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TITLE: HISTORICAL PERSPECTIVE OF CRITICALITY SAFETY IN THE UNITED STATES

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HISTORICAL PERSPECTIVE OF CRITICALITY SAFETY IN THE UNITED STATES

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Criticality safety in this country is 35 years old, so should be approaching maturity. My purpose is to review the stages through which this discipline has progressed, and to speculate about present signs of maturity.

Early History

Criticality safety was conceived with the Oak Ridge Gaseous Diffusion Plant and the Hanford Chemical Processing Plant that supplemented the early Plutonium Production Reactors. But the birth of this discipline had to await design and construction of these plants. activities that proceeded at a pace that now seems unimaginable.

Let me remind you that Hanford construction started in June 1943 and three production reactors and the Chemical Processing Plant were operating successfully summer of 1945.[1] (Crawford Greenewalt of duPont Company, the Hanford contractor, is quoted as saying that construction expenditures there, $750 million in two years, had not been equalled before or since.[2]) The monstrous diffusion plant (Fig. 1) came through on a nearly identical schedule, also with design completed just 4-1/2 years after the discovery of fission was announced.

By necessity, both plants had to proceed without reliable criticality information. This lack was not crucial because the processing plant was designed generously and gaseous UF_6 of the diffusion plant was too ethereal for criticality. But increased demands on the processing plant could be foreseen, and the diffusion plant's UF_6 could condense if heat were lost or could break down and deposit if there were inleakage of moist air. The resulting desire for criticality guidance was incompletely satisfied by calculation, and led to critical experiments as soon as enriched uranium and plutonium became available.

While being shown through the Clinton Laboratory on my last day at Oak Ridge (in August 1945), I witnessed a demonstration that was impressive but meant little more to me at the time. It must have been nearly the first critical assembly, being "played" by a maestro from Los Alamos --the rate of counter clicks increasing and diminishing as his hands approached and withdrew from a stack of cubes. Much later, I learned that this was an assembly of U(24)$_3O_8$-fluorocarbon* blocks interspersed with polyethylene to simulate a deposit in the Diffusion Plant (Fig.2).[3]

This study was expanded early in spring 1946 when Dixon Callihan, Clifford Beck, Raymond Murray and several others from Oak Ridge joined Louis Slotin for experiments at Pajarito Site in Los Alamos.[4] The fissile composition was a mixture of U(95)F_4 and polytetrafluoroethylene to simulate UF_6C. Critical assemblies were constructed of one-inch-cubic compacts of this material latticed with polyethylene (Fig. 3). (There was insufficient ^{235}U, nearly 50 kg, to attain criticality

*U(24) means uranium containing 24 wt% ^{235}U.

without hydrogen moderation.) Objectives achieved were stated as follows.

"(a) Considerable information on critical masses under conditions of interest to K-25 was established."
"(b) Experience was gained by the Oak Ridge group, which could then continue into further investigations as necessary for the safety of the plants at Oak Ridge."

The further Oak Ridge investigations began later in 1946 with a series similar to that at Los Alamos, but with U(30).[5] Fig. 4. This was followed immediately by the first of many critical assemblies of enriched-uranium solutions.[6] Critical plutonium solutions were studied first as crude assemblies at Los Alamos (1945),[7] then in a temporary facility at Hanford (beginning 1950) as a comprehensive series of clean assemblies.[8] Thus, experimental data for checking criticality calculations began to accumulate shortly after the beginning of plant operations. This interaction between experiment and theory fell outside the usual province of plant designers and operators, so led to the criticality safety specialist who remains entrenched to this day.

Evolution

Originally, batch processes with administrative control of batch sizes were characteristic of plants for purification and reduction of enriched uranium and plutonium. For example, hand operations in glove boxes or hoods were typical of metal production (Fig. 5). But as criticality data accumulated and the specialists contributed to process design, it became practical to reduce reliance upon administrative controls. The means, of course, was geometrically favorable equipment, and, in some cases, nonaqueous processing (Figs. 6 and 7). The transition to more positive criticality control, however, often lagged far behind the available technology while awaiting funds for rebuilding. Because of their secondary importance, scrap-recovery plants often found themselves on a back burner.

Accident Experience

A rash of process criticality accidents, six from 1958 to 1964, all occurred in scrap-recovery plants (Table I).[9,10] Why? Maybe guards were down because of a lucky decade. Out-of-date equipment was involved in a couple of instances. Inventory increases may have had some influence. And recovery plants were most vulnerable because their aqueous processes required flexibility for treating a variety of materials.

As a result of these accidents, the industry was shocked into more effective criticality control, and there was increased appreciation for the importance of safety specialists. The consequent elevation of criticality safety as a discipline appears to be maintained.

Maturity

I suggest that the best gauge of maturity is the status of standards in the field. Those standards now in effect are listed in Table II, and Table III gives others under consideration. In two of the instances,

preparation of the standards awaits further experimental data. In addition, lack of data has limited the scope of a number of the standards that have been drafted.

Among these, the standard "Raschig Rings as Solid Neutron Absorbers in Solutions of Fissile Material" is being reissued to include low-enrichment uranium as though the ^{238}U were not present. An improvement would reduce conservatism if there were experimental data to allow for the effect of ^{238}U. There is still no experimental basis for correcting other deficiencies, namely, inapplicability to Pu-U solution except by ignoring the ^{238}U, to ^{233}U solution, and to absorbers such as polyvinyl chloride and boron steel that would be suitable for solutions that attack glass.

The "Guide for Nuclear Criticality Safety in the Storage of Fissile Materials" is being expanded to include enriched-uranium solutions as well as solids. Applications to storage of Pu-U, ^{233}U solutions, or undermoderated hydrogenous mixtures, might be derived from calculations verified by individual units, but would be strengthened if based on array experiments. Further, there is insufficient experimental information for generalizing the effect of an activated sprinkler on array criticality.

"Nuclear Criticality Safety Guide for Pipe Intersections Containing Aqueous Solutions of Enriched Uranyl Nitrate" orginally included ^{235}U, ^{239}Pu, and ^{233}U solutions and was revised to include low-enrichment uranium in solution. An experimental basis for extension to Pu-U and ^{233}U-Th solutions would be desirable.

"Nuclear Criticality Control and Safety of Homogeneous Plutonium-Uranium Fuel Mixtures Outside Reactors" may be excessively conservative because of considerable reliance on computed data. Improved experimental guidance should be useful for PuO_2-UO_2, and would be essential for a companion standard on ^{233}U-Th.

The draft standard "Soluble Neutron Absorbers for Criticality Control" has been prepared for boron in enriched uranium solutions, and is being extended to gadolinium in plutonium solutions. More experiments are required for application to other absorbers and to solutions of Pu-U and ^{233}U-Th.

Finally, the draft "Criticality Control of Special Actinide Elements" applies to all transuranic elements except ^{239}Pu. Because of scanty integral experimental guidance and reliance upon measured cross sections, its critical-mass estimates are extremely conservative. These estimates should be subject to improvement as significant quantities of the elements become available.

In summmary, experimental criticality information has led to an impressive number of safety standards. Nevertheless, numerous valuable additions await further experiments. I conclude from this that criticality safety as a discipline is mature but by no means senile.

TABLE I

ACCIDENTS IN PROCESSING PLANTS

DATE	PLANT	TOTAL FISSIONS	FIRST PULSE	DOSES (RADS)	Notes
6/16/58	Y-12	1.3×10^{18}	$\sim 7 \times 10^{16}$	365, 339, 327, 270, 236, 69, 69, 23	U-235 Solution Washed into Drum
12/30/58	LASL	1.5×10^{17}	1.5×10^{17}	~4400 (Fatal), 135, 35	Pu Concentrated in Solvent Layer
10/16/59	Idaho CPP	4×10^{19}	$\sim 10^{17}$	50 R, 32 R, Mostly Beta	U-235 Solution Siphoned into Tank
1/25/61	Idaho CPP	6×10^{17}	6×10^{17}	None	U-235 Solution Forced into Cylinder by Air
4/7/62	RECUPLEX	8.2×10^{17}	$\sim 10^{16}$	87, 33, 16	Pu Solution in Sump Sucked into Tank
7/24/64	Wood River Junction	1.3×10^{17}	$\sim 10^{17}$	10000 (Fatal), Two 60-100	U-235 Solution Poured into Tank
8/24/70	Windscale	10^{15}	$\sim 10^{15}$	Negligible	Pu Concentrated in Trapped Solvent
10/17/78	Idaho CPP	3×10^{18}	Unknown	None	U-235 Builduip in Diluted Scrub Solution

Table II

CURRENT CRITICALITY SAFETY STANDARDS

ANS-8.1, ANSI N16.1-1975: Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors

ANSI/ANS-8.3-1979: Criticality Accident Alarm System

ANSI/ANS-8.5-1979: Raschig Rings as Solid Neutron Absorbers in Solutions of Fissile Material

ANS-8.6, ANSI N16.3-1975: Safety in Conducting Subcritical Neutron-Multiplication Measurements In Situ

ANS-8.7, ANSI N16.5-1975: Guide for Nuclear Criticality Safety in the Storage of Fissile Materials

ANSI/ANS-8.9-1978: Nuclear Criticality Safety Guide for Pipe Intersections Containing Aqueous Solutions of Enriched Uranyl Nitrate

ANS-8.10, ANSI N16.8-1975: Criteria for Nuclear Criticality Safety Controls in Operations where Shielding Protects Personnel

ANS-8.11, ANSI N16.9-1975: Validation of Calculational Methods for Nuclear Criticality Safety

ANSI/ANS-8.12-1978: Nuclear Criticality Control and Safety of Homogeneous Plutonium-Uranium Fuel Mixtures Outside Reactors.

Table III

PROPOSED CRITICALITY SAFETY STANDARDS

In Preparation

ANS-8.13.1: Evaluation of Neutron Interaction by the Solid Angle Method

ANS-8.14: Soluble Neutron Absorbers for Criticality Control

ANS-8.15: Criticality Control of Special Actinide Elements

Under Consideration

ANS-8.17: Criteria for Nuclear Criticality Safety of Reactor Fuel Elements

ANS-8.YX: American National Standard Administrative Practices for Nuclear Criticality Safety

Awaiting Experimental Data

ANS-8.12.1: Processing Mixtures of Uranium and Plutonium Oxides

ANS-8.16: Maximum Subcritical Limits for Slightly Enriched Uranium Compounds Processed in the LWR Fuel Cycle

FIGURE CAPTIONS

Fig. 1. The Oak Ridge Gaseous Diffusion Plant. The U-shaped structure is the original plant, designated K-25. Each leg of the U is one-half mile long.

Fig.2. Layout of the first critical experiments at the Clinton Laboratories. In case of emergency, support of the table leaf could be collapsed, dropping part of the assembly.

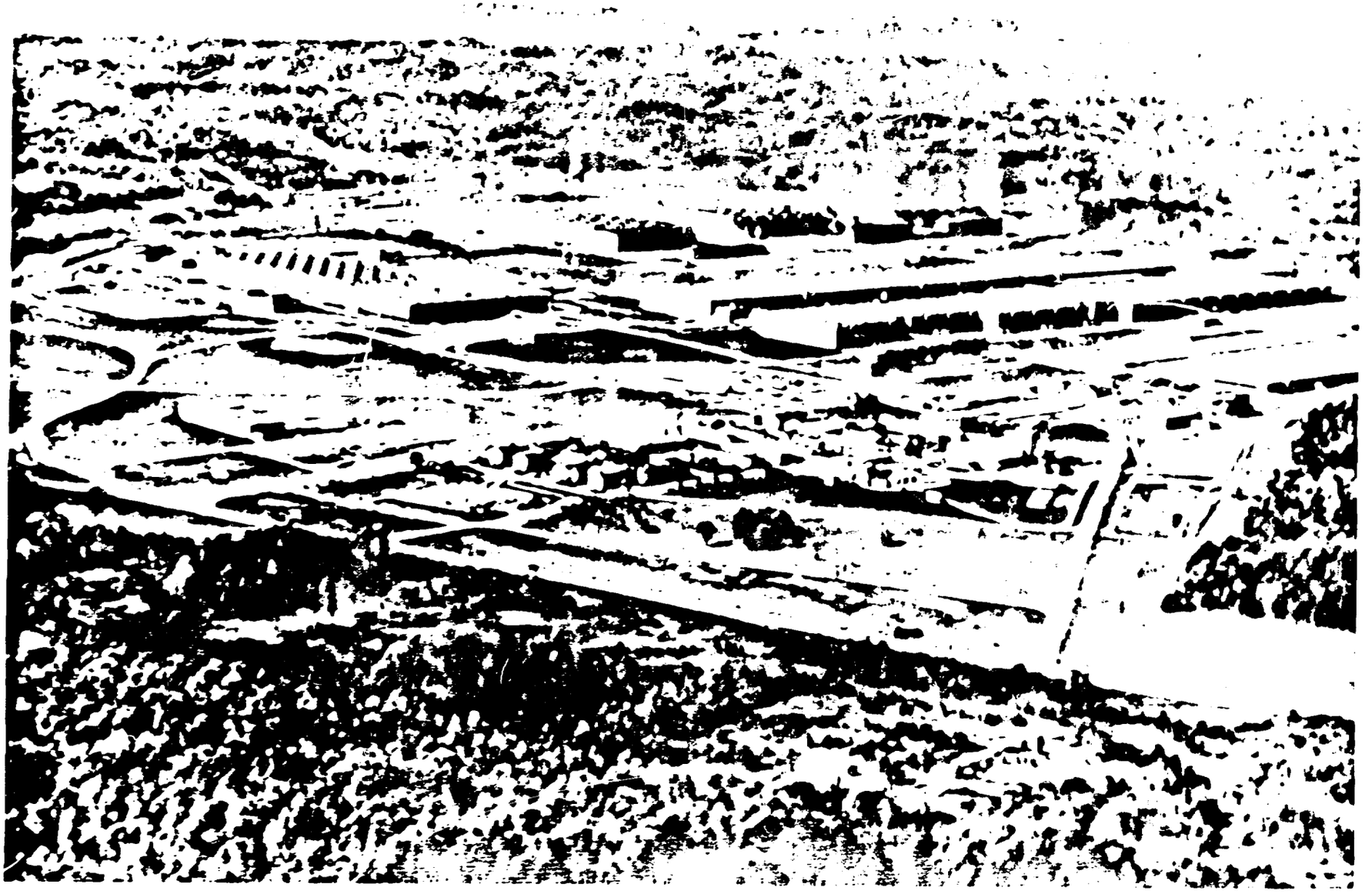
Fig. 3 Setup for the 1946 Los Alamos critical experiments with which Oak Ridge Personnel were active.

Fig. 4. Oak Ridge continuation of the 1946 Los Alamos critical experiments.

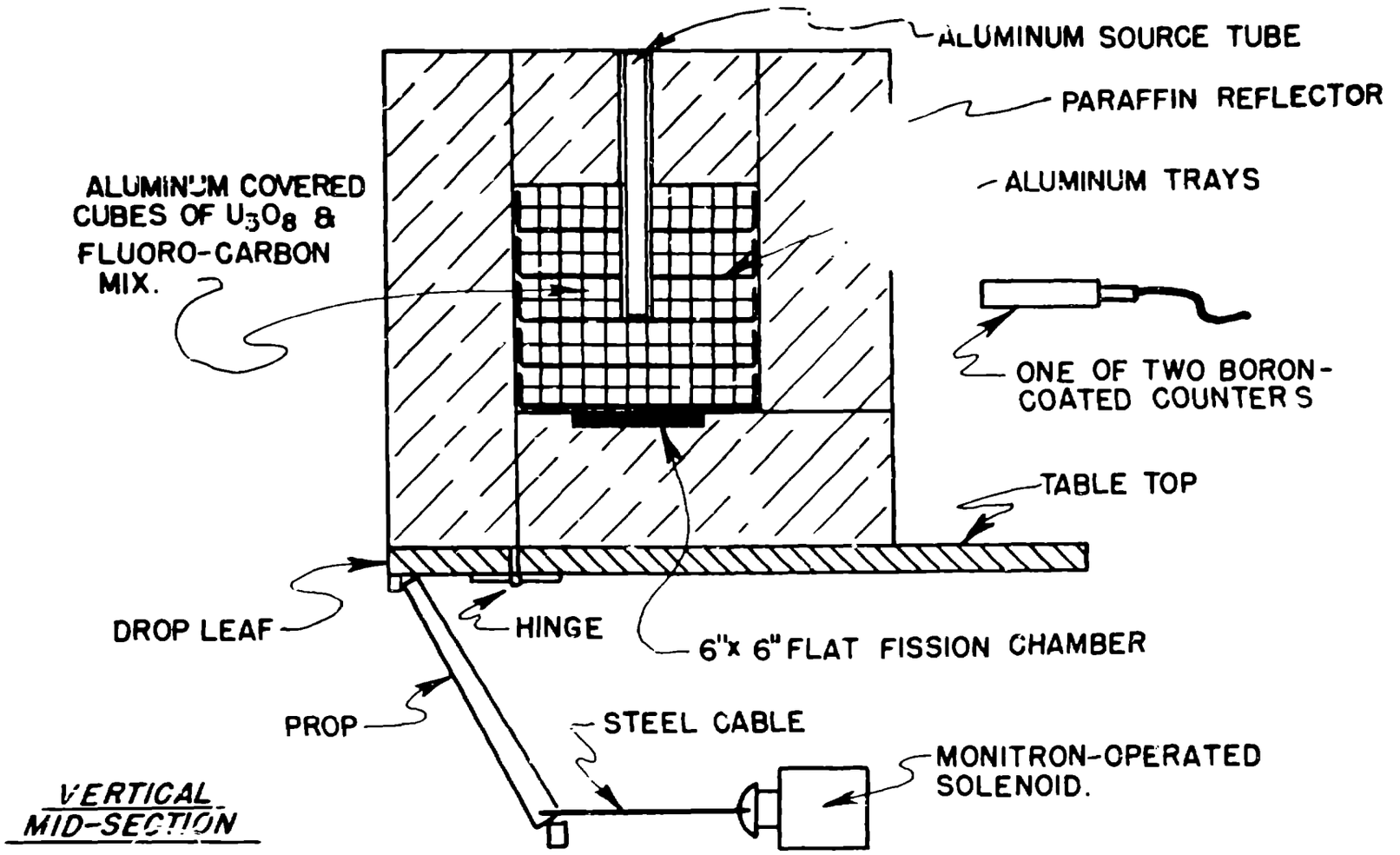
Fig. 5. Early glove boxes for batch precipitation of plutonium hydroxide.

Fig. 6. Modern glove-box complex for automated plutonium-purification line.

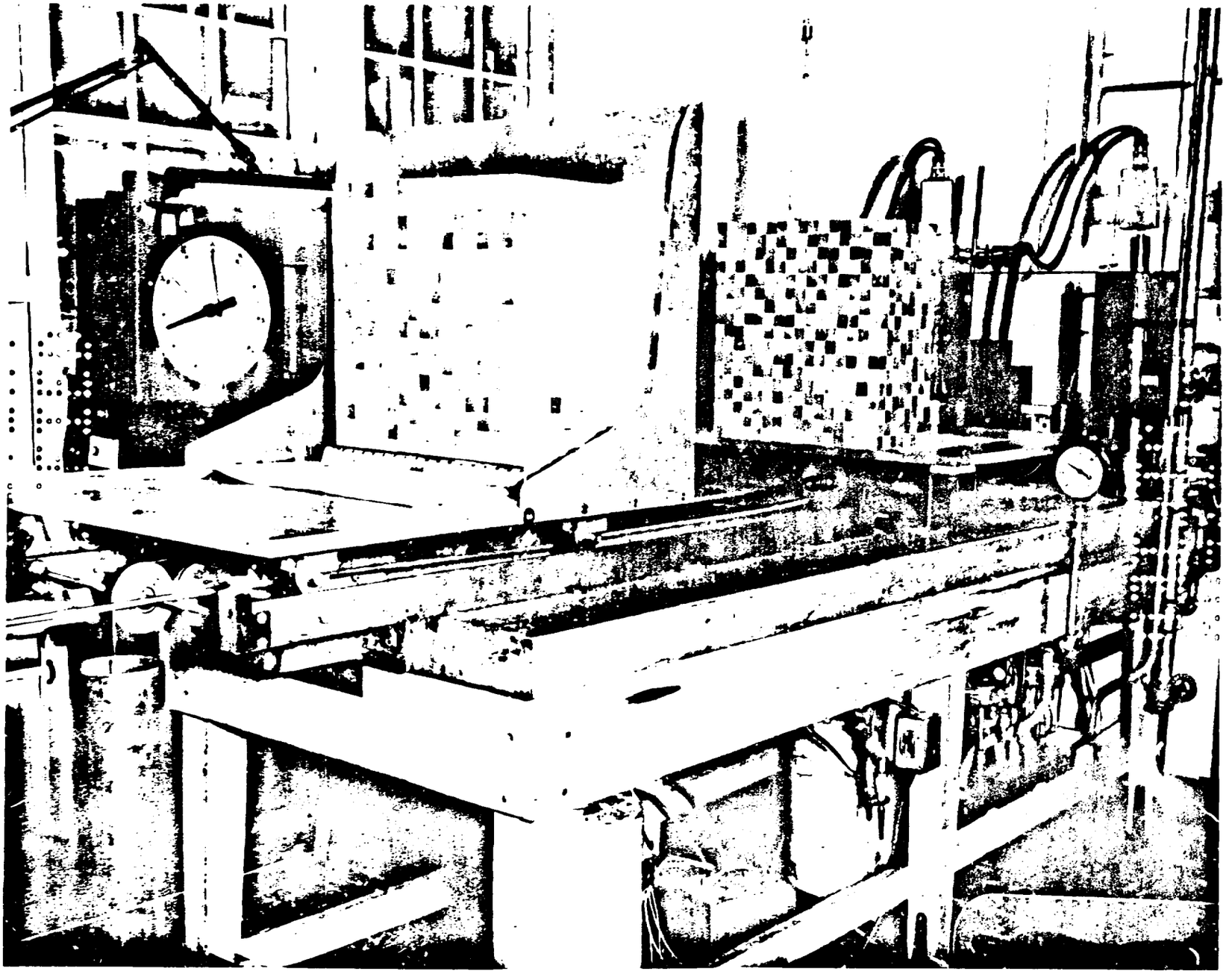
Fig. 7. Geometrically favorable equipment within the present plutonium-purification line. Typical batches are 1-kg Pu as compared with 160-320 g when the only control was administrative.

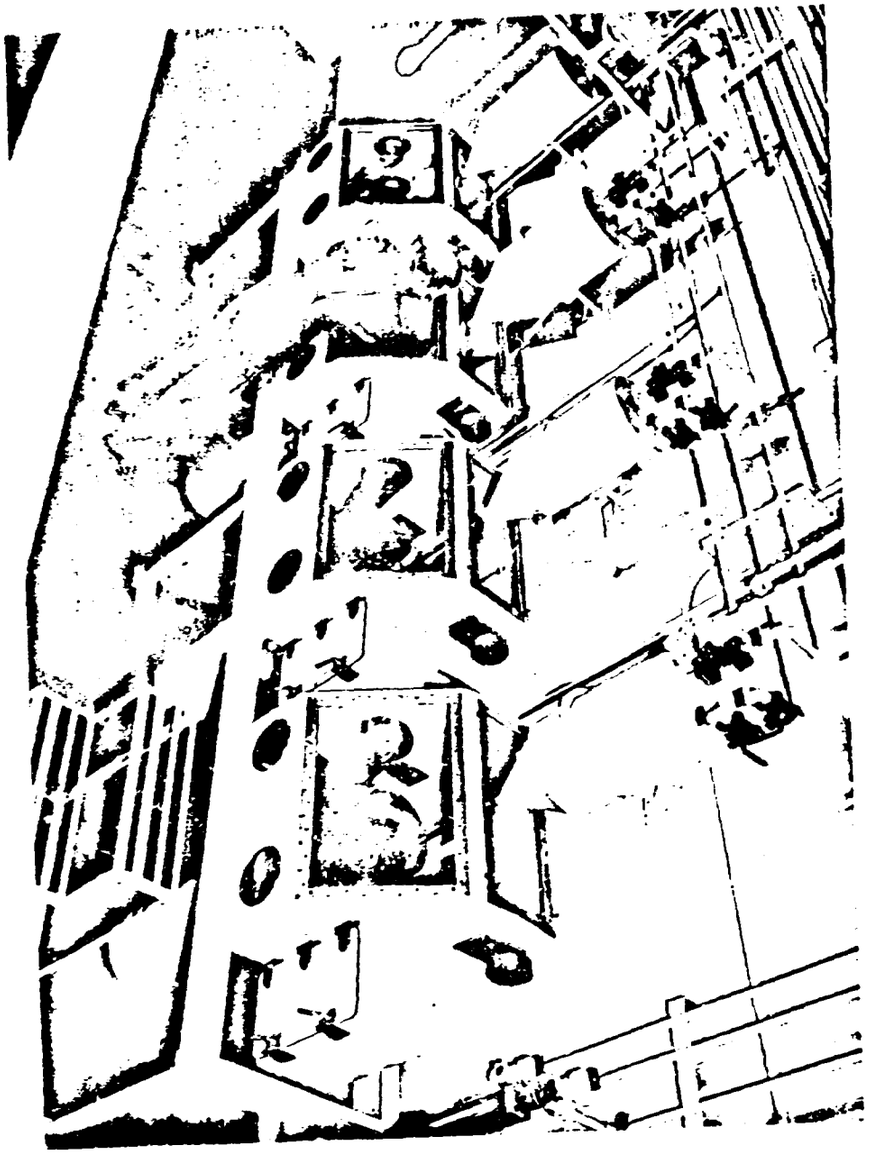


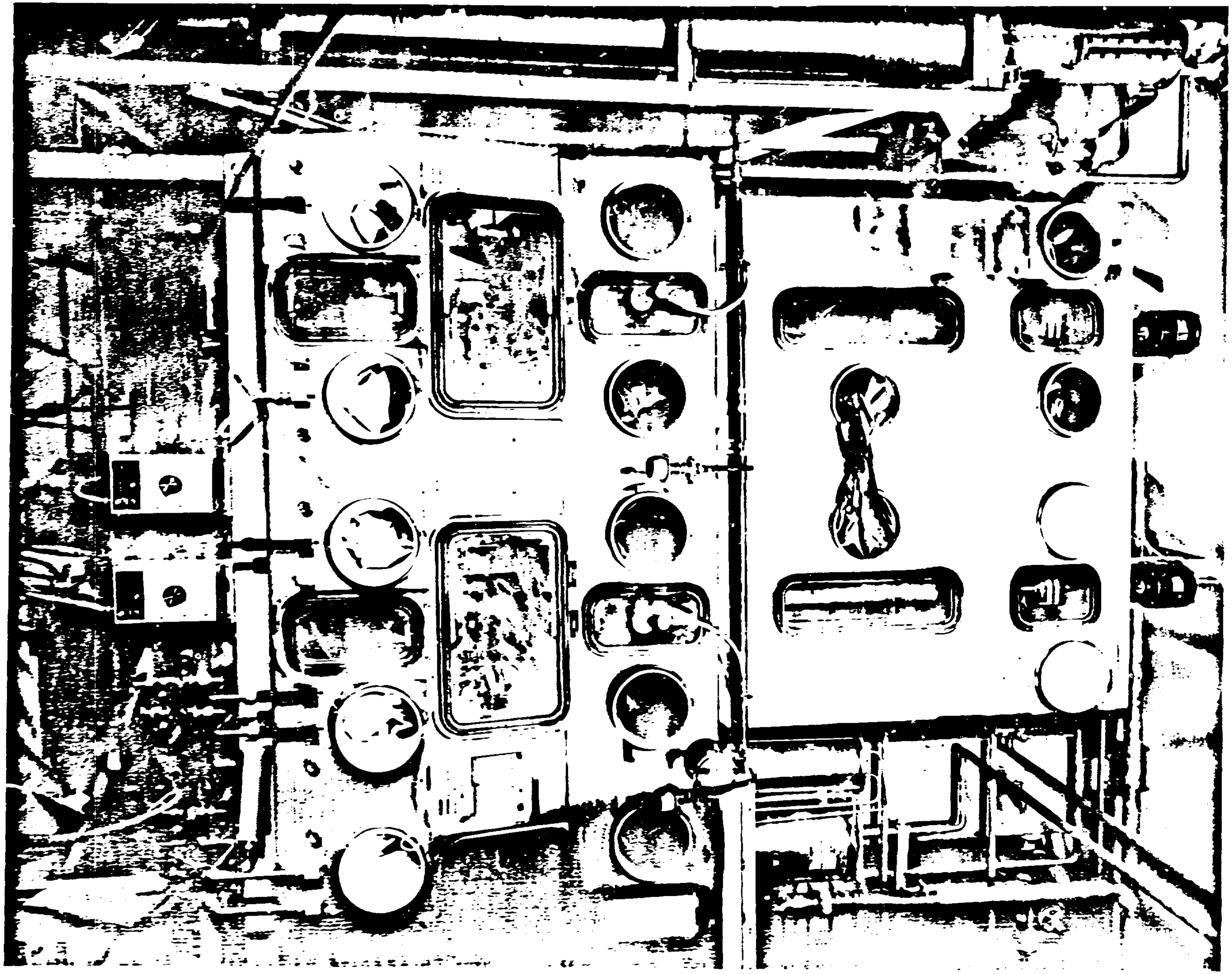
EXPERIMENTAL ARRANGEMENT OF CRITICAL ASSEMBLIES



VERTICAL MID-SECTION







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