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CRITICAL MASS STUDIES, PART VII

AQUEOUS URANIUM SLURRIES

D. F. Cronin
Dixon Callihan

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CRITICAL MASS STUDIES, Part VII

AQUEOUS URANIUM SLURRIES

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ABSTRACT

A series of experiments has been performed with aqueous slurries of uranium tri-oxide, enriched to 93% in U-235, as a preliminary study of the nuclear stability of critical and near critical suspensions. The slurries were contained in a 12" diameter cylinder and were mixed by a centrally located variable speed stirrer in a manner giving small surface disturbances. Critical masses were measured at concentrations ranging from 40 to 300 gm U/L at several stirrer speeds.

The dependence of the critical parameters of well mixed slurries on concentration is essentially the same as found earlier with solutions. The critical masses of incompletely mixed slurries were measured to be 5% to 10% less than those of well mixed ones under otherwise similar conditions, an amount not strongly dependent on the chemical concentration. Neutron activity excursions of corresponding magnitude were observed in near critical quantities of the slurry following stopping the stirrer in a well mixed system and, in some instances, after starting agitation of one which had settled. Although the activity changes lagged the alterations of the stirrer speed, in most cases requiring a few tens of seconds to develop, the magnitudes of the excursions are sufficient to warrent consideration of this effect in the safety features of reactors designed to use slurry fuels. The critical mass of a uranyl fluoride solution in the same vessel increased monotonically as the stirrer speed was increased over the same range, the total change being about 3%, due in part at least, to some vortex formation at the higher speeds.

It is concluded that the activity excursions in the slurry result from changes in the uranium distribution more complex than those which occur in settling alone. An analysis of the distribution, relating it to the neutron activity, has not been made so the results of the experiments are not completely interpretable.

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I. INTRODUCTION

The chemical stability of the oxides of uranium has warranted their consideration as a reactor fuel although their insolubility, if used in an aqueous system, poses some potential problems of nuclear safety. Significant activity excursions may result from the uranium concentration gradients caused by the settling of the oxide particles in the aqueous carrier as the agitation of the suspension is changed or as the hydrodynamic pattern of a circulating slurry is altered. The magnitudes and the signs of the activity changes will depend upon a number of parameters including, in addition to the uranium distribution, the shape and dimensions of the containing vessel and the relative hydrogen to uranium content of the slurry. Settling of the particles in a cylindrical vessel will, for example, decrease the height to diameter ratio of the active material and will probably decrease the moderation. The settling also leaves a volume of water above the uranium column, now of reduced height, thereby providing at least a partial neutron reflector had none been formerly present. Although an analytical treatment of the critical behavior of a slurry as the particles settle had been made,[1] the complexity of the problem justified an exploratory experimental program. The results of some preliminary observations of the critical conditions in an aqueous suspension of enriched uranium oxide and a comparison of them with the behavior of a solution of uranyl fluoride when similarly agitated are reported here. The work was done in the ORNL Critical Experiments Laboratory. It is to be emphasized that these experiments were performed in a particular assembly and that the results should be extrapolated with caution.

II. APPARATUS

The critical conditions of the aqueous slurry were determined in equipment which could be remotely controlled and which consisted, essentially, of a loop in which the slurry could be kept mixed by circulation, a test vessel with stirrer, and appropriate safety and control devices.

During a particular experiment the slurry was continuously circulated by a pump through a spray cooler, a storage volume, and a flow control valve. The pump was driven by two motors with rotors canned in stainless steel which were balanced by a side stream of the suspension. This balancing stream was remotely controlled by a valve driven by one of a pair of selsyn transmitters upon signal from the other. A pair of connections between the circulating system and the test vessel were

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1. Kasten, Paul, "Prediction of Critical Behavior of a Slurry upon Settling", ORNL-CF-53-1-294, January 26, 1953.

[redacted]

fitted with air operated valves allowing remote control of flow of the slurry into and out of the reactor. An additional drain was provided through a normally open, spring operated plug type valve, 3" in diameter, which led into a reservoir also 3" in diameter and having a capacity of about two gallons. By this safety device a significant quantity of uranium could be rapidly removed from the reaction vessel in response to a signal originated either manually or by the radiation level near the assembly. The entire system except the reactor vessel was fabricated from stainless steel and is shown schematically in Fig. 1. Figure 2 is a photograph of the storage system and circulating pump showing the liquid by-pass for balancing the rotors.

The critical tests were made in an aluminum cylindrical vessel 12" in diameter and 24" high having a hemi-spherical bottom to minimize the deposit of solids. The two connections, one 3/8" in diameter for normal filling and emptying, and the other, the 3" bottom plug for emergency draining, are shown in Fig. 3, a photograph of the inverted cylinder. Vertical baffles, 1/2" wide, were fastened perpendicularly to the inside wall of the cylinder. A propeller, 3" in diameter and having three blades, was located 2" above the bottom of the vessel and was attached to a 3/4" solid aluminum shaft placed on the axis of the cylinder. The shaft was driven by a motor through a Graham variable speed transmission which could be operated remotely by a pair of selsyn transmitters permitting stirrer speed adjustments between zero and 1000 rpm. The stirrer and the baffles were designed to produce vertical circulation of the slurry with only slight surface disturbance or vortex formation. A small direct current generator, coupled directly to the propeller shaft, had an output voltage proportional to the speed and served as a tachometer.

The height of the liquid in the reactor was indicated by a device[2] based on the change in capacity of an immersed cylindrical condenser and, in later experiments, by direct measurement. These values were converted to volumes by an empirical calibration curve.

The 12" vessel was mounted in a tank about nine feet in diameter which was available and into which water could be pumped to serve as a neutron reflector on the bottom and sides of the uranium container. No reflector was provided above the slurry. A spring loaded sheet of cadmium could be injected between the slurry and the water reflector as a part of the safety system. Additional safety was provided by a

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2. Sidman, R. D., "A Liquid Level Indicator" Q-1331, ORNL Instrument Department Report X-11-C-2, Sept. 24, 1952. See also Instrument Research and Development Quarterly Progress Report for Period Ending October 20, 1952, ORNL-1436, pg. 5.

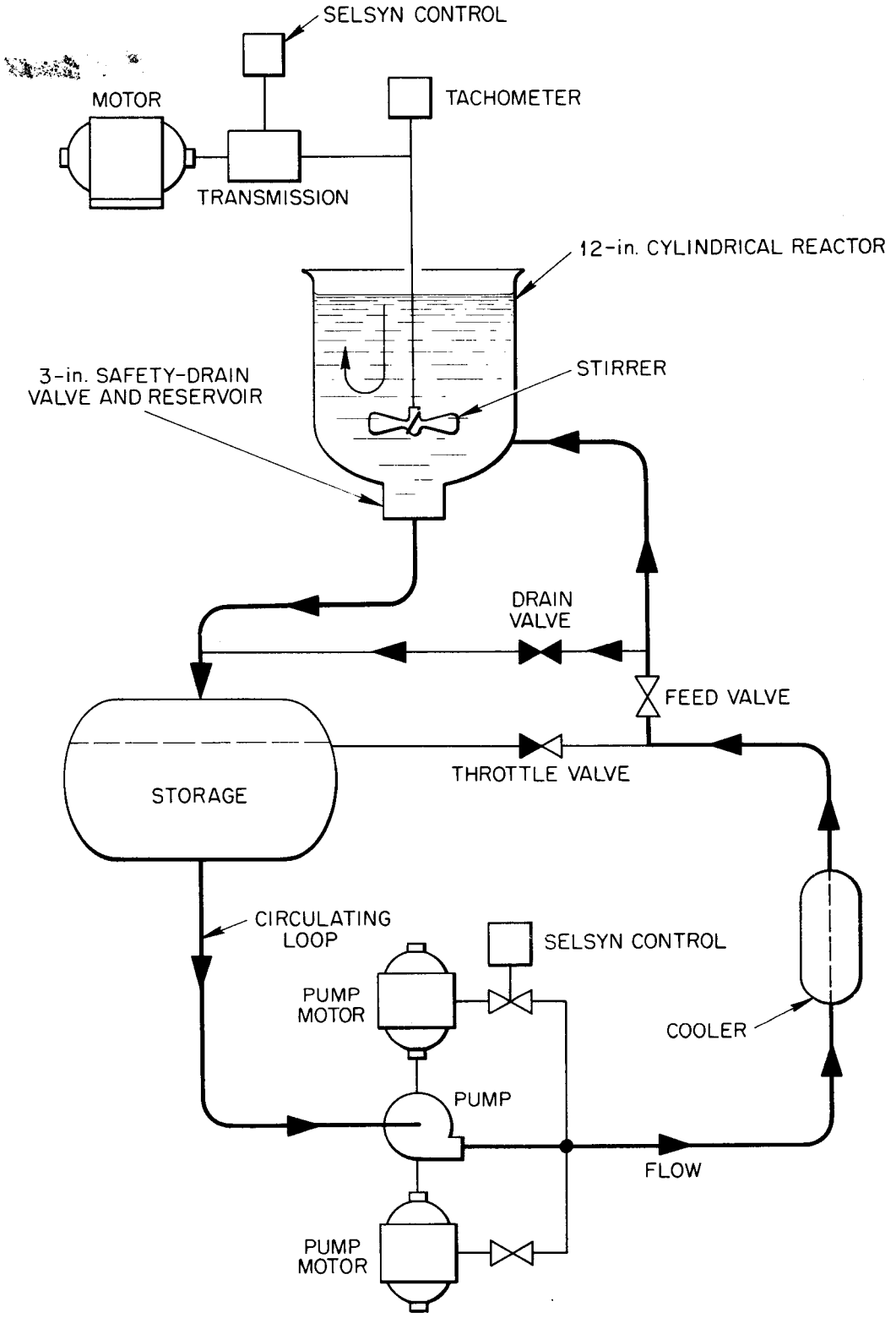

Fig. 1. Slurry Test System.

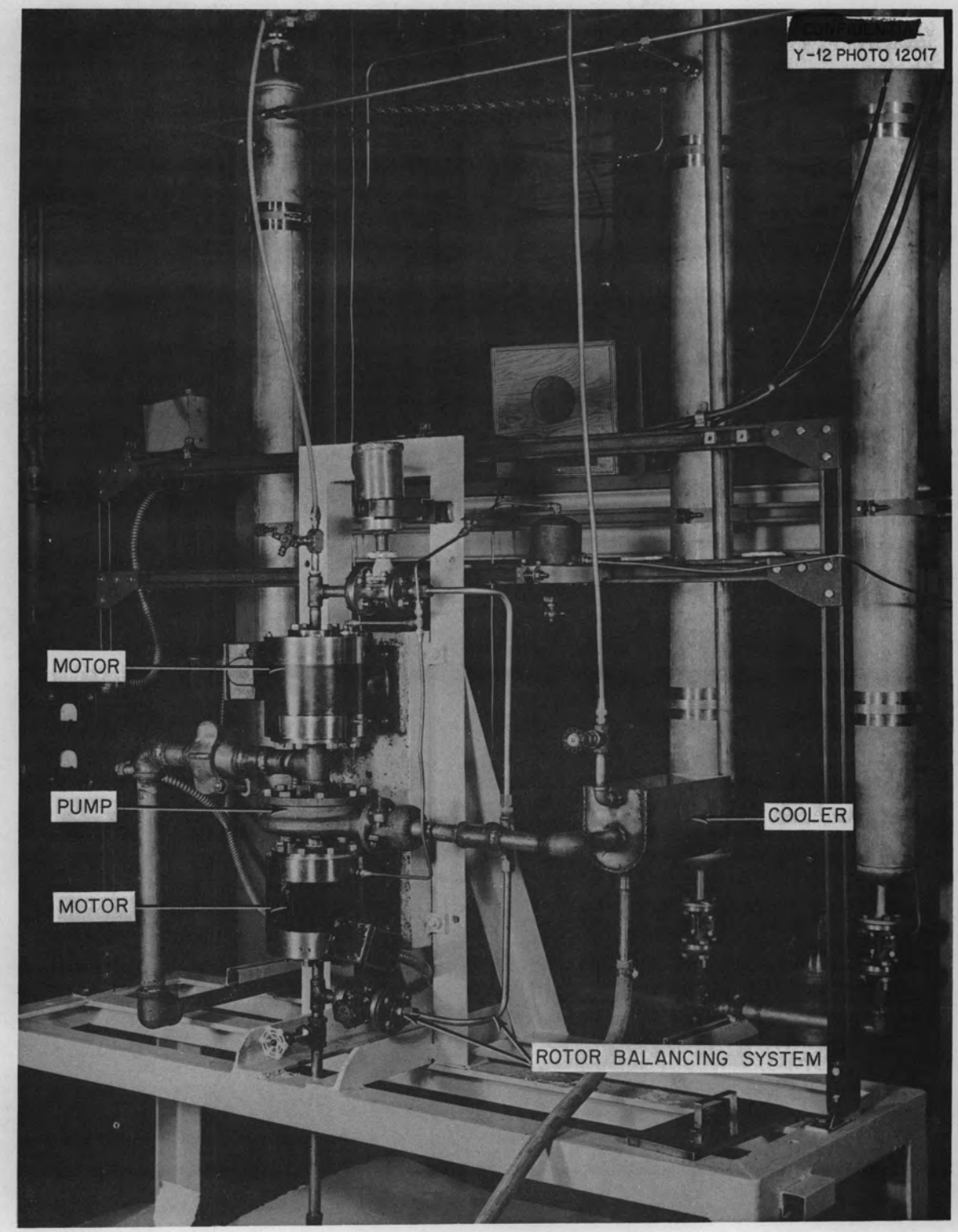

Fig. 2. Slurry Pump

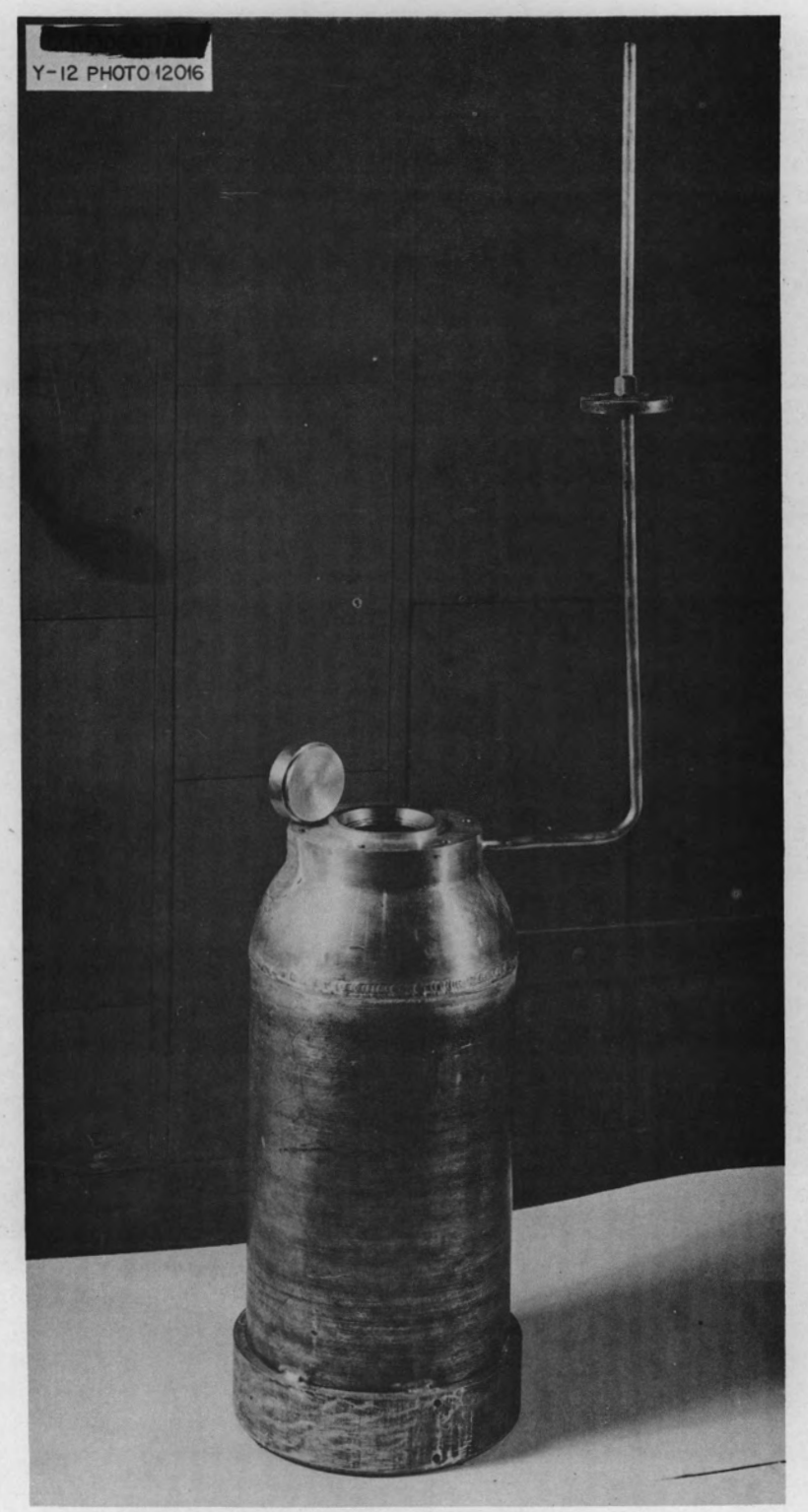

Fig. 3. Slurry Test Vessel

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normally open air operated 3" drain valve for the reflector water. A Po-Be neutron source of about 10^6 n/sec strength could be positioned in the water adjacent to the slurry and a 3/4" stainless steel tube, enclosing a cadmium sheet, could be adjusted in the slurry to effect small reactivity changes. Figure 4 is a view of the arrangement from above showing the 12" cylinder and auxiliary items. The long tubes at the left are thimbles, 6" in diameter, which contained the radiation detection instruments and were placed at least 6" from the edge of the reactor.

Conventional nuclear instrumentation was used and included two boron lined proportional counters with binary scalars, four BF_3 ion chambers which fed strip charts, and two gamma sensitive detectors. Four of these instruments were connected into the safety circuit which, at a pre-set radiation level, would drop the cadmium safety sheet, open both the slurry and reflector water drain valves, and stop the slurry and water pumps, all in about 100 milliseconds. Interlocks were provided whereby the safety system was actuated if the power supply to the stirrer failed. In some of the experiments an attempt was made to detect transients in reactivity by observing the output of gamma sensitive detectors on a Brush model BL-913 D. C. amplifier and a Brush model BL-202 oscillograph having a sensitivity of 1 mv/mm and a chart speed of 5 mm/sec.

III. EXPERIMENTAL MATERIALS

Uranium tri-oxide mono-hydrate was prepared by the Y-12 Chemical Division as a powder having a crystal density of 6.0 gm/cm^3 and with particles 5 microns in diameter and 25 to 35 microns in length. An aqueous slurry of this material which contained uranium enriched to 93.2% in the U-235 isotope, was used as the reactor fuel in these experiments. The uranyl fluoride solution which was also prepared by the Y-12 Chemical Division, contained an insignificant amount of HF and was enriched to 93.2% U-235. The densities, in grams of U-235 per liter of solution (or slurry), over a range of H:U-235 atomic ratios are shown in Fig. 5 to be practically equal. De-ionized water at 29 ± 1°C was used as the neutron reflector. Typical spectrochemical analyses of the materials are given in Table I.

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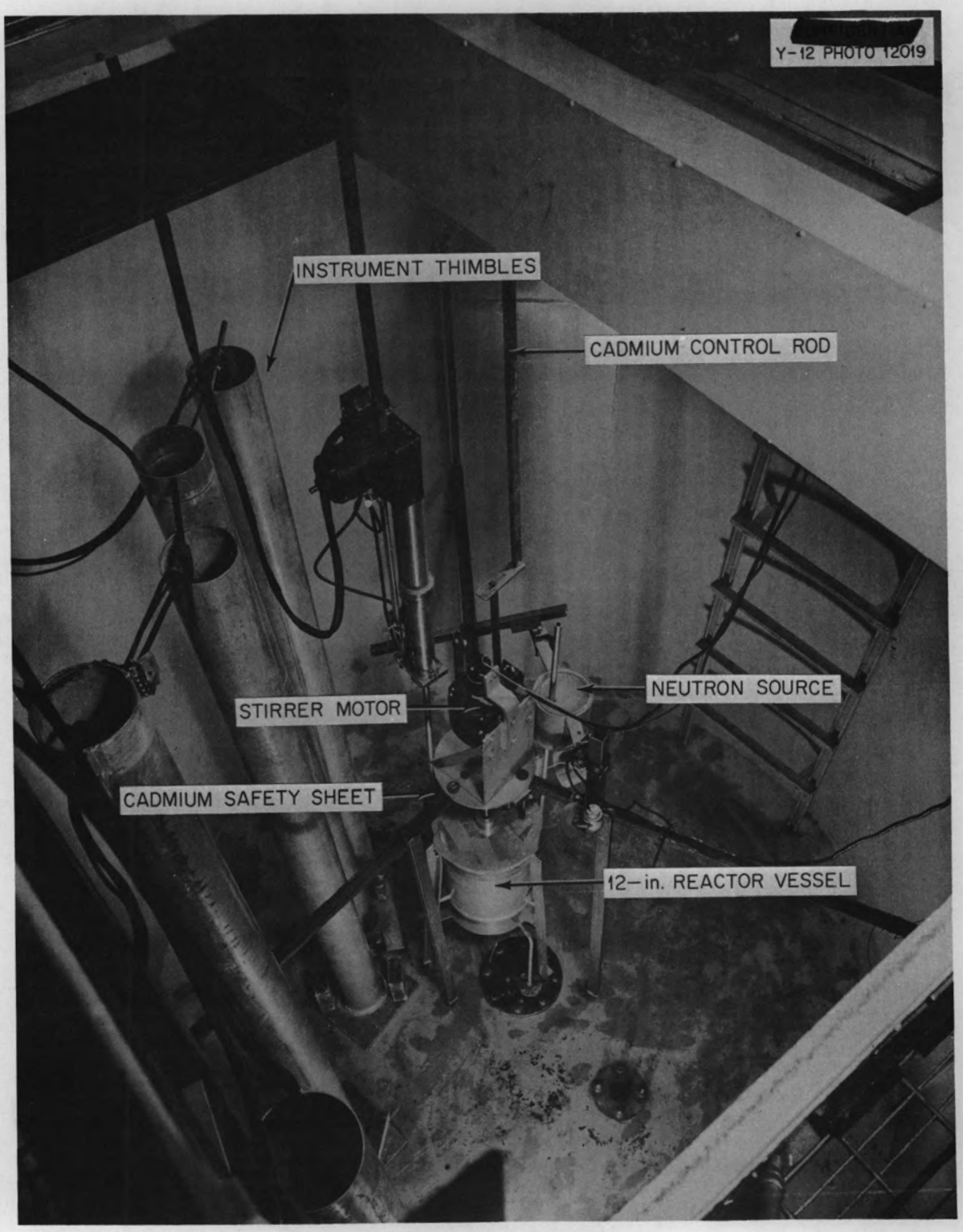

Fig. 4. Slurry Test System

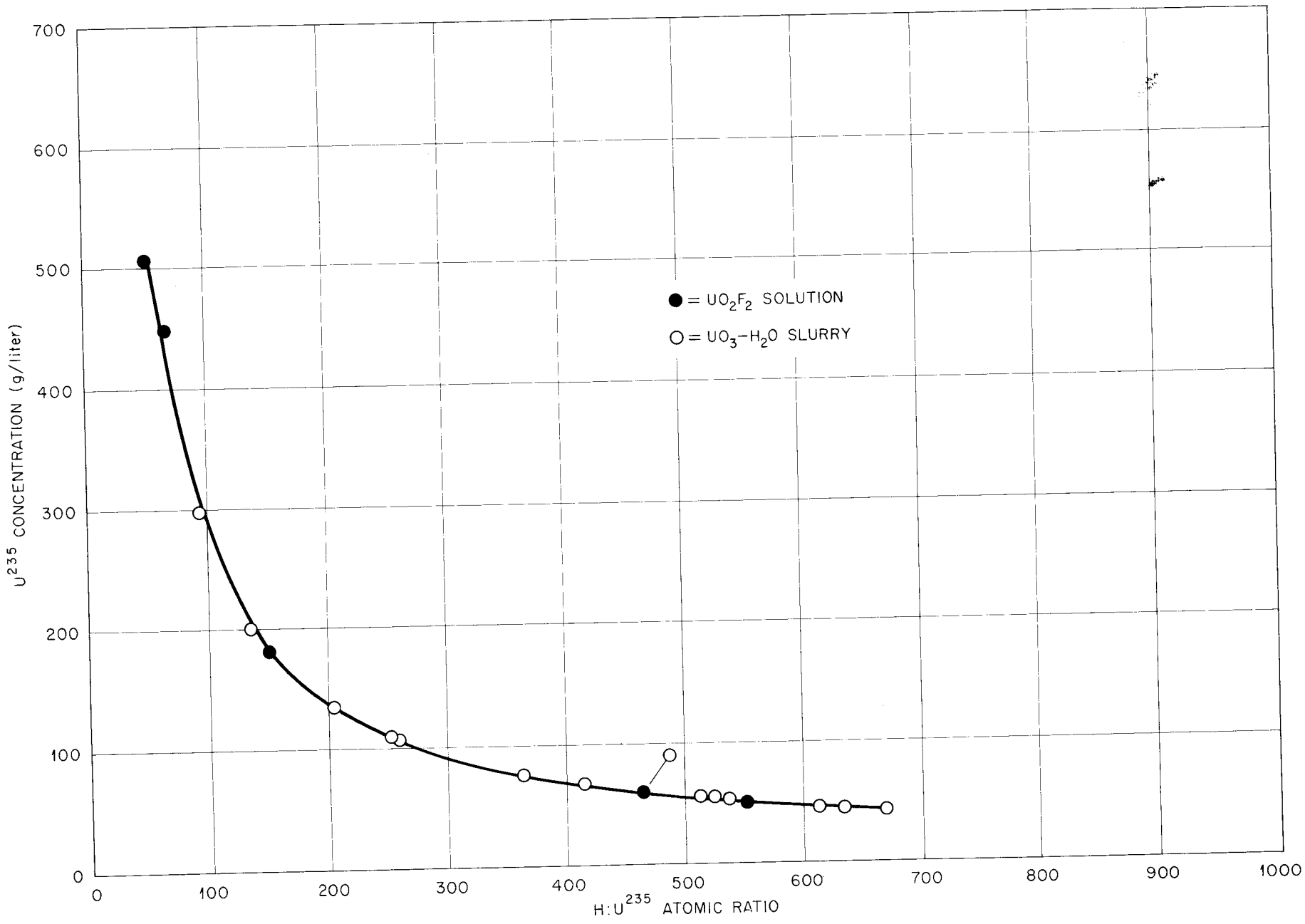

Fig. 5. Uranium Concentration vs. $H:U^{235}$ Atomic Ratio.

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TABLE I

Typical Spectrochemical Analyses of Experimental Materials

	UO_3	UO_2F_2	Reflector Water
Al	2 ppm	<1 ppm	0.06 ppm
As	<20	20	<0.05
Au	< 2	<4	<0.005
B	0.4	<0.1	0.006
Ba	<4	<4	<0.01
Be	<0.02	<0.02	<0.00005
Bi	<1	<1	<0.002
Ca	<10	<10	0.1
Cd	< 0.2	0.2	0.12
Co	<1	< 2	<0.002
Cr	<2	<4	0.01
Cu	10	10	0.12
Fe	18	12	0.2
Ga	<1	< 4	<0.002
Ge	70.4	<1	<0.001
Hg	<4	<4	<0.01
In	<2		0.01
Li	<0.2	< 0.2	<0.0005
Mg	<2	<2	0.01
Mn	<1	<1	<0.01
Mo	20	<2	0.015
Na	1	4	0.01
Ni	8	< 1	0.01
P	<100	<50	<0.25
Pb	<1	30	2.5
Pd	<1	<2	—
Sb	<4	<4	<0.01
Si	10	8	0.3
Sn	<1	<1	<0.002
Sr	<20	<20	<0.02
Ti	6	8	0.015
Tl	<4	<4	<0.01
V	<0.5	0.5	<0.001
Zn	<10	<10	0.1
Rare Earths		<14	

[redacted]

IV. EXPERIMENTAL RESULTS

Some preliminary qualitative investigations showed the occurrence of marked neutron activity excursions in near critical volumes of the UO_3 slurry as the agitation was changed. In general the activity increased, passed through a maximum, and decreased to essentially background as the agitation of an initially well mixed slurry was decreased either by interrupting the power supply to the stirrer or by reducing the rate of rotation step-wise. In some instances the activity peak was observed as the agitation was increased from zero stirrer speed. The behavior, which was independent of the slurry concentration over the range investigated*, is illustrated in Fig. 6 where relative activity is plotted against time. Initially the slurry was well mixed, then the power supply to the stirrer was first interrupted and subsequently restored. When the rate of rotation of the stirrer was changed step-wise, it was observed that the activity had a maximum at about 150 rpm and was essentially constant at speeds between 600 and 1000 rpm. It is to be noted that these measures of agitation refer to the vessel and stirrer used in these experiments and have relative value only.

The dependence of activity upon agitation was more quantitatively investigated by measuring the masses of U-235 in the UO_3-water slurries which were made critical at two stirrer speeds. One of these speeds was 200 ± 10 rpm, near the peak activity**, and the other was 600 ± 10 rpm, in the apparently stable well mixed region. Since no neutron reflection was provided at the top surface of the fuel an endeavor was made in all critical mass measurements to adjust the level of the lateral water reflector to that of the surface of the fuel. It was shown by experiment, however, that a water level 10 cm higher than the fuel surface introduced less than 2% error in the critical mass.

The results are given in Table II and in Figs. 7 and 8 where values of critical height and critical mass at both stirrer speeds are plotted as a function of the moderation. In all cases samples of the slurry were taken for chemical analyses directly from the reactor cylinder while the slurry was being well stirred and with the source and reflector water removed. The analytical results are, therefore, indicative of the average concentration. The method of sampling, however, may cause an error of as much as 10% in the analysis and hence in the absolute values of both the critical mass and the H:U-235 ratio. This error is not reflected in

---

* The concentrations studied varied between 40 gm U/L and 300 gm U/L. Lower concentrations, to be critical, required a volume greater than the capacity of the reactor vessel. Higher concentrations could not be satisfactorily mixed with the stirrer arrangement available.

**Unstable conditions at the speed of maximum activity precluded making mass measurements there.

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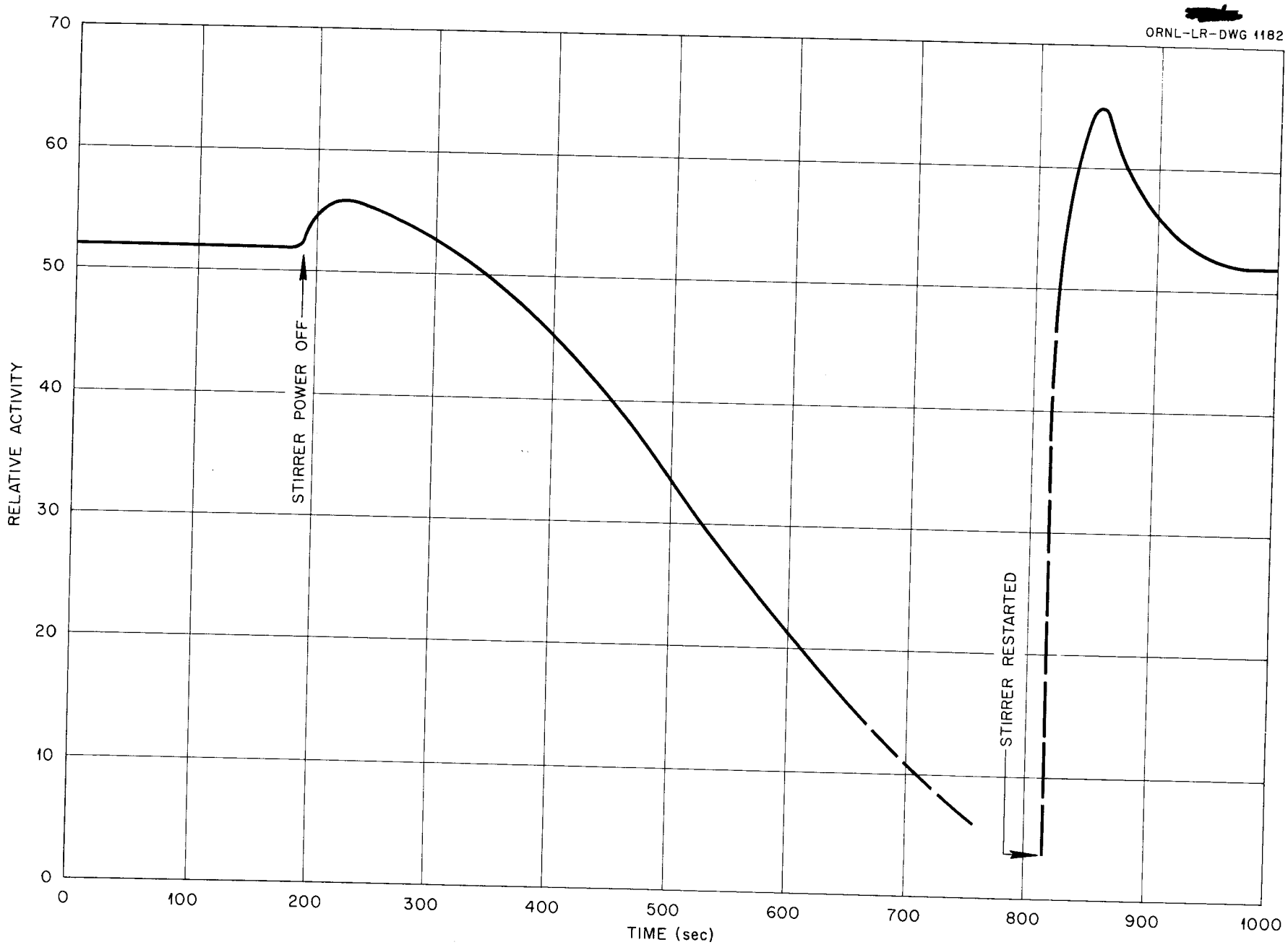

Fig. 6. Activity Changes in UO_3-H_2O Slurry: Concentration: ~40 g U/liter.

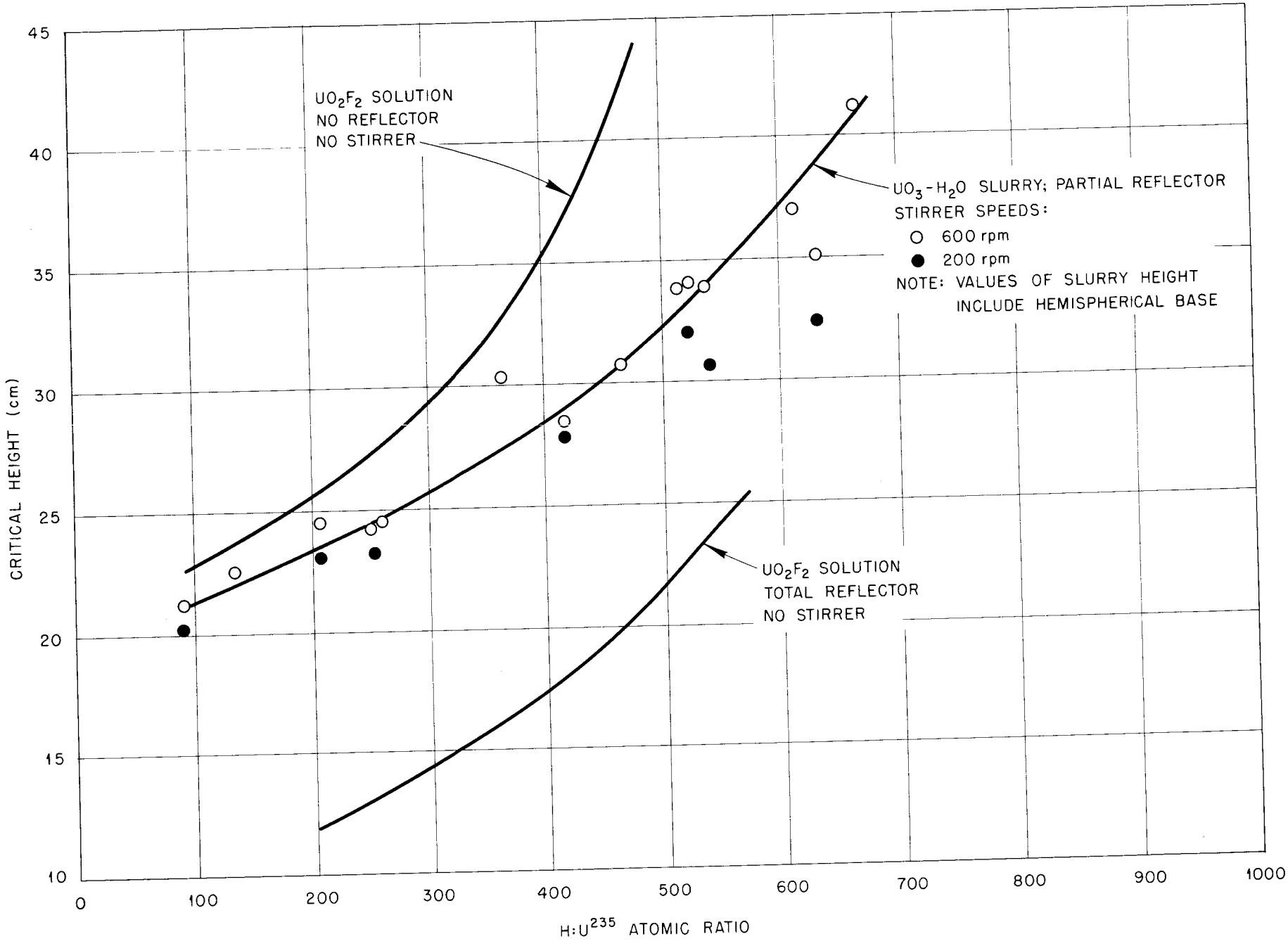

Fig. 7. Critical Height vs. Moderation; 12-in. Cylindrical Reactor.

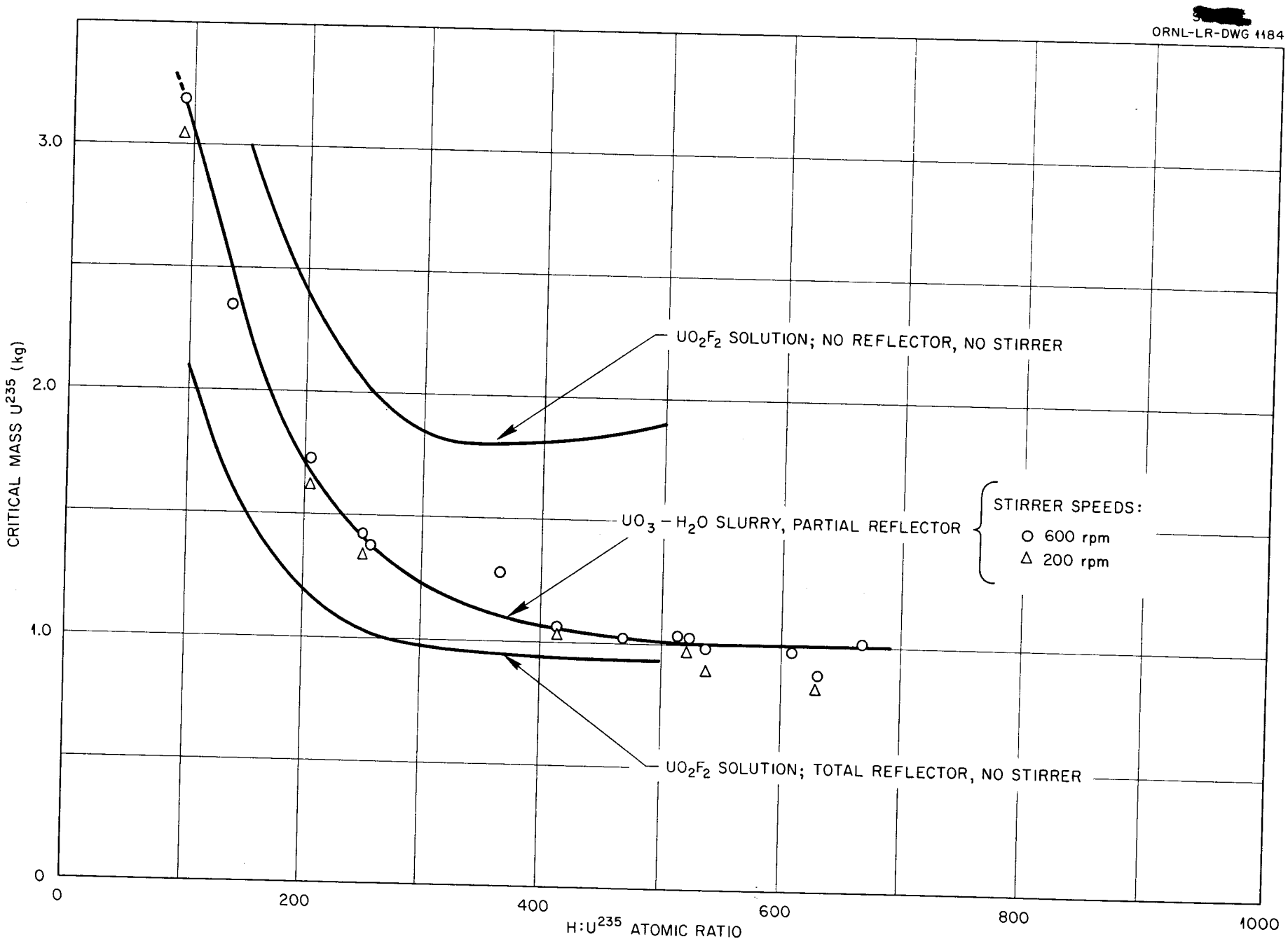

Fig. 8. Critical Mass vs. Moderation; 12-in. Cylindrical Reactor.

TABLE II

UO_3 - H_2O Slurry Critical Conditions in 12" Diameter Cylinder Reactor
Water Reflector on Sides and Bottom

			Stirrer Speed 600 rpm				Stirrer Speed 200 rpm			
H:U-235 Atomic Ratio	Slurry Concentration gm U / kg	Slurry Concentration gm U* / L	Reflector Height cm	Critical Height cm	Critical Volume L	Critical Mass U-235 kg	Reflector Height cm	Critical Height cm	Critical Volume L	Critical Mass U-235 kg
90.2	226.1	298.0	21.1	21.1	11.5	3.20	20.2	20.1	10.9	3.03
136	164.7	199.8	26.0	22.6	12.6	2.35				
204	117.7	134.6	27.6	24.3	13.8	1.73	27.6	23.1	12.9	1.62
251	98.50	110.0	24.1	24.1	13.7	1.40	23.2	23.2	13.1	1.34
259	95.75	106.6	28.0	24.3	13.8	1.37				
366	70.14	75.81	31.5	30.3	18.2	1.29				
413	62.78	67.28	28.5	28.5	16.8	1.05	27.8	27.8	16.3	1.02
467	55.90	59.45	33.0	30.8	18.5	1.02				
511	51.41	54.39	33.8	33.8	20.5	1.04				
523	50.36	53.22	34.0	34.0	20.8	1.03	34.0	32.0	19.3	0.96
538	49.02	51.73	33.3	33.8	20.5	0.99	33.3	30.7	18.5	0.89
610	43.51	45.63	38.0	37.0	22.9	0.97				
632	42.24	44.23	35.1	35.1	21.5	0.89	32.4	32.4	19.6	0.81
668	39.90	41.67	54.0	41.3	26.0	1.01				

* The laboratory analyses were reported as mass of uranium per unit mass of slurry sample. The specific gravities of the slurries were calculated from a value of the density of the UO_3- H_2O crystal assumed to be 6 gm/cc.

the relative values of the critical parameters measured at the two stirrer speeds with the slurry at the same concentration. For this reason, although there is some scatter in the data plotted in Figs. 7 and 8, the observation that the critical height and mass are 5% to 10% lower at the slower speed is believed to be significant. The error in the measurement of the slurry height is 1%. The critical conditions measured earlier[3] with UO_2F_2 solutions in 12" diameter cylinders, both reflected and unreflected, are included in the figures for comparison. The UO_2F_2 solutions, in the earlier experiments, were not stirred.

The effect of slurry agitation on near critical systems was studied further at several concentrations by noting the source neutron multiplication over a range of stirrer speeds. In these experiments, the apparent source neutron multiplication was measured when the activity had come to equilibrium at each stirrer speed thereby giving a qualitative measure of the activity*. One such set of results is shown in Fig. 9 for a slurry having a concentration of about 50 gm U/L in which the stirrer speed was increased from zero to 750 rpm and then decreased. The difference between the two curves is attributed to differences between the hydrodynamic patterns in the slurry resulting from speed changes of opposite sign rather than to lack of equilibrium. During these measurements the depth of the slurry in cylinder was 30.4 cm whereas 37.0 cm was required to be critical at a stirrer speed of 600 rpm. Similar data for other slurry concentrations initially more nearly critical are given in Figs. 10 and 11, where in the latter, two sets of measurements were made at heights somewhat differently removed from the critical height. All data are listed in Table III. These results indicate, at least qualitatively, that the variation in activity with stirrer speed is not strongly dependent on the slurry density over the range examined.

The time dependence of this change in activity, which occurred as the slurry agitation was altered, has been examined by first allowing a near critical system to equilibrate with the neutron source present, when well mixed at a stirrer speed of 600 rpm, and then turning off

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3. Beck, C. K., Dixon Callihan, J. W. Morfitt, R. L. Murray, "Critical Mass Studies, Part III," K-343, April 19, 1949.

* The counter-source-multiplying medium geometry, in these experiments, was not sufficiently good to determine the effective multiplication constant from neutron multiplication measurements.

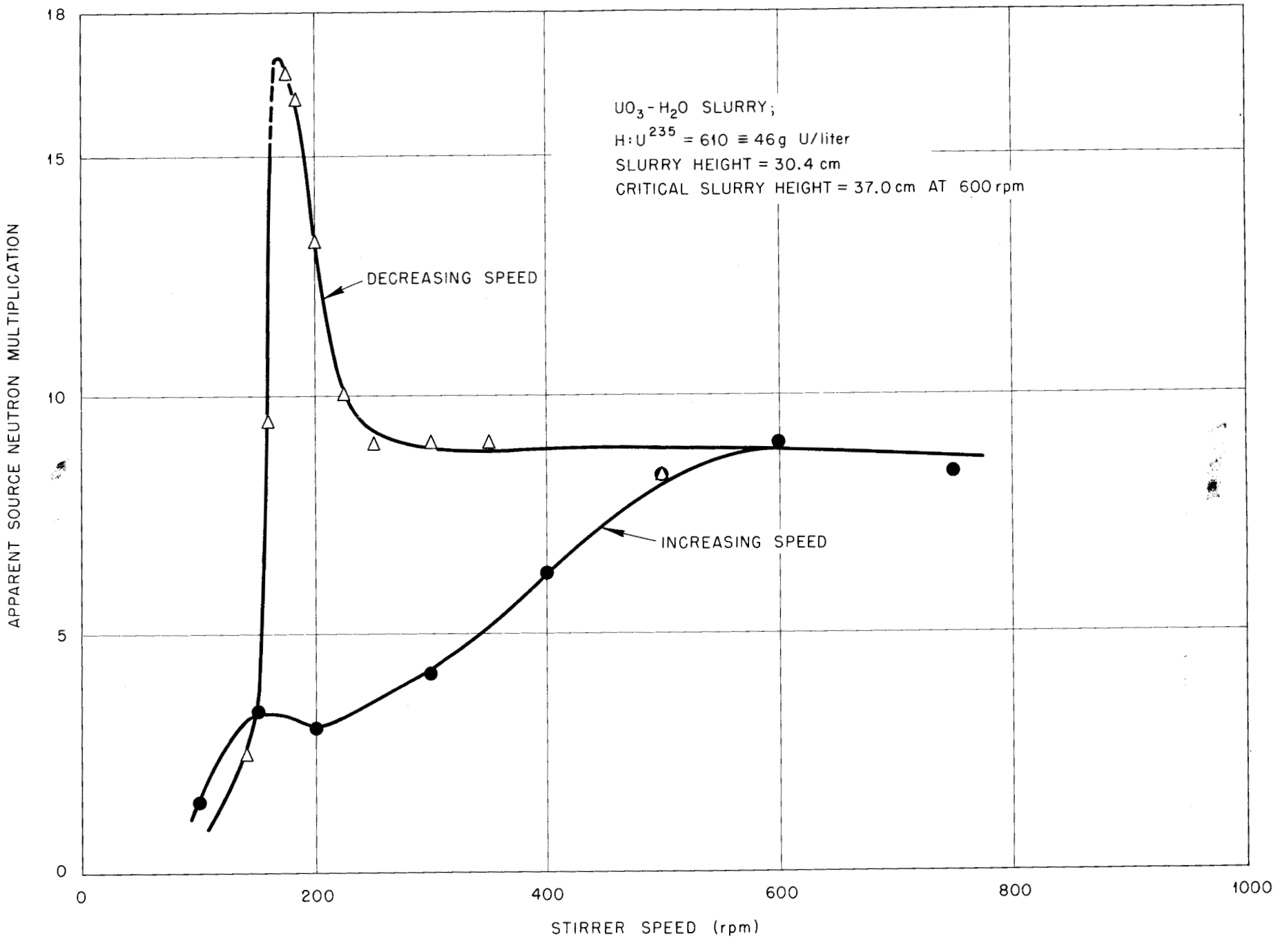

Fig. 9. Apparent Neutron Multiplication vs. Stirrer Speed.

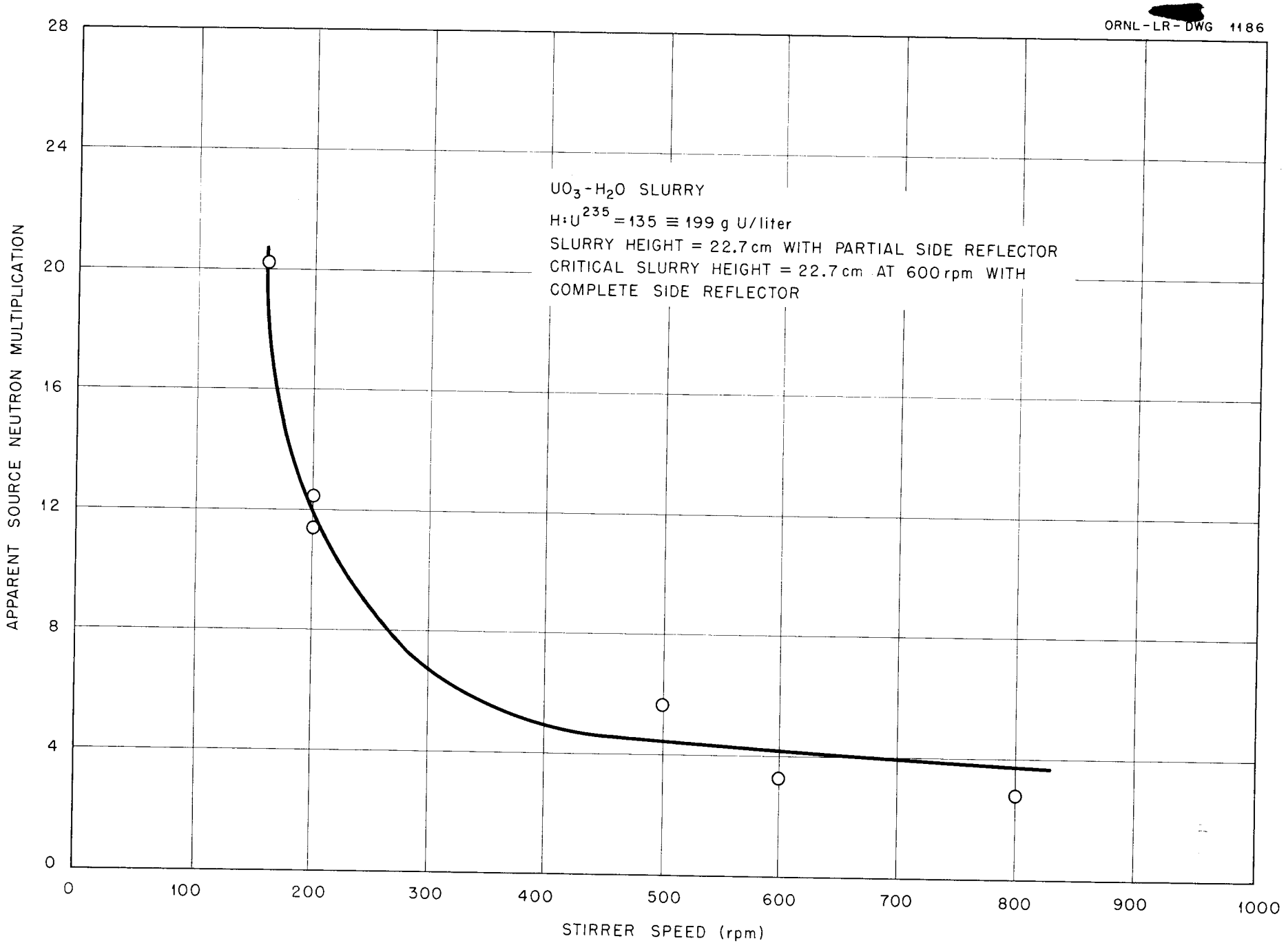

Fig. 10. Apparent Source Neutron Multiplication vs. Stirrer Speed.

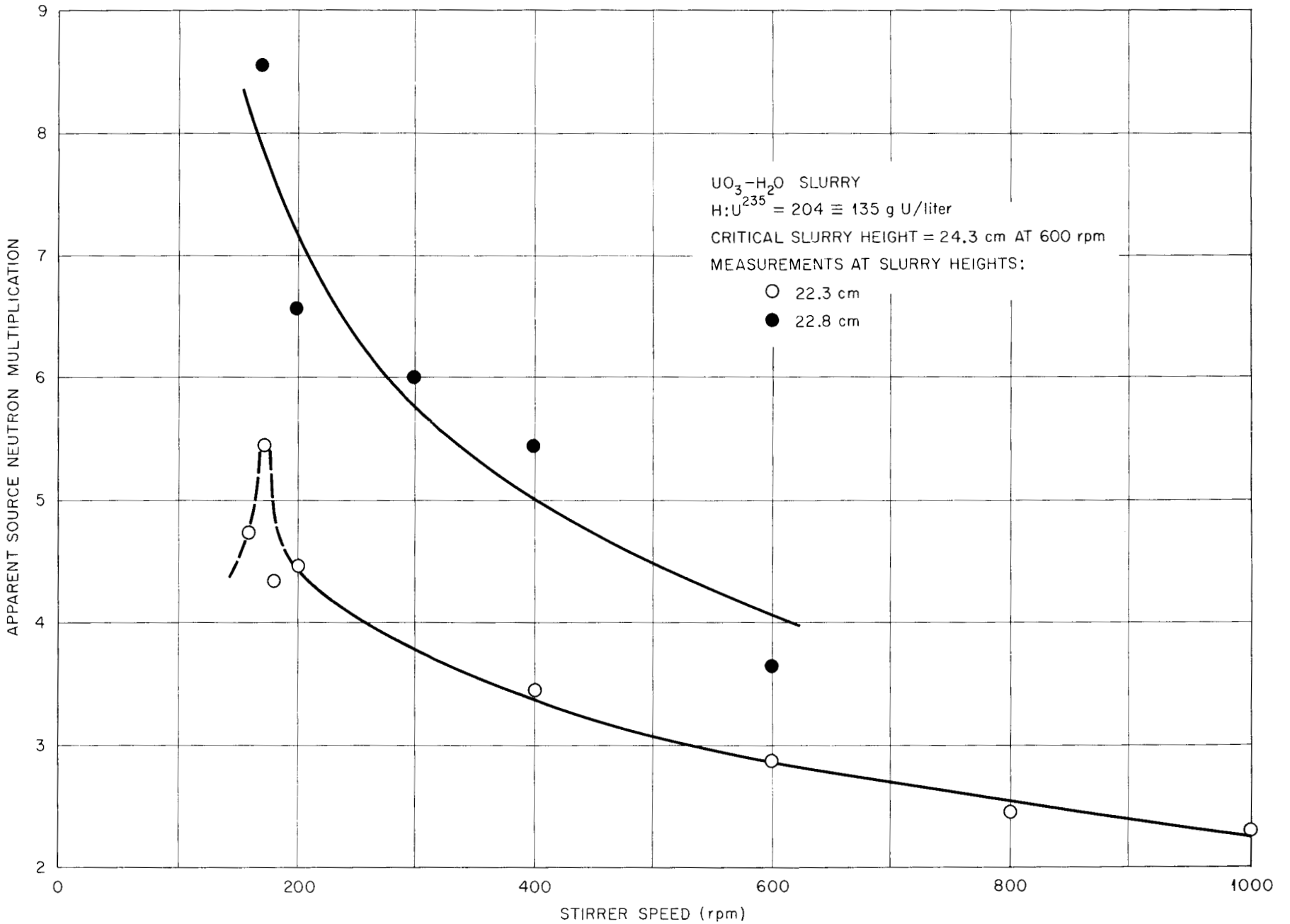

Fig. 11. Apparent Source Neutron Multiplication vs. Stirrer Speed.

TABLE III

Apparent Source Neutron Multiplication by Enriched Uranium Slurry
At Various Stirrer Speeds

H:U-235 = 610 Critical Slurry Height = 37.0 cm at 600 rpm		H:U-235 = 204 Critical Slurry Height = 24.3 cm at 600 rpm		H:U-235 = 135 Critical Slurry Height = 22.7 cm at 600 rpm	
Stirrer Speed rpm	Apparent Source Neutron Multiplication	Stirrer Speed rpm	Apparent Source Neutron Multiplication	Stirrer Speed rpm	Apparent Source Neutron Multiplication
Slurry Height = 30.4 cm		Slurry Height = 22.3 cm		Slurry Height = 22.7 cm With Partial Side Reflector	
100	1.47	1000	2.29	600	3.27
150	3.33	800	2.42	800	2.82
200	3.00	600	2.86	500	5.65
300	4.17	400	3.44	200	11.3
400	6.25	200	4.44	200	12.4
500	8.34	180	4.34	160	20.2
600	9.00	170	5.43		
750	8.34	160	4.75		
500	8.34				
350	9.00	Slurry Height = 22.8 cm			
300	9.00				
250	9.00	600	3.64		
225	10.0	400	5.44		
200	13.2	300	6.00		
185	16.4	200	6.58		
175	16.7	170	8.55		
160	9.45				
140	2.45				

the power supply to the stirrer. The activity recorded on a strip chart as a function of time, is reproduced in Fig. 12. As expected from earlier experiments, the activity reached a maximum in somewhat the same manner as with a stepwise reduction of the stirrer speed, and then decreased to background as the UO_3 settled further. When power was again applied to the stirrer, the activity returned to its initial level. In Fig. 13, also transcribed from a recorder chart, is plotted the relative activity as a function of the time during which changes were made in the stirrer speed. The system had initially an apparent source neutron multiplication of about three when stirred at 600 rpm. During the interval 400-600 sec the stirrer speed was slowly reduced to 400 rpm and the neutron level required 35 sec to equilibrate. At about 800 sec the speed was suddenly reduced to 200 rpm and the level allowed to become constant. The speed was subsequently decreased to 170 rpm and to 150 rpm and, in this range, the neutron activity passed through a maximum. Upon rapid restoration of the 600 rpm speed the activity returned to its initial value. The stirrer was subsequently stopped and in about 30 sec the activity passed through the familiar peak; upon restarting the stirrer at 600 rpm, the activity again returned to its original value.

An endeavor was made to observe activity excursions of short duration by simultaneously recording the stirrer speed and the signal from a γ-sensitive detector on a dual-pen Brush oscillograph having a chart speed of 5 mm/sec. Traces were made of signals originating in a BF_3 ion chamber-vibrating reed electrometer combination and of those from a scintillation detector. No significant differences were observed between these results and those described above indicating that no rapid transients were superimposed on the excursion. Three pairs of traces are reproduced as Fig. 14 showing the extent of the activity changes and the times at which they occurred relative to stirrer speed changes. The absolute magnitude of an excursion depends, of course, upon the extent to which the system is initially subcritical.

It is apparent from these results that although activity excursions of several percent in $\Delta k/k_{eff}$ occurred as the slurry agitation was changed, they tended to follow, in these instances, the motion of the slurry. After opening the power circuit to the stirrer, a few seconds were required for the propeller to stop rotating and an additional 25 to 60 seconds elapsed before the activity reached its peak. The interval between starting the stirrer and the activity peak was considerably shorter. Other results, although inconclusive, indicate that this latter interval depended upon the length of time the stirrer has been turned off, being longer for extended inactive periods than for shorter ones.

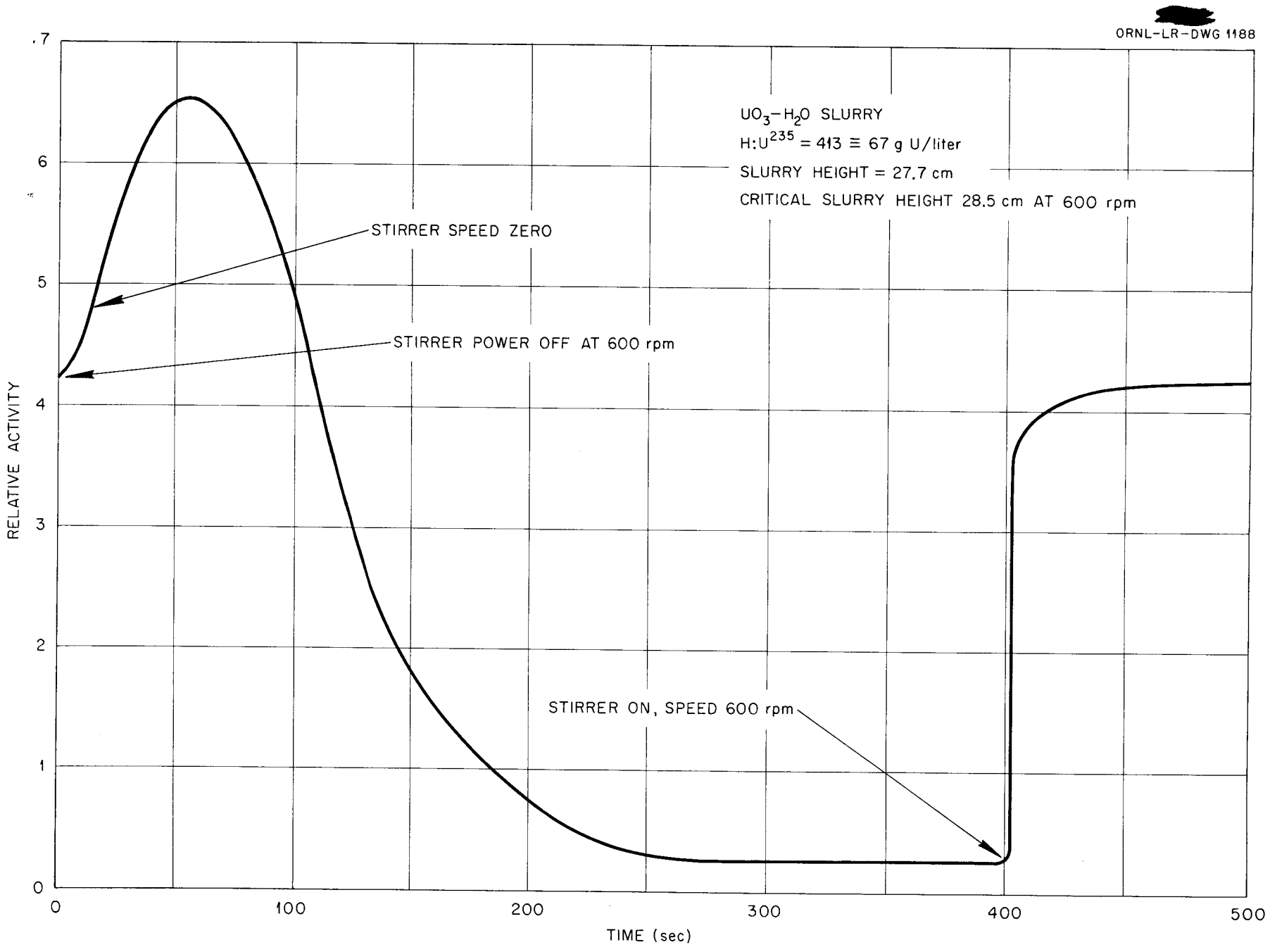

Fig. 12. Time Dependence of Reactivity Change.

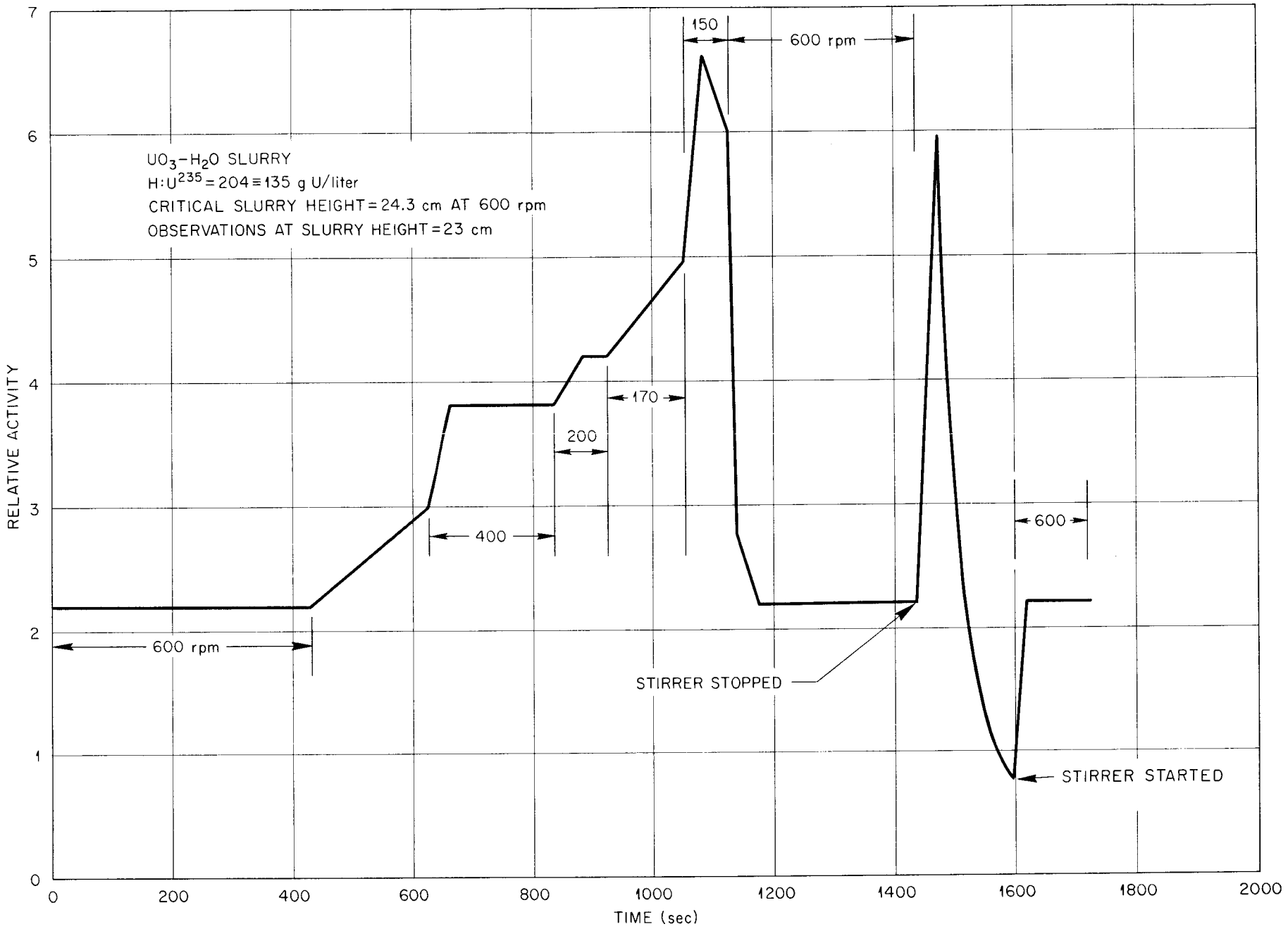

Fig. 13. Activity vs. Slurry Agitation.

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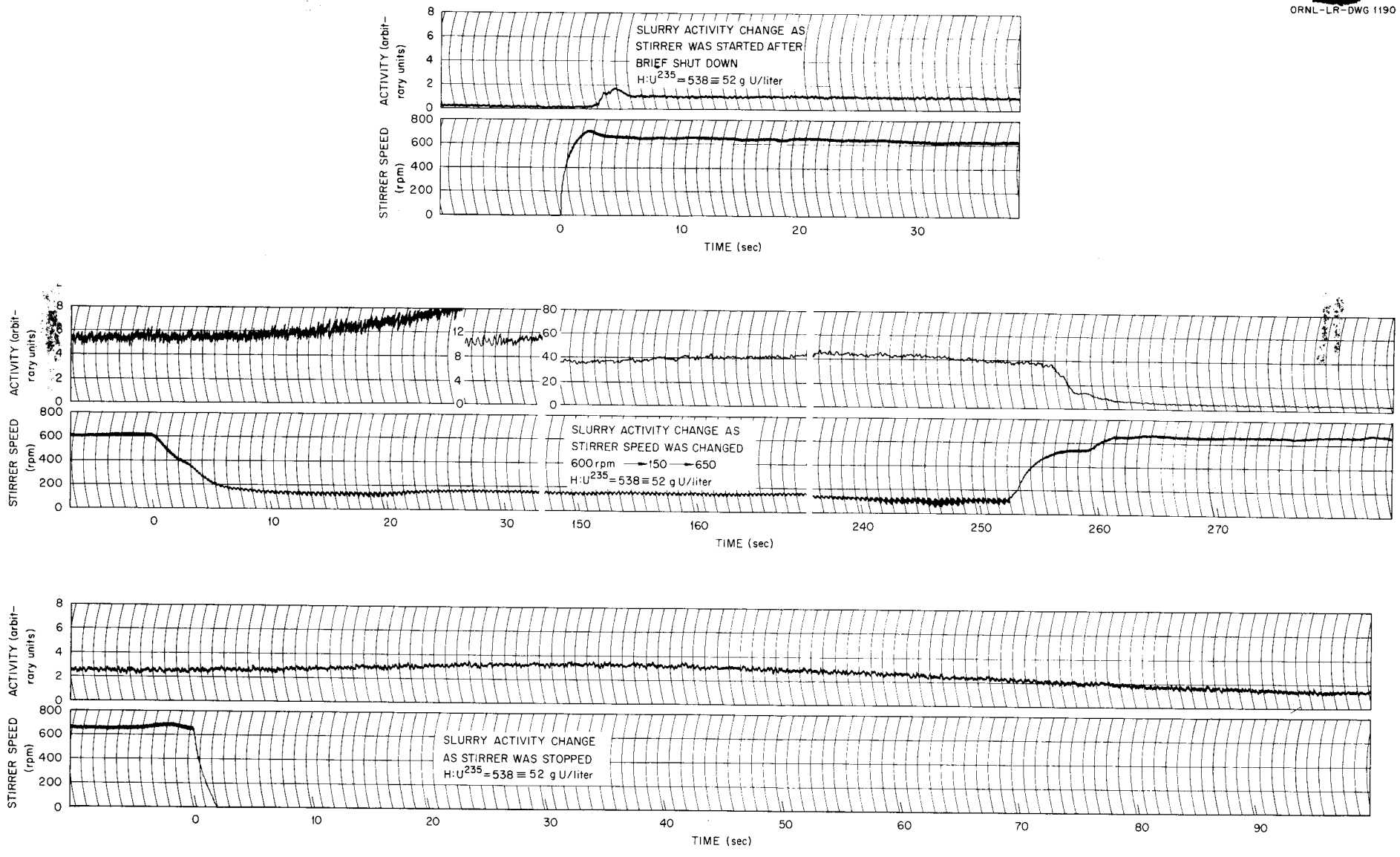

Fig. 14. Time Dependence of Activity Change.

A similar set of experiments was done using the same reactor vessel and associated equipment but with enriched uranyl fluoride solution as the fuel. In these experiments, the critical conditions were measured at several stirrer speeds and over a range of concentrations. The data are given in Tables IV and V and plotted in Figs. 15, 16, and 17. The first two graphs show the critical mass and critical height of the solution as a function of the chemical concentration with no stirring and with the stirrer at 600 rpm. The corresponding values measured with both a totally reflected and an unreflected cylinder without stirring reported earlier[3] and the slurry critical parameters are also plotted for comparison. Figure 17 shows the critical mass increased monotonically by about 3% as the stirrer speed was increased from zero to 600 rpm, a result due primarily to the formation of a vortex in the surface at the higher speed and possibly, to a lesser degree, to the distribution of the delayed neutron precursors within the solution.

V. DISCUSSION AND CONCLUSIONS

The following conclusions can be drawn from the results of these experiments:

1. The variation of critical parameters with neutron moderation for solutions and well mixed slurries of enriched uranium are about the same over the range studied.

2. Severe activity excursions, of the order of 10% in $\Delta k/k_{eff}$, were observed as the configuration of the mixed slurry was altered. The observations were not entirely reproducible.

3. In most instances these excursions developed slowly, requiring a few tens of seconds to reach their peak and seemed to accompany the changes in motion of the slurry. Increases in activity by a factor of e in the order of ten seconds, in the presence of a neutron source, were observed. The activity peak accompanying a start-up of the stirrer occurred within a shorter time than did those following the stopping of the stirrer.

4. Solutions of UO_2F_2, in the same apparatus with the same means of agitation, showed a small decrease in activity as the stirrer speed was increased and with no maximum observed at intermediate speeds.

The analysis, made[1] earlier of the critical behavior of a slurry, assumes the absence of a neutron reflector above the fuel as settling begins and

TABLE IV

UO_2F_2 Solution-Critical Conditions in 12" Diameter Cylinder Reactor (Hemi-spherical Base) Water Reflector on Sides and Bottom

Atomic Ratio H:U-235	CONCENTRATION gm U / gm	Sp. Gr.	gm U / L	STIRRER SPEED 600 RPM Reflector Height	CRITICAL Height	CRITICAL Volume	CRITICAL Mass U-235	STIRRER SPEED ZERO Reflector Height	CRITICAL Height	CRITICAL Volume	CRITICAL Mass U-235
50.4	0.322	1.57	506.2					24.7 cm	19.4 cm	10.3 L	4.86 kg
58.0	0.297	1.51	447.4	20.1 cm	20.1 cm	10.8 L	4.50 kg	19.6	19.6	10.5	4.38
150	0.150	1.20	180.0					21.2	21.2	11.6	1.94
463	0.0566	1.06	60.2	31.1	31.1	18.7	1.05	30.9	30.9	18.6	1.04
553	0.0475	1.05	49.8	35.7	35.7	22.0	1.02	35.6	35.6	21.9	1.02

TABLE V

Critical Height of UO_2F_2 Solution at Several Stirrer Speeds
H:U-235 = 58.0 = 447.4 gm U/L

Stirrer Speed rpm	Critical Height
0	19.6 cm
200	19.7
400	19.8
600	20.1

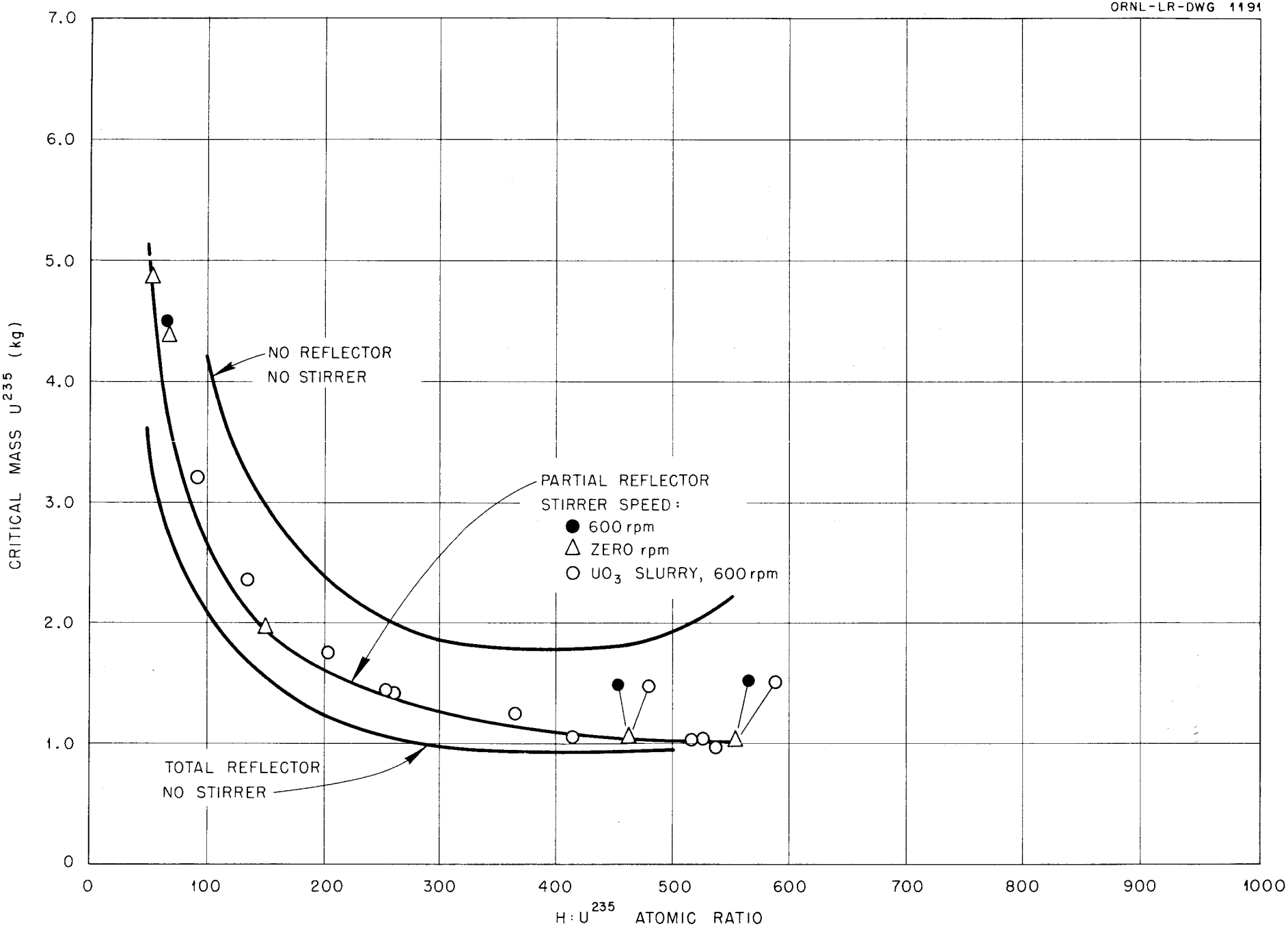

Fig. 15. UO_2F_2 Solution; Critical Mass vs. Moderation, 12-in. Cylindrical Reactor.

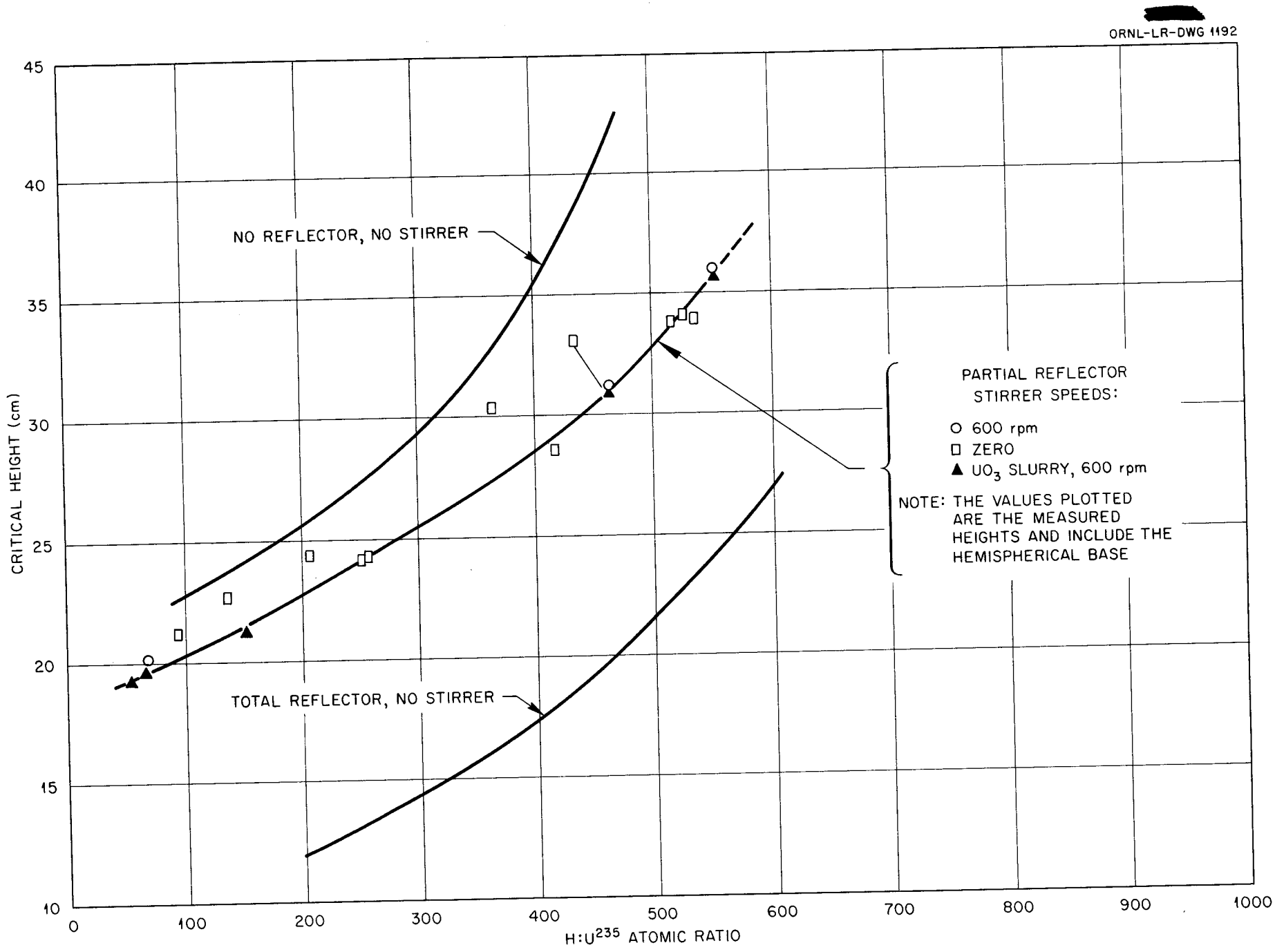

Fig. 16. UO_2F_2 Solution; Critical Height vs. Moderation; 12-in. Cylindrical Reactor.

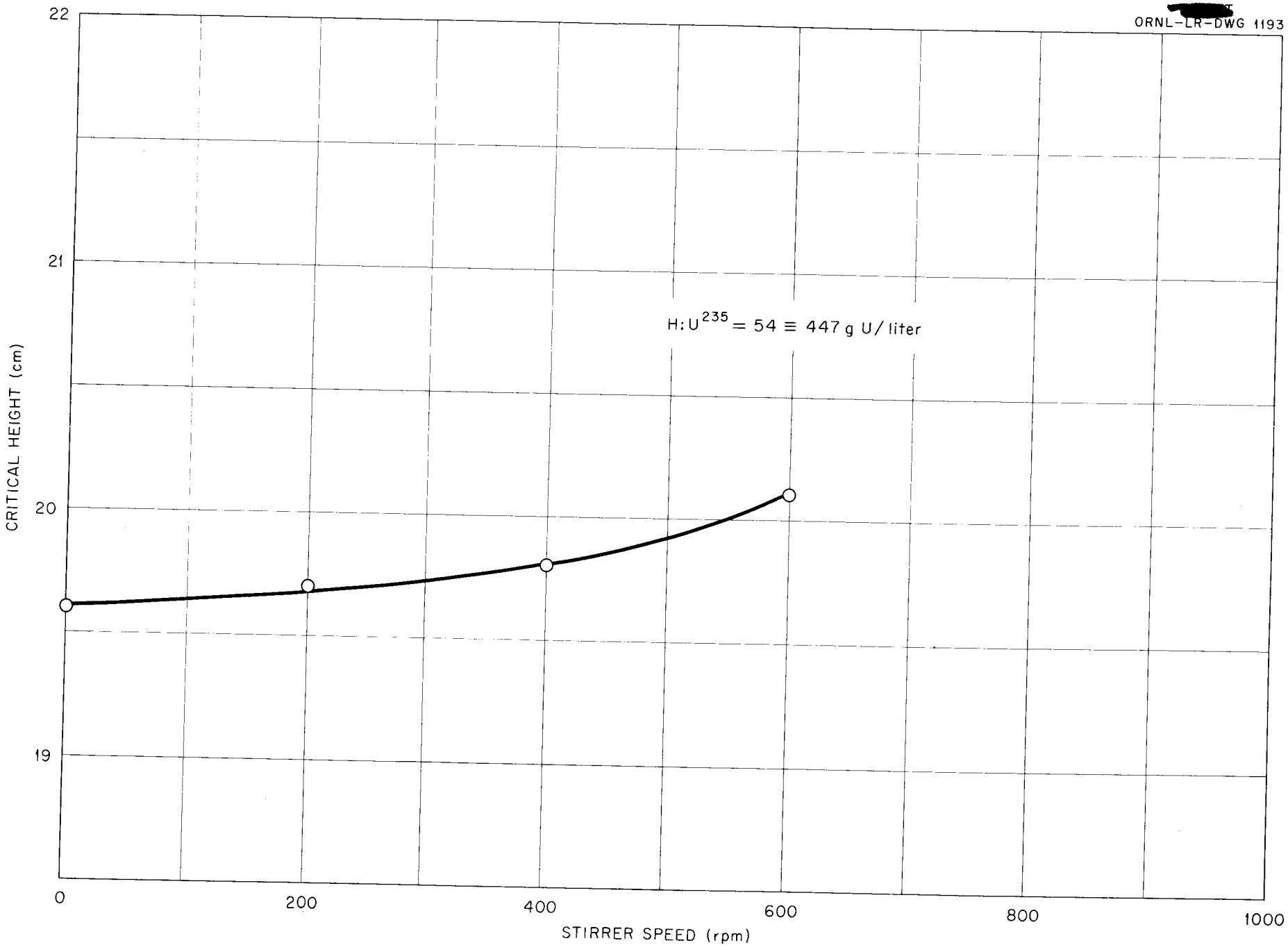

Fig. 17. UO_2F_2 Solution; Critical Height vs. Stirrer Speed.

[redacted]

the distribution of the uranium to remain homogeneous as its concentration increases during settling. In this process the supernatant water becomes a neutron reflector of increasing thickness. From considerations of these concentration and top reflector changes, it was possible, by a one group modified diffusion calculation, to predict a critical behavior and to estimate a value of the chemical concentration below which a just critical system would become super-critical upon settling. At higher initial concentrations the slurry would settle to a less reactive configuration. The experiments reported here illustrate a significant dependence of neutron reactivity upon the uranium distribution, within a 12" diameter cylinder, which is not readily describable. As the agitation is stopped, the pattern changes, apparently, in a manner different from free settling of the particles and the rather large reactivity changes which occur mask the prediction based on settling alone. This is especially true at the higher concentrations. Although the changes in the stirrer speed may be quite rapid, the resulting reactivity peak is delayed, apparently accompanying alterations in the slurry distribution. The magnitudes of the excursions, however, are sufficient to warrent serious consideration because of potential hazards. The apparatus in which these experiments were performed probably gave distributions of uranium which were very different from those encountered in a nuclear power reactor in which the aqueous fuel suspension is circulated through a core region, hence the results reported here may not be directly applicable to such designs. They are, however, indicative of the caution which must be exercised in the design and operation of reactors using liquid fuels in which heterogenities may occur or in which the uranium distribution may change.

VI. ACKNOWLEDGEMENTS.

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[redacted]