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THERMAL AND FISSION NEUTRON FLUX MEASUREMENTS IN PROCESS TUBING AND IN NEUTRON ACTIVATION TEST LOCATIONS

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INTRODUCTION

Neutron activation analysis provides the extremely high sensitivity necessary for the measurement of many trace elements. Activation techniques are presently being used to measure the concentrations of various undesirable parent isotopes in reactor process tubing, slug jackets and their corrosion film, and in process water and water treatment chemicals. The interpretation of results from activation analysis are sometimes complicated by the fact that a given radioisotope may be produced by fission neutrons (threshold reactions) from other elements as well as by the more common thermal neutron (n, γ) reactions. Shielding the sample with cadmium prior to irradiation or irradiating in the graphite reflector will minimize the contributions from thermal or fission neutrons, respectively.

The purpose of this study has been to measure the ratios and magnitudes of the thermal and fission fluxes in process tubing and to compare these with values at locations where neutron activations can be performed. Also, it was of interest to measure the ratios and magnitudes of the thermal and fission fluxes from a point in the lattice to a point well into the graphite reflector, and to determine if the fission neutron spectrum, as measured by two threshold reactions, changes appreciably over this region. These data will aid in selecting the most favorable available positions for activations with thermal neutron reactions or with threshold reactions.

The ratio of the thermal to fission neutron fluxes at various points in the lattice, at the outer edge of the graphite reflector, and in the aluminum process tubing was of primary interest in this study, whereas, their precise magnitude, which depends on somewhat uncertain cross section values, was of secondary interest.

The thermal neutron flux was usually based on either the Fe^{58} (n, γ) Fe^{59} or the Cu^{63}(n, γ)Cu^{64} reaction. The fission flux measurements were based on the threshold reactions, Al^{27}(n, α)Na^{24} or Fe^{54}(n,p)Mn^{54}. The fission to thermal ratios were based on either the formation rate of Mn^{54} and Fe^{59} from Fe^{54} and Fe^{58} or the formation of Na^{24} and Cu^{64} from Al^{27} and Cu^{63}, respectively. The ratio of the reaction rates for the threshold reactions Fe^{54}(n,p)Mn^{54} and Al^{27}(n, α)Na^{24} was used to detect any significant change in the shape of the fission spectrum which would affect threshold reactions.

Flux measurements similar to the F reactor Quickie facility (E test hole) measurements considered here were performed at a lower power level in the precursor to this facility in 1953,[1] using a different set of flux detectors. Also, a recent measurement of the thermal flux in the Quickie facility has been reported.[2]

SUMMARY AND CONCLUSIONS

The thermal and fission neutron fluxes have been measured in aluminum process tubing, in an empty process tube and in several pockets of the E test hole facility, with the stringer extending approximately 5" further into the reflector than in its normal position. In the E hole facility, an increase in the thermal to fission neutron flux ratio of about 20 was observed when the detector was moved from a point 11 inches inside the lattice to a point 25 inches outside (2 inches from the outer edge of the graphite). The fission spectrum, as measured by two different energy threshold reactions, did not change significantly over this region. Measurements of the production rates of Fe^{59} and Mn^{54} from iron or Na^{24} and Cu^{64} from Al^{27} and Cu^{63} were found to be suitable methods of determining the thermal to fission flux ratios at various locations in the reactor.

THEORETICAL

Several of the neutron activated radioisotopes which are formed in the corrosion film of aluminum process tubing and slug jackets and are subsequently released to the effluent water are formed by two or more reactions. For example P^{32} (14.7 day) is formed by:

$$P^{31}(n,\gamma)P^{32}$$
$$S^{32}(n,p)P^{32}$$
$$Cl^{35}(n,\alpha)P^{32}$$

In the use of activation analysis for measuring the concentrations of the trace quantities of parent isotopes in material of interest, it is often desirable to irradiate, as nearly as possible, with either thermal or fission neutrons so that the production of the daughter may be limited to n, γ or threshold reactions. It is also desirable to know the fission to thermal flux ratio at the positions of activation and at the corrosion film surface, since measurements at test positions must be extrapolated to the corrosion film surfaces.

Isotopes of iron can be conveniently used to measure the thermal and fission neutron fluxes.

	σ (mb)
$Fe^{58}(n,\gamma)Fe^{59}$	980 (thermal)
$Fe^{54}(n,p)Mn^{54}$	89 ± 9 (fission)

The Fe^{59} production serves as an indicator for the thermal flux (plus resonance, which contributes about six percent or less) and the Mn^{54} as an indicator for the fission flux.

In aluminum process tubing, the production of Na^{24} from aluminum and Cu^{64} from the trace copper present permits a very precise measurement of the thermal and fission neutron fluxes.

	σ (mb)
$Al^{27}(n,\alpha)Na^{24}$	0.60(3)
$Cu^{63}(n,\gamma)Cu^{64}$	4.3×10^3

Since the half-lives of Na^{24} and Cu^{64} are short, 15.0 hours and 12.8 hours, respectively, radioequilibrium is attained in a week or less of reactor operation. The fission flux is calculated directly from the Na^{24} concentration in a sample of aluminum tubing. The thermal flux calculation requires a measurement of both the Cu^{64} and the inert copper concentrations in the aluminum.

The conventional thermal neutron flux can be calculated from the rate of formation of a radioisotope by the equation

$$F = \frac{P}{\sigma N} \qquad (1)$$

N = Number of parent nuclei.

σ = Activation cross section for thermal neutrons.

P = Product nuclei formed per second.

The contribution from resonance neutrons can be determined and corrected for by covering the sample with a 30 mil cadmium liner and measuring the n,γ reaction rate.

The conventional thermal flux is the product of neutron density and thermal velocity (2200 meters per second) and can be obtained from activation of a 1/v absorber. A general discussion of reactor cross sections, flux measurements and experimental methods is given in reference 3.

The fission spectrum flux can be calculated by use of the "effective cross sections" for threshold reactions.(4)

$$F = \frac{P}{\sigma_e N} \qquad (2)$$

F = Total flux density of fission neutrons of all energies whether or not they contribute to the reaction.

σ_e = Effect cross section.

N = Number of target nuclei.

P = Number of product nuclei formed per unit time.

In using a single threshold reaction to measure the fission neutron flux, it is assumed that the energy distribution in that part of the fission neutron spectrum which contributes to threshold reactions does not change appreciably with sample location in the reactor. This assumption is reasonable, since the effect of moderated neutrons on the fission spectrum in the threshold reaction region (above 2 Mev) can usually be neglected.(4) Any significant change in the spectrum could be observed by measuring the ratio of the reaction rates (cross sections) of two or more threshold reactions which vary considerably in threshold energies. Experiments of this type have been performed in the BEPO reactor at Harwell.(5) Several threshold cross sections were measured in a hollow uranium bar and at various locations in the reactor, and no significant differences in cross section were observed.

In the work considered here, the cross sections for the reactions $Fe^{54}(n,p)Mn^{54}$ and $Al^{27}(n,\alpha)Na^{24}$ were compared in the test position of interest. The measured threshold for the Al^{27} reaction is about 6.5 Mev.(6) The calculated threshold for the Fe^{54} reaction(3) is Q = -0.16 Mev; however, because of the potential barrier for particle emission, the actual threshold is positive.(7) In an actual comparison of the relative reaction rates for these reactions at positions extending from the outer edge of the graphite reflector into the lattice, no significant change was observed.

EXPERIMENTAL

The E test hole facility (Quickie facility at F reactor) was used for the measurement of the thermal and fission fluxes at locations between the outer edge of the graphite reflector and the second and third rows of process tubes. The KE reactor "Quickie facility" was used for making flux measurements near the outer edge of the graphite and in the central flux zone of the reactor. The thermal and fission fluxes in these facilities were determined from the production rates of Fe^{59} and Mn^{54} in iron specimens. The thermal and fission fluxes in a poison column (process tube containing no fuel elements) were determined from the reaction rates of $Na^{23}(n,\gamma)Na^{24}$ and $Al^{27}(n,\alpha)Na^{24}$, respectively. The thermal and fission fluxes in aluminum process tubing samples were calculated from their Na^{24}, Cu^{64} and inert copper concentrations.

Measurements in the E Test Hole of F Reactor

The general layout of the E test hole facility is shown in Figure 1. This facility is similar to its precursor,(1) but it is water-cooled and it can be positioned approximately 11 inches further into the reactor. The position used for the measurements considered here is shown in Figure 1. An additional single measurement was made with pocket 18 moved out to two inches from the thermal shield.

The thermal flux was measured using the reaction $Fe^{58}(n,\gamma)Fe^{59}$ (σ = 0.98 barns)(6) as the thermal neutron detector. The contribution from epithermal neutrons was observed to be six percent in pocket No. 1. Since this is small, and would be expected to drop at about the same rate as the fission flux, it has not been corrected for in these measurements.

The fission flux was determined using the $Al^{27}(n,\alpha)Na^{24}$ and the $Fe^{54}(n,p)Mn^{54}$ reactions. The effective cross section for the Al^{27} reaction was taken as 0.60 mb.(4) Because of the large variation in reported cross section for the Fe^{54} reaction,(4) the cross section was measured relative to the Al^{27} reaction and found to be 89 ± 9 mb. "Specpure"* aluminum and iron samples were used for the irradiation to reduce interference from impurities. The irradiated specimens were dissolved in acid and aliquots taken for the measurements. Na^{24} and Fe^{59} were measured from the 4 Mev addition peak and the 1.32 Mev photopeak, respectively, using a gamma ray spectrometer equipped with a five-inch diameter by five-inch thick well crystal which had been calibrated with 4π standardized sources. The Mn^{54} was chemically separated and its counting rate compared on a gamma-ray spectrometer with a standard Mn^{54} source which was calibrated by X-ray--γ-ray coincidence counting.

* Johnson, Matthey, and Co., Ltd., 73/83 Hatton Garden, London, E.C.1.

The thermal and fission fluxes and the thermal to fission flux ratios are plotted in Figures 2 and 3, respectively. The ratio of the fission fluxes as measured with Al^{27} and Fe^{54} detectors are listed in Table I.

TABLE I

RELATIVE FORMATION RATES OF TWO RADIOISOTOPES BY THRESHOLD REACTIONS

Sample Position (inches)*	Cross Section Mn^{54}/Na^{24}**
10.75	0.91
9	0.99
5.5	1.03
2	1.02
1.5	1.19
- 5	0.93
- 8.5	0.92
- 12	1.12
- 15.5	1.06

* Relative to outside rows of fuel elements.
** Average of the nine values normalized to unity.

Since the ratio is constant within experimental error, the shape of the fission spectrum as measured by these two threshold reactions does not change significantly over these locations.

Similar flux measurements were made in the KE reactor Quickie facility. The observed fluxes at two positions in this facility are included with a summary of other flux measurements in Table II.

Flux Measurements in the Poison Column

Thermal and fission neutron fluxes were measured in a poison column at DR reactor (a process tube which contained aluminum dummies rather than the normal fuel load). This measurement entailed the irradiation of a sodium standard and an aluminum specimen in the "central zone" of a reactor and a calculation of the thermal and fission fluxes from the Na^{24} produced in the two samples.

	σ (mb)
$Al^{27}(n,\alpha)Na^{24}$	0.60
$Na^{23}(n,\gamma)Na^{24}$	536

The thermal and fission fluxes at this location were 1.0×10^{14} and 3.1×10^{12}. The thermal to fission flux ratio was thus 32. They are compared with other test locations in Table II.

TABLE II

SUMMARY OF FLUX MEASUREMENTS IN NEUTRON ACTIVATION TEST LOCATIONS

Test Facility and Position	Thermal Flux	Fission Flux	Ratio Thermal/Fission
F Reactor Quickie Facility Pocket No. 1*	9.0×10^{13}	9.0×10^{11}	33
In reflector about 2" from thermal shield.	2.5×10^{12}	3.2×10^{9}	794
KE Quickie Facility Pocket No. 2	7.6×10^{13}	4.3×10^{12}	17.7
In reflector about 2" from thermal shield.	5.7×10^{12}	1.47×10^{10}	387
BR Poison Column Near center of reactor.	1.0×10^{14}	3.1×10^{12}	32

* See position from Figure 1.

Flux Measurements in Aluminum Process Tubing

The thermal and fission neutron fluxes in process tubing were calculated from measurements of the Na^{24}, Cu^{64} and inert copper present in samples of aluminum tubing from B and C reactors. The samples were taken from near the center of the process tubes. The observed fission and thermal fluxes and their ratios are listed in Table III.

TABLE III

THERMAL AND FISSION FLUXES IN PROCESS TUBE METAL

Tube No.	Reactor	Thermal Flux	Fission Flux	Thermal/Fission
3764	B	4.98×10^{13}	1.13×10^{13}	4.39
Unknown	C	3.41×10^{13}	8.28×10^{12}	4.11

The Na^{24} was measured as described in the section "Measurements in the E Test Hole." The Cu^{64} was measured by a standard electroplating procedure with inert copper added as a carrier. The inert copper in a sample of the aluminum tubing was measured after the initial Cu^{64} had decayed, by a neutron activation and subsequent analysis of the Cu^{64}.

DISCUSSION

These studies were designed to determine the approximate fission and thermal fluxes and their ratios at the various neutron activation test locations and also in aluminum process tubing. Samples of aluminum (from process tubing) with its associated copper impurity served as detectors for the precise measurement of fission and thermal fluxes in process tubing. Since both the Na^{24} (daughter of Al^{27}) and Cu^{64} (daughter of Cu^{63}) have short half-lives, 15 hours and 12.8 hours, respectively, radioequilbrium is attained in a few days of reactor operation. The precision of the flux measurement is thus independent of past reactor operation.

The lattice spacing in KE reactor is 7.5 inches compared with 8.375 inches in the old reactors. This accounts for the lower thermal to fission flux ratio in the KE reactor test facility than in the F reactor facility (see Table II).

In performing an activation analysis where the daughter radioisotopes can be formed from both (n,γ) and threshold reactions, the location for minimum interference from the threshold reactions would be near the edge of the reflector of F reactor (see Table II). On the other hand, interference from an n,γ reaction would be a minimum in the control zone of KE reactor. (A cadmium absorber would probably be used to remove thermal neutrons in any case.)

The n,γ contribution from resonance neutrons has been neglected in all of the flux measurements. The cadmium ratio in pocket 1 of the F reactor Quickie facility was 16 for the $Fe^{58}(n,\gamma)Fe^{59}$ reaction, indicating about a six percent contribution from resonance neutrons. The contribution as the sample is moved into the graphite reflector would drop off at about the same rate as the fission flux, thus becoming extremely small. By the same reasoning, the resonance contribution in the KE reactor lattice would be larger than that observed at F reactor.

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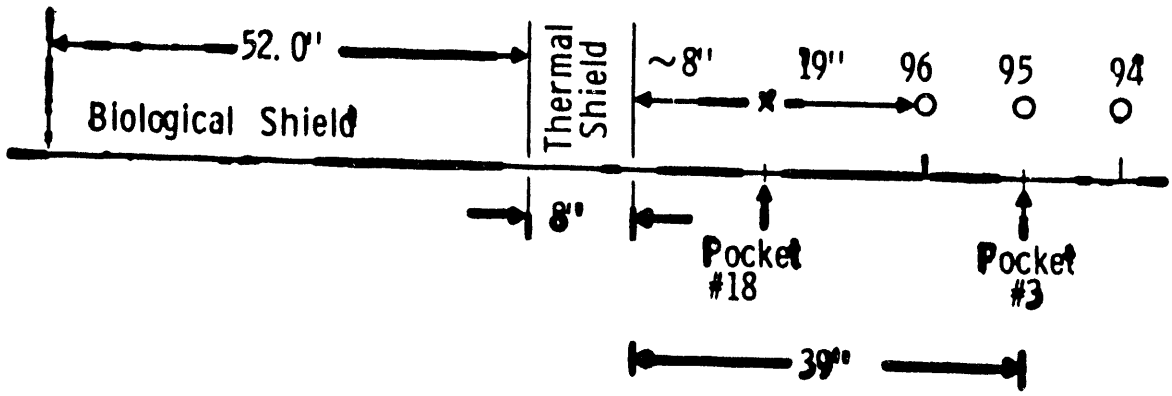
General Layout



Exposure Rod Enlargement

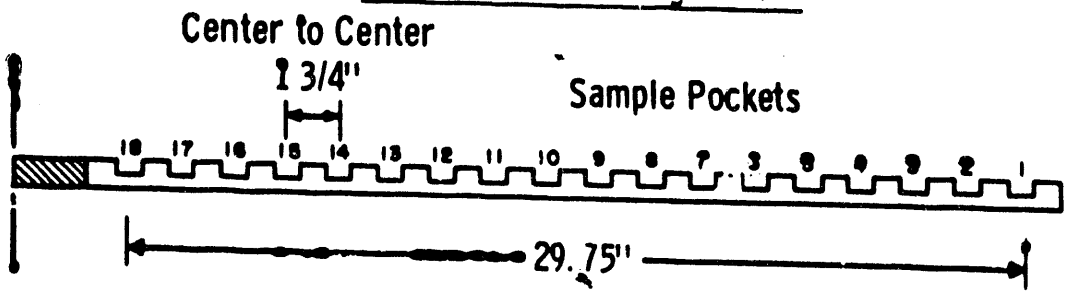


FIGURE 1

QUICKIE TEST FACILITY AND POSITION OF THE ALUMINUM ROD SAMPLE HOLDER USED FOR THE IRRADIATION

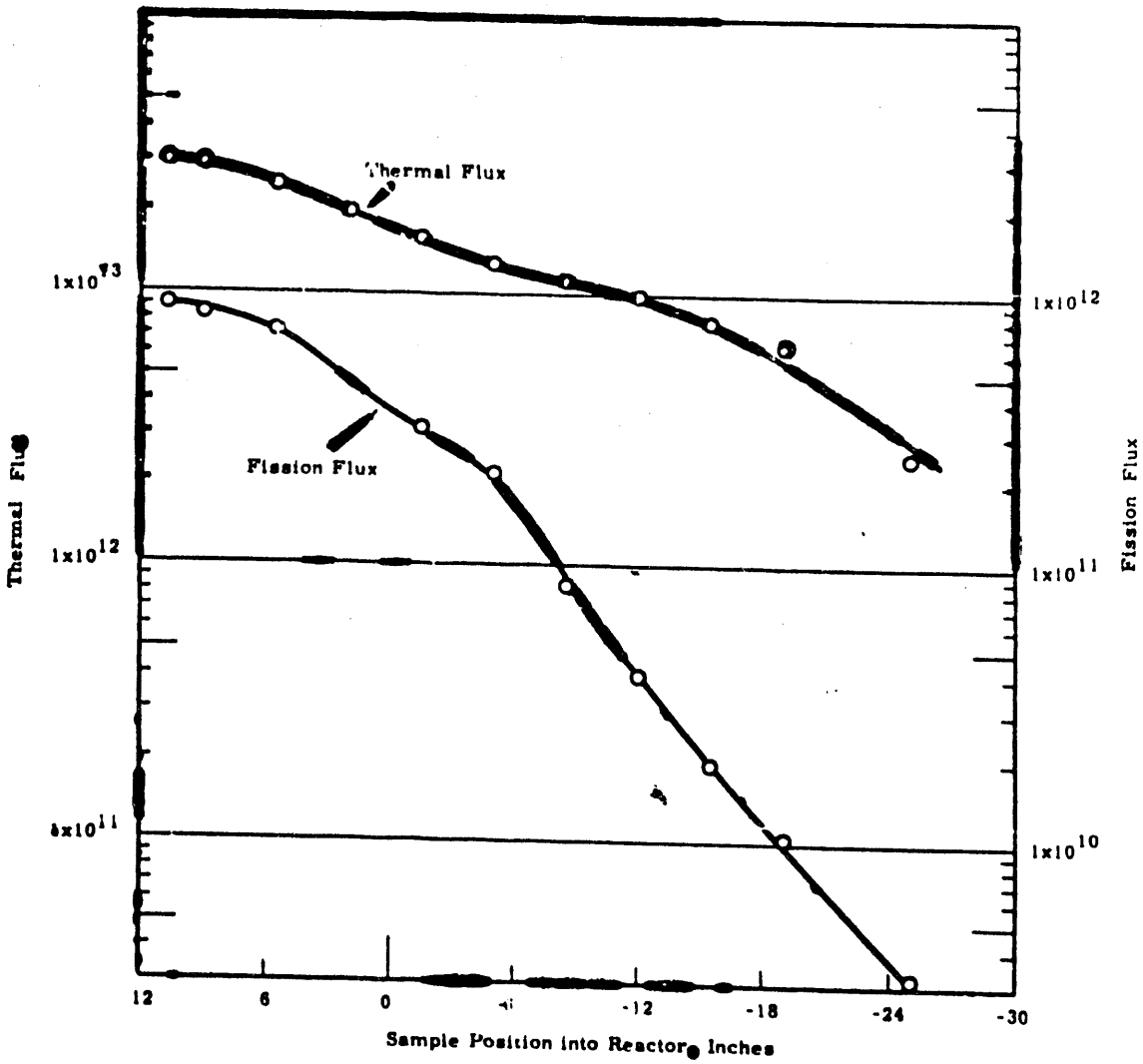


FIGURE 2

Thermal and Fission Fluxes in the Quickie Facility. Distance Measured from Center of Outside Row of Fuel Elements.

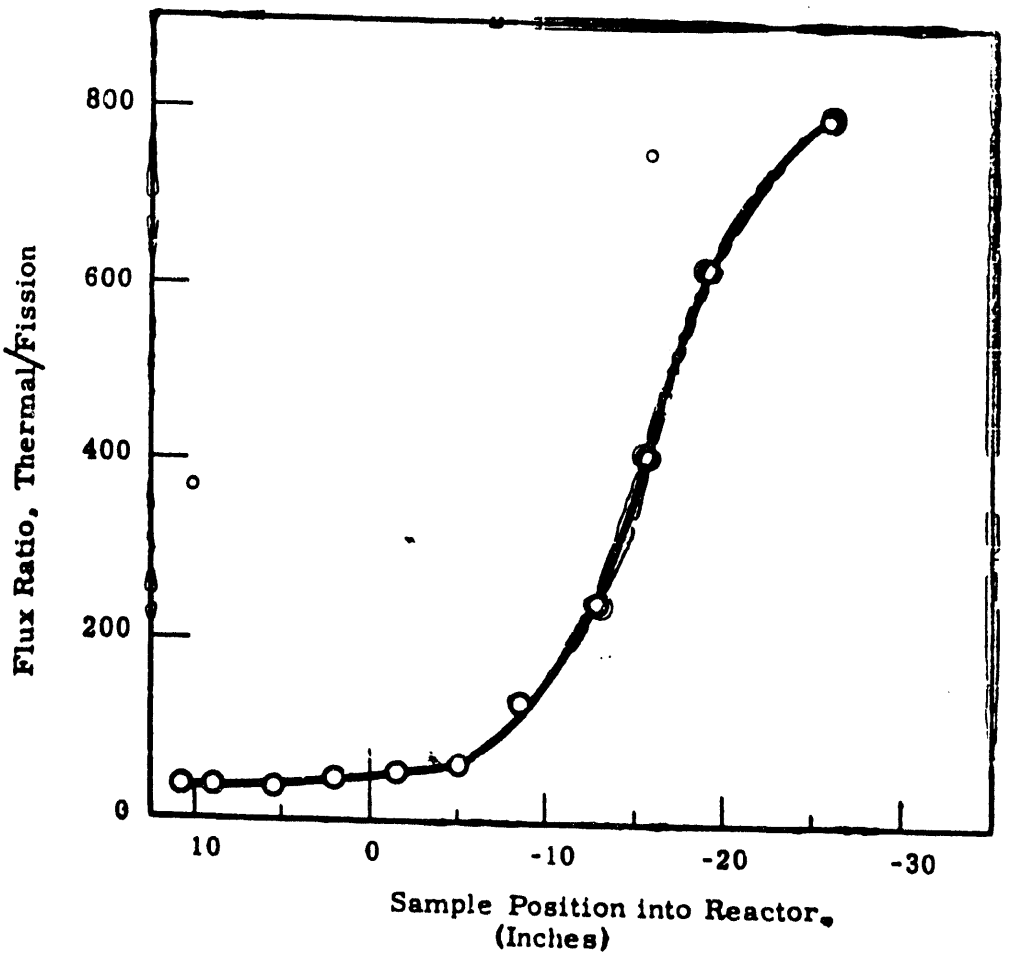


FIGURE 3

Ratio of Thermal to Fission Fluxes in the Quickie Facility. Distance Measured from Center of Outside Row of Fuel Elements

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