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LOS ALAMOS SCIENTIFIC LABORATORY OF THE UNIVERSITY OF CALIFORNIA ○ LOS ALAMOS NEW MEXICO

NUCLEAR-CRITICAL ACCIDENT AT THE LOS ALAMOS
SCIENTIFIC LABORATORY ON DECEMBER 30, 1958

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LOS ALAMOS SCIENTIFIC LABORATORY
OF THE UNIVERSITY OF CALIFORNIA LOS ALAMOS NEW MEXICO

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**NUCLEAR-CRITICAL ACCIDENT AT THE LOS ALAMOS
SCIENTIFIC LABORATORY ON DECEMBER 30, 1958**

by

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This report is an informal account of the findings of a Los Alamos Scientific Laboratory committee appointed to investigate the circumstances surrounding the nuclear-critical accident that occurred at the Laboratory in December of 1958. It has been prepared to distribute the information as promptly as possible and presents the facts known at the time of writing.

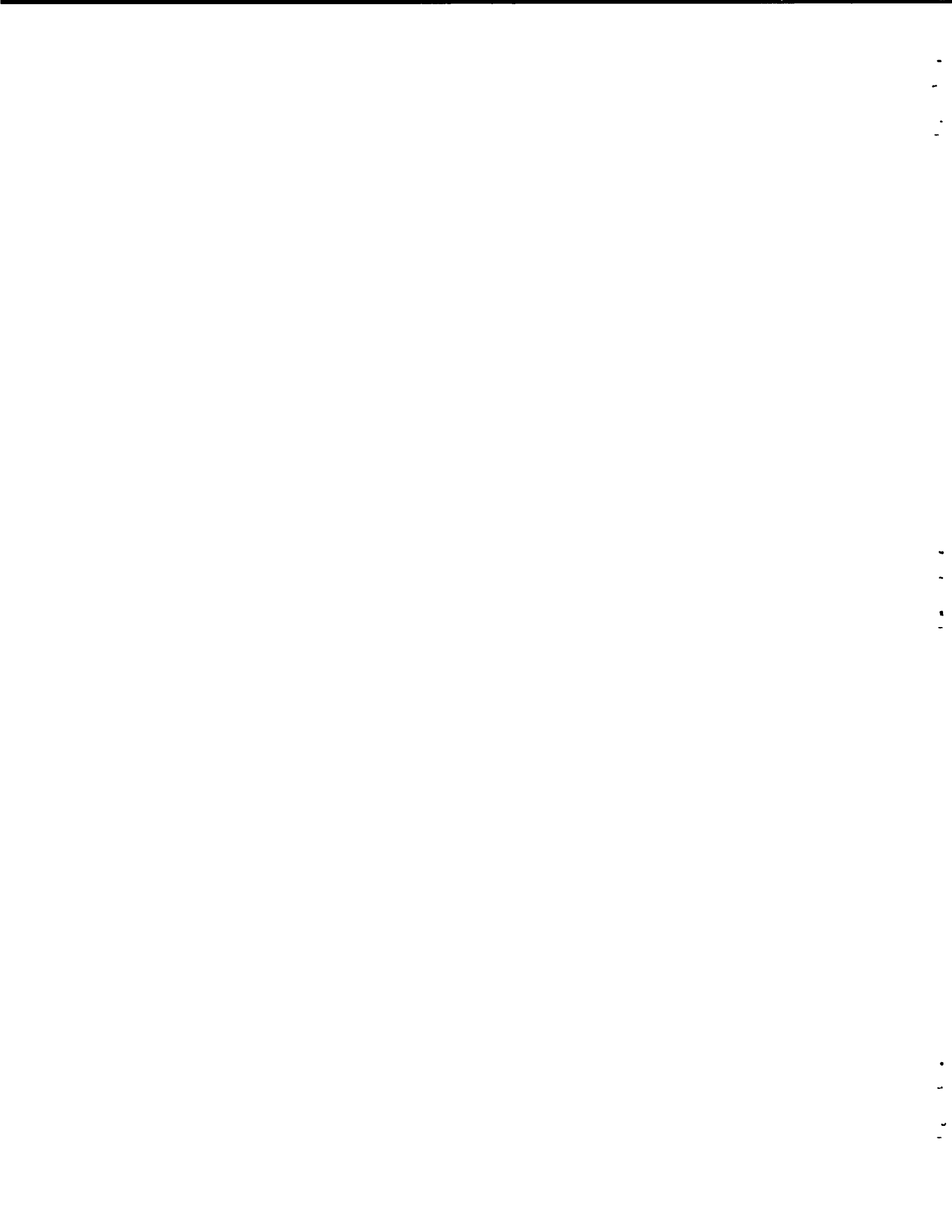


ABSTRACT

The nuclear-critical accident that occurred in the plutonium processing plant at the Los Alamos Scientific Laboratory on December 30, 1958, resulted from an unusual and complex set of circumstances. Reconstruction of the steps that preceded the accident and analysis of the materials involved give a reasonably specific picture of the conditions at the time of the radiation burst.

ACKNOWLEDGMENTS

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INTRODUCTION

On Tuesday, December 30, 1958, at 4:35 p.m., a nuclear-critical accident which resulted in the fatal radiation injury of a Laboratory employee occurred in the plutonium recovery operation at DP Site of the Los Alamos Scientific Laboratory. An official investigation of the circumstances surrounding the incident was conducted, and this report summarizes the findings concerning the events leading to the accident, the situation at the time of the critical burst, the removal and analysis of the solution that caused the burst, and the steps being undertaken to prevent a recurrence.

OPERATIONAL BACKGROUND

The layout of the site at which the accident occurred and the specific area involved (Rooms 213-218) are shown in Fig. 1. The part of the plutonium recovery operation involved in the accident was the tri-n-butyl phosphate (TBP) solvent-extraction process used to concentrate and purify plutonium from so-called "lean" residues, typically, solutions containing less than 0.1 g/liter of plutonium and traces of americium. A simplified flowsheet for the process is given in Fig. 2, and Fig. 3 shows the corresponding layout of equipment in the plant. Figures 4 to 9 delineate the steps involved in the procedures shown in Fig. 2.

Slag, crucible (from Metal Preparation), and other lean residues are appropriately dissolved, and the resulting solution is clarified by filtration. The removed solids are treated separately by the method shown in Fig. 4. The filtrate is contacted in columns with TBP carried in organic solvent to extract the plutonium, then the plutonium is back-extracted into an aqueous phase that is concentrated by evaporation. This operation is shown in Fig. 5. The concentrated product, which contains plutonium at a few grams per liter, is further purified by a TBP extraction in a mixer-settler operation (Fig. 6) and the product material removed for metal preparation.

The raffinate from the column extraction operation contains some residual plutonium and small amounts of americium. After chemical treatment of this solution (which causes the formation of a suspension of finely divided silicates), the plutonium and americium are removed by batch-contacting with

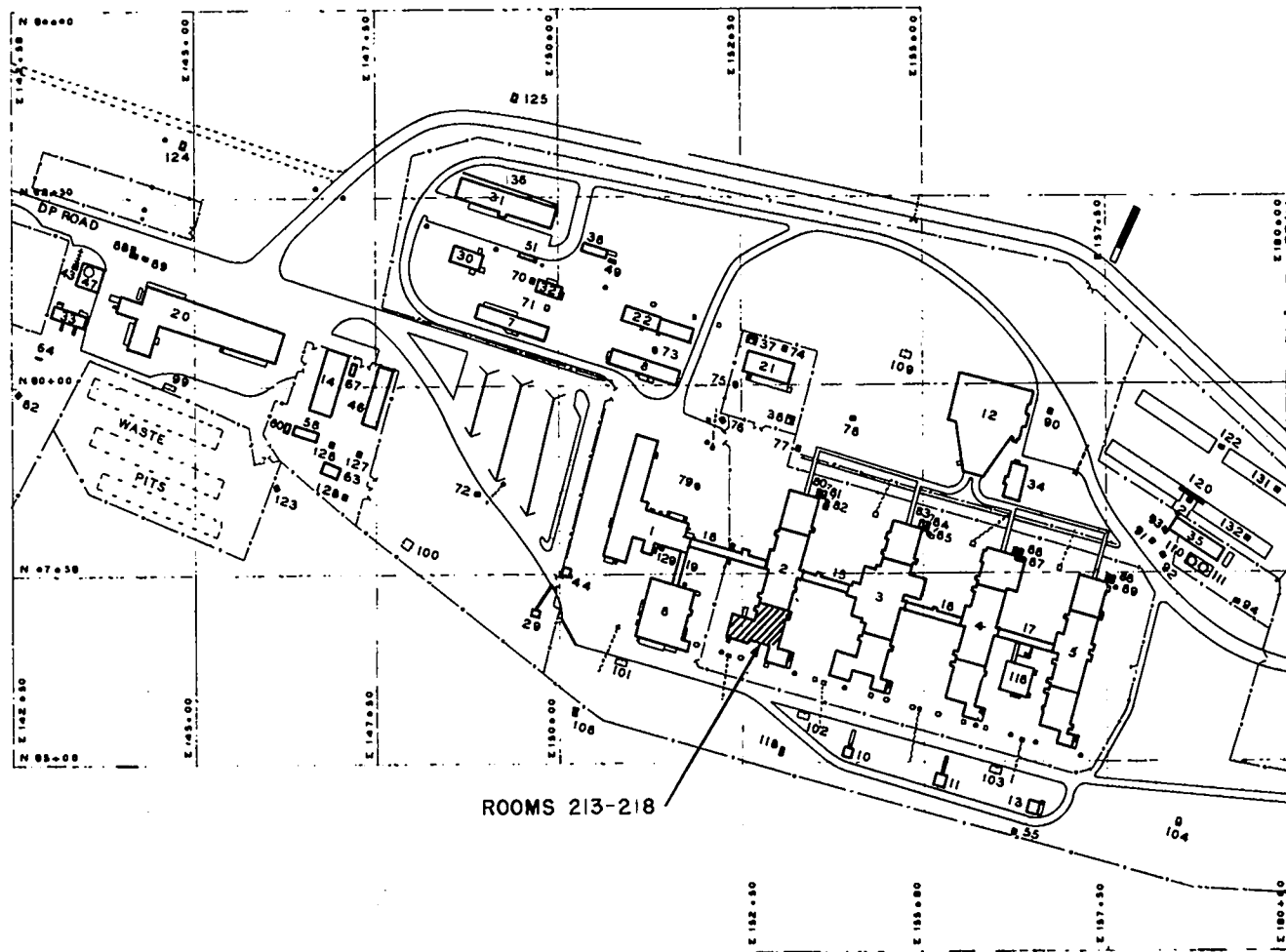


Fig. 1 Map of DP Site. Shaded area indicates location of accident.

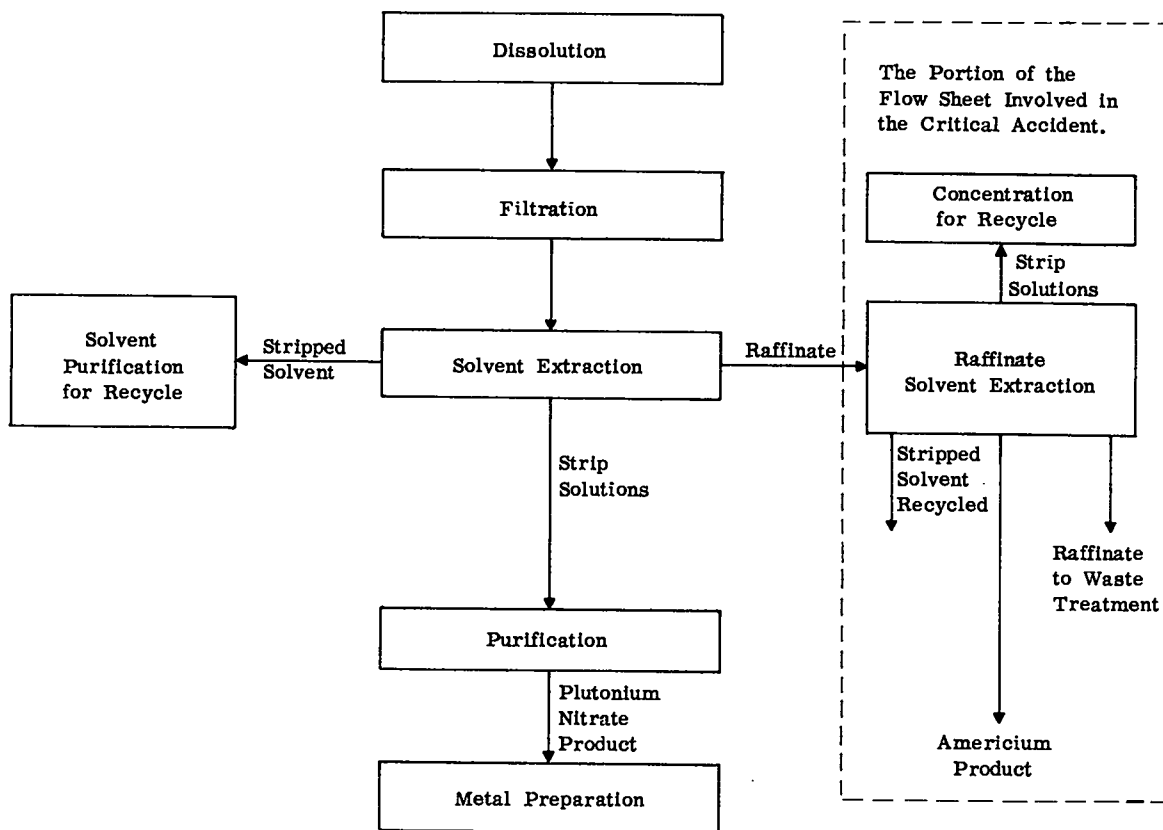


Fig. 2 Schematic flowsheet for the recovery of plutonium from lean residue.

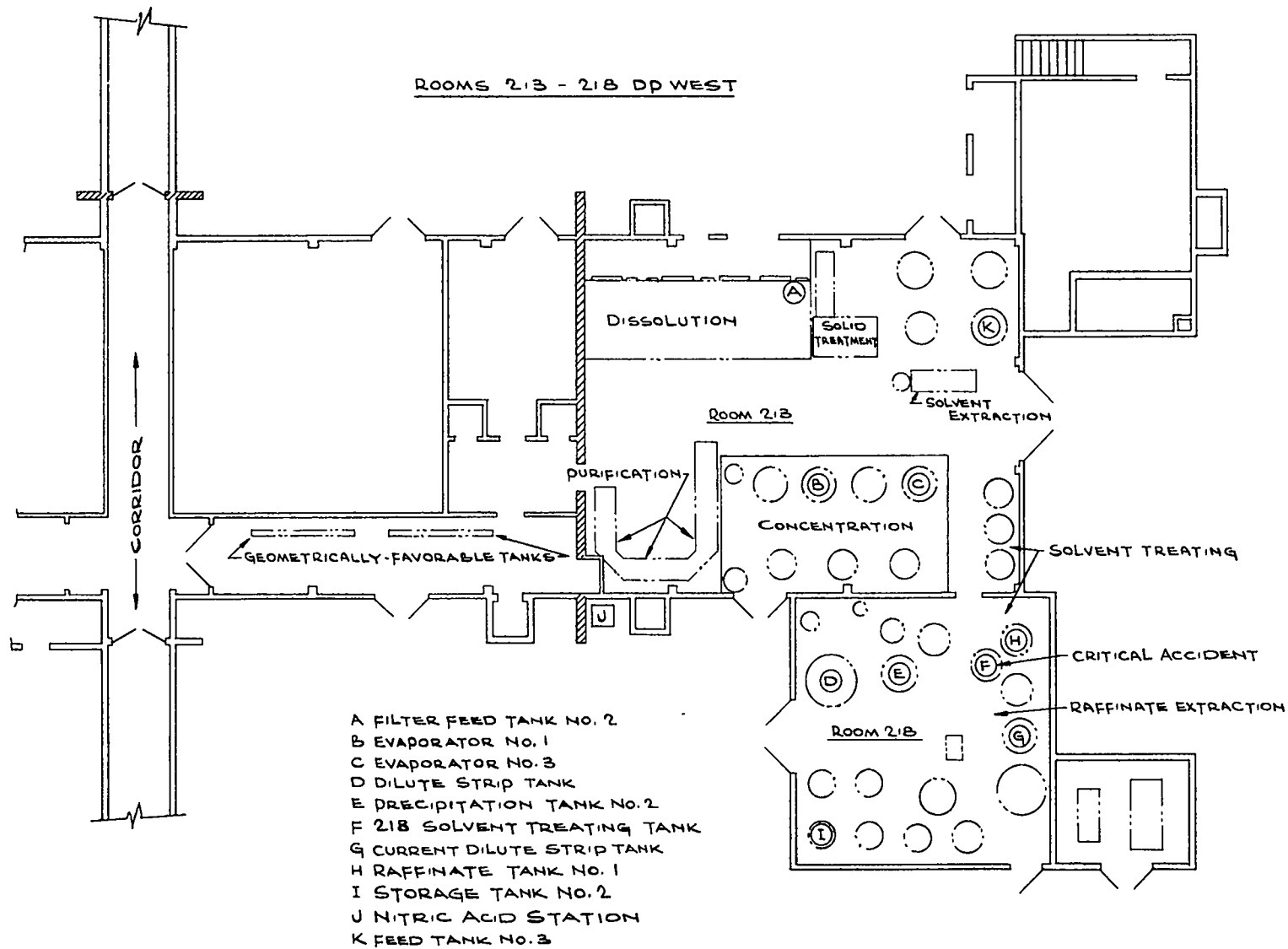


Fig. 3 Layout of equipment in area where accident occurred.

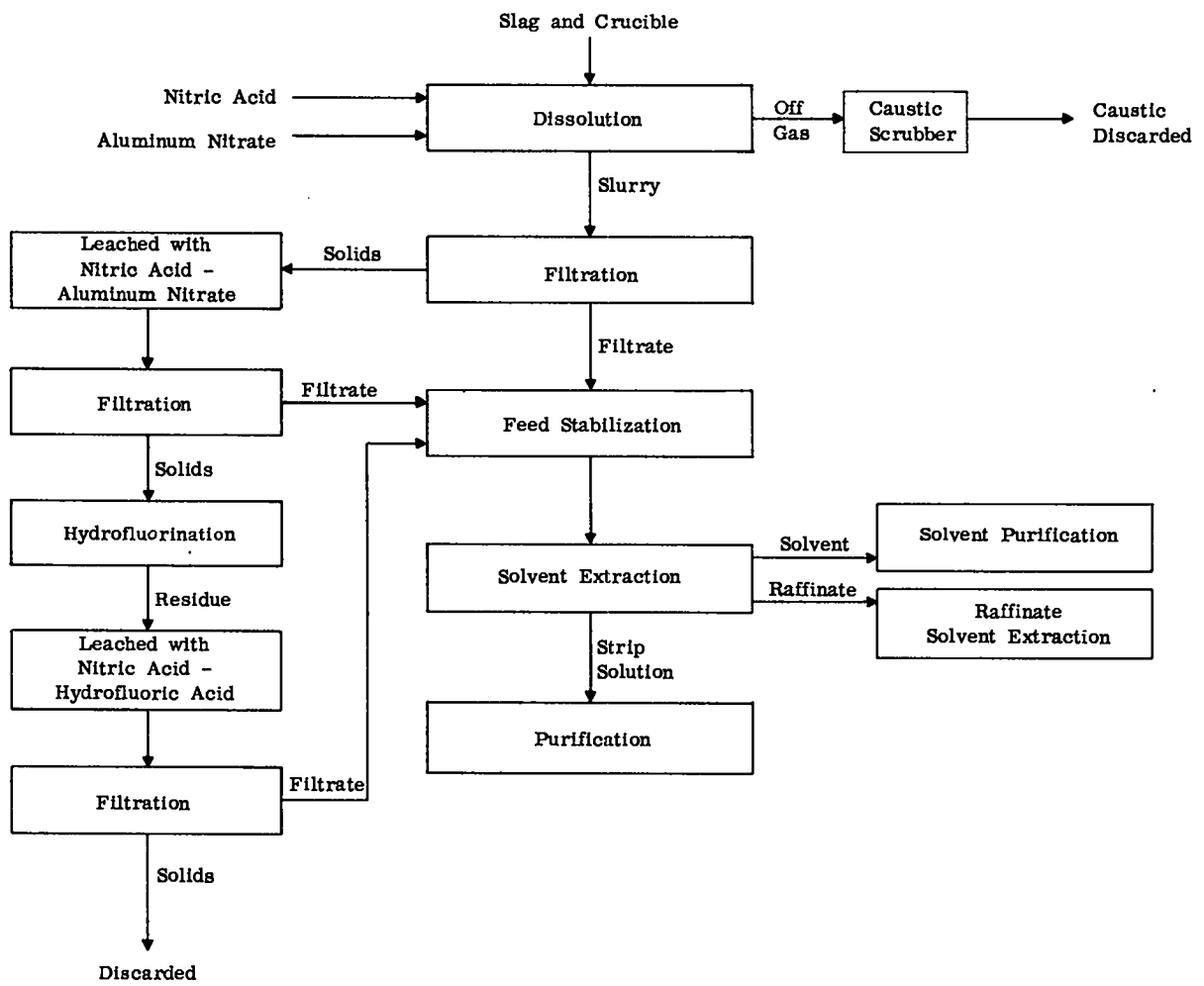


Fig. 4 Flowsheet showing dissolution process.

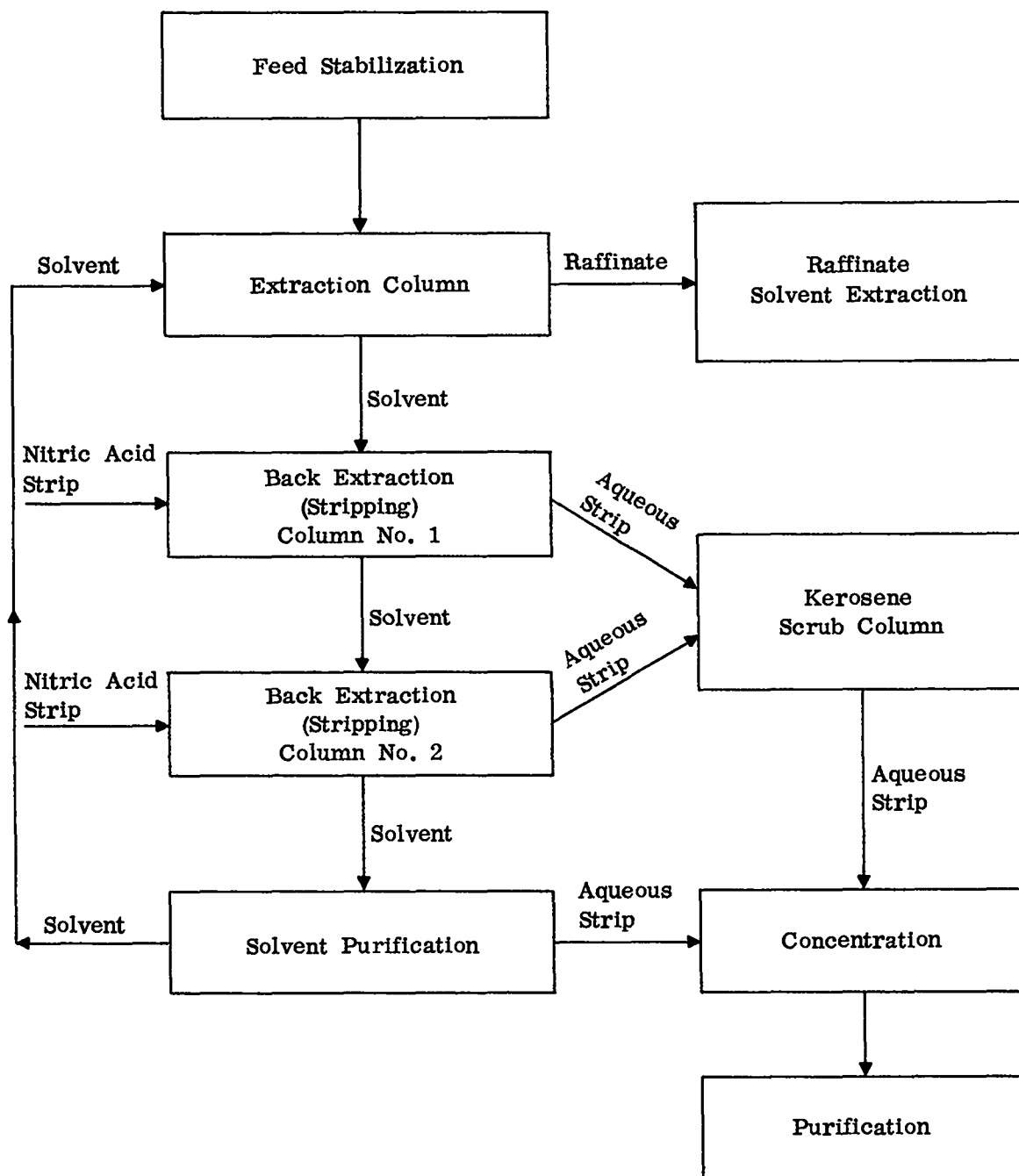


Fig. 5 Flowsheet showing solvent-extraction process.

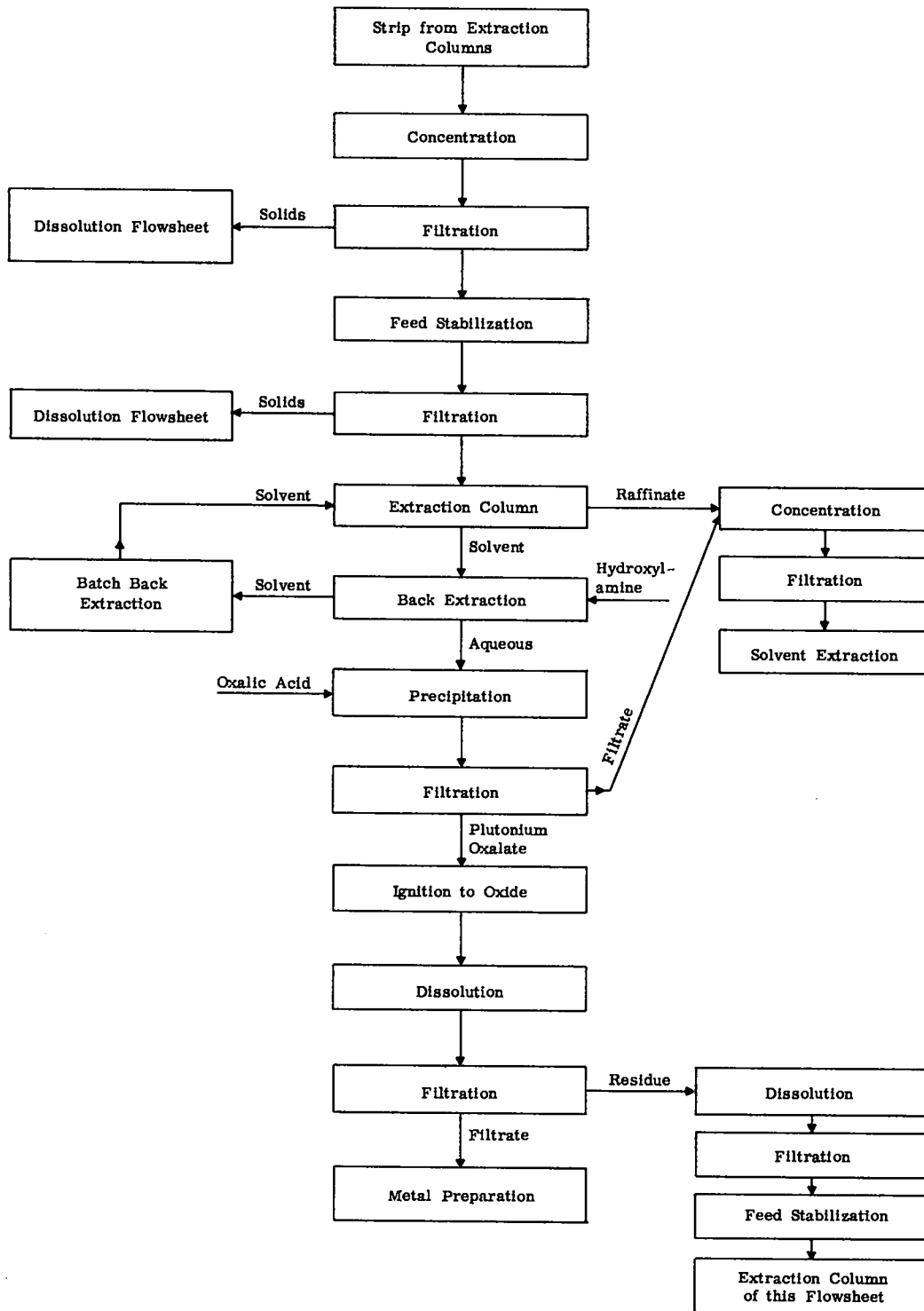


Fig. 6 Flowsheet showing purification process.

TBP (Fig. 7). These are back-extracted into an aqueous strip solution, which is concentrated by evaporation. The concentrate is stored for filtration and future processing back through the columns. Continued recycle of this material ultimately allows the separation of an americium product solution.

The actions of heat, nitric acid, and alpha particles emitted by plutonium on TBP result in its hydrolysis into mono- and dibutyl phosphates, both of which form plutonium-rich compounds. Treatment of the solvent with caustic precipitates these plutonium-containing materials, forming an emulsion. The caustic is acidified, which leaves the hydrolysis products precipitated, and the slurry is recycled through the filtration and column-extraction system. After acidification, the solvent is acceptable for re-use. This process is shown in Fig. 8.

Dilute aqueous streams are concentrated by evaporation for recycle (Fig. 9).

EVENTS LEADING TO THE ACCIDENT

At the time of the accident, a physical inventory was in progress in the TBP-solvent-extraction plant. The normal flow into 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 analysis of the clarified solutions and removed solids.

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 gallons of material was transferred to the stainless steel solvent-treating tank in Room 218 in which the accident occurred (Figs. 10 and 11). This tank already contained about 80 gallons of caustic-stabilized aqueous-organic emulsion that had resulted from the second step in the precipitation of TBP hydrolysis products.

Material transfers leading up to the accident were as follows (refer to Fig. 3 for container locations).

1. Dilute Strip Tank: Fifty gallons of water had been previously added and agitated to attempt to dissolve an estimated 20 lb of solids in the bottom of this tank. This slurry was transferred into the Current Dilute Strip Tank, and the remaining solids were flushed into that tank with a 15-gallon stream of water.

2. Evaporator No. 1: Out of a 110-gallon batch of concentrated dilute strip and peroxide filtrate, 100 gallons was transferred to Filter Feed Tank No. 2 for filtration. (This is the capacity of the tank.) The remaining 10

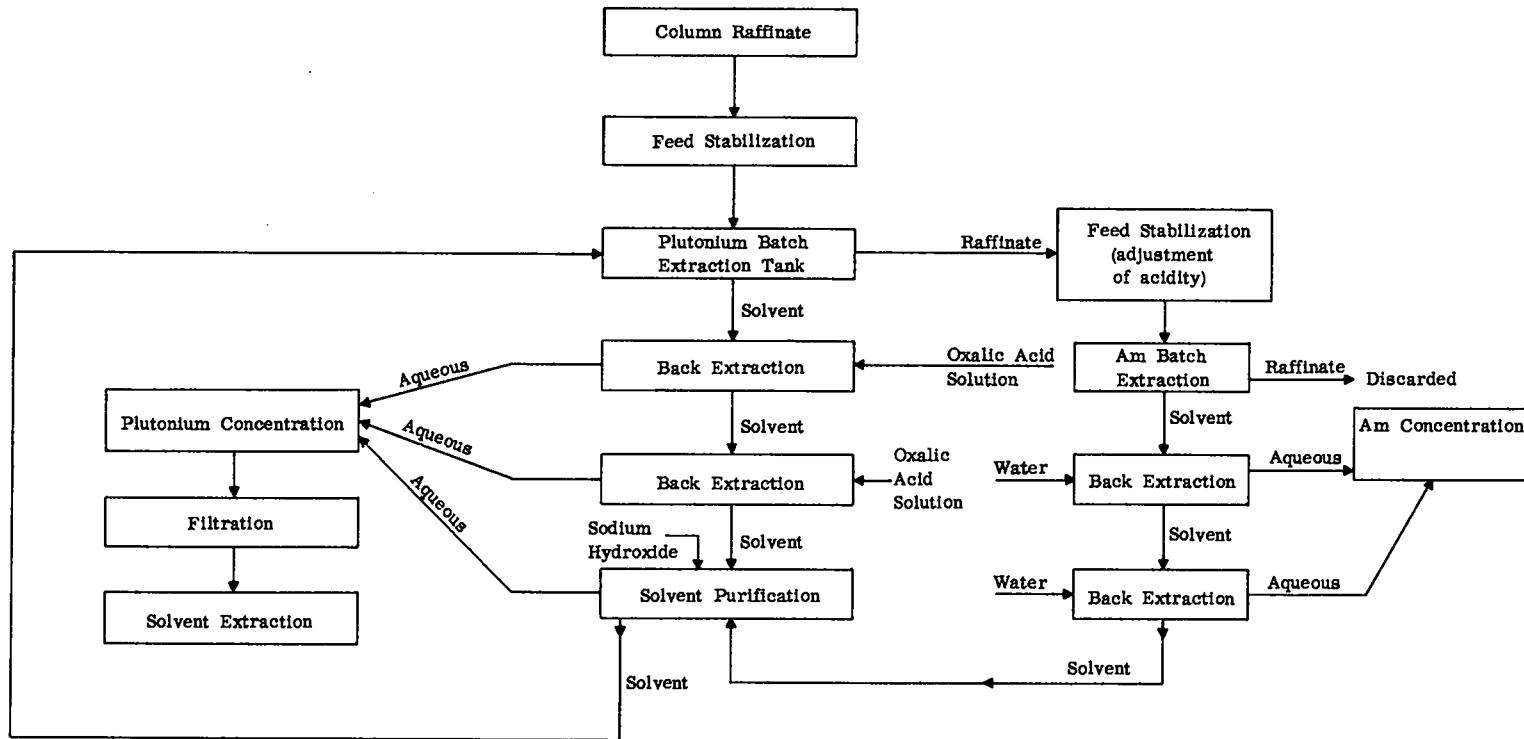


Fig. 7 Flowsheet showing raffinate solvent-extraction process.

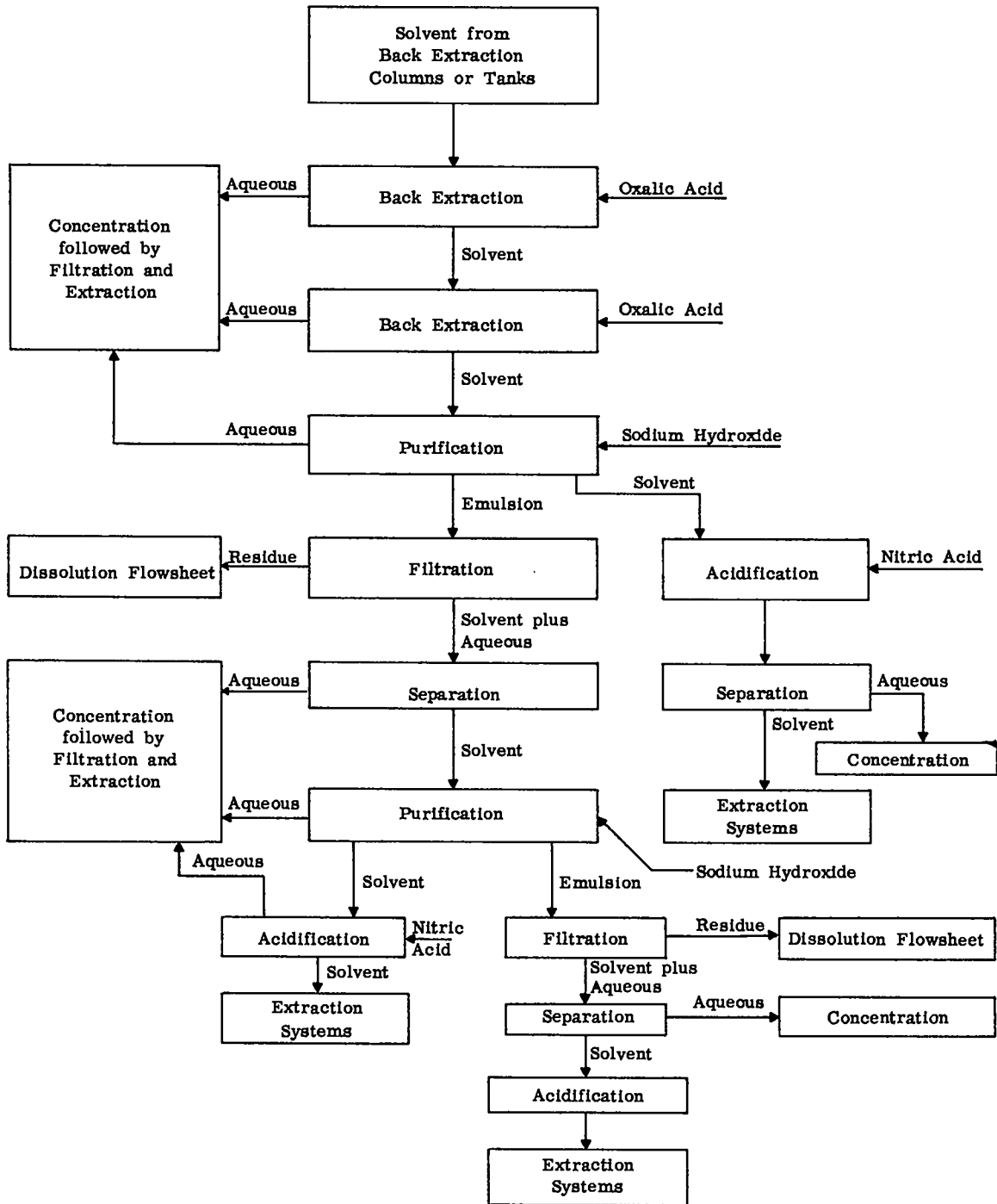


Fig. 8 Flowsheet showing solvent-purification process.

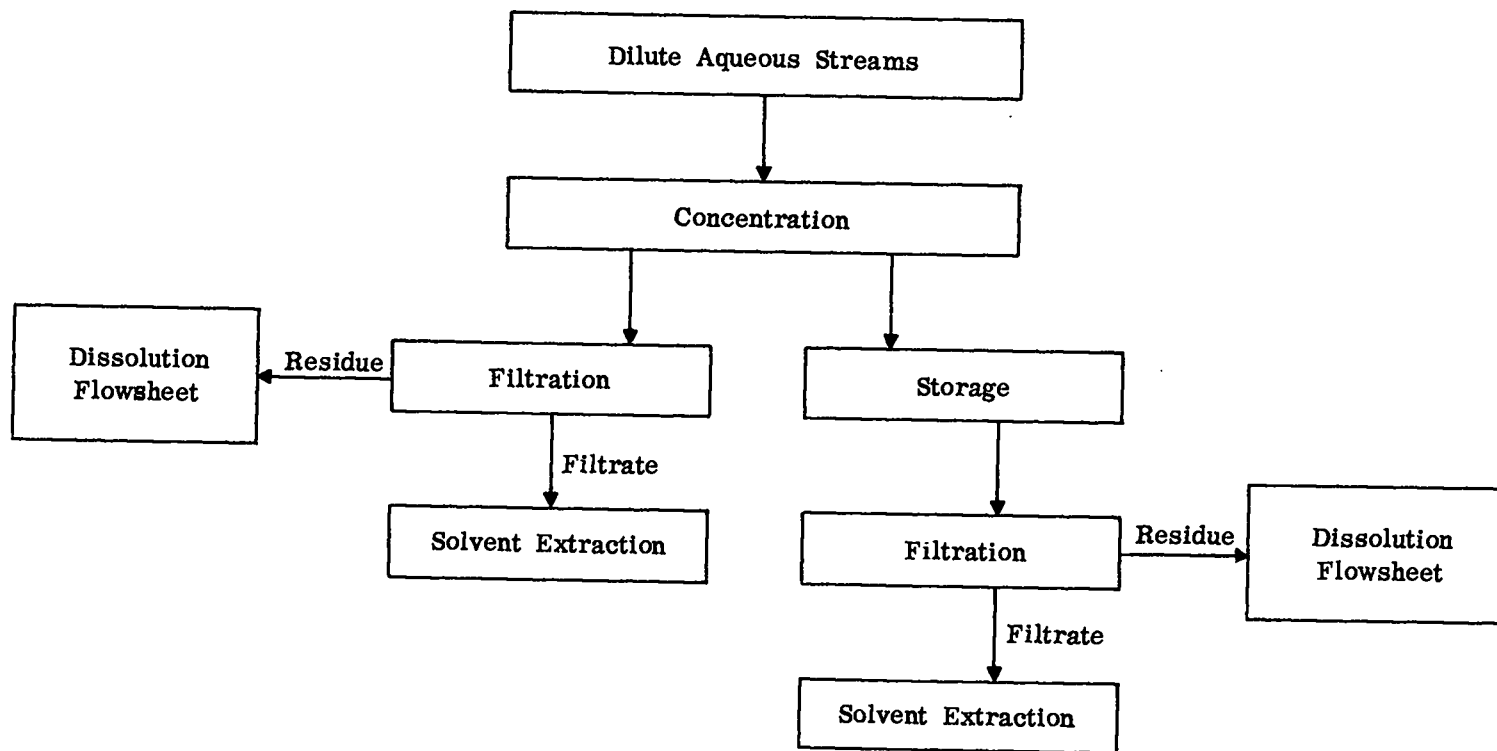


Fig. 9 Flowsheet showing concentration process for aqueous solutions.

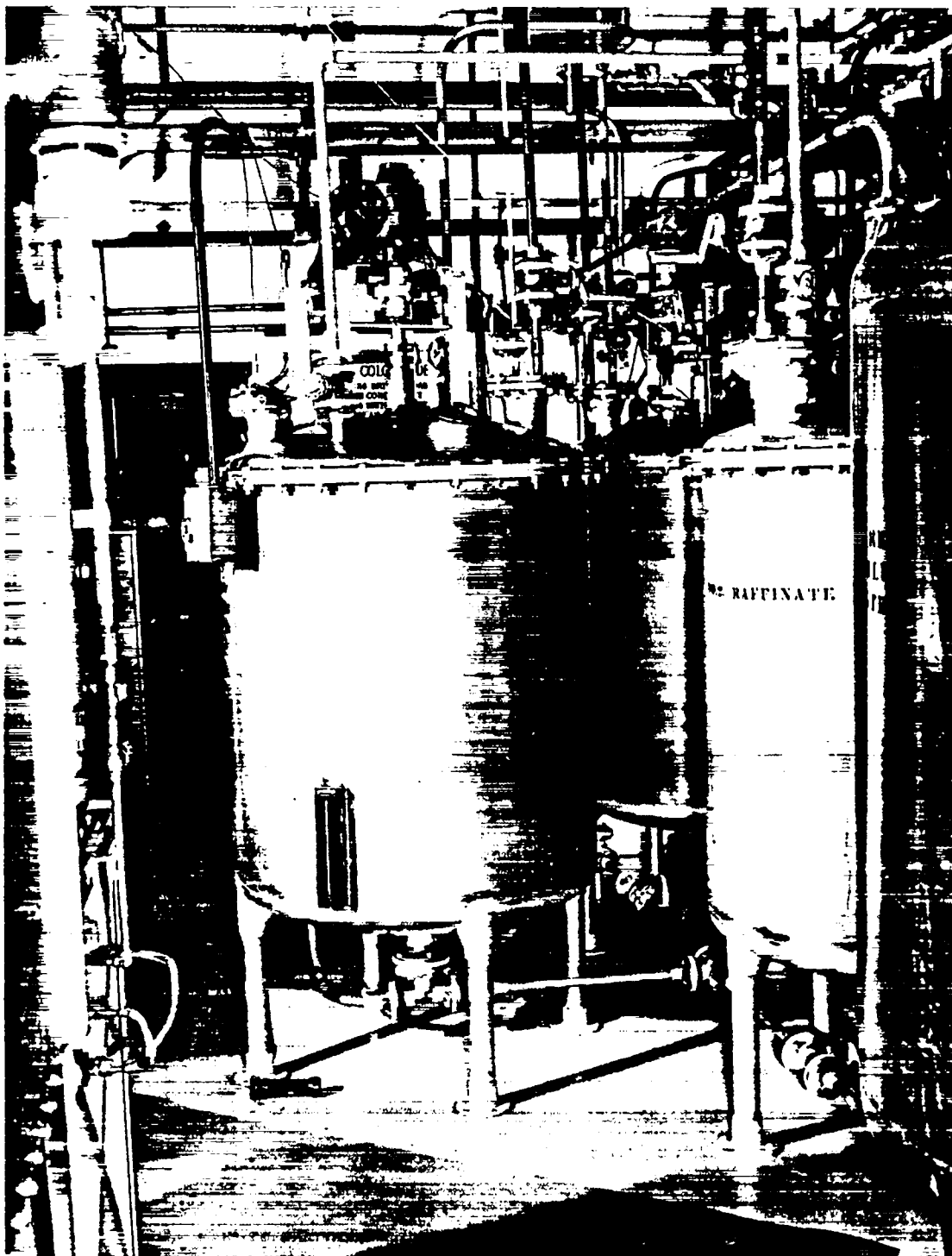


Fig. 10 218 Solvent Treating Tank.

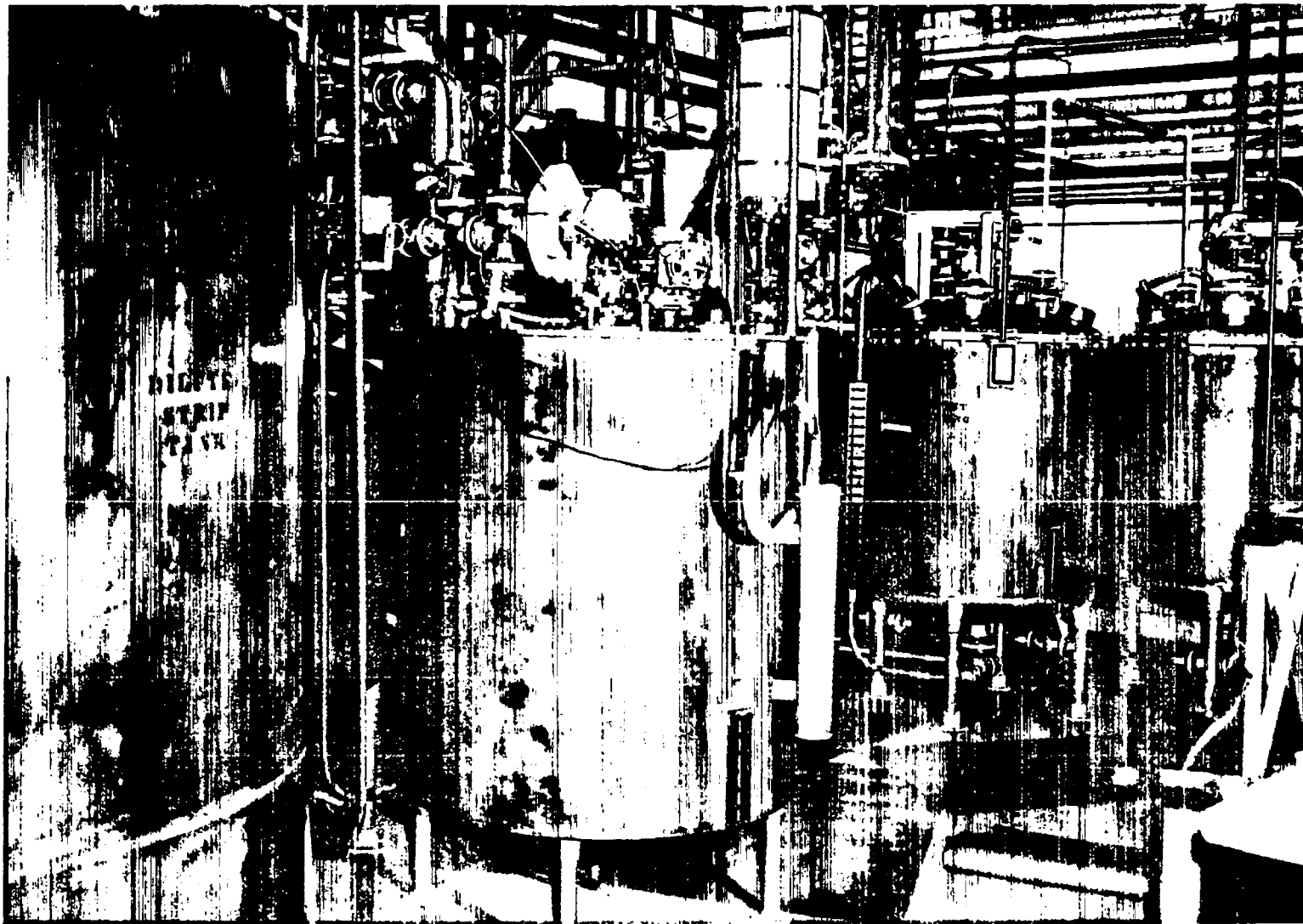


Fig. 11 218 Solvent Treating Tank.

gallons of slurry was transferred to the Current Dilute Strip Tank.

3. Current Dilute Strip Tank: In addition to the above solutions this tank contained an estimated 320 gallons of dilute strip, 100 gallons of acidified caustic from solvent treating, 15 gallons of solvent residual from phase separation of dilute strip, and 2 gallons of a special recovery solution containing 4 g of Pu.

4. 218 Solvent Treating Tank: A 145-gallon batch of treated solvent containing 2.9 g of Pu was transferred from this tank into Raffinate Tank No. 1.

5. Storage Tank No. 2: This tank contained an 80-gallon batch of an emulsion resulting from the second treatment of a batch of solvent. This material was transferred into the 218 Solvent Treating Tank.

6. Current Dilute Strip Tank: From prior experience it is believed that the solutions in and added to this tank contained

<u>Solution</u>	<u>Volume, gal</u>	<u>Plutonium, g</u>
Dilute Strip	320	30
Acidified Caustic	100	40
Solvent	15	1
Special Recovery Solution	2	4
Rinse Water	65	10
Evaporator No. 1 Solution	10	12
Total	512	97

The quantity of plutonium in the TBP hydrolysis product solids transferred from each of the tanks cannot be estimated individually; however, the total added is now known to have been 3.4 kg. A 308-gallon batch of aqueous solution, containing 181.2 g of Pu, was transferred into Precipitation Tank No. 2, and 165 gallons, with 50 g of Pu, was transferred into Evaporator No. 3. The balance of aqueous solution and solvent was transferred into the 218 Solvent Treating Tank. A 13-gallon lot of concentrated nitric acid was used to slurry residual solids from this vessel into the solvent-treating tank.

The addition of solutions and accompanying solids to the Current Dilute Strip Tank is accomplished through a bottom inlet, and each transfer is followed by a vigorous air-sparging of the tank contents. This treatment al-

solvent layer on top of the solution. Subsequent transfer of this plutonium-rich solvent and the nitric acid slurry of residual solids in the bottom of the tank into the 218 Solvent Treating Tank (Figs. 10, 11, and 12) acidified the emulsion, freeing additional solvent for further extraction of any remaining hydrolysis products into the combined organic phase. As in the case of the Current Dilute Strip Tank, mixing and extraction resulted from the air-sparging of this material through the bottom inlet of this tank. Upon cessation of the air-sparging the solvent phase separated into an 8-in. layer on top of the aqueous which was subcritical at this time. This thickness was determined by evaluation of the solutions in the tank after the accident. Phase separation took about 2-1/2 minutes.

The standard operating procedures require that slurries and solids residual in the various processing tanks be separately transferred from each tank to the Filter Feed Tank and into the filtration area, where the solids are removed, sampled for analysis, and processed in geometrically-favorable equipment. When solids cannot be transferred from a tank in the form of a slurry, the tank is opened and the solids are mechanically removed. The mixing of slurries with TBP is to be avoided, not only for purposes of criticality control, but also from the standpoint of acceptable processing procedure. The contacting of slurries with TBP generally results in a very stable emulsion caused by the silicates present.

Although filtration procedures had been performed on the solutions and slurries resulting from the first treatment of the material transferred into the 218 Solvent Treating Tank from Storage Tank No. 2, they were not carried out on the slurries transferred from Evaporator No. 1 and the Dilute Strip Tank into the Current Dilute Strip Tank, and from that vessel into the solvent-treating tank. It is concluded that the major source of plutonium was the solids from these tanks.

THE ACCIDENT

The situation in the 225-gallon, 38-in.-diameter, solvent-treating tank immediately before the accident is believed to be as illustrated in Fig. 12: 87.4 gallons of aqueous solution contained 40 g of Pu, and on top, a 42.2-gallon layer of solvent contained 3.27 kg of Pu. Solids (containing 60 g of Pu) 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. 13, 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 g/liter (the estimated critical thickness is 8-1/4 inches, and the actual configuration was roughly 5 dollars subcritical).

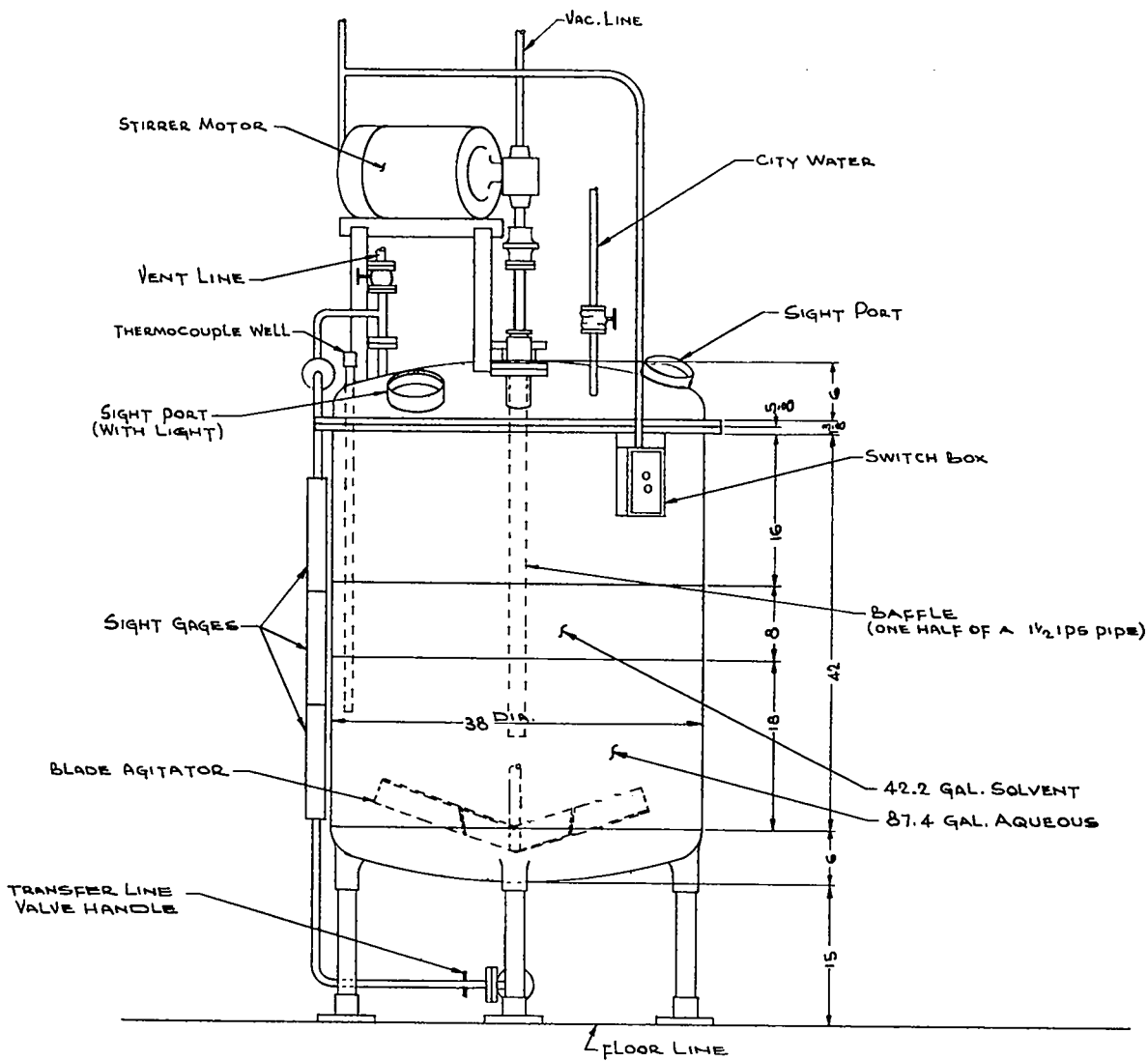


Fig. 12 Diagram of solvent-treating tank, showing levels of aqueous and organic phases before stirrer was activated.

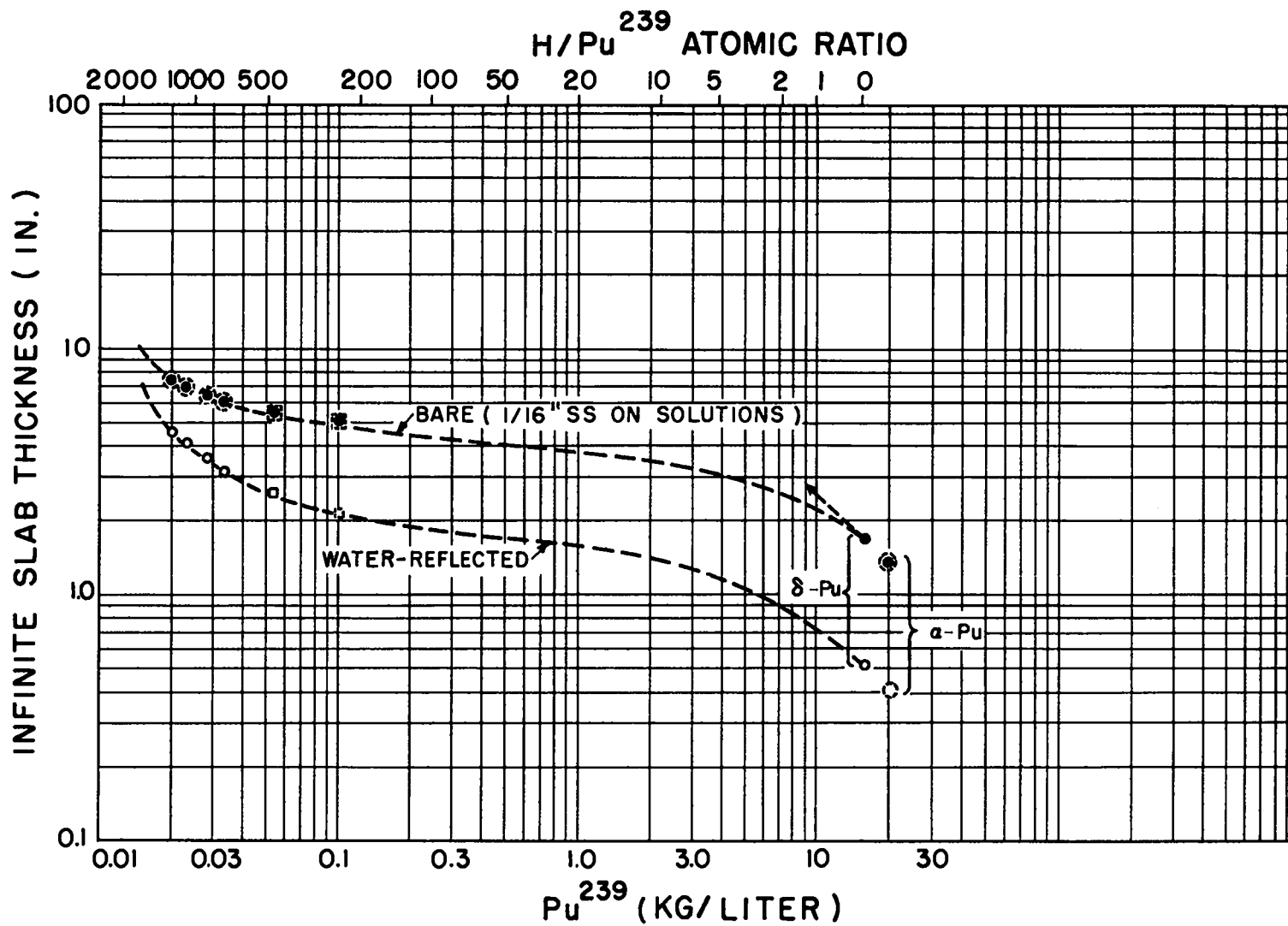


Fig. 13 Estimated critical thicknesses of infinite slabs of homogeneous water-moderated Pu^{239} in the absence of NO_3 and Pu^{240} . Based on data from F. E. Kruesi, J. O. Erkman, and D. D. Lanning, Hanford Report HW-24514, May 1952 (classified).

A chemical operator started the motor driving the mechanical agitator 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 (Figs. 10 and 12), was knocked off the two-step ladder on which he stood. Though the shock displaced the tank about 3/8 inch at its supports, the tank was not ruptured and no plutonium escaped. The operator apparently turned the stirrer motor off, then on again (this time noticing a rumbling sound), ran out a nearby exterior door, and called that he was "burning up." A second operator, some 40 feet away in an adjoining room, saw a reflection of the light on the walls ("like a photo-flash"), heard the report, went to help, and was joined by a third operator. Returning to lead the first man to a shower (on request, apparently as the result of suspecting an acid burn), they passed the tank, and the second operator turned off the stirrer motor.

The burst activated a radiation alarm located 175 feet away in a nearby building. This unit, connected to an indicating instrument, was set to sound at 10 mr/hr. The entire plant was evacuated.

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 about 0.4 inch, corresponding to an average radius decrease of 1/2 inch, 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.

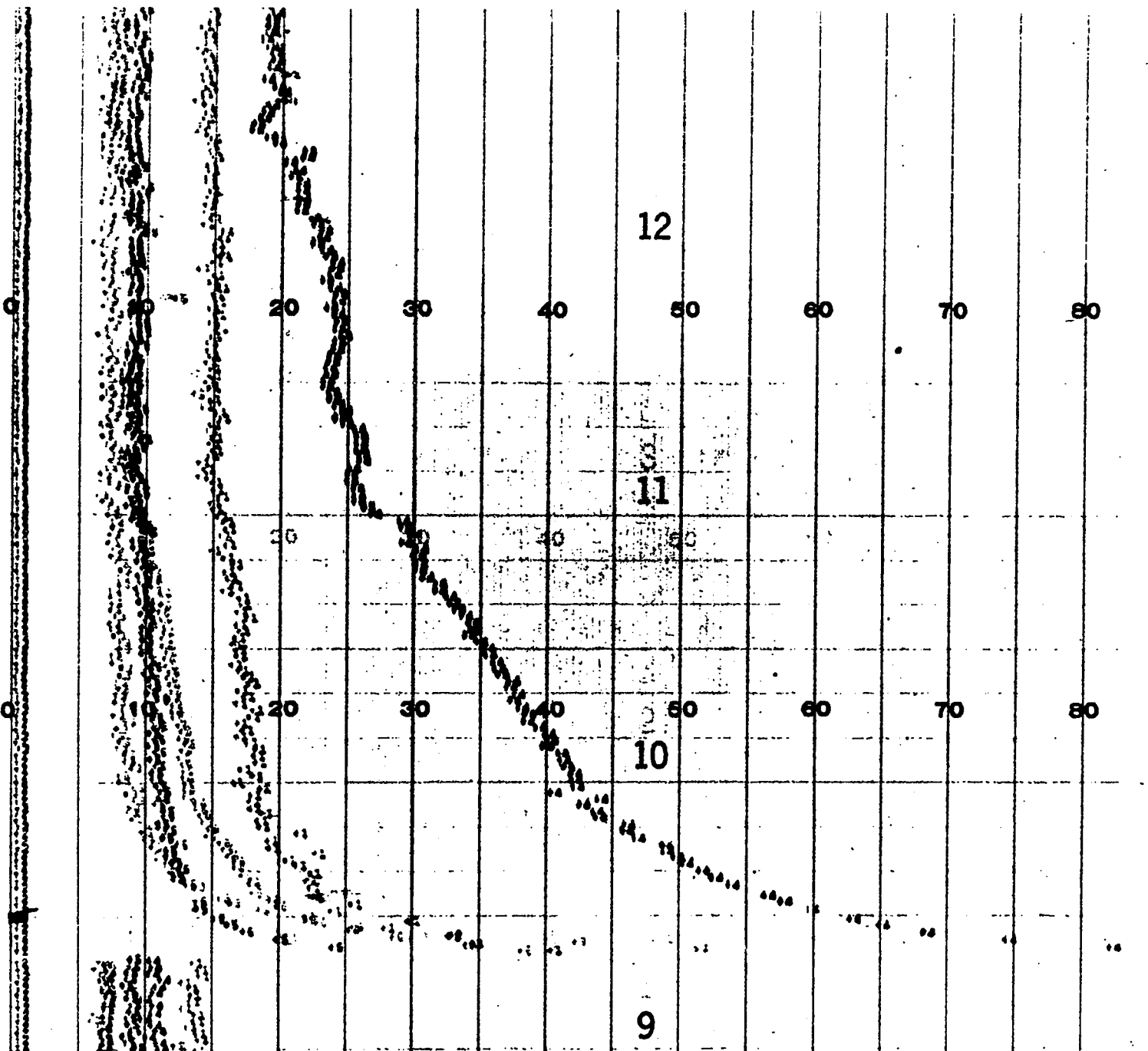
None of the gamma-sensitive recording detectors within range of the accident showed an ideal trace. They suggest, however, that there was but a single burst -- see, for example, the record reproduced in Fig. 14.

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.

EXPOSURES

Results of radiation monitoring in the vicinity of the solvent-treating tank after the accident are summarized in Table 1, and Table 2 lists radiation dosages received by personnel at the plant.

The chemical operator at the solvent-treating tank went into deep shock



Time interval on horizontal scale is 10 min

Fig. 14 Trace of slow-acting recorder (Point No. 4) located 160 ft from tank.

TABLE 1

RESULTS OF RADIATION MONITORING OF ROOM 218

<u>Date</u>	<u>Time</u>	<u>r/hr</u>	<u>Direct Sight Distance from Tank, ft</u>
12/30/58	4:40 p.m.	20	25
	5:10 p.m.	45	7.5
	5:15 p.m.	0.2	110
	5:35 p.m.	>50	4.5
	7:30 p.m.	12	1.5
	9:45 p.m.	0.1	25
	12/31/58	8:20 a.m.	0.05
1:40 p.m.		10	Contact
3:55 p.m.		-	7.5
10:01 p.m.		5	Contact
1/1/59	9:00 a.m.	2	Contact

TABLE 2

DOSAGE ESTIMATE FOR OPERATING PERSONNEL

<u>Location of Chemical Operators*</u>	<u>Dosage, rem</u>
Against 218 Solvent Treating Tank	12,000 \pm 50%
Behind Feed Tank No. 3 (43 ft)	134
In purification area (35 ft)	53
In other areas (>75 ft from supercritical burst)	<4

*See Fig. 3.

within 15 minutes of exposure. He regained consciousness about 6 hours later and remained rational and comfortable until nearly the time of his death, 3:15 a.m. on 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 dosage indicates, of course, that there was no succeeding burst while they were near the tank.

SOLUTION REMOVAL AND ANALYSIS

Immediately after the incident, a bank of ten 6- and 5-in.-diameter cylinders was fabricated and set up about 100 feet from the point of the accident (see Figs. 3, 15, and 16). 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 was therefore accomplished from behind a temporary shield by means of a 10-foot extension handle. The solution was vacuum-transferred into the bank of cylinders without complication on January 1, 1959. 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.

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 3. Not only was the bulk of the plutonium found to be in the organic solvent phase, but a concentration of Zr^{95} 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 was concentrated in the solvent layer when the burst occurred is given by the long-lived gamma activity of the stainless steel baffle (type-347) that was located about 6 inches within the wall of the solvent-treating tank (Fig. 12). Scanning indicated a peak in activity about 1 inch below the mid-plane of the solvent layer.

NUCLEAR-SAFETY CONTROL

A nuclear-criticality review of the entire plutonium plant had been completed about a month before the accident. Changes in equipment

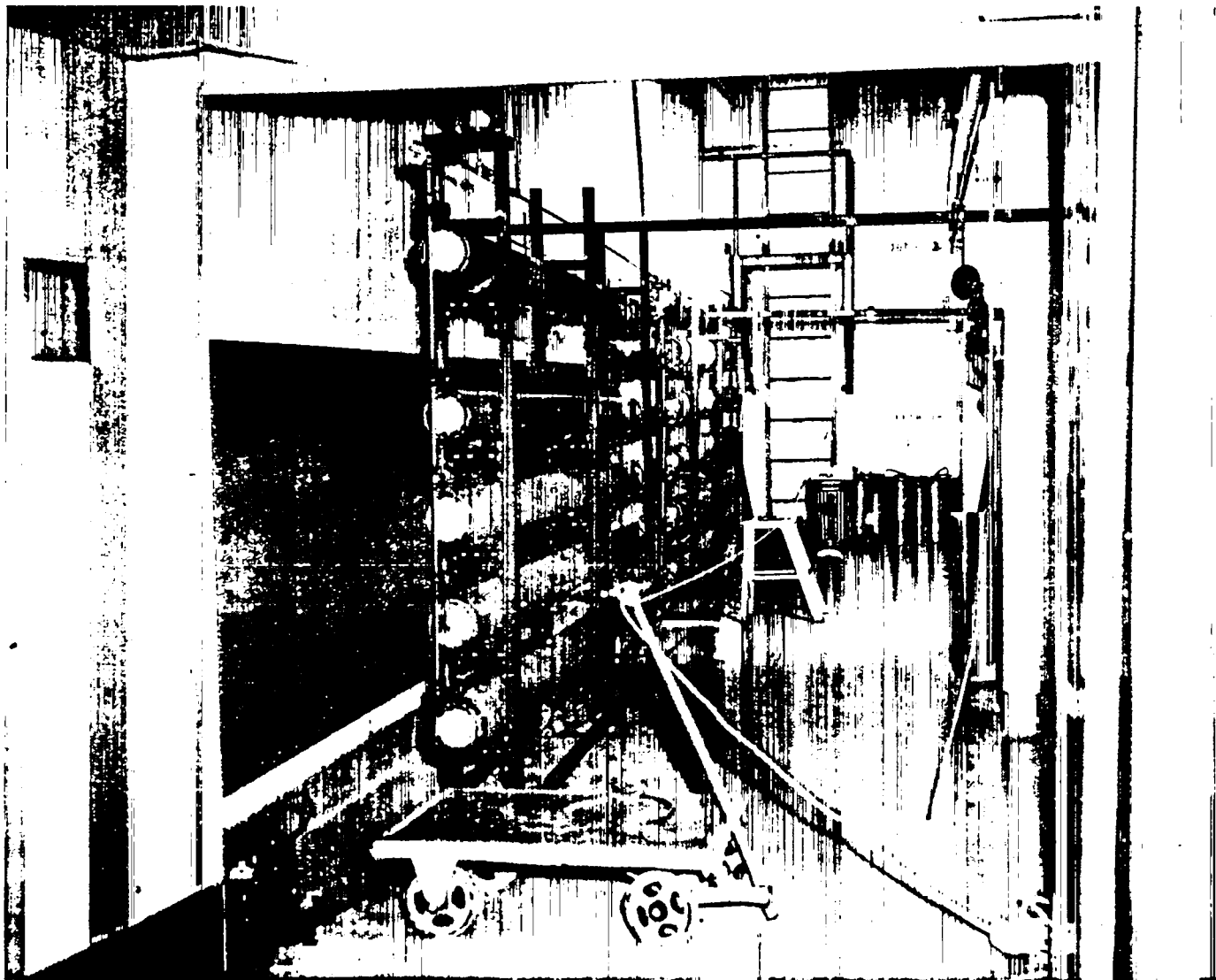


Fig. 15 Geometrically-favorable tanks used to receive the solution.

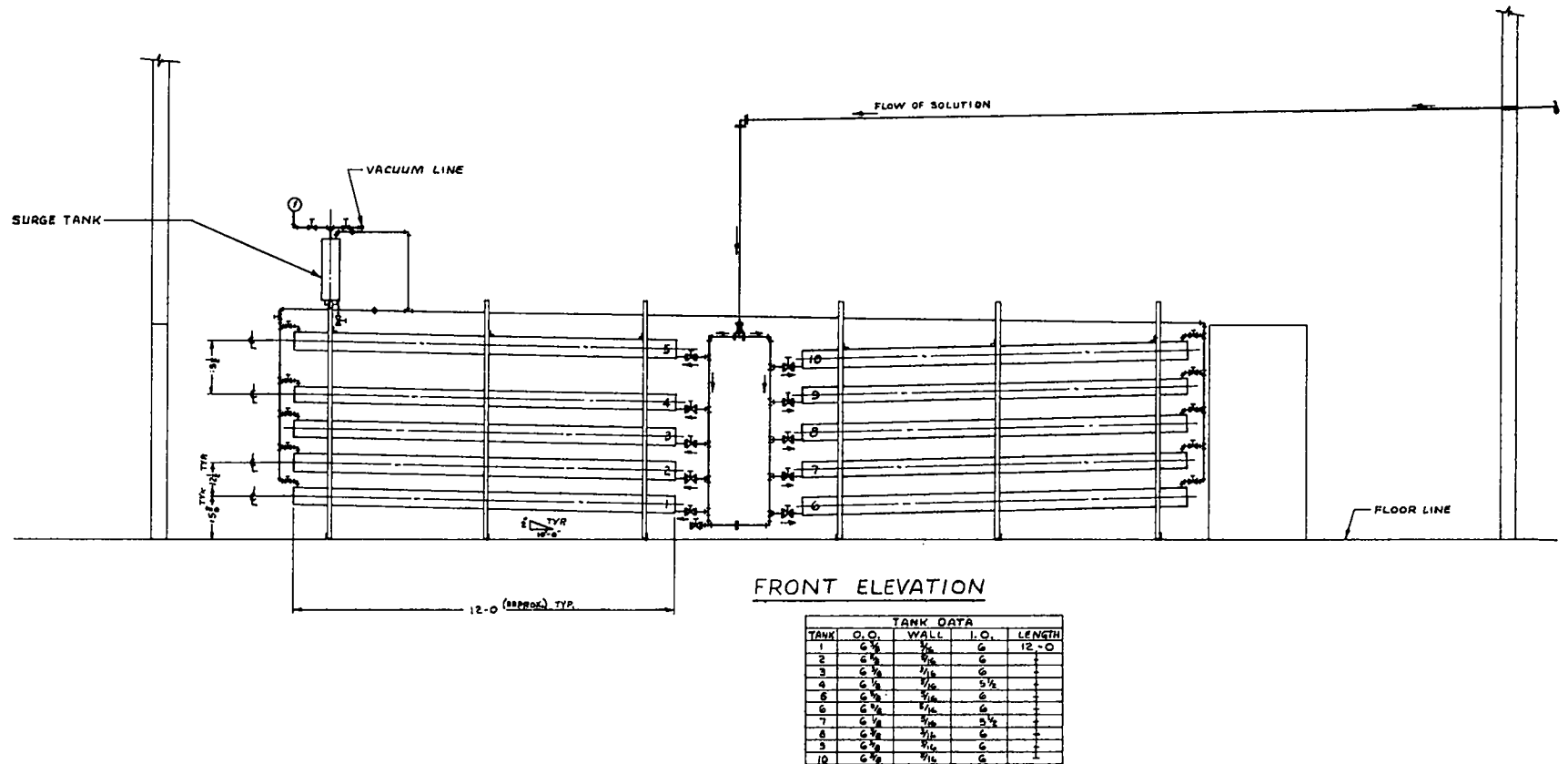


Fig. 16 Schematic diagram of geometrically-favorable storage cylinders.

TABLE 3

EVALUATION OF Pu IN THE GEOMETRICALLY-FAVORABLE TANKS
FROM THE 218 SOLVENT TREATING TANK

Tank No.	Aqueous		Organic		Solids
	Volume, l	Pu, g	Volume, l	Pu, g	Pu, g
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
Total	330.9	37.2	160.0	3272	47.5

Avg. conc., g/l 0.112 20.4

Estimated solids not removed from Tank Nos. 3 and 8: $(2,000 \text{ g})(0.01) \approx$
20 g of Pu

<u>Material</u>	<u>Volume, l</u>	<u>Pu, kg</u>
Organic	160	3.27
Aqueous	331	0.04
Solids		0.06
Total	<u>491</u>	<u>3.37</u>

associated with a hopefully-improved solvent-extraction process were planned to reduce dependence upon procedural control of batch sizes for nuclear-safety purposes, although this type of control had been used successfully during the 7-1/2 years that the plant was in operation. Installation of the new equipment was scheduled for June and July 1959. Depending upon the efficiency of the new extraction process, the possibility of eliminating the succeeding raffinate solvent-extraction and concentration stages (Fig. 2) 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 as a result of the review 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, of course, there has been further detailed review of the entire plutonium plant. As a result, additional safety precautions have been adopted to prevent recurrence of an accident, and work on those already planned has been accelerated. Following is an outline of the steps being undertaken as a result of the review prior to the accident and the investigation following it:

1. Geometrically-favorable dissolvers and feed tanks for filtration and solvent extraction have been designed, and it is expected that the equipment will be installed before operations are resumed.

2. Supplemental transfer lines, such as those required for emergency procedures, will be blocked to minimize the opportunity for abnormal interchanges.

3. Cadmium nitrate solution has been placed in vent tanks and vacuum-buffer tanks 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, although unequal distribution between phases in the one case, and plutonium deposition on surfaces in the other, present difficulties.

4. Neutron detectors are being calibrated and tested for indicating abnormal deposits of plutonium.

5. Measurements to give nuclear-safety data for hydrogen to plutonium ratios in the region of 10 to 1 are being planned. A computational program

is needed to interpolate between experimental points.

6. Gamma-sensing alarms are to be located in all areas where a criticality-accident potential exists. Equipment was temporarily installed at the site where the accident occurred, and a permanent installation has been designed which will be in operation by July 1, 1959.

7. Coupled with alarm devices, emergency procedures have been established and employees trained to follow them. As part of the training program, practice actions for emergency situations will be conducted at reasonable intervals. Physical changes in buildings, processes, gates, and fences will require a review and re-evaluation of evacuation procedures.

8. Although nuclear criticality is admittedly a complex and sophisticated subject, it is recognized that certain general safety fundamentals exist which can be taught to employees at the technician and operator level, e.g., the nature and consequences of criticality accidents. That such training is not scare tactics has been repeatedly demonstrated in the explosives industry, where violent demonstrations have been used effectively. Where an experimental criticality safety facility exists, it might well be used to show employees graphically the development of ionizing radiation from critical assemblies. It is planned to use the Pajarito laboratory of LASL group N-2 for this purpose.

APPENDIX

PLUTONIUM CRITICALITY DATA

Criticality data for homogeneous plutonium-water systems are summarized in Figs. A.1 and A.2.* Critical masses and critical volumes are given for bare spheres and water-reflected spheres. Deduced from these values are the corresponding critical radii of infinite cylinders and critical thicknesses of infinite slabs shown in Figs. A.3 and 13 of the text. For interconversions between critical spheres (radius r_s) and cylinders (height h_c , radius r_c), the following relation, which has been checked in detail for U^{235} solutions, is consistent with the limited plutonium solution data:

$$\left(\frac{\pi}{r_s + \delta_s} \right)^2 = \left(\frac{2.405}{r_c + \delta_c} \right)^2 + \left(\frac{\pi}{h_c + 2\delta_c} \right)^2,$$

where δ_s and δ_c are given in Fig. A.4.

Figure A.5 is a guide for the use of cadmium in aqueous plutonium solutions as a secondary criticality safeguard. Any other strong thermal poison may be substituted by adjusting its macroscopic cross section to equal that indicated for cadmium. The curve of Fig. A.5 was computed from the relation

$$k_\infty = \frac{\nu \left(\Sigma_f^{\text{th}} + \Sigma_f^{\text{res}} \frac{\Sigma_a^{\text{th}}}{\Sigma_s} \right)}{\Sigma_a^{\text{th}} + \Sigma_a^{\text{res}} \frac{\Sigma_a^{\text{th}}}{\Sigma_s}} = 1,$$

where Σ_f^{th} and Σ_a^{th} are thermal macroscopic cross sections for fission and total absorption, Σ_f^{res} and Σ_a^{res} are the corresponding resonance integrals, and Σ_s is the macroscopic scattering cross section of hydrogen.

*Solution data are from F. E. Kruesi, J. O. Erkman, and D. D. Lanning, Hanford Report HW-24514, May 1952 (classified).

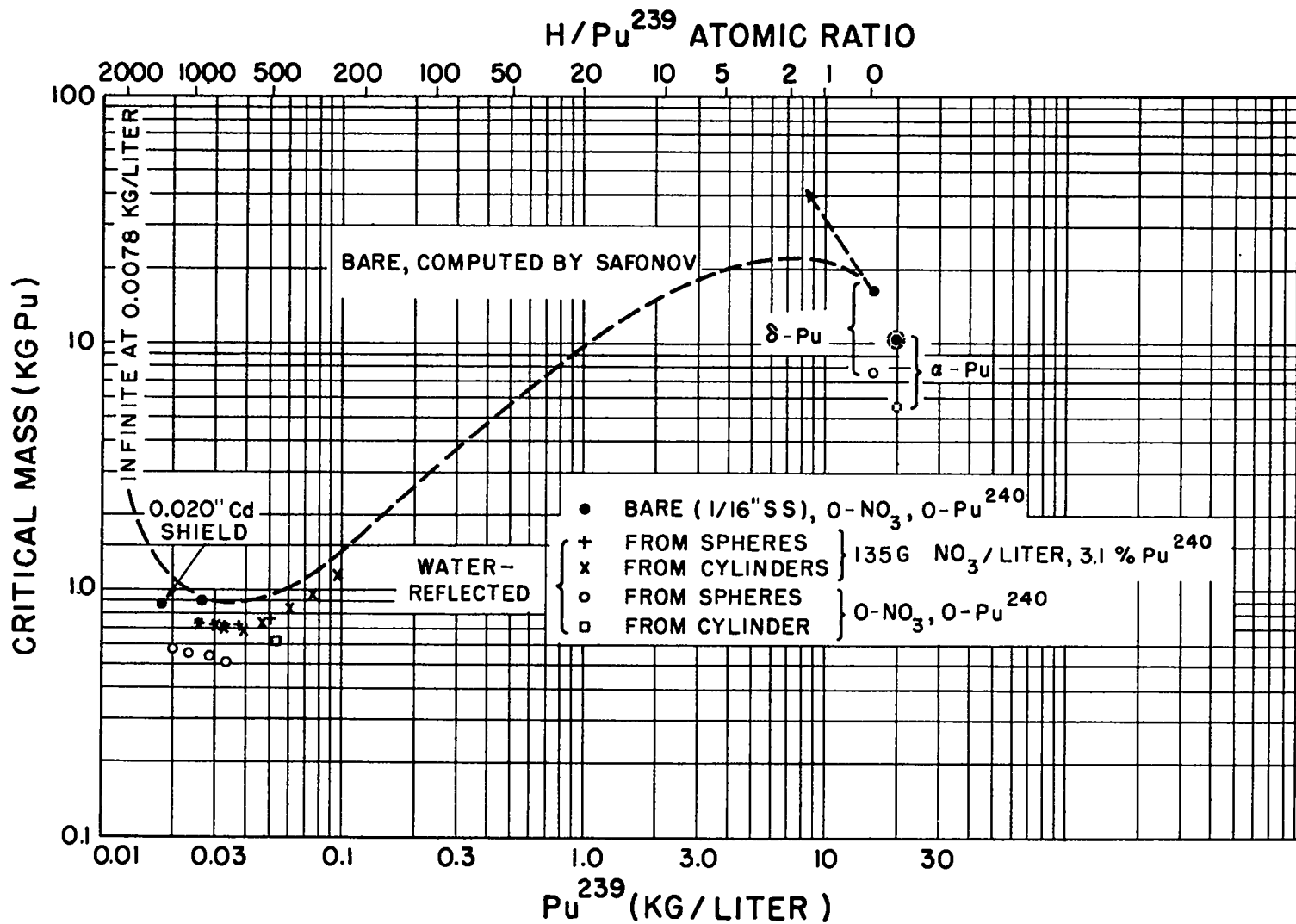


Fig. A.1 Critical masses of homogeneous water-moderated Pu^{239} spheres.

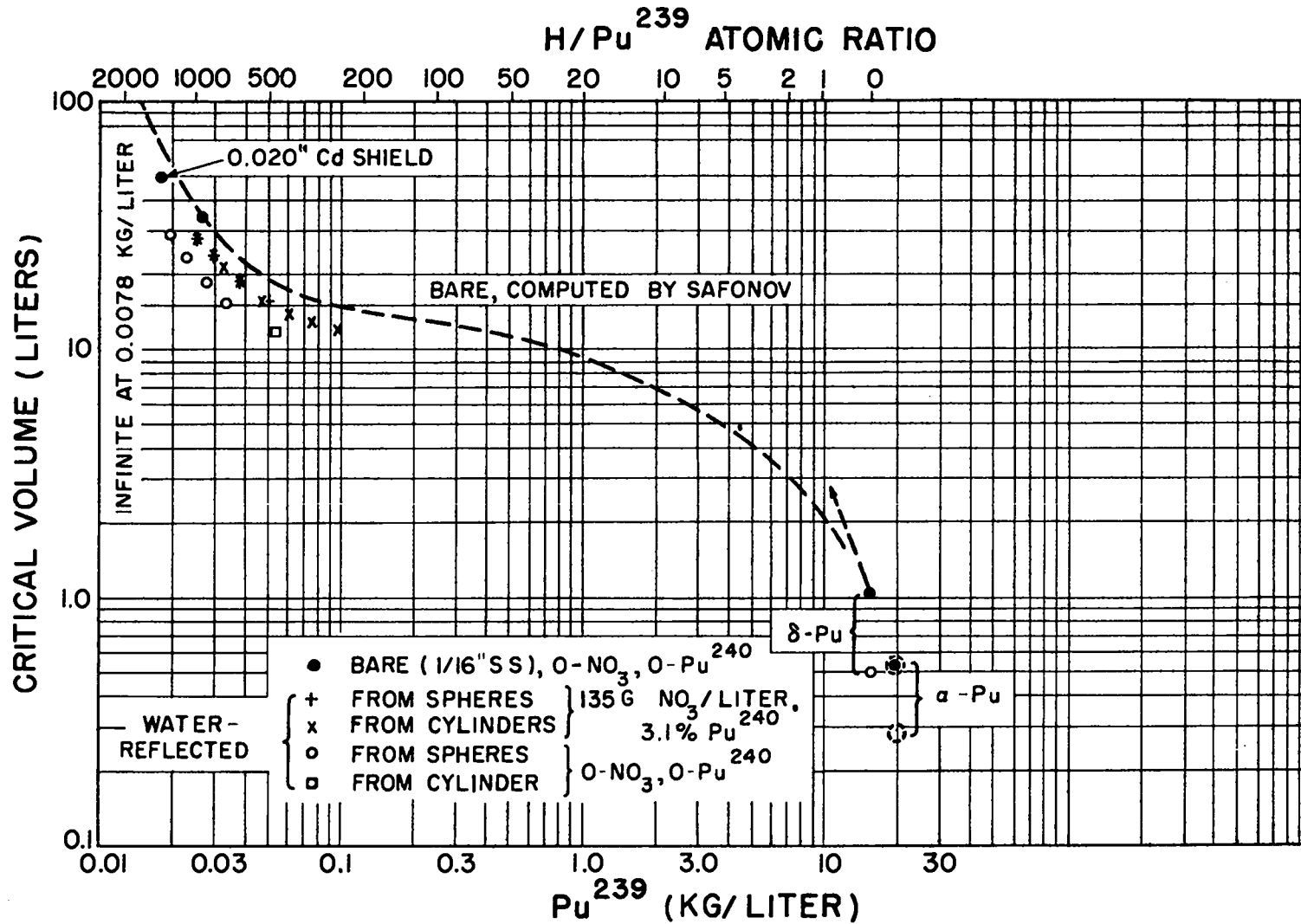


Fig. A.2 Critical volumes of homogeneous water-moderated Pu^{239} spheres.

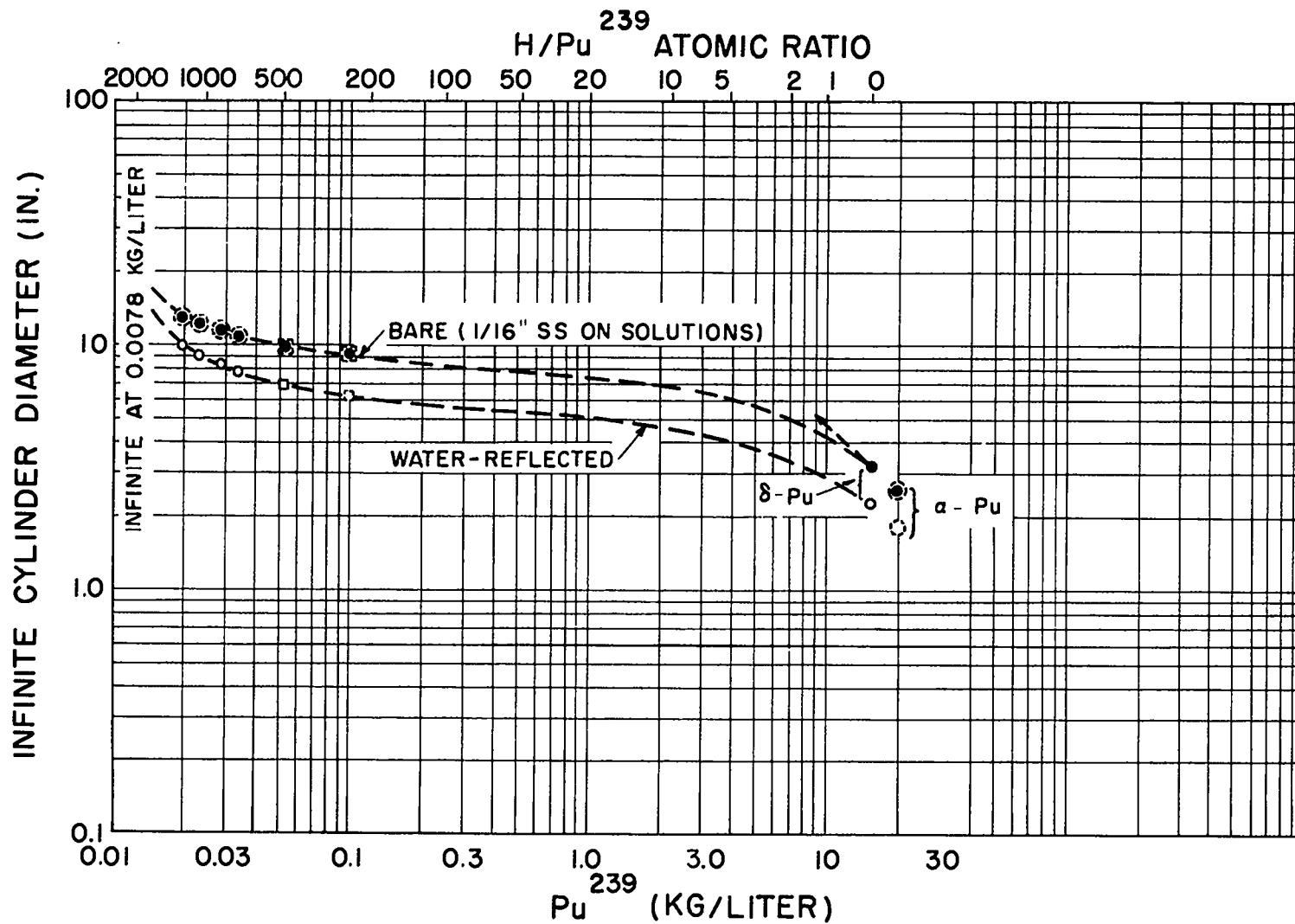


Fig. A.3 Estimated critical diameters of infinite cylinders of water-moderated Pu^{239} in the absence of NO_3 and Pu^{240} .

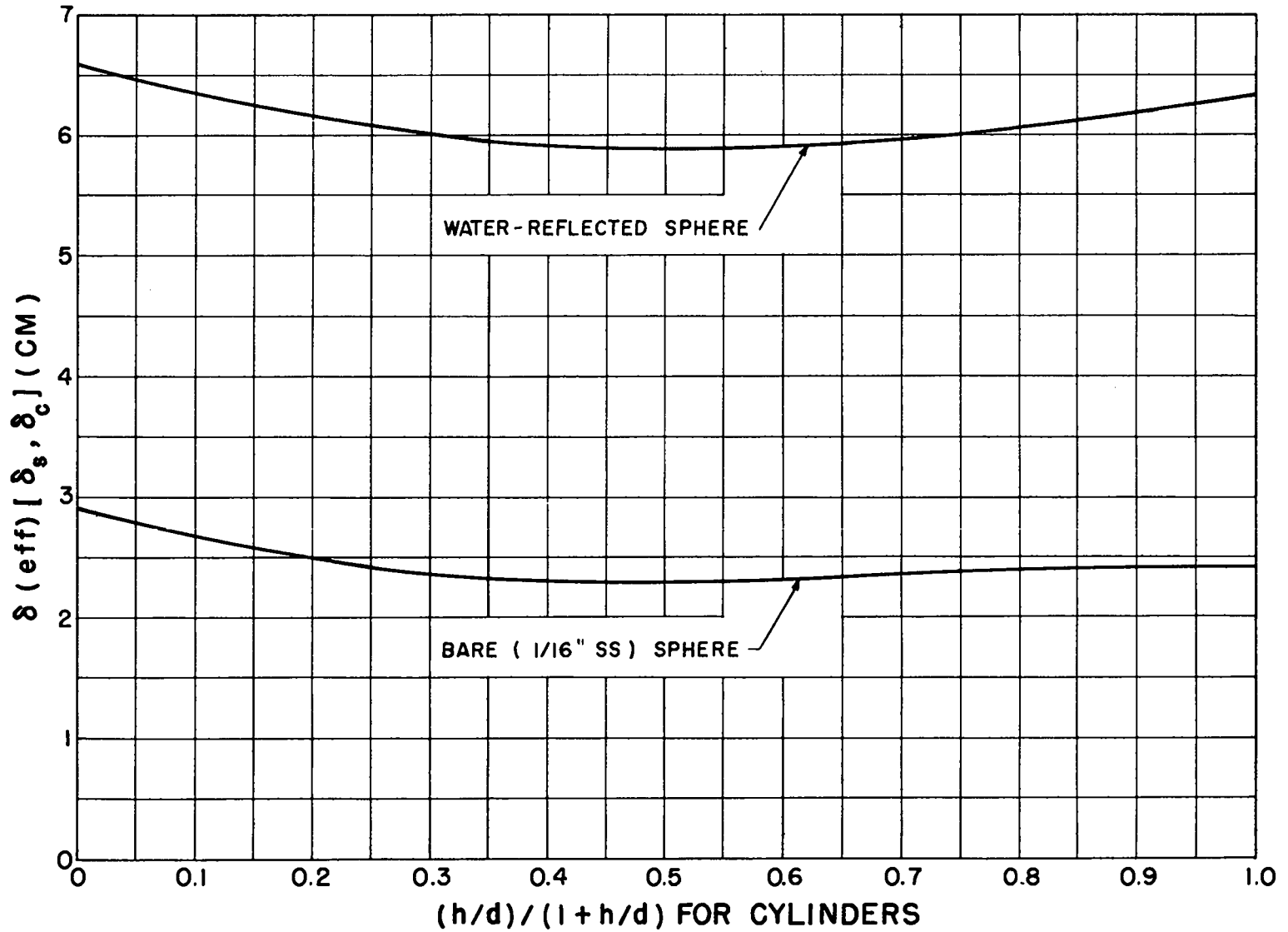


Fig. A.4 Effective extrapolation lengths for sphere-cylinder conversion of critical solutions.

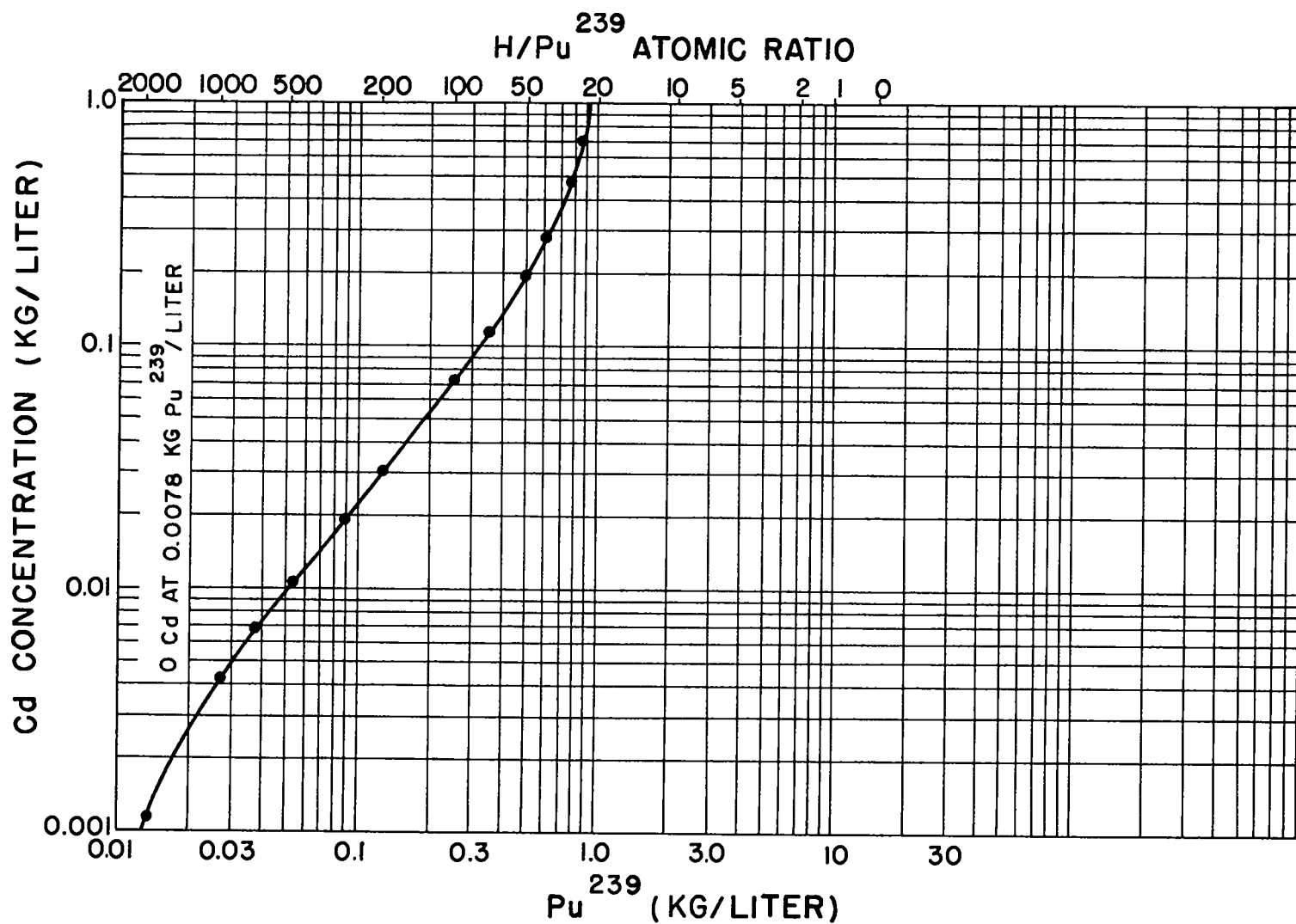


Fig. A.5 Uniform Cd concentrations required to "safe" Pu solutions regardless of volume.