

FIG. 1. FLOW SHEET for plant in which accidental criticality occurred. Portion involved is at right

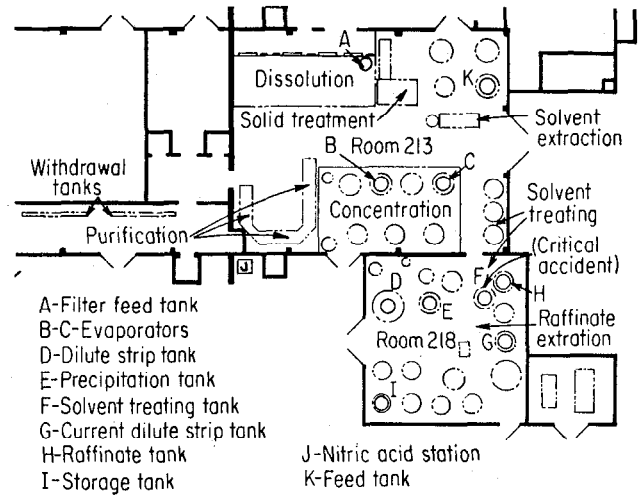


FIG. 2. PLANT LAYOUT shows relation of different processing areas and tanks set to receive near-critical liquids



Los Alamos Criticality Accident, December 30, 1958

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EDITORS' NOTE: A sudden criticality in a processing vessel at the Los Alamos plutonium-processing plant occurred on December 30. As a result a technician died 36 hr later (*NU*, Feb. '59, p. 21). **NUCLEONICS** presents here a report of the official investigation of the accident. The report tells that the accident occurred during an inventory in which normal plant flow was interrupted. An unusual amount of plutonium had collected in a processing tank. When a stirrer was actuated an aqueous solution at the bottom of the tank flowed up around the plutonium-rich organic phase above it, producing criticality.

THIS NUCLEAR CRITICAL ACCIDENT resulted from an unusual and complex set of circumstances. Reconstruction of the steps that preceded it and analyses of the materials involved now give a reasonably specific picture of the conditions at the time of the radiation burst.

Operational Background

The portion of the plant in which the accident took place is used for concentration and purification of "lean" residues from a plutonium recovery operation. Typically, solutions contain less than 0.1 gm/liter of plutonium and traces of americium. The plutonium and americium are separated by repeated batch extraction into tri-*n*-butyl phosphate (TBP) carried in organic solvent, followed by back extraction into an aqueous phase that is concentrated by evaporation.

The resulting solution, containing plutonium at a few grams per liter, then is fed back into an earlier recovery stage.

These processes are complicated by hydrolysis of TBP into mono- and dibutyl phosphates from the actions of heat, nitric acid and alpha-particle radiation. Both hydrolysis products tend to form tightly bound plutonium complexes, and periodically they must be precipitated from the TBP-solvent mixture. These plutonium-rich solids are filtered and sent to another part of the plant for processing in small batches. Figure 1 is a simplified flowsheet for the residue-recovery operation, and Fig. 2 shows the corresponding layout of equipment.

At the time of the accident a physical inventory was in progress in the TBP-solvent-extraction plant. The normal inflow to this area was interrupted, and residual materials in all process vessels were to be evaluated for plutonium content. This requires filtration of solutions supplemented by thorough cleaning of vessels and analyses of clarified solutions and removed solids.

Before the Accident

Reconstruction of significant events indicates that plutonium-rich solids, which normally would have been handled separately, were washed from two other vessels into a large vessel that contained dilute aqueous and organic solutions. After removal of most of the aqueous solution from this vessel, the remaining 40 gal of material

was transferred to the stainless-steel solvent-treating tank in which the accident occurred (Fig. 3). This tank already contained about 80 gal of caustic-stabilized aqueous-organic emulsion that had resulted from the second step in the precipitation of TBP hydrolysis products. In addition, 13 gal of concentrated nitric acid was used to wash solids from the bottom of the large vessel into the solvent-treating tank. Transfers into the bottom of the tank were followed by air-sparging, which mixed the contents, so that the emulsion and plutonium originally in the solids was extracted into the solvent. Phases then separated; about 2.5 min was required.

The resulting situation in the 225-gal, 38-in.-diameter, solvent-treating tank immediately before the accident is now believed to be as illustrated in Fig. 4: 87.4 gal of aqueous solution contained 40 gm of plutonium, and on top, a 42.2-gal layer of solvent contained 3.27 kg of plutonium. Solids (containing 60 gm of plutonium) were suspended in both the aqueous and organic phases and at the interface; few solids settled to the bottom of the tank. Estimates based on Fig. 5, taking into account the tank diameter and effects of neutron poisons, indicate that the 8-in.-thick solvent layer was barely subcritical at the plutonium concentration of 20 gm/liter (the estimated critical thickness is 8 1/4 inches, and the actual configuration was roughly 5 dollars subcritical).

Continued on next page

The Accident

A chemical operator started the motor driving the stirrer in the solvent-treating tank. There was a "blue flash" and a muffled report. The operator, who was looking into a sight glass at the top of the tank, (Fig. 4) was knocked off the two-step ladder on which he stood. The operator apparently turned the stirrer motor off and then on again (this time noticing a rumbling sound), ran out a nearby exterior door and called that he was "burning up." Though the shock displaced the tank about $\frac{3}{8}$ in. at its supports, the tank was not ruptured and no plutonium escaped.

The tank in which the accident occurred was not bolted to the floor. Tangential shift of one of the supporting legs was apparent because the initial position was outlined by the latest coat of floor paint. Apparently the tank pivoted about the opposite foot.

We cannot be certain that there was no following low-order burst when the stirrer was turned on the second time, but it seems unlikely. No accompanying blue flash was reported, and it is doubtful that the few seconds presumed to have elapsed while the stirrer was off would have been sufficient to re-establish a potentially critical situation. Radiation-detector traces are consistent with a single-burst hypothesis, but do not exclude the possibility of two closely spaced pulses. The stirrer rumbles when turned on under normal conditions; it appears that degree of rumble could easily have been misjudged. We will never know for sure.

At the time of the burst a second operator some 40 ft away in an adjoining room saw a reflection of the light on the walls ("like a

TABLE I—Pu in Withdrawal Tanks*

Tank no.	Aqueous solution		Organic solution		Solids Pu (gm)
	Volume (liters)	Pu (gm)	Volume (liters)	Pu (gm)	
1	63.5	7.54	0	0	1.33
2	67.2	8.80	0	0	1.09
3†	43.5	3.38	15.8	362	9.89
4	0	0	56.2	1129	7.20
5	0	0	0	0	0
6	68.0	8.02	0	0	1.50
7	54.0	7.07	0	0	0.76
8†	34.7	2.43	25.0	518	18.3
9	0	0	63.0	1263	7.4
10	0	0	0	0	0
TOTALS	330.9	37.2	160.0	3272	47.5

Average concentrations (gm/liter)

0.112

20.4

Estimated solids not removed from tanks 3 and 8, and other solids

60

* Tanks shown in Fig. 6 are numbered successively from bottom to top. The nearer ones are Tanks 1-5; the farther ones, Tanks 6-10. They were filled from bottom upward.

photoflash"), heard the report, went to help, and was joined by a third operator. They joined the first operator who had by this time run out of a nearby door. All three men returned past the accident tank to lead the injured operator to a shower. By this time he suspected that he was suffering from an acid burn. As the three passed

the tank in which the accident occurred, the second operator turned off the stirrer motor.

The burst activated a radiation alarm located 175 ft away in a nearby building. (This unit was connected to an indicating instrument and set to sound at 10 mr/hr. After a report of the alarm, the entire plant

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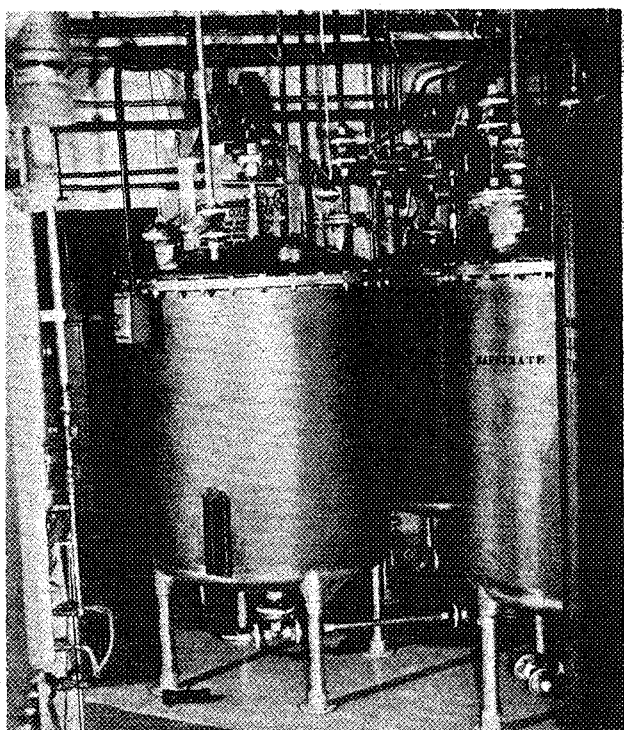


FIG. 3. ACCIDENT TANK, shown at left, was moved $\frac{3}{8}$ in. by shock that occurred when stirrer was turned on

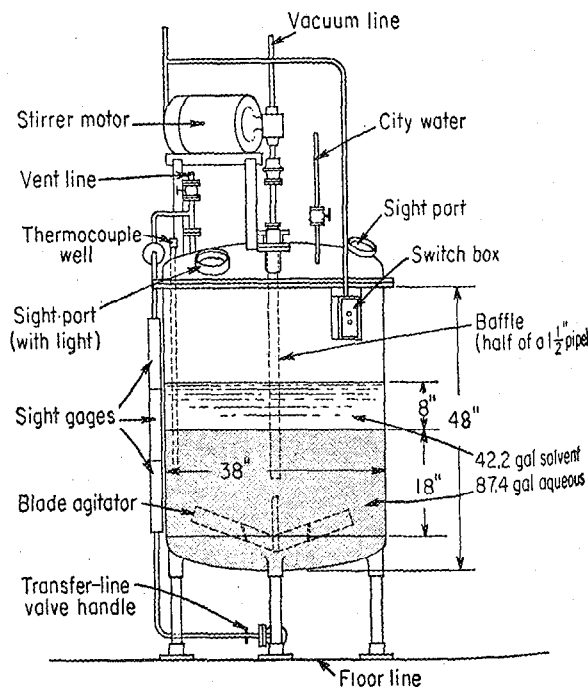


FIG. 4. RECONSTRUCTION OF SITUATION at time of accident indicates tank had barely subcritical layer of organic solution above aqueous layer

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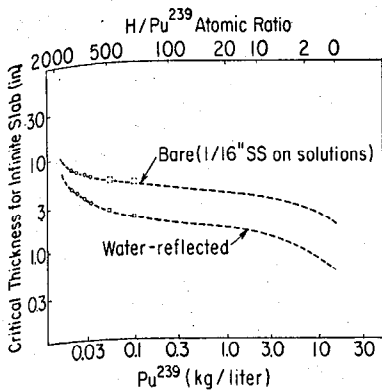


FIG. 5. CRITICAL THICKNESSES of infinite slabs can be used to estimate amount by which tank was subcritical before stirring

was evacuated. The time was 4:35 p.m., December 30.

The Burst

The initial action of the stirrer in the solvent-treating tank forced aqueous solution up along the wall, displacing the outer portion of the solvent layer and thickening the central portion. An average increase in solvent-layer thickness of ~0.4 in., corresponding to an average radius decrease of $\frac{1}{2}$ in., could account for the obviously supercritical configuration. The burst was certainly terminated by the violent disturbance that it generated, and continuation of the stirring diluted the plutonium beyond the point at which a critical reaction could recur.

Radiochemical analyses for Mo⁹⁹ (concentrated mostly in the aqueous phase) indicate that the total number of fissions in the burst was about 1.5×10^{17} . This value may be subject to refinement when analyses for other fission products are complete.

Solution Removal

Immediately after the incident, a bank of ten 6- and 5-in.-diameter withdrawal cylinders was fabricated and set up about 100 feet from the point of the accident (Figs. 2 and 6). This storage array was connected to a transfer line that leads from the bottom outlet of the solvent-treating tank.

As the disposition of plutonium in the tank was then unknown, opening of the bottom outlet valve was a suspect operation, and therefore it was accomplished from behind a temporary shield with a 10-ft extension handle. The solution was vacuum-transferred into the bank of cylinders without complication on January 1. The solution level within the cylinders was followed by gamma-sensing instruments (maximum reading at contact was about 1 r/hr, and indications along the wall of the solvent-treating tank immediately after emptying were less than 50 mr/hr).

Analysis. Successive 9-liter lots of solution were removed from the storage cylinders, and each batch was sampled for chemical analysis. Results of analysis are summarized in Table 1. Not only was the

Special Committee Reviews Report

A technical report* describing the circumstances of the Los Alamos criticality accident has been made available by the Atomic Energy Commission. It has been written by the authors of this article and can be got at the Office of Technical Services, Department of Commerce, Washington 25, D. C.

General Manager A. R. Luedeker of the AEC appointed an investigation-review committee of AEC personnel to evaluate the report. On the committee were Marvin M. Mann, Division of Inspection, Clifford K. Beck, Division of Licensing and Regulation, Charles L. Dunham, Division of Biology and Medicine and Kenner F. Hertford, manager of the Albuquerque Operations Office.

The committee concluded that the accident was directly attributable to errors on the part of the operator who was killed. There has been some disagreement about how strongly the blame should be fixed. It has been established that the operator did not follow precisely the directions of his supervisor, who had outlined the procedure before the day's operations began.

The operator was an experienced man who knew the processes he was carrying out and the reasons for them. He had always followed directions and had a reputation for reliability. The manner in which he departed from the prescribed procedure led him to handle several batches of material together instead of one at a time. The altered procedure made for easier inventory and a neater process. The investigators feel that this may have been his motivation in making the alteration.

The committee found that closer supervision might have prevented the accident and commended Los Alamos personnel on measures they have taken to prevent a recurrence.

* H. C. Paxton *et al*, LANS-2293 (Feb. 20, 1959)

bulk of the plutonium found in the organic solvent phase, but a concentration of Zr⁹⁰ in a suspension at the aqueous-organic interface suggested that the situation was the same at the time of the accident. Furthermore, the chemical form in which the plutonium occurred in the solvent was consistent with the hypothesis that it had been complexed with TBP hydrolysis products.

Additional evidence that the plutonium

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was concentrated in the solvent layer when the burst occurred is given by the long-lived gamma activity of the type-347 stainless-steel baffle located ~6 in. within the wall of the solvent-treating tank (Fig. 4). Scanning indicated a peak in activity about 1 in. below the mid-plane of the solvent layer.

Radiation Exposures

Results of radiation monitoring near the solvent-treating tank after the accident are summarized in Table 2. The fatally injured man received a radiation dose estimated at 12,000 rem \pm 50%. The next highest doses received by any of the operating personnel are estimated at 134 and 53 rem. Other exposures from the supercritical burst were less than 4 rem.

The chemical operator at the solvent-treating tank went into deep shock within 15 min of exposure. He regained consciousness about 6 hr later and remained rational and comfortable until nearly the time of his death at 3:15 a.m., January 1. The operators who went to his assistance show no physical effect of exposure other than typical changes in blood count in the case of the man who arrived first and switched off the stirrer motor. Both have continued on regular duty. The fact that they received predominantly gamma exposure indicates, of course, that there was no succeeding burst while they were near the tank.

Nuclear-Safety Control

Ironically, a nuclear-criticality review of the entire plutonium plant had been completed about a month before the accident. Changes in equipment associated with a hopefully improved solvent-extraction process were planned so as to reduce dependence on procedural control of batch sizes for



FIG. 6. TEN WITHDRAWAL TANKS established safe geometry for vacuum removal of solutions after accident occurred

nuclear-safety purposes. This type of control had been used successfully during the 7½ years that the plant was in operation. Installation of the new equipment was scheduled for next June and July. Depending on efficiency of the new extraction process, the possibility of eliminating the succeeding raffinate solvent extraction and

TABLE 2—Results of Radiation Monitoring of Room 218

Date	Time	Dose rate r/hr	Distance from tank (ft)
Dec. 30	4:40 p.m.	20	25
	5:10	45	7.5
	5:15	0.2	110
	5:35	>50	4.5
	7:30	12	1.5
Dec. 31	9:45	0.1	25
	8:20 a.m.	0.05	25
	1:40 p.m.	10	Contact
Jan. 1	10:01	5	Contact
	9:00 a.m.	2	Contact

concentration stages (Fig. 1) was to be explored.

Because primary dependence had to be placed on procedural controls for a while longer, they were bolstered by re-emphasis and improved solids-sampling methods. Other measures undertaken were replacement of a product vessel for concentrated solution by one of safer geometry, review of operating procedures, improvement of the nuclear-safety training of personnel, design and installation of gamma-sensing radiation alarms, and updating of evacuation procedures.

The accident occurred in a process that was believed to be relatively safe because of small throughput. Long-term holdups of material, however, resulted from the requirement that certain plutonium accounts be balanced before material in other accounts could be processed. So, even here, procedural control was recognized as a safety requirement.

Since the accident there has been further detailed review of the entire plutonium plant. Additional or accelerated safety steps that resulted are:

- Design of geometrically favorable dissolvers and feed tanks for filtration and solvent extraction. This has been completed, and it is expected that the equipment will be installed before operations are resumed.
- Blocking of supplemental transfer lines. Lines like those required for emergency procedures will be blocked to minimize the opportunity for abnormal interchanges.
- Placing cadmium nitrate solution in vent tanks and vacuum-buffer tanks. This is to protect against criticality in case of accidentally introduced plutonium solutions. The use of both soluble and fixed neutron poisons in large process equipment is being studied, though unequal distribution between phases in the one case, and plutonium deposition on surfaces in the other, present difficulties.
- Calibrating and testing neutron detectors for indicating abnormal deposits of plutonium.

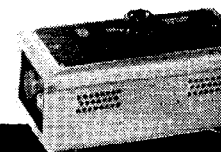
Previous Accident Reports:

Kellogg, NUCLEONICS 15, No. 12, 42 (1957)
insdale, NUCLEONICS 15, No. 12, 43
W(1957)
Oak Ridge Y-12, NUCLEONICS 16, No. 11,
138 (1958)

PHIL

equipment for

Single channel
pulse-height analyser
type PW 4082
shown together with
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type PW 4072
and anode supply
PW 4029:
at left
pre-amplifier
type PW 4071.



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for info
Radiation Safety
Number 88-4

LOS ALAMOS SCIENTIFIC LABORATORY CRITICALITY ACCIDENT

The Los Alamos Scientific Laboratory experienced a criticality accident on December 30, 1958, in a plutonium recovery area when solutions from four tanks were transferred into one 850-liter (225-gallon), 96.5-cm (38-inch) -diameter tank.

Large, unfavorable geometry tanks were allowed in the recovery area due to a small (0.1 kg) plutonium inventory and low concentration (0.1 g per liter) solutions. Nevertheless, intentions during plutonium inventory were that each vessel containing fissile material be emptied and cleaned individually. Instead, acid solutions and organic residues from four tanks were transferred into the 850-liter tank. It is presumed that this transfer included 3.23 kg of plutonium that had accumulated in the process equipment during 7.5 years of operation prior to the accident.

After transfer, the organic and aqueous phases separated resulting in the extraction of 3.27 kg of plutonium in the organic phase at a concentration of 20.5 g per liter. The organic phase floated like oil on water on the aqueous phase which contained 60 g of plutonium at a concentration of 0.18 g per liter. The system was subcritical due to the low concentration in the aqueous phase and the slab or disk-like geometry of the organic phase. Only a slight change in the geometry of the organic phase was necessary to cause criticality, and this change occurred when the stirrer in the 850-liter tank was started (see Fig. 1).

An operator looking into a sight glass on the tank (Fig. 2) was exposed to the radiation from 1.5×10^{17} fissions in the 2 seconds before continued stirrer action mixed the two phases diluting the plutonium to a subcritical concentration. This operator died 35 hours later, and two other operators nearby received significant doses of radiation.

The plutonium recovery plant had been scheduled to operate for six more months but was retired immediately. A new facility was constructed with equipment geometrically favorable for criticality safety. Large auxiliary vessels were filled with borosilicate glass Raschig ring neutron absorbers, and unnecessary solution transfer lines were blocked. Written procedures and the criticality safety training program were improved. Portable survey instruments were acquired to detect plutonium buildup in the process equipment, and improved gamma-ray-sensing radiation alarms were installed to ensure more complete coverage of process areas.

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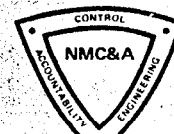
SAFETY



ENVIRONMENTAL



NUCLEAR
CRITICALITY



NUCLEAR MATERIAL
CONTROL

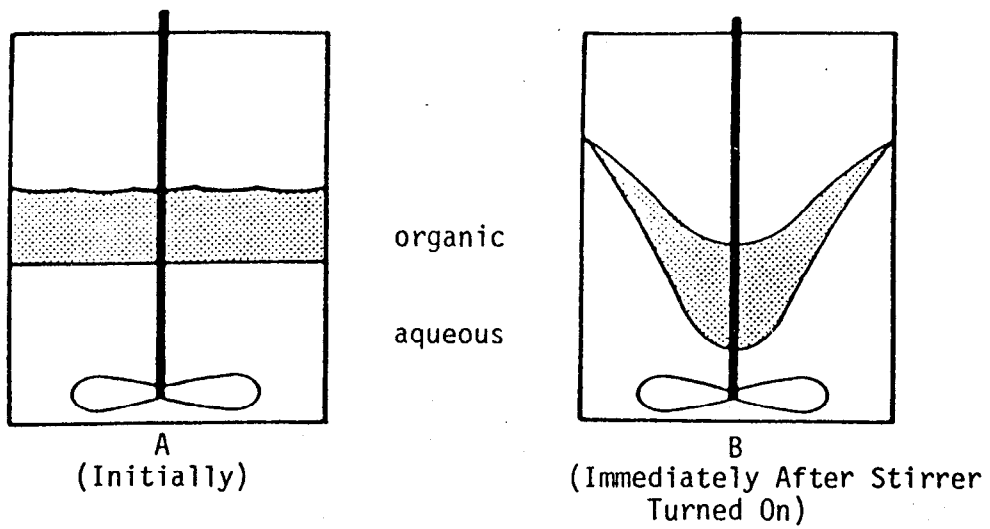


Fig. 1. Solutions in the 850-Liter Tank

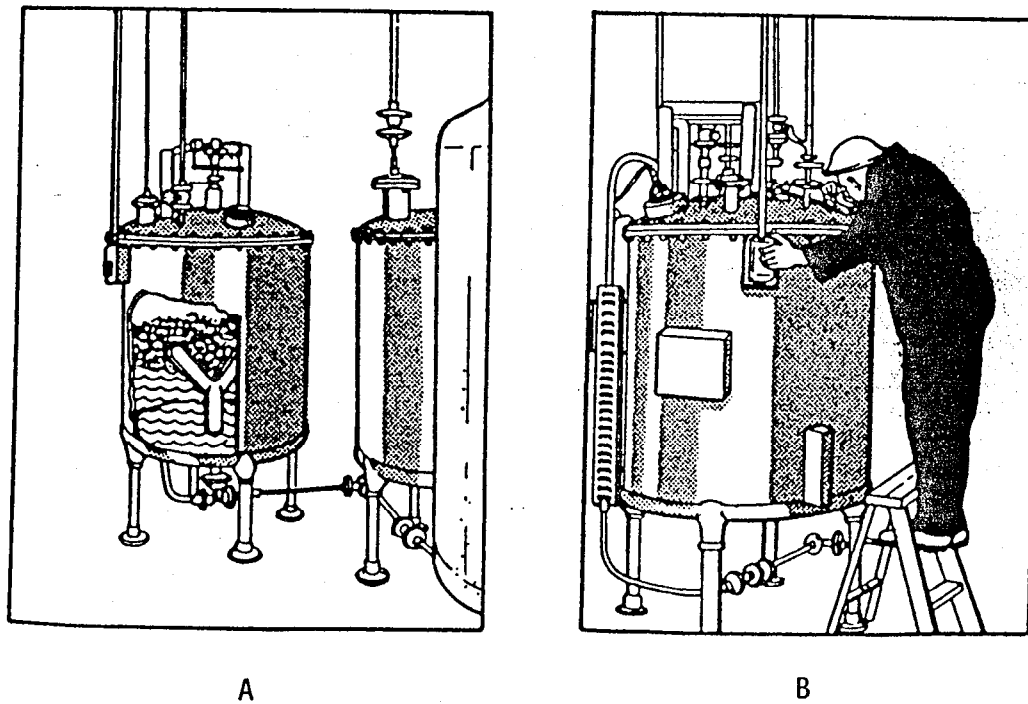


Fig. 2. The plutonium-processing tank (A) cutaway and (B) with the operator in position at the time of the LASL criticality

D. D. Butcher
 April 21, 1988

A CASE HISTORY

An example of a radiation accident is presented here in detail to illustrate the circumstances that may lead to an accident, the radiation injuries caused, and the clinical course of radiation sickness. The following description consists of partial quotations* from a lengthier report published by the investigators who were directly in charge of the treatment of the victim.

The Los Alamos accident was a criticality accident and resulted in one fatality. The victim was exposed to one of the largest doses ever reported for any accident. He died of direct damage to his heart, although he must have also suffered from a severe gastrointestinal syndrome. Death followed within less than 2 days after the accident. With the high supralethal dose involved nothing could be done to save his life. This accident, like many others, was thoroughly studied by experts; their studies have been a significant contribution to the knowledge of human radiation injury. The following are the details of the 1958 Los Alamos accident:

On the 30th of December, 1958, an accident occurred at the Los Alamos Scientific Laboratory which was of particular interest because of the extremely high dose of radiation delivered to the principal victim. The accident occurred in a complex of buildings known as DP West, situated some thousands of feet from any housing areas or from any other concentration of work. DP West is primarily concerned with the chemical and metallurgical processing of plutonium. The procedure being carried out when the accident occurred was the recovery of plutonium from liquid wastes (a lengthy process involving many steps). Not more than a few hundred grams of plutonium were normally processed at any one time. At the time of the accident, the operation was nearing the final step with plutonium in a tank containing water and a solvent, tributyl phosphate. The solvent and aqueous phases were in separate layers, but were to be mixed by stirring.

This was an extremely complicated accident resulting from several quite unrelated factors. In the first place, the process was the normal routine, one that had been carried out many times over a three-year

period; it was the end-of-the-year cleanup in preparation for the annual inventory of plutonium. It is difficult to imagine that the simple action of a stirring device in a tank could draw a subcritical configuration of fissionable material into a truly critical geometry. The system had been in operation for a number of years and many batches of plutonium had been processed. With each batch a little more went in than came out, and it was assured that the deficit was irrecoverable loss which had gone down the drain. What was not realized was that over the years, the system had actually retained, bit by bit, a total of almost 3.0 kg of plutonium.

The chemical operator "K" was a man of no great technical education, but with many years of practical experience in this and other related operations. He was repeating a process he had carried out many times before. It is possible that over the years he had introduced a few short-cuts in the process without the knowledge of his supervisors. On the afternoon of the accident, "K" was standing on a short stepladder, looking through a viewing port into the tank (where plutonium was). Within seconds after the stirrer was started, there was a muffled boom and "K" fell backwards off the stepladder. The blades of the stirrer drew material down in the centre and forced it up the outer sides of the tank and for an unfortunate instant the geometry in the solvent layer brought the material together in a critical configuration.

There was only a single critical excursion without subsequent oscillations such as occurred at Oak Ridge. Later calculations showed that there had been a burst of 1.5×10^{17} fissions. Fortunately, "K" was the only man in the room, but there were two men in the adjacent room. There were a great number of tanks of various sizes which fortunately shielded the other two men, D and R. Both these men heard the boom of the critical excursion. In a matter of seconds D had left his work station to see what had happened in the next room. By the time he got there, "K" had already picked himself up off the floor and had gone to and opened the outside doors. When D reached him, "K" was standing outside in the snow. D found "K" ataxic and disoriented. He needed support to remain erect, and all he could say was "I'm burning up. I'm burning up." "K's" face appeared flushed even at this early time.

Thinking that "K" had been the victim of chemical contamination, D guided and supported "K" back into the room where they were met by R and the three continued on to an emergency shower. D and R stripped of his outer clothes and held him under the shower because he could not stand unaided. Perhaps five minutes after the accident, he was virtually unconscious. While R called for assistance, D returned to the room of the accident. He certainly passed within a few feet of the tank at least two or more times.

The plant nurse arrived on the scene approximately ten minutes after the accident and was puzzled to find a patient obviously in shock and unconscious, but with nice, rosy-pink cheeks. She did not realize that his color was due to radiation-induced erythema. The patient was nearly pulseless. The man was admitted to the emergency room of the Los Alamos Medical Center twenty-five minutes after the accident.

The patient was a powerfully-built man of 38; he weighed approximately 170 lbs and was 71 inches tall. By the time he arrived at the hospital, he was semiconscious, but disoriented. He was moving around restlessly on the stretcher and all visible skin areas were of a dusky purplish color. He seemed to be in severe pain, apparently abdominal. His conjunctivae were markedly reddened, but his excessive restlessness made careful examination difficult. He retched frequently, but vomited only small amounts of watery fluid. About 10 minutes after admission, he had an episode of explosive watery diarrhea. Some of this faecal fluid was radioassayed and showed a significant content of ^{24}Na , indicating a copious passage of fluids into the gastrointestinal tract.

His blood pressure was found to be 80/40 mm Hg with a pulse rate of 160 per minute. He had repeated mild shaking chills, and his restlessness was so great that he had to be restrained. An indication that the dose had been massive was the fact that a portable gamma survey instrument held to the surface of the body gave a reading of 15 mR/hour.

The patient was placed in an oxygen tent. His hypotension and his rapid pulse still persisted and his rectal temperature was found to be 103°F. Physical examination did not reveal impressive findings. His optic nerves were normal, but the mucous membranes were intensely injected. His eyes looked as though they should have been painful, but the patient denied any discomfort. There was definite redness over the anterior surface of the body down to the level of the knees.

About 5 hours after the accident, the patient appeared to be in a satisfactory condition. He was rational, comfortable, and emotionally at ease. By this time, it was also apparent from the dosimetric studies that his radiation exposure had unquestionably been supralethal and of greater magnitude than in any of the cases previously reported. The changes in his white cell counts reflected this very definitely. The total white cell count rose steadily to a peak of 28,000 per mm⁵, but the lymphocytes had virtually disappeared from the circulating blood in less than 6 hours. This we regarded as a very grave prognostic sign. A very dramatic finding was the marked degree of urinary retention. There was a total urinary output of less than 600 ml with a total fluid intake of approximately 14 litres.

On the second evening more than 30 hours after the accident, the patient's condition deteriorated rather abruptly. He developed increasing abdominal cramps and fairly heavy sedation failed to control his restlessness. Despite administration of oxygen by mask, he showed increasing cyanosis. Sedation was given and he lapsed into a coma from which he never roused. Death supervened from cardiac arrest 34 3/4 hours after the accident, his heart having been the target of nearly 12,000 rad of ionizing radiation.

The neutron dose was determined by measurement of induced ²⁴Na activity in the blood, in selected body tissues, and in the whole body, as well as from induced activity in other materials such as brass overall buttons, and nearby chemicals. It now appears that the combined neutron and gamma dose delivered to K's anterior chest wall and, thus, to the right side of the heart and the anterior wall of the stomach was apparently 12,000 rads. The total dose to the face and to the front of the skull was less, but still in excess of 10,000 rads. The dose to the lower legs was probably less than 1000 rads.

At autopsy, the most striking find, was the edematous, water-logged appearance of practically all tissues except the lungs. The general picture was quite characteristic of acute right heart failure resulting from right-sided myocarditis complicated by excessive fluid intake. The first loop of the jejunum, the gastric pyloric bulb and the surface of

the left lobe of the liver contained numerous petechial hemorrhages. The spleen was wrinkled and flabby. The right side of the heart was dilated and filled with blood, while the left heart was in systole. Externally, the right auricle and the anterior portion of the right atrium also showed hemorrhages similar to those in the pericardium.

This man had received more than enough radiation to his bone marrow to kill him in 3 or 4 weeks, had he not had any other injuries. The injury to his gastrointestinal tract would have killed him in 1 or 2 weeks had not a more vital insult killed him first. In our case, the man received, at the same time, another and quite distinct injury to his heart, one which physiologically, was quite overwhelming. It seems clear that the injury to the heart muscle, in this case, must be regarded as the primary cause of death.

* From Shipman, T. L., 1961. A radiation fatality resulting from massive overexposure to neutrons and gamma rays. In "Diagnosis and treatment of acute radiation injury." International Documents Service, New York, Pp. 113-133.