ICPP Criticality Event of October 17, 1978

By Nuclear Safety Staff

[Editor's Note: This article is adapted from Investigation of the 10-17-1978 Criticality Incident in the Uranium Extraction Process at the Idaho Chemical Processing Plant, a report issued by the Idaho National Engineering Laboratory in November 1978. The probable cause of the criticality incident described in this report was the failure of management to rectify deficiencies in both administrative control and instrumentation, both of which had been previously identified as being required. However, the incident produced no personnel injury, neither onsite nor offsite contamination, and no damage to equipment or property.]

The Idaho Chemical Processing Plant (ICPP) is owned and administered by the U.S. Department of Energy. The plant is operated by Allied Chemical Corporation and is one of the principal facilities at the Idaho National Engineering Laboratory located west of Idaho Falls, Idaho.

The primary purpose of the ICPP is the recovery of uranium from spent reactor fuel. The fuels processed consist primarily of uranium clad in aluminum, zirconium, or stainless-steel alloys. Fission products formed by the fissioning of uranium are also present in the fuel and are separated from the uranium during processing.

Spent fuel elements from test, research, and power reactors plus elements from the U.S. Navy’s ship propulsion reactors are received and stored at the ICPP before processing. The actual processing begins with dissolution of the fuel in acid. The reaction products, containing uranium and the fission products, are subsequently treated to reduce the corrosivity of the solution and to convert the uranium compounds to uranyl nitrate. This solution is then brought into contact with an organic solvent, tributylphosphate (TBP), in a countercurrent perforated-plate column where selective extraction of the uranyl nitrate by the solvent separates the uranium from the radioactive fission products. The uranium is stripped from the solvent by a weak nitric acid solution and, to purify it further, is processed with another organic solvent in two more extraction stages. The product of the third extraction cycle is a uranium nitrate solution from which practically all fission products and other impurities have been removed. The uranyl nitrate is then converted to granular uranium oxide for safe handling, and it is shipped to off-site fuel-fabrication facilities for return to the nuclear fuel cycle.

The criticality incident occurred in the first-cycle (TBP) extraction system previously mentioned. The equipment associated with this system is located in heavily shielded cells and adjacent areas within the process building (building CPP-601) at the ICPP.

DESCRIPTION OF THE INCIDENT

On Oct. 17, 1978, and immediately before, the first-cycle solvent extraction system was in operation recovering highly enriched uranium from salvage solutions. A flowsheet for this operation is shown in Fig. 1. The first-cycle solvent extraction system shown in Fig. 1 consists of extraction (1A), scrubbing (1B), stripping (1C), and solvent washing (1D) columns, with an intercycle product evaporator. The auxiliary equipment is similar to that found in a conventional Purex cycle.

On about September 18 a gradual decline in the concentration of aluminum nitrate in the 1BS [aqueous scrub stream feeding column H-100 (1B)] started to occur. This was not detected until after the
criticality event because of the arrangement of the makeup tank (PM-106-0) and feed tank (PM-107-0) for this stream and the absence of the density recorder alarm instrument on the stream. The makeup tank is sampled for composition control. A material balance of the aluminum content of the makeup tank, which was made after the event, traces the chronology of the decline in aluminum concentration. This aluminum material balance is based on solution volumes and after-the-fact analyses of samples that had been taken for impurity analysis (Table 1).

During normal operation of the solvent extraction cycle with 0.75 M aluminum scrub solution, a small concentration (and mass) of uranium is returned to the extraction column feed via the 1BR stream. As the aluminum nitrate concentration in 1BS and 1BR decreases, the distribution coefficient for uranium [defined as the equilibrium concentration in the organic phase divided by the concentration in the aqueous phase (\(D_{o/a}\))] decreases, and the uranium concentration in the 1BR increases. Data for the distribution of uranium are given in Table 2. On the basis of these data and on consideration of the recycle condition in the system, it is estimated that on the evening of October 17 a concentration of ~21 to 22 g of uranium per liter prevailed in the 1BR stream which fills the base of the column.

With the lower concentration of aluminum nitrate in the scrub solution (1BS), uranium concentration profiles in the 1B column change, with sharply higher uranium concentrations developing in the aqueous phase in the base of the column. Conceptual descriptions of the uranium concentration profiles under normal and low aluminum scrub solution concentrations are shown in Figs. 2 and 3, respectively. Under these conditions with higher masses of uranium being

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<th>Analyzed</th>
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<tr>
<td>9/15/78</td>
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<td></td>
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<tr>
<td>9/16/78</td>
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<td></td>
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<tr>
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<td></td>
</tr>
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<td>9/27/78</td>
<td>0.43</td>
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<td>10/14/78</td>
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<td>10/16/78</td>
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<td></td>
</tr>
<tr>
<td>10/17/78</td>
<td>0.08</td>
<td></td>
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<tr>
<td>10/18/78*</td>
<td></td>
<td>0.084†</td>
</tr>
</tbody>
</table>

*The plant was in intermittent operation during this period.
† Analyzed after the fact.
**Table 2** Distribution of Uranium into 5% TBP as a Function of Aluminum Nitrate Concentration*

<table>
<thead>
<tr>
<th>$\text{Al(NO}_3)_2$, M</th>
<th>$\text{HNO}_3$, M</th>
<th>Distribution coefficient ($C^o/A$) for uranium</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0</td>
<td>0.0</td>
<td>0.003</td>
</tr>
<tr>
<td>0.25</td>
<td>0.0</td>
<td>0.5</td>
</tr>
<tr>
<td>0.5</td>
<td>0.0</td>
<td>1.2</td>
</tr>
<tr>
<td>0.25</td>
<td>0.5</td>
<td>1.9</td>
</tr>
<tr>
<td>1.0</td>
<td>0.0</td>
<td>13</td>
</tr>
<tr>
<td>0.25</td>
<td>1.0</td>
<td>15</td>
</tr>
</tbody>
</table>

*In the chemical environment normally present at the base of the IB column (0.75 M aluminum nitrate and ~0.1 M HNO$_3$), the uranium is held in the organic phase by a relatively high distribution coefficient. However, as the aluminum nitrate concentration drops below 0.5 M, the distribution coefficient decreases rapidly, and uranium is forced into the aqueous phase.

On October 17, during the period of approximately 8:15 to 8:40 p.m., a criticality event occurred in the base of the IB column, H-100. The inventory of medium- to short-lived fission products used to determine the number of fissions (Table 3) indicates that the criticality occurred in column H-100 aqueous phase, and the sampling of the column wall, with counting of the filings, clearly indicates that the event occurred in the column base (Fig. 4). As shown in Table 3, this criticality event produced an estimated $2.74 \times 10^{18}$ fissions.

Figure 2 shows that a small increase in uranium concentration exists in both the aqueous and organic phases at about the midpoint of the column during normal operation. The increase is due to internal circulation (reflux) of uranium between the two phases. This occurs frequently in solvent extraction columns and is an accepted and sometimes desirable phenomenon.

Under abnormal conditions (Fig. 3), the uranium concentrations associated with this internal recirculation are quite high, representing a substantial mass of uranium at relatively high concentration literally suspended in the small-diameter (203.2-mm) section of the column. We believe that at 8:40 p.m. on October 17 the solution in the base of H-100 had been in a delayed critical condition at ~22.2 g of uranium (82)* per liter (at 20°C). The power production rate can be expected as $21 \exp (1.67 t)$ kW, with $t$ in hours.

*Uranium(82) indicates uranium enriched to 82% $^{235}$U.
The excursion appeared to have a stable positive period of 0.6 h on the basis of the two process makeup area constant air monitors. This period corresponds to a $k_{eff}$ of 1.00004, or 0.66 excess reactivity. As the power rate was rising, the temperature would also increase. The reactivity would decrease owing to the temperature rise by some 0.0001 $\Delta k/^{\circ}\text{C}$; so the average lower head concentration would need to rise by some 0.006 g of uranium per liter to compensate for each degree of temperature increase. The rise in temperature would then cause both a direct negative-reactivity effect and thermal mixing with the higher uranium concentration in the central column to increase reactivity. The two effects appear to have balanced each other over the 25 min of the stable period. There was also an increase in concentration due to continuing extraction from the organic phase.

The operator noticed a perturbation in the column instrumentation and took the corrective action of manually reducing the pressure on the jackleg and allowing a substantial flow of aqueous-phase solution out of the bottom of the column and through the jackleg. This would allow the more concentrated solution from the column (Fig. 3) to flow into the base of the column. This would raise the $k_{eff}$ of the lower head from the stable 1.00004 to prompt critical. The period would change from 36 min to some $10^{-6}$ s. The creation of voids associated with radiolysis gases and likely localized boiling and with temperature rise probably quenched the nuclear reaction temporarily.

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**Table 3** Number of Fissions—Summary of Vessel Analyses

<table>
<thead>
<tr>
<th>Vessel</th>
<th>Measured fissions*</th>
<th>Percent of event</th>
</tr>
</thead>
<tbody>
<tr>
<td>H-100 organic</td>
<td>$3 \times 10^{16}$</td>
<td>1</td>
</tr>
<tr>
<td>H-100 aqueous</td>
<td>$2.66 \times 10^{15}$</td>
<td>97</td>
</tr>
<tr>
<td>H-103 organic</td>
<td>$1 \times 10^{15}$</td>
<td>0.04</td>
</tr>
<tr>
<td>H-103 aqueous</td>
<td>$5.7 \times 10^{14}$</td>
<td>2.1</td>
</tr>
<tr>
<td>H-108</td>
<td>$8 \times 10^{2}$</td>
<td>0.01</td>
</tr>
<tr>
<td>H-126</td>
<td>$2 \times 10^{2}$</td>
<td>0.01</td>
</tr>
<tr>
<td>H-131</td>
<td>$8 \times 10^{1}$</td>
<td>0.01</td>
</tr>
<tr>
<td>H-134</td>
<td>$1 \times 10^{1}$</td>
<td>0.04</td>
</tr>
<tr>
<td>H-136</td>
<td>$9 \times 10^{1}$</td>
<td>0.01</td>
</tr>
<tr>
<td>G-108</td>
<td>$3 \times 10^{1}$</td>
<td>0.01</td>
</tr>
<tr>
<td>G-111</td>
<td>$2.4 \times 10^{1}$</td>
<td>0.07</td>
</tr>
<tr>
<td>Total</td>
<td>$2.74 \times 10^{18}$</td>
<td>100</td>
</tr>
</tbody>
</table>

*Based on average number of fissions, as measured by $^{144}\text{Ce}$ and $^{137}\text{Ce}$.  
†Single analyses and uncertain sample homogeneity; fissions based on $^{93}\text{Sr}$.  

Final shutdown was achieved by either of two mechanisms: (1) the flow of aqueous-phase solution out through the jackleg (dumping) swept out the concentrated solution, and a subcritical concentration was achieved due to the flow of dilute aqueous-phase solution from the upper portion of the plate section of the column into the column base; or (2) the nuclear reaction was suppressed by elevated temperature during the brief period that it took the operator to shut down the streams and set the jackleg controller on manual at full pressure (no flow from the base of the column). Under these circumstances, the continued operation of the pulser would cause mixing throughout the column and lead to subcritical concentrations in the base. The preceding interpretation and the operator's statements are consistent with the instrument strip charts for the three column instruments—the interface controller, the jackleg pressure, and the column liquid level.

**CONCLUSIONS**

**Findings**

1. The aluminum nitrate feed to column H-100 was not within specifications and caused the aqueous solution to become a stripping agent rather than a scrubbing agent. This resulted in a buildup of uranium within columns G-111 and H-100 to the point that a critical concentration was ultimately reached in H-100.
2. The potential for criticality in the event that out-of-specification aluminum nitrate was used as feed had been recognized before the incident occurred. Section 6.3 of the safety review document indicated that instrumentation was provided to detect and warn of this condition. However, installed instrumentation had become inoperative before the incident, and additional instrumentation requested by Operations had not yet been installed.

3. Documented requirements for sampling of feed tank PM-107-0 existed and, if followed, would have prevented the incident. Several revisions of the procedure (Standard Operating Procedure 4.16.0) were available to operating personnel, both with and without the requirement for sampling PM-107-0.

4. Information indicating the gradual dilution of the material in PM-106-0 was available but was not noted by personnel. Consequently, corrective actions were not taken.

5. A safety review document had been prepared and published in 1974 for the extraction process. This document identified the criticality risk if the aluminum nitrate scrub feed were to become dilute but also incorrectly assumed that stoppage of the scrub feed was necessary. In any case, appropriate safety limits and limiting conditions for operation had not been formulated to ensure that diluted scrub solution would not be used. Further, the safety review document did not define the operator actions expected if instrument alarms on the scrub solution column did occur.

6. There is a lack of formality in the conduct of operations and the adherence to written requirements—specifically, lack of compliance with standard operating procedures and assignment of uncertified personnel to perform operating tasks.

7. Management systems in place are not adequate to assure control of plant configuration—specifically, a steam hose connected to tank PM-107-0 which should have been removed and omission of a functioning density recorder alarm on either tank PM-106-0 or tank PM-107-0.

8. Feedback systems within Allied Chemical Corporation do not appear adequate to provide definitive information as to the lack of discipline and formality with which operations are conducted. However, in spite of the system weakness, almost all supervisory and management people interviewed were aware that procedural compliance was not the practice.

9. Instrument maintenance and calibration are not being performed based on known equipment failure rates or other equipment reliability determinations.

10. The criticality occurred in a heavily shielded and suitably ventilated cell with the result that there was no personnel injury, no on- or off-site containment, and no damage to the equipment or property.

Probable Cause

The probable cause of this incident was the failure of the management and review systems to provide for or to prevent the deterioration of administrative controls and alarming instrumentation, both of which had previously been identified as being required. These omissions resulted in an inadvertent criticality in a column in the first cycle of the uranium extraction system.

Judgment of Needs

1. Conservative safety limits [safety limits and requirements (SL&R) and limiting condition for operation (LCFO)] for parameters important to the criticality safety of the fuel extraction operation should be determined and incorporated into the operating restraints.

2. The Safety Review Board (SRB) should be formally charged with a search-out function to keep informed on plant and operational status and to follow up to ensure that SRB requirements are carried out.

3. Consideration should be given to transferring responsibility for safety review document (SRD) preparation to the Technical Support unit which should have primary responsibility for the SRD. As a minimum, the interface between those currently preparing the SRD (Operational and Environmental Safety) and the Technical and Operating units should be improved.

4. A management policy requiring strict compliance with procedures and other requirements important to safety (such as emergency plans and training requirements) should be thoroughly and effectively implemented.

5. The document control system should be strengthened to ensure that current policies, Standard Operating Procedures, or other requirements important to safety are available to operating personnel and that outdated documents are withdrawn.

6. Administrative requirements should be strengthened to ensure that all appropriate personnel are familiar with new requirements [Standard Operating Procedures (SOPs) Emergency Plans, etc.] at the time of their issuance.

7. The independent audit function should be strengthened. Coverage should be greatly expanded,
and frequent audits of changes should be made as they take place.

8. Requirements to control physical changes to the plant should be developed and implemented. These requirements should include inspection and acceptance of completed work by the operating unit.

9. Those positions within the plant requiring certified personnel should be formally defined.

10. A more comprehensive and formal maintenance and calibration program should be instituted. The program to obtain data to determine maintenance and calibration frequencies should be better implemented.

11. Training for postcriticality recovery should be formally included in the training program.

12. Sufficient analyses should be performed of off-normal conditions to allow definition in the SOPs of actions to be taken by operators in the event various operating instruments sound an alarm.

13. Requirements for operational readiness reviews should be formally established and implemented.

14. Interfaces between Technical Support and Operations and between Maintenance and Operations should be strengthened to ensure proper support for plant production operations.

15. Technical details such as time history of aluminum nitrate dilution, uranium buildup, and subsequent criticality should be reconstructed and documented by Allied Chemical Corporation.

Events Resulting in Reactor Shutdown and Their Causes

Compiled by R. L. Scott and R. B. Gallaher

The licensee event reports (LERs) that were received at the Nuclear Safety Information Center (NSIC) in May and June 1980 were reviewed, and those events which involved a reactor shutdown are listed in Table 1. Both operating research reactors and operating commercial power reactors were considered.

Prior to Vol. 19 of *Nuclear Safety*, each issue carried a more extensive listing of reportable events; now as a space-saving measure only those events involving a reactor shutdown are listed. Thus Table 1 presents only a fraction of the reportable events submitted to the Nuclear Regulatory Commission and accessioned by NSIC. Additional information on these events can be obtained by contacting the NSIC (see the inside front cover) where a continuously updated computerized file of the reportable events has been maintained for more than 12 yr.

The format used to list the reported shutdowns has been developed to concisely present the more important data. The first column gives the report date, under which is the date of the event. A brief description of the event is in the second column, and the cause is in the third. The fourth column gives the component involved, and immediately below is the system involved. Abbreviations are used for many of the system titles because of their length. In the last column is the facility name, and immediately below that is the docket number. With this information, interested readers can obtain copies of the reports from the Nuclear Regulatory Commission Public Document Room, 1717 H Street NW, Washington, D.C. 20555.

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<tr>
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<td>La Crosse 50-409</td>
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<td>Vital Bus Inverter Failure Results</td>
<td>Electrical</td>
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<td>092079</td>
<td>In Reactor Trips and Safety Injects</td>
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<td>122879</td>
<td>Reactor Coolant Leakage Into</td>
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<tr>
<td>122879</td>
<td>Safety Pressurizer Relief Tank Caused EML</td>
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(Table continues on the next page.)
This accident occurred in a shielded cell of a fuel reprocessing plant where solutions from the dissolution of irradiated reactor fuel are processed to remove fission products and recover the enriched uranium using a solvent extraction process.

Solvent extraction is a process whereby immiscible aqueous and organic streams counterflow with intimate contact, and through control of acidity a material of interest is transferred from one stream to the other. In this operation (Fig. 1) the first stage (1A column) had as a feed into the column top the aqueous recovery solution containing less than 1 gm enriched uranium per liter. Less dense organic, consisting of a mixture of tributyl phosphate and kerosene, was fed into the bottom of the column. A vertical string of perforated plates along the axis of the column was externally driven up and down to increase the effectiveness of contact between the two streams. This made it a "pulsed column". (Fig. 2) As the two streams passed through the pulsed column the uranium was stripped from the aqueous by the organic. The large diameter regions at the top and bottom of the column are disengagement sections where the aqueous and organic are given the opportunity to separate more completely. The aqueous waste stream (raffinate) from the bottom of the 1A column was sampled to verify compliance with discard limits before being sent to waste storage tanks. The organic product stream, containing about 1-gm uranium per liter, from the top of 1A became feed into the bottom of the scrub column 1B.

In 1B the organic product was contacted by a clean aqueous stream to scrub out residual fission products. This aqueous stream, fed into the top
Fig. 1 Simplified flowsheet for first-cycle extraction.
Fig. 2 Typical column.
Idaho Chemical Processing Plant

of IB, was buffered with aluminum nitrate to a concentration of 0.75 M to prevent significant transfer of uranium from the organic to the aqueous. In normal operation some uranium would be taken up by the aqueous, to a concentration of about 0.15 g/L, so the aqueous output of IB was fed back and blended with the dissolver product going into IA. The organic product stream from IB, normally about 0.9 g uranium per liter, went on to IC where the uranium was stripped from the organic by 0.005 M nitric acid. The output of the stripper column went to mixer settlers where additional purification took place.

Further downstream the uranium solution went to an evaporator where it was concentrated to permit efficient precipitation of the uranium.

As is often true, several factors contributed to this accident. An evaporator had plugged and operations were suspended for several weeks while instrumentation difficulties were corrected. During this downtime a valve leaked water into the aluminum nitrate makeup tank used for preparation of the aqueous feed to the scrubber column (IB). This leakage over time diluted the feed solution from 0.75 M to 0.08 M. The 13,400 liter makeup tank had been equipped with a density gauge that would have indicated this discrepancy but this gauge was not operable. A density gage was to be installed on the 3,000 liter process feed tank which was filled as necessary from the makeup tank. This density gauge had not yet been installed on the feed tank. The makeup tank was instrumented with a strip chart recorder showing the solution level in the tank, but the leak into the tank was so slow that the change in level was not discernable
without pulling out several days of the chart length. Procedures required that the density in the process feed tank be obtained after each transfer from the makeup tank. Results of sample analyses were not available until after the accident.

The out-of-specification aqueous feed to the scrubber column caused it to operate as a stripper rather than as a scrubber. Much of the enriched uranium was removed from the 1B organic and recycled into the input of 1A. This partially closed loop resulted in a steady increase in the uranium inventory in these two columns.

Each time diluted solution was added to the feed tank from the makeup tank, the aluminum nitrate concentration in the feed was further reduced, and stripping became more effective.

Analyses of the aqueous feed for the 1B column (feed tank PM-107-0) showed the proper 0.7 M aluminum nitrate on September 15, 1978. Samples taken on September 27 and October 18 had concentrations of 0.47 M and 0.084 M.

Concentrations of aluminum nitrate less than 0.5 M are insufficient to prevent some stripping of uranium from the organic, and the final concentration would result in almost all the uranium being stripped from the organic.
CHRONOLOGY OF ACCIDENT

The process feed tank (PM-107-0) was filled with aluminum nitrate solution from the make-up tank (PM-106-0) at approximately 6:30 PM on October 17. Chemical analysis of this solution (available after the accident) showed an out-of-specification 0.08 molarity. Approximately an hour and a half later the process operator was having difficulty controlling pulsed column H-100. During his efforts to maintain proper operation he reduced the pressure on the jackleg, thus permitting increased aqueous flow from H-100 back to G-111. At approximately 8:40 PM a radiation alarm activated, probably because of fission products in the plant stock gasses. Shortly after this alarm, several others activated and the stack monitor gave a full-scale reading. The Shift Supervisor and the Health Physicist went outside the building and detected radiation intensities up to 100 millirem/hr. At 9:03 PM the shift supervisor ordered the building evacuated, and by 9:06 an orderly evacuation had been accomplished. Appropriate road blocks were established and proper notification was provided management.

In the evacuation the process operator shut off all feed to the first-cycle extraction process, but did not stop the pulsation of the columns.

It seems probable that as the uranium inventory in the bottom of H-100 increased because of the lean ammonium nitrate scrub solution, the system achieved the delayed-critical state. As further uranium was carried down to the lower section the system became slightly super critical and the
increasing power level raised the temperature to compensate for the presence of additional uranium. This process would continue so long as the uranium addition was slow and until the reduced pressure on the jacklog permitted more rapid addition of uranium and a sharp increase in reactivity. The system is thought to have approached prompt criticality, at which time the rate of power increase would have been determined by the neutron lifetime which would be on the order of milliseconds.

The continuation of the pulse action after the feed was turned off probably led to improved mixing of the solution in the bottom section of H-100 and terminated the reaction.

**A POSSIBLE COURSE OF THE ACCIDENT**

The last sample from PM-107-0 prior to this evening of the accident was on September 29, some 19 days earlier. The process was not in operation from September 29 until October 13, because of difficulties with a down-stream evaporator. Operations were conducted from October 13 till October 16, when there was a brief interruption with restart the same day. The September 29 sample indicated an aluminum nitrate concentration in the aqueous feed to the H-100 column of 0.47 M, but this result was not available until after the accident.

At 6:30 PM, when PM-107-0 was last filled, the uranium inventory in H-100 must have been about 5 kg as a result of extended operation with below-specification aluminum nitrate solution.

During the 2.5 hours between the time the feed tank was filled with very lean solution and the turning off of all feed to the process, the inventory in
the first two stages of the process would have increased by an amount equal to the input to the process, or 566 liter/hr. x 0.79 g/l x 2.5 hrs., or slightly more than 1 kg. Most of this increase would appear in H-100 in the 150 liters of lean aqueous added after 6:30. This material, coming down the column, would be at a concentration about 7 g/liter greater than the aqueous already in the bottom section of H-100 (~18 g/l) but at a lower density (~1.04 vs ~1.12). Depending on the amount of mixing that took place, the bottom section of the tank might have been about half full of material at ~18 g/liter with 25 g/liter solution on top. This configuration would have been more reactive than a uniform solution at the average concentration, this explaining the termination of the reaction as mixing continued.

While this explanation is clearly conjectural, it does seem to fit the observed situation.

The reaction clearly took place in the lower section of H-100, with most of the fissions occurring in the upper portion of that section. Records indicate the reaction rate increased very slowly until late in the sequence, when a sharp rise in power occurred. The total number of fissions during the reaction was estimated to be $2.7 \times 10^{18}$ or an energy release of about 165 m wt sec. The average power level during the approximately 1/2 hour of the reaction was then a little less than 100 kwt.
No significant personnel exposures occurred and there was no damage to process equipment. As a direct result of this event the plant suffered an extended and expensive shutdown as all operating procedures were reviewed in detail and revised as appropriate. Increased emphasis was given to plant maintenance and operator training.

In summary, the need for adherence to well-developed operating procedures was re-emphasized by this accident.
Fig. 6  Simplified flowsheet for first-cycle extraction.
This accident occurred in a shielded cell of a fuel reprocessing plant where solutions from the dissolution of irradiated reactor fuel are processed to remove fission products and recover the enriched uranium, using a solvent extraction process.

Solvent extraction is a process whereby immiscible aqueous and organic streams counterflow with intimate contact, and through control of acidity a material of interest is transferred from one stream to the other. In this operation (Fig. 1) the first stage (1A column) had as a feed into the column top the aqueous recovery solution containing less than 1 gm enriched uranium per liter. Less dense organic, consisting of a mixture of tributyl phosphate and kerosene, was fed into the bottom of the column. A vertical string of perforated plates along the axis of the column was externally driven up and down to increase the effectiveness of contact between the two streams. This made it a "pulsed column". (Fig. 2) As the two streams passed through the pulsed column the uranium was stripped from the aqueous by the organic. The large diameter regions at the top and bottom of the column are disengagement sections where the aqueous and organic are given the opportunity to separate more completely. The aqueous waste stream (raffinate) from the bottom of the 1A column was sampled to verify compliance with discard limits before being sent to waste storage tanks. The organic product stream, containing about 1-gm uranium per liter, from the top of 1A became feed into the bottom of the scrub column 1B.

In 1B the organic product was contacted by a clean aqueous stream to scrub out residual fission products. This aqueous stream, fed into the top
Fig. 1  Simplified flowsheet for first-cycle extraction.
Fig. 2 Typical column.
of 1B, was buffered with aluminum nitrate to a concentration of .75 M to prevent significant transfer of uranium from the organic to the aqueous.

In normal operation some uranium would be taken up by the aqueous, to a concentration of about 0.15 g/L, so the aqueous output of 1B was fed back and blended with the dissolver product going into 1A. The organic product stream from 1B, normally about 0.9 g uranium per liter, went on to 1C where the uranium was stripped from the organic by .005 M nitric acid. The output of the stripper column went to mixer settlers where additional purification took place.

Further downstream the uranium solution went to an evaporator where it was concentrated to permit efficient precipitation of the uranium.

As is often true, several factors contributed to this accident. An evaporator had plugged and operations were suspended for several weeks while instrumentation difficulties were corrected. During this downtime a valve leaked water into the aluminum nitrate makeup tank used for preparation of the aqueous feed to the scrubber column (1B). This leakage over time diluted the feed solution from 0.75 M to 0.08 M. The 13,400 liter makeup tank had been equipped with a density gauge that would have indicated this discrepancy but this gauge was not operable. A density gage was to be installed on the 3,000 liter process feed tank which was filled as necessary from the makeup tank. This density gauge had not yet been installed on the feed tank. The makeup tank was instrumented with a strip chart recorder showing the solution level in the tank, but the leak into the tank was so slow that the change in level was not discernable
without pulling out several days of the chart length. Procedures required that the density in the process feed tank be obtained after each transfer from the makeup tank. Results of sample analyses were not available until after the accident.

The out-of-specification aqueous feed to the scrubber column caused it to operate as a stripper rather than as a scrubber. Much of the enriched uranium was removed from the 1B organic and recycled into the input of 1A. This partially closed loop resulted in a steady increase in the uranium inventory in these two columns.

Each time diluted solution was added to the feed tank from the makeup tank, the aluminum nitrate concentration in the feed was further reduced, and stripping became more effective.

Analyses of the aqueous feed for the 1B column (feed tank PM-107-0) showed the proper 0.7 M aluminum nitrate on September 15, 1978. Samples taken on September 27 and October 18 had concentrations of 0.47 M and 0.084 M.

Concentrations of aluminum nitrate less than 0.5 M are insufficient to prevent some stripping of uranium from the organic, and the final concentration would result in almost all the uranium being stripped from the organic.
The process feed tank (PM-107-0) was filled with aluminum nitrate solution from the make-up tank (PM-106-0) at approximately 6:30 PM on October 17. Chemical analysis of this solution (available after the accident) showed an out-of-specification 0.08 molarity. Approximately an hour and a half later the process operator was having difficulty controlling pulsed column H-100. During his efforts to maintain proper operation he reduced the pressure on the jackleg, thus permitting increased aqueous flow from H-100 back to G-111. At approximately 8:40 PM a radiation alarm activated, probably because of fission products in the plant stock gasses. Shortly after this alarm, several others activated and the stack monitor gave a full-scale reading. The Shift Supervisor and the Health Physicist went outside the building and detected radiation intensities up to 100 millirem/hr. At 9:03 PM the shift supervisor ordered the building evacuated, and by 9:06 an orderly evacuation had been accomplished. Appropriate road blocks were established and proper notification was provided management.

In the evacuation the process operator shut off all feed to the first-cycle extraction process, but did not stop the pulsation of the columns.

It seems probable that as the uranium inventory in the bottom of H-100 increased because of the lean ammonium nitrate scrub solution, the system achieved the delayed-critical state. As further uranium was carried down to the lower section the system became slightly super critical and the
increasing power level raised the temperature to compensate for the presence of additional uranium. This process would continue so long as the uranium addition was slow and until the reduced pressure on the jacklog permitted more rapid addition of uranium and a sharp increase in reactivity. The system is thought to have approached prompt criticality, at which time the rate of power increase would have been determined by the neutron lifetime, which would be on the order of milliseconds.

The continuation of the pulse action after the feed was turned off probably led to improved mixing of the solution in the bottom section of H-100 and terminated the reaction.

A POSSIBLE COURSE OF THE ACCIDENT

The last sample from PM-107-0 prior to this evening of the accident was on September 29, some 19 days earlier. The process was not in operation from September 29 until October 13, because of difficulties with a down-stream evaporator. Operations were conducted from October 13 till October 16, when there was a brief interruption with restart the same day. The September 29 sample indicated an aluminum nitrate concentration in the aqueous feed to the H-100 column of 0.47 M, but this result was not available until after the accident.

At 6:30 PM, when PM-107-0 was last filled, the uranium inventory in H-100 must have been about 5 kg as a result of extended operation with belowSpecification aluminum nitrate solution.

During the 2.5 hours between the time the feed tank was filled with very lean solution and the turning off of all feed to the process, the inventory in
the first two stages of the process would have increased by an amount equal to the input to the process, or 566 liter/hr. x 0.79 g/l x 2.5 hrs., or slightly more than 1 kg. Most of this increase would appear in H-100 in the 150 liters of lean aqueous added after 6:30. This material, coming down the column, would be at a concentration about 7 g/liter greater than the aqueous already in the bottom section of H-100 (~18 g/l) but at a lower density (~1.04 vs ~1.12). Depending on the amount of mixing that took place, the bottom section of the tank might have been about half full of material at ~18 g/liter with 25 g/liter solution on top. This configuration would have been more reactive than a uniform solution at the average concentration, this explaining the termination of the reaction as mixing continued.

While this explanation is clearly conjectural, it does seem to fit the observed situation.

The reaction clearly took place in the lower section of H-100, with most of the fissions occurring in the upper portion of that section. Records indicate the reaction rate increased very slowly until late in the sequence, when a sharp rise in power occurred. The total number of fissions during the reaction was estimated to be $2.7 \times 10^{18}$ or an energy release of about 165 m\text{\textasciitilde}w\text{\textasciitilde}t\text{\textasciitilde}sec. The average power level during the approximately 1/2 hour of the reaction was then a little less than 100 kwt.
No significant personnel exposures occurred and there was no damage to process equipment. As a direct result of this event the plant suffered an extended and expensive shutdown as all operating procedures were reviewed in detail and revised as appropriate. Increased emphasis was given to plant maintenance and operator training. An extensive plant protection system was installed, including redundant automatic safety controls. In summary, the need for adherence to well-developed operating procedures was re-emphasized by this accident.
Fig. Simplified flowsheet for first-cycle extraction.
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