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ORNL - TM - 540
Addendum

28

DRAFT

TECHNICAL PROGRESS REVIEW

NUCLEAR SAFETY

VOLUME 4, NO. 4

[redacted]

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To: Distribution Listed in ORNL-TM-540 Addendum

The two incident reviews in the attached addendum to the draft copy of Nuclear Safety, Vol. 4, No. 4, are to be a part of Section 6 of the finished copy of Vol. 4, No. 4. It had been intended that the reviews be in the draft issue, but they were not available in time. If you have any comments or criticisms concerning the contents of this addendum, please send copies to Mr. Thomas W. Hazard of DTI, Mr. H. F. Carroll of Declassification, and to me. Inasmuch as the final draft of this issue is scheduled to go to press early in June, your comments will have to received by June 1 in order to be considered for inclusion in the final publication. Mr. Hazard's and Mr. Carroll's addresses are as follows:

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Yours truly,

Wm. B. Cottrell by AWS

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TECHNICAL PROGRESS REVIEW
NUCLEAR SAFETY

Volume 4, No. 4

Prepared for U.S. Atomic Energy Commission
by Oak Ridge National Laboratory

DATE ISSUED

MAY 20 1963

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
operated by
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U.S. ATOMIC ENERGY COMMISSION

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MTR FISSION-BREAK INCIDENT

R. A. Costner, Jr.

A fission break[1] occurred at the Materials Testing Reactor (MTR) on November 13, 1962 because of the melting of a small portion of one of the 19 fuel plates in a single fuel element. There was a total loss[2] of about 0.7 g of U^{235}. The plate melted as a result of insufficient cooling caused by a restriction of the flow of the primary cooling water by debris that was later identified as a gasket material from the floating roof of the seal tank (a 17,000-gal water-supply tank). The reactor automatically shut itself down, and personnel were evacuated from the reactor building for 12 to 15 min. There were no significant personnel exposures and no consequences beyond the site boundary. A brief description of the MTR, as well as some aspects of its operation, was included in the March 1962 issue of Nuclear Safety.[3]

A fission-break incident that occurred at the Engineering Test Reactor was reviewed in the June 1962 issue of Nuclear Safety.[4]

The Incident and Immediate Events

The reactor had been operating at 40 Mw since November 2, 1962, except for five short-duration power reductions. On November 13, 1962, approximately 15 sec before the scram, which occurred at 1426 hr, the fuel-assembly differential-pressure monitor annunciated[5] to indicate that the differential pressure across one of the fuel elements had been reduced by 2 psi. (This instrumentation is set to initiate a full scram if the drop is 4 psi.) During the annunciation, the servo-controlled (regulating) rod moved erratically over a span of 2 in. (i.e., between 15 and

[redacted]

17 in. withdrawn). At 1426 hr a full scram was initiated by the N^{16} instrument and by the low differential-pressure system. Owing to the coincidence of the signals, it was difficult to determine which signal occurred first.

One minute after the scram the reading of the seal-tank activity monitor, which is in the primary-cooling-water exit line just upstream from the seal tank, increased from 100 mr/hr to 9.8 r/hr. (Full scale on this instrument is 10 r/hr.) During this same period, the reading of the vent-seal activity monitor, which monitors all gaseous effluent removed from that portion of the primary cooling water that passes through the flash evaporators, increased from 970 mr/hr to greater than 10 r/hr. (Full scale for this instrument is 10 r/hr and it remained off-scale for about 44 min.) By 1428 hr, several outlying area radiation monitors had begun to alarm. Health physics survey teams responded immediately and surveyed all areas, including the process-water building and the fan house. At 1431 hr, personnel were evacuated from the reactor, reactor services, and reactor wing buildings. Health physics survey teams determined that all radiation readings were due to radiation from systems containing primary cooling water and that there was no air activity in the reactor building at that time. Personnel were then allowed to return to the buildings in 12 to 15 min, although personnel movement in the reactor-building basement was under the surveillance of health physics personnel until the direct readings subsided.

Preliminary Investigation and Remedial Action

Following the scram, the primary coolant flow was maintained at the normal rate of 22,500 gpm in order to reduce the deposition of fission

products on the walls of the reactor system and to aid in the preliminary investigation of the cause of the incident. At 1428 hr an inspection of the fuel-assembly differential-pressure monitors indicated that the differential pressure across element J-385 in position L-32 was only 8 psig, that is, 5 psi less than normal, which was more than sufficient to initiate a scram. The differential pressure across all other elements was normal.

At 1500 hr, air activity of low concentration was detected in the reactor-building basement, on the main floor, and at the reactor top. A sample of the primary cooling water was taken at 1505 hr that indicated a total beta-gamma contamination of 630,000 cpm/ml (following a 2-hr decay), which was 13 to 16 times normal. A comparison was made of the results of a detailed analysis of this sample and of a sample taken several hours prior to the scram, and it was found that the activity from specific nuclides increased by the following ratios: I^{131} and I^{133}, 62; I^{135}, 126; Ba^{140}, 25; and Sr^{91}, 36.

At 1655 hr, flow of the primary cooling water was stopped in order to drain the water from the overhead working reservoir to a retention basin. By 1935 hr, all the water in the reactor primary system had been flushed to the basin and replaced with fresh demineralized water. Flush samples taken at the completion of this operation showed that the total beta-gamma activity was 124,000 cps/ml (following a 2-hr decay). By 1925 hr, all areas were free of air activity.

Postincident Inspection and Operation

The reactor top plug was removed at 2025 hr, and one minor release of air activity occurred during the removal operation. The bottom of

the plug read 1 r/hr at 1 ft. An inspection of the reactor core revealed a piece of black material in element J-395 in position L-32. A considerable portion (estimated as 40%) of the area on top of the fuel plates in this element was covered by this material. Small pieces of this material were also found on the elements in positions L-35 and L-37; however, differential-pressure readings made prior to stopping the flow of the primary cooling water were normal for the latter positions.

On November 14 the reactor core loading was changed, the top plug was reinstalled, and the flow of the primary cooling water was re-established (22,500 gpm). After 2 hr the core and reflector pieces were inspected, and no additional debris was found.

On November 15 the Reactor Physics and Engineering Staff and the MTR Safeguards Committee approved returning the MTR to power under the following conditions:

1. Primary cooling water was to be circulated at the normal operating condition for 1 hr prior to shim rod withdrawal. During this period and thereafter, each fuel-element differential-pressure monitor was to indicate a value comparable to that observed during the previous 2-hr circulation test (when the core was found to be free of debris).

2. Satisfactory results were to be obtained during shim rod drop tests.

3. The output of the reserve servo system was to be monitored on a fast-response recorder (to warn of the onset of nucleate boiling) during each increase in reactor power level. The reactor power level was to be increased in 5-Mw steps until 30 Mw was reached; power level increases above 30 Mw were to be made in 2.5-Mw steps.

4. The fuel-element differential-pressure monitoring system was to be given rigid surveillance during the remainder of Operating Cycle 182.

Later, on November 15, preliminary work was started to return the reactor to power under these conditions; however, a 2-psi drop in the differential pressure across the element in position L-34 was indicated. Inspection of the core revealed no debris, but inspection of the differential-pressure monitors for positions L-34 and L-35 resulted in replacement of the overrange pressure bellows in the monitors. By 0800 hr on November 16, a power level of 10 Mw was attained. The full reactor power of 40 Mw was attained at 1056 hr on November 16. As a precaution the fuel-element differential-pressure instrument set-point was adjusted to initiate a full scram when a 2-psi drop occurred.

The total beta-gamma contamination in the primary cooling water rose to 120,000 cpm/ml (following a 2-hr decay of the sample). This level had decreased to 70,000 to 80,000 cpm/ml (following a 2-hr decay of the sample) when Cycle 182 was terminated on November 19.

Iodine-132 tests indicated definitely that fuel element J-385 had a fission break, and therefore the element was transferred to the MTR-ETR hot cell for examination.[2] This examination revealed that a small portion of the seventh plate (from the convex side) had melted, and approximately 0.7 g of U^{235} was lost. (The total U^{235} inventory in element J-385 was calculated to be 177 g based on 11.5% burnup.) It also appeared that the channels on both sides of the damaged plate had been partially blocked, but there must have been sufficient cooling water on the convex side of the sixth plate and the concave side of the eighth plate to prevent damage to these plates.

During the Cyle 183 shutdown (November 19 through November 22, 1962) the seal gasket on the seal-tank floating roof was found to be in poor condition, and replacement of a major portion of the seal was necessary. The gasket material was analyzed and appeared to be the same as that found in element J-385.

Subsequent Related Events

On November 25, 1962, a scram was initiated by the differential pressure for position L-12.[6] The core was inspected, and the fuel assembly was found to contain debris described as "red rubber." Later, on December 24, 1962, a scram was initiated by the differential-pressure monitor for position L-19.[7] Again a "red-rubber" type of material was found in the fuel elements in positions L-16, L-17, L-19, and L-35. The material covered approximately 40 to 50% of the plate-channel area of the element in position L-19; much less material was found in the other elements. Emission spectrometer data indicated a similarity with the material found in position L-19 on November 25, 1962; however, an infrared analysis did not correspond to that of the L-19 material.

During the shutdown for Cycle 185 (January 2 through January 5, 1963), the primary side of flash evaporator C was inspected,[8] and numerous pieces of debris were found in the spray nozzles. Therefore, evaporators A and B were also inspected, and debris was again found but not in the quantity found in evaporator C. All the debris was removed and examined. Several pieces of the "red-rubber" material were included in the debris.

Conclusions and Recommendations

The lessons to be learned from the above events are the following:

1. A method of screening reactor systems to minimize or eliminate obstructions to fuel element cooling flow is desirable. Such screens should, as far as possible, isolate the cooling-water inlet from equipment containing moving parts and from areas where gasket material is expected to function under dynamic conditions. The openings in these screens should be smaller than the spacing between plates in the fuel elements. Periodic inspection of the screens for the presence of debris may warn the reactor-operations staff of difficulties before they become serious.

2. When events such as those described above occur in spite of elaborate precautions and instrumentation systems, there is no substitute for a well-trained staff. The response of health physics, maintenance, and MTR-operations personnel was excellent.

3. The value of an internal review by groups familiar with the particular reactor but not directly involved in reactor operation is illustrated by the thorough, but straightforward, procedures developed for returning the reactor to power.

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ACCIDENTAL NUCLEAR EXCURSION IN RECUPLEX OPERATION AT HANFORD IN APRIL 1962

Dixon Callihan

Criticality was accidentally achieved at about 11 a.m. on Saturday, April 7, 1962 in essentially unshielded plutonium-recovery equipment at the Hanford Atomic Products Operation of the General Electric Company. A volume of plutonium solution, which had been separated from the uranium and fission products with which it was associated earlier, was drawn by vacuum into a cylindrical vessel where it became supercritical. Oscillations between supercritical and subcritical apparently followed during the succeeding 37 hr and generated 8×10^{17} fissions. Dosimetric observations showed that three employees near the site of the accident received exposures to neutron and gamma radiation totaling 110, 43, and 19 rem, respectively. No clinical symptoms attributable to these exposures have been observed. None of the 19 other persons in the building received more than 2 rem. These exposures are not inconsistent with the plausible power-versus-time pattern of the excursion, which was developed from the nuclear properties of the materials concerned and an estimate of their quantity and their location. The power excursion caused no rupture of process lines or damage to equipment, and the plutonium contamination problem, which is normally present in these operations and requires that the equipment be provided with containment barriers, was in no way aggravated by the accident. The following discussion of the nuclear power excursion is a summary of the report[1] of the investigating committee that was appointed by the Manager, Hanford Operations Office, Atomic Energy Commission, and

was composed of staff members of the Hanford Operations Office and the Hanford Atomic Products Operation.

It is interesting to observe that this is the fifth criticality accident to occur in chemical operations within the history of nuclear energy in the United States. The first of these was at the Oak Ridge Y-12 Plant[2-6] in mid-1958. Another occurred at the Los Alamos Scientific Laboratory,[7,8] and two occurred at the Idaho Chemical Processing Plant, National Reactor Testing Station.[9-12] In the first two accidents, operations with unirradiated fissile material were being directly performed by personnel, as was also true in the Hanford accident, whereas the Idaho accidents were in remotely operated well-shielded equipment for processing irradiated reactor fuel. Accordingly the exposures to individuals in the vicinity of the Y-12 accident were as high as 461 rem; the exposure which resulted in a fatality at Los Alamos was estimated at 20,000 rem; and those in Idaho did not exceed 8 rem internal or 50 rem to the skin. It is equally interesting to note that all the accidents, and particularly those in directly operated equipment, occurred in fissile-material salvage processes where inventory uncertainties are more highly probable and where operations are less routine than in production operations.

Description of the Facility

The accident occurred in a multipurpose recovery system designated Recuplex. The equipment for this operation is located, in part, in Room 221, Building 234-5, as shown in Fig. 1. In this operation, plutonium was being salvaged from various waste streams arising both within the Hanford production complex and in offsite sources. The impurities from

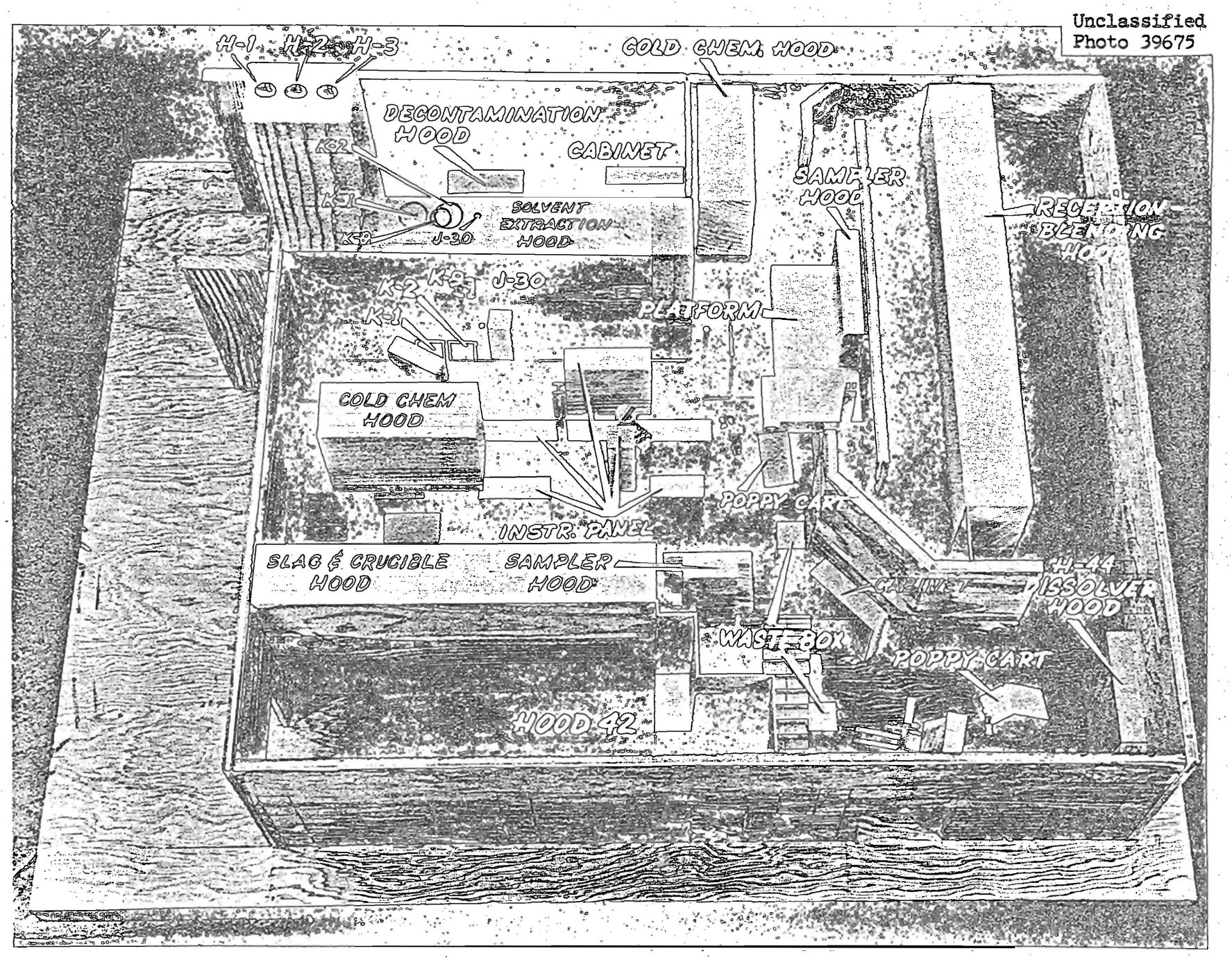

Fig. 1. Model of Hood Arrangement in Room 221. (Fig. IV-2 of ref. 1, with additions to show positions of solvent extraction columns H-1, H-2, and H-3 of Fig. 2.)

which the plutonium was separated were primarily ionic, rather than uranium or fission products. The Recuplex system contained dissolvers, feed-preparation tanks, solvent-extraction contactors, and the miscellaneous auxiliary equipment necessary for chemical separations by solvent extraction. The organic extractant was tributyl phosphate in carbon tetrachloride, which essentially quantitatively removed plutonium from the aqueous feed solution in the extraction column and, also essentially quantitatively, left the impurities behind. The ionic impurities that were carried into the organic stream were washed back into an aqueous phase by scrub streams introduced at the bottom of the first extraction column. One of these scrub streams derived from the stripping column, where plutonium was returned to an aqueous phase, and was, in fact, the product of the stripping column. In this recycling operation, plutonium was not only purified but was concentrated to about 100 g/liter prior to final discharge as Recuplex product.

Since plutonium adheres strongly to dibutylphosphate, which is a decomposition product of tributylphosphate, a small amount of plutonium was carried out of the stripping column in the organic effluent. It was necessary to treat the effluent to remove this "unstrippable" plutonium and, at intervals, the dibutylphosphate. This treatment for plutonium recovery was carried out in vessels auxiliary to the principal stream and consisted of an additional solvent extraction of the plutonium into an aqueous solution of ferrous ammonium sulfate, sulfamic acid, and nitric acid. Prior to achieving a plutonium concentration of 3 g/liter, this aqueous "cap" was transferred to subsequent process steps via a transfer tank designated K-9. It was in tank K-9 that the critical volume of

plutonium solution was accumulated. Tank K-9 was a cylindrical vessel about 18 in. in diameter with a capacity of 69 liters; its lateral wall was 3/8-in.-thick Pyrex glass; its top and bottom were 1-in.-thick steel plates.

The natural hazards associated with processing plutonium require equipment containment to limit the dispersion of radioactive contamination arising in normal operations, such as the machining of the metal, or from accidental occurrences, such as the rupture of a liquid-carrying process line. Containment is often provided by compartments constructed of transparent materials, plastic or glass, and equipped with openings tightly fitted with long gloves to allow manipulations within the compartments. These are called "glove boxes" or "hoods," even though they may be the order of a hundred feet long to accommodate extended process trains. The recuplex system occupied several such hoods, including one for the solvent extraction equipment in which tank K-9 was located. In some instances the hoods were equipped with relatively thin shields that were designed to protect the operators from the natural radioactivity of plutonium and its companion elements but which were not intended to protect against radiation arising in a nuclear accident of the magnitude discussed here.

Nuclear safety, that is, protection from accidental achievement of criticality, was effected in the Recuplex process by imposing, individually, several of the usual limitations, such as those on chemical concentration and on the dimensions and shape of equipment, and, in some cases, by combinations of two or more of these. Some equipment, for example, was described as "geometrically unfavorable," meaning that the dimensional

restrictions assured safety provided the plutonium content, or the plutonium concentration, did not exceed a predetermined value. Solutions of cadmium nitrate were added to the process stream in some instances. There was no provision, however, for the addition of solid neutron-absorbing materials, such as rings of borosilicate glass. It is obvious that tank K-9 could not safely accommodate its full capacity of plutonium solution without concomitant restrictions, so, as noted above, the process design limited the plutonium concentration to 3 g/liter, which is well below the minimum concentration required for criticality in an aqueous solution. The concentration was administratively controlled by analyses at points immediately upstream of tank K-9.

Operations at the Time of the Accident

During a period of several weeks prior to the accident, the extraction section of the Recuplex process had been used for cleaning the hood in which that section was located. This housekeeping chore was demanded by an accumulation of liquid and solid materials, and some in-between phases, on the floor of the box. The accumulation apparently included both organic and aqueous solutions of plutonium and reagents from leaks in the process plumbing; there were also neoprene gloves and plastic containers that had been temporarily disposed of by dropping them to the floor. All in all the result was a conglomerate mess that was somewhat rich in plutonium. Much foreign material had collected on the walls of the hood and thereby reduced visibility of the interior, a factor of some significance. The cleanup entailed adding successive volumes of aluminum nitrate solution and nitric acid to the floor. These solutions

were sampled and analyzed and then transferred into tank K-9 in batches, with the size of the batch established by the analyses. Cadmium nitrate solution had previously been added to tank K-9 as a safety measure. The transfer from a sump in the floor to tank K-9 was by vacuum through a temporarily installed 1-in.-diam plastic tube that was attached, through two valves, to the bottom of tank K-9 and had not yet been removed at the time of the accident, although the floor cleanup, per se, had been satisfactorily completed some days previously. There remained to be done, however, a necessary cleaning of the undissolved tarry materials from the extraction system itself before normal process operations could be resumed. Difficulties with plugged lines, flooding of columns, and system leaks were encountered in this clean out, although the operation, which was not routine, had been described, as was the entire cleaning task, by procedures properly prepared prior to its commencement.

A piece of equipment of some importance and appropriately introduced into the discussion at this point was a geometrically favorable product-receiver tank, designated J-1, that was immediately downstream of the stripping column. It could receive solutions with plutonium concentrations of up to 100 g/liter from the column. For the past several years, tank J-1 had apparently been equipped with an overflow that spilled solution directly to the floor of the hood instead of into a catch tank, where an overflow of tank J-1 could have been detected, as was formerly true. That it could overflow directly to the floor in this manner was not known to the directly associated operating organization and was not reflected in the operating procedures. The reasons for allowing solution to accumulate knowingly outside the process piping were not made clear.

In one phase of the rather complex operations, it was necessary to use tank K-9 to transfer solutions of low plutonium content from one process vessel to another, and this transfer could be effected through either or both of two paths. Transfer by one of the paths was prescribed by operating procedures; transfer by the other path although not prescribed, was not prohibited by the procedures. On occasion the latter path was employed because, in the opinion of some of the operators, the transfer could be made more expeditiously through it. Flow through this path was controlled by two valves in series, valves a and b, Fig. 2. It is probable that not only was this unprescribed path used during the shift preceding the one in which the accident occurred but also that, at the conclusion of the transfer, both valves were inadvertently left open.

Between the two valves in the unspecified flow path was a tee to which was also connected, through a third valve (valve c), the temporary plastic tube leading to the sump in the floor of the hood through which the floor-cleaning salvage solution had been removed. It was intended, of course, that this third valve be closed at all times except when removing liquid from the sump. The valve was found closed after the accident.

Immediately preceding the accident some 200 liters of organic solution containing about 2 g of plutonium per liter had been transferred via tank K-9 from one vessel to another preparatory to further removal of "unstrippable" plutonium. A few tens of liters of aqueous phase, probably quite lean in plutonium, which had been collected unintentionally atop the organic solution, was then transferred by vacuum back to tank K-9. The next step, the addition of reagents to the 200 liters of organic

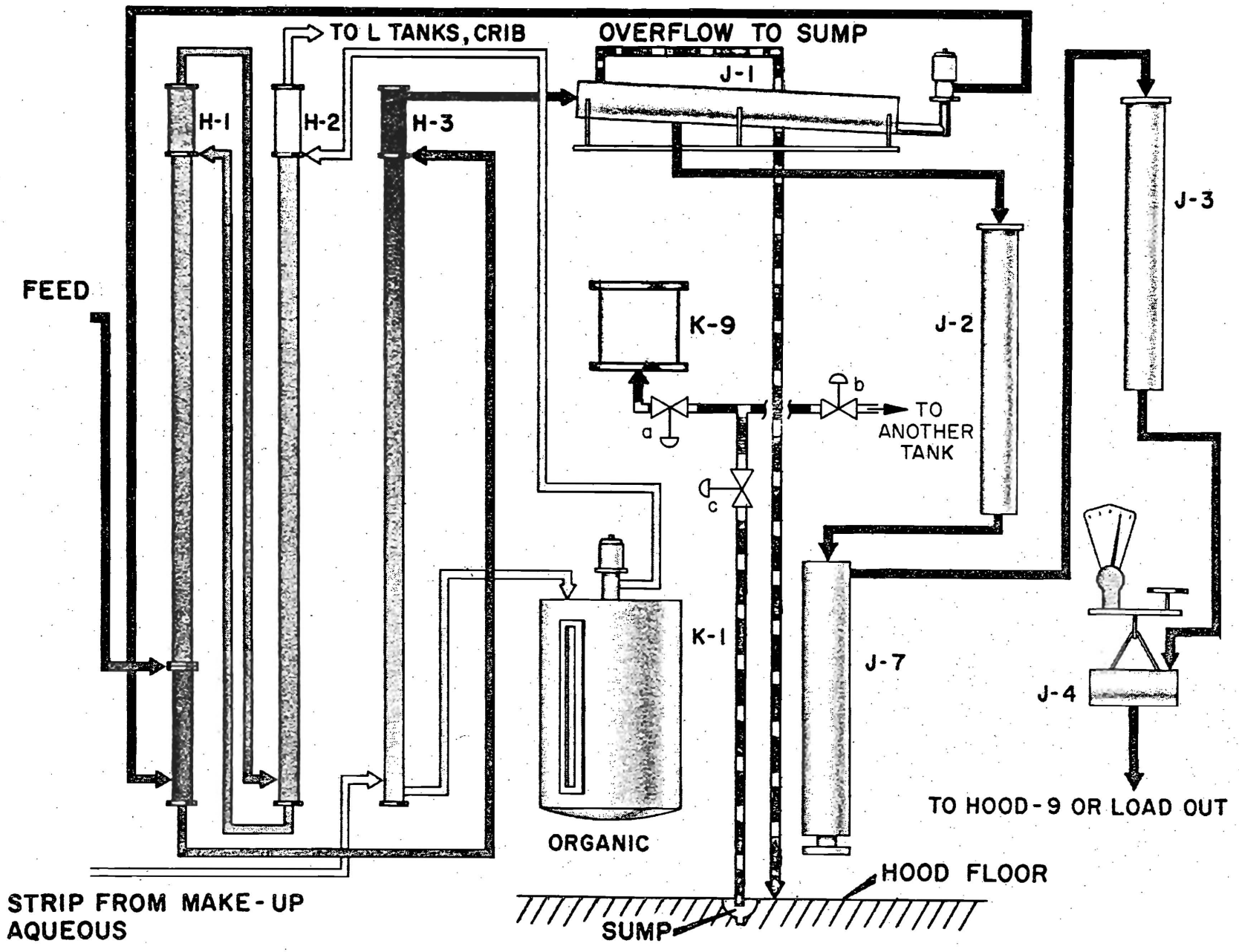

Fig. 2. Flow Diagram of Solvent Extraction Process. (Fig. IV-5 of ref. 1, with additions to show valves a, b, and c.)

solution, was interrupted by the excursion which was evidenced by a flash of blue light, the response of radiation detection instruments, the sound of emergency alarms, and, according to one observer, a sound resembling that of an electric arc. Evacuation of the area by personnel was immediate.

Cause of the Accident

In order to establish the cause of the accident, which obviously resulted from a critical accumulation of plutonium in tank K-9, it was necessary to ascertain the source of the plutonium and to postulate a reasonable method of transfer. From many pieces of information and observations, including examination of valve conditions and tests of their integrity, chemical analyses of samples from all process streams and vessels, reviews of records from continuously operating process instrumentation, and interrogation of operating personnel, the following reconstruction of events leading to the accident was derived. Even so there remain inconsistencies and uncertainties, some possibly caused by the delay in sampling enforced by the area being inaccessible.

The conclusions point to the presence of 45 to 50 liters of solution in vessel K-9 at the time of the accident. The solution contained 1400 to 1500 g of plutonium and was dilute in nitric acid and other chemicals. Within a few hours prior to the incident, an unobserved overflow of an estimated 48 liters of product solution, having a plutonium concentration of about 45 g/liter (2160 g of plutonium), probably occurred from the product recovery tank J-1 and collected on the floor of the hood. The deposit of obscurant materials on the inside of the plastic walls of the hood had reduced visibility so that an overflow of such a quantity

might not have been observed. The presence of liquid on the floor was so common that product solution there at this time probably elicited no concern, even if it were noted. A likely source of the requisite quantity of plutonium was therefore established.

Mention has been made of the continued presence of the plastic tube, which was temporarily installed for the hood cleanup, and of the fact that one of the two valves (valves a and c in Fig. 2) separating the tube from tank K-9, was found open after the accident. The end of the tube was located in the floor sump. The one valve (c) found closed after the accident must have been opened, at least momentarily, to substantiate the most reasonable reconstruction of the accident. No evidence of the valve being opened was presented by operating personnel. The valve was of a ball-plug type requiring only a 90° turn of a bar handle from full open to full closed. The handle was inside the glove box and an accidental turn of it while manipulating another was unlikely but possible.

When re-entry of the area was made, tank K-9 was found vented to the atmosphere, a consequence, no doubt, of remote manipulation of the three-way (vent vacuum K-9) valve during the activities whereby the safety of re-entry was assured. There is little question that tank K-9 was under vacuum at the time of the accident. Therefore a plausible path and means of movement of the solution existed.

An hypothesized power-versus-time pattern of the release, supported by data and calculations, demands that the approach of the solution to criticality was at a much slower rate than that possible upon the addition of approximately 50 liters in the time available. Reasonable means existed, however, for further addition of reactivity in increments

sufficiently small to account for the observations and analyses. This final increment of reactivity may have been provided through dilution of the larger volume by the aqueous "cap" of low plutonium concentration, which actually was added to the believed-to-be-empty tank K-9; the increment of reactivity may have been added by deaeration of the contents of K-9 following the addition of the "cap"; or the reactivity increase may have resulted from settling of low-plutonium-containing organic material (carbon tetrachloride, a neutron absorber) from the plutonium solution. Any one or a combination of these three possibilities would be consistent with the findings.

Personnel Response and Exposure

Following the audible radiation alarm, prompt evacuation of personnel from the entire building was affected according to well-established emergency procedures. From the local assembly point, all personnel were transferred to the area First Aid Building, where a "Quick Sort" examination* not only correctly identified the three employees who were shown later by more sophisticated dosimetry to have received doses greater than 2 rem but also excluded, with possibly one exception, all other personnel from the significant-exposure category. These three exposed employees were in the room containing the extraction hood and were at distances from the accident estimated to range from 5 to 26 ft. They, together with a fourth employee whose exposure was initially uncertain, were hospitalized. The fourth employee, who was later shown to have received

*In this test a Geiger-Müller tube is placed against the subject's abdomen, he bends his body over the tube, and neutron-induced activity in the body is detected.

an exposure of about 1 rem, was discharged the following day. The other three were discharged on the ninth day.

A complete medical report describing the radiation effects on the three individuals receiving the greatest exposure will not be issued until the completion of relatively long-term clinical tests. A variety of immediate clinical tests and radioactivity determinations (including blood pattern studies, testicular biopsies, and measurement of induced radioactivity in body fluids, in hair, in fingernails, and in toenails) gave results that were not inconsistent with exposures derived from physical dosimeters. There was some variance because the exposures occurred at short-range and therefore varied from part to part of an individual. The presently reported whole-body exposures are, respectively, for the four individuals: (1) 63 r gamma and 24 rads neutron, (2) 23 r gamma and 10 rads neutron, (3) 13 r gamma and 3 rads neutron, and (4) 1.4 rem (estimated). Based on a value of 2 for the fast-neutron RBE factor, these whole-body exposure values confirm the total exposures listed earlier. No deleterious effects attributable to these exposures had been observed in any of the individuals at the time the report[1] of the incident was issued, that is, August 1962.

Behavior and Reconstruction of the Accident

It is of interest to review the physical behavior of the critical volume, the actions taken to suppress it, and the reconstruction of its cause and behavior. Radiation surveys in and around Building 234-5 during the hour following the excursion revealed (1) a persisting gamma-ray field and (2) the absence of alpha-particle contamination; the first signified

continuation of the nuclear reaction, and the second implied the containment of the affected plutonium within the extraction hood. The continuation of the reaction was soon confirmed by measurements within the building of neutron and gamma fields as great as 0.25 rad/hr and 2.5 r/hr, respectively. Since within the next several hours the chain reaction appeared to be decreasing in intensity, it was decided to allow it to extinguish itself, as it did, about 37 hr after its initiation.

The most complete record of the history of the reaction was obtained from a recording neutron-sensing device located within Building 234-5 about 350 ft from tank K-9. Although the recorder was off scale early in the period, a power pattern can be constructed. There was a rather sharp initial power peak of uncertain duration owing to the recorder being off scale until 28 min following the excursion (except for some indication of an increase in the neutron field above background during the first 1.5 min, which possibly defines an increasing multiplication of the ambient Pu^{240} neutron population as the solution approached criticality). In the second half-hour period the intensity decreased exponentially, and then it gradually further decreased during the next 34 hr (until 10 p.m. on April 8). It then decreased rapidly to background some 24 hr later. It is postulated that the volume became subcritical about midnight April 8 (37 hr after its initiation) and that background neutron multiplication continued for another 24 hr. Imposed on this pattern were many power oscillations.

Radiochemical analyses of liquid recovered from tank K-9 and the volume involved, determined from a ring around the Pyrex cylinder, gave the energy release as 8.2×10^{17} fissions (6.4×10^{6} calories), of which

about 20% appeared in the initial unrecorded, half-hour interval. The decay during this interval was assumed to be exponential from its inception, with the period observed during the second half-hour interval. This may be a gross, but unimportant, over simplification. The pattern is not inconsistent, however, with the energy yield in the initial stages, as ascertained from personnel exposures and from dosimeters.

Cessation of the chain reaction by means then unknown demanded assurance, as a preliminary to contact action and investigation, that the chain reaction would not be restarted. At that time the exact location of the accident was not unequivocally fixed, the momentary observation of the glow having been not entirely definitive. The use of a robot to gather information and to effect remedial measures is perhaps the most fascinating part of the reconstruction. This mobile one-handed device with a closed-circuit television eye was operable from a point 100 ft distant and around some corridor corners from the extraction hood, and the operator was therefore provided with considerable radiation protection in the event the reaction started again. It was established that the reaction occurred in tank K-9, for example, by equipping the robot with a highly directional gamma-ray detector and demanding that he scan the hood. Tank K-9 was the only strong gamma-ray source observed. The robot also placed lights and instruments, read meters, moved equipment, and turned valves. The valve connecting tank K-9 to the vacuum header, for example, was turned to vent, and observations on detectors previously placed by the robot showed no resulting effect on the nearby neutron field. The culmination of many such actions was the reasonably assured safe entry by personnel who, still by operations moderately remote,

drained much of the liquid from tank K-9. Cadmium nitrate solution was added to all vessels in the Recuplex system not geometrically favorable and all parts of the system were sampled.

Instrumentation was positioned (some by the robot) whereby the neutron field could be observed as the solution was removed from tank K-9 and an estimate could be made of a critical volume by an apparent neutron multiplication curve taken in reverse time. The conditions necessary for criticality in the cylinder were also calculated by reactor-physics computer codes based on reasonable plutonium concentrations and reflector conditions. These many considerations, not entirely internally consistent and suffering from unknowns in such basic parameters as the mass of plutonium involved (for example, solids bearing plutonium which might have been suspended during at least a part of the period were found on the bottom of the tank), allowed a formulation of a sequence of remarkably coherent events in view of the complexity of the normal process and, particularly, of the special-purpose operations in progress at the time.

The hypothesized sequence of events leading to and during the excursion is described in the following summary. Approximately 1450 g of plutonium in a solution with a concentration of less than 30 g of plutonium per liter was added to tank K-9 from the hood sump, and it occupied a subcritical volume. The slow addition of dilute nitric acid, bringing the total volume to perhaps 45 to 50 liters, gradually increased the reactivity and led to a small and slowly developing initial pulse of approximately 10^{16} fissions, consistent with personnel exposure. Repeated pulses were then formed and each was terminated by the presence of radiolytic gas bubbles until, after about 20 min, boiling ensued (at 60°C

under the pressure in tank K-9). As boiling proceeded to reduce the solution volume, the reactivity decreased, forcing a decline in power. Such a decline would be expected to be exponential and, if this analysis is correct, was in progress about the time the neutron recorder came back on scale. After 2 to 3 liters of solution had boiled off, the declining reactivity could no longer maintain bulk boiling; this condition developed about 1 hr after initiation of the reaction. Following termination of boiling, evaporative cooling and heat losses to the environment reduced the temperature and thereby added slight reactivity through the negative temperature coefficient and established a quasi-equilibrium condition that persisted until the solution reached ambient temperature. Then, with the source of reactivity no longer present, the system became subcritical owing to further evaporation which, in fact, continued until the evacuation system was valved off. Since the volume of the solution in tank K-9 at the time of personnel re-entry was 39 liters, approximately 10 liters was removed by boiling and evaporation.

Restoration of Operations

Processes other than Recuplex occupying the same general area at Hanford were shut down at the time of the accident on April 7 as a precaution against personnel exposures that might arise from the continuing or aggravated nuclear reaction. Processes in buildings other than Building 234-5, that is, those in buildings beyond a 1500-ft-radius exclusion area, were reactivated on April 16. Those within Building 234-5, except Recuplex, were restarted on April 30. As of April 1963, the Recuplex processing system had not been returned to service, and it will probably be replaced by a more modern salvage system.

Comments and Conclusions

The criticality accident that occurred in the Hanford Operations in April 1962 is the latest of the remarkably few accidents of this kind that have occurred in the country's chemical and metallurgical processing of fissile materials. The fact that its consequences were not more severe is a tribute to a well-planned and well-organized emergency procedure whereby personnel evacuated the affected area in a prompt and orderly manner. As has been true in related instances elsewhere, the cause was not an unexpected physical or chemical phenomenon but, according to the most plausible reconstruction of events, was a combination of shortcomings, some in the design of the process and the equipment, some in supervision and administration, and some in the operation itself. The value of the accident to other operations is a re-emphasis of the need for care and caution on the part of all persons concerned in order to more completely assure safe operations.

Although there were many items that contributed to the accident, it is difficult to single out dominant ones, particularly since there are uncertainties in the description of the way in which it happened. Measures that would correct many of these items can be read into the description of the occurrence. It is possible, in the opinion of this reviewer, to make two general comments. Salvage operations, because of the variety, nonuniformity, and uncertainty in the process materials, must be clearly prescribed and carefully carried out. This need is particularly apparent in those processes with fissile materials where the safety of the processing depends, in part, upon parameters administratively controlled, such

as the chemical concentration of process solutions, and, in part, upon more firmly fixed controls, such as the dimensions of equipment. Transfers of material between these areas must be done carefully and with an understanding of the concomitant shift in the manner of assuring safety.

The second comment has to do with an apparently simple matter — good housekeeping — upon which all types of safety so strongly depend. It is rather apparent, in retrospect, that the recovery process was made difficult by poor visibility of operations, that the source of the plutonium existed unnoticed for the same reason, and that this particular cleanup was necessitated by a long-standing accumulation of waste materials.

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