

# TRANSACTIONS

OF THE  
AMERICAN NUCLEAR SOCIETY

June 3-7, 1984  
The New Orleans Hilton and Towers  
New Orleans, Louisiana

Volume 46  
TANSO 46 1-877 (1984)  
ISSN: 0003-018X

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2. G. PENNELLE, "Description et Analyse de l'Accident de Criticité survenu au Reacteur VENUS à Mol en date du 30 Décembre 1965," *Proc. First Int. Congress Radiation Protection*, Rome, Italy, 1966, p. 1223.
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6. T. G. HUGHES, "Criticality Incident at Windscale," *Nucl. Eng. Int.*, p. 95 (Feb. 1972).

### 5. The Hanford Pulsar Accident, E. D. Clayton (PNL)

Eight criticality accidents are known to have occurred within the United States in operations external to reactors, excluding experimental systems where the intent was the study or measurement of criticality itself.<sup>1</sup>

In the realm of criticality accident experience, the Hanford accident of April 7, 1962, remains one of the more interesting and complex of any to date. Since details have

never been broadly circulated, the accident will be reviewed for the lessons gained. This accident, and subsequent recovery operations, were unusual for the following reasons:

1. The reaction continued for ~37 h before termination, whereas criticality accidents normally terminate within periods ranging from seconds to minutes. No super critical chain reaction had remained uncontrolled for this long before. (Excluding criticality that occurred in the earth in the Republic of Gabonaise in primeval time).<sup>2</sup>
2. There was no spread of contamination.
3. There was no physical damage.
4. There was no serious radiation dosage to any staff member.
5. A remotely controlled mechanical robot was used for the first time in the aftermath of such an accident to perform various operations, position detectors, conduct surveys, turn valves, etc. (see Fig. 1).
6. *In situ* multiplication data were obtained on the solution in the vessel in a reverse approach-to-criticality during the recovery operations, which led to interesting conclusions regarding the shutdown mechanism.
7. The method of shutdown was not the result of simple evaporation or boiling off of solution by itself from fission heating, nor of expulsion of solution from the vessel.
8. Both administrative errors and mechanical failures combined in a sequence of events leading to criticality.

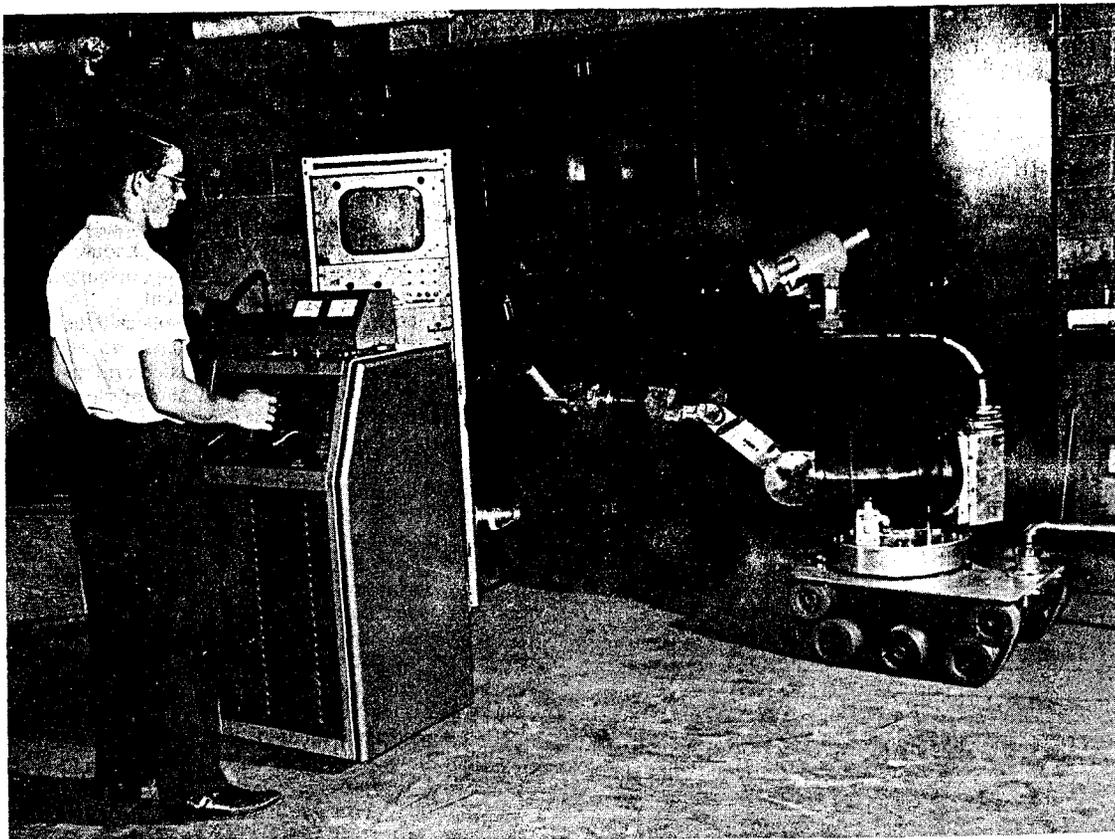


Fig. 1 Remotely controlled mechanical robot used in plant recovery operation.

9. Some mystery remained concerning final cause of the accident. The final reason given by the investigation committee was the operation of a valve "contrary to oral instructions," . . . "no one admits to having operated the valve but no other plausible explanation is available."<sup>3</sup>

The location of the accident was in the Recuplex (chemical plutonium scrap recovery) portion of the 234-5 building, in one of the waste cleaning hoods. Operators reported seeing a "blue flash," criticality alarms sounded and the plant was evacuated promptly. Three men received overexposures of radiation: 110, 45, and 19 rem. The highest radiation dose was lower, however, than a level at which clinically observable effects would be expected.

The source of the excursion was a 69-ℓ cylindrical glass tank with a 17.5-in. i.d. normally used for transfer of dilute side stream from solvent-extraction columns. The unit became critical when ~46 ℓ of aqueous plutonium nitrate solution containing some 1500 g Pu at ~33 g Pu/ℓ including ~1 ℓ of light organic solution was inadvertently transferred into the vessel. The total yield of  $8.2 \times 10^{17}$  fissions was distributed over some 37 h with ~20% of the fissions appearing in the first half hour. Radiation monitoring of the area around the building following evacuation showed the presence of both neutron and gamma radiation, indicating a continuing low-level fission chain reaction. Reconstruction of events indicated that an initial spike of  $\sim 10^{16}$  fissions occurred followed by smaller spikes (pulsing).

*In situ* multiplication measurements made on the vessel during draining showed that criticality could not have been reinitiated through the simple process of adding water to the vessel containing the plutonium.<sup>4</sup> There was an apparent deficiency in plutonium of ~140 to 150 g. (The missing plutonium was later located.)

It is postulated that the reaction went critical with a small amount of light organic phase (~1 ℓ) on top of the aqueous solution. The reaction terminated itself as a consequence of (a) plutonium mass transfer from the aqueous to the TBP-DBP phase; as the density of the organic increased above the aqueous density, it would have settled to the bottom of the vessel and also (b) through aqueous evaporation. (However, the mass transfer and subsequent settling would have reduced the concentration to below the critical point, irrespective of evaporation.)

In summary, the accident occurred because product solution had overflowed to the hood sump coincident with an organic leak. A temporary line to the sump that was used for cleanup had not been removed following its use. A valve was operated "contrary to oral instructions" that resulted in a sufficient quantity of plutonium entering the transfer vessel from the sump via this line to initiate criticality. In this case, there were two administrative errors and two equipment malfunctions, at least three of which had to exist simultaneously.

This accident serves to illustrate the seriousness accorded criticality accidents. Even though there may be no physical consequences of the accident itself, the plant recovery operation and investigative process can be detailed and time consuming.

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2. R. NAUDET, "Rapport de Synthèse sur le Phénomène D'Oklo," *Bull. Inf. Sci. Tech.*, No. 193 (1974).
3. R. F. NOWAKOWSKI, "4 Men Exposed to Radiation a Year Ago Show No Effects," *Tri-City Herald* (Apr. 7, 1963).
4. E. D. CLAYTON, "Further Considerations of Criticality in Recuplex and Possible Shutdown Mechanisms," HW-77780, General Electric Co. (May 31, 1963).

## 6. The 1963 Livermore Criticality Accident Revisited, R. L. Kathren (PNL)

On March 25, 1963, at approximately midnight, an unplanned nuclear excursion occurred in a beryllium and polyethylene reflected enriched metallic uranium assembly during an experimental approach to criticality in a shielded concrete vault. Even after two decades it is instructive to examine this accident again as it resulted in several unique experiences of interest and importance to those concerned with nuclear criticality safety.

The assembly in which the accident occurred consisted of three nested concentric hollow cylinders of 93% enriched uranium metal into which a solid cylinder (a "ram") of similar material was inserted from below to achieve increased multiplication. This operation was accomplished remotely by personnel at a control console outside the vault. The accident was believed to have resulted from a slight misalignment of the parts, which caused the ram to raise the innermost hollow cylinder as it was inserted. When the hollow cylinder fell back down into position around the ram, a prompt criticality burst ensued. The yield, calculated from fission product activity, was  $3.76 \times 10^{17}$  fissions, equivalent in explosive force to ~6 lb of TNT.

The  $2.6 \times 10^6$  calories produced by the excursion were sufficient to cause the uranium to ignite; an estimated 15 of the 47 kg in the assembly were oxidized and an estimated 10 kg melted and flowed over the floor of the vault before resolidifying. Nearby combustibles did not burn or scorch although some of the polyethylene reflector melted. Physical damage was generally limited to the assembly and its tubular aluminum support apparatus, including two polyethylene moderated ionization chambers, which were knocked to the floor.

Relatively little radioactivity was released from the vault, which was not a sealed containment structure, although air monitoring instruments in nearby buildings detected both the prompt gamma pulse and subsequently showed significantly increased air concentrations of particulate radioactivity, suggestive of an external plume that seemed to linger in the vicinity of the building largely because of highly stable atmospheric conditions. Particulate activity 350 m north of the building housing the vault was  $1.3 \times 10^9$   $\mu\text{Ci}/\text{cm}^3$  at ~1 h after the event, and about an order of magnitude below the first measurements of airborne activity made in the vault, ~5 h after the accident.

Off-site environmental samples collected as early as a few hours after the accident revealed no significant or detectable fission product activity. As determined from film badges, the maximum exposure to any of the four persons in the building at the time of event was 120 mrem from penetrating photons; neutron exposures were less than the minimum detectable levels of 50 mrem (fast) and 1 mrem (thermal). The small exposures observed may not have resulted wholly or even partially from the accident as the badges had been worn several days prior to the event. Urinalyses, nasal wipes, and thyroid scans were all negative, suggesting no internal exposure. Protection of personnel was clearly provided by the shielded vault, which, although not sealed, also minimized fission product release to the environment.

The excursion left a secondary criticality problem in the form of the three hollow uranium cylinders, which had fused into a single mass estimated to contain 23 kg of enriched uranium. The degree of oxidation of the uranium was unknown, and the problem was further complicated by the presence of an external sleeve of the beryllium reflector that was fixed into position. The large mass of uranium was subsequently removed to a hot cell facility where the beryllium sleeve was removed and the remaining mass of