REFERENCE 60

P. LECORCHE AND R. L. SEALE, "A REVIEW OF THE EXPERIMENTS PERFORMED TO DETERMINE THE RADIOLOGICAL CONSEQUENCES OF A CRITICALITY ACCIDENT," OAK RIDGE CRITICALITY DATA CENTER REPORT Y-CDC-12 (NOVEMBER 1973).

Y-CDC-12
UC-46 -- Criticality Studies
UNION CARBIDE CORPORATION
NUCLEAR DIVISION
OAK RIDGE Y-12 PLANT
operated for the
U.S. ATOMIC ENERGY COMMISSION

A REVIEW OF THE EXPERIMENTS PERFORMED TO DETERMINE THE RADIOLOGICAL CONSEQUENCES OF A CRITICALITY ACCIDENT

Pierre Lécorché

Commissariat à l'Énergie Atomique
Direction de la Protection et de la Sûreté Radiologiques
Service d'Études de Criticité

and

Robert L. Seale

University of Arizona

Printed in the United States of America. Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road, Springfield, Virginia 22151
Price: Printed Copy $5.45; Microfiche $0.95

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Robert L. Seale

University of Arizona
Tucson, Arizona 85721

Oak Ridge Y-12 Plant
P. O. Box Y, Oak Ridge, Tennessee 37830

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by UNION CARBIDE CORPORATION-NUCLEAR DIVISION
under Contract W-7405-eng-26

Date Issued: NOV 3 1973

PREFACE

The Criticality Data Center again has the opportunity to make available in translation a report of an interesting and informative series of experiments performed by the Section Expérimentale d'Études de Criticité at Valduc under the aegis of the Service d'Études de Criticité of the French Commissariat à l'Énergie Atomique. The summary was prepared by Professor Robert L. Seale in collaboration with M. Pierre Lécorché. The Center acknowledges the permission granted by the Commissariat to report this research.

OAK RIDGE CRITICALITY DATA CENTER

The Criticality Data Center was established under the auspices of the U.S. Atomic Energy Commission for the development of methods allowing extension and application of data derived from experiments and from analyses to problems in nuclear criticality safety, as well as for the review and evaluation of the data themselves. A necessary part of this program is a medium whereby information germane to the intent of the Center is made available. This report series has been inaugurated for that purpose.

The first five reports were published by and identified with the Oak Ridge National Laboratory. Subsequent reports, however, issued from the Y-12 Plant, are identified by a number sequence including the prefix Y-CDC.

Inquiries should be directed to E. B. Johnson, P. O. Box Y, Oak Ridge, Tennessee 37830.

Previous Reports in This Series:

ORNL-CDC-1 Criticality of Large Systems of Subcritical U(93) Components by J. T. Thomas (1967).

ORNL-CDC-2 Calculated Neutron Multiplication Factors of Uniform Aqueous Solutions of ^{233}U and ^{235}U by J. Wallace Webster (1967).

ORNL-CDC-3 Estimates of Maximum Subcritical Dimensions of Single Fissile Metal Units by W. H. Roach and D. R. Smith (1967).

ORNL-CDC-4 The Effect of Unit Shape on the Criticality of Arrays by J. T. Thomas (1967).

ORNL-CDC-5 Minimum Critical ^{235}U Enrichment of Homogeneous Hydrogenous Uranyl Nitrate by S. R. Bierman and G. M. Hess (1968).

Y-CDC-6 Some Effects of Interspersed Moderation on Array Criticality by J. T. Thomas (1969).

Y-CDC-7 Uranium Metal Criticality, Monte Carlo Calculations and Nuclear Criticality Safety by J. T. Thomas (1970).

Y-CDC-8 Use of Borosilicate Glass Raschig Rings as a Neutron Absorber in Solutions of Fissile Material by J. P. Nichols, C. L. Schuske, and D. W. Magnuson (1971).

Y-CDC-9 Criticality of Liquid Mixtures of Highly ^{235}U-Enriched Uranium Hexafluoride and Hydrofluoric Acid by Robert Caizergues, Edouard Deilgat, Pierre Lécorché, Louis Maubert, and Henri Revol (1971).

Y-CDC-10 The Criticality of Cubic Arrays of Fissile Material by J. T. Thomas (1971).

Y-CDC-11 The S_n Method by K. D. Lathrop
The Monte Carlo Method as Applied to Nuclear Criticality Safety Calculations by G. E. Whitesides (1972).

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A REVIEW OF THE EXPERIMENTS PERFORMED TO DETERMINE THE RADIOLOGICAL CONSEQUENCES OF A CRITICALITY ACCIDENT

Pierre Lécorché and Robert L. Seale

ABSTRACT

The series of nuclear criticality excursions in aqueous solutions of uranium highly enriched in the ^{235}U isotope, conducted by the Service d'Études de Criticité of the French Commissariat à l'Énergie Atomique from 1968 through 1971, are summarized. A total of 40 excursions were observed under a wide range of solution concentrations and ramp reactivity insertions. Results for the pulse trains that developed include determination of peak fission rates, pulse widths, integrated yields, and consequent gamma-ray dose rates. The analysis of these results suggests the strong influence of gas evolution, as well as temperature, as a mechanism limiting pulse size.

I. INTRODUCTION

In November 1968, the Service d'Études de Criticité of the French Commissariat à l'Énergie Atomique initiated a program designed to provide data on the consequences of exceeding delayed criticality with various aqueous solutions of uranyl nitrate. In this program, designated "Consequences Radiologiques d'un Accident de Criticité" (CRAC), a series of experiments was performed in which aqueous uranyl nitrate solutions at various concentrations were injected into a large diameter cylinder to heights in excess of the critical height. The uranium was 93% enriched in ^{235}U.

The results of the experiments have provided bases for evaluating analytical models describing the consequent transient phenomena. In addition, the results give insight into the behavior expected in accidental supercritical accumulations of fissile materials which may occur in chemical processes.

II. EXPERIMENTAL MATERIALS AND PROCEDURES

Experiments with the uranyl nitrate solution in a 300-mm-diam cylinder (3 mm wall) (CRAC 01 through 29) and in an 800-mm-diam cylinder (4 mm wall) (CRAC 37 through 44) have been completed and are summarized here. The reports describing the experiments are listed in Appendix A. In the experiments conducted in the 300-mm-diam cylinder, critical heights varied from 193.9 cm in CRAC 04 to 27.47 cm in CRAC 25; the corresponding ^{235}U concentrations were 48.2 g/liter and 298 g/liter, respectively. The cylinder was placed inside a large airtight enclosure to contain any gas or liquid ejected from the reacting volume. Figure 1 is a plan view of the experimental area showing the location of various radiation detectors. The experimental vessel is shown in Fig. 2. The length of the cylinder could be extended by adding 1-m-long sections.

The normal experimental procedure was to inject solution into the cylinder at a constant rate varying from 104 liters/h in CRAC 12 to 1864 liters/h in CRAC 25. Addition of solution at a constant rate resulted

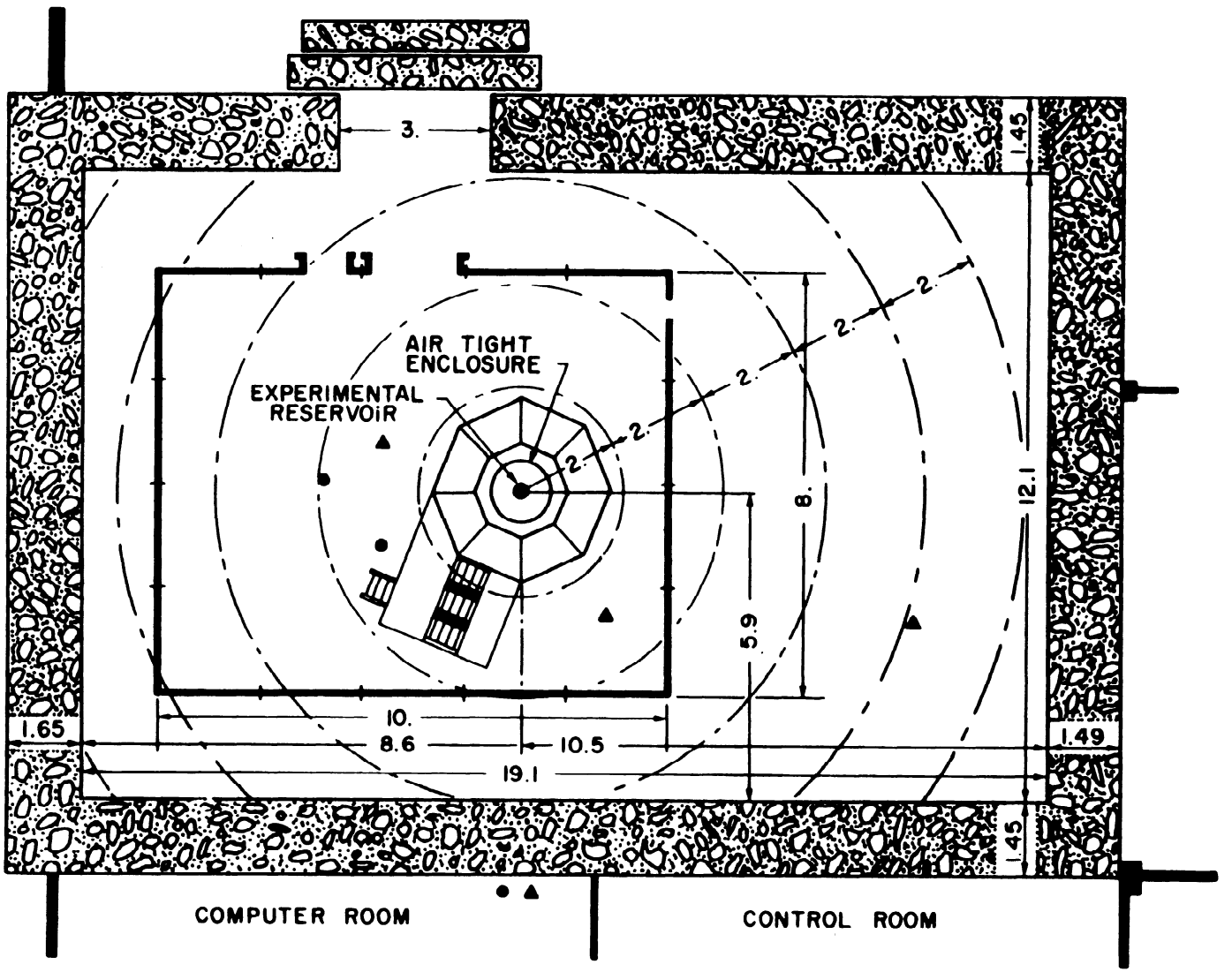

Fig. 1. Plan View of CRAC Experimental Facility.

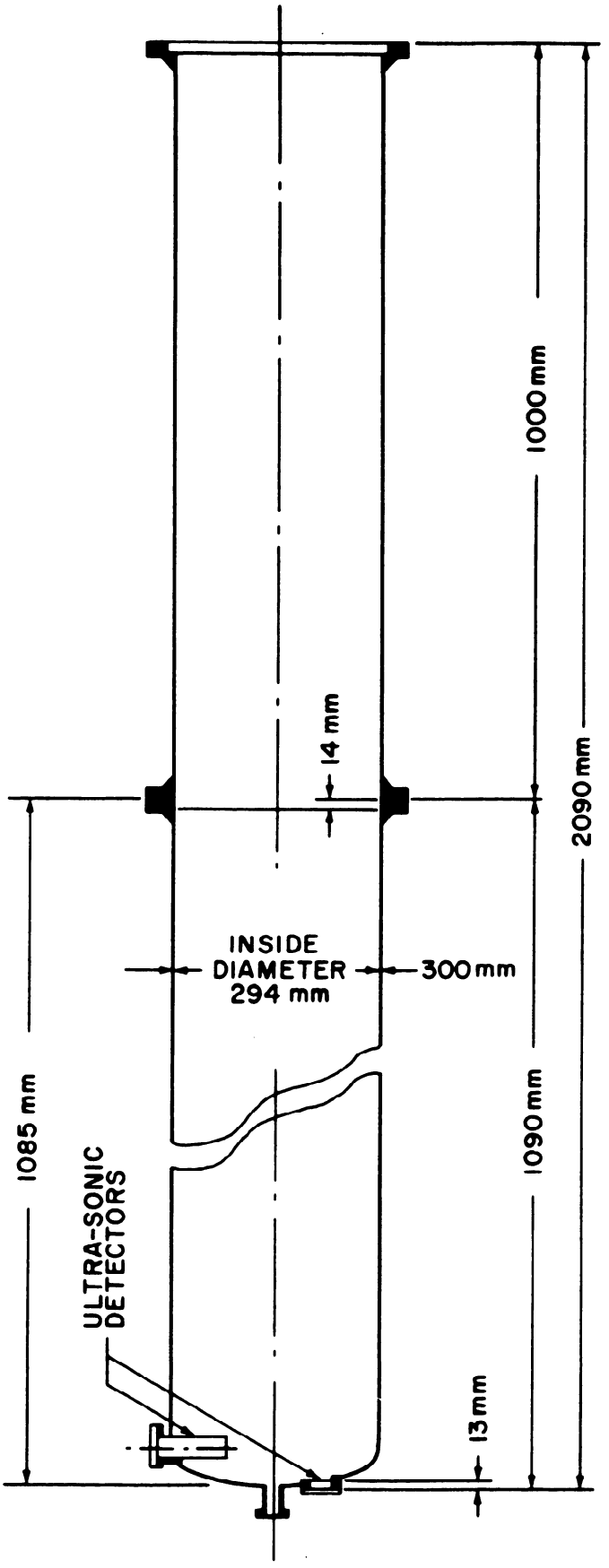

Fig. 2. Schematic of an Experimental Vessel.

initially in a uniform ramp-rate increase in reactivity. In most experiments the ramp was initiated with a subcritical volume of solution. In CRAC 15, 16, 17, and 18, however, the volume was initially critical at a low steady power when the ramp was initiated. Of the experiments in which the solution in the cylinder was initially subcritical, CRAC 14, 20.1, 20.2, and 20.3 were conducted in the presence of an external neutron source. The remaining experiments were performed without an external neutron source being present.

After a supercritical volume of solution was accumulated in the cylinder, power increased to a peak corresponding to overriding the excess reactivity by thermal expansion and gas formation. This initial pulse died out and was followed by a series of peaks of generally decreasing size. In all these cases, solution was added through the duration of the first peak and, in many instances, was continued until after several peaks were observed. Several flux traces are shown in Figs. 3, 4, and 5 as representative of the experiments. Eventually fissions generated in the solution caused heating, material ejection, and gas formation such that the excess reactivity made available by the excess fuel was balanced and a constant power was achieved.

In addition to the "excursion experiments" resulting from the extended ramp insertion of reactivity, there were cases in which a slightly supercritical volume of fuel was put into the reaction vessel for a "step" insertion of reactivity and the development of the power history monitored. The stable period of the resulting "slow excursion" allowed direct determination of the reactivity worth of incremental additions of solution to a critical volume.

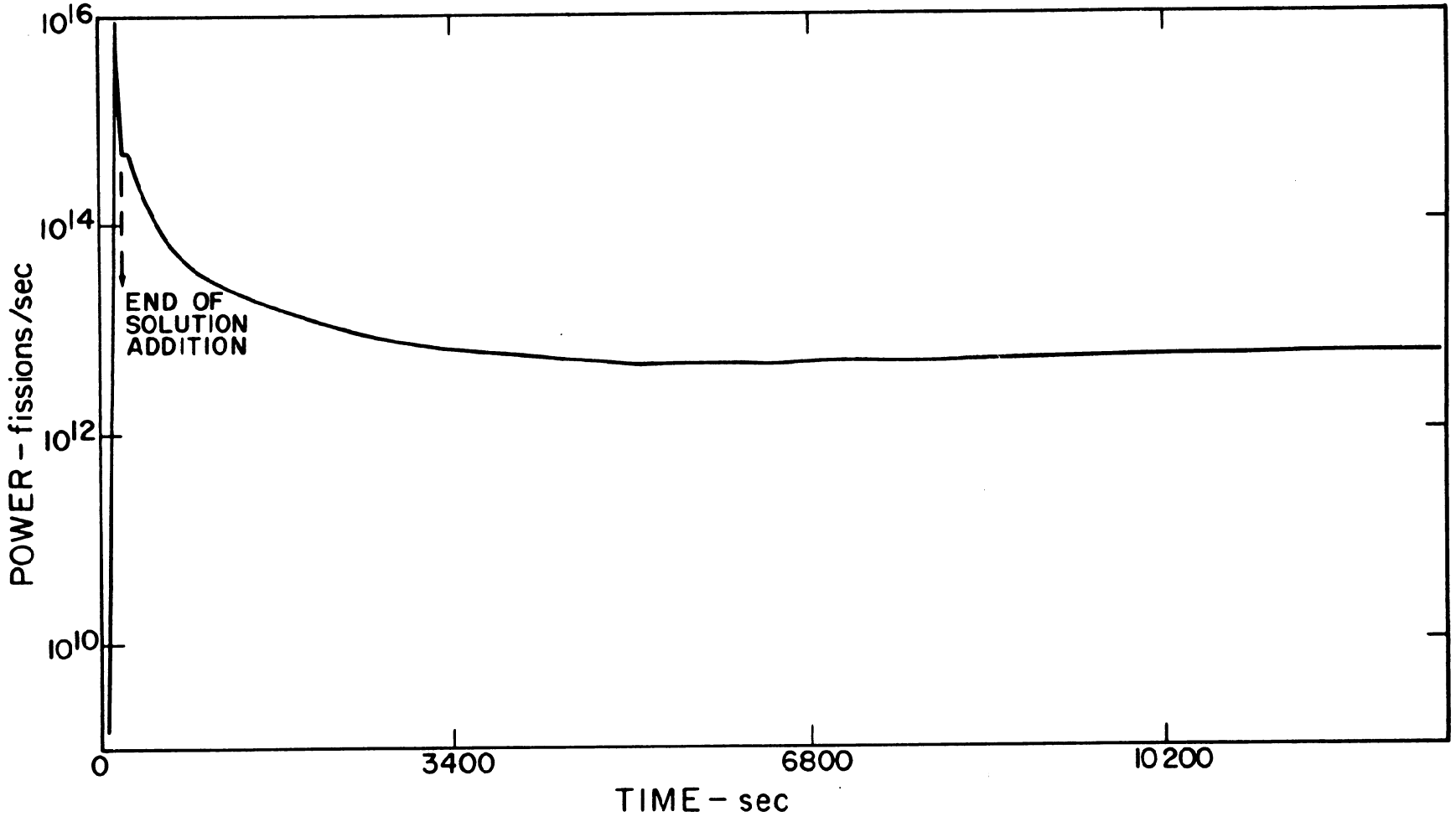

Fig. 3. Fission Rate in CRAC 04 as a Function of Time.

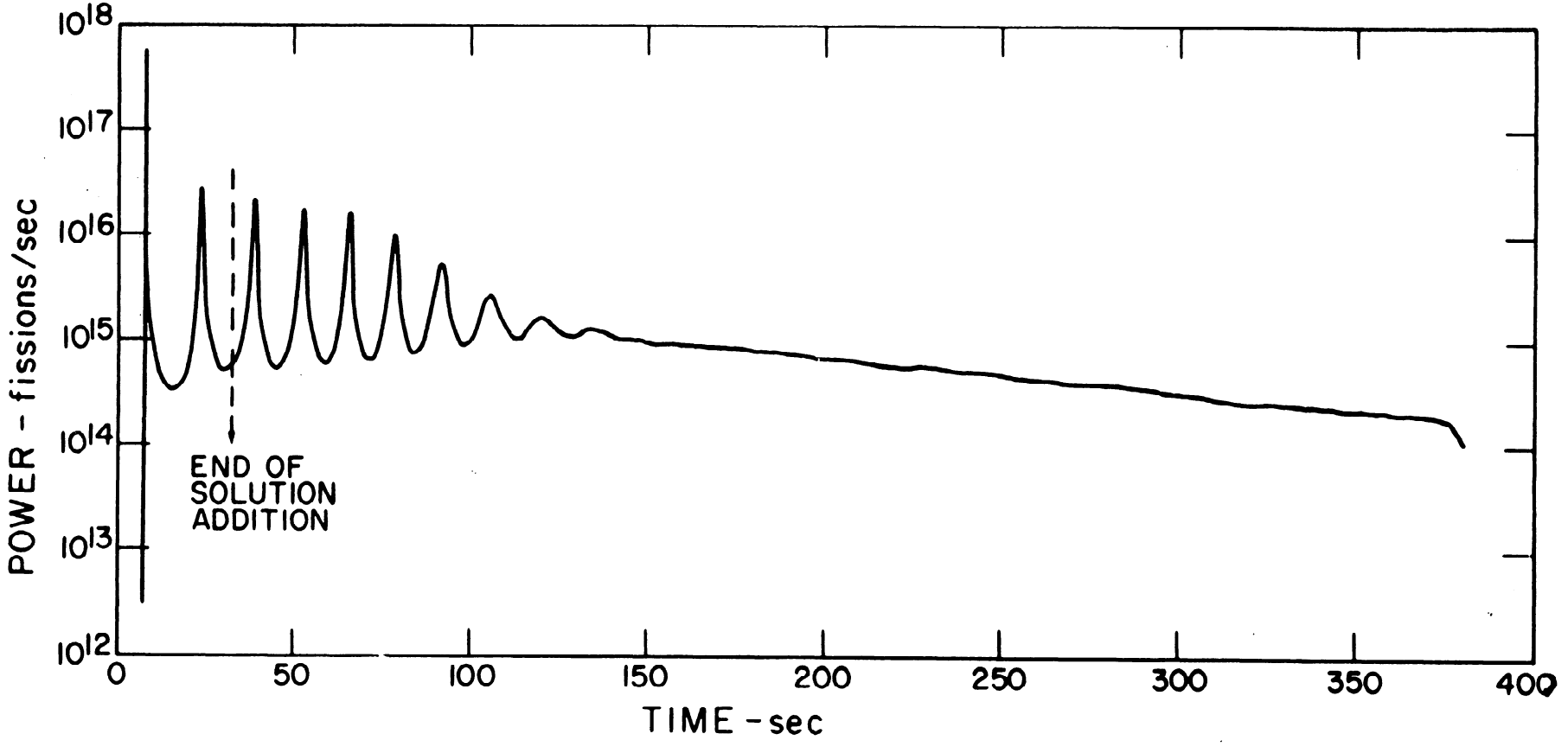

Fig. 4. Fission Rate in CRAC 13 as a Function of Time.

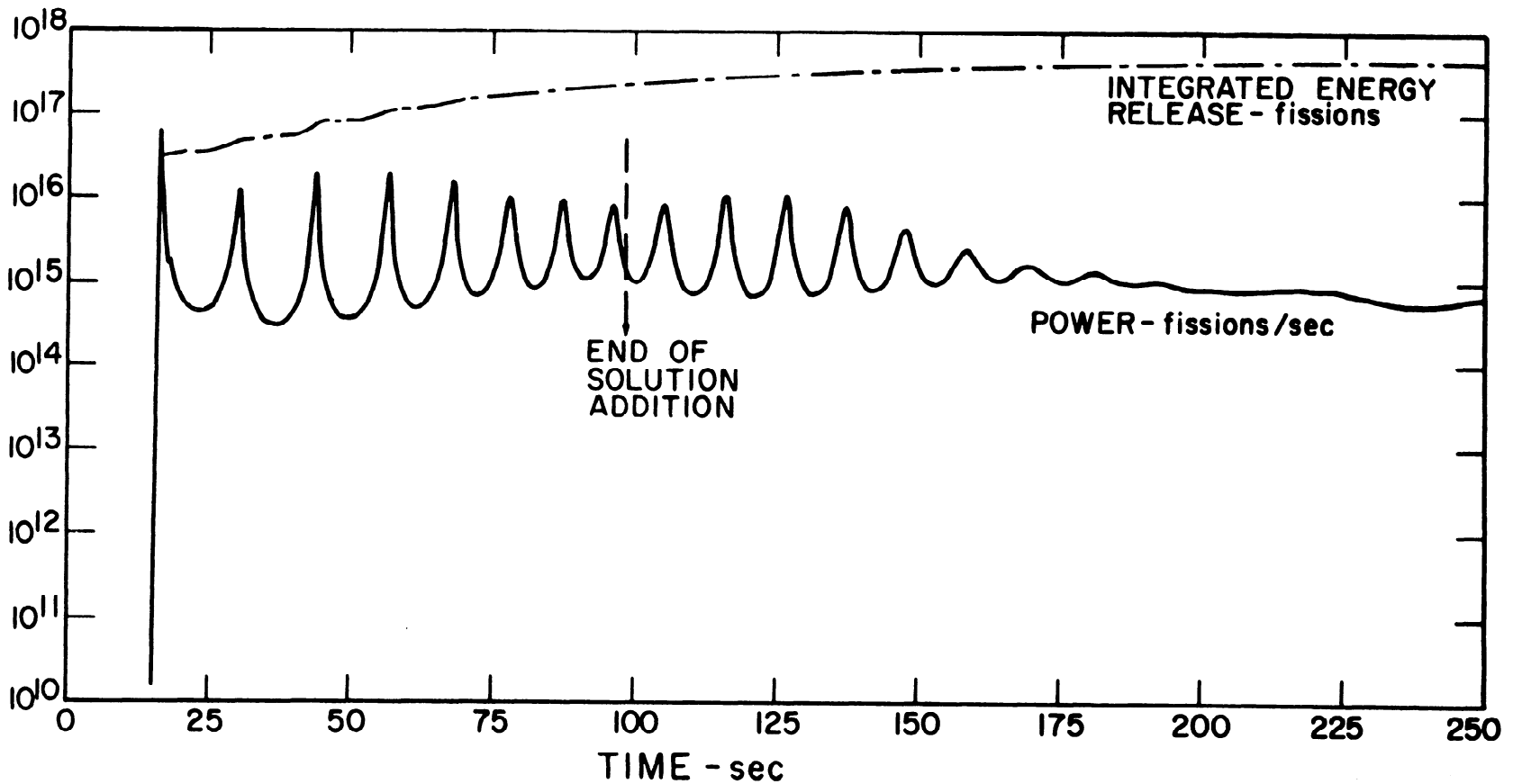

Fig. 5. Fission Rate and Integrated Fission Energy Release in CRAC 19 as a Function of Time.

III. THE EXPERIMENTAL PARAMETERS AND RESULTS

A summary of the parameters has been assembled for the experiments from the CRAC program. The data as presented are taken directly from the appropriate report in the CRAC series. The critical dimensions of the cold uranyl nitrate solutions in the 300-mm- and 800-mm-diam cylinders are given as a function of uranium concentration in Figs. 6 and 7, respectively. These data are presented for each CRAC experiment in Tables 1 and 2. In addition to the "excursion experiments," for each particular geometric set described below, a group of preliminary measurements was made to provide information on the basic nuclear characteristics of various concentration solutions used in the program. In this latter category of experiments, referred to as the "slow excursion" experiments, a series of stable reactor period measurements was made for a fixed slightly supercritical volume of solution of a given concentration. The reactivity worth of an incremental volume of solution in excess of the critical volume was thus determined. In the terminology adopted in the original reports, this group of tests is identified by the insertion of the letter "D" before the experiment number followed by a slow excursion number. Thus, the first slow excursion performed using the solution made up for CRAC 08 experiment has been identified as CRAC D 08-01. The results of the slow excursions are given in Table 3. Also included are values of neutron lifetime and β_{eff} for the various solution concentrations. Reactivity and β_{eff} are reported in pcm:

$$\rho(\text{pcm}) = (k_{ex} - 1) \times 10^5 \quad .$$

Plotting the reactivity in dollars versus the fractional change in height above delayed criticality for various solutions in the 300-mm-diam vessel suggests that reactivity initially increases linearly with height above delayed criticality (Fig. 8). From the slopes of the lines in Fig. 8, the reactivity change as a function of height is obtained for each solution concentration. Since the radius is constant, this also gives the reactivity change as a function of solution volume.

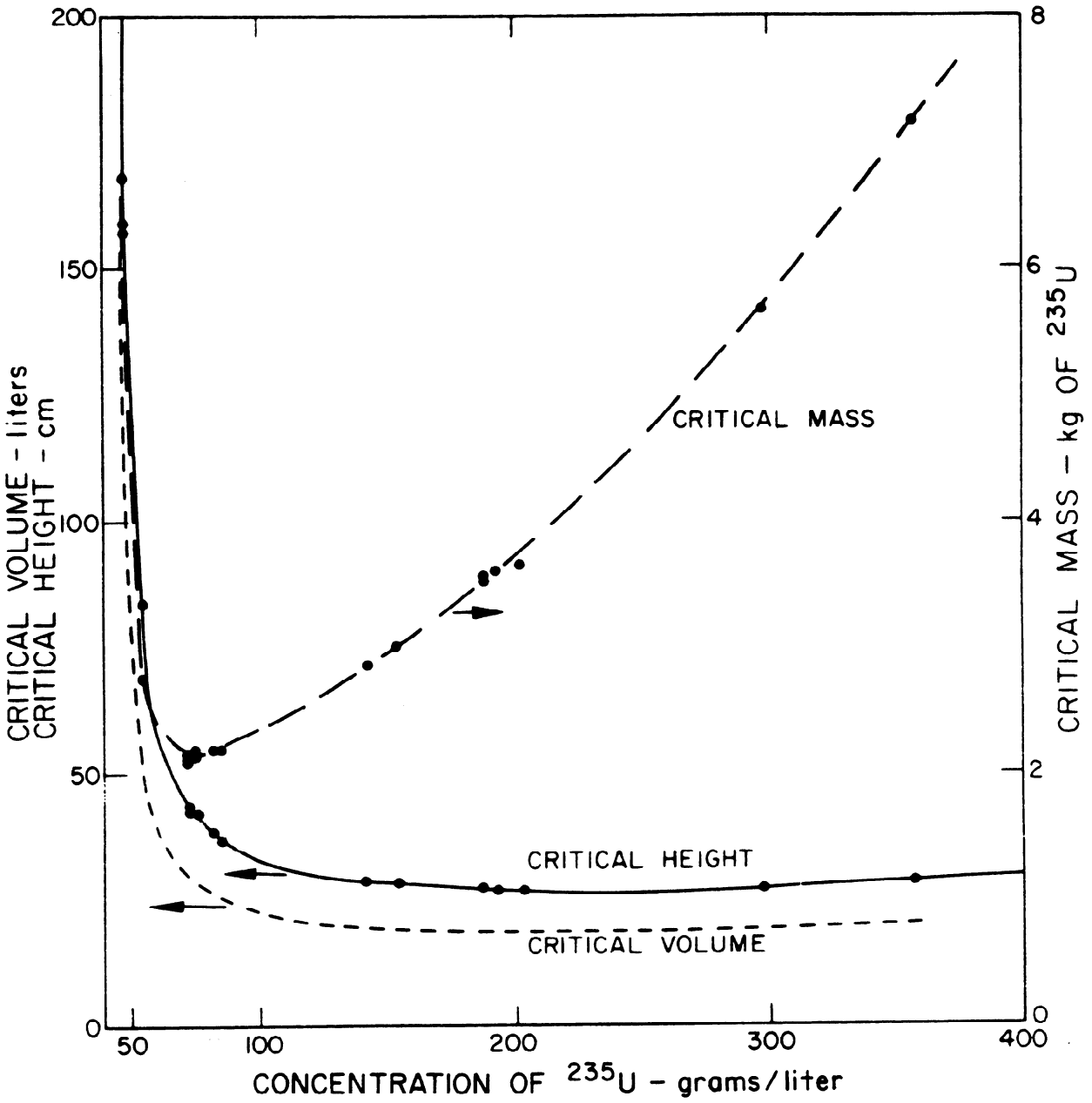

Fig. 6. Critical Dimensions of Cold, Clean Uranyl Nitrate Solution in the 300-mm-diam Cylinder.

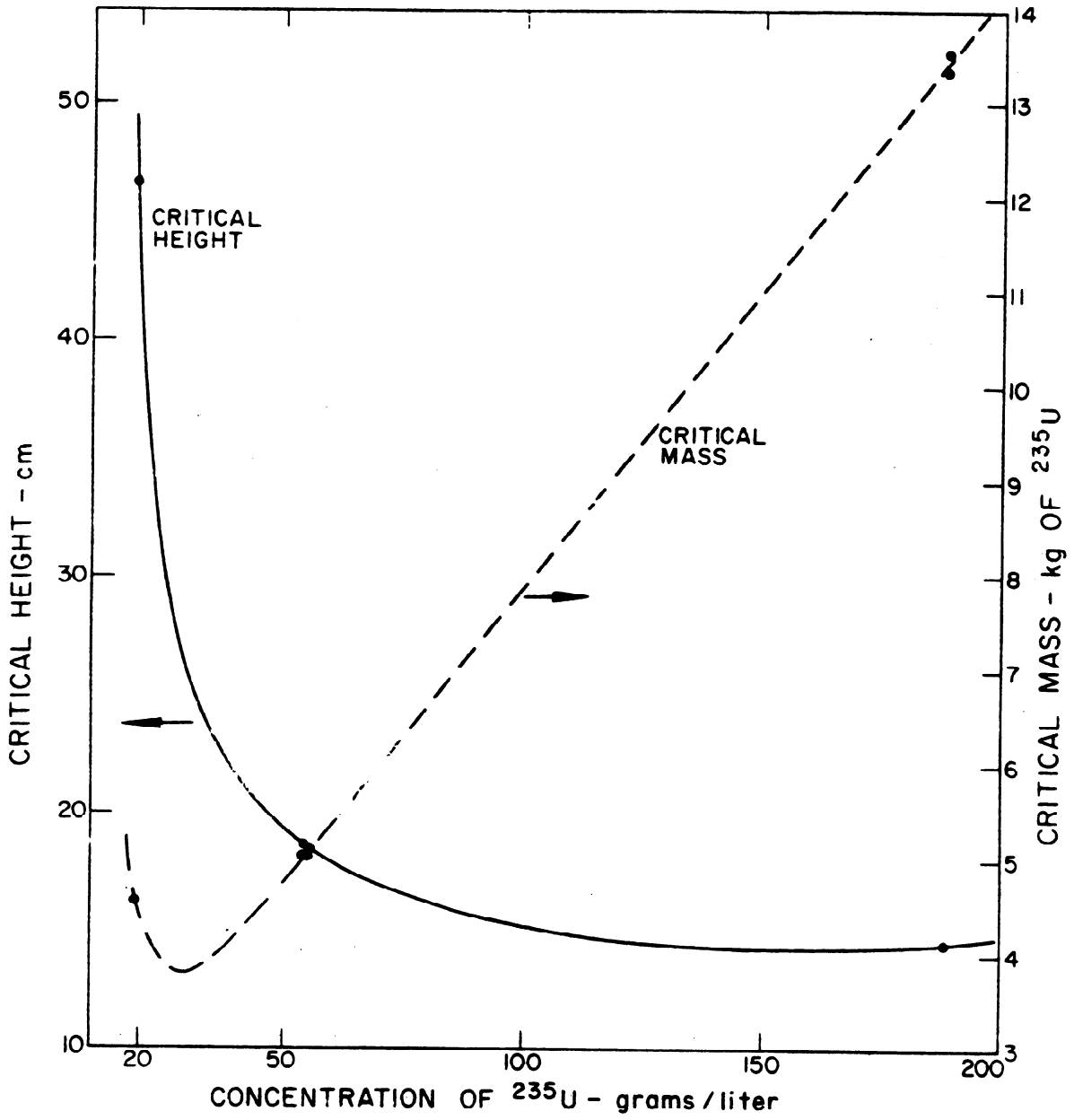

Fig. 7. Critical Dimensions of Cold, Clean Uranyl Nitrate Solution in the 800-mm-diam Cylinder.

Table 1. Summary of Critical Data for Experiments in the 300-mm-diam Cylinder.

Experiment Number	Uranium Concentration (g/liter)	^{235}U Concentration (g/liter)	Critical Dimensions			
			Solution Height, H_c (cm)	Solution Volume, V_c (liter)	Uranium Mass (kg)	^{235}U Mass (kg)
CRAC 01	52.0	48.4	191.3	129.8	6.75	6.28
CRAC 02[a]	{139 liter of 51.5 g/liter; 50 liter of 62.3 g/liter}	----	206.5	140.0	7.21	6.72
CRAC 03	52.2	48.6	182.1	123.5	6.45	6.00
CRAC 04	51.7	48.2	193.9	131.5	6.80	6.34
CRAC 05	61.1	56.9	69.05	47.1	2.88	2.68
CRAC 06	61.1	56.9	68.06	46.5	2.84	2.64
CRAC 07	202	188	27.02	18.7	3.78	3.52
CRAC 08	202	188	27.25	18.9	3.81	3.55
CRAC 09	78.3	72.9	42.90	29.5	2.31	2.15
CRAC 10	78.3	72.9	42.61	29.3	2.29	2.13
CRAC 11	383	357	28.94	20.0	7.66	7.14
CRAC 12	77.9	72.5	43.53	29.9	2.33	2.17
CRAC 13	77.9	72.5	48.90	33.5	2.61	2.43
CRAC 14	77.9	72.5	42.30	29.0	2.26	2.11
CRAC 15	82.3	76.6	41.15	28.3	2.33	2.17
CRAC 16	82.3	76.6	42.00	28.8	2.37	2.21
CRAC 17	79.6	74.1	41.90	28.8	2.29	2.13
CRAC 18	79.6	74.1	41.97	28.8	2.29	2.13
CRAC 19	82.0	76.4	41.50	28.5	2.34	2.18
CRAC 20.1	218	203	27.10	18.8	4.09	3.81
CRAC 20.2	218	203	27.10	18.8	4.09	3.81
CRAC 20.3	218	203	27.10	18.8	4.09	3.81
CRAC 20.4	218	203	27.10	18.8	4.09	3.81
CRAC 20.5	218	203	27.10	18.8	4.09	3.81
CRAC 21	80.2	74.7	41.85	28.7	2.31	2.15
CRAC 22	207	193	26.91	18.6	3.86	3.60
CRAC 23	91.8	85.5	37.00	25.5	2.34	2.18
CRAC 24	153.2	142.7	28.85	20.0	3.06	2.85
CRAC 25	320	298	27.47	19.0	6.09	5.67
CRAC 26	165.9	154.5	28.41	19.7	3.26	3.04
CRAC 27	89.0	82.9	38.35	26.4	2.35	2.19
CRAC 28	89.0	82.9	38.35	26.4	2.35	2.19
CRAC 29	81.0	75.4	42.37	29.1	2.36	2.19

a. The critical dimensions are those for the solution of concentration 51.5 g of U/liter (48 g of ^{235}U/liter). In the experiment, 139 liters of this solution was transferred to the test vessel, followed by 50 liters of solution containing 62.3 g of U/liter (58 g of ^{235}U/liter). The final volume, 189 liters, was assumed to be homogeneous and of concentration 50.5 g of ^{235}U liter.

Table 2. Summary of Critical Data for Experiments in the 800-mm-diam Cylinder.

Experiment Number	Uranium Concentration (g/liter)	^{235}U Concentration (g/liter)	Critical Dimensions			
			Solution Height (cm)	Solution Volume (liter)	Uranium Mass (kg)	^{235}U Mass (kg)
CRAC 37	21.8	20.3	45.0	221.9	4.84	4.50
CRAC 38	21.4	19.9	46.75	230.5	4.93	4.59
CRAC 39	30.6	28.5	27.25	134.4	4.11	3.83
CRAC 40	58.7	54.7	18.65	92.0	5.40	5.03
CRAC 41	59.3	55.2	18.55	91.4	5.42	5.05
CRAC 42	203	189	14.56	71.8	14.54	13.54
CRAC 43	202	188	14.38	70.9	14.35	13.36
CRAC 44	Introduction of 46 liters of fissile solution (uranium concentration 200 g/liter) into 200 liters of 2N acid.					

Table 3. Reactivity Worth of Various Solutions Determined by Stable Period Measurements.

Experiment Number[a]	Uranium Concentration (g/liter)	^{235}U Concentration (g/liter)	β_{eff} (pcm)	Neutron Lifetime (μsec)	Critical Solution Height, H_c (cm)	Incremental Change in Height, ΔH (cm)	$\Delta H/H_c$	Reactivity (pcm)	Reactivity (dollars)
CRAC D 01-02	52.0	48.4	777	41	198	21.9	0.111	96.4	0.124
						32.1	0.162	140	0.180
						41.3	0.209	179	0.230
						60	0.303	256	0.332
CRAC D 05-01	61.1	56.9	774	37	68	1.99	0.0293	180	0.2325
-02						3.07	0.0451	270	0.3488
-03						4	0.0589	351	0.4535
CRAC D 09-01	77.9	72.5	814	31	43.26	0.40	0.00925	134.5	0.1652
-02						0.81	0.0187	264	0.3243
-03						1.31	0.0303	409	0.5024
CRAC D 08-01	203	189	845	13.5	27.5	0.21	0.00764	204	0.2414
-02						0.35	0.0127	332	0.3929
-03						0.46	0.0167	434	0.5136
CRAC D 11-03	325	303	854	----	26.19	0.45	0.0172	467	0.547

a. These experiments are different from those reported in Tables 1 and 6 which may account for slight differences in the critical quantities.

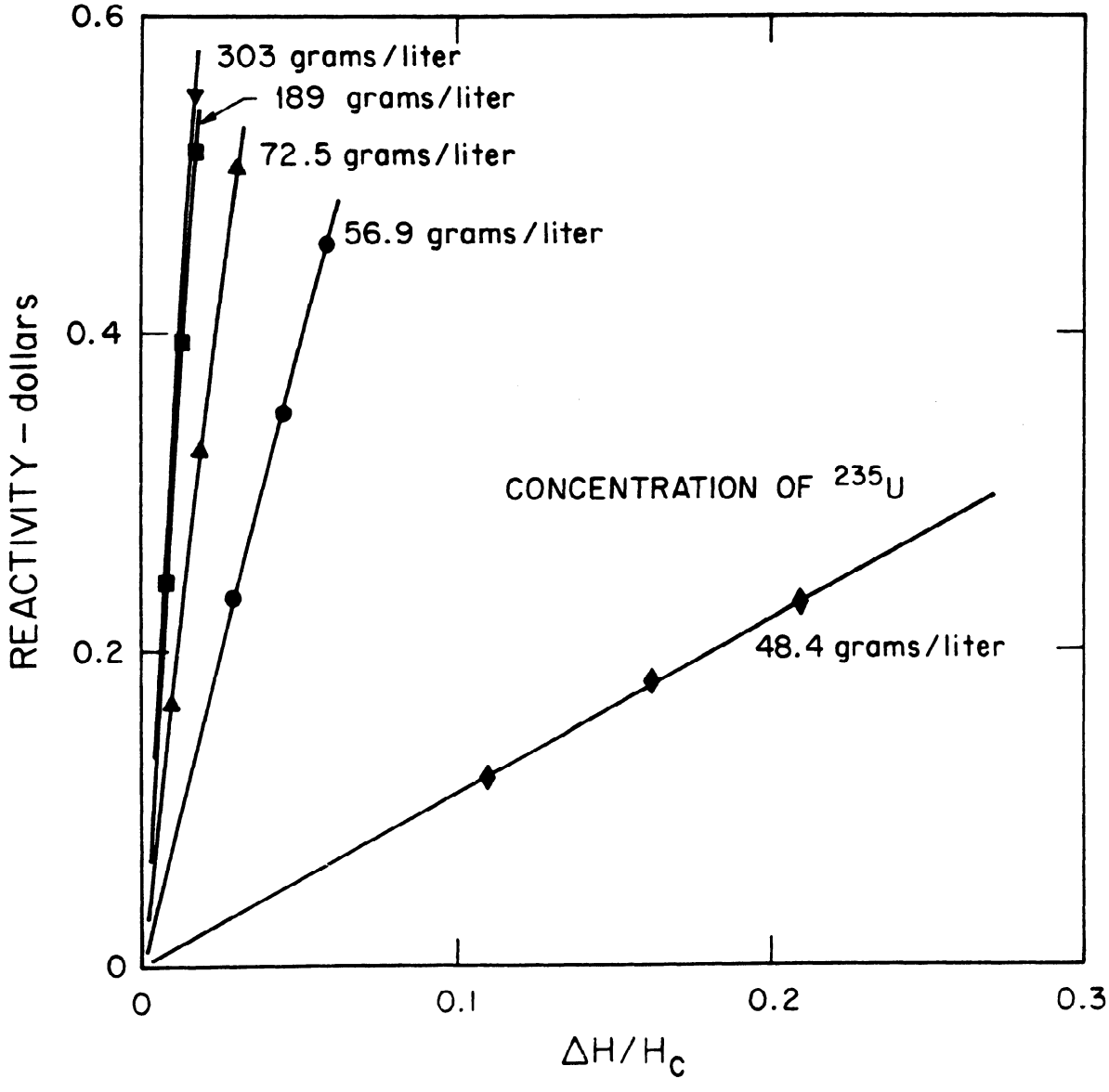

Fig. 8. Positive Reactivity Increment Resulting from a Step Increment of Solution Above Criticality for Various Solution Concentrations in the 300-mm-diam Cylinder.

Some of the "stable period" experiments made using the slow excursions were allowed to progress through the occurrence of a "pulse." These pulses were quite broad, with the minimum period being ~4 sec and, correspondingly, the peak fission rate was less than 10^{16} fissions/sec. In these experiments, the excess reactivity was added in a time short compared to the time to achieve peak power and hence can be considered as resulting from the addition of a step change in reactivity. The reactivity step was, in every case, less than 500 pcm. The results of these slow excursions are summarized in Tables 4 and 5.

The stable period experiments for each solution yield a measure of reactivity addition per unit change in solution volume near criticality in the 300-mm-diam cylinder; i.e., $\Delta\rho/\Delta V$. From these results can be obtained the rate of ramp addition of reactivity, in dollars/sec, per unit rate of solution addition, in liters/h. This result for the 300-mm-diam cylinder is plotted in Fig. 9.

The excursions were produced by injecting solution into the experimental vessel at a fixed rate. Generally, solution was added to the reacting volume well past the occurrence of the initial pulse. In addition to the power level, other measurements during an experiment provided a more complete record of the sequence of events. A typical set of experimental data from CRAC 23 is described. In Fig. 10 the power and the integrated energy release traces are shown as a function of time. Also plotted is the computed reactivity of the experimental assembly based on a dynamic solution of the inhour equation and appropriate values of neutron lifetime and β_{eff} by an on-line computer. In Fig. 11 a more detailed trace of power as a function of time through the first pulse is shown. Gamma-ray dose rates were measured during most of the experiments. A recorder trace of the dose rate for CRAC 23 is shown in Fig. 12. Two temperature traces are shown in Fig. 13, one obtained from a thermocouple placed on the center line of the cylinder and the other from a thermocouple located on the external wall of the vessel. These results are typical of the data taken during each experiment; complete sets of data are available in the individual experiment reports.

Table 4. Slow Excursions in the 300-mm-diam Cylinder.

Experiment Number[a]	Uranium Concentration (g/liter)	^{235}U Concentration (g/liter)	Critical Dimensions: Solution Height (cm)	Solution Volume (liter)	Uranium Mass (kg)	^{235}U Mass (kg)	Final Dimensions: Solution Height (cm)	Solution Volume (liter)
CRAC D 01-02	52.0	48.4	198	134.3	6.98	6.50	258	179
CRAC D 05-02	61.1	56.9	68	46.4	2.84	2.64	72	49
CRAC D 08-03	203	189	27.5	19.0	3.86	3.59	27.96	19.4
CRAC D 11-03	325	303	26.19	18.2	5.92	5.51	26.64	18.5

Experiment Number[a]	Final Mass: Uranium (kg)	Final Mass: ^{235}U (kg)	Excess Height, ΔH (cm)	Duration of Experiment (min)	Total Energy (fissions)	Solution Height at Peak Power (cm)	Solution Volume at Peak Power (liter)	Minimum Doubling Time (sec)	Reciprocal Period (sec^{-1})
CRAC D 01-02	9.31	8.66	60	10	2.0×10^{17}	258	179	10.4	0.0666
CRAC D 05-02	2.99	2.79	4	5	6.8×10^{16}	72	49	5.18	0.134
CRAC D 08-03	3.94	3.67	0.46	4	3.5×10^{16}	27.96	19.4	3.75	0.185
CRAC D 11-03	6.01	5.61	0.45	5	4.1×10^{16}	26.64	18.5	3.11	0.222

Experiment Number[a]	Peak Power (fissions/sec)	Time to End of Pulse (sec)	Total Energy in Pulse (fissions)	Specific Power at Peak Power (fissions/cm^3-sec)	Specific Energy in Pulse (fissions/cm^3)	β_{eff} (pcm)	Reactivity Added (pcm)
CRAC D 01-02	1.6×10^{15}	600	2.0×10^{17}	0.89×10^{10}	1.1×10^{12}	777	256
CRAC D 05-02	1.1×10^{15}	290	6.8×10^{16}	2.2×10^{10}	1.4×10^{12}	774	351
CRAC D 08-03	7.8×10^{14}	230	3.5×10^{16}	4.0×10^{10}	1.8×10^{12}	845	434
CRAC D 11-03	9.8×10^{14}	270	4.1×10^{16}	5.3×10^{10}	2.2×10^{12}	854	467

a. These experiments are different from those reported in Tables 1 and 6 which may account for slight differences in the critical quantities.

Table 5. Slow Excursions in the 800-mm-diam Cylinder.

Experiment Number[a]	Uranium Concentration (g/liter)	^{235}U Concentration (g/liter)	Critical Dimensions				Final Dimensions	
			Solution Height (cm)	Solution Volume (liter)	Uranium Mass (kg)	^{235}U Mass (kg)	Solution Height (cm)	Solution Volume (liter)
CRAC D 37-02	21.4	19.9	46.77	230.6	4.93	4.59	47.57	234.5
CRAC D 39-02	30.6	28.5	27.24	134.3	4.11	3.83	27.54	135.8
CRAC D 40-02	58.7	54.7	18.61	91.8	5.39	5.02	18.79	92.6

Experiment Number	Final Mass		Excess Height, ΔH (cm)	Duration of Experiment (min)	Total Energy (fissions)	Solution Height at Peak Power (cm)	Solution Volume at Peak Power (liter)	Minimum Doubling Time (sec)	Reciprocal Period (sec^{-1})
	Uranium (kg)	^{235}U (kg)							
CRAC D 37-02	5.02	4.67	0.80	5	3.9×10^{17}	47.57	234.5	6.76	0.103
CRAC D 39-02	4.16	3.87	0.30	5	3.0×10^{17}	27.54	135.8	3.24	0.214
CRAC D 40-02	5.44	5.07	0.18	5	2.7×10^{17}	18.79	92.6	2.48	0.279

Experiment Number	Peak Power (fissions/sec)	Time to End of Pulse (sec)	Total Energy in Pulse (fissions)	Specific Power at Peak Power (fissions/cm^3-sec)	Specific Energy in Pulse (fissions/cm^3)	β_{eff} (pcm)	Reactivity Added (pcm)
CRAC D 37-02	5.0×10^{15}	320	3.9×10^{17}	2.1×10^{10}	1.7×10^{12}	711	288
CRAC D 39-02	6.2×10^{15}	300	3.0×10^{17}	4.6×10^{10}	2.2×10^{12}	747	404
CRAC D 40-02	7.4×10^{15}	260	2.7×10^{17}	8.0×10^{10}	2.9×10^{12}	780	460

a. These experiments are different from those reported in Tables 2 and 7 which may account for slight differences in the critical quantities.

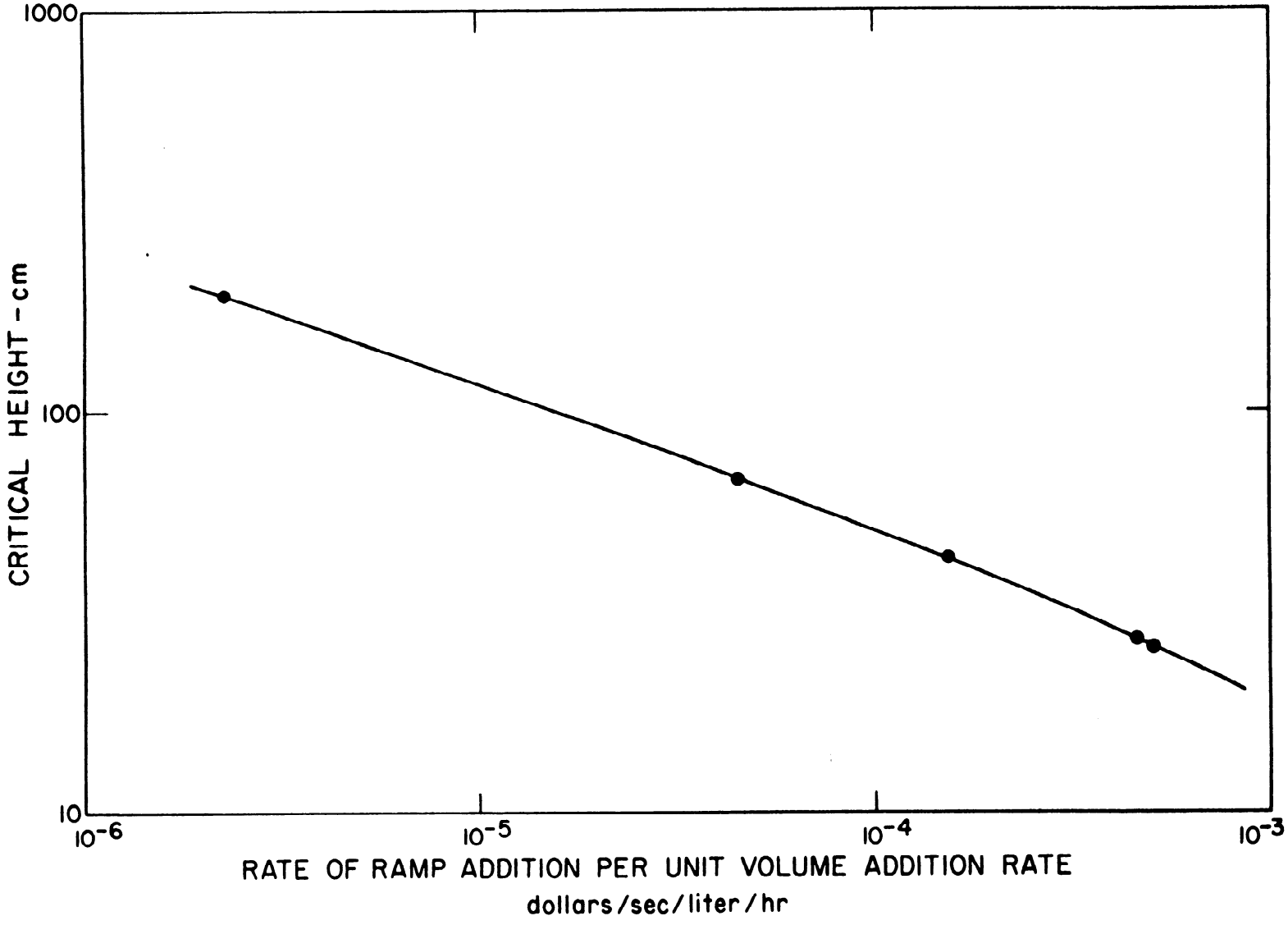

Fig. 9. Rate of Ramp Addition of Reactivity per Unit Volume Addition Rate Near Criticality as a Function of the Critical Height in the 300-mm-diam Cylinder.

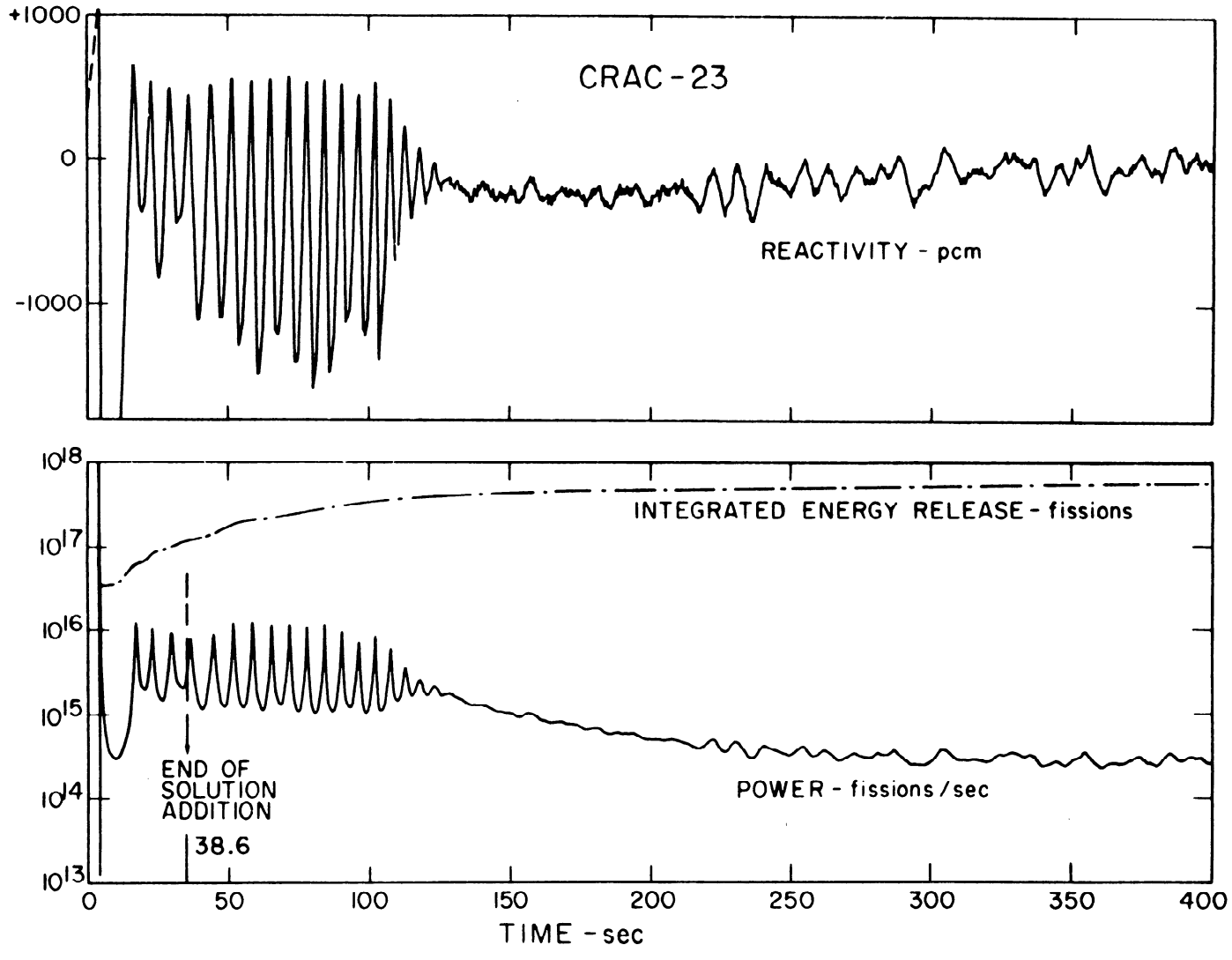

Fig. 10. Instantaneous Power, Integrated Energy Release, and Reactivity as a Function of Time.

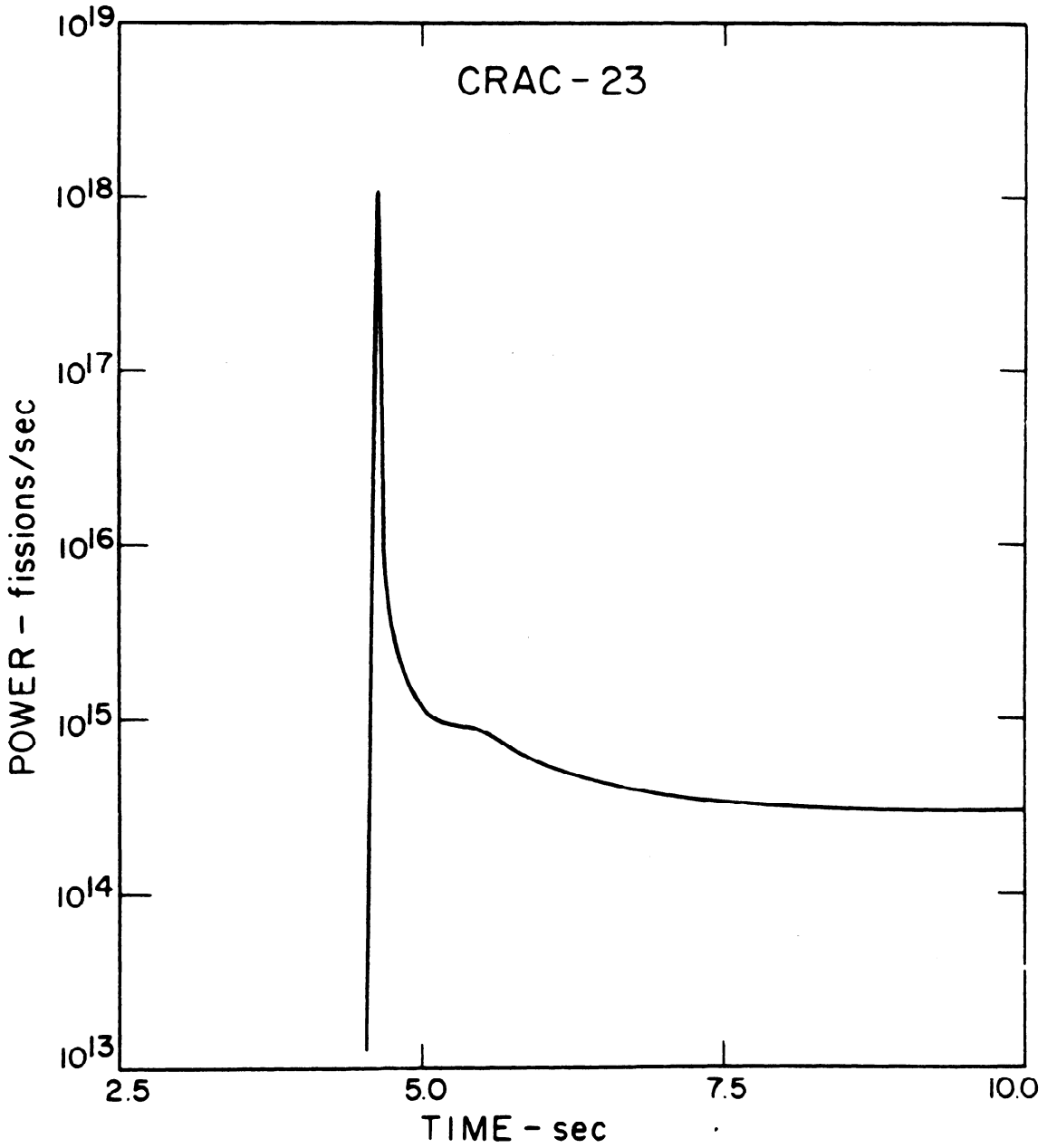

Fig. 11. Power Trace for the First Pulse in CRAC 23.

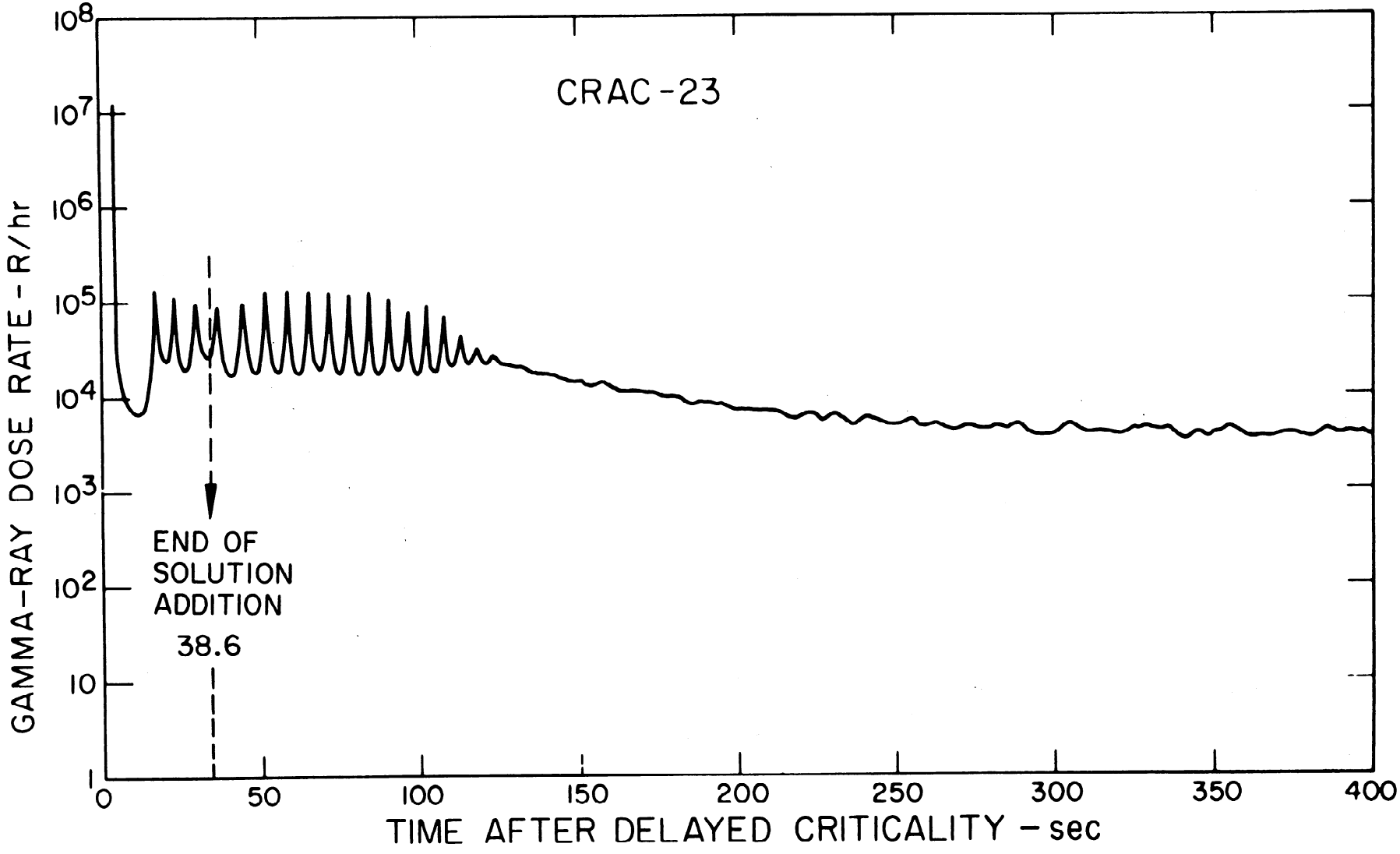

Fig. 12. Gamma-Ray Dose Rate as a Function of Time at a Detector Located 3 m from the CRAC Assembly.

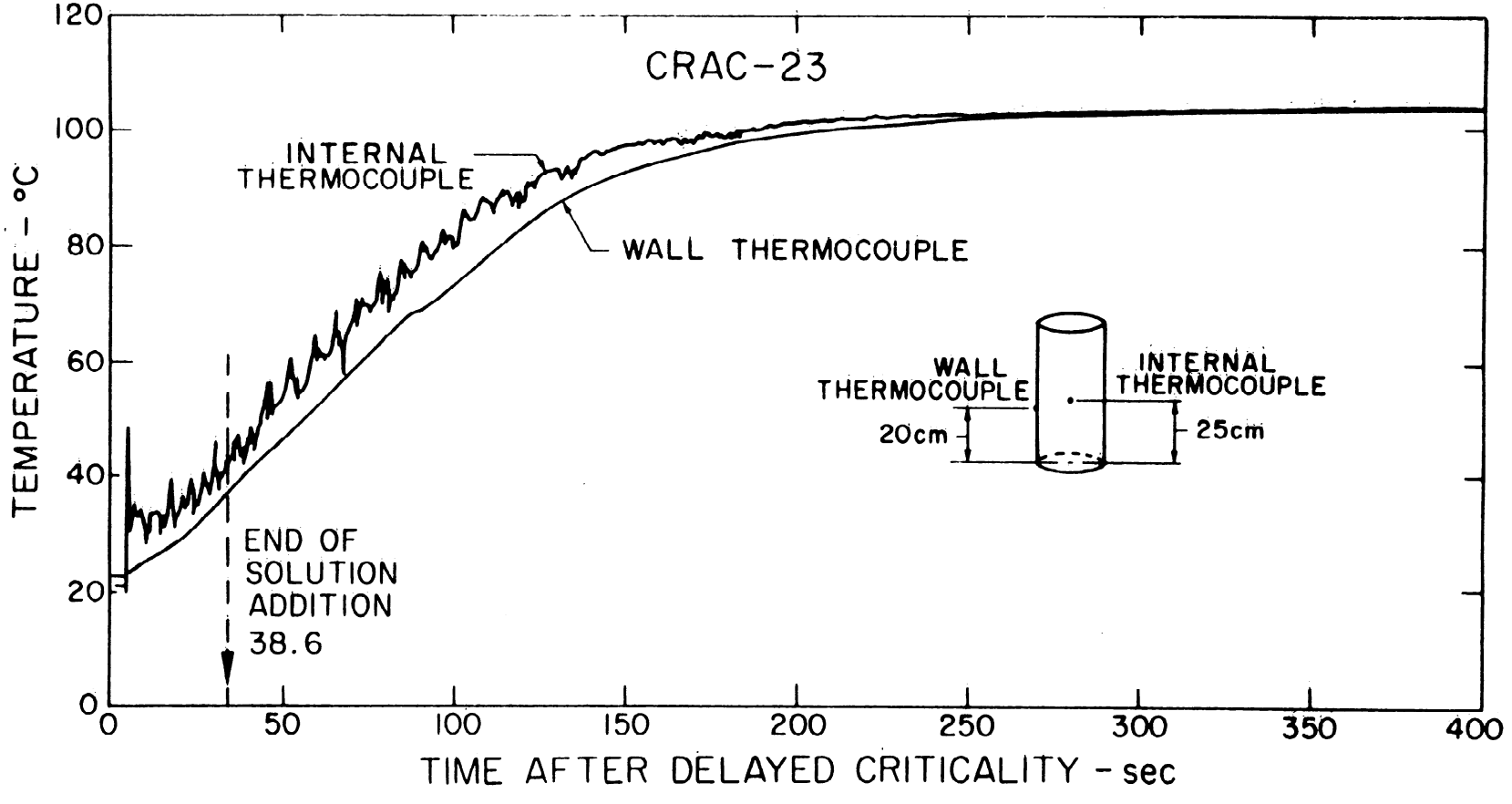

Fig. 13. Temperature Traces for CRAC 23. Critical height for cold solution was 37 cm. Solution addition terminated at 38.6 sec.

Several experiments were carried out under special conditions which include the following:

a. In CRAC 13, samples of a fuel solution, with and without $CuSO_4$ added, were placed at the center of the test vessel to study the value of $CuSO_4$ as a catalyst in the recombination of evolved radiolytic gases. No apparent effect was observed.

b. In CRAC 15, 16, 17, and 18, solution in excess of that necessary for cold, clean criticality was allowed to come to equilibrium power by the production of radiolytic gas prior to starting the pump to inject more fuel into the vessel. Hence, an initial neutron population was present when reactivity was added.

c. In CRAC 14, 20.1, 20.2, and 20.3, a source was located near the vessel to provide neutrons to start the first fission chain early in the reactivity ramp. CRAC 20.4 did not have a nearby external source; CRAC 20.5 was also done without an external source but took place 30 min after CRAC 20.4.

The results of the CRAC excursions are assembled in Tables 6 and 7. The criticality parameters (concentrations, critical height, critical volume, and critical mass) for the various experiments are given in Tables 1 and 2. In addition to the initial and final solution heights, the solution addition rate, the solution addition time, the experiment duration, and the total yield are given.

Of particular interest are the parameters describing the first peak in the pulse chain. These data are shown in Tables 8 and 9 for the experiments in the 300-mm- and 800-mm-diam cylinders, respectively.

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Table 6. Summary of Excursion Data from the 300-mm-diam Cylinder.

Experiment Number	Solution Height at Start of Addition (cm)	Duration of Addition (sec)	Solution Addition Rate		Final Solution		Final Mass		Excess Above Critical Height, $H_f - H_c$ (cm)[c]	Time Required for $(H_f - H_c)$ Addition[a] (sec)	Duration of Experiment (min)	Time to End of Oscillations[a] (sec)	Total Yield in	
			Height (cm/sec)	Volume (liter/h)	Height, H_f (cm)	Volume, V_f (liter)	Uranium (kg)	^{235}U (kg)					Oscillations (10^{17} fissions)	Experiment[b] (10^{17} fissions)
CRAC 01	186.1	313	0.592	1440	371.4	251.5	13.08	12.18	180.1	304.2	260	10 000	4.6	5.0
CRAC 02[c]	196.3	121	0.683	1663	279.0	189.0	10.27	9.57	72.5	106.2	22	350	4.7	6.6
CRAC 03	127.1	1116	0.215	524	367.4	248.8	12.99	12.10	185.3	860.6	40	1 000	3.2	4.0
CRAC 04	183.2	235	0.698	1699	347.3	235.2	12.16	11.33	153.4	219.7	260	10 000	3.7	4.0
CRAC 05	13.23	143	0.609	1483	100.4	68.3	4.17	3.89	31.4	51.4	19	160	2.8	3.8
CRAC 06	10.03	143	0.629	1531	100.1	68.1	4.16	3.87	32.0	50.9	20	160	2.6	3.6
CRAC 07	10.06	33	0.687	1672	32.8	22.6	4.56	4.24	5.8	8.3	10	120	2.2	3.1
CRAC 08	10.04	34	0.666	1622	32.8	22.6	4.56	4.24	5.6	8.2	10	120	2.2	2.7
CRAC 09	10.10	69	0.655	1594	55.3	37.8	2.96	2.76	12.4	18.9	6	100	2.1	2.9
CRAC 10	12.23	267	0.202	492	66.3	45.2	3.54	3.30	23.7	117.0	13	235	5.0	5.8*
CRAC 11	10.10	40	0.618	1504	34.8	24.0	9.19	8.57	5.9	9.5	6	--	--	--
CRAC 12	9.98	1239	0.043	104	62.8	42.9	3.34	3.11	19.3	451.0	17	600	4.3	4.8
CRAC 13[d]	10.02	101	0.587	1429	69.3	47.3	3.69	3.43	20.4	34.8	7	150	3.5	4.6
CRAC 14[e]	41.78	99	0.255	620	67.0	45.8	3.57	3.32	24.7	97.0	13	240	4.9	5.8
CRAC 15[f]	41.27	39	0.612	1489	65.1	44.5	3.66	3.41	24.0	39.0	5	190	5.2	5.5
CRAC 16[f]	42.84	205	0.191	466	82.1	56.0	4.61	4.29	40.1	205.0	13	--	--	9.0
CRAC 17[f]	42.29	97	0.204	497	62.1	42.5	3.38	3.15	20.2	97.0	6	200	4.3	4.9
CRAC 18[f]	42.34	1404	0.043	104	102.4	69.7	5.54	5.16	60.4	1404	360	--	--	50
CRAC 19	10.06	252	0.210	512	63.1	43.1	3.53	3.29	21.6	102.7	5	200	4.3	4.9
CRAC 20.1[g]	4.86	44	0.602	1466	31.4	21.7	4.72	4.39	4.3	7.1	4	190	1.9	2.0
CRAC 20.2[g]	4.90	46	0.611	1488	33.0	22.8	4.96	4.62	5.9	9.7	3	180	2.8	2.8*
CRAC 20.3[g]	4.92	41	0.617	1502	30.2	20.9	4.55	4.23	3.1	5.1	0.3	--	--	0.22*
CRAC 20.4	4.92	42	0.612	1489	30.6	21.2	4.61	4.29	3.5	5.8	0.3	--	--	0.61*
CRAC 20.5	4.91	51	0.551	1340	33.0	22.8	4.96	4.62	5.9	10.7	3	180	2.7	2.7*
CRAC 21	9.93	257	0.207	505	63.3	43.2	3.47	3.23	21.5	103.5	5	200	4.2	4.9
CRAC 22	7.90	58	0.438	1066	33.3	23.0	4.76	4.43	6.4	14.6	3	120	2.0	2.2
CRAC 23	7.92	90	0.566	1377	58.9	40.2	3.69	3.44	21.9	38.6	7	140	4.3	5.3
CRAC 24	10.03	--	--	--	57.5	39.3	6.03	5.61	28.7	--	6	--	--	5.8
CRAC 25	10.14	30	0.766	1864	33.0	22.7	7.28	6.78	5.5	7.2	0.2	--	--	0.39
CRAC 26	9.97	49	0.624	1520	40.6	27.9	4.63	4.30	12.2	19.5	3	140	2.9	3.0
CRAC 27	10.11	82	0.631	1537	61.9	42.3	3.77	3.51	23.6	37.3	1.5	--	--	2.8
CRAC 28	10.27	109	0.452	1102	59.6	40.8	3.63	3.38	21.3	47.1	5	150	4.1	5.1*
CRAC 29	10.17	262	0.207	505	64.6	44.1	3.58	3.33	22.2	107.5	3	180	4.3	4.3

a. Zero time was achievement of criticality.
b. The yield was determined by radiochemistry except those designated * were from power detectors.
c. The critical dimensions are those for the solution of concentration 51.5 g of U/liter (48 g of ^{235}U liter). In the experiment, 139 liters of this solution was transferred to the test vessel, followed by 50 liters of solution containing 62.3 g of U/liter (58 g of ^{235}U liter). The final volume, 189 liters, was assumed to be homogeneous and of concentration 50.5 g of ^{235}U/liter.
d. The use of $CuSO_4$ as a catalyst was tested in this experiment.
e. A 100 mCi source of neutrons was placed near the vessel.
f. The solution, initially supercritical, had come to equilibrium by the production of radiolytic gas prior to the initiation of the ramp, thereby providing a source of neutrons. The solution boiled in CRAC 16 and 18.
g. A 200 mCi neutron source was placed under the bottom of the vessel.

Table 7. Summary of Data from Experiments in the 800-mm-diam Cylinder.

Experiment Number	Solution Height at Start of Addition (cm)	Duration of Addition (sec)	Solution Addition Rate		Final Solution		Final Mass	
			Height (cm/sec)	Volume (liter/hr)	Height (cm)	Volume (liter)	Uranium (kg)	^{235}U (kg)
CRAC 37	44.0	322	0.0252	448	52.1	256.9	5.60	5.22
CRAC 38	44.0	115	0.0741	1315	52.5	259.0	5.54	5.15
CRAC 39	26.0	38	0.0787	1397	29.0	142.9	4.37	4.07
CRAC 40	18.0	28	0.0468	830	19.3	95.2	5.59	5.21
CRAC 41	18.0	212	0.0055	97	19.2	94.5	5.60	5.22
CRAC 42	14.0	21	0.0719	1276	15.5	76.4	15.47	14.41
CRAC 43	14.0	19	0.0793	1407	15.5	76.4	15.46	14.40
CRAC 44	Introduction of 46 liters of fissile solution (uranium concentration 200 g/liter) into 200 liters of 2N acid				49.9	246	9.20	8.57

Experiment Number	Change in Solution Height (cm)	Time of Addition Above Critical Height (sec)	Duration of Experiment (min)	Yield in Experiment (from Radiochemistry) (10^{18} fissions)
CRAC 37	7.1	282	7	3.2
CRAC 38	5.8	78	4	2.7
CRAC 39	1.7	22	3	1.6
CRAC 40	0.65	14	2	1.08
CRAC 41	0.62	112	4	1.02
CRAC 42[a]	0.94	13	1	0.46
CRAC 43[a]	1.1	14	3	1.05
CRAC 44[a,b]	--	--	1	1.7

a. Solution was ejected from the vessel.
b. The cylinder supports were bent and there was some damage to vessel.

Table 8. Characteristics of the First Pulse of the Experiments in the 300-mm-diam Cylinder.

Experiment Number	Time to Peak of Pulse[a] (sec)	Solution: Height at Peak Power (cm)	Solution: Volume at Peak Power (liter)	Rate of Reactivity Addition (dollars/sec)	Minimum Doubling Time (sec)	Inverse Period (sec^{-1})	Peak Power (fissions/sec)
CRAC 01	232	329	226	0.00341	2.9	0.24	1.1×10^{16}
CRAC 02	72	255	173	--	0.18	3.9	6.6×10^{16}
CRAC 03	427	274	186	0.00141	5.0	0.138	4.7×10^{15}
CRAC 04	197	331	225	0.00391	3.2	0.216	8.7×10^{15}
CRAC 05	21.6	82.2	56.0	0.0667	0.060	11.6	6.3×10^{16}
CRAC 06	22.8	82.4	56.2	0.0740	0.050	13.9	6.6×10^{16}
CRAC 07	3.9	29.7	20.5	0.786	0.00157	442	5.0×10^{18}
CRAC 08	3.1	29.3	20.3	0.746	0.00069	1004	3.0×10^{19}
CRAC 09	6.4	47.1	32.3	0.247	0.015	46	2.9×10^{17}
CRAC 10	6.6	44.0	30.2	0.0772	0.0176	39.4	2.0×10^{17}
CRAC 11	--	--	--	--	--	--	--
CRAC 12	65	46.3	31.8	0.0156	0.275	2.52	1.0×10^{16}
CRAC 13	7.5	53.3	36.5	0.157	0.012	57.7	5.3×10^{17}
CRAC 14	12.0	45.4	31.1	0.0992	0.049	14.1	4.5×10^{16}
CRAC 15	4.0	43.6	29.9	0.253	0.033	20.8	1.1×10^{17}
CRAC 16	11.2	44.1	30.3	0.0755	0.242	2.86	1.6×10^{16}
CRAC 17	11.7	44.3	30.4	0.0820	0.177	3.92	2.0×10^{16}
CRAC 18	43.8	43.8	30.1	0.0168	0.52	1.33	7.7×10^{15}
CRAC 19	16.2	44.9	30.8	0.0870	0.036	19.2	6.7×10^{16}
CRAC 20.1	2.2	28.4	19.7	0.674	0.0061	114	5.3×10^{17}
CRAC 20.2	2.2	28.4	19.7	0.684	0.0063	110	5.2×10^{17}
CRAC 20.3	2.4	28.6	19.8	0.691	0.0066	105	4.5×10^{17}
CRAC 20.4	3.4	29.2	20.2	0.685	0.00118	587	1.0×10^{19}
CRAC 20.5	2.5	28.5	19.7	0.616	0.0058	120	5.8×10^{17}
CRAC 21	17.0	45.4	31.1	0.0833	0.032	21.6	7.6×10^{16}
CRAC 22	4.6	28.9	20.0	0.501	0.00147	471	5.4×10^{18}
CRAC 23	4.6	39.6	27.2	0.310	0.0058	120	1.2×10^{18}
CRAC 24	--	--	--	--	--	--	--
CRAC 25	7.1	33.0	22.2	0.871	0.00153	453	4.9×10^{16}
CRAC 26	8.3	33.6	23.1	0.638	0.0027	252	2.6×10^{18}
CRAC 27	4.7	41.3	28.4	0.315	0.012	58	3.8×10^{17}
CRAC 28	8.8	42.3	29.1	0.226	0.011	63	4.1×10^{17}
CRAC 29	12.3	44.9	30.8	0.0808	0.032	21.7	7.3×10^{16}

Table 8 (Cont'd)

Experiment Number	Time to End of Pulse[a] (sec)	Total Energy in Pulse (10^{16} fissions)	Specific Power at Peak (fissions/cm^3-sec)	Specific Energy in Pulse (10^{12} fissions/cm^3)	β_{eff} (pcm)	Reactivity Added Through First Pulse (pcm)
CRAC 01	300	27	4.7×10^{10}	1.2	777	440
CRAC 02	90	18	3.8×10^{11}	1.0	---	---
CRAC 03	500	17	2.5×10^{10}	0.93	777	---
CRAC 04	300	22	3.9×10^{10}	1.0	777	---
CRAC 05	36	6.3	1.1×10^{12}	1.1	---	795
CRAC 06	36	6.7	1.2×10^{12}	1.2	---	805
CRAC 07	4	4.0	2.4×10^{14}	2.0	845	1230
CRAC 08	3.2	8.0	1.5×10^{15}	4.0	845	1680
CRAC 09	14	4.4	9.0×10^{12}	1.4	814	950
CRAC 10	12	4.3	6.6×10^{12}	1.4	814	---
CRAC 11	--	--	--	--	---	---
CRAC 12	75	3.7	3.2×10^{11}	1.2	814	725
CRAC 13	15	5.2	1.5×10^{13}	1.4	---	---
CRAC 14	18	4.0	1.5×10^{12}	1.3	814	840
CRAC 15	10	3.7	3.7×10^{12}	1.2	814	875
CRAC 16	18	3.6	5.3×10^{11}	1.2	814	760
CRAC 17	18	3.7	6.5×10^{11}	1.2	814	775
CRAC 18	50	3.7	2.6×10^{11}	1.2	814	680
CRAC 19	22	3.5	2.2×10^{12}	1.1	814	860
CRAC 20.1	3	2.0	2.7×10^{13}	1.0	841	990
CRAC 20.2	3	2.1	2.6×10^{13}	1.1	841	---
CRAC 20.3	3	2.0	2.3×10^{13}	1.0	841	---
CRAC 20.4	3.5	5.9	5.0×10^{14}	2.9	841	1430
CRAC 20.5	3	2.2	2.9×10^{13}	1.1	841	---
CRAC 21	23	3.1	2.5×10^{12}	1.0	---	---
CRAC 22	10	4.2	2.7×10^{14}	2.1	845	1320
CRAC 23	9	4.2	4.5×10^{13}	1.6	815	1060
CRAC 24	--	--	--	--	---	---
CRAC 25	10	3.9	2.2×10^{14}	1.7	854	1240
CRAC 26	12	3.9	1.1×10^{14}	1.7	838	1200
CRAC 27	10	3.5	1.3×10^{13}	1.2	---	---
CRAC 28	14	3.7	1.4×10^{13}	1.3	815	---
CRAC 29	19	3.3	2.4×10^{12}	1.1	---	960

a. Time measured from delayed criticality.

Table 9. Characteristics of the First Pulse of the Experiments in the 800-mm-diam Cylinder.

Experiment Number	Time to Pulse Peak[a] (sec)	Solution: Height at Pulse Peak (cm)	Solution: Volume at Pulse Peak (liter)	Rate of Reactivity Addition (dollars/sec)	Minimum Doubling Time (sec)	Inverse Period (sec^{-1})	Peak Power (fissions/sec)
CRAC 37	71	46.8	230.8	0.00381	0.345	2.01	8.6×10^{16}
CRAC 38	30	49.0	241.6	0.00368	0.102	6.80	2.8×10^{17}
CRAC 39	11	28.1	138.6	0.01383	0.031	22.4	8.8×10^{17}
CRAC 40	4.1	18.8	92.9	0.1494	0.018	38.5	1.2×10^{18}
CRAC 41	50	18.8	92.9	0.01746	0.183	3.79	5.8×10^{16}
CRAC 42	1.8	14.7	72.5	0.3062	0.0031	224	1.5×10^{19}
CRAC 43	3.2	14.6	72.0	0.3377	0.0050	139	5.6×10^{18}
CRAC 44	--	--	--	--	0.0014	495	3.1×10^{19}

Experiment Number	Time to End of Pulse[a] (sec)	Total Energy in Pulse (10^{17} fissions)	Specific Power at Pulse Peak (fissions/cm^3-sec)	Specific Energy in Pulse (10^{12} fissions/cm^3)	β_{eff} (pcm)	Reactivity Added Through First Pulse (pcm)
CRAC 37	80	3.4	3.7×10^{11}	1.5	711	630
CRAC 38	40	3.2	1.2×10^{12}	1.3	711	750
CRAC 39	12	1.8	6.4×10^{12}	1.3	747	900
CRAC 40	5	1.3	1.3×10^{13}	1.4	780	940
CRAC 41	51	0.80	6.2×10^{11}	0.86	780	735
CRAC 42	2.5	1.7	2.1×10^{14}	2.3	825	1090
CRAC 43	4	1.3	7.8×10^{13}	1.8	825	995
CRAC 44	--	1.9	--	--	--	--

a. Time measured from delayed criticality.

IV. CORRELATION OF RESULTS

The results of these experiments demonstrate several points of interest. The peak fission rate per unit volume (specific power) developed in the experiments with the 300-mm-diam cylinder is plotted versus the rate of ramp addition of reactivity in Fig. 14. Since CRAC 15, 16, 17, and 18 were started from delayed criticality, the pulse size achieved from their initial ramp rate was smaller than for the other experiments. CRAC 14, 20.1, 20.2, and 20.3 were started from below delayed criticality in the presence of an external neutron source. According to the standard neutron kinetics analysis of an assembly in the presence of a source,[1] the peak fission rate in the first pulse should vary as the ramp rate. In Fig. 14, a line of slope unity is drawn through the CRAC 14 data and passes fairly close to the CRAC 20.1, 20.2, and 20.3 cluster. Since CRAC 20.5 was performed within 30 min after CRAC 20.4, photoneutron production in the solution from residual gamma-ray activity apparently acted as a start-up source.

The remaining CRAC experiments were conducted without an external source and demonstrate the quantitative behavior expected for assembly in the presence of a "weak source."[2] At low ramp rates (< 0.05 dollar/sec) all pulses have peak yields which fall near the standard kinetics family; however, as the ramp rate increases above 0.05 dollar/sec, solutions assembled without an external source show an increasing tendency for significant delays in the initiation of the pulse and result in larger pulse peaks. As ramp rates approach 1 dollar/sec, pulses having peak yields $\simeq$ 100 times that of "standard kinetics" are observed (CRAC 08). CRAC 20.4, which was the same as CRAC 20.1, 20.2, and 20.3 except that no source was present, had a peak yield $\simeq$ 20 times larger than the pulses produced with a source present.

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1. G. R. Keepin, Physics of Nuclear Kinetics, Addison-Wesley, Reading, Mass., p. 313 (1965).
2. G. E. Hansen, Nucl. Sci. Eng. 3, 709 (1960).

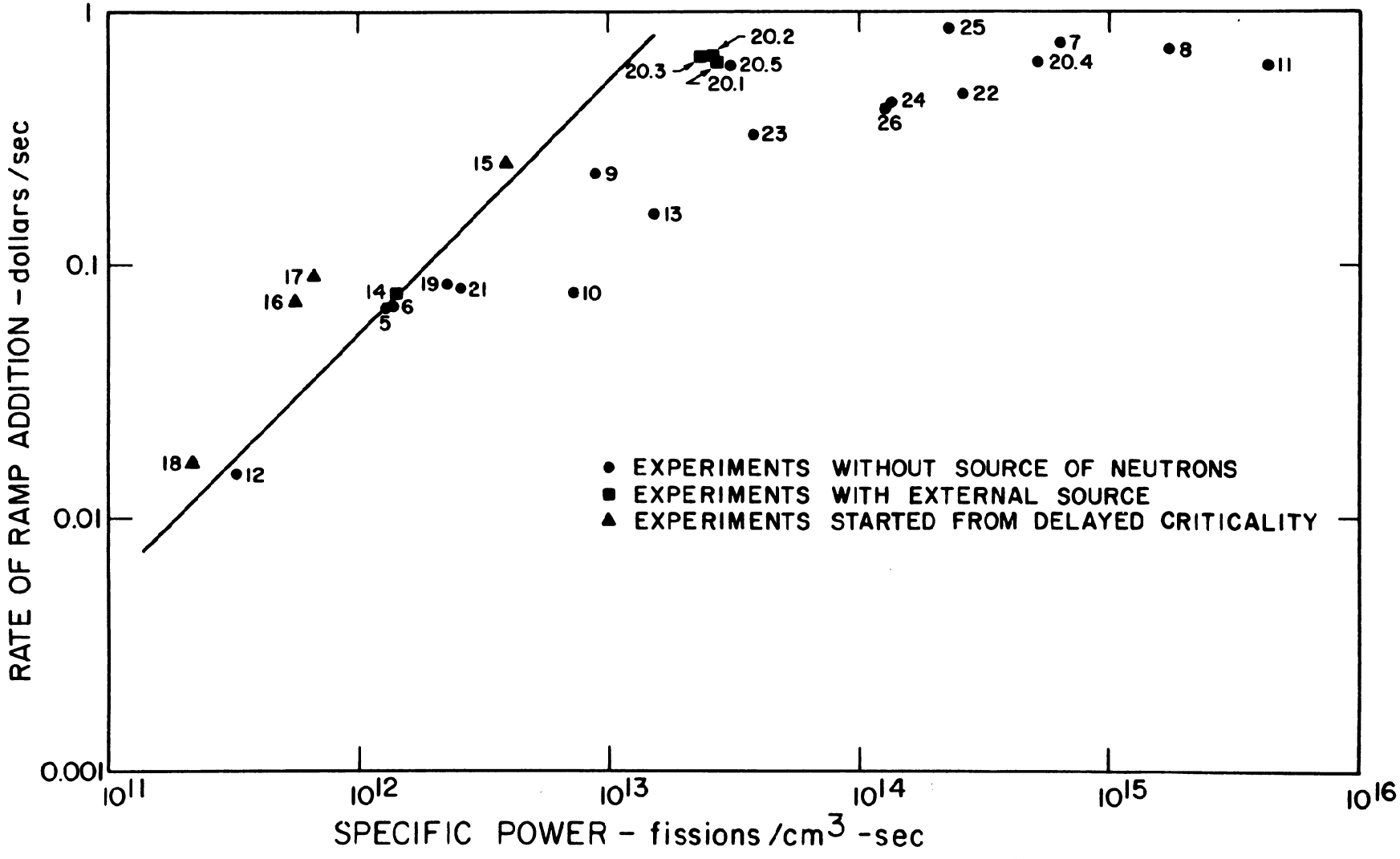

Fig. 14. Rate of Ramp Addition of Reactivity as a Function of Specific Peak Power for Experiments in the 300-mm-diam Cylinder. The numerics identify the experiments.

An attempt was made to relate the total fission yield during the chain of pulses of an experiment to the total reactivity available. In a typical experiment there was a chain of pulses of decreasing size until the fission rate damped to a more or less steady rate. The total number of fissions in the pulsing portion of an experiment, normalized to the total volume of the solution (and hence to its heat capacity), fit an expression of the form:

$$\frac{\text{Yield}}{\text{Total Volume}} \simeq K \rho_{\text{total}}$$

where

$$1 \times 10^{12} \leq K \leq 2 \times 10^{12} \text{ fissions/liter-pcm} .$$

The Fuchs-Nordheim model with simple temperature feedback predicts that for short reactor periods (or large pulses), peak pulse yield approaches proportionality to the square of the inverse minimum period. During the KEWB program,[3] an excursion model for aqueous-solution-fueled reactors based on the Fuchs-Nordheim model, but also containing a negative reactivity coefficient due to radiolytic gas formation (with the dissolved gas concentration proportional to energy and the nucleation rate proportional to power), was developed which predicted that peak power would be proportional to inverse minimum period to the 3/2 power for large pulses. For the CRAC experiments, specific peak power as a function of inverse period is plotted for the 300-mm- and 800-mm-diam cylinders in Figs. 15 and 16. For reciprocal periods in excess of 100 sec^{-1}, the line has a slope of 3/2. The agreement is remarkable considering the range of pulse sizes covered and the extreme simplicity of the model.

During the course of the CRAC experiments, gamma-ray dose rates were measured with dosimeters located 3 or 4 m from the solution container. The detected dose at 4 m was $(1.8 \pm 0.4) \times 10^{-15}$ R/fission for the experiments performed in the 300-mm-diam vessel. The integrated yield in a pulse is relatively insensitive to the peak fission rate

3. M. Dunenfeld, Kinetic Experiments on Water Boilers - "A" Core Report - Part II, Analysis of Results, NAA-SR-5416, North American Aviation (1962).

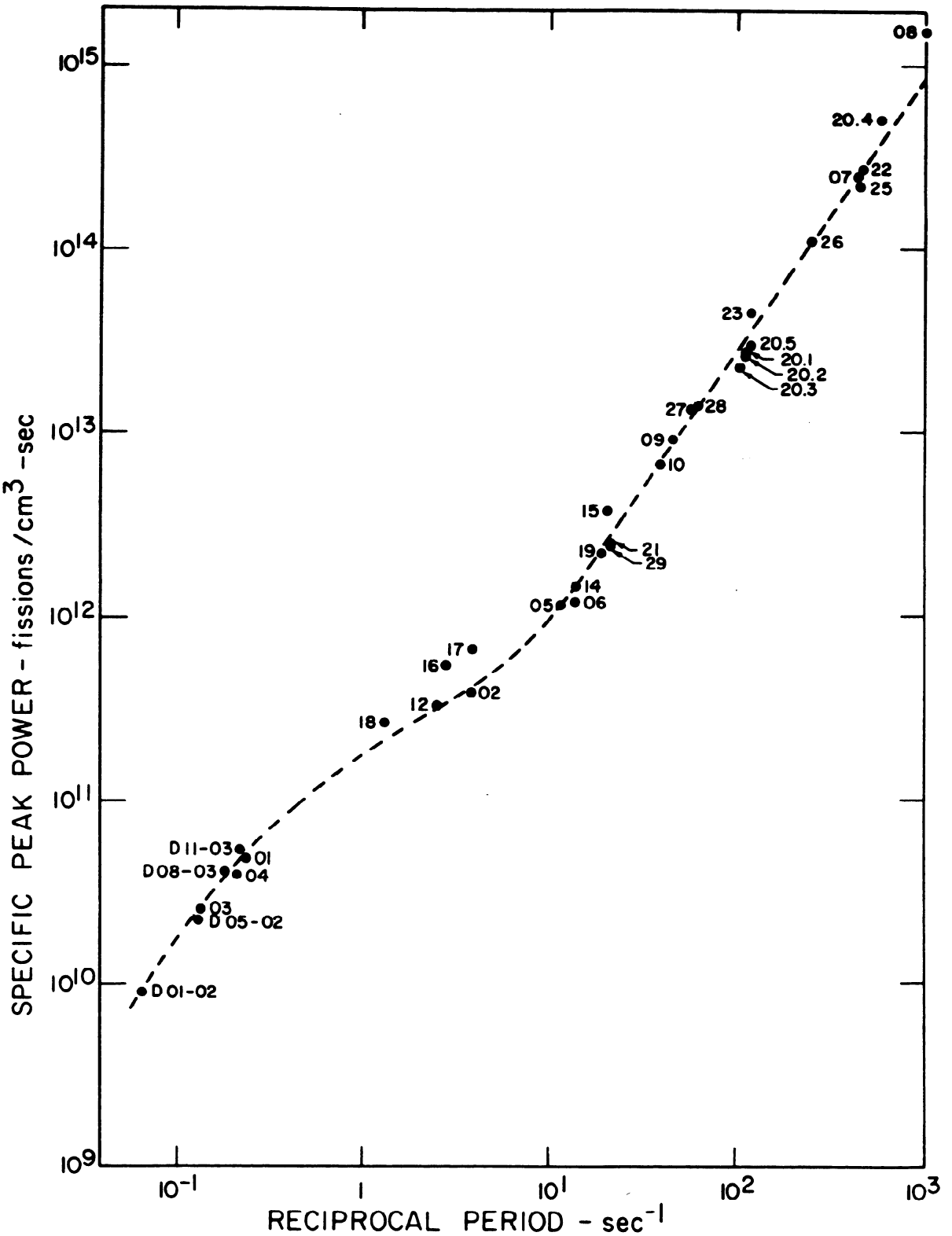

Fig. 15. Specific Power at the Peak of the First Pulse as a Function of the Reciprocal Period for Experiments in the 300-mm-diam Cylinder. The numerics identify the experiments.

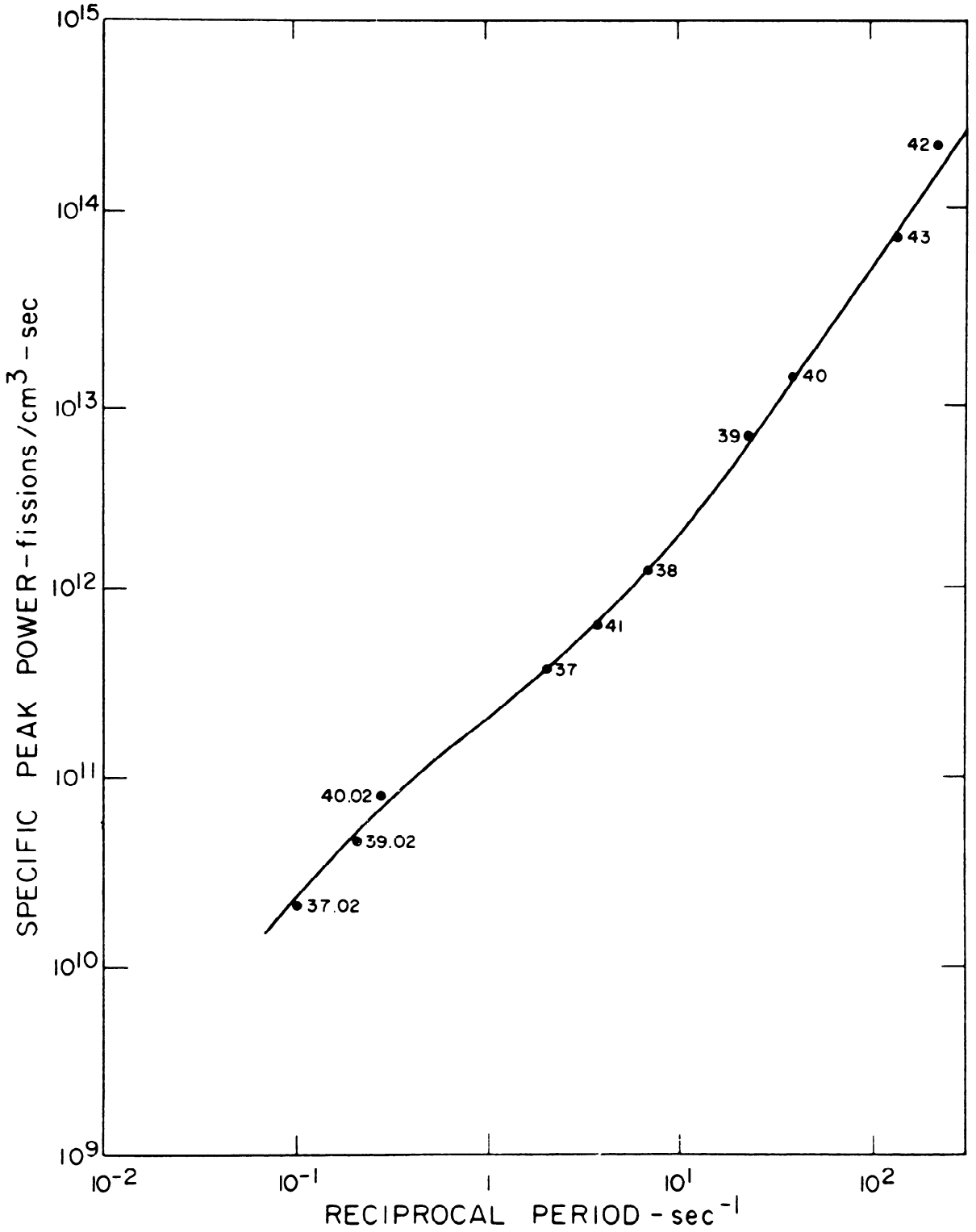

Fig. 16. Specific Power at the Peak of the First Pulse as a Function of the Reciprocal Period for Experiments in the 800-mm-diam Cylinder. The numerics identify the experiments.

since the pulse tends to broaden as the peak rate decreases. Thus, the integrated dose during the first pulse at a point 4 m from the assembly generally ranged between 40 and 550 R, a factor of ~14. The greater integrated dose occurred for the relatively broad low-peak pulses (CRAC 01 or 02) rather than the high, narrow pulses (CRAC 08 or 20.4).

In the 800-mm-diam vessel, the dose rate at 4 m was $\sim 5 \times 10^{-16}$ R/fission. The relation between the specific peak power in the first pulse and the reactivity introduced up to the time of the pulse is shown in Figs. 17 and 18 for the 300-mm- and 800-mm-diam cylinders, respectively.

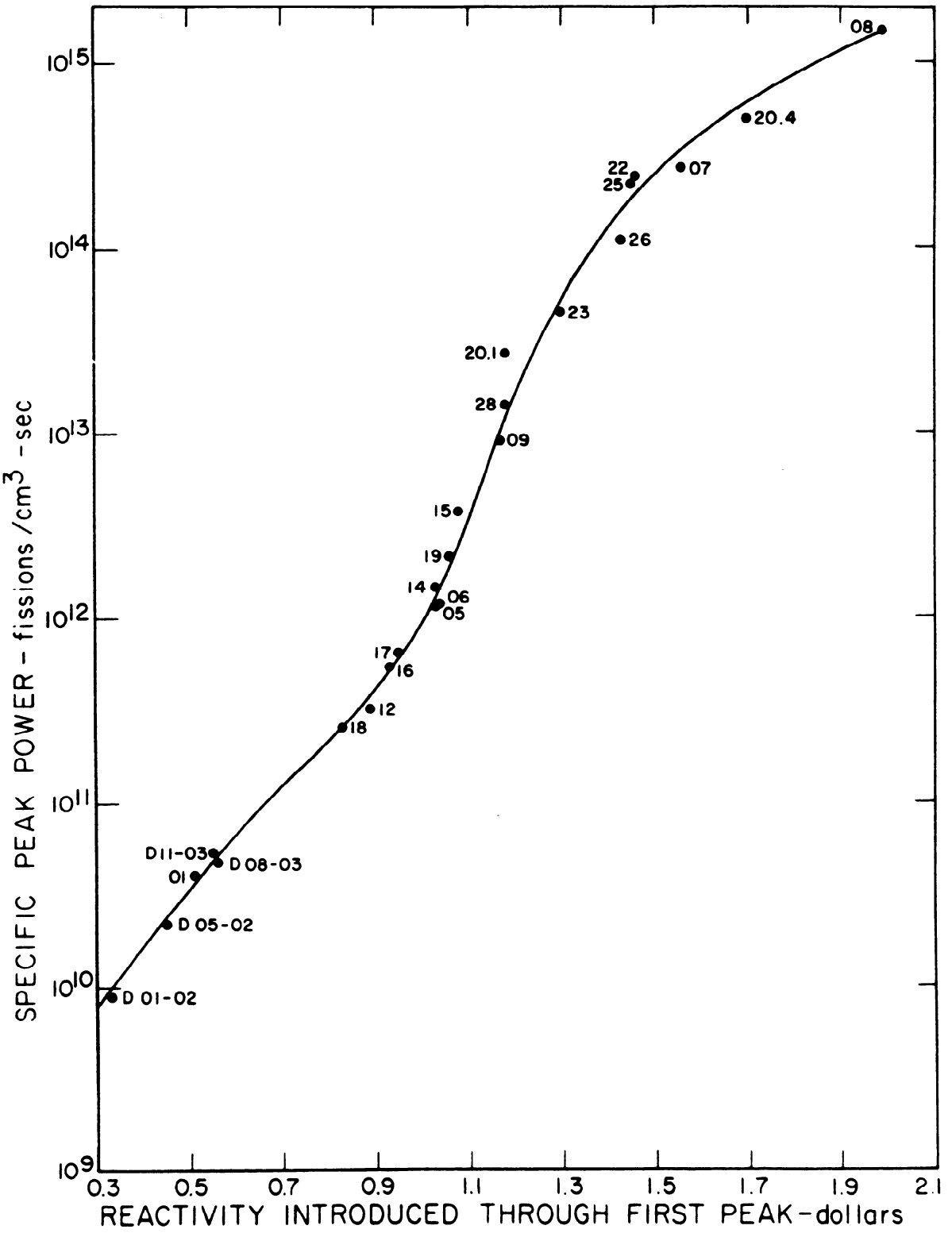

Fig. 17. Specific Peak Power as a Function of Reactivity Introduced Through the Peak of the First Pulse of Experiments in the 300-mm-diam Cylinder. The numerics identify the experiments.

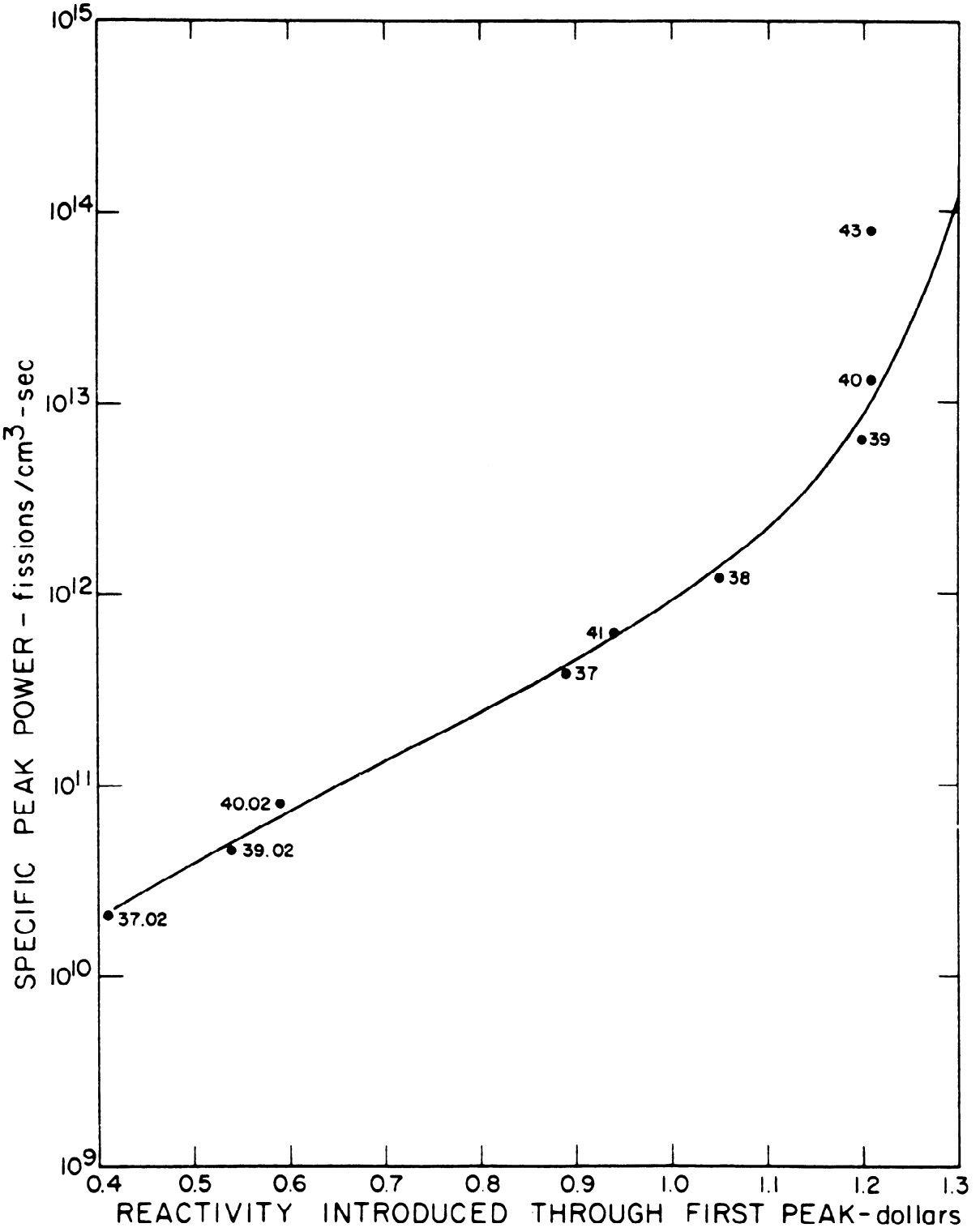

Fig. 18. Specific Peak Power as a Function of Reactivity Introduced Through the Peak of the First Pulse of Experiments in the 800-mm-diam Cylinder. The numerics identify the experiments.

V. CONCLUSIONS

The CRAC experiments provide a wealth of information on the behavior of solution reactors and an insight to the general criticality characteristics of these solution cylinders over a wide range of dimensions. The critical heights in the experiments with the 300-mm-diam vessel ranged from ~260 to 2000 mm. The geometry of the solution in the 800-mm-diam vessel was rather squat cylinders with critical heights ranging from ~140 to ~450 mm. This range of configurations can be readily extrapolated to fuel processing operations.

As far as the pulse experiments are concerned, most of the analyses thus far have been confined to the characteristics of the first of what may be a chain of pulses. Evidence indicates that the characteristics of the first peak are in agreement with a reactivity shutdown model which includes a negative temperature-dependent feedback and the effect of radiolytic gas evolution as shutdown mechanisms. The model that was developed for the KEWB reactor at least qualitatively fits the results obtained in these experiments. Some further analysis appears desirable with a more detailed evaluation of the entire chain of pulses, including the effect of the gradually increasing solution temperature. Indeed, for many of the CRAC experiments, the magnitude of the entire energy release was such that a significant portion of it was converted into kinetic energy by the expansion of the solution, thereby inducing the shutdown. This phenomenon should be investigated in more detail.

In addition to these results, there is practical information which should be of value in establishing the procedures and practices to be followed in fuel processing operations. Certainly the observed wide variation in peak fission yield in the instances where no external neutron source was available leads to the recommendation that neutron sources be used to provide "background neutrons" in various processing operations. The importance of this procedure is not so much to limit the gamma-ray dose received by personnel in the vicinity of the excursion, since the analysis of gamma-ray yields indicates that they are relatively insensitive to pulse size per se, but rather to limit the

size of the first peak in order to keep the resulting pressure pulses below the level that would result in significant equipment damage with consequent dispersal of solution.

ACKNOWLEDGEMENTS

The authors would like to thank all the people who took part in the program: R. A. Caizergues and L. M. Maubert of the Service d'Études de Criticité's staff at Saclay who made the theoretical studies prior to the experiments, the Section Expérimentale d'Études de Criticité at Valduc headed by H. F. Revol, who designed, constructed, and operated the experimental set, and particularly F. Y. Barbry, who was in charge of the measurements and put the results in usable form.

One of the authors (RLS) performed much of his portion of this review while serving as a consultant at the Los Alamos Scientific Laboratory during a part of the summers of 1970 and 1971. He would particularly like to express his appreciation to Dr. H. C. Paxton for calling his attention to this very interesting series of experiments and for his interest and encouragement during the subsequent analysis. Of course, the spirit of cooperation and confidence in which he was accepted by M. Lécorché and his associates as a collaborator in the preparation of this review is greatly appreciated.

APPENDIX A

CRAC PROJECT REPORTS

Report		Experiment	Date
C.E.A., D.P.S.R., S.E.E.C.*	No. 57	CRAC 01	December 1968
	59	CRAC 02	January 1969
	60	CRAC 03, 04	March 1969
	64	CRAC 05, 06	April 1969
	68	CRAC 07, 08	May 1969
	69	CRAC 09, 10, 11	August 1969
	70	CRAC 12, 13, 14	October 1969
	74	CRAC 15, 16, 17	January 1970
	77	CRAC 18, 19	February 1970
	79	CRAC 20.1, 20.2, 20.3, 20.4, 20.5	March 1970
	83	CRAC 21, 22, 23	May 1970
	91	CRAC 24, 25, 26	July 1970
	92	CRAC 27, 28, 29	August 1970
	96	CRAC 37, 38	December 1970
	98	CRAC 39, 40, 41	March 1971
C.E.A., D.S.N., S.E.E.C.**	103	CRAC 42, 43, 44	August 1971

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* C.E.A., D.P.S.R., S.E.E.C. -- Commissariat à l'Énergie Atomique, Direction de la Protection et de la Sûreté Radiologiques, Section Expérimentale d'Études de Criticité

** C.E.A., D.S.N., S.E.E.C. -- Commissariat à l'Énergie Atomique, Département de Sûreté Nucléaire, Section Expérimentale d'Études de Criticité