

OAK RIDGE NATIONAL LABORATORY operated by UNION CARBIDE CORPORATION NUCLEAR DIVISION for the **U.S. ATOMIC ENERGY COMMISSION**

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JANUARY 30, 1968

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JUNE 1968

OAK RIDGE NATIONAL LABORATORY Oak Ridge, Tennessee operated by UNION CARBIDE CORPORATION FOR THE U. S. ATOMIC ENERGY COMMISSION Contract No. W-7405-eng-26

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ABSTRACT

A volume of an aqueous solution of uranyl nitrate, in which the 233 U content of the uranium was 97.6%, was inadvertently made super prompt critical in the Oak Ridge Critical Experiments Facility. The fission yield was 1.1 x 10^{16} . There was neither internal nor external measurable exposure of any individual to radiation; there was no property damage or material loss. Fission products decayed sufficiently within twenty-four hours to allow unrestricted occupancy of the affected room. Solution containing an estimated 15 g of uranium was spilled into the reflector-water tank as the result of a dislodged rubber stopper.

INTRODUCTION

Unexpected criticality was achieved in a volume of an aqueous solution of a salt of ²³³U during a series of routine critical experiments in progress in a well-shielded assembly area of the Oak Ridge Critical Experiments Facility, Building 9213, Y-12, at 10 a.m., Tuesday, January 30, 1968. The criticality-radiation alarm system functioned as designed, the evacuation of personnel from the building was prompt and orderly, and the excursion was terminated expeditiously by a negative coefficient of reactivity and was prevented from recurring by the action of the safety devices. The fission yield was 1.1 x 10¹⁶. Gamma-ray sensitive personnel dosimeters read immediately following the excursion showed no direct exposure greater than 5 mr to any person present, a result confirmed by reading processed film from the security badges of the three individuals located in the control room adjacent to the assembly area. Bioassay of samples from these three individuals as well as from other occupants of the Facility showed no internal exposure above tolerance. Results from the threshold detector unit in the control room also showed no dose equivalent greater than the detector sensitivity. There was no property damage or loss of fissile materials. The solution and the atmosphere within the test area were contaminated with short-lived fission products which decayed within 24 hours to a level permitting free access to the affected area.

The purpose of the experiment in progress was to establish the critical concentration of a sphere of the solution of uranyl nitrate surrounded by a thick water reflector.

EQUIPMENT AND MATERIALS

The spherical container for the solution was of thin aluminum and was mounted near the center of a cylindrical water container. The sphere was 22.3 cm in diameter and had a capacity of 5.84 liters; the cylinder was 91 cm in diameter and 132 cm high. Water at least 20 cm thick surrounded the aluminum sphere. As shown on the sketch of the equipment, page 4, both the solution and the water systems, in addition to the above mentioned sphere and cylinder, were comprised of a storage vessel, a line of large diameter in which was located a normally open valve caused to open upon high radiation signal, a pump, and a remotely operable normally closed valve in the line connecting the pump with the vessel. An auxiliary, normally open, remotely operable drain valve and line connected the sphere and the solution storage vessel. The solution storage vessel and the test vessel were also connected by a flexible tube lightly attached to the top of the sphere and to the storage. Air was transferred through this connection as the liquid levels were simultaneously changed, thereby successfully reducing α -particle contamination of the room.

Attached to the solution system was a device to facilitate fine adjustment of the quantity of solution in the sphere. This device was simply a 5-cm-diam pipe, with axis vertical, mounted on a drive. In normal operation the usual solution supply system, i.e., the pump and feed control valve, designated as B on the sketch, were turned off and closed, respectively, and the auxiliary pipe raised or lowered. Solution then flowed to or from the sphere through a flexible transparent tube.

The level of solution in the sphere was indicated by the corresponding level in an adjacent sight glass, with transmission of information to the control point by a level-seeking photocell with selsyn indicators. The water level was indicated by an ordinary sight glass. A thermocouple near the center of the sphere was connected to a recorder in the control area.

The solution was aqueous uranyl nitrate in which the uranium contained 97.6% ²³³U. The uranium concentration was 167 g/liter and the specific gravity was 1.23.

A more complete description of equipment typical of that outlined above together with characteristic performance, experience, exemplifying

FLEXIBLE TUBING 91 SCREW DRIVE . . 0 3.8 GLASS DIA.FINE ADJUSTMENT PIPE and the big of the big NORMALLY CLOSED TESPHERE, SIGHT 132 n WATER WATER . REFLECTOR SOLUTION 2.5 3 FROM E 7 PUMP FLEXIBLE TUBING 1.30 0 B WATER DUMP VALVE NORMALLY OPEN 7.8 DIA. CLOSED TO WATER STORAGE ן ה DRAIN VALVE C REMALLY SOLUTION. DUMP VALVE NORMALLY OPEN 7.8 DIA. 1070 $\boldsymbol{\lambda}$ SYMBOLS 501 53 E -174 -VALVE, REMOTELY OPERATED 10 20 JOLAND 50 X PUMP

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SCHEMATIC OF SYSTEM FOR SOLUTION CRITICAL EXPERIMENT

procedures, and actions put into effect upon emergency are given in Ref. 1. The available rate of adding liquid into test vessels at the time of the excursion was 150 cm³/sec, maximum. (It is pointed out that this is the rate were the solution feed value to remain open continuously.) The solution removal rate was eight times as great, or 1200 cm³/sec. From the volume coefficient of reactivity under the conditions of the excursion, 1.5 cents/cm³, the maximum reactivity addition rate was 225 cents/sec. The response time of the value for solution removal was 220 msec.

^{1. &}lt;u>A Safety Analysis of the Oak Ridge Critical Experiments Facility</u>, ORNL-TM-349 Rev. 1 (1967). The excursion occurred in the room designated as South Assembly in Fig. 2 of this reference.

THE EXPERIMENT

Criticality of the sphere on the preceding day was apparently achieved only by overfilling into a small cylindrical tube attached to the top of the sphere. Although 1.5 cents/cm³ had been measured as the volume coefficient of reactivity, there remained an uncertainty in the actual height of the solution in the tube." The purpose of the measurement in progress at the time of the excursion was to establish the critical solution height and to evaluate the reactivity of the perturbation introduced by the short column of liquid above the sphere. In the procedure to accomplish this purpose the sphere was made slightly supercritical, the feed valve was closed, and the pump stopped. An attempt to remove solution into the small fine-adjustment pipe did not give the expected response, so solution was drained from the sphere and subcriticality was attained. Removal of air, visually observed from the control point to be trapped in the flexible line to the fine-adjustment pipe, was attempted by again draining solution to storage (the reactivity decreased), then raising the small pipe (the reactivity increased). The responses to these actions were normal. This sequence was repeated at least twice. At a time when no adjustments were being knowingly made, the reactivity increased rapidly, all the shutdown devices functioned, and the radiation alarm sounded.

^a For the benefit of readers unfamilar with the operation of critical experiments, it is pointed out that there is no safety significance to the absence of knowledge of an absolute dimension of an assembly.

POST EXCURSION FINDINGS

Normal occupancy of the entire building except the affected assembly area and the adjacent control room was re-established immediately upon completion, within an hour, of a health physics survey.

Extensive investigation of the experimental area was postponed until 22 hours after the excursion, a convenient time in the normal work schedule, when the air-borne activity was below tolerance and the contamination of the floor in the assembly area averaged 300 α and 10,000 β - γ disintegrations per minute over a 100 cm² smear sample.

The rubber stopper, denoted by A in the sketch, at the connection between the top of the sphere spout and the return line to storage, was observed to have tipped (not completely removed) sufficiently to allow liquid to be expelled. The tubing above the stopper contained solution. Vapor generated within the sphere no doubt displaced solution into this line, dislocated the stopper, and forced some solution out of the system. Some of this solution fell into the reflector water as it drained, for, in fact, a sample analysis showed eight parts of uranium per million of water. Since the reflector-water system contained 600 liters, five g of uranium, or 30 cm³ of solution probably spilled into the reflector as it drained. Additional solution spattered over the neck of the sphere, onto the water reflector tank, and onto a limited area of the platform supporting the tank. On the sphere and in the tank it appeared as drops that had dried. Contamination outside the tank was detectable only by its radiation. All of this dispersed liquid contained an additional estimated 10 g, which was recovered by a dilute acid wash.

Radiochemical analysis of a sample of the solution showed 5.6×10^{11} fissions per cm³, extrapolated to 10 a.m., January 30. Analysis was made of ⁹⁹Mo and ⁹²Sr taking 4.8% and 6.5% as the respective fission yields.² The volume of solution in the system was ~20 liters giving a yield of 1.1×10^{16} fissions, which indicates that prompt criticality was achieved.

2. E. I. Wyatt, ORNL, personal communication (1968).

The recorded signal from a thermocouple located at the center of the sphere showed a minimum temperature of 50° C during the excursion, an increase of 26° . (Although the recorder pen pegged at 50° C, the pen-return trace showed that the actual temperature was likely not much greater.) The spatial power distribution, the mass, and the specific heat of the solution gave an estimated energy release of the order 10^{16} fissions.

Results from the threshold detector unit in the assembly area 16 ft from the ²³³U solution showed a dose equivalent of less than 50 mrem due to low energy neutrons. There was no detectable activity in the fast neutron sensors. These results are consistent with the presence of the thick water reflector.

CAUSE OF THE EXCURSION

The cause of the excursion is related to the dislodgment of the air bubble from the flexible connection of the fine-adjustment device and to a resulting transient displacement of solution into the sphere. For example, inertial effects in the solution volume resulting from draining could have set up oscillations in the liquid in the flexible connection containing the entrapped air. Such oscillations could result in a transient in the volume contained in the sphere. Alternatively, the bubble might have moved toward the vertical connection to the sphere thereby forcing an increment of solution into the sphere. Either of these postulates is a mechanism whereby reactivity could be added to the sphere. By visual observation from the control point the volume of the air bubble in the tube was estimated to be 30 cm³. This volume of solution would have added about 45 cents reactivity allowing for hydrostatic pressure differences whereas 67 cm³ was necessary to increase the reactivity from delayed to prompt criticality. Since the sphere was definitely subcritical immediately before the effort to remove the bubble, a volume even greater than 67 cm³ was required for prompt criticality. An additional source of solution, and of reactivity, could have been a flow from the

fine-adjustment tube following breakage of an air lock established in the flexible tube by the bubble. Since the capacity of a 1-in.-long section of the fine-adjustment pipe is 47 cm³, an "out-of-level" condition of as little as 1 in. could provide more than the necessary additional re-activity to cause the excursion.

A supplemental cause was the failure, in manipulations for bubble removal, to drain sufficient solution to offset the postulated transient addition. Although the procedure followed was customary, reasonable and acceptable, in retrospect it would have been prudent to abort the experiment instead of attempting to remove the bubble.

It is emphasized that any quantitative discussion of the excursion is highly speculative because information necessary to such an analysis, such as the amount of reactivity added, the rate of its addition, and even the reactivity of the sphere before the addition are not known. Further, the information is unattainable except through some type of repeat experiment. Any result would then be descriptive of a single unique situation. These remarks have, however, provided a reasonable explanation. Any more extensive or detailed investigation cannot be justified by the scant return of technically valuable information.

REMEDIAL MEASURES

Since movement of entrapped air is postulated to be the cause of the excursion, consideration was given to the source and means of eliminating it. The solution feed valve, B in the sketch, was located 94 cm above the tee connection for a bypass line to storage, point C in the sketch. Upon closure of B in the stepwise process of adding solution to the sphere, drainage of the solution from this section of 1.9-cm-diam pipe separating valve B from point C could occur. The air thereby entrapped would flow toward the sphere upon subsequent opening of B for the next solution addition. Valve B has now been lowered to a position immediately adjacent to the tee. No entrapped air has been observed in the lines since that change was made.

INCIDENTAL OBSERVATIONS

The audible alarm was silenced after serving its purpose by initiating It was, however, automatically reactivated about 15 minutes evacuation. after the first signal. Immediate inspection of the continuous records of operational instrumentation associated with the 233U experiment, as contrasted to alarm instrumentation, showed a steadily decreasing ambient radiation field. Additional surveys identified no other source. The second alarm was, therefore, spurious and is believed to have been caused in the following way. Each of the five monitor stations within the Facility is comprised of three detectors, each with associated circuitry including a meter relay making contact at 75% full scale deflection. Contact of two relays at one station is required for an alarm (two-outof-three coincidence). A characteristic of the circuitry requires that reactivation of the alarm, once silenced, can occur only if the meter relay contacts have been first broken. In this instance, as the field at the detectors nearest the ²³³U experiment decayed to the alarm trip point, the meter relays chattered, thereby again fulfilling the conditions for producing an alarm.

It is interesting to note that one person located in the building 110 ft from the source heard the response of his portable Personal Radiation Monitor to the pulse of radiation before the area alarm sounded, an experience comparable with that during the purposeful production of pulses by the Army Pulse Radiation Facility Reactor at ORNL in 1967. The operators of the experiment under discussion reported action of shutdown devices also before the alarm.

Two additional items, of only local interest and importance, were considered and corrected as a result of the experience with the 233 U solution. The operation of the radio transceivers located at the emergency assembly stations was undependable and they have been replaced by Bell telephones. The second item is a change in the emergency alarm circuitry whereby the signal to a traffic control light can be deactivated independently of the signal to the audible alarms. Both of these changes are recorded in Ref. 1.

SUMMARY

About 5.8 liters of aqueous uranyl nitrate solution, in which the uranium contained 97.6% ³³³U, was made super prompt critical in the Oak Ridge Critical Experiments Facility producing 1.1×10^{16} fissions. There was no measurable personnel exposure, no property damage, and no loss of fissile material. About 15 g of uranium, spilled from the system through a loosened joint, was recovered. After two weeks the natural decay of the fission products produced in the solution had reduced their contribution to the activity of the solution to only 20% of the normal background. During this decay period the program of experiments was continued with other solution available in the Facility. Effort totalling 13 man-days was required for decontamination, substitution of solution, and for the equipment changes described.

The primary cause of the excursion is identified as an accumulation of air in a line auxiliary to the main flow system. The secondary cause was a manipulation removing the air. Changes correcting the primary cause have been effected. Observation of less important matters has led to additional improvements in overall operations.

That the real losses and consequences of the occurrence are far less than those of many industrial accidents attests to the adequacy of the Facility and its staff for events of this type.

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