MASSACHUSETTS INSTITUTE OF TECHNOLOGY
DEPARTMENT OF NUCLEAR ENGINEERING
Cambridge, Massachusetts 02139

MIT-3944-4

MITNE-111

REACTOR PHYSICS PROJECT PROGRESS REPORT NO. 2

September 30, 1969

Contract AT(30-1)-3944
U.S. Atomic Energy Commission

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AEC Research and Development Report
(TID-4500, 47th Edition)

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AEC Research and Development Report

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ABSTRACT

This is the second annual report in an experimental and theoretical program to develop and apply single and few element heterogeneous methods for the determination of reactor lattice parameters.

During the period covered by the report, October 1, 1968 through September 30, 1969, work was primarily devoted to measurement of the heterogeneous fuel element parameters (Γ, η and A) of 19- and 31-rod clusters of plutonium-containing fuel. Methods development research focused on determination of the epithermal absorption constant, A. Calculations and an analysis of data reported in the literature were made to assess the applicability of heterogeneous methods to H_2O-moderated systems.

Advanced gamma spectrometric methods using Ge(Li) detectors were applied to the analysis of prompt and delayed gamma spectra from fertile and fissile materials and from fuel elements. These methods were used successfully for nondestructive analysis of the composition of fuel elements. A feasibility study was performed on an in-pile gamma spectrometer.

Two fuel pins irradiated to a burnup of approximately 20,000 MWD/MT in the Dresden reactor were received and preparations made for their analysis and use in reactor physics experiments.

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1. INTRODUCTION

1.1 Foreword

This is the second annular progress report of the Reactor Physics Project of the Massachusetts Institute of Technology. This project was initiated January 1, 1968 with the objective of developing and applying single and few element methods for the determination of reactor physics parameters. Development of these methods should increase the ability to evaluate the reactor physics characteristics of new and promising types of reactor fuel at very low cost.

The project, scheduled for completion during the coming contract year, has focused on two demonstration applications of single element methods. The first involves an investigation of 19- and 31-rod clusters of plutonium-containing fuel, and the second involves irradiated fuel pins containing both plutonium and fission products.

This report summarizes the work completed during the period of October 1, 1968 through September 30, 1969, which has primarily been concerned with clustered fuel.

1.2 Research Objectives and Methods

The basic objective of the present research is the experimental determination of those parameters of heterogeneous reactor theory which characterize the neutronic properties of a fuel element. They are:

Γ = asymptotic thermal neutron flux at the fuel element's surface per thermal neutron absorbed by the element.

η = number of fast neutrons emitted by the fuel element per thermal neutron absorbed in the fuel element, and

A = number of epithermal neutron absorptions by the fuel element per unit slowing-down density.

Previous work at M.I.T., summarized in Ref. 1, showed that η and A can be inferred from in-rod foil activation experiments and that Γ can be determined from radial foil activation traverses external to the rod. Later work, done as part of the present project (2), developed a method for measuring η relative to a standard, through cadmium ratio determinations external to the fuel element. Chapter 3 of this report deals with the measurement of the parameter A by irradiation of molybdenum and gold foils on the surface of the fuel element. Thus, it is now possible to determine all three parameters without cutting into the fuel, which may contain plutonium and fission products, and thereby to avoid the attendant contamination problem. In Chapter 2 a series of demonstration experiments are described in which the methods were applied to clusters of plutonium recycle fuel rods in a simulated D_2O-moderated reactor application.

In addition to the foil activation methods which form the basis of the experimental-heterogeneous approach, high resolution gamma spectroscopy using Ge(Li) detectors has proven useful for the non-destructive assay of fuel element composition. Chapter 4 of this report summarizes applications involving both prompt and delayed gammas. This work will become even more important during the coming year since the fuel pins to be investigated have been previously irradiated to a burnup of approximately 20,000 MWD/MT, and there is no other satisfactory way to establish their composition as a prelude to further reactor physics experimentation.

1.3 Staff

The project staff, including thesis students, during the report period was as follows:

M. J. Driscoll, Associate Professor of Nuclear Engineering
T. J. Thompson, Professor of Nuclear Engineering (through 6/1/69)
I. Kaplan, Professor of Nuclear Engineering (on sabbatical leave during report period)
D. D. Lanning, Professor of Nuclear Engineering (since 6/1/69)
N. C. Rasmussen, Professor of Nuclear Engineering

F. M. Clikeman, Associate Professor of Nuclear Engineering

A. T. Supple, Jr., Engineering Assistant

G. E. Sullivan, Technician

V. Agarwala, Research Assistant (since 2/15/69)

J. N. Donohew, Research Assistant (through 9/15/69)

G. T. Hamilton, S.M. student (through 6/15/69)

T. L. Harper, AEC Fellowship, Part-time Data Analyst (through 9/1/69)

Y. Hukai, Research Assistant, Sc.D. student

T. J. Kelley, S.M. student (through 6/15/69)

T. C. Leung, Research Assistant, S.M. student (through 6/15/69)

E. L. McFarland, Research Assistant, S.M. student

S. S. Seth, Research Assistant, Sc.D. student

J. M. Sicilian, B.S. student (through 6/15/69)

1.4 References

(1) Heavy Water Lattice Project Final Report, MIT-2344-12, MITNE-86, September 30, 1967.

(2) Reactor Physics Project Progress Report, MIT-3944-1, MITNE-96, September 30, 1968.

2. PARAMETER MEASUREMENTS AND CALCULATIONS FOR 19- AND 31-ROD PLUTONIUM-FUELED CLUSTERS

S. Seth

2.1 Introduction

Since January 1968 the Reactor Physics Project at M.I.T. has been carrying out experimental and analytical research with the primary objective of perfecting and applying single element methods for the determination of reactor physics parameters, primarily those of heavy water moderated lattices typical of pressure-tube type reactors. The major demonstration experiment, employed to assess the ultimate utility of the single element method, involved measurements on 19- and 31-rod clusters of Type B Simulated Burned Fuel from the USAEC/AECL Cooperative Program. Full lattices of this same plutonium-containing fuel had previously been investigated extensively, both experimentally and analytically, by the Savannah River Laboratory (1).

Details concerning the development of the experimental methods have already been reported (2), (3). Thus, this chapter will be concerned only with a brief review of the work and will primarily be concerned with the experimental results obtained. A complete topical report on this entire research area is being prepared:

> S. S. Seth, M. J. Driscoll, I. Kaplan, T. J. Thompson, D. D. Lanning, "A Single Element Method for Heterogeneous Nuclear Reactors," MIT-3944-3, MITNE-109, (May 1970 est.)

2.2 Experimental Methods

Methods have been developed for the determination of η, Γ, and A in single element exponential experiments using the MITR heavy water exponential facility (2), (3). The parameter η is determined by measuring the cadmium ratio of gold foils in the moderator external to the fuel

element; the parameter Γ is determined by measuring the distance to the thermal flux peak in the moderator by using bare gold foils; and the parameter A is determined by measurement of cadmium-covered gold and molybdenum foil activities on the rod surface. The parameters are measured relative to a known standard: a one-inch-diameter, natural uranium metal rod in the present work.

Tables 2.1 through 2.3 and Fig. 2.1 describe the characteristics of both the standard fuel assembly and the test fuel assemblies for which Γ, η and A were determined through application of the experimental techniques.

2.3 Comparison of Results with SRL Lattice Data

With the methods and formulae described in last year's annual report (2), the infinite lattice multiplication factor, k_∞, and the material buckling, B_m^2, can be calculated from the single rod parameters Γ, η and A. It is also possible to compute the thermal utilization, f, from Γ and the resonance escape probability, p, from A.

Table 2.4 summarizes the results of the present work together with that of SRL for comparison. The five parameters η, p, f, k_∞ and B_m^2 are compared. Excellent agreement is evident between the MIT and SRL results.

Based on these results, it may be concluded that single element methods can compete favorably with full-lattice methods for the determination of many reactor physics parameters of new types of fuel.

2.4 References

(1) Baumann, N. P. et al., "Lattice Experiments with Simulated Burned Fuel for D_2O Power Reactors," DP-1122, February 1968.

(2) Seth, S. S., Calculation of Lattice Parameters, Chapter 2, in Reactor Physics Project Project Report, MIT-3944-1, MITNE-96, September 30, 1968.

(3) Chapter 3, this report.

TABLE 2.1

Fuel Elements Tested

Run Number	Fuel Element	Geometric Characteristics	Active Length
1	Single Natural Uranium Rod	Tube Housing: 1.01-inch O.D.	54 inches
2	19-Rod UO_2-PuO_2 Cluster	Tube Housing: 3.125-inch O.D. Pitch: 0.597 inch	54 inches
3	31-Rod UO_2-PuO_2 Cluster	Tube Housing: 4.000-inch O.D. Pitch: 0.597 inch	54 inches

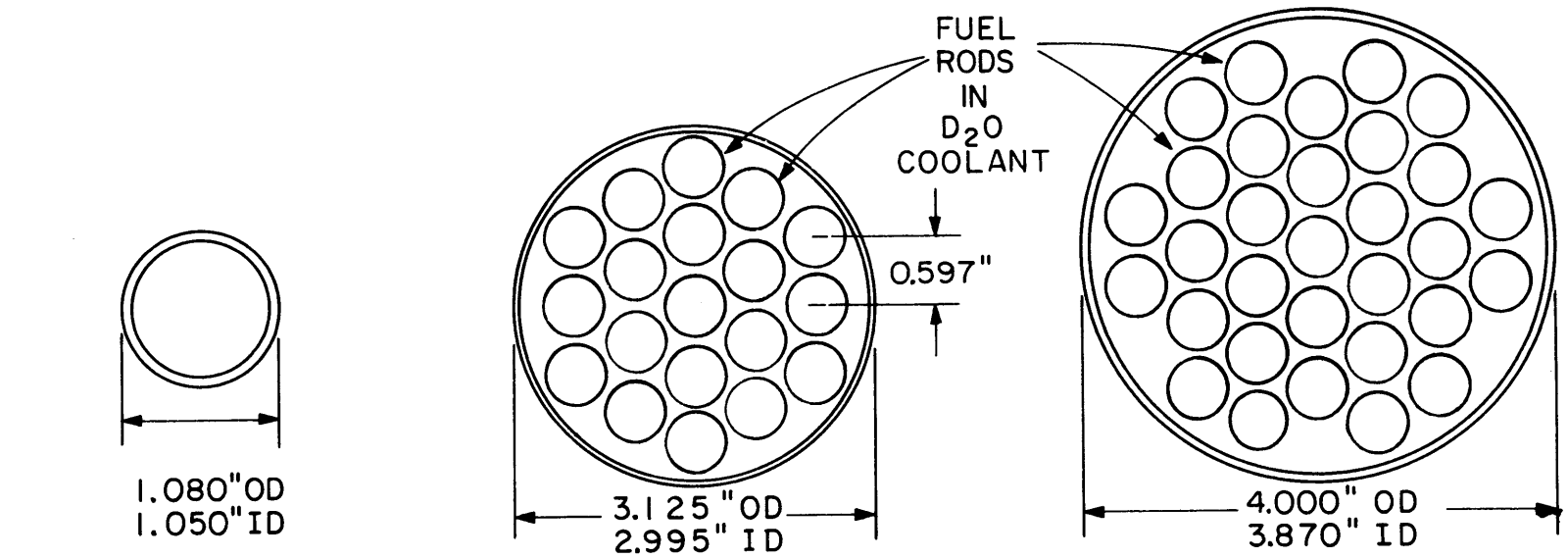

FIG. 2.1 FUEL ELEMENT ARRANGEMENTS

TABLE 2.2

Description of Fuel and Cladding

Run No.	FUEL				CLADDING		
	Material	% Fissile	ρ (gm/cm^3)	O.D. (in.)	Material	O.D. (in.)	Thickness (in.)
1	U-Metal	0.71 (U-235)	18.9	1.01	1100 Al	1.080	0.028
2, 3	UO_2-PuO_2	0.298 (U-235) 0.250 (Pu-239)	10.4	0.500	6063-T Al	0.547	0.020

TABLE 2.3

Properties of Type B, USAEC-AECL Cooperative Program, Simulated Burned Fuel

A) Isotopic Composition, Wt % of Total U + Pu

U-238	U-235	Pu-239	Pu-240	Pu-241	Pu-242
99.431	0.30	0.25	0.016	0.002	0.001

B) Individual Fuel Rods

Pellets: sintered coprecipitated oxide, 95% theoretical density, 0.500 ± 0.002-inch diameter

Clad: 6063-T6 aluminum, I.D. 0.507 ± 0.004 inch, wall thickness 0.020 ± 0.002 inch, length 54 inches

Color Code: Gold

TABLE 2.4

Comparison of Single Element and Lattice Results
for D_2O Moderated and Cooled, Plutonium-Containing Fuel Clusters

TYPE OF RESULT	η	p	f	k_∞	B_M^2, $cm^{-2} \times 10^6$
A. 19-Rod Cluster					
9.33-Inch Lattice Spacing					
(1) MIT Single Element	1.385	0.8750	0.9793	1.1868	540 ± 45
(2) SRL Calculation	1.407	0.8556	0.9610	1.1566	484
(3) SRL Lattice Expt.	—	—	—	—	524 ± 15
B. 31-Rod Cluster					
9.33-Inch Lattice Spacing					
(1) MIT Single Element	1.438	0.7926	0.9863	1.1241	458 ± 43
(2) SRL Calculation	1.451	0.79	0.9621	1.1028	425
(3) SRL Lattice Expt.	—	—	—	—	501 ± 15
C. 31-Rod Cluster					
12.12-Inch Lattice Spacing					
(1) MIT Single Element	1.412	0.8684	0.9733	1.1937	472 ± 44
(2) SRL Calculation	1.419	0.8486	0.9513	1.1458	416
(3) SRL Lattice Expt.	—	—	—	—	429 ± 20

3. DETERMINATION OF THE EPITHERMAL PARAMETER, A

E. McFarland

3.1 Introduction

In heterogeneous reactor theory the parameter A is defined as the number of epithermal absorptions per unit slowing-down density. It is related to the resonance escape probability, p, of unit cell theory:

$$p = 1 - A/V_m, \tag{3.1}$$

where V_m is the cross-sectional area of the moderator in the unit cell.

A method has been developed to determine A in a single element exponential experiment by measuring the ratio of gold to molybdenum foil activities at the surface of the fuel element (1). A multigroup computer code is used to establish relative correlation lines (see, for example, Fig. 3.1) relating the foil activity ratio to A, and absolute normalization is achieved by comparison of the unknown with a standard element for which the value of A is known.

This procedure was used to measure A for the 19- and 31-rod fuel clusters discussed in Chapter 2.

3.2 Theoretical Basis for the Method

As noted by Lefevre (2), the relation between A and the resonance escape probability, Eq. 3.1, suggests that A can be related to the ratio of slowing-down densities above and below the major U^{238} and U^{235} absorption resonances. Calculations show that gold, with a resonance at 4.9 eV, and molybdenum, with a resonance at 480 eV, span a region covering over 90% of the resonance absorptions of concern. The choice of these two materials has an additional advantage: the half-lives of the activation products are nearly identical, 67 hours for Mo^{99} and 65 hours for Au^{198}. We thus have suitable foil materials with which to measure the relative ratio of fluxes or slowing-down densities above and below the resonance region. The following simple considerations

illustrate the basic principles underlying the relationship between the measured foil ratios and the parameter A, and also serve to introduce the complicating factors which require use of more sophisticated numerical methods and introduction of a standard for comparison.

Consider a line source emitting one neutron at $\tau = 0$ and absorbing (1 - p) neutrons in a resonance sink at $\tau = \tau_r$. Then, for energies 1 and 2 bracketing the resonance, the self-induced slowing-down density at the origin is:

$$q_1 = \frac{1}{4\pi\tau_1} , \tag{3.2}$$

$$q_2 = \frac{1}{4\pi\tau_2} - \frac{(1-p)}{4\pi(\tau_2-\tau_r)} . \tag{3.3}$$

Upon dividing Eq. 3.3 by Eq. 3.2 and with (1 - p) replaced by using Eq. 3.1, the resulting expression becomes:

$$\frac{q_2}{q_1} = \frac{\tau_1}{\tau_2} - \left(\frac{\tau_1}{\tau_2 - \tau_r}\right) \frac{A}{V_m} . \tag{3.4}$$

Finally, since the ratio of gold to molybdenum activities, R, is proportional to q_2/q_1, a general relation for A is:

$$R = a - bA . \tag{3.5}$$

Equation 3.5 suggests that the foil activity ratio is quite simply related to the epithermal parameter A, a circumstance which motivated further evaluation of the proposed method.

The simple analytic model presented above has some important defects. In particular, an actual fuel rod is not a line source or sink and finite size effects must therefore be taken in account. To remedy these defects the most elegant means at hand, the ANISN computer code, was used to generate relative curves of R versus A for both unknown and standard fuel rods. The introduction of a standard serves an important purpose. Absolute determination of the activity ratio, R, would require knowledge of parameters such as self-shielded resonance absorption cross sections for gold and molybdenum. Use of a standard, however, permits determination of a normalization factor relating

calculated and experimental foil ratios:

$$f = \frac{R}{R_e} . \tag{3.6}$$

The parameter f is determined from the calculated and measured foil ratios for the standard rod and then applied to the experimental data for the unknown rods to obtain R values consistently normalized to the computer-generated data.

3.3 Use of ANISN Calculations

The modified ANISN computer code used in this work is described in detail in Ref. 3. Basically, ANISN is a one-dimensional multigroup S_N transport program. It is used in the present instance to solve the neutron transport equation for a cylindrical, D_2O-moderated, exponential tank containing a centrally located cylindrical fuel element. Exponential experiments are simulated by adding an effective in-leakage cross section, $-D\gamma^2$; where γ^2 is the axial buckling, determined by trial and error to achieve a steady-state neutron balance.

Group 9 of the 16-group ANISN calculations covers the resonance of Mo^{98}, while group 12 includes that of Au^{198}. Thus the ratio of group 12 to group 9 fluxes at the surface of the fuel rod was used as the calculated, relative foil activity ratio, R. To prepare the R versus A plot, the effective A value was varied for each fuel type considered by changing the self-shielding of the U^{238} absorption cross sections in the multigroup set. Since A_{28} is directly proportional to the resonance integral, this mathematical artifice permitted arbitrary variation of A over the range of interest. Figure 3.1 shows the R versus A plots prepared in this manner for the standard and unknown fuel rods used in the present work. The "exact" value of A for the standard one-inch-diameter, uranium metal rod was taken to 20.39 cm^2, as calculated from the relation:

$$A = \left[N^{28} ERI^{28} + N^{25} RI_{\infty}^{25} \right] \frac{V_f}{\xi \Sigma_s} , \tag{3.7}$$

where ERI^{28} = effective resonance integral for U^{238} as determined

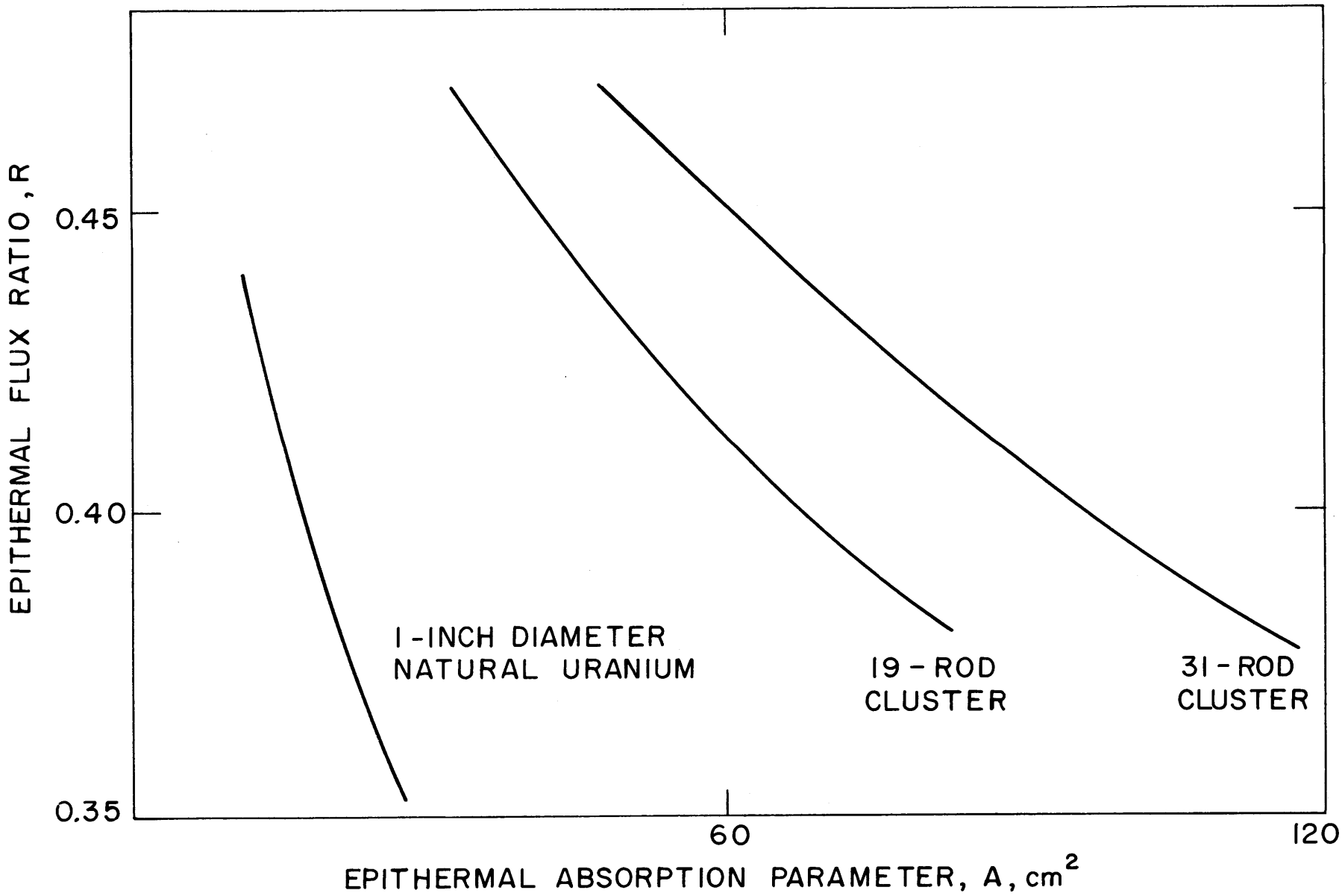

FIG. 3.1 EPITHERMAL FLUX RATIO (R) VERSUS EPITHERMAL ABSORPTION PARAMETER (A)

from Hellstrand's correlations (4).

Calculations were also performed with the ANISN code to show that the differences between single-rod and full-lattice experiments were not consequential for present purposes. For example, the mean age to resonance capture for a single one-inch rod was found be be 77.3 cm^2, as opposed to 81.2 cm^2 for the same rod in a typical complete lattice.

3.4 Experimental Procedure and Analysis of Results

The experiments were carried out in the MITR's 36-inch-diameter, D_2O-moderated exponential facility. Three fuel elements were tested: a one-inch-diameter, natural uranium metal rod (the standard) and the 19- and 31-rod UO_2-PuO_2 fuel clusters described in Chapter 2.

The 0.25-inch-diameter foils used for the experiment had the following characteristics: 1.24 wt % Au in Al 3 mils thick, and 23.8 wt % Mo^{98} metal 25 mils thick. The foils were irradiated together inside a disc-shaped can of 30-mil-thick cadmium. Four foil packets were used for each experimental run. Three were attached to the fuel element surface using mylar tape, and the fourth was used as a standard to correct for residual activity from previous irradiations.

The foil packets were irradiated from 12 to 36 hours and counted for 60 minutes so as to accumulate at least 50,000 counts. The counting system employed was a standard NaI well-counter, and all foils were integral-counted with a baseline setting just below the gold photopeak at 0.412 MeV. All foil activities were corrected for background, residual activity and foil weight. Table 3.1 shows the resulting foil activity ratios.

TABLE 3.1

Experimental Au/Mo Foil Activity Ratios

Fuel	Re
1-Inch-Diameter, Natural Uranium Metal	0.9510 ± 0.0006
19-Rod Cluster	1.044 ± 0.028
31-Rod Cluster	1.025 ± 0.013

From Fig. 3.1, at A = 20.39 cm^2 for the standard rod, R = 0.387 and therefore the normalization factor f = 0.387/0.951 = 0.407. Entering Fig. 3.1 at f times Re for the two unknowns yields values of A of 58.0 and 81.5 cm^2 for the 19- and 31-rod clusters, respectively.

3.5 Discussion of Results

The values of A obtained in the above manner were used to calculate resonance escape probabilities for complete lattices of the subject fuel. These values were then compared with values determined by Savannah River Laboratory (5) in conjunction with their critical lattice assembly studies of the same fuel. Table 3.2 shows the results of this comparison.

TABLE 3.2
Comparison of SRL and MIT Results

Clusters	Lattice Spacing	Resonance Escape Probability	
		SRL	MIT
19-Rod	9.33-Inch	0.856	0.875
31-Rod	9.33-Inch	0.790	0.793
31-Rod	12.12-Inch	0.849	0.868

The relatively good agreement between the SRL and MIT values is evident. The largest difference occurs at the largest lattice spacing where corrections for the nonuniform slowing-down density across the unit cell become less accurate.

It is concluded that the Au/Mo foil technique is an adequate method for the determination of the epithermal parameter A in single-element experiments. It has the major advantage of not requiring in-rod foil activation experiments. The major shortcoming is the reliance upon concurrent computer code calculations. This restriction could be removed, however, if standard elements were available having the same equivalent diameter as the unknown fuel assemblies. In such circumstances, Fig. 3.1 could be generated using only experimental data.

3.6 References

(1) McFarland, E., "Experimental Determination of the Epithermal Absorption Parameter, A," M.S. Thesis, M.I.T. Nuclear Engineering Department, November 1969.

(2) Lefevre, Y.-M., Determination of the Epithermal Parameter, A, Chapter 6 in "Reactor Physics Project Progress Report," MITNE-96, MIT-3944-1, September 30, 1968.

(3) Donohew, J. N. and J. D. Eckard, Numerical Methods, Chapter 7 in "Reactor Physics Project Progress Report," MITNE-96, MIT-3944-1, September 30, 1968.

(4) Hellstrand, E., "Measurement of Resonance Integrals," in Reactor Physics in the Resonance and Thermal Regions, Vol. 2, A. J. Goodjohn and G. C. Pomraning, eds., M.I.T. Press, 1966.

(5) Baumann, N. et al., "Lattice Experiments with Simulated Burned-Up Fuel for D_2O Power Reactors," DP-1122, February 1968.

4. GAMMA SPECTROSCOPY

Y. Hukai, N. C. Rasmussen, J. M. Sicilian, and T. Kelley

4.1 Introduction

The objective of the work described in this chapter has been the application of nondestructive gamma spectrometry methods to the analysis of fuel elements of interest to the single-element physics program. Previous work in this area has been reported in last year's annual report (1), and a topical report is under preparation which describes this phase of the research program in detail:

> Y. Hukai, N. C. Rasmussen, M. J. Driscoll, "Some Applications of Ge(Li) γ-Ray Spectroscopy to Fuel Element Assay," MIT-3944-5, MITNE-113 (April 1970 est.).

During the report period, work was carried out on two major applications involving both prompt and delayed gamma analysis; and feasibility studies were completed evaluating the construction of an in-pile spectrometer and the analysis of irradiated fuel from a power reactor. Results of this work are summarized in the following sections of this chapter.

4.2 Prompt Gamma Analysis

This phase of the work has been primarily concerned with determination of reference prompt gamma spectra for the nuclides U^{238}, Th^{232}, U^{235} and Pu^{239}. Data on the first two materials were reported in last year's annual report (1). During the past year, work was concentrated on the difficult task of measuring prompt capture gamma spectra of the fissile species. Preliminary results are available for U^{235}, which will be reported in this chapter. Complete results, including an application of the data for fuel assay, will be presented in MITNE-113.

The apparatus used for these measurements was again the versatile facility constructed at MITR beam port 4TH1 by Orphan, Harper and Hukai, as described in Refs. 2 and 3. This facility can be used to measure prompt capture gamma spectra produced by a 4.2×10^8 n/cm^2-sec flux of well thermalized neutrons (cadmium ratio $\approx$ 80)

using a combination of NaI and Ge(Li) detectors in free, triple-coincidence and Compton-suppression modes. The 10-cc Ge(Li) detector has an energy resolution of 3.2 keV at 0.511 MeV and 6.5 keV at 6.5 MeV.

The major difficulty encountered in capture gamma studies of fissile materials is the high background produced by the fast fission neutrons emitted by the sample. Thus, the key to achieving acceptable results proved to be the evolution of a suitable filter and collimator design: four inches of paraffin followed by one inch of LiF and 0.25 inch of lead were used as a filter together with the standard 1-inch and 0.75-inch I D. lead collimators commonly used for less demanding applications.

Preliminary results for the prompt gamma line spectra associated with neutron absorption by U^{235} are shown in Table 4.1. Some of the higher energy features of the spectrum appear to be different for U^{235} and Pu^{239}; this is currently under investigation.

4.3 Delayed Gamma Analysis

The objective of this phase of the spectrometry work has been the development of a fuel assay method based on the difference in fission product yields between Pu^{239} and U^{235}. This has been possible, based upon use of a fuel rod containing uranium of a known enrichment and foils of enriched uranium and plutonium.

Consider, as an example, the Zr^{92} fission product chain:

$$Kr^{92} \xrightarrow{3.0\ \text{sec}} Rb^{92} \xrightarrow{5.3\ \text{sec}} Sr^{92} \xrightarrow{2.7\ \text{hrs}} Y^{92} \xrightarrow{3.6\ \text{hrs}} Zr^{92} .$$

The activity of the Sr^{92} member of this chain is given by the relation:

$$A_s = N \sigma \phi F(\lambda_k, \lambda_r, \lambda_s, Y_k, Y_r, Y_s, ti, td) , \qquad (4.1)$$

where

N = number of atoms of fissile element,

σ = fission cross section of fissile element,

ϕ = neutron flux.

TABLE 4.1

Prompt Gamma Lines Associated with Thermal Neutron Absorption by U^{235}

Line Number	Energy (keV)	Relative Intensity*
1	2195.9	1.0
2	2393.7	1.3
3	2554.5	0.6
4	2569.1	1.4
5	2639.2	1.1
6	2754.2	1.1
7	2790.4	1.1
8	2868.2	0.8
9	2942.9	0.9
10	3286.9	1.3
11	3314.0	1.0
12	3575.2	1.4
13	3599.7	1.7
14	4039.4	0.4
15	4076.3	1.0
16	4136.0	0.5
17	4179.4	0.4
18	4366.1	1.0
19	4451.5	1.4
20	4884.0	0.4

*Relative to line No. 1 assigned a value of 1.0

F = a complicated function of the decay constants of Kr^{92}, Rb^{92} and Sr^{92} (λ_k, λ_r, λ_s), their fission yields (Y_k, Y_r, Y_s), and the irradiation and decay times (ti, td).

For ti and td on the order of hours, the function F simplifies to:

$$F = (Y_k + Y_r + Y_s)(1 - e^{-\lambda_s ti})\, e^{-\lambda_s td} . \tag{4.2}$$

Thus, fission product activities are a direct measure of the fissile nuclide content. Irradiation of standard and unknown fuel rods will thus give rise to nuclide activities obeying the following relations:

$$A^s = N_5^s \sigma_5 \phi^s F_5 \epsilon , \tag{4.3}$$

$$A^x = N_5^x \sigma_5 \phi^x F_5 \epsilon + N_9^x \sigma_9 \phi^x F_9 \epsilon , \tag{4.4}$$

where the subscripts 5 and 9 refer to U^{235} and Pu^{239}, respectively; the superscripts s and x refer to the standard and unknown fuel rods, respectively; and ϵ is the total detection efficiency.

If Eqs. 4.3 and 4.4 are written for two different fission products, the resulting set of equations can be solved for the Pu^{239} and U^{235} content of the unknown:

$$\frac{N_9^x}{N_5^s} = \frac{\phi^s}{\phi^x} \frac{\dfrac{A^{1x}}{A^{1s}} - \dfrac{A^{2x}}{A^{2s}}}{R_1 - R_2} , \tag{4.5}$$

$$\frac{N_5^x}{N_5^s} = \frac{\phi^s}{\phi^x} \frac{R_1 \dfrac{A^{2x}}{A^{2s}} - R_2 \dfrac{A^{1x}}{A^{2s}}}{R_1 - R_2} . \tag{4.6}$$

The superscripts 1 and 2 refer to the two fission products, and:

$$R_1 = \frac{\sigma_9 F_9^1}{\sigma_5 F_5^1} , \tag{4.7}$$

$$R_2 = \frac{\sigma_9 F_9^2}{\sigma_5 F_5^2} . \tag{4.8}$$

Thus, the concentration ratios are functions of the monitored flux ratios, the gamma peak activity ratios and the quantities R_1 and R_2. The latter quantities can be determined using U^{235} and Pu^{239} reference foils and the relation:

$$R_1 = \frac{A_9^1}{A_5^1} \cdot \frac{\phi_5}{\phi_9} \cdot \frac{W_5}{W_9} \cdot \frac{239}{235}, \qquad (4.9)$$

where W is the foil surface density (mg/cm^2).

The above method was employed to analyze a rod of mixed Pu/U fuel similar to that described in Chapter 2 (Type A of the SRL simulated burned fuel), using a 1.30% enriched fuel rod as the standard. All samples were irradiated in a thermal neutron flux of 2×10^8 n/cm^2 sec for 15 hours, cooled for 5 to 10 hours, and then counted for 160 minutes (livetime). The fission products I^{135} and $Rb^{88} + Kr^{88}$ were used for the analysis. Preliminary results confirm the nominal 0.24% Pu^{239} content for this fuel as specified by SRL. Work is continuing on more sophisticated data analysis procedures which sum all photopeaks appearing in the spectrum of a given nuclide (e.g., some 5 for I^{135}) and which include use of a third fission product ($Sr^{92} + Y^{92}$), all of which should improve the statistical precision.

4.4 Evaluation of In-Pile Spectrometer

A feasibility study was carried out to determine whether it would be possible to construct a spectrometer which could monitor prompt gamma spectra emitted by a single fuel element during irradiation in the MITR exponential facility (4). The key question proved to be whether the background dose rate could be reduced to less than approximately 50 mR/hr, which previous experience with out-of-pile Ge(Li) spectrometers has shown to be an acceptable upper limit.

In order to obtain data upon which to base the evaluation, gamma dose rate measurements were made in and around the exponential facility during operations in which the tank was filled with pure D_2O moderator. Ionization-type integrating dosimeters were used for all runs. The following results were obtained:

(1) In the graphite-lined hohlraum beneath the exponential tank, the measured gamma dose rate was 6000 R/hr for a bare dosimeter, 4700 R/hr for a boral-covered dosimeter, and 30,000 R/hr for a cadmium-covered dosimeter.

(2) In the annular region surrounding the exponential tank, dosimeters shielded with borated plastic gave dose rates ranging from 500 to 1000 R/hr.

(3) Inside the tank, at the mid-height and over a region extending out to within about 20 cm of the tank wall, the measured dose rates averaged about 100 R/hr for a boron-shielded dosimeter and 10 R/hr when a two-inch-thick lead shield was added.

(4) On the top cover of the tank, dose rates measured 80 to 100 R/hr.

The major conclusions drawn from these data were that carbon capture gammas in the hohlraum are the major contributors to the observed dose rates in the exponential tank, but that capture gammas from the cadmium lining on the outer surface of the tank are also a significant contributor. The latter source could be removed by re-lining the tank with boral or borated plastic, but the former would be very difficult to shield against at the source or around the tank periphery. While technically feasible, use of 8 to 10 inches of lead around the detector itself would involve a major redesign of the tank structure to support the added weight requirements. Therefore, it was decided not to proceed further with design of an in-pile spectrometer. Since this approach was considered as an alternate to the foil methods described in Chapter 2, which have now been shown to be quite adequate, and since the out-of-pile experiments described earlier in this chapter have not indicated that the in-pile spectrometer would have any unique capabilities not otherwise attainable, this decision does not detract from the achievement of project objectives.

4.5 Analysis of Dresden Fuel Pins

During the past year, arrangements were completed and shipment made from G.E. to M.I.T. of two irradiated fuel pins. These two fuel rods had been irradiated to about 20,000 MWD/T, the first 6000 MWD/T in the Vallecitos Boiling Water Reactor and the remainder in the Dresden Nuclear Power Station. These rods contain both fission products and plutonium. Table 4.2 summarizes the information provided by G.E. on the projected current status of this fuel.

TABLE 4.2

Characteristics of Dresden Fuel Pins

Average Burnup	Pin Number A4 20,200 MWD/TU	Pin Number A28 21,200 MWD/TU
% U^{235}	0.935	0.880
% Pu	0.575	0.606
% Pu^{239} + Pu^{241}	0.398	0.400
wt. U^{235}	5.70 gm	5.44 gm
wt. Pu	3.51 gm	3.75 gm
wt. Pu^{239} + Pu^{241}	2.42 gm	2.47 gm
wt. U	594.57 gm	602.37 gm

A feasibility study was carried out to evaluate the handling procedures and to establish shield and transfer flask design requirements for conducting out-of-pile gamma spectrometry on these fuel pins (5). It was concluded that presently available facilities are adequate for these experiments and that the analytic methods of Ref. 6 should be applicable for the inference of fuel irradiation history despite the long out-of-pile decay time experienced by the Dresden fuel pins.

During the coming year, the major concern of the Reactor Physics Project will be these two partially burned fuel pins. Prompt and delayed gamma spectra will be obtained out-of-pile to evaluate the current composition of the fuel and to confirm its past irradiation history. This will be followed by in-pile studies in which the foil methods used in Chapter 2 will be used to determine the heterogeneous reactor physics constants characterizing the fuel. It is anticipated that these latter tests will be carried out in H_2O rather than D_2O in view of the encouraging results cited in Chapter 5 and the fact that the subject fuel rods are, in fact, light water BWR-type fuel.

4.6 References

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5. EVALUATION OF HETEROGENEOUS METHODS FOR H_2O SYSTEMS

G. T. Hamilton

5.1 Introduction

Heterogeneous reactor theory, upon which the Single Element Method is based, has been developed for and applied to heavy water- and graphite-moderated systems. While there are good qualitative reasons to anticipate problems in extending this work to light water-moderated systems, it was felt worthwhile to evaluate H_2O lattice data available in the literature in order to assess the precise limitations in a more quantitative manner (1). In addition, the availability of the modified ANISN program, described in Chapter 3, made it possible to perform a numerical single-element experiment in H_2O for comparison with similar numerical and physical experiments using D_2O moderator.

5.2 Behavior of Integral Parameters δ_{25}, ρ_{28}, δ_{28}, C^*

From a purely empirical viewpoint, one of the more useful results of heterogeneous theory has been its use to justify the use of linear correlations of foil activity ratios versus volume fraction fuel (2), (3).

The integral parameters which are the subject of such correlations are the following four familiar quantities:

$$\rho_{28} = \frac{\text{Epicadmium capture rate in } U^{238}}{\text{Subcadmium capture rate in } U^{238}},$$

$$\delta_{25} = \frac{\text{Epicadmium fission rate in } U^{235}}{\text{Subcadmium capture rate in } U^{235}},$$

$$C^* = \frac{\text{Capture rate in } U^{238}}{\text{Fission rate in } U^{235}},$$

$$\delta_{28} = \frac{\text{Fission rate in } U^{238}}{\text{Fission rate in } U^{235}}.$$

Pilat (3) has derived expressions of the following form for the variation of these parameters with cell volume:

$$\rho_{28} = \frac{C}{\xi\Sigma_s} \frac{V_f}{V_c}, \tag{5.1}$$

where

V_f = fuel volume in unit cell,

V_c = total unit cell volume,

$\xi\Sigma_s$ = mean slowing-down power.

For the homogenized unit cell:

$$\xi\Sigma_s = \frac{V_f(\xi\Sigma_s)_f + V_m(\xi\Sigma_s)_m}{V_c}. \tag{5.2}$$

Since $(\xi\Sigma_s)$ for the fuel is much less than that for the moderator and V_f is less than the moderator volume,

$$\xi\Sigma_s \approx (\xi\Sigma_s)_m \frac{V_m}{V_c}, \tag{5.3}$$

so that

$$\rho_{28} = C' \left(\frac{V_f}{V_m}\right). \tag{5.4}$$

The difference between Eqs. 5.1 and 5.4 is not significant for D_2O-moderated lattices, since V_f/V_c is typically less than 0.05 for the uniform lattices studied at M.I.T. For light water lattices, however, V_f/V_c can be on the order of 0.5 and thus one must follow the more accurate expression, Eq. 5.4, and correlate integral parameters versus the fuel-to-moderator ratio and not the volume fraction fuel.

Figures 5.1 through 5.7 display data culled from the literature for uranium metal and oxide fueled lattices moderated by H_2O. As can be seen, the linear correlation forms suggested by heterogeneous theory appear to be valid. These results provide considerable circumstantial evidence favoring the applicability of heterogeneous methods to light water lattices.

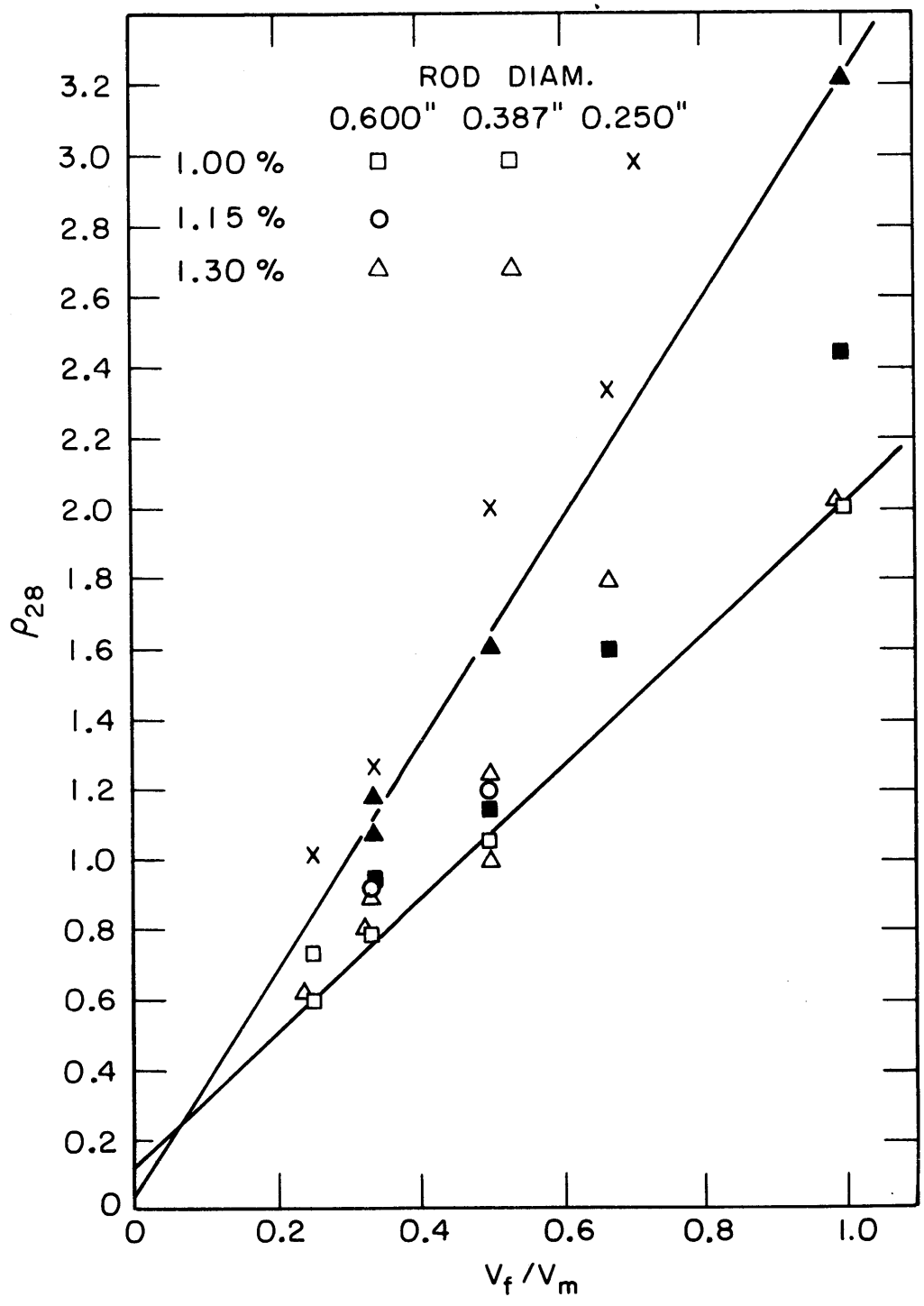


FIG.5.1 ρ_{28} FOR URANIUM METAL RODS IN H_2O

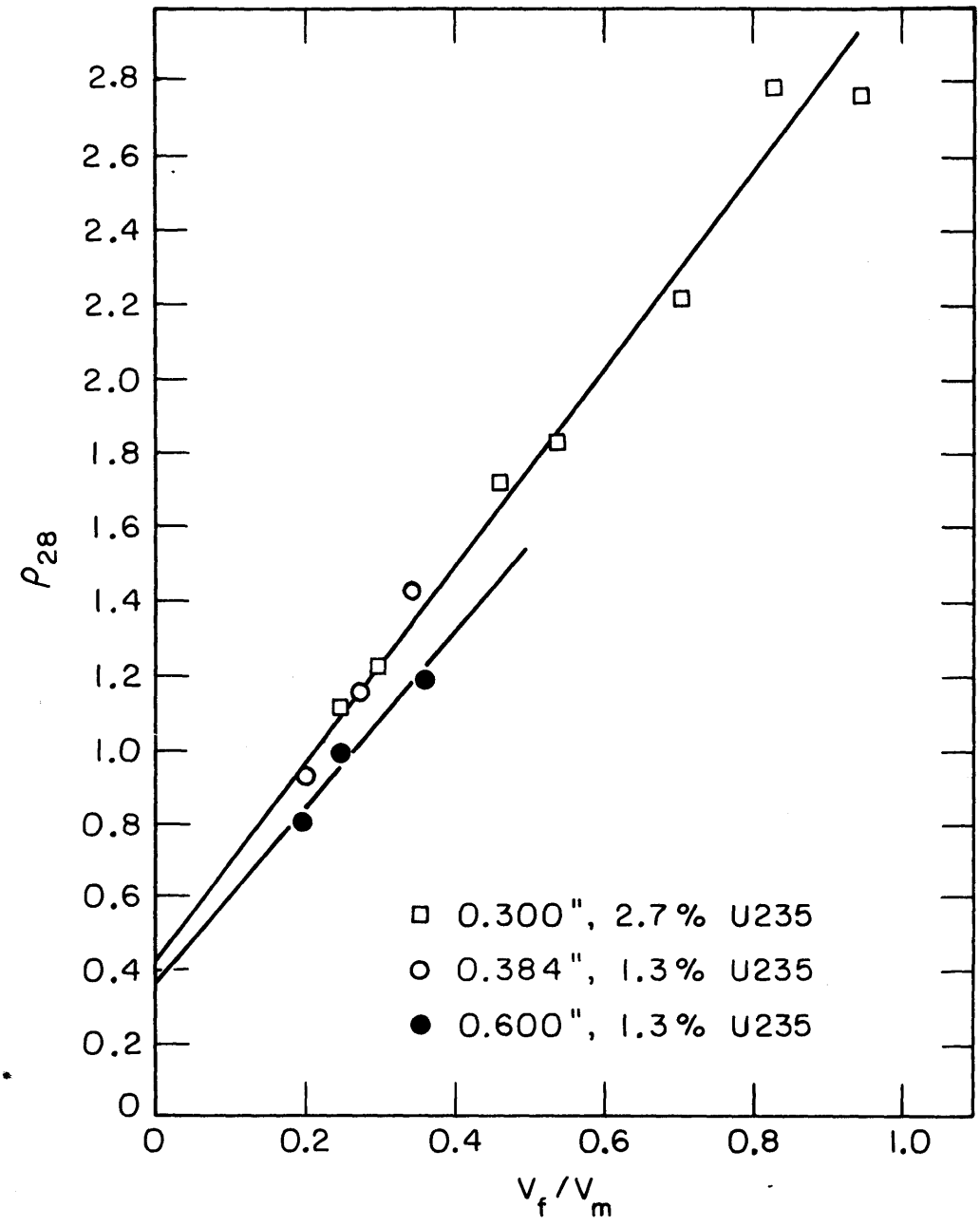


FIG. 5.2 ρ_{28} FOR UO_2 RODS IN H_2O

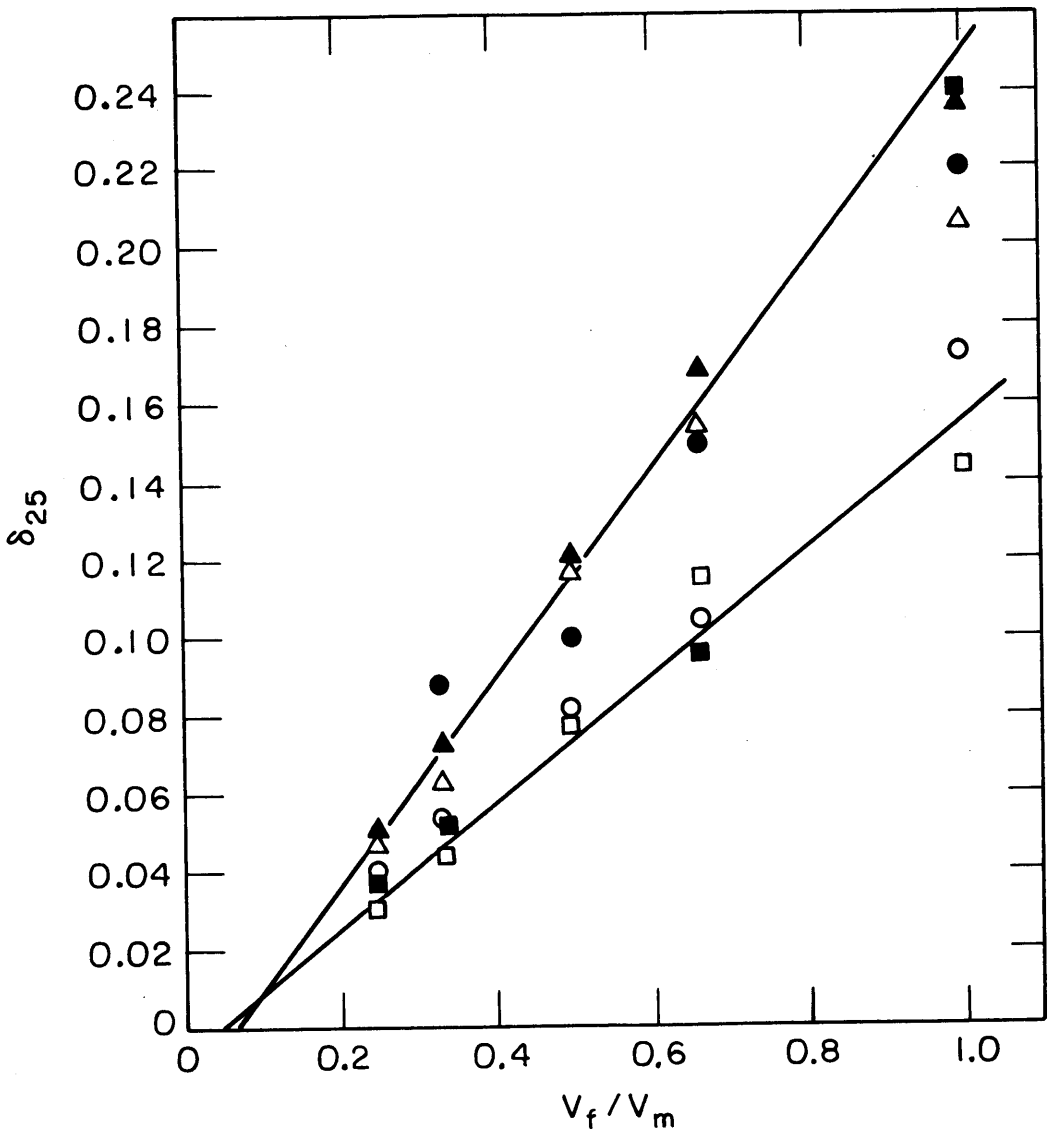


FIG. 5.3 δ_{25} FOR URANIUM METAL RODS IN H_2O

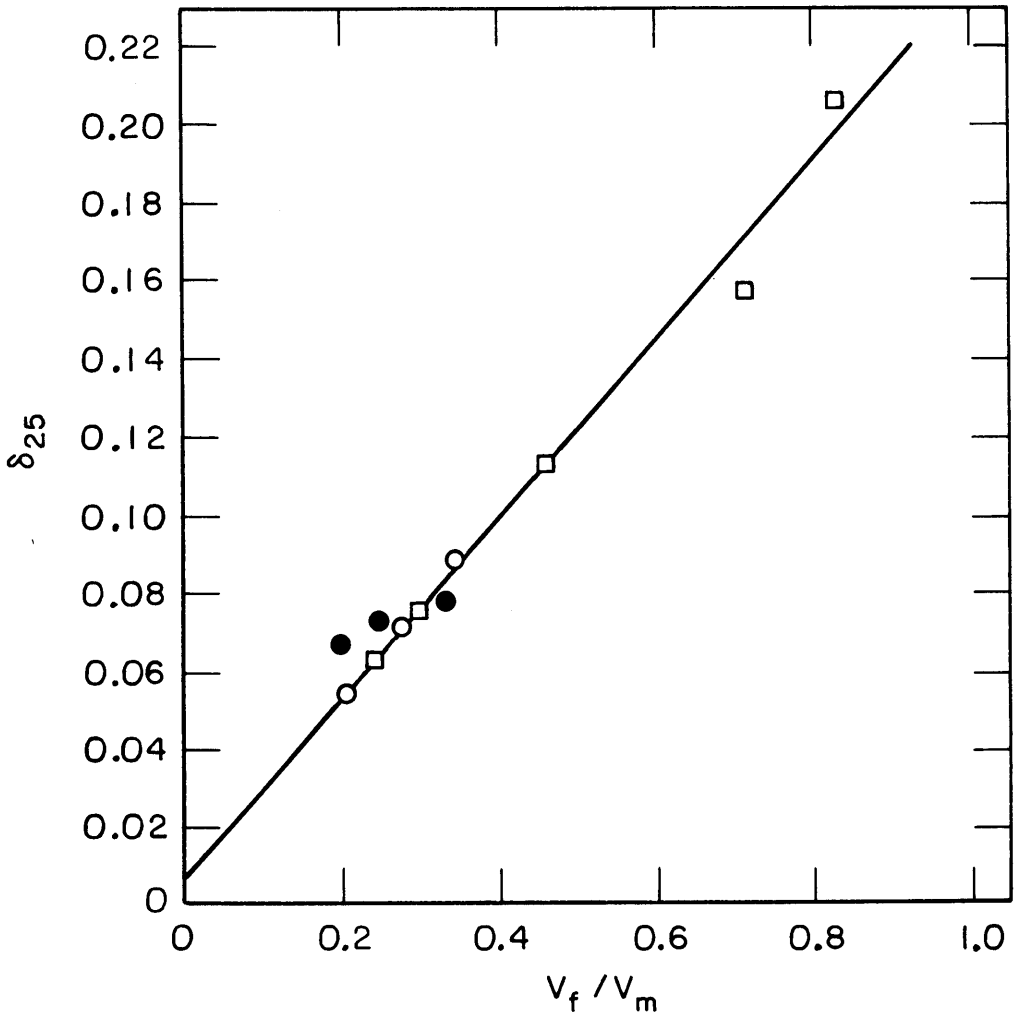
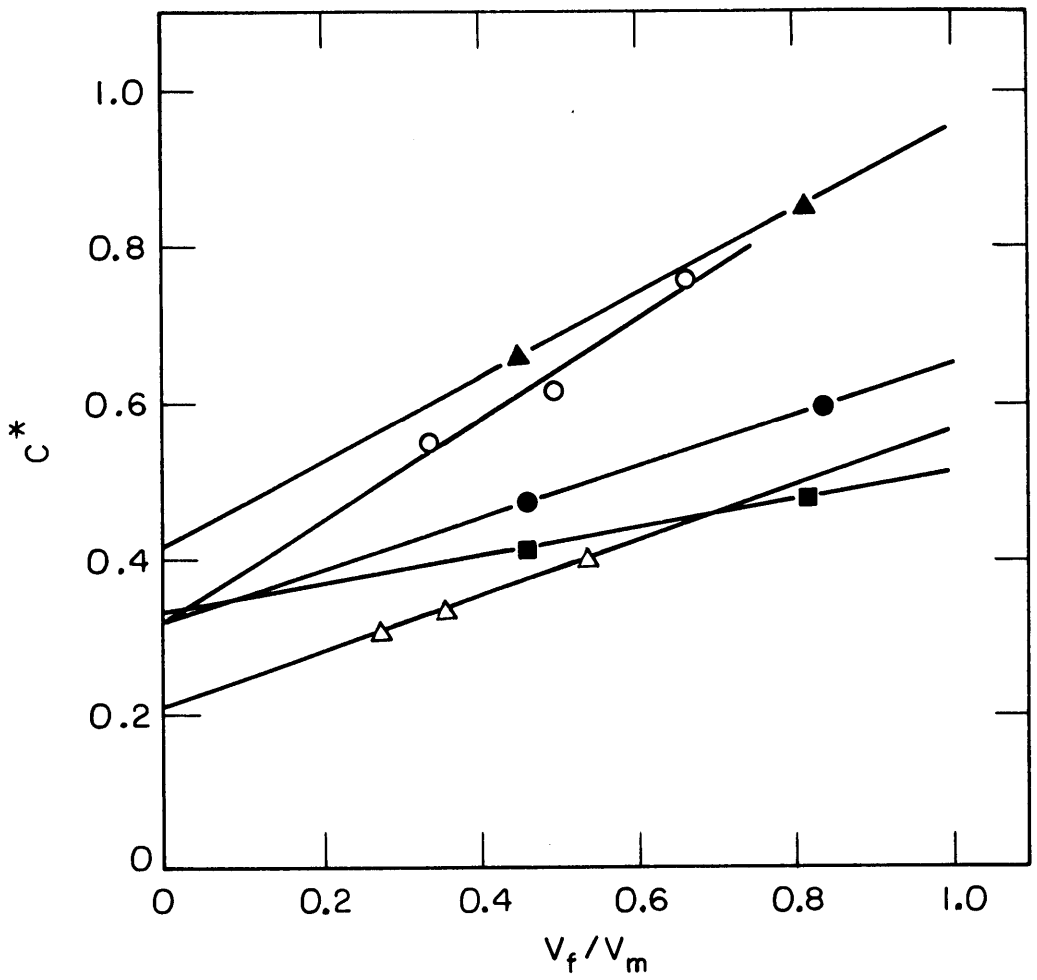


FIG. 5.4 δ_{25} FOR UO_2 RODS IN H_2O



▲ 0.300", 1.6% UO_2
● 0.300", 2.7% UO_2
■ 0.300", 3.7% UO_2
△ 1.194", 3.4% UO_2
○ 0.600", 1.3% U METAL

FIG. 5.5 C^* FOR H_2O MODERATED LATTICES

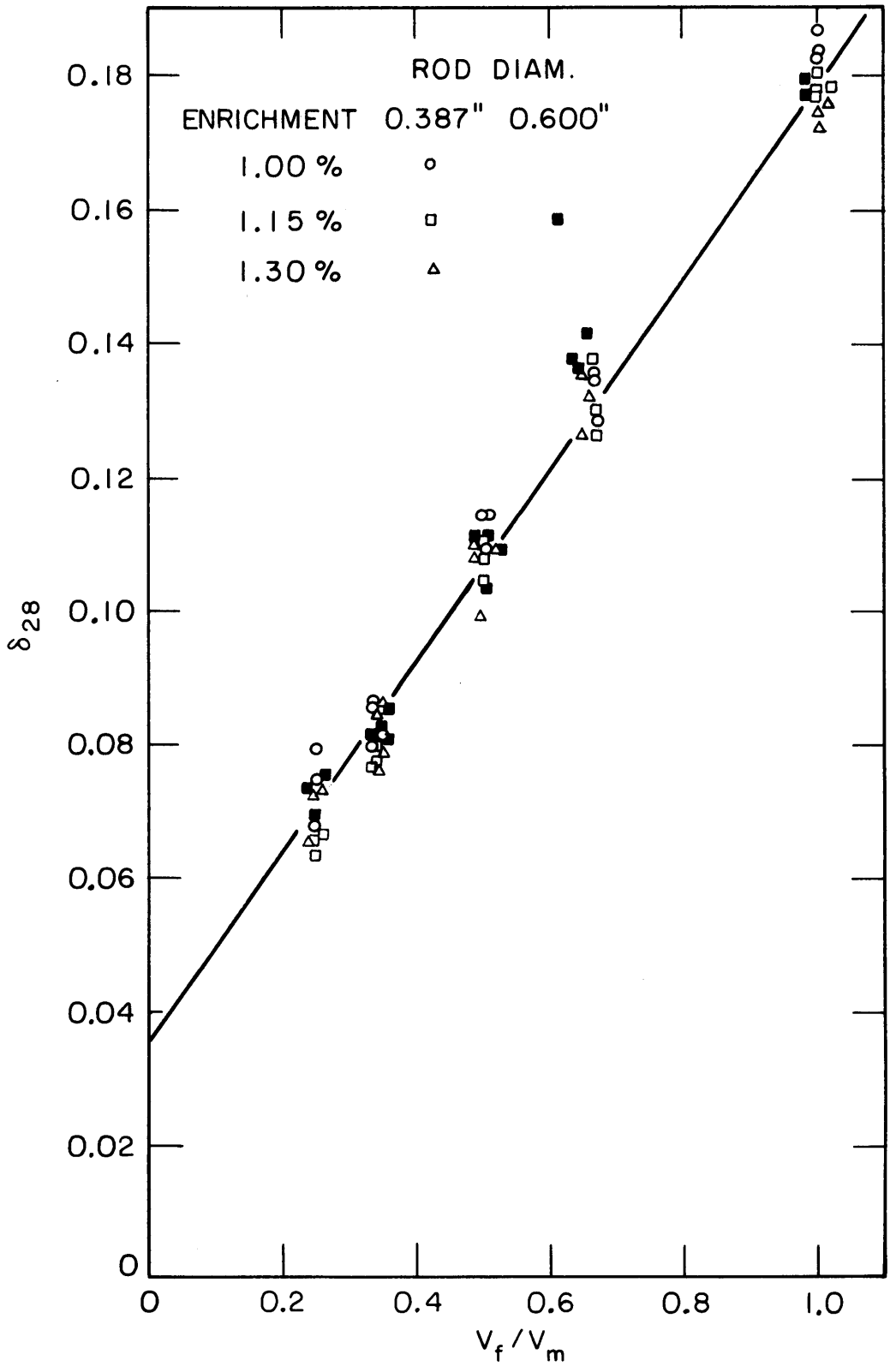


FIG. 5.6 δ_{28} FOR URANIUM METAL RODS IN H_2O

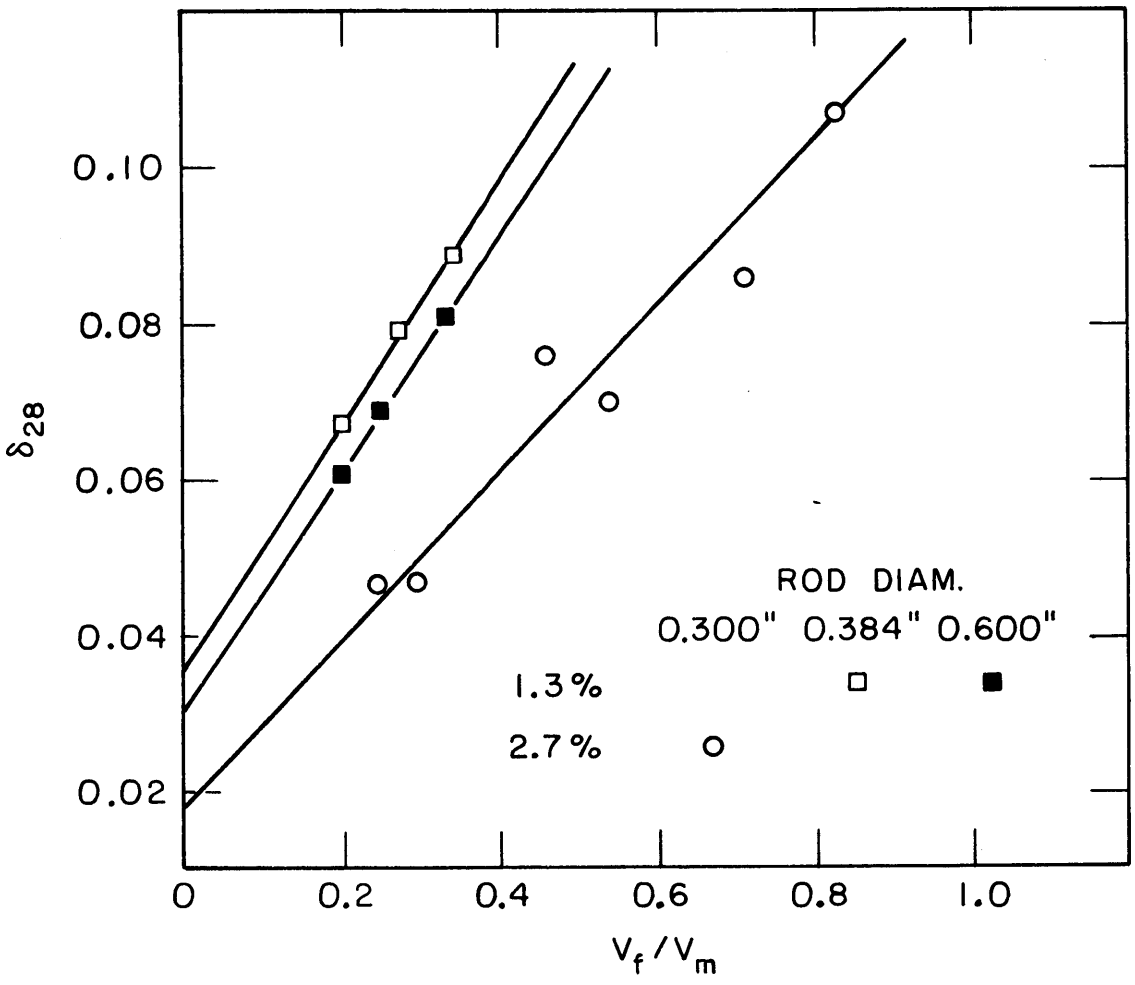


FIG. 5.7 δ_{28} FOR UO_2 RODS IN H_2O

5.3 Evaluation of Γ, η, and A

The heterogeneous parameters Γ, η and A can be related to the integral parameters discussed in the previous section and to the thermal utilization (4); specifically,

$$\eta = \frac{[\nu_{25} + (\nu_{28} - 1 - \alpha_{28}^{F})\,\delta_{28}](1+\delta_{25})}{C^{*}\left(\frac{1+\delta_{25}}{1+\rho_{28}}\right) + 1 + \alpha_{25}^{th}} \tag{5.5}$$

and

$$A = \frac{\left[\rho_{28} C^{*} \frac{1+\delta_{25}}{1+\rho_{28}} + \delta_{25}\left(1+\alpha_{25}^{e}\right)\right] f V_m}{C^{*}\left(\frac{1+\delta_{25}}{1+\rho_{28}}\right)\left(1+f\rho_{28}\right) + \left(1+\alpha_{25}^{th}\right) + f\delta_{25}\left(1+\alpha_{25}^{e}\right)} . \tag{5.6}$$

$$\Gamma = \frac{\frac{1}{f} - 1 - \Delta}{\Sigma_{\alpha m} V_m} , \tag{5.7}$$

where

$$\Delta = \frac{V_c}{4\pi L_m^2}\left[\frac{\ln(V_c/V_f)}{1-(V_f/V_c)} - \frac{3}{2} + \frac{1}{2}\frac{V_f}{V_c}\right] ,$$

and standard symbols are used to define all quantities. For example, α is the capture-to-fission ratio for the subscripted nuclide (e.g., $25 = U^{235}$), averaged over the superscripted spectrum (e.g., F = fission spectrum, e = epithermal spectrum, th = thermal spectrum).

Equations 5.5 through 5.7 were used to calculate Γ, η, and A from experimental data published for H_2O-moderated lattices. The results are plotted in Figs. 5.8 through 5.10. If heterogeneous theory in its simplest form provided an exact description of the lattices in question, the three parameters would be constants, characteristic only of the fuel rods and not of the lattice spacing. Figure 5.8 shows that the parameter Γ satisfies this criterion quite well. Figure 5.9, however, shows that η increases linearly with the fuel-to-moderator ratio.

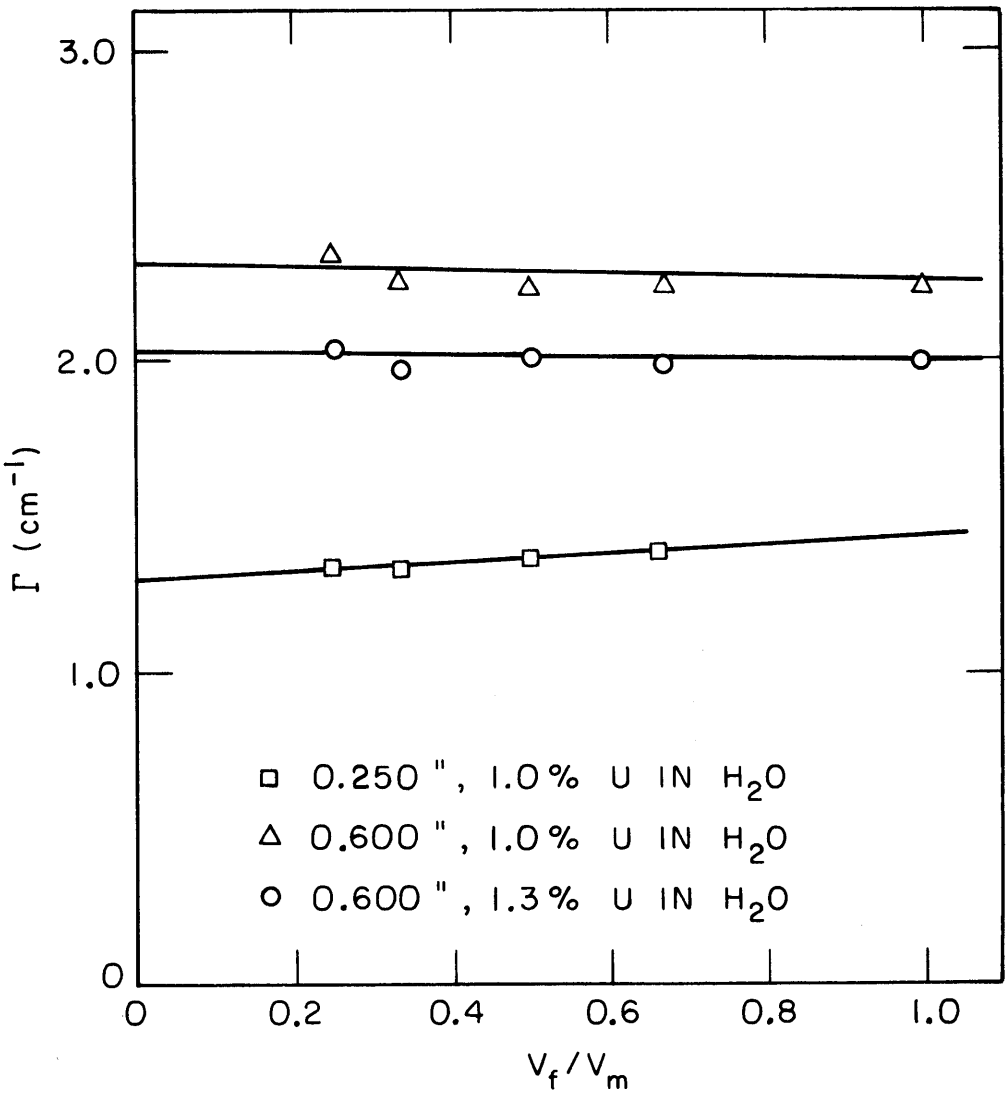


FIG. 5.8 THERMAL CONSTANTS FOR URANIUM METAL RODS IN LIGHT WATER LATTICES

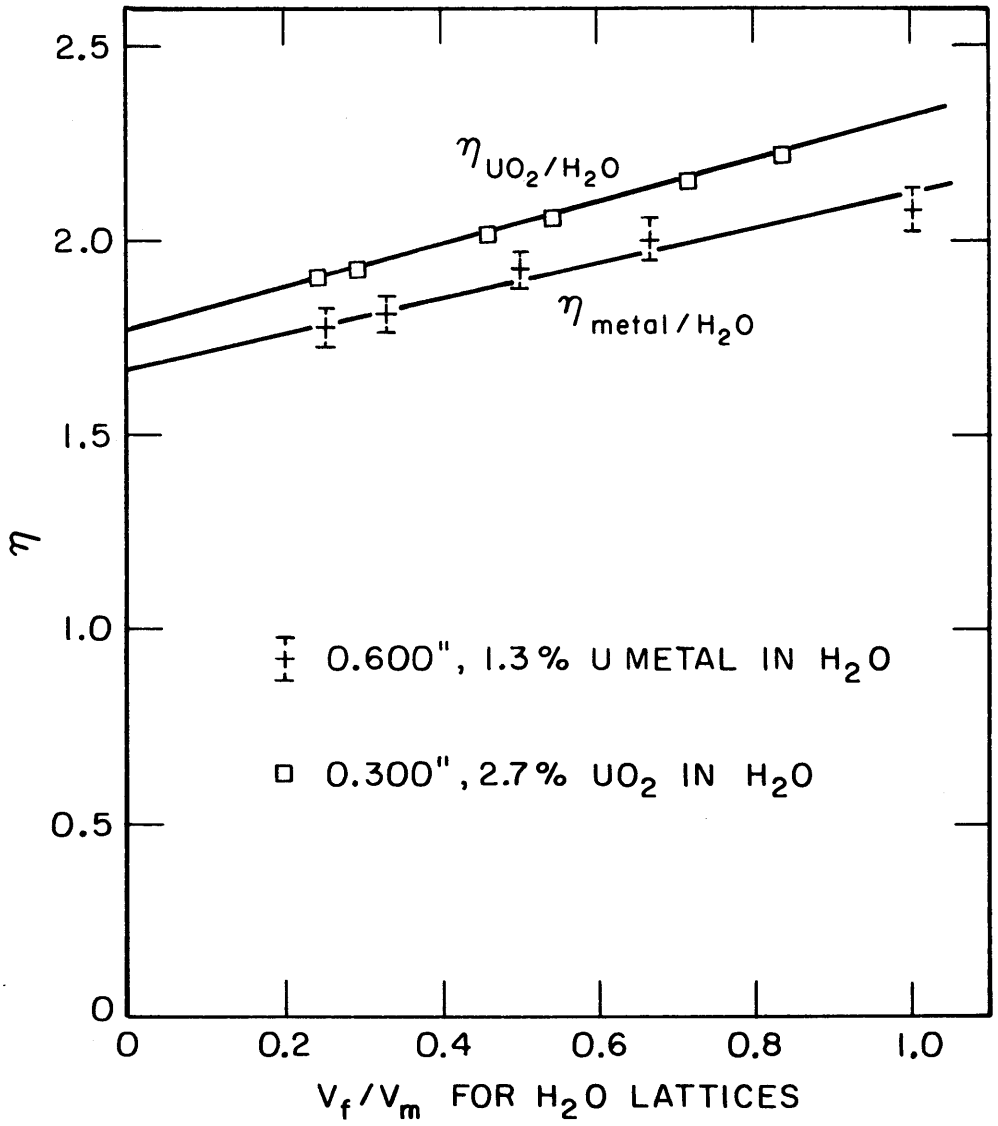


FIG. 5.9 VARIATION OF THE NEUTRON YIELD WITH THE FUEL TO MODERATOR VOLUME RATIO

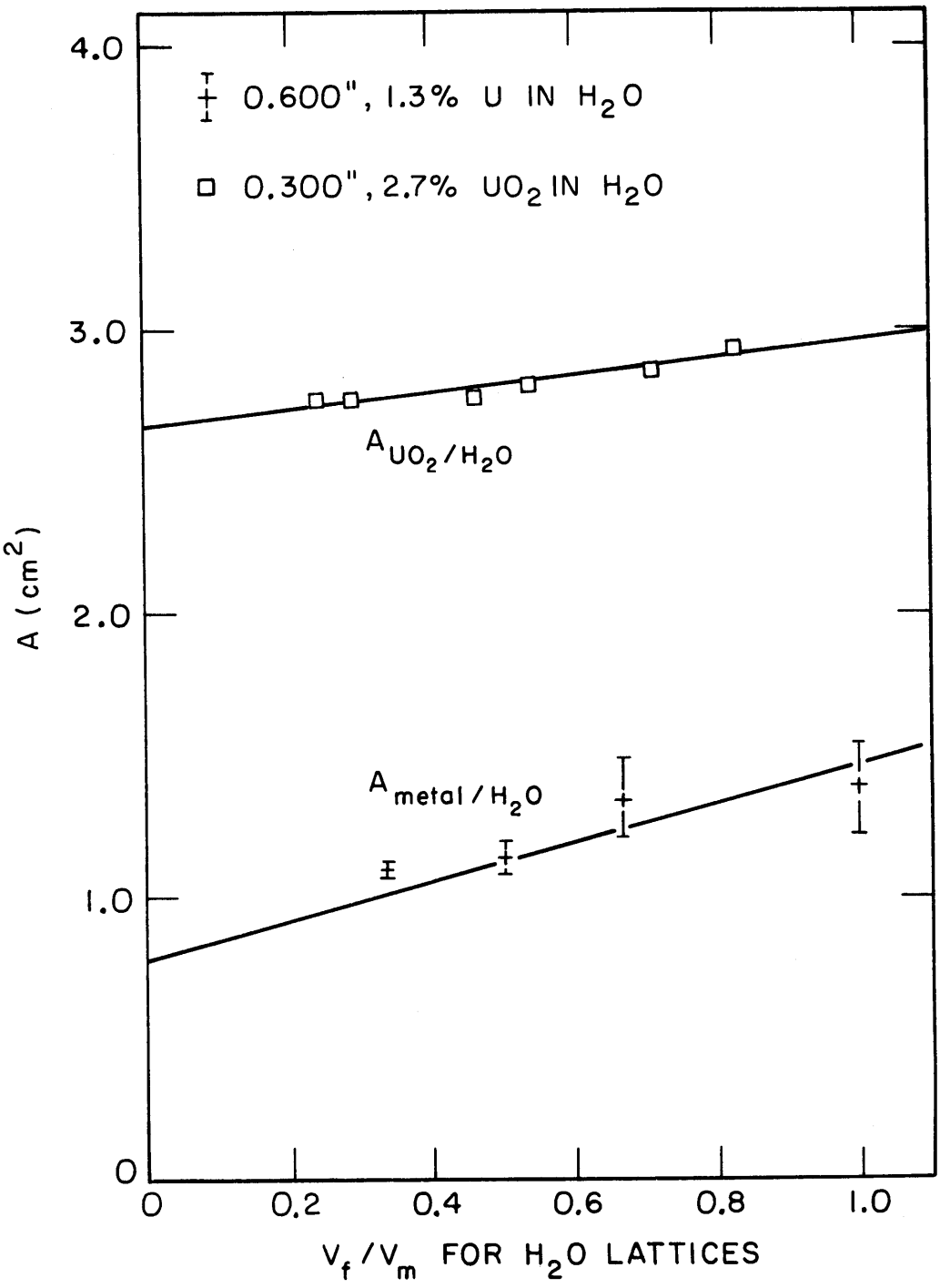


FIG. 5.10 VARIATION OF THE EPITHERMAL ABSORPTION PARAMETER WITH THE FUEL TO MODERATOR VOLUME RATIO

A more detailed evaluation shows that this dependence can be quantitatively accounted for by taking into account the increased fast and epithermal fissions as inter-rod spacing is decreased. Figure 5.10 shows that the parameter A also increases slowly and linearly with the fuel-to-moderator ratio, in contrast to its behavior for D_2O-moderated systems, where a comparable linear decrease is noted. More work is needed before this behavior can be accurately predicted.

In short, the behavior of Γ, η and A does not appear to be any more complex for H_2O-moderated systems than has already been successfully dealt with in D_2O systems. Presumably, therefore, if one can measure the single-element value of these parameters (the ordinate intercepts at $V_f/V_m = 0$ in Figs. 5.8 through 5.10), the analytic formalism either exists or can be developed to permit development of the entire linear map and thereby to characterize complete lattices of the subject fuel.

5.4 Numerical Experiments

In order to provide some basis upon which to evaluate the feasibility of single rod experiments in H_2O, a series of numerical calculations was performed using the ANISN code. The code was used to calculate multigroup flux profiles around a fuel element in simulated single rod exponential experiments using both D_2O and H_2O moderator.

Analysis of the results of these calculations led to the following conclusions:

1) Measurements of the parameters η and A in H_2O should prove no more difficult than in D_2O.

2) Precautions against and corrections for horizontal foil holder tilt are more important for H_2O systems because of the more rapid attenuation of the axial flux in H_2O. Calculations for the 36-inch-diameter MITR exponential tank showed axial exponential behavior of approximately $\exp - 0.17\ \Delta Z$ in H_2O and $\exp - 0.05\ \Delta Z$ in D_2O, where ΔZ is the change in relative axial position in centimeters.

3) Additional theoretical work may be required to extract Γ from radial thermal flux profiles in H_2O systems because of the greater importance of transport effects.

5.5 Conclusions

The experimental data examined in this chapter have been found to be interpretable in terms of heterogeneous reactor theory. Correlation methods which were originally developed for D_2O lattices also apply to H_2O systems if the fuel-to-moderator ratio is used as a correlation parameter in place of the volume fraction fuel. Integral parameter data can be used to calculate the heterogeneous constants Γ, η and A, and the changes in these parameters with system variables are as well-behaved as for D_2O-moderated lattices. Finally, numerical experiments simulating the techniques developed at M.I.T. for measurement of Γ, η and A, using foil activation data measured external to a single fuel element, appear to be extendable to the H_2O-moderated systems.

5.6 References

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