ in the corresponding thermal activation values.

The curves of Figure 4, i.e. the total activation ratios of Lu^{176} normalized to those of Lu^{175} , exhibit a trend just opposite that of the curves of Figure 1. This behaviour is very simply explained by the large epi-Cd activation of Lu^{175} in the light-water-moderated lattices, which is mainly due to a resonance in the Lu^{175} neutron activation cross section at about 2.6 ev; such activation, which obviously increases with decreasing lattice pitch, prevails over the slowly varying behaviour of the Lu^{176} total activation. In fact, when the sub-cadmium activation ratios for Lu^{176} and Lu^{175} are inferred from the data of Figures 2, 4 and 5, the corresponding curves (Figure 6) show the correct trend, in agreement with the curves of Figure 1.

It follows that, in not well-thermalized lattices as light-water-moderated cores, small errors in the Cd-ratios for Lu^{175} may result in rather large uncertainties in the corresponding sub-Cd Lu^{175} activations (e.g. in the $W/U = 2$ lattice a $\pm 1\%$ error in the Lu^{175} Cd-ratio introduces a $\pm 8\%$ error in the corresponding thermal activation value). This is a serious limitation to the accuracy attainable when using Lu^{175} as the

thermal neutron detector for the Lu-activation method applied to the analysis of such lattices. Conversely, the large epi-cadmium activation of Lu^{175} may give good information about the epi-thermal component of the neutron spectrum. This is put in evidence of Figure 7, which shows the strong dependence of the quantity " $(\text{Lu}^{175}$ lattice epi-Cd activation/ Lu^{175} Maxwellian activation)/(Dy lattice total activation/Dy Maxwellian activation)" - as inferred from the data of Figures 1, 4, and 5 on the moderator-to-fuel volume ratio of the lattices investigated.

In conclusion, it appears that the Lu^{176} - Dy set is altogether more adequate for the thermal spectrum analysis of light-water-moderated lattices, than is the set Lu^{176} - Lu^{175} . Thermal spectrum data obtained using the latter set have been presented here, only to give evidence for the general trend of the relative curves (Figure 6). On the other side, the epi-cadmium response of Lu^{175} may be compared to the thermal response of Dy (or of another thermal neutron detector) to yield rather accurate integral data for the epi-thermal component of the neutron spectrum. Such procedure, which provides the epi-thermal-spectrum information directly from the analysis of the composite activation of the main detector, e.g. the lutetium, shows some advantages over the other methods to obtain the same

information, which require irradiations of an auxiliary resonance detector, as Au or In.

References:

- 1) L. C. Schmid and W. P. Stinson
HW-66319 (1960), HW-64866 (1960)
- 2) C. H. Westcott, W. H. Walker and T. K. Alexander
A/CONF. 15/P/202
- 3) R. Sher, S. Tassan, E. Weinstock and A. Hellsten
Nuclear Science and Engineering, 11,4,369-376 (1961)
- 4) H. Kouts and R. Sher
BNL-486 (1957)
- 5) H. Honeck, Private Communication, August, 1965
- 6) H. Honeck, BNL 5826 (1961)
- 7) M. Nelkin, Physics Review 119, 791 (1960)

LIST OF FIGURES

Fig. 1: Lu 176 Total Activation Ratios (Lattice/Maxwellian)
Normalized to Dy Total Activation Ratios

Fig. 2: Lu 176 Cd-Ratio

Fig. 3: Dy 164 Cd-Ratio

Fig. 4: Lu 176 Total Activation Ratios (Lattice/Maxwellian)
Normalized to Lu 175 Total Activation Ratios

Fig. 5: Lu 175 Cd-Ratio

Fig. 6: Lu 176 Sub-Cd Activation Ratios (Lattice/Maxwellian)
Normalized to Lu 175 Sub-Cd Activation Ratios

Fig. 7: Lu 175 Epi-Cd Activation Ratio Normalized to Dy
Total Activation Ratio

$$\left(= \frac{\text{Lu 175 Epi-Cd Lattice/Lu 175 Maxwellian}}{\text{Dy Total Lattice/Dy Maxwellian}} \right)$$

TABLE I EXPERIMENTAL RESULTS

$\frac{W}{U}$	Medium	$\frac{Lu\ 176}{Dy}$	$\frac{Lu\ 176}{Lu\ 175}$	Cd Ratio Lu 176	Cd Ratio Lu 175	Cd Ratio Dy
1:1	Fuel	1.654	0.1170	11.70	1.070	20.5
		± 0.01	± 0.0005	± 0.2	± 0.007	± 0.5
	Moderator	-	-	-	-	-
1.5:1	Fuel	1.493	0.1535	14.0	1.130	29.1
		± 0.010	± 0.0015	± 0.4	± 0.015	± 1.5
	Moderator	1.450	0.1655	15.0	1.150	32.8
		± 0.015	± 0.0010	± 0.4	± 0.015	± 1.0
2:1	Fuel	1.402	0.1847	18.1	1.150	41.3
		± 0.010	± 0.0010	± 0.4	± 0.005	± 0.8
	Moderator	1.381	0.1976	18.8	1.160	-
3:1	Fuel	1.320	0.2391	22.5	1.229	-
		± 0.007	± 0.0015	± 0.4	± 0.005	
	Moderator	1.289	0.2560	26.2	1.280	76.5
		± 0.010	± 0.0010	± 0.3	± 0.015	± 1.5
4:1	Fuel	1.276	0.2843	27.5	1.317	70.0
		± 0.010	± 0.0015	± 0.5	± 0.008	± 2.0
	Moderator	1.250	0.2972	33.4	1.343	97.8
		± 0.010	± 0.0025	± 0.4	± 0.015	± 1.5

$\frac{Lu\ 176}{Dy}$ = Total Activation Ratio for Lu 176 (lattice/Maxwellian) normalized to Total Activation Ratio for Dy

$\frac{Lu\ 176}{Lu\ 175}$ = Total Activation Ratio for Lu 176 (lattice/Maxwellian) normalized to Total Activation Ratio for Lu 175

Lu 176 Total Activation Ratios (Lattice/Maxwellian)
Normalized to Dy Total Activation Ratios

Figure 1

Lu 176 Cd-Ratio

Figure 2

DY 164 Cd-Ratio

Figure 3

Lu 175 Cd-Ratio

Figure 5

Lu 175 Epi-Cd Activation Ratio Normalized to Dy Total Activation Ratio

Figure 7