ORNL-2452 **Criticality Studies** 

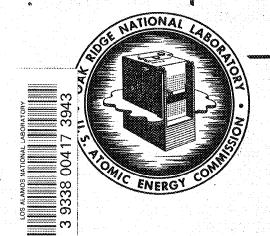
CII

# RADIATION EXCURSIONS AT THE ORNL CRITICAL EXPERIMENTS LABORATORY

1 May 26, 1954

Il February 1, 1956

J. T. Thomas D. Callihan



.....

## OAK RIDGE NATIONAL LABORATORY

operated by UNION CARBIDE CORPORATION for the

### U.S. ATOMIC ENERGY COMMISSION

1

### Printed in USA, Price 1.00 cents. Available from the

N. 18.14

( (S.))

action,

1.11

1.16

Office of Technical Services U. S. Department of Commerce Washington 25, D. C.

#### -LEGAL NOTICE

S. All

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

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

 B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.
 As used in the above, "person acting on behalf of the Commission" includes any employee or

constructor of the Cammission to the extent that such employee ar contractor prepares, handles or distributes, or provides access to, any information pursuant to his employment or contract with the Cammissian.

.

#### ORNL-2452 Criticality Studies TID-4500 (13th ed., Rev.)

Contract No. W-7405-eng-26

Applied Nuclear Physics Division

RADIATION EXCURSIONS AT THE ORNL CRITICAL

EXPERIMENTS LABORATORY

I May 26, 1954

II February 1, 1956

J. T. Thomas and D. Callihan

DATE ISSUED

MAY 271958

OAK RIDGE NATIONAL LABORATORY Oak Ridge, Tennessee operated by UNION CARBIDE CORPORATION for the U.S. ATOMIC ENERGY COMMISSION

#### ABSTRACT

Two prompt critical power excursions have occurred in enriched  $UO_2F_2$  solutions used in critical experiments at Oak Ridge National Laboratory. The first resulted from a mechanical failure in the equipment, and the second was due to a redistribution of the solution caused by the insertion of a safety device into a near critical volume. Although the safety mechanism operated normally in both instances, the order of  $10^{17}$  fissions occurred, corresponding to an energy release of about 1 kwhr. No significant property damage occurred and personnel exposures were limited to a few hundred milliroentgens. Experiments were resumed in a few days. On the basis a partial reconstruction of the first event, a semiquantitative analysis has been made; a similar treatment of the second was not attempted.

### Table of Contents

		Page N	io.
	Abstract	1	-
	Introduction	7	5
I.	Excursion of May 26, 1954	կ	F
	<ol> <li>Description of Equipment</li> <li>The Excursion</li> <li>Radiation Levels and Exposure</li> <li>Immediate Operations</li> <li>Post-Excursion Experiments</li> <li>Probable Chronology of Events</li> <li>Qualitative Description of the Excursion</li> <li>Analysis of the Excursion</li> </ol>	1 10 11 13 19 20 23	701350
II.	Excursion of February 1, 1956	28	3
	1. Description of Equipment2. Chronology of Events3. Cause of the Excursion4. Observations and Results of Analyses	28 28 29 29	B 9
ÍII.	Acknowledgements	, 3	1

#### INTRODUCTION

Two power excursion have occurred in assemblies of  $U^{235}$  accidently made prompt critical in the ORNL Critical Experiments Laboratory. The first of these occurred in May 1954 and the other in February 1956. In both assemblies the uranium, as an aqueous solution of  $UO_2F_2$ , was contained in a nominally unreflected open cylinder and no damaging pressure developed. Although the safety devices operated normally and the reactions were automatically terminated, the energies released in both excursions were about equal and occurred in unmeasured times. Preliminary descriptions of both accidents have been reported<sup>1</sup> but, for completeness, most of the details will be repeated here.

It was possible to reconstruct singly the several operational steps leading to the first event and those instrumental in stopping it, thereby allowing a semiquantitative analysis to be made. The complexity of the mechanism causing the second precluded even a qualitative study.

The uranium in these experiments was enriched in  $U^{235}$  to 93.2%. The chemical concentration in May 1954 was 0.33 g of  $U^{235}/ml$  and that in February 1956 was 0.47 g of  $U^{235}/ml$ .

 A. D. Callihan, "The Radiation Excursion of May 26, 1954," CF-54-6-40 (June 8, 1954);
 A. D. Callihan, "Radiation Incident of February 1, 1956," CF-56-2-105 (Feb. 15, 1956).

#### I. EXCURSION OF MAY 26, 1954

#### 1. Description of Equipment

The floor plan of the critical experiments facility, showing the location of the critical assembly in question and the permanent shielding, is given in Fig. 1. The critical assembly areas are separated from the rest of the building by 5-ft-thick concrete walls which serve as radiation shields. The roof is of conventional construction providing little shielding from scattered radiation.

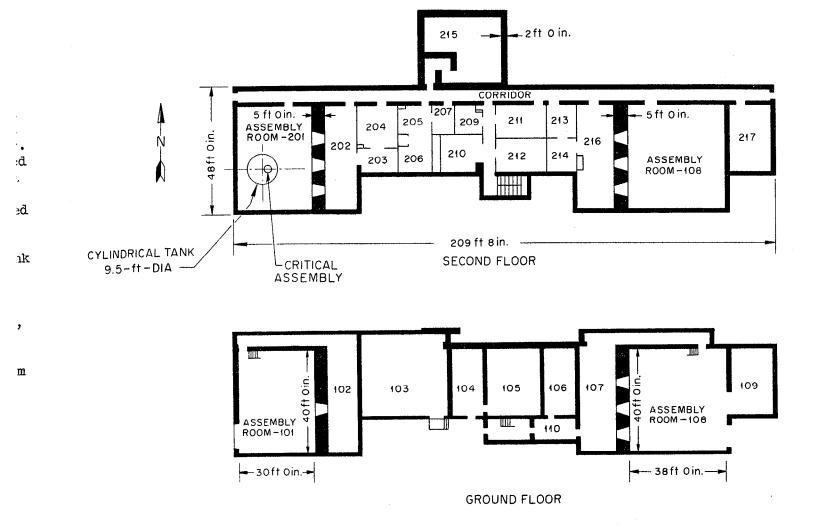
The program in progress at the time of the 1954 excursion was one of a series designed to study critical conditions of aqueous solutions in annular cylindrical containers. The experiments involved the study of the effect on critical mass of varying the inner and outer radii of the annulus and the contents of the inner cylinder. Air, water, cadmium, and combinations of these were the latter variables. All cylinders were 6 ft long and fabricated of 1/16-in.-thick 2S aluminum. The bottom of the outer cylinder was fastened to the top of a Plexiglas table by lugs welded to the outside of the cylinder. The inner cylinder was positioned at the lower end by a pin which was received by a recess in the bottom of the outer cylinder. The upper end was held by a downward compressive force from a 120-deg spider, the legs of which were bolted to the top flange of the outer cylinder. The assemblies were contained in a 9.5-ft-dia x 9 ft cylindrical tank which could be filled with water to provide a neutron reflector if desired. The location of this tank in room 201 is indicated in Fig. 1. The uranyl fluoride solution was stored in a bank of 5-in.-dia cylinders in room 102 which was connected through a 1/2-in.-dia line to a 2-in.-dia pipe directly under the test assembly. The annular assembly could be drained through the 2-in. connection directly into a dump system consisting of a 5-ft length of 5-in.-dia pipe through an air-operated, spring-loaded, normally open, 3-in. diaphragm type valve (Fig. 2) which could be opened automatically by a signal from radiation monitoring instruments. Following such an event the solution could be held in the dump system until such time as it is desirable to drain it back into the normal storage system or into shipping containers.

The neutron source, which was used during the approach to critical, was positioned by a drive mechanism located below the assembly. The source was inserted into the bottom of the assembly through a stainless steel tube located in and coaxial with the 2-in. manifold.

A superstructure above the large cylindrical tank supported a surfacecontact, solution-level indicator and a 3/4-in.-dia cadmium-steel safety rod which was magnetically supported. The safety rod mechanism and the level indicator could be moved vertically by motor-driven racks and pinions and their positions indicated by selsyns.

Prior to the excursion fourteen experiments had been successfully completed in the program with a 10-in.-dia outer cylinder and inner cylinders 6, 4, and 2 in. in diameter.

CYLINI 9.5



- 5 -

UNCLASSIFIED ORNL-LR-DWG 26239

Fig. 1. Floor Plan of Critical Experiments Laboratory. (Room numbers are shown).

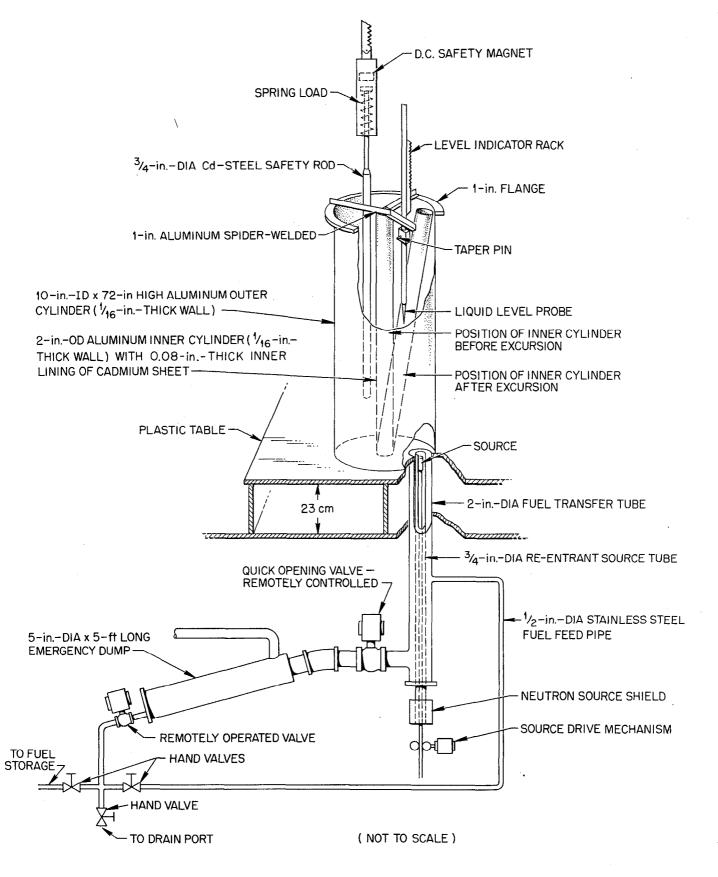
rs

1

£

1

t t



~ 6 -

#### 2. The Excursion

38

ЗE

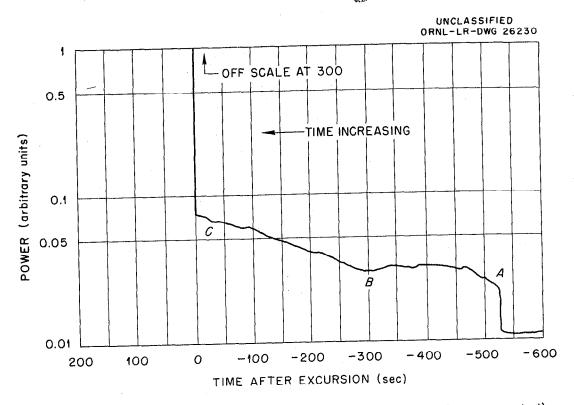
EEL

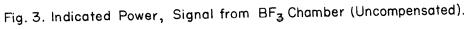
Μ

The experiment in progress at 1 P.M. on May 26 was one in which the 2-in.-dia cylinder contained a lining of 0.08-in.-thick cadmium and was filled with water. The outer 10-in.-dia cylinder was unreflected. The annulus had been filled to a height of 45 in. without becoming critical and the U02F2 solution had been drained back to about 20 in. in order that the cadmium lining inside the inner cylinder could be extended to the full height. A record of the subsequent approach to critical is shown in Figs. 3 and 4. The power level and period meters were connected electronically to and received their signal from a BF3 ionization chamber located approximately 3 ft from the assembly. The chamber was not compensated for gamma radiation, but under ordinary conditions this is not significant. The labeled points in Figs. 3 and 4 indicate changes in neutron level during the subsequent approach to the previous height of 45 in. Between A and B the source was partially withdrawn and reinserted to check the response of instruments. (A greater response was observed on more sensitive instruments.) Between B and C the source was in position, and solution was being added as indicated by the gradual rise in the power trace. The period fluctuations during this interval are statistical and background noise. A fuel height of 40 in. was observed at time C. As the fuel level indicator was being raised and, concurrently, solution being added at a slow rate, a flash occurred and the radiation detection instruments went off scale.

At this time the solution level was below 45.9 in., the terminal position of the indicator. Although the safety systems functioned properly, the instruments did not come back on scale immediately. A survey instrument on the control room indicated a radiation level of the order of 1 r/hr at a water-filled viewing window in the shield wall. The persons conducting the experiment and others near the control room began evacuation of personnel from the adjoining area. Although an immediate survey of the central area of the building revealed tolerable radiation levels, personnel were directed into room 108 in order to take advantage of distance and the second radiation shield wall. Immediate examination of both neutron and gamma-ray personnel monitors showed exposures to have been of the order of a few tenths of a roentgen.

Inspection of the equipment on the day following the excursion showed its cause was a displacement of the inner cylinder, effectively a poison rod, to a region of less importance. This displacement resulted from a dislocation of the positioning spider by a pin, used to connect sections of the liquid-level-indicator rack, protruding beyond the side of the rack and engaging a leg of the spider as the indicator was raised. Removal of the compressional force from the top of the inner cylinder allowed it to fall against the inside of the lO-in.-dia cylinder. A photograph of the top of the assembly after the incident (Fig. 5) shows the distorted spider leg and the top of the inner cylinder against the outer one. Although the displacement was small, it was sufficient to cause a large increase in the effective neutron multiplication.





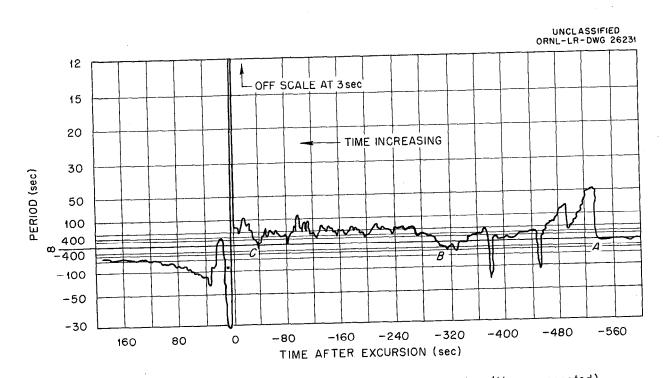


Fig.4. Indicated Period, Signal from  $BF_3$  Chamber (Uncompensated).

-8-

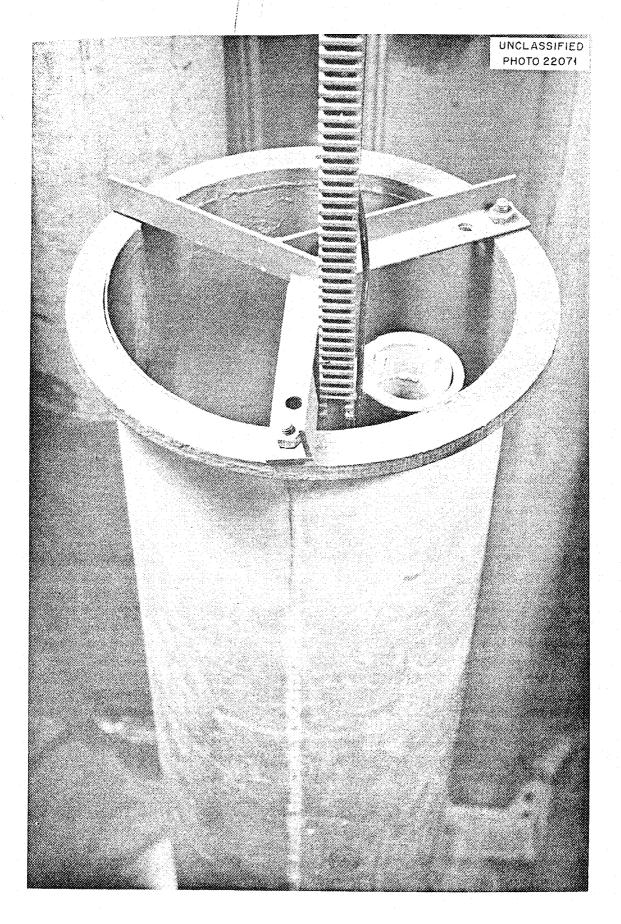


Fig.5. Photograph of Assembly After Excursion on May 26, 1954.

#### 3. Radiation Levels and Exposures

A series of radiation surveys within the building was begun a few minutes after the incident and continued for several days. Personnel film badge exposures ranged from 0.08 to 0.90 rem with an average of approximately 0.3 rem, the largest having been incurred at the guard shelter, which is about 100 ft from the critical assembly. Exposures of persons in and near the control room averaged about 0.5 rem. The gamma radiation doses received by personnel throughout the central part of the building differed from each other by no more than a factor of five even though some were more than 100 ft from the primary shield wall. Radiation detecting film packets distributed in the building were not sensitive enough to establish a gradient. The existence of appreciable amounts of air-scattered radiation, "skyshine", is implied by these observations since, as pointed out earlier, the roof of the test cell was built to meet structural requirements only. This is borne out by the fact that exposures behind the second 5-ft-thick shield were the order of 0.1 rem not much less than some of those observed in the central part of the building.

Early in the afternoon all members of the group received examinations for internal exposure. Nasal swabs showed those who had made the initial radiation surveys to have experienced some internal exposure, a condition substantiated by measurements of urinary excretion of fission products during the succeeding 20 hr. The maximum internal exposure indicated was approximately 0.3 µc.

At the time of the excursion, the exhaust fans in the assembly room were operating and the truck door was open, allowing fission products, expelled from the solution, to be blown immediately from the room since a complete change of air in the room occurred every 4 to 5 min. The ventilating system for the building and the fans in the assembly room were turned off some 15 min later. Data from air samples taken in and around the building, exclusive of the assembly room, within 30 min after the excursion indicated below-tolerance concentrations.

An hour prior to the occurrence the wind was in an ENE direction at 6 mi/hr. At a distance of 1200 ft downwind from the building there would occur a dilution factor\* of approximately  $10^4$ . Since the Laboratory is a greater distance from other occupied areas, it is extremely unlikely that the excursion resulted in significant contamination elsewhere. On the morning of May 28, the ventilating system for all but the assembly room was turned on and occupancy of that part of the building was normal in the afternoon.

A compliant summary of observed radiation levels in mr/hr is given in Table 1. The locations, with respect to the assembly, may be ascertained

\* Furnished by R. F. Myers of the Oak Ridge Weather Bureau Office.

1 1 1

r

(

(

( ( (

1

from Fig. 1. A typical decay curve of radiation level as a function of time, measured at the door to the assembly room, is given in Fig. 6. Other data show the radiation level had dropped by a factor of more than an order of magnitude 30 min after the excursion.

		rels at Three Inter sion (mr/hr)	rvals After
Location	1/2 hr	4 hr	24 hr
utside door to 102	380	11	1.1
ruck door to 101	2650	250	16
orridor door to 201	900	85 .	5.
orridor door to 202	40	2.5	< 0.1*
orridor door to 204	15.5	0.85	≤0.1*
orridor west of door 201	200	19*	1.1*
100m 205	10.3	0.97*	~0.1*
hd-building corridor	10.3	0.97*	~0.1*

#### Table 1. Partial Summary of Building Radiation Levels

\* These values were found by long extrapolations from observed points.

#### 4. Immediate Operations

On May 27, a survey of the reactor assembly room showed not more than a few tens of cubic centimeters of solution to have been displaced from the cylinder and this spillage was confined in the large surrounding tank. Most of the solution was distributed between the reactor vessel and the dump system with a small quantity in the storage tanks of room 102. This last quantity had been drained into the reservoir shortly after the excursion to assure that the safety devices had brought the system subcritical. The radiation field at the top of the reflector tank, a few feet from the solution remaining in the reactor vessel, 24 hr after the excursion was about 0.9 r/hr.

<u>Samples.</u> Several hundred cubic centimeters of the solution were removed directly from the reactor vessel via a transfer line traversing the 5-ft-thick shield. The gamma radiation from small samples of the solution sealed in cylindrical Lucite capsules was monitored. A radiochemical fission-product analysis was made of an additional sample.

Storage of Solution. On May 28 the irradiated solution was transferred from the system to stainless steel cylinders which were then stored in a shielded room during the decay of the residual fission-product activity. By early September the radiation field adjacent to the cylinders had decreased from 600 to 20 mr/hr and the solution was returned to use without decontamination.

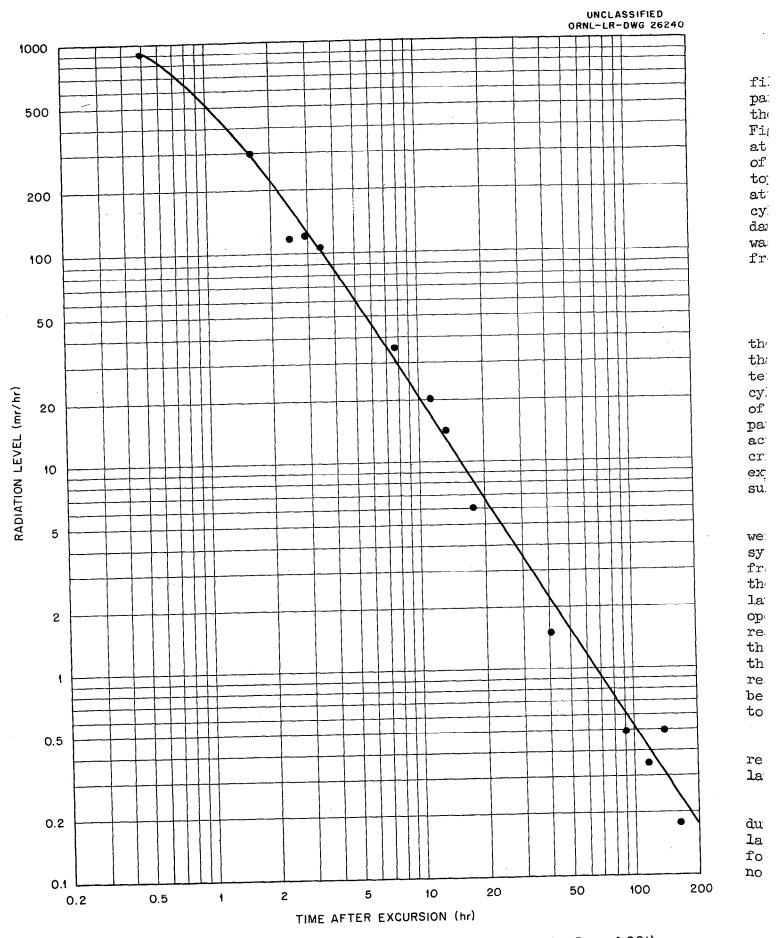


Fig. 6. Typical Decay Curve for Building Radiation Level (Outside Hall Door of 201).

-12-

¢

Exposure of Film to Gamma Radiation. On June 2, 35 dental-sized Du Pont film, Type 552, were placed on the outside of the empty 10-in.-dia cylinder, parallel to the axis, and exposed for 20 hr. The film exposure resulting from the induced activity in the aluminum expressed in milliroentgens is plotted in Fig. 7 as a function of the distance from the top of the cylinder. The peak, at 40 in. from the top, is interpreted as representing the nubilous location of the center of reactivity during the excursion. The gradual rise near the top of the cylinder remains unexplained. The steep rise at the bottom is attributed to the fact that approximately 20 in. of solution remained in the cylinder for two days. During this time a minor but observable radiation damage occurred to the surface of the aluminum. A change in surface appearance was sharply defined at the position of the solution surface by a transition from the typical dull gray of wrought aluminum to a more bright gray.

#### 5. Post-Excursion Experiments

Additional experiments were required to evaluate the reactivity, to check the total energy released, and to better understand the details of the events that occurred. The action and response time of the safety devices were determined, and the time required for the inner cylinder to tilt against the outer cylinder was measured. Also the critical height, and hence the critical mass, of the solution as a function of the position of the center cylinder along its path was measured. These data allowed an estimation of the rate at which reactivity was added and of the lengths of time the system was delayed and prompt critical, respectively. Finally, unirradiated samples of UO2F2 solution were exposed to the known neutron flux in an ORNL reactor, the LITR, and the resultant decay curves were compared with those of samples from the excursion.

<u>Safety System Response</u>. The safety devices installed in the experiment were a spring-loaded cadmium-lined steel safety rod and the solution dump system. These devices could, of course, be actuated manually or by a signal from radiation detection instruments. Of the electronic circuits in service, the one having the shortest response time derived its signal from a scintillation crystal sensitive to gamma radiation. Ten milliseconds was required to operate the photomultiplier tube and its relay after the radiation level reached the preset trip-value; after an additional 60 msec, on the average, the power relays opened. From some estimates of the instrument sensitivity at the time of the excursion, it is assumed that the radiation intensity did not reach the trip-level until the system became prompt critical. Since, as will be shown below, prompt criticality was reached 0.36 sec after the cylinder began to tip, the power supply to the safeties was not interrupted until 0.43 sec.

The force of a spring acted during the first 6 in. of the rod fall, which required 0.10 sec; in an additional 0.20 sec it was fully inserted. In the latter position the rod suppressed reactivity amounting to 0.005 (\$0.65).

The signal from the photomultiplier also interrupted the power to the dump valve, releasing the air pressure which held the valve closed. This latter operation required at least 0.84 sec following which the average rate for draining 10 in. of solution was measured as 4.7 in./sec. The solution did not begin to drain, therefore, until 1.27 sec.

200

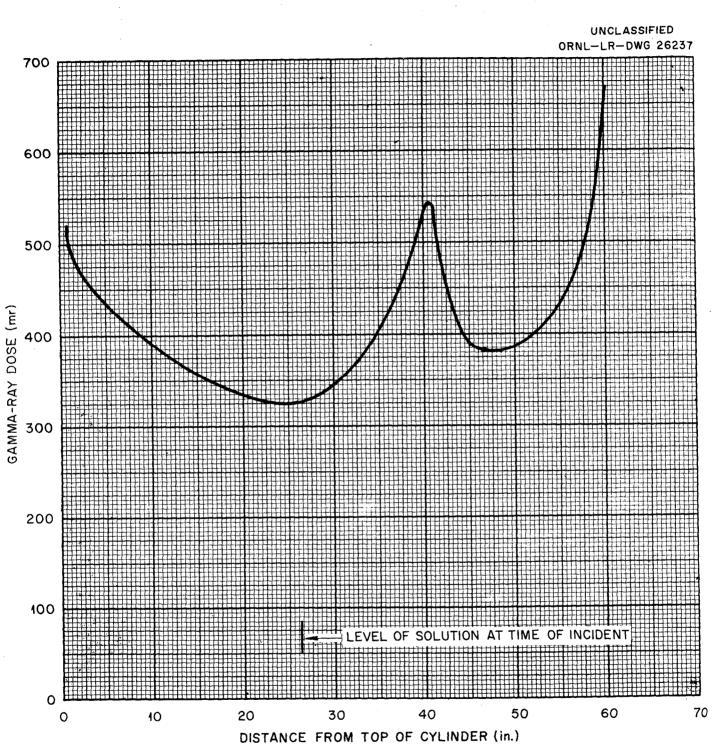


Fig. 7. Gamma-Ray Exposure of Du Pont Type 552 Film Due to Induced Activity of 10—in.—dia Aluminum Cylinder.

.

- 14 -

<u>Tilting Cylinder</u>. The position of the top of the inner cylinder as a function of the time was determined from a recorder trace of a series of signals from electrical contacts made along a radius of the 10 in. cylinder as the inner cylinder fell. Although the measurements were made with water instead of UO2F2 solution, the effect of the differences in viscosities is assumed negligible. The results plotted in Fig. 8 show the motion to be approximately linear and requiring 0.91 sec for the inner cylinder to make contact with the outer one.

Critical Height and Period Measurements. The critical height of the UO2F2 solution was determined as a function of the displacement of the top of the inner cylinder along a radius of the outer cylinder and the results are shown in Fig. 9. Also depicted are the positions of the cylinder for delayed and prompt critical at a solution height equal to that at the time of the excursion. The prompt critical point was estimated from the results presented in Fig. 10. The four experimental points in Fig. 10 give reactivities resulting from solution-height increments made to the delayed critical system with the central cylinder in its extreme position and the line is the result of a two-group analysis assuming the radial buckling remains constant. It is observed that an increment of about 4 in. corresponds to one dollar of reactivity if the effective delayed neutron fraction is 0.0075. If this correspondence is assumed, in turn, to be independent of height over the range of interest, the position of the central rod when the 45-in. column became prompt critical can be estimated. Since the inner cylinder reached its full displacement before the liquid began to drain and probably before the safety rod became effective, the maximum reactivity was about 0.021 (\$2.8) resulting from the solution then being 12 in. above the delayed critical height of 33 in.

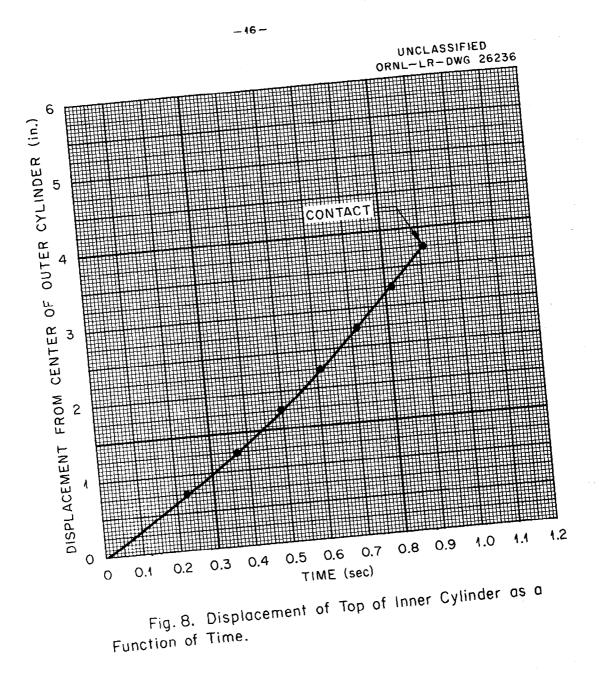
Using Figs. 8 and 9 and the relation  $f = 1.8 \times 10^{-3} \Delta h$  (in inches) from Fig. 10, the time rate of change of reactivity was determined to be essentially constant above prompt critical. The results are showin in Fig. 11.

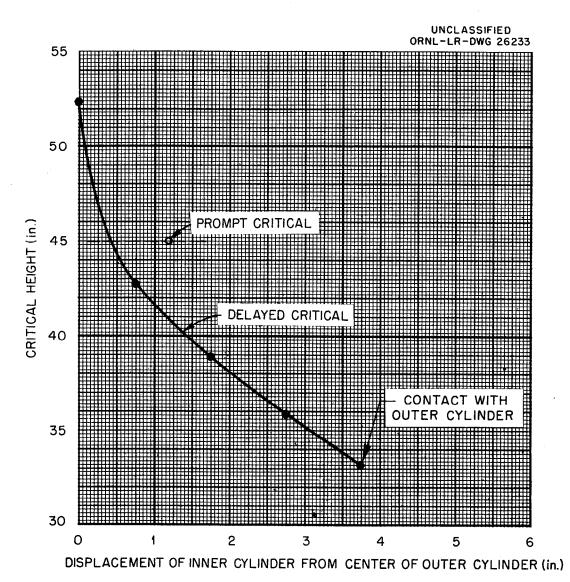
#### 6. Probable Chronology of Events

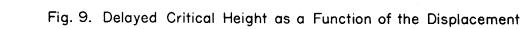
On the basis of the above experiments, the events may be approximately located, in sequence, on a time scale having zero at the start of the motion of the inner cylinder as follows:

ime (se	c) Event
0	Inner cylinder began to tip
0.13	System entered delayed critical state (estimated from cylinder position) 4.35 What
0.36	System entered prompt critical state (estimated from cylinder position)
0.36	Radiation sufficiently intense to actuate safety circuit (assumed)
0.37	Photomultiplier relay operates
0.43	Safety circuits de-energized
0.53	Cadmium rod inserted 6 in. into the solution.

70







of the Top of the Inner Cylinder Along a Radius of Outer Cylinder.

- 17 -

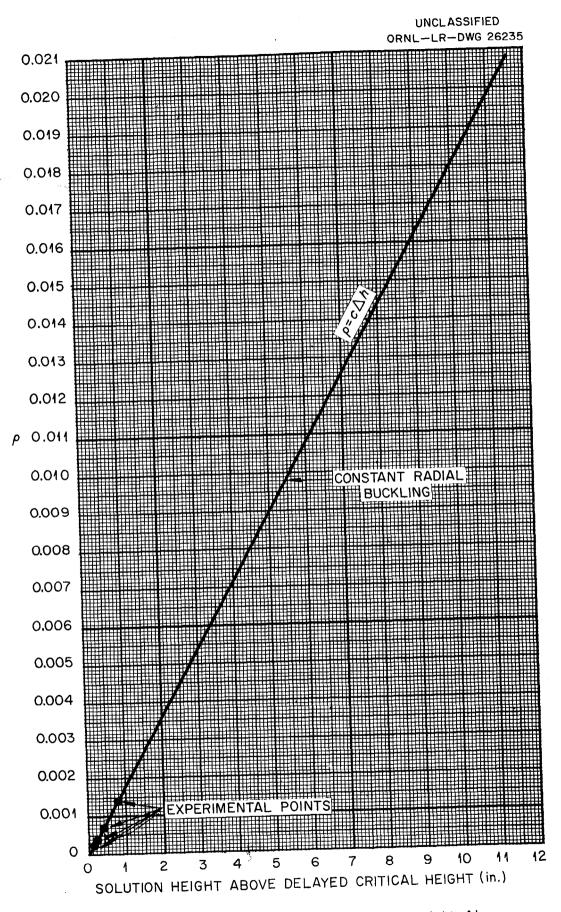
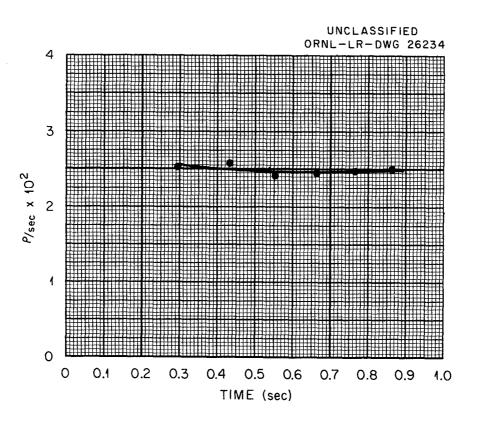


Fig. 10. Reactivity as a Function of Solution Height Above Delayed Critical Height with Top of Inner Cylinder Against Outer Cylinder.



- 19 -

Fig. 11. Time Rate of Change of Reactivity as a Function of Time.

Time (sec	e) Ev	ent		1
0.73	Cadmium rod at end of travel		•	
0.91	Inner cylinder reaches maximum di	splacement		
1.27	Liquid begins to drain			
2.33	System reenters delayed critical	state		
3.22	System becomes subcritical			

#### 7. Qualitative Description of the Excursion

The quantitative observations and post-event experiments were analyzed for a qualitative description which characterizes unscheduled prompt critical assemblies. These include such items as the number of fissions, the energy release, the associated gamma- and neutron-radiation fields, and the method of termination of the excursion.

Energy Release. The total energy released during the excursion was determined from radiochemical analyses for fission products in a sample of the irradiated UO<sub>2</sub>F<sub>2</sub> solution and their yields and disintegration constants. The results from the analyses for five fission products showed that about  $10^{12}$  fissions/ml had occurred. Table 2 summarizes the energy release based on 197 Mev/fission<sup>2</sup> and a solution volume of 55 liters.

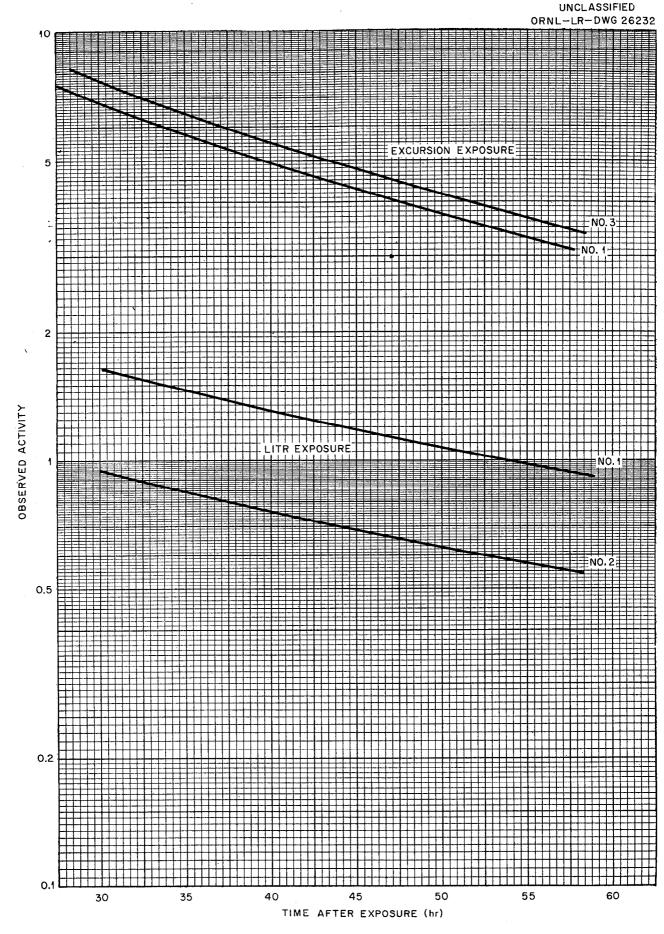
Radio- isotope	Half- life (days)	Fission Yield (%)	Total Number of Fissions (x10-17)	Total Energy Released (Mev x 10-19)
Ba140	12.5	6.1	0.93	1.8
Ce <sup>143</sup> M <sub>0</sub> 99 I131	1.38	5.4	1.2	2.4
M <sup>99</sup>	2.67	6.2	1.1	2.2
II31	8.14	2.97	1.1	2.2
$z_r 95$	65	6.7	1.5	3.0

Table 2. Total Energy Release in the Excursion

The most reliable result is that from the Ba<sup>140</sup> analysis, since the counter efficiencies were better known for this isotope, where the uncertainty is less than  $\pm 10\%$ . On the basis of this value, the total energy released during the transient was 2.9 x  $10^6$  joules, or  $\sim 0.8$  kwhr.

<u>Gamma-Ray Measurements</u>. Approximately 40 mg of the irradiated U02F2 solution was sealed in each of two Lucite capsules, 5/16 in. in diameter and 1/4 in. high, and the delayed-gamma-ray decay curve for each was determined for the interval between 28 to 60 hr after the excursion. The curves are plotted in Fig. 12. Two 50-mg samples of unexposed U02F2 solution were

2. A. M. Weinberg and E. P. Wigner, "Theory of Neutron Chain Reactors," CF-56-11-43, 1, 525.



1

Fig. 12. Decay of Gamma-Radiation Activity of Exposed Samples of  ${\rm UO}_2F_2$  .

-21-

exposed in the LITR in a flux of  $3.4 \times 10^{13}$  n/cm<sup>2</sup>-sec for one second while monitored by a cobalt foil. These were subsequently diluted to an activity corresponding to  $\sim 10^{12}$  fissions/ml and their gamma-ray activities were followed on the same counters and for the same period as were the samples from the excursion.

The observed activity of the samples irradiated to the known flux of the LITR is proportional to the exposure and depends upon the decay prior to the activity measurement. The activity of the samples irradiated in the excursion is proportional to the exposure (number of fissions per unit volume) and to the volume and depends also upon the decay time. If the decay schemes of the two samples are assumed identical, the exposure in the transient is readily calculated to be  $1.01 \times 10^{12}$  fissions/ml corresponding to  $5.6 \times 10^{16}$  fissions in the whole volume. It is noted, however, that the decay curves of the two pairs of samples are not parallel, the slope of those from the LITR being smaller, so this evaluation of the energy release is probably too low, as is shown by comparison with the result from the barium analysis. It is known that shorter exposure times result in steeper slopes of decay curves and, since the LITR exposures were approximately one second in duration, the major portion of fissions which took place in the excursion must have occurred in a time not greater than a second.

 $U^{235}$  Burnup and Solution Temperature Rise. The concentration of the original solution was 0.33 g or 8.49 x 10<sup>20</sup> atoms of  $U^{235}$  per milliliter. If, as shown by the barium analysis, 1.7 x  $10^{12}$  fissions occurred in each milliliter, the  $U^{235}$  burnup was 2.0 x  $10^{-74}$ , corresponding to 6.7 x  $10^{-10}$  g of  $U^{235}/ml$ , a total of 36 µg in the 55 liters irradiated. As may be expected, this change was not detected by an isotopic analysis of the solution. Assuming no heat was lost from the solution and taking its specific heat to be unity, the average rise in temperature was  $10.6^{\circ}C$ .

Associated Gámma-Ray and Neutron Fields. Although the intensity of the gamma-radiation field in the vicinity of the excursion may be estimated in a number of ways, a value for the neutron field is not available.

A detector employing an anthracene crystal and a photomultiplier tube located in the assembly room was used to accuate a safety circuit and its operation was checked daily with a 5-mg radium source. The circuit remained actuated during the time the radiation level was in excess of a preset value and was automatically reset when the level receded below that value. Thirtythree minutes after the excursion it was observed that the photomultiplier circuit was still actuated; 4 hr later the circuit had reset. Since the detector was located 92 in. from the surface of the cylinder containing the  $UO_2F_2$  solution and its sensitivity was such that it could be actuated by a field of 1.73 r/hr, the dose rate at a point 1 in. from the surface of the cylinder 35 min after the excursion was, from an inverse square law, greater than  $1.46 \times 10^4 \text{ r/hr}$ . Assuming that the decay of fission products follows the (time)<sup>-1.2</sup> relation, the calculated dose rate 10 sec after the excursion was greater than  $8.3 \times 10^6 \text{ r/hr}$ . Since the circuit is known to have reset automatically at some time less than 4 hr after the excursion, an upper limit of the gamma-ray field may be obtained in the same manner to be 9.1 x  $10^7$  r/hr. The limits of the dose rate are thereby defined as

$$8.3 \times 10^6$$
 r/hr  $< D < 9.1 \times 10^7$  r/hr.

Knowledge of the total number of fissions determined from the Ba  $^{140}$  analysis enable an estimate of the gamma-ray dose rate if the number of gamma photons per fission and their energy are assumed. The time rate of gamma-ray energy emission at time t after fission is given<sup>2</sup> as  $0.90t^{-1.2}$  where the emission is expressed in Mev/fission-sec and the value of t, in seconds, lies between 10 sec and one day. Taking the mean energy of the gamma photons as 0.7 Mev, the total number of fissions as  $9.3 \times 10^{16}$ , and using the conversion factor<sup>4</sup> 7.5  $\times 10^{5}$ photons/cm<sup>2</sup>sec = 1 r/hr, the dose rate at the reactor approximately 10 sec after the excursion was  $1.6 \times 10^{7}$  r/hr.

The gamma-ray dose rate at the open truck door to Room 101 approximately 15 ft from the reactivity center of the solution 28 min after the excursion was 2.5 r/hr measured by a recently calibrated survey meter. The inverse square and the  $t^{-1.2}$  laws give the dose rate 1 in. from the reactor surface 10 sec after the excursion to be 3.8 x 10<sup>7</sup> r/hr. A similar observation with another survey instrument at a different location and time gives 7.9 x 10<sup>7</sup> r/hr. Table 5 summarizes these estimates.

> Table 5. Summary of Delayed Gamma-Ray Dose Rate Levels After the Excursion

Method	Intensity 1 in. from Reactor 10 sec After Excursion (r/hr)		
Photomultiplier Circuit	8.3 x 10 <sup>6</sup> <d<9.1 10<sup="" x="">7</d<9.1>		
Radiochemical Analysis	$1.6 \times 10^{7}$		
Survey Meter No. 1	$3.8 \times 10^7$		
Survey Meter No. 2	$7.9 \times 10^{7}$		

Accepting a decay factor of at least one order of magnitude during the first 10 sec following the excursion, in order to include the prompt and shortlived gamma-ray contributions, the dose rate at the reactor during the excursion was  $\sim 10^{\circ}$  r/hr.

#### 7. Analysis of the Excursion

The following is a description of a possible sequence of events occuring during the excursion. It was observed from the post-event experiments that about 0.36 sec was required for the system to become prompt critical, that the safety rod was inserted in 0.73 sec and that the solution began to drain in

3. K. Way and E. P. Wigner, Phys. Rev. 73, 1318 (1948); see also Katcoff, Finkle, Elliott, Knight, and Sugarmann, Metallurgical Laboratory Report CC-1128 (Dec. 7, 1945).

4. E. P. Blizard, "Introduction to Shield Design, II," CF-51-10-70 (Mar. 7, 1952).

1.27 sec, zero time being the inauguration of the motion of the inner cylinder. The time required to drain to the delayed critical height with the inner cylinder against the outer one and with the safety rod inserted was 3.22 sec. The energy release to be expected in this interval would have been a few orders of magnitude greater than that observed. Although the safety devices were responsible for the final termination, it is evident that some other mechanism limited the power surge commensurate with the observed energy release in the available time.

The number of fissions,  $\phi$  , occurring during a burst may be characterized by the differential equation?

$$\frac{d^2\phi}{dt^2} = \alpha(t) \frac{d\phi}{dt}$$
(1)

provided the assembly to prompt critical has been slow. The quantity  $\alpha(t)$ is the neutron multiplication rate and is composed of two terms

$$\alpha(t) = \alpha_1(t) - \alpha_2 \tag{2}$$

where  $\boldsymbol{\kappa}(t)$  is the multiplication rate resulting from mechanical changes in the assembly which are time dependent and, for a constant rate of assembly, is equal to at where a is the time rate of change of the neutron multiplication rate; & is the rate of the competing effect due to the disassembly forces and is assumed equal to the product of  $\phi$  and a constant, <u>b</u>. In this treatment it is further assumed that the temperature remains unchanged. The duration of the burst, which is considered symmetrical in intensity about its midpoint in time, is 2t<sub>o</sub>, given by

$$\frac{2}{at_0^{\circ}} \exp\left(\frac{at_0^{\circ}}{2}\right) = \frac{2a}{b\phi_0}$$
(3)

The fission rate,  $\phi_0$ , is that occurring as the system entered the prompt critical state and is approximated by entergent = 7,5×10 = 7,5×10 = 7,5×10

$$\dot{\phi}_{0} = S \sqrt{\frac{\pi \, q_{R}}{2 (\gamma \beta)^{2} \dot{D}}}$$

where

 $\alpha_{\rm R}$  = multiplication rate at the beginning of prompt critical (100 sec<sup>-1</sup> based on experience with the HYPO Water Boiler),

 $\gamma\beta$  = effective delayed neutron fraction, taken to be 0.0075,

D = rate of addition of reactivity, expressed in dollars per second S = strength of the Po-Be neutron source present during the excursion.

<sup>5.</sup> G. E. Hansen, "Burst Characteristics Associated with the Slow Assembly of Fissionable Materials," LA-1441 (July, 1952). See also LA-596 by K. Fuchs (Classified). Much of the following analysis is taken from these reports.

On the basis of this model the total number of fissions which would occur during a single burst may be shown to be  $^{\rm 2}$ 

$$\phi = \frac{2at_0}{b}$$
(5)

The coefficients a and b are derived in the following manner from the expressions\*

$$k_{\text{eff}} = \frac{k_{\boldsymbol{\omega}}}{(1+L^2B^2)(1+\boldsymbol{\tau}B^2)}$$
(6)

(7)

and

 $\alpha = \frac{k_{eff} - 1}{\ell}$ 

where

# $\mathcal{L}$ = mean lifetime, in seconds, of the prompt neutrons in the solution.

The change in  $k_{eff}$  is the net results of an increase due to the tilting of the central cylinder and a decrease caused by the disassembly action now assumed to be the density variation due to dissociation gases in the aqueous solution. Equation 7 then becomes

$$\boldsymbol{\alpha} = \frac{2}{\boldsymbol{\lambda}} \left[ \frac{(1+L^2B^2) \boldsymbol{\gamma} + (1+\boldsymbol{\gamma}B^2) L^2}{(1+L^2B^2)(1+\boldsymbol{\gamma}B^2)} \right]_{\mathbf{0}} \left[ \frac{\pi^2 \text{ st}}{(h_0+2\boldsymbol{\lambda})^3} - \left( B_0^2 - \frac{\pi^2 h_0}{(h_0+2\boldsymbol{\lambda})^3} \right) \frac{f}{v_0} \boldsymbol{\phi} \right]$$
(8)

where

- s = positive rate of change in reactivity due to the tilting cylinder expressed as an effective rate of increase on solution height (35 cm/sec),
- $\lambda$  = unreflected extrapolation distance (3 cm),
- f = volume of gas formed in UO<sub>2</sub>F<sub>2</sub> solution per fission<sup>6</sup> =  $1.03 \times 10^{-16}$  liters/fission,
- V = volume of solution which would be delayed critical with the inner cylinder tilted,
- t = time the system has been prompt critical,
- h<sub>o</sub> = delayed critical solution height with the inner cylinder in its position of maximum displacement

and the subscript zero refers to the delayed critical conditions for the tilted inner cylinder.

- \* The notation used is that of S. Glasstone and M. C. Edlund, "The Elements of Nuclear Reactor Theory," Van Nostrand, New York, 1952.
- 6. J. W. Boyle et al., "The Decomposition of Water by Fission Recoil Particles," Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, 7, Paper 741 (1955). It is also shown by B. R. Leonard, Jr. in report HW-24327 that the time for bubble formation is short compared to the duration of a burst.

In Eq. 8 the coefficient of t is a and the coefficient of  $\emptyset$  is b, the constants determining  $\alpha_1$  and  $\alpha_2$ , respectively. For a UO<sub>2</sub>F<sub>2</sub> solution with a density of 0.33 g of U<sup>235</sup>/ml, a consistent set of two-group parameters gives

$$a = 34.7s \text{ sec}^{-2}$$
  
 $b = 1.66 \times 10^{-13} \text{ sec}^{-1}$ 

For a source strength, S, of  $3.8 \times 10^7$  n/sec and a rate of assembly from Fig. 11 of 3.3/sec,  $\phi_0$  is  $3.5 \times 10^{10}$  fissions/sec. Hence, from Eq. 3,  $t_0 = 0.16$  sec, and, from Eq. 5,  $\phi = 2.3 \times 10^{15}$  fissions. The total duration of the burst, due to the symmetric character of the solution of Eq. 1, is 0.32 sec. These results are inconsistent with the observed energy release and estimated duration of the excursion.

The duration of the excursion, i.e., the time above prompt critical, was ample for the system to have behaved in the oscillatory manner depicted in LA-596.<sup>5</sup> In this model the assembly and disassembly forces alternately dominate, resulting in a rapid sequence of bursts. However, adoption of this model would require the disassembly forces to be completely removed at the end of a cycle and the reactivity negated by this force to be returned during the succeeding cycle in a time equal to one-half the preceding burst width. This is tantamount to requiring the bubbles formed due to fission to pass out of the solution in a time not compatible with the physical situation. Recent work<sup>7</sup> indicates that the residence time for bubbles in such a situation is of the order of 2 to 3 sec. It is further suspected that there is a delay time for bubble formation. It seems reasonable, therefore, to seek the delay time required to produce the observed number of fissions.

Accepting the hypothesis of a bubble residence time in excess of 2 sec, it is evident that only a single burst would have been possible. This burst would have terminated a short time before the inner cylinder had completely tilted. At this time the system would have been subcritical, and, since the safety rod would have already become effective, the reactivity added by the tilting cylinder during the remainder of its travel would have at most brought the system back to delayed critical for a very short time.

Postulating a delay time,  $t_d$ , the number of fissions occurring before bubbles begin to appear is approximated by the expression

$$\phi = 2\phi_0 \frac{\exp\left(at_d/2\right)}{at_d}$$
(9)

(10)

and after time  $t_A$  the burst is described by

$$\phi = \frac{2 \alpha_1}{b}$$

7. D. L. Hetrick et al., "Preliminary Results on the Kinetic Behavior of Water Boiler Reactors," NAA-SR-1896 (April, 1957). h

which is the relation describing instantaneous assembly of a prompt critical assembly.<sup>5</sup> A delay time of 0.151 sec yields  $6.6 \times 10^{10}$  fissions from Eq. 9 and 2.5  $\times 10^{16}$  fissions from Eq. 10, giving a total of 9.1  $\times 10^{16}$  fissions, which is to be compared with the energy release determined by other methods. The associated peak fission rate is  $1.7 \times 10^{19}$ /sec corresponding to a peak power of 500 Mw.

#### II. EXCURSION OF FEBRUARY 1, 1956

#### 1. Description of Equipment

The program in progress at the time of the 1956 excursion was a series of experiments designed to evaluate certain reactor parameters by measuring stable reactor periods. The equipment was essentially that used at the time of the previous accident and was in the same location. The experimental setup is described in Fig. 2 modified by removing the inner test cylinder, enlarging the outer one from 10 to 30 in. in diameter, and replacing the cadmium-steel safety rod by a steel-clad sheet of cadmium 6 in. wide. Although the plumbing for the solution was the same as before, somewhat more detailed reference will be made here to the solution handling procedure since it figured in this occurrence to a greater extent than in the earlier one.

Transfer of solution from storage to the test cylinder was effected by the application of air pressure to the storage vessel and flow was controlled by a remotely operated valve in the 1/2-in.-dia line. With the control switch in the "feed" position this valve was open and the air pressure was applied; with the switch in the "drain" position the valve was also open but the air supply was turned off and the storage vessels were vented to the atmosphere. When the switch was in the intermediate "neutral" position the valve was closed and the storage vessels were vented.

#### 2. Chronology of Events

On February 1, 1956 the 30-in.-dia cylinder was being made critical by the successive addition of small increments of solution having a concentration of 0.47 g of  $U^{235}/ml$  and a specific gravity of 1.58. The  $U^{235}$  enrichment of the uranium was 93.2%. After several additions to the reactor it was apparent from the control instruments that another increment would be needed to achieve a critical system at the desired power level. The volume of the solution in the cylinder was then 58.8 liters, about 100 ml less than the critical volume. The addition was made and the transient period decreased rapidly to approximately 30 sec where it seemed to remain constant. Removal of the source was started at about this time and shortly thereafter the fuel control switch was placed in the "drain" position. The period meter again indicated a rapid increase in reactivity. The safety devices were then actuated about simultaneously by both manual and instrument signal, the instrument trip-point having been set at a 10-sec period. All recording instruments, including a logarithmic amplifier, were observed to be off scale showing that a power excursion had occurred so the laboratory was evacuated immediately except for an emergency team.

A favorable wind and the isolation of the laboratory made it possible to purge the test cell in which the accident occurred using ventilating fans installed during construction for that purpose. The small amounts of betaray and gamma-ray activity which fell out in other parts of the building were removed and, except for the test cell, occupancy was normal the morning of February 2. After the removal of the irradiated solution from the system to a shielded area on February 2, the background radiation was sufficiently low to allow unobstructed access to the test cell.

#### 3. Cause of the Excursion

The excursion was initiated by an unintentional over-addition of solution to the reactor. Later observations showed that addition of solution to the reactor could have continued for several seconds after the control switch was placed in the drain position if insufficient time were allowed for the operating pressure to be vented. Another increment was, therefore, probably added after the switch was thrown, accounting for the observed positive period.<sup>\*</sup> Extrapolation of some measurements of excess reactivity as a function of solution height shows, however, that the rate at which solution could be added in this manner was insufficient to raise the reactivity from delayed to prompt critical in the time of the excursion. In fact, even the rate with full operating pressure was too low to account for the rapid rise. It is necessary, therefore, to consider other mechanisms by which the solution could have been made prompt critical.

It has been observed that the critical heights of cylindrical volumes of this solution, having diameters in the range considered here, are very insensitive to the diameter. The critical height of a 30-in.-dia cylinder is about 5 in., only 0.5 in. less than that of a 20-in.-dia cylinder. Any disturbance reducing the effective diameter of the solution would result in a concomitant increase in height to a value in excess of the critical height. Such a disturbance was probably caused by the insertion of the safety sheet. No definitive experiments were performed to establish this mechanism although it is substantiated by qualitative observations of the sheet falling into water.

#### 4. Observations and Results of Analyses .

The total neutron and gamma-ray exposures of persons in the building are shown in Fig. 13 at their locations at the time of the excursion. These results were obtained from film badges carried by the individuals and agree with the exposures shown by their neutron- and gamma-ray-sensitive dosimeters. The values are confirmed by film meters distributed throughout the building. The counting room on the second floor has 2-ft-thick walls and roof and it is to be noted that the exposure there was unmeasurable, additional evidence for the scattering of radiation into other parts of the building. Also shown on Fig. 13 are the fast (>7 kev) and thermal (<0.5 ev) neutron doses (nvt) at three locations within the building. The results were obtained from the activities induced in plutonium, in gold, and in cadmium-covered gold foils.

Samples of the irradiated solution became available on February 2 for radiochemical analyses and for direct activity measurements. The following number of fissions which occurred per unit volume of the solution were derived from these analyses.

\* This pneumatic system of solution transfer has been replaced by a cannedrotor pump which was, in fact, on order at the time of the excursion.

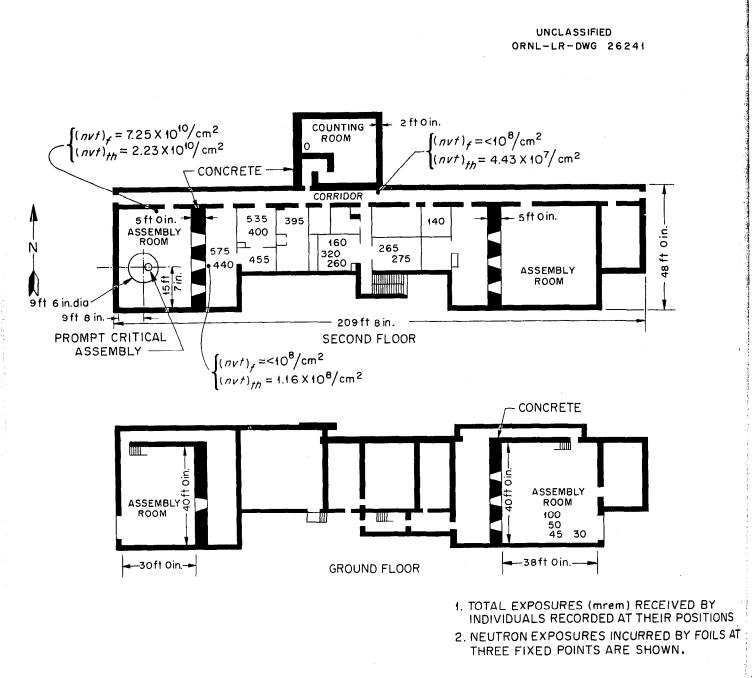


Fig. 13. Radiation Exposures from February 1956 Excursion.

-30 -

Isotope <sup>*</sup>	Fissions/ml (x 10 <sup>-12</sup> )
Ba <sup>140</sup>	2.6
La <sup>140</sup>	2.8
Sr <sup>91</sup>	1.6
Ru <sup>103</sup>	3.5
Cr <sup>143</sup>	3.9
1 <sup>131</sup>	3.5

\* The La<sup>140</sup> result is from a direct gamma-ray measure of a sample of the solution; the others are from radiochemical analyses.

As stated above, about 59 liters of solution had been made critical. Using the more reliable  $Ba^{140}$  and  $La^{140}$  analyses and assuming the total energy release to be 197 Mev/fission, 1.6 x  $10^{17}$  fissions occurred with an energy release of 3.1 x  $10^{19}$  Mev, 5.0 x  $10^{6}$  joules or 1.4 kwhr. The activity induced in a plutonium foil located 27 ft from the cylinder resulted from an exposure to 7.25 x  $10^{10}$  fast neutrons/cm<sup>2</sup>, which, in turn, would have been produced in the order of  $10^{17}$  fissions. No temperature measurements were made in the solution. The volume of gas formed was about 12 liters.<sup>6</sup> Approximately 60 µg of  $U^{235}$  was consumed.

It has not been possible to estimate the excess reactivity or the duration of the excursion.

A considerable volume of solution was forcibly ejected from the cylinder, requiring a laborious chemical (not radioactive) decontamination of the assembly room. The bottom of the 30-in.-dia cylinder, made of type 2S aluminum 0.5-in.thick, was noticably distorted by a downward force. No light was observed by those who saw the displacement of the solution, perhaps a consequence of the liquid being near the bottom of a tall cylinder.

3Y TIONS DILS AT

#### ACKNOWLEDGEMENTS

The problems associated with these two experiences were minimized and the observations were evaluated only through the cooperation of a large number of individuals. Very special reference is made to the calm and orderly behavior of the members of the laboratory staff at the times of the occurrences and their assistance until normal operations could be completely resumed. Acknowledgement is also made of the generous assistance in matters of Health Physics supplied by the ORNL staff under K. Z. Morgan and the Y-12 groups then under L. C. Emerson. The radiochemical analyses were made by M. T. Kelley, S. A. Reynolds and E. I. Wyatt. The control samples were irradiated in the LITR by J. A. Cox and staff.

100.

ב 2 3 4-9 6

7-26 27

28

50 50

32.

ORNL-2452 Criticality Studies TID-4500 (13th ed., Rev.) February 15, 1958

#### INTERNAL DISTRIBUTION

1. C. E. Center 2. Biology Library 3. Health Physics Library 4-5. Central Research Library 6. Reactor Experimental Engineering Library 7-26. Laboratory Records Department 27. Laboratory Records, ORNL R.C. 28. A. M. Weinberg 29. L. B. Emlet (K-25) 30. J. P. Murray (Y-12) 31. J. A. Swartout 32. E. H. Taylor 33. E. D. Shipley 34. A. H. Snell 35. M. L. Nelson 36. W. H. Jordan 37. G. E. Boyd 38. R. A. Charpie 39. S. C. Lind 40. F. L. Culler 41. A. Hollaender 42. J. H. Frye, Jr. 43. M. T. Kelley 44. J. L. Fowler 45. R. S. Livingston 46. K. Z. Morgan 47. T. A. Lincoln 48. A. S. Householder 49. C. P. Keim 50. C. S. Harrill 51. C. E. Winters

52. D. S. Billington 53. D. W. Cardwell 54. D. Phillips 55. A. J. Miller 56. E. P. Blizard 57. M. J. Skinner 58. R. R. Dickison 59. J. A. Harvey 60. A. Simon 61. F. C. Maienschein 62. C. E. Clifford 63. J. K. Fox 64. L. W. Gilley 65. A. D. Callihan 66. R. Gwin 67. J. T. Thomas 68. D. F. Cronin 69. D. Scott 70. E. R. Rohrer 71. L. C. Emerson 72. L. C. Johnson 73. W. Zobel 74. F. L. Keller 75. L. B. Holland 76. C. D. Zerby 77. C. A. Preskitt 78. F. F. Friedman (consultant) 79. H. Goldstein (consultant) 80. H. Hurwitz, Jr. (consultant) 81. L. W. Nordheim (consultant) 82. R. R. Wilson (consultant)

83. ORNL - Y-12 Technical Library,

Document Reference Section

#### EXTERNAL DISTRIBUTION

84. H. F. Henry, Union Carbide Nuclear Company (K-25 Plant) 85. J. D. McLendon, Union Carbide Nuclear Company (Y-12 Plant) 86. H. K. Clark, duPont Company, Aiken 87. C. L. Schuske, Dow Chemical Co., Rocky Flats 88. Frank Woltz, Goodyear, Portsmouth 89. H. C. Paxton, Los Alamos Scientific Laboratory 90. R. B. Lemon, Phillips Petroleum Co. (NRTS) 91. E. D. Clayton, General Electric Co., Hanford 92. James Carothers, University of California Radiation Laboratory, Livermore 93. R. C. Baker, Union Carbide Nuclear Company (C-31 Plant) 94. John Flora, Atomics International 95-97. AEC, Washington (1 copy ea. to C. K. Beck, Rogers McCullough, and Marvin Mann) 98. W. J. Ozeroff, General Electric, Valecitos Laboratory, San Jose, Calif. 99. T. A. Fox, National Advisory Committee for Aeronautics, Cleveland 100-646. Given distribution as shown in TID-4500 (13th ed., Rev.) under Criticality Studies category (75 copies - OTS)

647. Division of Research and Development, AEC, ORO