

# ACCIDENTAL RADIATION EXCURSION AT THE OAK RIDGE Y-12 PLANT—I DESCRIPTION AND PHYSICS OF THE ACCIDENT

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(Received 13 November 1958)

**Abstract**—An aqueous solution of enriched uranium is inadvertently flowed into a 55 gal drum in a process area in Oak Ridge in June 1958, establishing a prompt-critical neutron chain reaction in which about  $10^{18}$  fissions occurred before the system finally became subcritical by the addition of water. The solution contained about 2.5 kg of  $U^{235}$ . Records of the radiation field show the power excursion to have continued about 20 min during which the reaction oscillated a number of times. This paper describes the accident and presents a reactor-physics analysis yielding reactivities in an unperturbed system as great as 1.3 per cent which were above zero for a time consistent with observations. A plausible sequence of events during the excursion is enumerated. The emergency and health physics procedures and the medical observations of exposed personnel will be given in subsequent papers of this series.

## INTRODUCTION

ESTIMATES have been made of the expected consequences to personnel and equipment of an uncontrolled neutron chain reaction outside a nuclear reactor ever since the inception of chemical and metallurgical processes with fissionable materials. These estimates have been primarily extrapolations of the experiences with nuclear accidents in experimental critical assemblies<sup>(1)</sup> although at least one analytical treatment of an assumed set of conditions has appeared in the literature.<sup>(2)</sup> The accident that occurred in one of the salvage areas in Oak Ridge in June 1958 is the first recorded radiation excursion in a process area and resulted from the inadvertent accumulation of an aqueous enriched-uranium solution in a process vessel. This occurrence, which has been reported fully elsewhere,<sup>(3)</sup> was an important experience in applied health physics since the primary and immediate concern was an evaluation of personnel exposure to penetrating

radiation and the establishment of necessary remedial measures. A study of the health physics problems and of the medical findings will be presented in subsequent papers in a series of which this is the first. The cause of the accident and an analysis of it from a reactor physics viewpoint, both of which are discussed in this paper, are of less overall importance except as they aid in establishing the radiation levels to which personnel may have been exposed. Therefore, only a brief description of the cause and a review of what is believed to be the sequence of events leading first to the critical condition within the solution and to final return to subcritical are presented here. One plausible reactor physics analysis of the power excursion is also considered.

## DESCRIPTION AND ANALYSIS

The nuclear accident occurred in an area in which salvable enriched uranium ( $\sim 90\%$   $U^{235}$ ) is recovered from various materials by physicochemical methods in a complex of equipment. This recovery process was being remodeled at the time, and the situation was

\* Operated for the U.S. Atomic Energy Commission by Union Carbide Nuclear Company.

further aggravated by an inventory then in progress. The latter required disassembly, cleaning, reassembly and leak testing of certain pieces of equipment, particularly several long 5 in. diameter pipes used for storage of aqueous solutions of  $U^{235}$  whose shape and dimensions prevent the establishment of nuclear chain reactions. The leak testing operation consisted in filling the pipes with water from a 55 gal stainless steel drum (approximately 22 in. in diameter and 33 in. high), observing the joints in the system and subsequently draining the water back into the same drum. These inventory procedures extended over several days and it was not possible or economic to schedule them concurrently throughout the several stages of the salvage train. As a matter of fact, operations had been re-established in the step immediately ahead of the scene of the accident some hours prior to its occurrence and a part of the bank of storage pipes in question had been leak tested a few days previously. As a consequence of both this time factor and irregularities in the function and operation of some valves, a quantity of solution was inadvertently transferred from the area already in operation into the one still undergoing leak testing. Further, this transfer was into one of the 5 in. diameter pipes which had already been tested. It has been established subsequently that the flow pattern from these pipes into the drum intended to receive the leak-test water was such that the accumulated solution preceded the water. The dimensions of the drum and the concentration of the solution permitted the system to become critical. The reaction was terminated sometime later by the inflow of a relatively larger volume of the water believed to have been the only liquid present in the system. A photograph of the drum in the position occupied during the accident is shown in Fig. 1.

A specification of the manner and rate of establishment of the neutron chain reaction system, the determination of the time which elapsed between its first becoming critical and its final return to subcritical together with the power pattern within this interval, and the mechanism by which the nuclear reaction was ultimately terminated would constitute a mini-

mal description of the event. Although the process of transfer of liquid from one vessel to another is fundamentally simple, it is correct to infer from the above description of the present operation that many of the details of this transfer are not known even after some careful attempts at reconstruction with non-reactive solutions. It should be pointed out, parenthetically, that although the liquid transfer can certainly typify chemical operations in which accidents of this kind may be expected to occur, it is not believed that this same series of events would ever again ensue, thereby duplicating the consequence. For these reasons any value

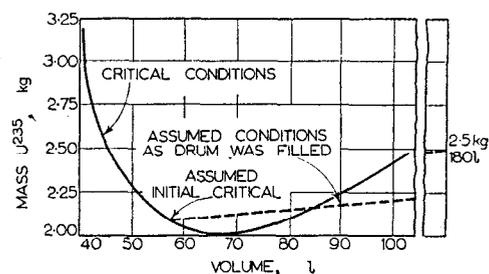


Fig. 2. Mass-volume relations in 55 gal drum during radiation excursion.

of a detailed analysis to the field of reactor physics is doubtful. Although there is no evidence of any basically unexpected physical phenomenon, a complete analytical description of the critical event, agreeing with the observations, would be gratifying and would satisfy the scientific curiosity of many readers. Unfortunately such a description is not possible. A great many observations have been combined to present here a qualitative discussion of the course of events.

A chemical analysis showed 50 g of  $U^{235}$  per l. to be the most concentrated solution available for transfer to the drum and 2.5 kg of  $U^{235}$  as the mass transferred. A plot, Fig. 2, of a short extrapolation of measured critical dimensions of  $U^{235}O_2F_2$  solutions ( $\sim 90\%$   $U^{235}$ ) gives critical masses as a function of critical volumes in a 21.75 in. diameter unreflected steel cylinder. It is seen that the above values of the chemical concentration and mass set 7.6 and 17.2 in. as the lower and upper limits on the critical height. Since both the sequence of

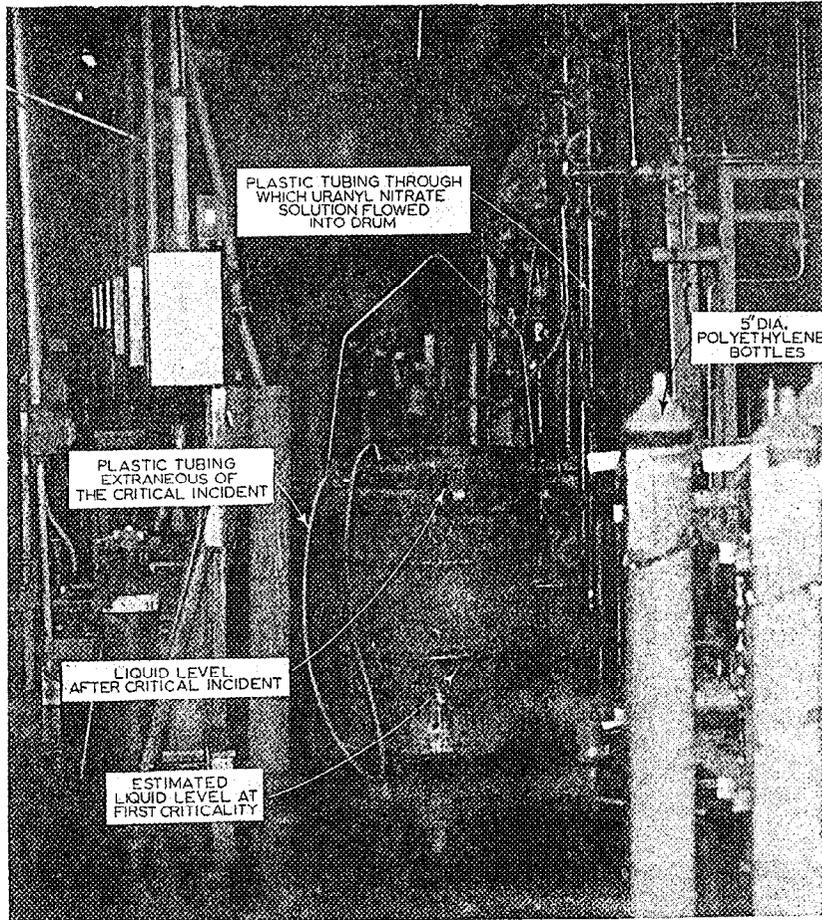


FIG. 1. Drum (55 gal) in which the critical incident occurred.\*

\* *Correction:* The wording on the figure "Plastic tubing extraneous of the critical incident", should read "Plastic tubing extraneous to the critical incident".

Table 1. Calculated reactivity during radiation excursion

Time (min)	Solution				Critical Mass† U <sup>235</sup> (kg)	Reactivity × 10 <sup>3</sup>
	Height		Volume (l.)	Mass U <sup>235</sup> (kg)		
	(cm)	(in.)				
0.0*	23.45	9.23	56.2	2.10	2.10	0
1.8	25.07	9.87	60.1	2.11	2.04	7.5
5.4	27.12	10.68	65.0	2.12	2.02	11.4
9.0	29.20	11.50	70.0	2.13	2.03	12.4
12.5	31.29	12.32	75.0	2.15	2.07	11.8
15.3	32.82	12.92	78.7	2.16	2.15	7.5
20.0	35.67	14.04	85.5	2.18	2.18	0

\* The drum was delayed critical at this point.

† This mass in the volume shown at the left will be critical.

valving operations postulated and the data from the hydraulic reconstruction experiments stipulate some dilution of the original solution as it flowed into the drum, a volume of 56.2 l. containing 2.10 kg of U<sup>235</sup> standing at a height of 23.45 cm (9.23 in.) is selected as the initial delayed critical configuration. This selection is justified by these factors: the reactor analysis which has been made, based on these initial conditions, yields a time interval consistent with what is believed to be the observed duration of the excursion, and the assumed critical height agrees both with the liquid level estimated in the drum by the individual standing nearby at the time of the first indication of a reaction and with the distribution of induced activity in the walls of the drum described below. If it is assumed that the concentration of the solution subsequently added to the drum was uniform and that the volume in the drum reached 180 l. when the entire 2.5 kg of U<sup>235</sup> had been transferred, the mass-volume relation in the drum is described by the straight line on the plot. It is recognized that this simplifying assumption is somewhat unrealistic and overestimates the time interval between delayed and prompt critical. It does, however, provide a lower limit to the reactivity available as a function of time. The details of the analysis are given in the Appendix and the results are in Table 1. The reactivity as a function of the solution height in the drum and of the time after delayed critical

is shown in Fig. 3. The time scale was derived from some of the post-accident hydraulic measurements, particularly the rate of flow of liquid into the drum. The duration of the excursion, by this analysis, was 20 min. The effects of the neutron absorption by the nitrogen

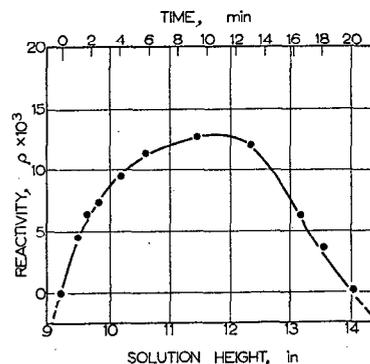


FIG. 3. Calculated reactivity in drum during radiation excursion.

and of the neutron reflection by the concrete floor, located approximately 3 in below the drum, were somewhat compensating and have been neglected. The bases for, and the results of, the above analysis are also not inconsistent with the following additional significant observations.

#### Records from radiation monitors

During the excursion a radiation detection instrument, sensitive to both neutrons and  $\gamma$ -rays, was operating some 1400 ft distant and

cross the wind from the scene of the accident. Fig. 4 is a reproduction of its output recorded during that time. The following discussion is based on a  $\times 7$  enlargement of this record, although most of the points are discernible on the reproductions shown here. The radiation intensity is observed first to have increased extremely rapidly from point (a), driving the pen off scale, to have decreased to point (b), and then to have repeated the sequence to point (c), all in about 15 sec as determined by the chart drive speed. During the next interval, the signal oscillated an indeterminate number of times, finally decreasing to about five times background 2.8 min after the first rise in level. The upper and lower limits of some of these pulses, discernible on the enlarged trace, are indicated by  $u$  and  $l$ , respectively. This (average) high-intensity field was then followed by a slowly decreasing level of some 18 min duration, again characterized by pulses. One peak, at 61 on the scale, is separated inordinately in time from adjacent portions of the trace and may be due to a momentary peculiarity of this detector, particularly since it is not readily identified on the charts from either of the air monitors referred to below. Although this neutron detector is equipped with two sensitivity ranges (25 and 125 mr/hr, full scale, respectively), it is believed to have remained on the more sensitive scale during the entire period, discounting the inference that some of the discontinuities are due to scale changes.

An enlargement, Fig. 5, of a section of the chart reproduced in Fig. 4 illustrates qualitatively the power pulses which occurred during the extended period of relatively lower activity.

The overall duration of the excursion is shown by this trace to have been 21 min. The absence of a strong neutron field within the drum as it initially became critical may mean that the critical height was reached prior to the initial energy release, that is, even though the system was critical, it did not manifest itself until it was "triggered" at a low power level, in a statistical manner, by ambient neutrons. This dormant period may have been a few tens of seconds, well within the accuracy of the above estimate.

Two additional radiation monitoring instruments were operating during the time of interest, both being air samplers which detect the  $\gamma$ -radiation from particulates collected on a filter surrounding a Geiger tube. Figs. 6 and 7 are copies of the records from these instruments. Each chart shows the direct radiation from the excursion and, subsequently, the arrival of the air-borne activity. The differences in the interval between the detection of these two activities at the two locations, about 12 min and 48 min, respectively, can be qualitatively correlated with the recorded wind direction at that time. The former was down-wind from the accident. The latter was located in an area adjacent to the site of the detector discussed above, i.e. cross wind from the accident, so the delay in the arrival of air-borne activity is expected to be comparable and equal to about  $\frac{3}{4}$  hr. This observation is presented as evidence favoring the interpretation of the extended, low-level activity shown in Figs. 4 and 5, being direct radiation. In addition, of course, Fig. 5 does not typify a radioactivity decay curve. No other quantitative interpretation is made of Figs. 6 and 7.

There are a number of undocumented observations made with portable radiation detection instruments in the vicinity of the accident to the effect that the radiation level remained constant for times of from 5 to 15 min, which is at least supporting evidence that the source of radiation was extended in time.

#### *Analysis of induced activity in the drum wall*

Activity was induced by neutrons in the components of the stainless steel of which the drum was constructed. Analyses of these activities yield at least relative values of the neutron exposure and, hence, of the neutron flux at various elevations along the side of the drum. The fast neutron measure was derived from the activity of  $\text{Co}^{58}$  arising in the  $\text{Ni}^{58}(n, p)\text{Co}^{58}$  reaction. The thermal neutrons were evaluated from the  $\text{Cr}^{51}$  activity from the  $\text{Cr}^{50}(n, \gamma)\text{Cr}^{51}$  reaction. (An analysis of the steel showed that it contained 17.99 and 11.84 wt % chromium and nickel, respectively.) The activation data are recorded in Table 2 and are

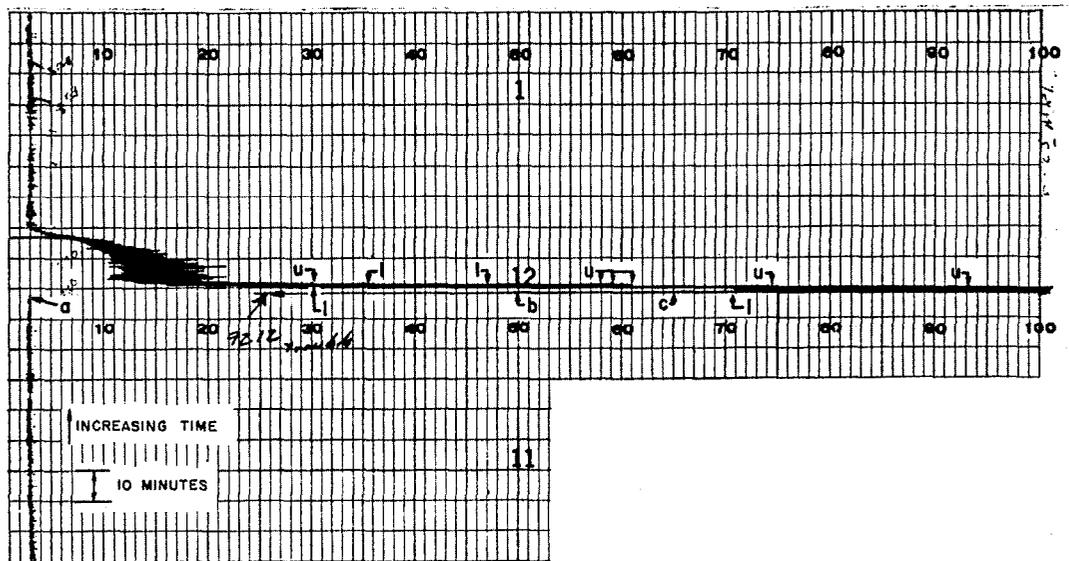


FIG. 4. Recorder chart from neutron detector showing direct radiation,

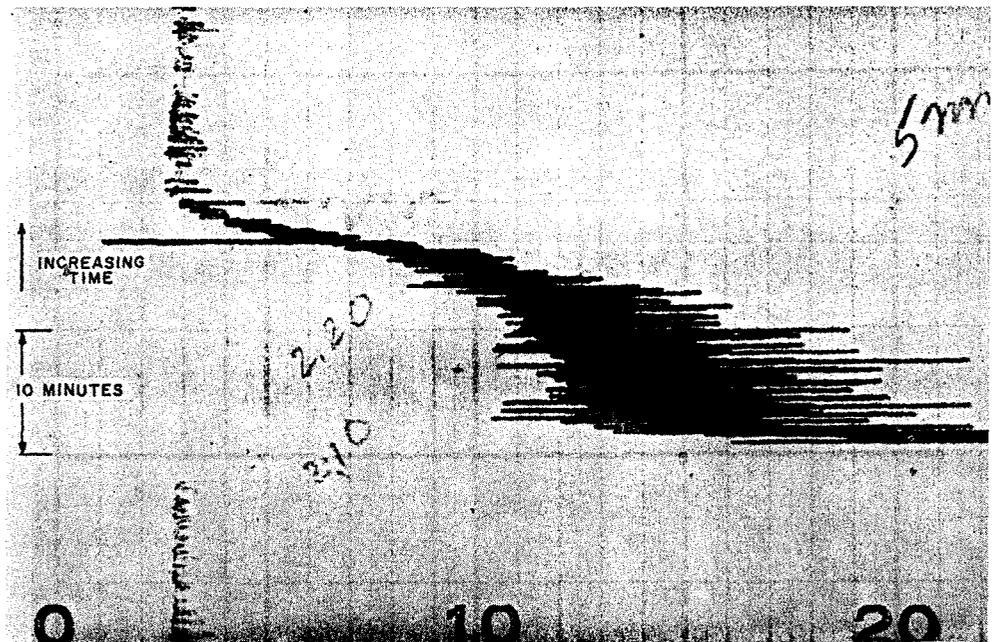


FIG. 5. Section of recorder chart of Fig. 4. Enlarged approximately four times.

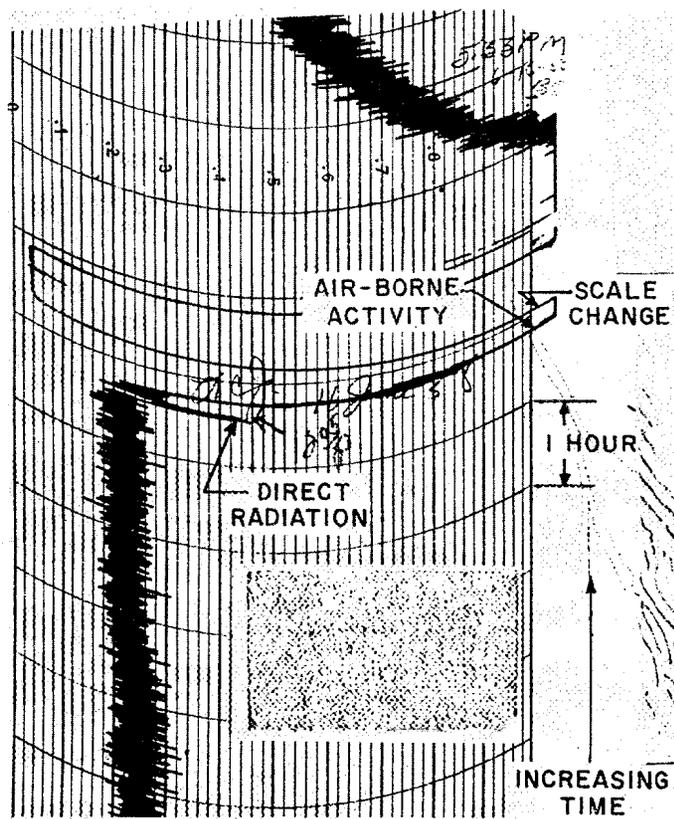


FIG. 6. Recorder chart from air sampler located down wind from accident.

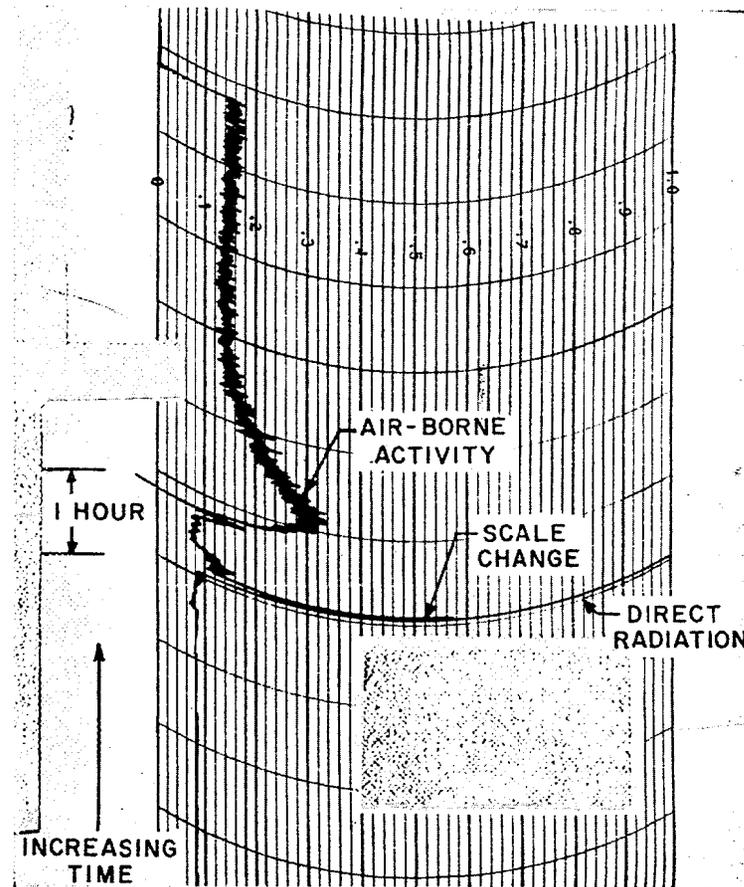


FIG. 7. Recorder from air sampler located crosswind from the accident.

Table 2. Relative activities of stainless steel samples from drum

Height from bottom of drum (in.)	Activation* (arbitrary units)	
	Thermal neutron	Fast neutron
15½	1.0	4.8
13½	1.1	4.9
11½	1.2	6.1
9½	2.1	9.1
7½	2.9	13
5½	3.8	14
3½	3.9	14
1½	3.8	11
center bottom	18	28

\* The values were obtained by  $\gamma$ -ray spectrometry; radiochemical analysis of three typical samples gave fast-neutron activations from 5 to 15 per cent lower.

plotted in Fig. 8. The results from additional samples from peripheral locations at three elevations show no significant asymmetry in the flux pattern in horizontal planes.

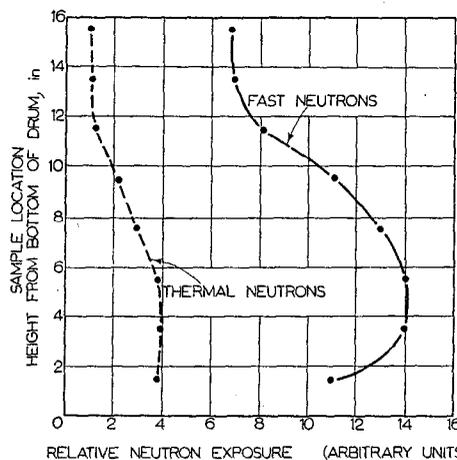


Fig. 8. Relative neutron exposure of samples from side of stainless steel drum.

It is interesting to note that the maximum activation occurred between 3 and 5 in. from the bottom, and that there is some evidence of asymmetry in the thermal neutron distribution, implying an effect of the stainless steel covered concrete floor as a reflector. If the peak activity is associated with some weighted center

of reactivity of the supercritical system, an effective reactor height of 10 in. is not inconsistent with the assumptions in the above analysis. No estimate of the energy in the excursion has been made from these values of the steel exposure.

#### Chemical and radiochemical analyses; energy release

The number of fissions which occurred during the power excursion, and hence the energy release, has been determined from radiochemical analyses of samples of the activated uranium solution. A sample of limited size was taken from the top of the liquid in the drum about 8 hr after the accident. Since this sample may not have been representative of the entire volume of the solution, a pair of samples was taken about 1 month later from the well-homogenized solution as it was then stored in shielded containers. It must be pointed out that some dilution of the solution occurred upon transfer from the drum to the storage containers which accounts for differences observed in the specific activities and the solution volumes. This, of course, in no way invalidates the method, provided the volume is measured at the time of sampling. From the concentration of appropriate fission products (obtained by measuring their characteristic radiation),

Table 3. Fission densities of solution determined by radiochemical analyses of samples  
Uranium concentration:\*

sample I, taken 8 hr after accident: 14.0 g of  $U^{235}$  per l.  
sample II, taken 1 month after accident: 9.6 g of  $U^{235}$  per l.  
sample III, taken 1 month after accident: 9.6 g of  $U^{235}$  per l.

Nuclide measured	Method†	Fission density (fissions/ml)		
		Sample I	Sample II	Sample III
$Mo^{99}$	$\beta$ count	$7.7 \times 10^{12}$		
$Ba^{140}$	$\beta$ count	$6.0 \times 10^{12}$	$2.8 \times 10^{12}$	$3.0 \times 10^{12}$
	$\gamma S^{\ddagger}$	$6.5 \times 10^{12}$		
$La^{140}$	$\gamma S^{\S}$	$4.6 \times 10^{12}$		
$Ba^{139}$	$\gamma S$	$2.2 \times 10^{12}$		
$Ce^{141}$	$\gamma S$		$5.8 \times 10^{12}$	$5.6 \times 10^{12}$
$Ce^{144}$	$\beta$ count**		$4.1 \times 10^{12}$	$4.0 \times 10^{12}$
$Zr^{95}$	$\gamma$ count		$3.5 \times 10^{12}$	$3.6 \times 10^{12}$
$Cs^{137}$	$\gamma S$		$0.6 \times 10^{12}$	$0.6 \times 10^{12}$
$Sr^{89}$	$\beta$ count		$0.5 \times 10^{12}$	$0.5 \times 10^{12}$
Weighted "best value"		$7 \times 10^{12}$	$5 \times 10^{12}$	$5 \times 10^{12}$

\* From chemical analyses.

† The activities were measured by  $\beta$ - or  $\gamma$ -ray counting ( $\beta$  count or  $\gamma$  count) or by scintillation spectrometry ( $\gamma S$ ).

‡ Assuming 21.5 per cent of the disintegrations yield 0.54 MeV  $\gamma$ -rays, which is based on unpublished data of LYON, Oak Ridge National Laboratory.

§ After several hours growth in separated barium.

\*\* The  $Pr^{144}$  beta particles were measured through an aluminum absorber ( $104 \text{ mg/cm}^2$ ) used to reduce the Ce beta particles. The presence of this absorber necessitated a 48 per cent correction to obtain the  $Pr^{144}$  beta yield.

Table 4. Estimates of energy release during accident

	Based on sample I	Based on samples II and III
Volume of solution yielding sample (l.)	180	252.8
Mass of $U^{235}$ in total volume (kg)	2.5	2.4
Total number of fissions	$1.3 \times 10^{18}$	$1.3 \times 10^{18}$
Energy release	$2.6 \times 10^{20} \text{ MeV} = 11 \text{ kWh}$	

together with their decay constants and fission yields and the elapsed time between the excursion and the analysis, the number of fissions which occurred per unit volume of the solution was obtained. All of the analytical results and a weighted "best value" of the energy released in the excursion,  $2.6 \times 10^{20}$  MeV from  $1.3 \times 10^{18}$  fissions, are given in Tables 3 and 4.

It will be noted that large discrepancies exist in the data of Table 3. A partial explanation lies in the existence of noble-gas precursors of most of the nuclides measured in the analysis. A list of these precursors is given in Table 5.

Table 5. Properties of fission product nuclides measured in analysis of solution

Nuclide	Fission yield fraction <sup>(4)</sup>	Gas precursor
Sr <sup>89</sup>	0.048	3.2 min Kr
Zr <sup>95</sup>	0.064	"short" Kr
Mo <sup>99</sup>	0.062	—
Cs <sup>137</sup>	0.059	3.9 min Xe
Ba <sup>139</sup>	0.063	41 sec Xe
Ba <sup>140</sup>	0.061	16 sec Xe
Ce <sup>141</sup>	0.060	1.7 sec Xe
Ce <sup>144</sup>	0.061	~1 sec Xe

Gases of longer half-lives obviously have higher escape probabilities from the liquid than those of short half-lives. Further confirmation of this explanation is obtained from observations on samples of solutions in which the fission concentrations have varied; the difference between the fission concentration values derived from Ba<sup>139</sup> and Mo<sup>99</sup> increases with increasing concentration, i.e. increasing heat output. The apparently low values of the fission concentration in the latter sample, based on Zr<sup>95</sup> and Ba<sup>140</sup>, reported in Table 3, may be explained by the well-known hydrolytic behavior of zirconium and possible similar loss of barium due to traces of sulfate (in addition to the loss of 16 sec Xe<sup>140</sup>). Disagreements between values from Ce<sup>141</sup> and Ce<sup>144</sup> have not been explained.

#### Hydraulic reconstruction experiments

Considerable effort was expended in attempts to reconstruct the flow patterns of the several

volumes of liquids as they were added separately to a somewhat complex system of piping, partly mixed therein, and finally drained into the 55 gal drum in a stream of variable uranium concentration. An aqueous solution of cadmium nitrate, adjusted in concentration to approximate the fluid properties of the mislocated uranium solution, together with the volume of water believed appropriate, were used in these tests. Flow rates into the drum were measured and frequent samples were obtained both from the top of the liquid in the drum and from the line as the drum was filled. Although, in principle, the analyses of these samples allow an estimation of the uranium inventory and concentration in the drum as a function of time, it is not certain they are truly representative of the conditions in the drum at the time of the accident. This uncertainty may be due, for example, to irreproducible mixing conditions, particularly since the first emission of nuclear energy caused at least local turbulence. The fill rate was used in the above reactivity analysis, but it has not been possible to correlate the time-uranium inventory data with the uranium concentrations required for criticality.

#### General observations

There are two additional observations which should be recorded for consideration. One of them is the absence of a strong ambient neutron field at the scene of the accident (the most likely source being the  $O(\alpha, n)Ne$  reaction between the U<sup>234</sup>  $\alpha$ -particles and the oxygen in the water) and, as a consequence, the system may have been above delayed critical before the power level increased from zero.

The second observation is that there was no evidence of the rapid production of large quantities of gas or vapor. There was, for example, no liquid on the floor under or adjacent to the drum, nor was there an inordinate amount of localized fission product contamination on the fill tube (see Fig. 1) except where it was in contact with the liquid. The nature of the process in the area precluded any meaningful  $\alpha$ -particle contamination survey for dispersed uranium. These observations minimize any assumption of vigorous boiling of

the solution. There is no clear explanation of why the solution was not dispersed outside the drum, although speculation can relate the violence of the turbulence to the rate and mode of the approach to critical, to the characteristics of the first power surge, and possibly, to the geometry of the vessel. Comparison of experiences with other critical accidents<sup>(1)</sup> with solutions shows that large as well as insignificant discharges of liquid have been observed in events with the same energy release.

#### DISCUSSION

An attempt has been made in the preceding paragraphs to record and interpret a rather wide variety of observations made in connection with the radiation accident. It is believed, unquestionably, that sufficient enriched uranium solution was added to a 55 gal drum to become critical, that the concomitant energy release occurred during an interval of a few minutes in which the effective reactivity and the power level oscillated a number of times, and that the chain reaction was ultimately stopped by the addition of water to the solution (since, very fortunately the valve, through which the solution was admitted, was left open as personnel evacuated the area). The quantity of uranium involved and the energy developed in the reaction are moderately well known; the uncertainty in the duration of the excursion and the fluctuation in the reactivity have not allowed an evaluation of the peak power. The potential personnel hazard from the ionizing radiation generated in the observed number of fissions is developed elsewhere in this series of papers and is compared with the exposures experienced by employees in the vicinity of the accident.

As pointed out earlier, it is impossible to reconstruct the reactivity-time pattern and there are, no doubt, several combinations of events which can account for the observations. It is intended to outline very briefly here one possible sequence.

With reference to the power-level relation, indicated by the radiation monitor record described in Fig. 4, the following sequence of conditions is suggested. In the absence of a source of neutrons, this system was prompt

critical before any energy was emitted. Once started, however, the power level rose quite rapidly to a high value. The energy from these fissions produced gases by dissociation,<sup>(5)</sup> reducing the density and driving the solution subcritical. Exit of these gas bubbles once more made the system prompt critical and, with the delayed neutrons as a source, the power level again rose. This cycling persisted for an estimated 2.8 min, during which, of course, the temperature of the solution increased. Boiling\* finally ensued, causing a sharp decrease in density and a concomitant return to subcritical indicated by the decrease in the instrument deflection to about scale reading "20," (Fig. 4). Following this steep descent, the system settled into an equilibrium condition somewhere in the delayed critical range where it was controlled for about 18 min by vapor formation and, to a lesser extent, by decomposition gases. The system remained delayed critical until the inflow of water reduced the concentration to a final subcritical value.

In previous experiences with accidental critical assemblies,<sup>(1)</sup> which have been limited to a single burst by some reactor shutdown mechanism, the energy release has been from  $10^{16}$  to  $10^{17}$  fissions, a not unreasonable estimate of the first of the several pulses in this case.

It is appropriate to consider, briefly, other courses the reaction may have taken and the consequences which could have resulted. For example, one shutdown mechanism for a supercritical solution, alternate to a dilution, is the removal of sufficient water to increase the chemical concentration beyond that which will support a nuclear chain reaction under the other existing conditions. This removal would be by dissociation and vaporization. In this particular instance, the above analysis shows (Fig. 2) the limiting concentration to be about 54 g of  $U^{235}$  per l. with a total of 2.5 kg of  $U^{235}$ , a value, incidentally, not much different from that of the original solution. Had no

\* The permanent deformation of a polyethylene liner, present in the drum during the excursion, into the convolutions of the drum is evidence that the temperature of the solution at least approached the boiling point. The energy release obtained from the fission product analyses was adequate to boil the solution.

water been added in the operation, the excursion might not have been as severe as the one experienced.

Another shutdown mechanism is a dispersal of the fissionable material, the causes of which are difficult to predict from past experience.

It is believed that the incident described here is a point of departure for predicting the causes and effects of possible future accidents. It does not set an upper limit to the consequence to be expected for, as pointed out above, there were associated with it a number of unique, fortunate circumstances which reduced the problem significantly. A study of this type of accident has been made,<sup>(2)</sup> which is supported in part by the findings reported here, and which, in the absence of externally applied shutdown mechanisms, predicts much more severe results.

*Acknowledgments*—The co-operation of many persons in making the study of the radiation excursion is gratefully acknowledged. Particular mention must be made of helpful discussions with F. S. PATTON and G. R. JASNY of the Y-12 Plant in Oak Ridge and with J. R. KNIGHT and J. C. BAILEY of the Oak Ridge Gaseous Diffusion Plant. The radiochemical analyses were made under the direction of S. A. REYNOLDS and E. I. WYATT of the Analytical Chemistry Division of the Oak Ridge National Laboratory.

#### APPENDIX

##### *Method of calculating reactivity*

The critical mass in an unreflected 21.75 in. diameter stainless steel cylinder was determined as a function of the critical volume by equating its geometric buckling to that of a similar cylinder 20 in. in diameter for which the critical parameters are known. Once the variation of critical mass with critical volume of the larger cylinder is known, an initial critical point on the curve, commensurate with facts observed after the excursion, is chosen. This point, *A*, in Fig. 9, represents 2.1 kg of  $U^{235}$  in 56.2 l. of solution. The subsequent masses and volumes, as additional solution enters the drum, are represented by line *AB*, assuming that the concentration of the incoming solution remains constant. It is further assumed that the final contents are 2.5 kg of  $U^{235}$  in 180 l.

In a two neutron-energy group analysis, the effective reactor multiplication factor, *k*, of critical and near critical assemblies is related

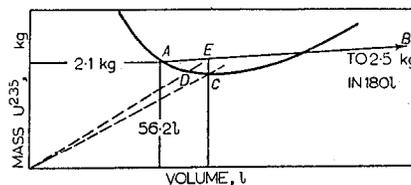


FIG. 9. Schematic diagram of critical mass in an unreflected 21.75 in. diameter stainless steel cylinder as a function of critical volume.

to the material and geometric properties of the assembly by

$$k = \frac{\eta f}{(1 + L^2 B^2)(1 + \tau B^2)}$$

where  $\eta$  = number of fission neutrons produced per neutron absorbed by  $U^{235}$

$f$  = thermal neutron utilization

$L^2$  = square of the thermal diffusion length

$B^2$  = geometric buckling of the reactor

$\tau$  = neutron age

Along the critical curve in Fig. 9, the equation has the value unity, of course, and the geometric and material buckling are equal.

As the cylinder continues to fill, the mass and volume increase to point *E* which describes a different (supercritical) combination of geometry and material. The nuclear properties of the latter are the same as those of the solution critical at point *D*, since a line through the origin represents a particular chemical concentration, and the values of  $\eta f$  at *D* and *E* are, therefore, equal. Since the geometric buckling at conditions *C* and *E* are the same and  $L^2$  and  $\tau$  are essentially constant over this concentration range, the multiplication constant at *E* is given by:

$$k_E = \frac{(\eta f)_D}{(\eta f)_C}$$

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\* This and similar designations refer to U.S. Atomic Energy Commission Research and Development Reports available from the Office of Technical Services, U.S. Department of Commerce, Washington 25, D.C.