WHC-MR-0437

A Brief History of the PUREX and UO_3 Facilities

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Date Published
November 1993

Prepared for the U.S. Department of Energy
Office of Environmental Restoration and
Waste Management

Westinghouse Hanford Company
P.O. Box 1970
Richland, Washington 99352

Hanford Operations and Engineering Contractor for the
U.S. Department of Energy under Contract DE-AC06-87RL10930

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Printed in the United States of America

DISCLM-1.CHP (1-91)

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Title: Brief History of the PUREX and UO3 Facilities

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BD-7600-062 (08/91) WEF074

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LIST OF TERMS

AEC	Atomic Energy Commission
DOD	Department of Defense
DOE	U.S. Department of Energy
DST	Double-Shell Tank
ERDA	Energy Research and Development Administration
FFTF	Fast Flux Test Facility
HOO	Hanford Operations Office
HW	Hanford Works
LOE	Level Operated Efficiency
MIBK	Methyl Isobutyl Ketone
NOX	Nitrous Oxide
NPH	Normal Paraffin Hydrocarbon
P&O	Pipe and Operating
PFP	Plutonium Finishing Plant
PRTR	Plutonium Recycle Test Reactor
PUREX	Plutonium-Uranium Extraction
REDOX	Reduction Oxidation
TOE	Time Operated Efficiency
UNH	Uranium Nitrate Hexahydrate
UO_3	Uranium Trioxide

METRIC CONVERSION CHART		
If you know	Multiply by	To get
inches	2.54	centimeters
feet	0.3048	meters
square feet	0.0929	square meters
gallons	3.7854	liters
pounds	0.4536	kilograms
Fahrenheit	F - 32 x 5/9	Centigrade

1.0 THE PUREX PLANT

1.1 GENESIS

The Plutonium-Uranium Extraction (PUREX) Plant, conceived amidst some of the direst imperatives of the early Cold War years, was a vehicle to increase significantly U.S. nuclear weapons production capacity. In September 1949, the Soviet Union had demonstrated its initial nuclear weapons capability by exploding a test atomic bomb over Siberia. Later that autumn, Soviet leader Josef Stalin signed a mutual assistance pact with Chinese Communist chief Mao Tse-tung, as Mao completed his defeat of nationalist Chinese forces and drove them to exile on Formosa (Taiwan). During the ensuing winter, some of the most famous spy cases in American history developed. Alger Hiss was convicted of perjury for denying that he had passed atomic secrets to the Soviets, and Klaus Fuchs confessed that he had conveyed similar secrets to Stalin's operatives over the six-year period during which he worked at the Los Alamos bomb development laboratory in New Mexico and at Britain's central atomic installation at Harwell. Through an obscure course of events, information from his confession led to the conviction and eventual execution of New York Communist Party members Julius and Ethel Rosenberg. In the United States, communism was perceived increasingly as a monolithic, worldwide threat--especially when the North Korean communist government of Kim Il Sung invaded South Korea in June 1950. That same year, the McCarran Internal Security Act was passed barring communists from working in defense plants, from obtaining passports, and from many other activities; and Senator Joseph McCarthy started his campaign of fear and accusation that communists had infiltrated multiple layers of the American government.

The United States responded to international events with vigor. When the Chinese Communist army joined the North Korean war effort in December 1950, President Truman declared a national emergency. Included in the emergency defense monies appropriated by the Congress were funds to construct many of the facilities that eventually formed the Atomic Energy Commission (AEC), now the U.S. Department of Energy (DOE) complex: the Rocky Flats, Savannah River, and Paducah Gaseous Diffusion Plants; the Nevada Test Site; the National Reactor Testing Station (now the Idaho National Engineering Laboratory); and others.

At the Hanford Works (HW), the mainstay of the nation's plutonium production, expansions already were underway. Since the earliest days of the Manhattan Project's plutonium separations experiments (1942), a continuous solvent extraction process had been desired. However, wartime deadlines had permitted only the development of batch-type separations processes based on repeated dissolution, precipitation, and centrifugation. Despite the nearly three-fold increase in processing rates in T Plant and B Plant between 1945 and 1950, the World War II plants could not hope to meet the defense projections drawn up by the AEC in the late 1940's.[1] By that time, however, the chemistry for the reduction oxidation (REDOX) process, the first continuous solvent extraction process in the world, was nearly perfected. Construction of the REDOX Plant began in 1949. Although it

was expected to be far more efficient than T Plant and B Plant, new defense projections drawn up by the AEC and the Department of Defense (DOD) showed a gap between capacity and need just as the Korean War escalated in late 1950 and early 1951. The REDOX chemistry, although workable, was not ideal. Because it was based on the use of 100 percent strength methyl isobutyl ketone (MIBK also known as hexone), an organic with a relatively low flashpoint (81 °F), many components of the REDOX Plant had to be built to explosion-proof standards. Also, the REDOX "salting agent," aluminum nitrate, could not be recycled and reused, thus rendering the plant expensive to operate both in terms of fresh chemical consumption and waste storage.[2]

Just as the Korean War entered its second full year in June 1951, the Hanford Operations Office (HOO) of the AEC began developing criteria and cost estimates for new facilities. At the request of AEC-HOO, a Facility Study Group was formed in September to examine Program X, the development of a new separations plant. It was determined that the new facility needed to process 200 metric tons of irradiated, aluminum-clad uranium (MTU) per month, exposed to neutron bombardment at an average level of 600 megawatt days per ton and aged not less than 90 days, given an 80 percent overall efficiency rate. (A complete explanation of the overall efficiency rate is in Section 1.3, Production Chronology.) Furthermore, the plant needed the potential to increase to 400 MTU per month with certain equipment changes. This capacity was necessary to handle the material generated by the twin "jumbo" reactors, 105-KE and 105-KW, new plants also planned for HW. The new separations plant was planned for the 200 East Area to provide a strategic distance from the West Area locations of the REDOX Plant and the Plutonium Finishing Plant (PFP) and their ancillary steam, electrical, water, and other support facilities. Also, extra, high-level waste storage capacity was available in the 200 East Area at C Tank Farm.[3]

Design engineering for the new processing facility began in July 1952 and construction in April 1953. The PUREX Plant was completed in April 1955. "Cold" runs (tests with unirradiated materials) were initiated in late 1955, and hot startup (work with radioactive substances) commenced in January 1956. Initial design costs totaled approximately $4.2 million while subsequent labor, construction, miscellaneous, and all-inclusive costs totaled approximately $74.4 million and involved 63,578 man-months of labor.[4] Figures 1, 2, and 3 show the PUREX Plant during construction.

1.2 DESIGN FEATURES

The original PUREX Plant was a concrete rectangle 1,005 feet long, 104 feet high (with approximately 40 feet below grade), and 61.5 feet wide. The shielding capacity of the concrete was designed so that personnel in non-regulated service areas would not receive radiation in excess of 0.1 millirem per hour.

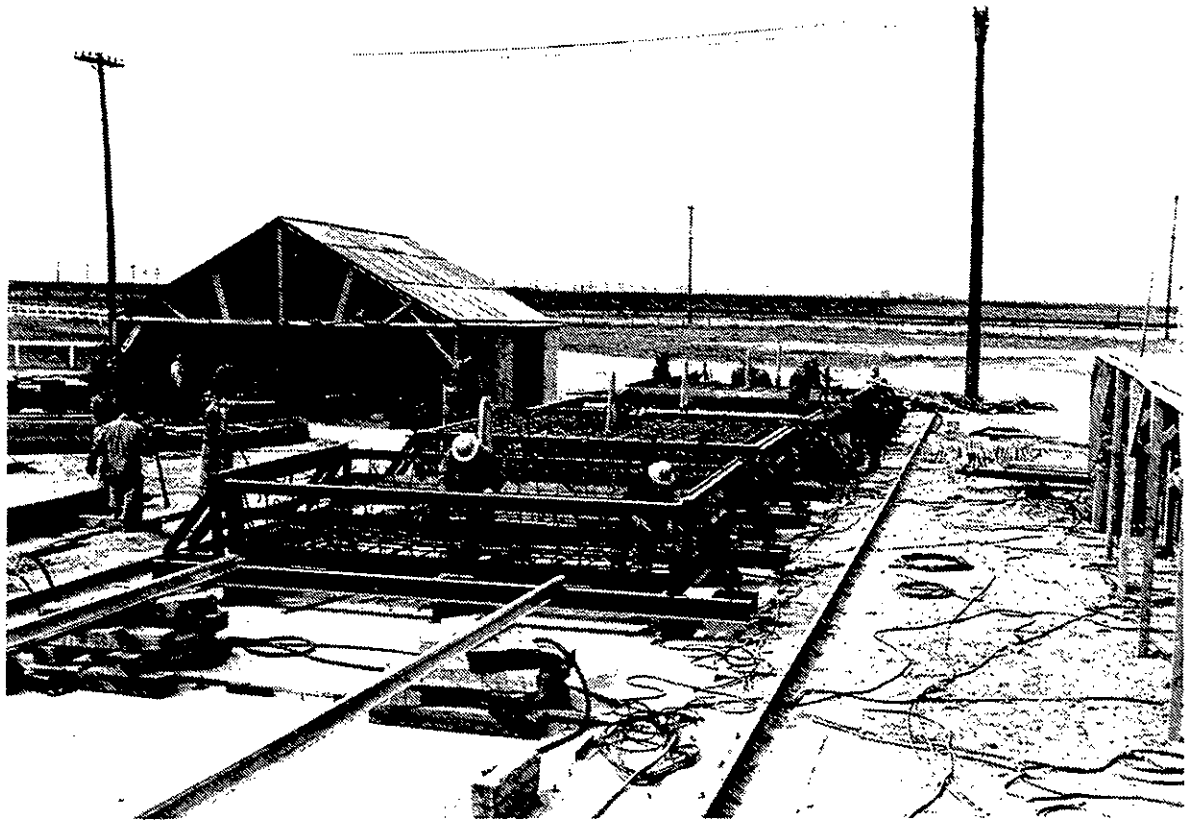

Figure 1. Reinforced concrete cell cover blocks being fabricated outside the 202-A (PUREX) Building, March 1954.

The Plant's main "canyon" portion, approximately 860 feet long, contained 11 cells designated A through H, J, K, and L. Each cell was 14 feet wide, 42.5 feet deep, and 39.5 feet deep from floor to underside of the reinforced concrete cover blocks. The cells were used as follows:

- A-, B-, C-Cells — housed metal dissolution equipment and activities
- D-Cell — contained equipment used in preparing dissolved metal into feed solution for the PUREX process
- F-Cell — housed waste treatment and acid recovery operations
- G-Cell — used for organic recovery (sometimes called first-cycle solvent recovery)
- H-, J-, K-, L-Cells — incorporated remaining solvent extraction and concentration steps.

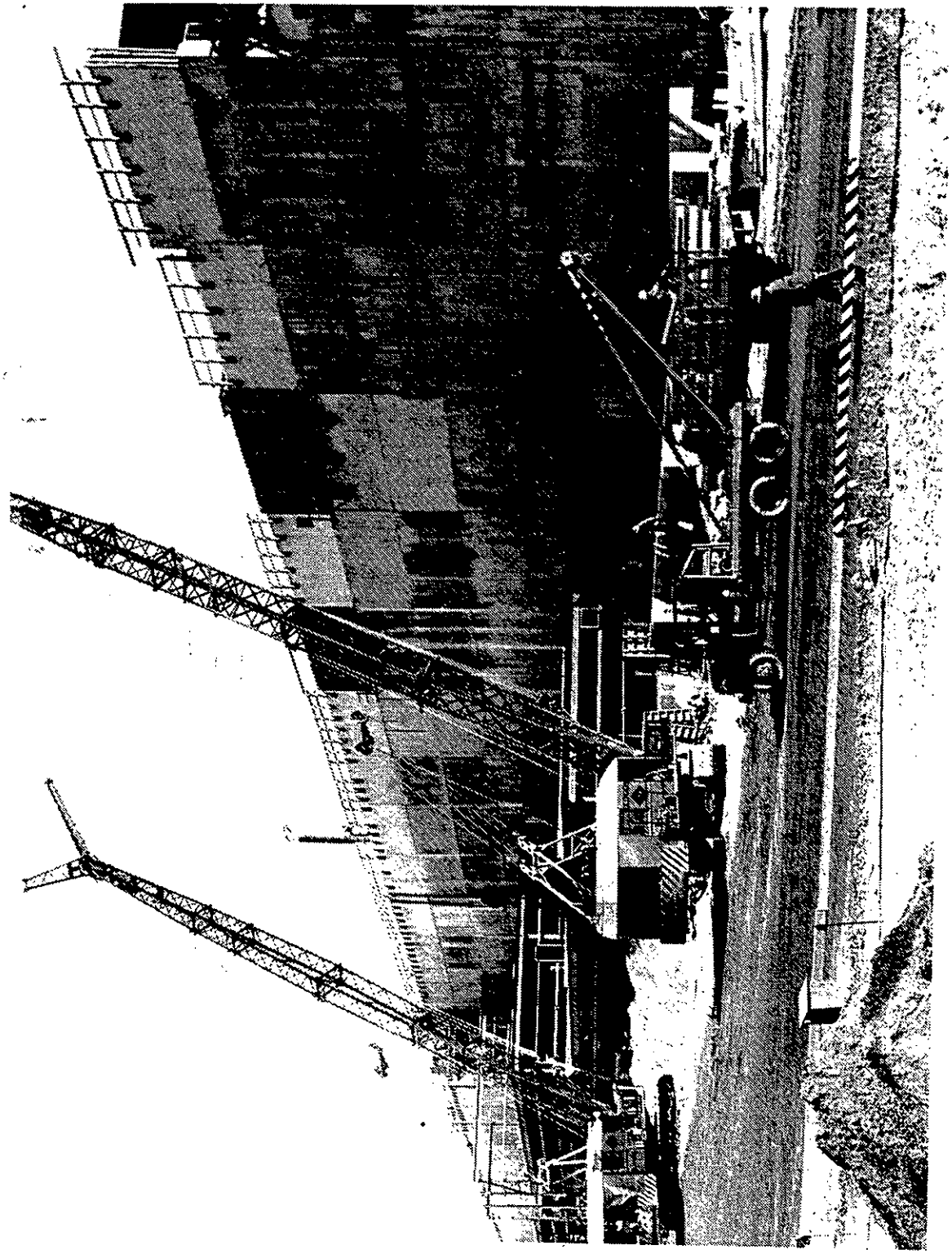

Figure 2. The PUREX Building under construction, April 1954.

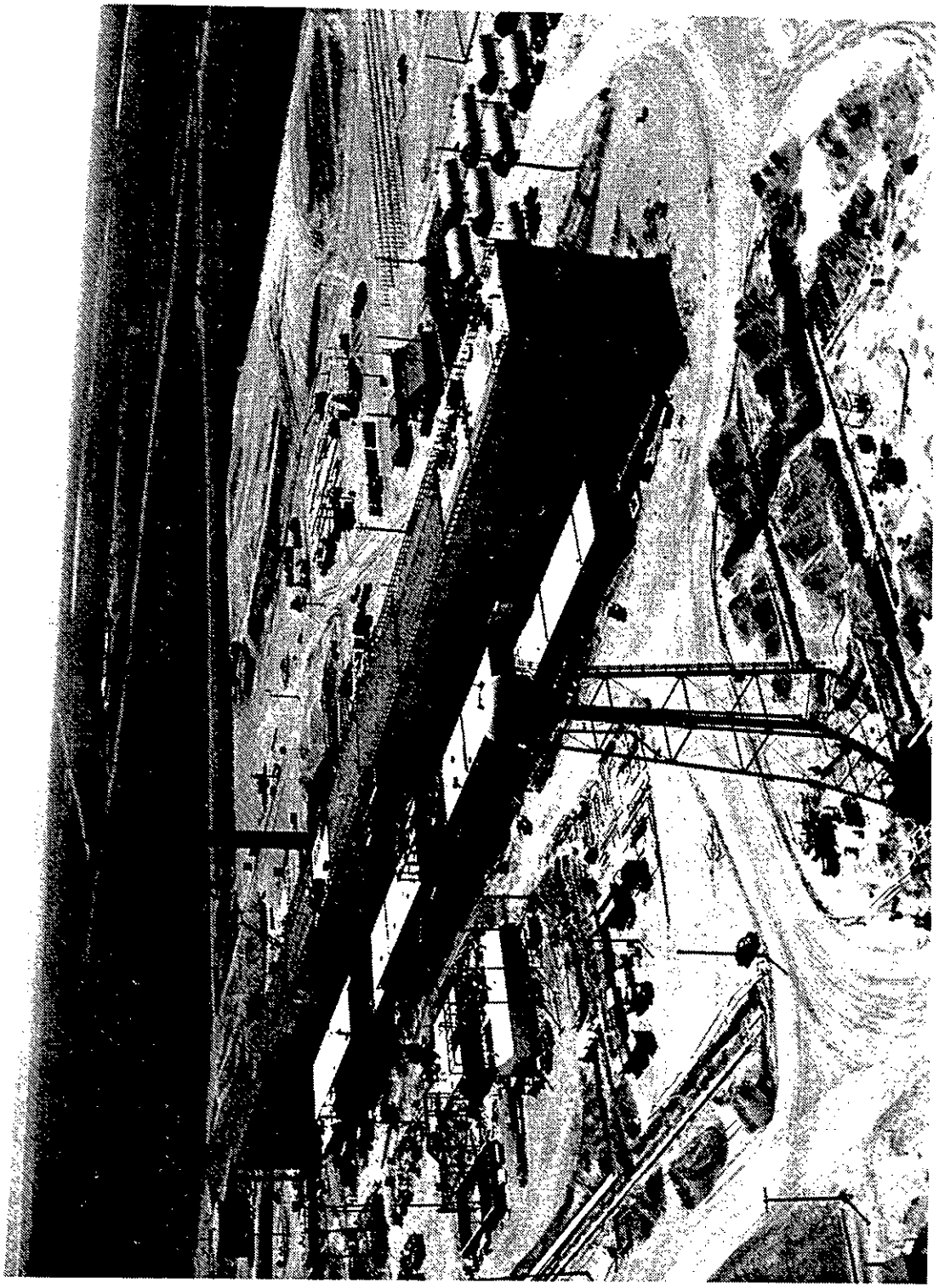

Figure 3. Aerial view of the PUREX Plant under construction showing the temporary construction buildings and huts, May 1954.

Figures 4 and 5 show J-Cell was used for uranium decontamination, and L-Cell for plutonium decontamination.[5]

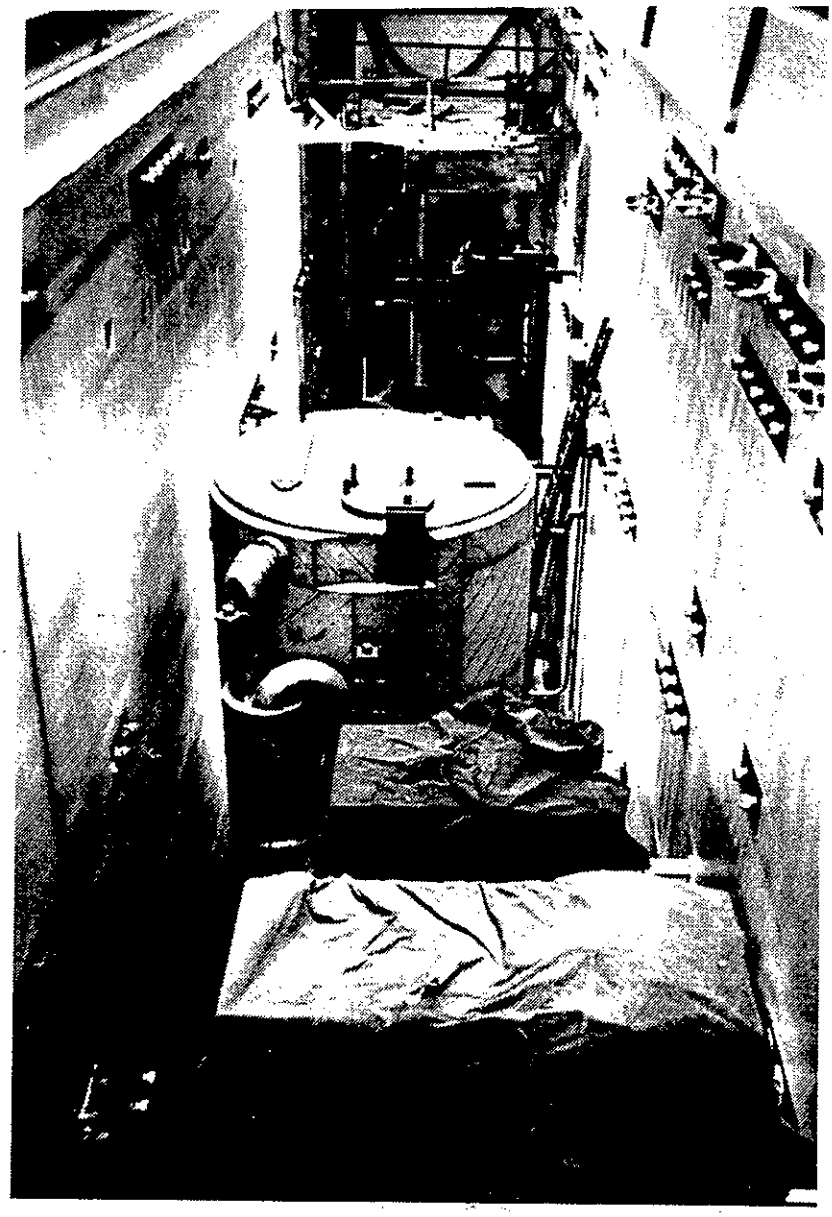

Figure 4. Processing equipment being lowered into J-Cell, PUREX Building, November 1954.

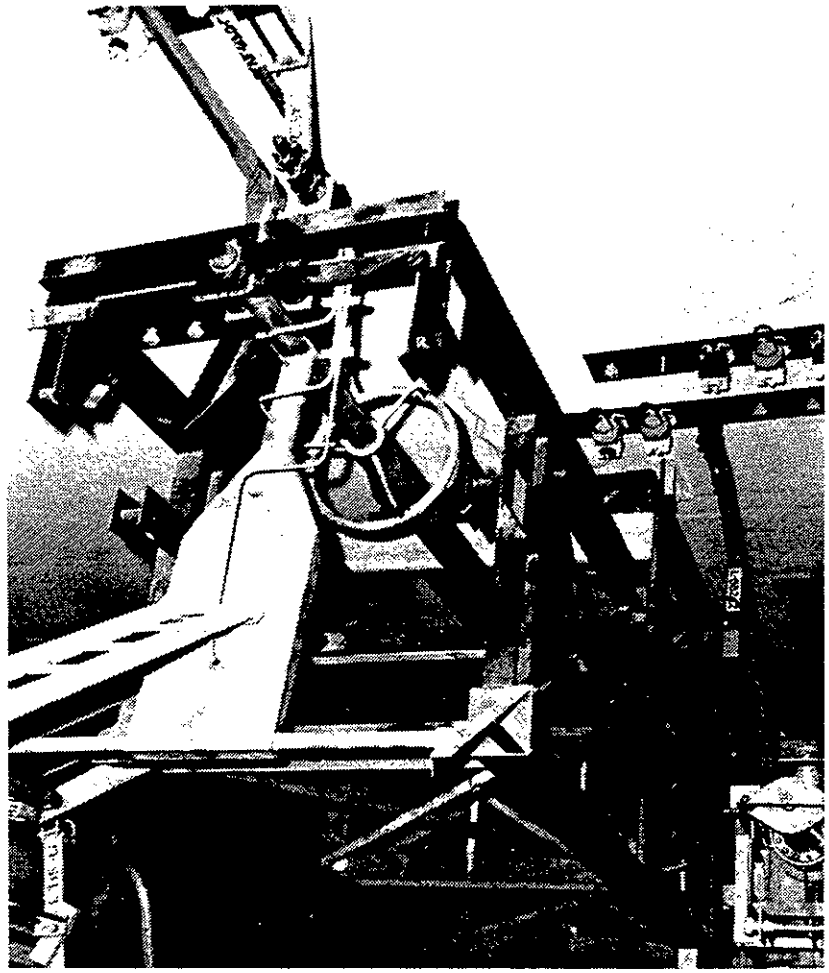

Figure 5. Plutonium decontamination equipment ready to be installed in L-Cell, PUREX Building, November 1954.

Additional portions of the PUREX Plant (the 202-A building) were comprised of a hot pipe trench (an 860-foot by 12-foot by 33-foot enclosure which housed pipes that transported radioactive solutions) and an air tunnel (an 860-foot by 11-foot by 7.5-foot space through which contaminated air from the canyon and the process ventilation system passed on its way to the filters and the 291-A Stack). See Figure 6. Also, paralleling the cells on the side opposite the hot pipe trench and air tunnel, were the crane cab gallery, the pipe and operating (P&O) gallery, the sample gallery, and the storage gallery. A 40-ton general utility or "main" crane (also known as the remote crane) serviced the canyon along with a 40-ton auxiliary or "slave" crane. The slave crane, which operated 17 feet above the main crane, was designed to handle tall pieces of equipment such as columns, concentrators, dissolver towers, and some pipe jumpers. The main crane was equipped with closed-circuit color television for viewing the canyon area, the first in a Hanford processing plant. Additionally, a regulated area of the 202-A Building contained M-Cell with a water pool for

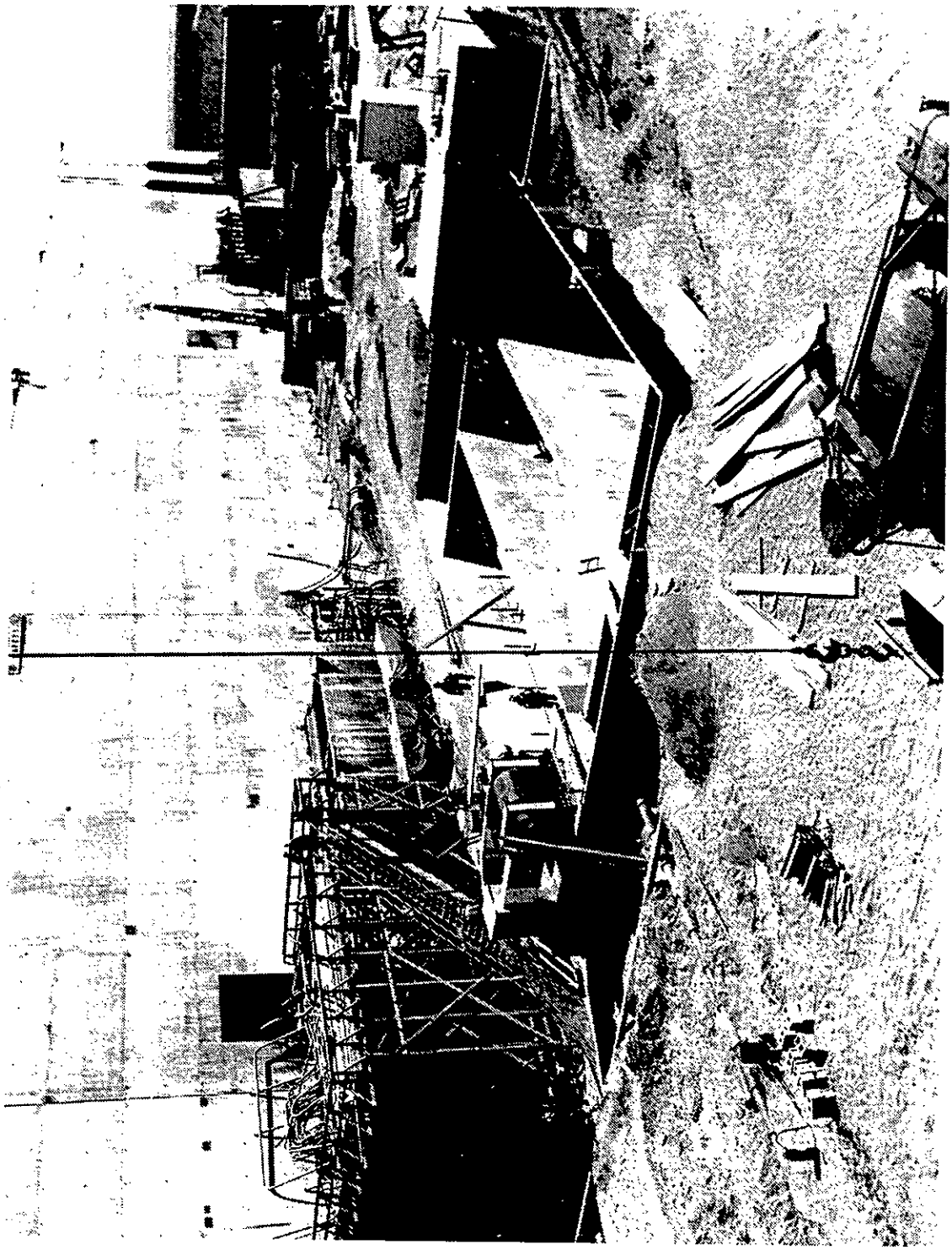

Figure 6. The PUREX filter building under construction, July 1954.

decontamination of major process equipment, a "hot shop," and a regulated shop for decontamination and repair of smaller equipment having varying levels of radioactivity. The decontamination cell incorporated leaded glass windows and flexible jumpers for the first time in a Hanford processing plant.

Adjoining the north wall of the main 202-A Building was a 750-foot long, 60-foot wide service area containing three control rooms (central, head end, and power unit), the PUREX process control laboratory, the aqueous make-up and storage area, and the acid concentration vault. An underground solvent storage and make-up facility was adjacent to the service side of the 202-A Building. Many other adjunct and service facilities were built close to the PUREX Plant. These included the following: the 203-A Uranium Storage and Pumping Station; the 207-A Retention Basin; 211-A Chemical Tank Farm (see Figure 7); the 252-A Electrical Substation; the 272-E Mock-Up Building; additions to the 282-E Raw Water Pump House, the 283-E Filter Plant, and the 284-E Power House; the 291-A Stack and Filter Building; a series of electrical and piping facilities, warehouses, rail systems; and many other structures. Waste storage and disposal facilities also were constructed (see Section 1.6, Waste Management).[6]

The design of PUREX equipment and systems incorporated several unique features. Of these, the most important for overall operations were the use of pulse columns to achieve chemical separation rather than packed columns or mixer-settlers, and the development of "liquid-liquid-solid-type centrifuges." Packed columns had been used in the REDOX Plant and had necessitated the construction of the tall silo or "penthouse" on the west end of that plant. Such columns were ruled out for the PUREX Plant because a height reduction of 50 percent with its attendant financial savings could be achieved by other means to attain contact between the aqueous and organic solvents and the feed solution. Pulse columns became the design choice because solids could be removed from them more easily than from mixer-settlers, and the smaller volume-per-unit capacity of pulse columns permitted faster detection of off-standard conditions. Between 1950 and 1953, 50 man-years of research and development were applied to perfecting the PUREX pulse column design. One of the new features was the insertion of swirl plates into the columns at several heights to improve contact and mixing of the aqueous and organic solutions by reducing channeling effects. The action of the plates increased mass transfer and resulted in more efficient column operation. The first "packed pulse" column to be used at the Hanford Site was installed in L-Cell as the 2A column. In addition, a special agreement with a subcontractor resulted in the development of the first three-phase, 48-inch bowl centrifuge of its kind in the world. It was designed during the first nine months of 1953 for use in the PUREX process.[7]

Other unique PUREX Plant features included the following: an irradiated fuel element (slug) storage basin, the first of its kind, located within a separations facility; a railroad tunnel that permitted unloading of contaminated cask cars without compromising the ventilation system; and a "soft wall" at the east end of the building consisting of concrete blocks and grout that could be removed for the installation of an additional crane or to enlarge the building at a future date. In the ventilation system, an initial glass wool filter was chosen instead of the sand filters used at earlier HW processing plants. The overall

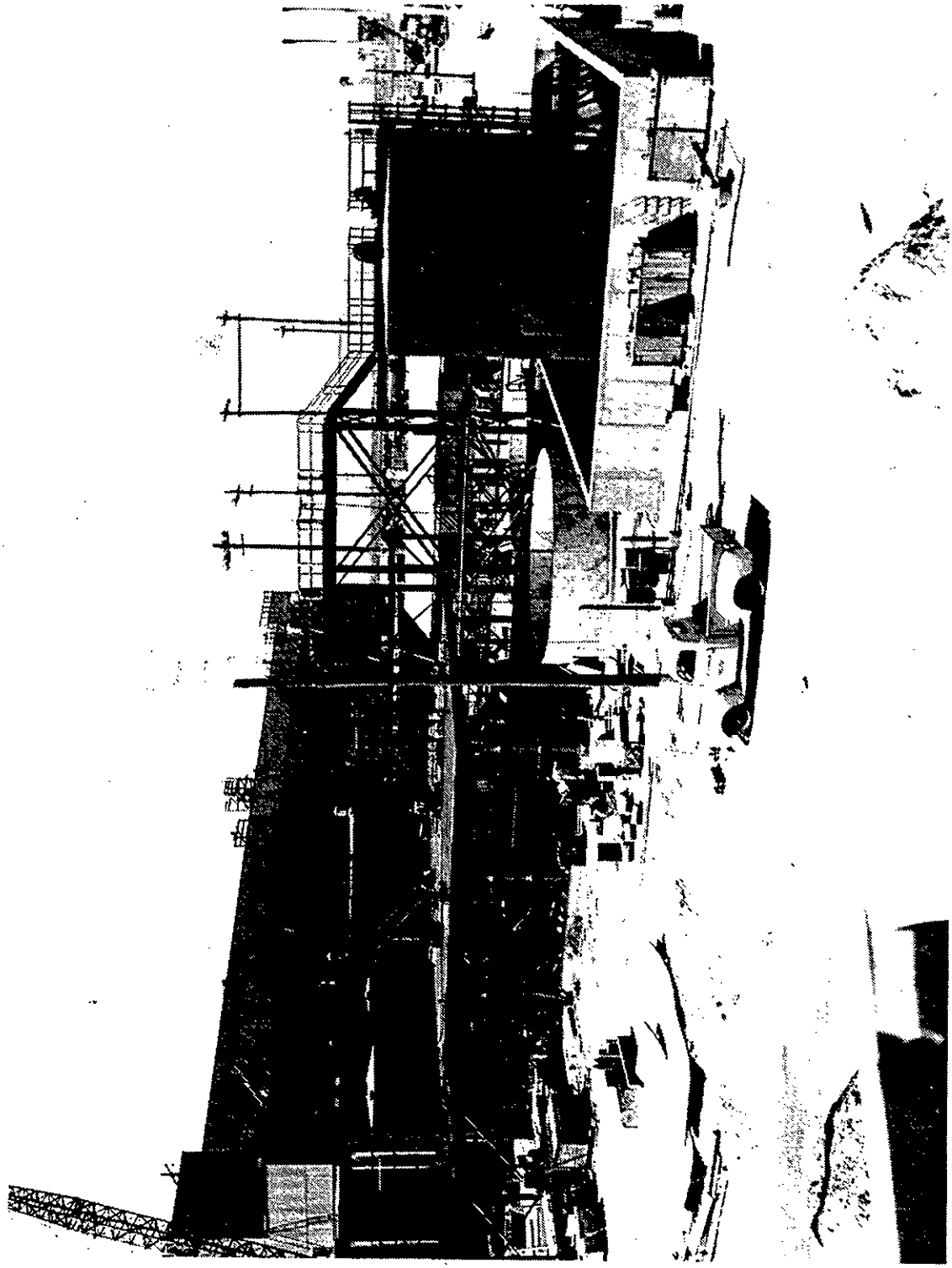

Figure 7. The 211-A Chemical Tank Farm, serving the PUREX Building, under construction, July 1954.

ventilation system was also designed to have three times the capacity of the REDOX system so that air of considerable force could enter the cells when cover blocks were removed and prevent the escape of contaminated particles from the cells into the canyon area. The PUREX jumpers, like most other equipment in the plant, were larger and longer than those at the REDOX facility because of the greater depth of the process cells. Like other PUREX apparatus, they were standardized to be removable and interchangeable from a remote location. A single drawing and specification could permit the construction of up to 27 jumpers. The jumpers utilized pipe bends instead of expansion joints to prevent the leakage that had occurred with such joints at other Hanford plants. "Debottlenecking," another unusual design characteristic within PUREX, enabled the piping to accommodate 50 percent more flow, allowing for future expansions.[8]

A number of unprecedented features were built into PUREX instrumentation systems and equipment. Some of these were so rare that commercial vendors could not supply them, and they had to be fabricated in 300 Area technical shops. New inventions included the X-ray photometer used to determine the relative X-ray absorption properties of various process solutions, and in-line gamma energy spectrum monitors, used to scan a representative portion of the process stream and record the results on a continuous basis. The monitors could scan 12 streams every 30 minutes without removing a single physical sample from the process, thus saving an estimated 3,000 hours of laboratory time per month. Miniaturized instrumentation in a graphic panel display board exhibited the readings of the crucial rotameters, pneumatic transmitters, electric interface detecting probes, and sampling instruments. The Gilmont pipette sampler,[a] designed for the most radioactive substances, was another key innovation. The sampler was hydraulically filled and discharged by an operator in the sample gallery and attached to a separate ventilating hood.

The vertical "basket" type tube bundles used in the PUREX concentrator heat exchange units also were designed specifically for the plant. They were 12 feet long and 3 feet in diameter, and they contained 630, 12-gauge stainless steel tubes, each 10 feet long with a 1-inch outside diameter. A number of remote maintenance puzzles and difficulties with the large remote flange were overcome in the design of this equipment. The PUREX agitators also were unique in that they had no gear boxes and no bearings below the motor to guide the long shafts. Such features were chosen to reduce the frequency of breakdowns, but they also necessitated running the agitators at the relatively slow speed of 580 rotations per minute. In another innovation, pump bearings serving the PUREX canyon vessels were lubricated with process solution, while the drive bearings were lubricated by a permanent grease reserve. Again, these features served the needs of remote maintenance and were designed to prolong equipment life.[9]

---

[a]No registered trademark can be found for Gilmont samplers in the time period of reference.

1.3 PRODUCTION CHRONOLOGY

The PUREX design production rate was based on two crucial concepts that tracked the efficiency of various types of operating plants including the 100 Area reactors and the 200 Area separations facilities: Time Operated Efficiency (TOE) and Level Operated Efficiency (LOE). The TOE rating referred to the amount of time that a facility actually ran compared to the amount of total time existing in a given period. At PUREX, the designed TOE rating was 90 percent, or 27 days per month, which allowed for maintenance outages, systems flushings, and unscheduled difficulties. The LOE rating referred to the amount of time during those periods when the facility actually ran at peak capacity. In other words, the startup time that it took reactors to achieve full power after shutdowns, and the initial dissolving periods that it took processing plants to fill all of their systems with solution after shutdowns brought the LOE rating to varying levels below 100 percent. For the HW processing plants, theoretical operation at an LOE rating of 100 percent was known as the capacity factor or the instantaneous rate, that is, the rate at which the plant could process material if its startup occurred instantly rather than over a period of hours. During the design phase, it was estimated the PUREX instantaneous processing rate would be 8.33 MTU per day and the LOE rating would be 89 percent. Combined with the TOE of 90 percent, the plant's overall efficiency rating was estimated at 80 percent or a planned production rate of 200 MTU per month.[10]

As the PUREX Plant was being built during 1954 and 1955, a simultaneous project was underway at HW to upgrade and increase the processing capacity at the World War II B and T Plants. The project was called the 4X Program because it planned to utilize the capabilities of all four Hanford processing plants (B, T, REDOX and PUREX). This endeavor actually completed renovations at B Plant in December 1955 and was fully involved in the design phase of T Plant upgrades. However, the first year of PUREX operations demonstrated such an overwhelming production capacity, along with economic efficiency as compared to the other separations plants, that the 4X Program was abandoned. T Plant was shut down as a processing facility, and B Plant, closed since 1952, was not restarted.[11]

During 1956, the PUREX Plant processed 56 percent of the total plutonium output of HW, a 59 percent increase over 1955. In September 1956, PUREX demonstrated a sustained, instantaneous rate of 16 MTU per day and an astounding on-line efficiency of 99 percent. In 1957, the total output of plutonium from HW increased 54 percent over 1956 with PUREX processing 71 percent of the new total. That October, the plant demonstrated an instantaneous production rate of 20 MTU per day with a reduction in the amount of residual fission product activity in the product of 25 times below design specifications. During 1958, the total HW output of plutonium increased 5 percent over 1957 with PUREX contributing 79 percent of that total. That year, the decision was made to send all of the standard irradiated uranium from the 100 Areas production reactors to PUREX and to process 94 Metal (enriched uranium containing 0.947 percent uranium 235 by weight) to the REDOX Plant. REDOX equipment was reconfigured and handling techniques were changed to accommodate the higher enrichment levels of the 94 Metal. Additionally in 1958, the

PUREX facility began the recovery of neptunium 237 (Np-237) on an occasional batch basis from its normal product stream. Immediately, the plant became the AEC's principal supplier of this isotope.[12]

The late 1950's and early 1960's witnessed even more spectacular production leaps at the PUREX facility. By October 1960, the plant had processed 22,000 tons of irradiated fuel, thus surpassing the combined total of B and T Plants and the REDOX facility for all of their years of operation. For short periods of time, PUREX demonstrated the capacity to operate at 3.6 times its design capacity. The plant also exceeded its previous Np-237 production figures by producing 7.4 kilograms, 92 percent of the national supply. From 1960 to 1964, the two HW separations plants processed approximately 35,000 tons of irradiated fuel, with PUREX handling about 80 percent of the total. During 1962, both REDOX and PUREX were modified to permit the continuous recovery of Np-237 without interfering with normal plutonium recovery operations. In late 1965, other PUREX systems were modified to allow the plant to sustain normal operations at a 4.0 capacity factor, that is, 33 MTU per day.[13]

Beginning in 1963, the PUREX Plant was modified to enable the processing of various fuel types including fuel from N Reactor. (Further discussion of PUREX Plant modifications dealing with N Reactor fuel is in Section 1.4, Projects and Equipment Changes.) N Reactor fuel elements were much larger than the 4- to 8-inch slugs from HW's single-pass reactors. The new elements were approximately 26 inches long, weighed about 52 pounds each, and had a tube-in-tube configuration. They were clad not in aluminum but in Zircaloy-2, an alloy comprised mostly of zirconium and small amounts of nickel tin, chromium, and iron. Zircaloy-2 was harder and slower to dissolve than aluminum cladding. Therefore, once the separation of N Reactor fuel began in 1967, the entire context of production rates and numbers changed for PUREX. Although processing 5,000 to 7,000 MTU per year had been normal for the plant during the early years of the 1960's when single-pass reactor slugs had comprised the irradiated portion of the feed material, the processing rate now fell to about 2,000 MTU per year for N Reactor fuel. Even lower rates occurred in 1970 when regular processing runs were interrupted by thoria fuel separations. In 1972, only about 1,000 MTU were processed for the normal recovery of plutonium because part of the plant's capacity was diverted to the separation of mixed oxide fuels from Hanford's Plutonium Recycle Test Reactor (PRTR). PUREX began a long shutdown period that September.

During 1965 and 1966, the PUREX facility processed powdered thorium oxide fuel targets that had been irradiated for the production of uranium 233 (U-233), an isotope desired for its potential use in weaponry, and because it could be made from plentiful, natural thorium. However, the processing campaign caused plugging and other equipment and contamination problems within PUREX. A more successful campaign in 1970 processed pelletized thorium oxide targets. Shortly thereafter and for reasons unrelated to PUREX, thorium oxide fuel was ruled out for large-scale development.[14]

When the REDOX Plant closed in 1967, PUREX became the sole, operating processing facility at the Hanford Site. In 1972, the PUREX Plant entered a temporary shutdown period that lasted 11 years. All of HW's single-pass reactors had closed, and most of the available (aged) N Reactor fuel had been processed. The initial PUREX shutdown was planned for 18 months to allow the accumulation of N Reactor fuel and to clean out selected high-level waste tanks in the A Tank Farm. Sluicing was to be used to remove solids and associated heat content so that the tanks could be reused for high-level waste storage. Other projects planned and completed during the same period included the installation of a condensate recycle system and a separate offgas system for ammonia-bearing effluents. As the shutdown period progressed, the initial sluicing effort took longer than expected, and a startup delay to 1976 was announced in mid-1973. This time period coincided with some of the biggest changes in waste disposal and environmental policy in the history of the Hanford Site. In the summer of 1973, the largest known high-level waste tank leak occurred in tank 241-T-106. Subsequent investigations by the Energy Research and Development Administration (ERDA), successor agency to the AEC, the operating contractor, and the media led to the decision to store new wastes only in double-shell tanks and to involve the public in the development of an Environmental Impact Statement on high-level defense wastes. The PUREX restart was delayed another two years as double-shell tank farm facilities and other projects were completed.[15]

Other issues arose during the shutdown period including environmental concerns that led to providing upgraded filtration systems, seismic safeguards, backup power sources, and many other projects. Increased safety concerns about shipping plutonium in nitrate form from PUREX in the 200 East Area to the Plutonium Finishing Plant in the 200 West Area led to an additional delay. The shutdown period was extended while facilities were added within the PUREX Plant to convert plutonium nitrate to plutonium oxide. Several additional upgrades and changes also were made. (Further discussion of these projects can be found in Sections 1.4 and 1.6, Project and Equipment Changes and Waste Management, respectively.)

In 1983, the PUREX Plant reopened with an operating limit that allowed it to process up to 3,000 MTU per year of N Reactor fuel, or about 8 MTU per day. However, the highest sustained but short-term production rates in the ensuing years hovered near 7 MTU and the highest long-term rates hovered near 3 MTU per day. Following a safety violation, the PUREX Plant closed for about six weeks in 1988. It closed again for a year beginning in December 1988 after steam pressures fell below levels necessary to support backup safety equipment. Additional equipment repairs and improvements to waste handling systems also occurred during that closure period. After a stabilization run lasting only a few weeks, the plant again closed in early 1990 to prepare additional environmental and safety documentation and facility upgrades. In October of that year, it was placed on standby status by Secretary of Energy James Watkins. A final closure order was issued by DOE in December 1992.[16]

1.4 PROJECTS AND EQUIPMENT CHANGES

The PUREX Plant underwent many changes to its systems and equipment over the years, becoming more simple and efficient and more flexible and multifaceted at the same time. Some of the changes were made even before operations began. A "cold semi-works," (a prototype that used unirradiated materials) to test some of the planned PUREX equipment, was installed in the 321 Building in 1953. A pulse column testing facility was built along with a long-tube evaporator for the concentration of uranium solutions (uranium nitrate hexahydrate [UNH]) to reuse in column tests. In the very first trial on July 1, the concentrator feed pump failed, and over concentration occurred to the point of incipient denigration. As a result, changes were made in the concentrator valving and instrumentation. Soon afterward, Hanford's C Plant (hot semi-works), a test facility in the 200 East Area, was fitted to conduct trials of PUREX equipment full-strength radioactive solutions. As a result of tests demonstrating the evolution and release of more ammonia than expected, an ammonia scrubber was added to the PUREX design.[17]

Additional design changes were incorporated into PUREX before the startup included a silica gel "tail-end" treatment facility and a low-pressure nitric acid fractionator. The fission products, ruthenium 103 and 106 (Ru-103/106) and zirconium and niobium 95 (Zr/Nb 95) were removed from the UNH product by running it through a bed of the silica gel. In particular, Ru-103/106 had caused contamination problems around the REDOX Plant from 1952 to 1954. A nitric acid concentrator, which was originally designed for F-Cell as part of the acid recycling system, was replaced with a unit outside the canyon in the 276-U structure. The unit could be operated under vacuum at much lower temperatures, thus preventing excessive corrosion.

In 1955, new limits were imposed on the permissible atmospheric emissions of iodine 131 (I-131). In response, backup radioiodine removal facilities were added to the PUREX Plant in 1956 (see Figure 8). The small, concrete 293-A structure, placed above ground between the PUREX canyon building and the 291-A Stack, provided increased nitric acid and iodine recovery. It enclosed two acid absorber columns with bubble-cap trays and two tanks on two levels (at and below grade). The second acid absorber column was operated with a sodium hydroxide scrub to remove radioiodine. However, the acid content reacted rapidly with the caustic, resulting in low iodine recovery and high waste volumes. When these reactions were observed, the operation of the column was changed to recover additional nitric acid. Overall radioiodine removal remained about the same.[18]

During the early years of PUREX operations, the most important changes were the placement of additional in-line, alpha and gamma monitoring instruments (1956) and new plutonium concentration equipment in L-Cell. (The former L-Cell unit had contained a stripper for organic steam stripping prior to concentration and two concentrators.) The new equipment contained a stripper and a single, more efficient concentrator. In 1957, the HA column, the first decontamination column in the primary decontamination cycle, was replaced. A series of column modifications reversed the organic and aqueous continuous operations, thus placing the interfaces at the opposite end from the product. The new

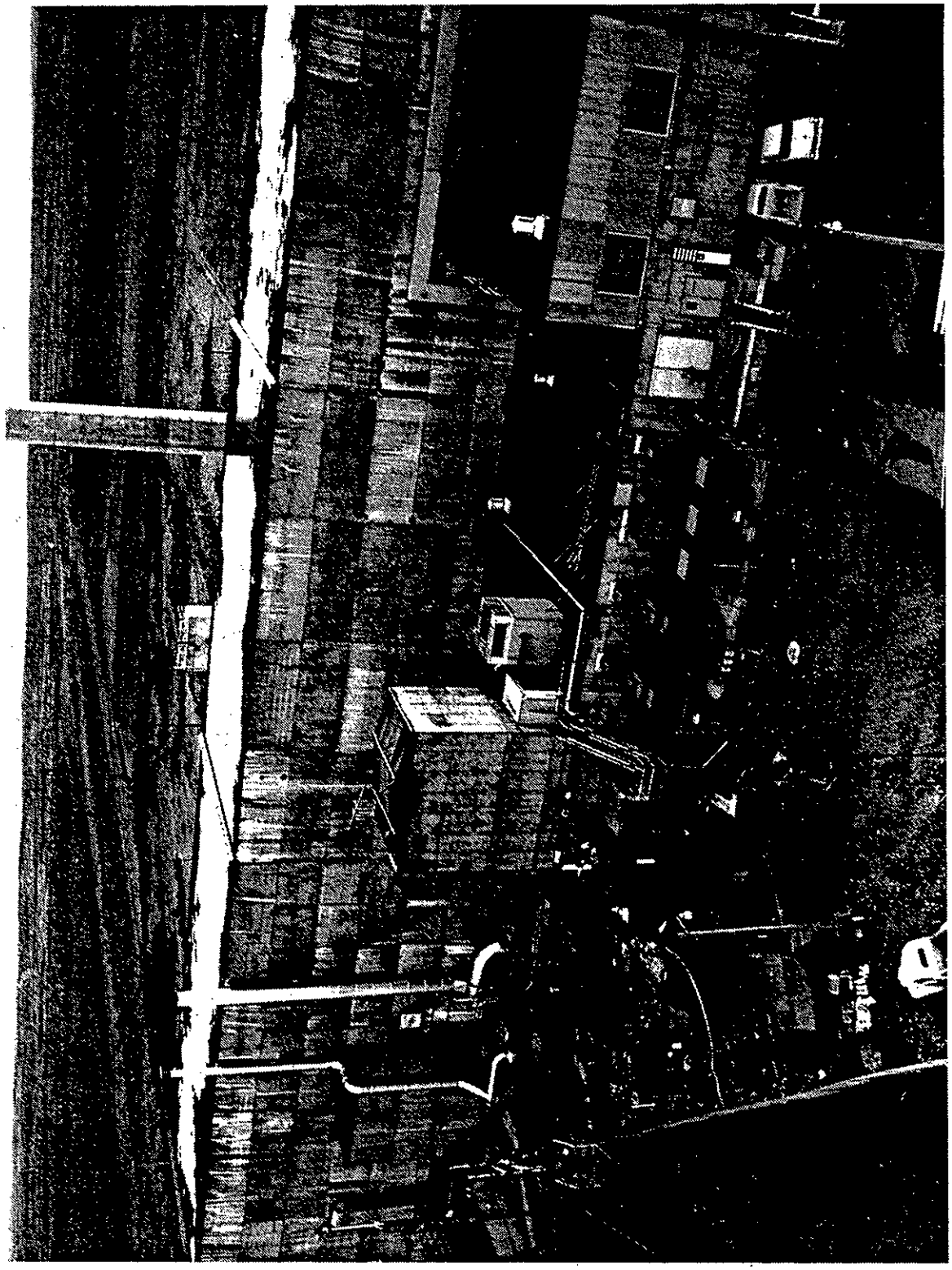

Figure 8. Additional PUREX offgas treatment facilities under construction, October 1956.

configuration minimized the carryover of interfacial materials with high fission product content, thus considerably improving the decontamination factors. That same year, additional isolated ventilation systems and shielding partitions were installed at the west end of the P&O gallery. These contained the effects of a significant contamination event that had occurred in February 1956. (Additional information on this event is in Section 1.7, Contamination Events.)

A very key, early 1958 project was the conversion of the PUREX extraction and stripping facilities (H-, J-, and K-Cells), and the waste concentration facilities (F-Cell) from a three- to a two-cycle system. Anion exchange, a new method of removing contaminants from the plutonium product when it was in the +4 valent state combined with strong nitric acid, was substituted for the older concentration methods to achieve better decontamination factors. The conversion changed the function of the HC stripping column to a scrub column associated with the primary decontamination column (HA). Column 1A was removed, and a backcycle waste system was established which used the former E-H4 concentrator to evaporate aqueous waste streams from the second uranium and plutonium cycles. These and other waste streams were combined and sent to the HA column to be recycled through the entire plant process. Operation of the E-F11 concentrator also was discontinued. The new system eliminated time and equipment in the processing cycle, thus freeing space in J-Cell for the eventual placement of continuous Np-237 recovery equipment (1962).[19]

The PUREX plutonium anion exchange unit was replaced with a larger unit (1959), and a series of "reliability improvements" were made (1960 to 1961). The changes included modifications to the following: solvent extraction columns; P&O gallery, solvent recovery, and storage piping; aqueous make-up and unloading areas; 211-A Chemical Tank Farm; canyon tanks; and instrument and electrical system retrofits. Together, the expansions allowed PUREX to operate at a 3.6 capacity factor. Also in 1961, secondary confinement systems (backup filters) were added to each dissolver offgas system. At almost the same time, the steel 212-A Fission Product Load-Out facility was added onto the south side of PUREX to provide a place for the transfer of liquid wastes from in-canyon vessels to suitable shipping containers. Successful pilot studies examined using table sugar rather than formaldehyde to de-nitrate PUREX acid wastes.

In 1962, new facilities enabled the continuous recovery of Np-237 (also known as "palm" or "birch"). The neptunium recovery cycle (J-Cell Package) consisted of a compact assembly of feed and product tanks, two solvent extraction pulse columns, assorted jumpers, and structural supports. The package was installed in the J2 cell position that previously had held the 1A column. Ion exchange facilities for the final purification of the Np-237 were installed in the Q-Cell area of the plant.[20]

Additional equipment changes in the following year enabled the processing of E Metal (fuel elements enriched to 1.75 percent uranium 235 [U-235] by weight), also known as Eisenhower or Ike, N Reactor fuel, mixed oxide fuels from HW's PRTR, and other fuels. The Zirflex process by which N Reactor fuels were declad involved the dissolution of the Zircaloy-2 coating with a mixture of ammonium fluoride and ammonium nitrate (AFAN),

subsequent metathesis with potassium hydroxide, centrifugation of the coating waste for product removal, and finally dissolution of the fuel elements themselves in nitric acid. PRTR fuels were processed only occasionally because of the small volume. Core dissolving of the mixed oxide fuels involved the use of a highly corrosive mixture of nitric acid and hydrofluoric acid, followed by the blending of the dissolver solution with recycled uranium to achieve criticality control. In 1964, flow through the original fiberglass ventilation filter for the main PUREX stack was restricted because of a buildup of solids in the filter. Occasional exposure of these solids, which contained a significant quantity of ammonium nitrate, to air with high moisture content resulted in the formation of a crust on the filter top. A steam regeneration process partially dissolved the ammonium nitrate in the crust and temporarily restored normal filter flow. However, the regenerated filter plugged again four months later. As a result, a second, deep-bed fiberglass filter was added to the plant. Subsequently, a project was undertaken to separate and reroute the ammonia-bearing effluents to another collection system (1978).

In 1965, a tantalum-lined concentrator and a titanium receiver tank were installed in N-Cell for U-233 processing. The new smaller equipment was designed to meet the more stringent criticality safety requirements for U-233 and to reduce metallic impurity contamination of the product by vessel corrosion. Also, for reasons of criticality control, the L-11 vacuum tank in the product handling and removal (PR) room was replaced at the same time. Later that year, the installation of larger columns, additional jumpers, miscellaneous controls, instrumentation, pumps, valves, and other equipment allowed PUREX to expand to a 4.0 production capacity factor (about 33 MTU per day).[21]

In February 1966, the commercial diluent that had been used with 30 percent volume tributyl phosphate to make up the basic PUREX solvent extractant was replaced with normal paraffin hydrocarbon (NPH) diluent. Because NPH was subject to less radiolytic and chemical degradation, fewer fission products accumulated in the solvent, and better decontamination factors were achieved in the final product.[22] Later, annular dissolvers (those having a smaller vessel inside a larger vessel) were placed in PUREX to allow the processing of increasingly enriched N Reactor fuel. Also, a "co-product insert" was placed in PUREX to process the driver fuel elements needed to facilitate irradiation of the lithium-aluminate targets that produced tritium in N Reactor. By 1967, 125 Metal (enriched uranium containing 1.25 percent U-235 by weight) was being processed through PUREX, always in conjunction with fuels of lower enrichment levels. The following year 210 Metal (driver material containing 2.10 percent U-235 by weight) was also being sent through the plant, again in conjunction with less reactive materials.[23]

Additional PUREX projects that were completed before the 1972 major shutdown included the installation of tank P6, a storage tank in the 203-A facility for concentrating UNH coming from the UO_3 Plant; the construction of the D5 cave, a shielded cell in the sample gallery; and additional fire protection facilities for the hot shop, regulated shop, and service and storage galleries. Importantly, a third plutonium solvent extraction cycle was placed in L-Cell, adding sophistication and efficiency to the final plutonium purification step.

A high-level cell was placed in the PUREX laboratory (1964), and further laboratory modifications were made (1967).[24] (Waste handling changes, which also took place during these years, are discussed in Section 1.6, Waste Management.)

During the 11-year shutdown, many projects and equipment modifications took place. One of the most important projects modified the M- and N-2 Cells (in the PUREX canyon) into maintenance and processing facilities so that plutonium nitrate could be converted to plutonium oxide as the final plant product. Four multi-cylinder "bank-tanks" were added to a partitioned section of M-Cell; and a complete precipitation, calcination, blending, and oxide product can loadout process was installed in N-Cell and in the adjacent pipe chase. The PUREX PR room also was modified to accommodate shipment of the oxide product to PFP, and additional changes were made to the loadout configuration of Q-Cell (the facility built in 1962 for continuous neptunium purification).[25] Additionally, upgrades were made to the primary and PR room stack systems, the treatment systems for nitrous oxide and ammonia bearing fumes, plant fire protection systems, facility security, the P&O gallery, the hot shop, the UNH storage area, the heating and ventilation air systems, the backup generators, the White Room filtration facilities, and the lag storage tank. Structural safeguards were implemented to ensure the integrity of PUREX in case of a seismic event.[26] For information on projects and modifications concerning waste handling facilities see Section 1.6, Waste Management.

Since the restarting of PUREX in 1983, few major changes have been undertaken. Significant among these, however, were improvements to canyon exhaust and auxiliary filter systems and upgrades to the in-plant equipment decontamination and repair facility, the fire protection systems, the perimeter intrusion detection system, the electrical system, the backup generators, and miscellaneous structures. Additionally, various process changes were undertaken to remediate old problems of foaming, burping, and drops in specific gravity in the dissolvers; and to address thorium, phosphorus, and titanium impurities in the oxide product. In 1985, prototype testing was undertaken in the 300 Area to modify the head-end of the PUREX Plant so cladding on N Reactor fuel elements could be sheared off rather than chemically dissolved. The physical shearing method was desired because the residues of chemically dissolved cladding complexed high-level tank waste. However, such modifications were delayed in the actual PUREX Plant until after the final cold standby order came for N Reactor (February 1988). At that time, the shearing concept was revived, and plans were initiated to modify the head-end of the PUREX Plant to permit the shearing of stainless-steel clad fuel from the Fast Flux Test Facility (FFTF). The bare fuel cores then would be dissolved in C-Cell. However, this project was never undertaken because of other problems at PUREX and uncertainties about the future of the FFTF.[27]

1.5 EQUIPMENT DECONTAMINATION AND REUSE

Soon after the PUREX Plant had demonstrated its production capacity in 1956, the decision was made to close the 221-T Building as a processing facility and to convert it to an equipment decontamination and repair facility. Because the latter operations quickly became

a success at T Plant, the 2706-T decontamination annex was added. Many pieces of PUREX equipment were decontaminated at these facilities in the ensuing years. Sometimes equipment was decontaminated just to make it safe for handling during burial; at other times equipment was completely refurbished and returned to PUREX for first-line service or storage as a ready spare. Decontaminated apparatus ranged from cell wrenches, gaskets, or saws worth about $1,000 each to dissolver towers worth approximately $235,000 each. The financial savings were considerable even after subtracting the repair costs and using a discounted value for decontaminated and returned equipment. For example, during 1969, PUREX equipment worth approximately $300,000 was decontaminated at the 221-T and 2706-T facilities at a cost of approximately $70,000. This resulted in a direct savings to PUREX of $230,000 (not counting additional procurement costs for new equipment).

The reduction in radiation levels was considerable as well. For example, PUREX jumpers reading 10 rads at 1 foot, or impact wrenches reading 8 rads at 1 foot, could be decontaminated to a level of 20 millirads (mr) at 6 inches. Centrifuges and agitators reading 12 rads per hour at 2 inches could be brought down to levels of 170 to 200 mr per hour at the same distance. Typical decontamination methods and materials included high pressure hosings, steam cleaning with various acids and commercial cleansers, filling and then flushing vessels with similar solutions, air spargings, and prolonged soakings in chemical mixtures.[28] Figure 9 shows the pulse generator from J-Cell undergoing decontamination.

1.6 WASTE MANAGEMENT

Because of the large variation in the strengths and radionuclide contents of wastes generated by the PUREX Plant, waste handling facilities were divided into those designated for high- and low-level liquid wastes and those for solid wastes. The original, low-level waste handling facilities consisted of seven 216 facilities or "cribs."[29] Each crib was made of rock and was filled to a minimum depth of 7 feet with 3 inches of aggregate stacked beneath the distribution header. Unlike the oldest Hanford cribs, there were no timbers or void spaces in the PUREX cribs.

Organic waste from solvent treatment operations and the waste disposal cell in the 202-A Building went to the small (20-foot^2) 216-A-2 crib via batch transfer through the 241-A-151 diversion box. The crib was located approximately 100 feet south of the main canyon building. Laboratory and cell drain waste went to another small crib, 216-A-4, located about 100 feet south of the 291-A PUREX Stack. Drainage from the 203-A facility (for storage of UNH recovered out of the PUREX process) and from the acid fractionator operating in the 276-U cell went to the 216-A-3 crib. This 30-foot by 200-foot facility was located about 200 feet north of the 203-A structure. Process condensate went to the 35-foot^2 216-A-5 crib located south of the west end of the PUREX canyon building. Steam condensate went to the large (100-foot^2) 216-A-6 crib located 1,000 feet east of the 202-A structure. The tiny (10-foot^2) 216-A-7 crib, located 200 feet east of the 241-A-152 diversion box, received the drainage from that facility. The 241-A-152 diversion box was the device that routed high-level waste to underground Tank Farms A and C. Condenser cooling water

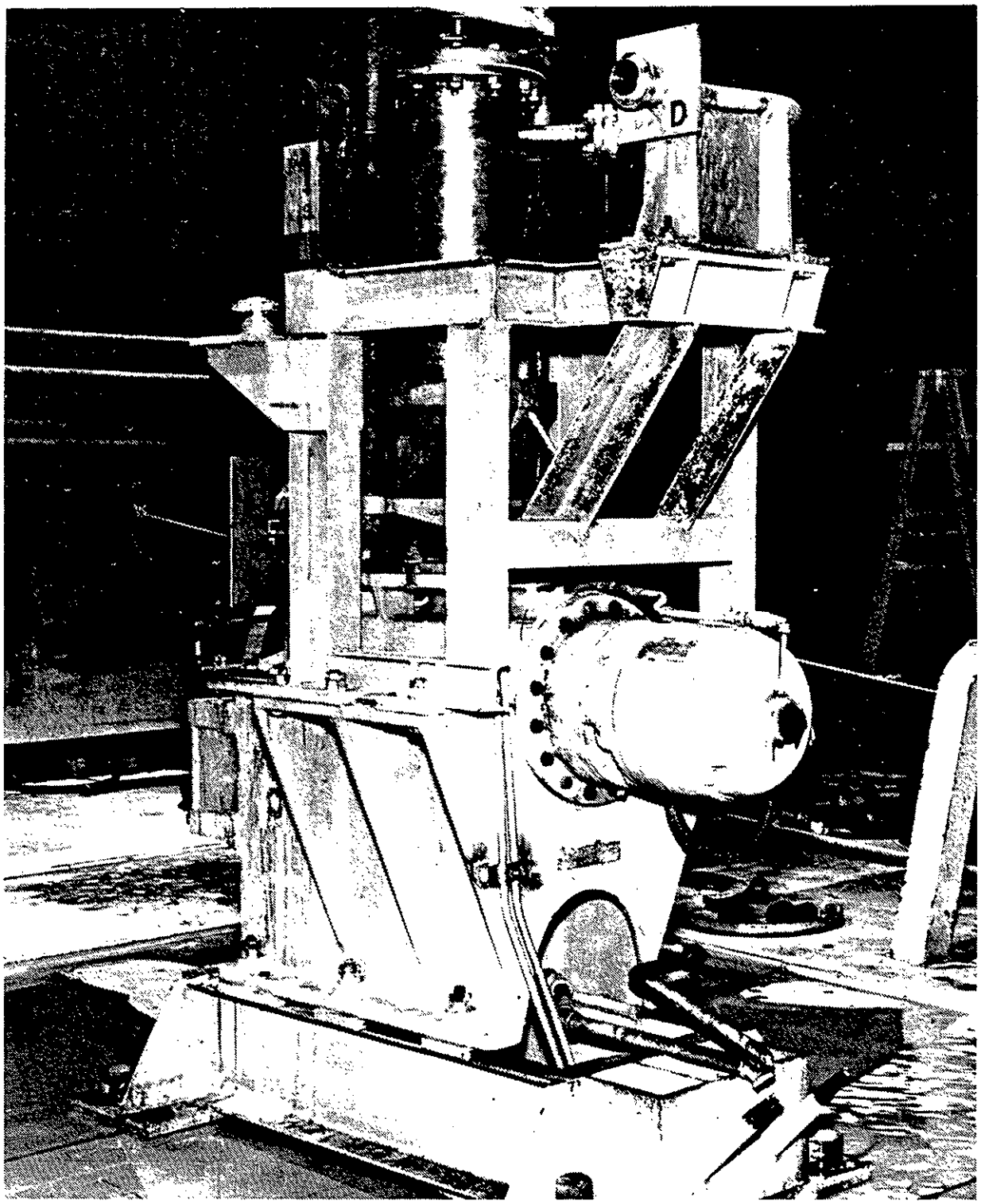

Figure 9. Pulse generator from J-Cell, PUREX Building, undergoing decontamination in T Plant, April 1961.

and condensate from the 241-A Tank Farm were disposed to the 80-foot^2 216-A-8 crib, located 700 feet northeast of that high-level waste facility.[30]

Raw water employed for cooling and general utility purposes was considered to be nonradioactive during the first several years of PUREX operations. It was routed out through the south wall of the canyon and through a collection header that sloped from west to east, parallel to the canyon building. Some of this water was collected in the 201-A steel storage tank for use in cooling the contact condensers of the 241-A Tank Farm to the northeast (see Figure 10). The majority of the water was piped through a vitrified clay line to the cooling water swamp southeast of the main PUREX Plant.

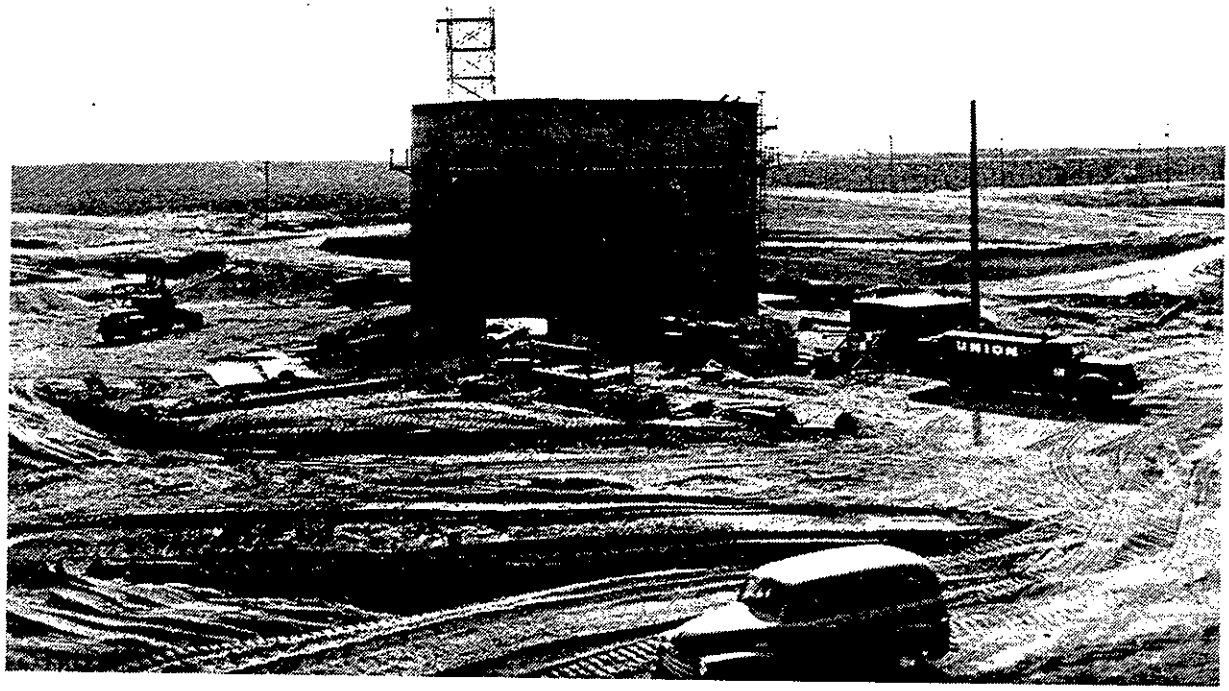

Figure 10. Cooling water tank for 241-A Tank Farm, under construction, March 1954.

The disposal of low-level liquid wastes from PUREX was complicated by a situation that had developed previous to startup. Mounds had developed in the groundwater beneath the 200 East and 200 West Areas as the result of cooling water discharges from B, T, REDOX, and U Plant, (with PFP as a lesser contributor to the 200 West Area mound). The mounds shifted natural hydraulic gradients and drainage patterns, sometimes intersecting with underground plumes of contaminated liquids and routing them towards areas of high permeability and transmissibility. Also, the pressure and weight of the mounds themselves increased the hydrostatic head, forcing all nearby or affected underground liquids to travel faster toward the Columbia River. By October 1955, three years after the closure of B Plant, the groundwater mound beneath the 200 East Area had dropped 10 feet from its peak of 405 feet. The main area of concern to Hanford hydrologists thus became the behavior of the huge mound beneath the 200 West Area.

The startup of the PUREX Plant in January 1956 changed this situation considerably. The facility's swamp discharges of 6.5 million gallons per day were more than three times that of any previous rate discharged to the area. In just over a year, the groundwater mound beneath the 200 East Area was 418 feet, higher than at any previous time. It migrated southeast toward highly transmissible substrata; and it also began to seep westward, filling the underground "valley" between 200 East and 200 West Area mounds. Furthermore, wastes from the 216-A-8 crib reached groundwater for the first time between December 1 to 12, 1956, causing "significant increases in beta-emitter activity in groundwater samples." During the next three months, activity in groundwater samples beneath this crib increased by 100,000 times.[31]

As a result, a project was undertaken to reroute PUREX cooling water. A widely accepted HW theory at the time was that the movement of underground water could be controlled by the strategic and deliberate formation of groundwater mounds that would act as hydrologic dams. Such mounds would counterbalance each other and enable scientists to divert underground water through desirable pathways by carefully controlling and rotating discharges to certain swamps. In the case of PUREX, it was believed that underground pathways of cooling water could be managed by distributing the majority of the water to the Gable Mountain Swamp, about 20 percent to the initial swamp, and about 10 percent to B Swamp (B Pond), located north, southeast, and east of the plant respectively.

By the time the diversion project was completed, the enormous PUREX production capacity had raised the average daily cooling water discharges to 9.8 million gallons per day. Wastes from the PUREX cribs continued to reach groundwater, intersect with the mound southeast of the plant, and travel into permeable zones. Construction was begun on interim waste crib 216-A-27 in late 1957. At year's end, an estimated 7,800 curies of activity had gone to the PUREX cribs; 75 percent of this total went to 216-A-8. Five months later, gross beta analysis of wells sampling the groundwater beneath this crib demonstrated readings as high as 6.0 X 10E-4 microcuries per cubic centimeter. (This reading was higher than the then-recommended limit for aqueous wastes discharged to the ground at HW by more than a

factor of 10.) At that time, the crib was taken out of service and replaced with crib 216-A-24, which received the condensates from PUREX tank farms 241-A and 241-AX until 1966.[32]

The effects of PUREX discharges on the groundwater beneath HW ranged from areas as far away as the pre-Hanford townsite to points of intersection with the Columbia River near the 300 Area. The effects continued and grew throughout the production leaps of the late 1950's and 1960's. Many changes and solutions were tried to mitigate the effects of operations on the groundwater. In late 1959, surface condensers were installed in the 241-A Tank Farm. This reduced the amount of tank farm condensates going to cribs to about 5 percent of their previous volumes. During 1960-1961, the 216-A-6 crib was replaced with the 216-A-10 crib because production increases had rendered the infiltration capacity of the former crib inadequate. Crib 216-A-2 also was replaced in 1961. Early that year, a Micro Pilot Plant was operating to conduct experiments in decontaminating PUREX tank farm condensates. Both natural and synthetic ion exchange resins were tried. Initial results showed that clinoptilolite was the most promising. A large percentage of the cesium 137 (Cs-137), a key isotope of concern in tank farm condensates, was absorbed by the natural zeolite. Percentages of strontium 90 (Sr-90), cerium 141-144 (Ce-141-144), Ru 103/106, and Zr/Nb-95 likewise were reduced.

The 1962 conversion from a two- to a single-cycle acid recovery system greatly increased activity levels in the waste going to the 216-A-10 crib. Chemical and process changes were instituted to try and reduce this activity, especially the radioruthenium content. However, in 1966, the 216-A-10 crib and the 216-A-10 steam condensate crib had to be replaced by the 216-A-38 crib and the 216-A-30 crib respectively. At the same time, the 216-A-36B ammonia scrubber waste crib was constructed, the 216-A-8 crib was reactivated, and the 216-A-24 crib was taken out-of-service. The period 1960 to 1965 was marked by the construction of other cribs and the discharge of the largest amounts of low-level waste at HW prior to the PUREX shutdown in 1972. Of the slightly more than 32 billion gallons released, 64 percent came from PUREX.[33]

During and following the 1972 to 1983 PUREX shutdown, many additional changes were made to reroute, treat, and minimize low-level waste discharges. Among the early upgrades was the installation of a tank farm condensate recycling system. The 216-A-8 crib was taken out of service for the second time in 1976, but it was reactivated for the 1983 startup. Leaking parts of the steam condensate line were bypassed and replaced along with pumping additions. Just prior to startup, monitors and diversion flow systems were installed for the chemical sewer; and improved sampling, monitoring, and flow totalizing capabilities were installed for the process waste condensate discharges. The 216-A-10 crib was designated to receive 202-A Building process condensate, and the 216-A-38 crib was to be used as a backup. The 216-A-37-1 crib was constructed in 1977 to receive process condensate from the 242-A Evaporator. Additionally, the 216-B-3 pond was expanded to receive increased PUREX cooling water discharges, and several effluent cribs were

upgraded during the 1980's. Pursuant to agreements made in the 1989 *Hanford Federal Facility Agreement and Consent Order* (Tri-Party Agreement), all untreated, low-level liquid discharges from the PUREX facility and from other Hanford plants must cease in 1995.[34]

The management of high-level wastes at the PUREX Plant originally was based on the six tanks of the 241-A Tank Farm, located approximately 1,500 feet northeast of the midpoint of the 202-A Building. Each tank had a diameter of 75 feet and a capacity of one million gallons, and each was constructed of reinforced concrete with mild steel liners on the bottoms and sides (see Figure 11). Although many coating materials were tried during the development stage of the tank liners, all became brittle, shrank, developed blisters, or melted and became soluble in the presence of highly corrosive, simulated PUREX high-level waste. Desiring a full field test, the builders of the PUREX tanks placed corrosion test inserts of various materials (mild steel, carbon steel, and two other steel alloys) into tank 241-A-101 to observe the differences in the materials' resistance over time. They already had learned some key lessons from the first experiences with self-boiling wastes at the REDOX Plant. In 1952 and 1953, radioactive wastes, which had boiled from the self-generated heat of the decay reactions taking place in them, had ruptured the cooling coils of the 241-S tanks and spread contamination to nearby areas. HW scientists called the phenomenon "bumping" because heat built up in the waste layers and then was released in periodic bursts. Techniques for refluxing condensate between various tanks (to control temperatures) and for mixing waste layers of separate temperatures within each tank had not been built into the 241-S Tank Farm.[35]

The builders of the PUREX tanks constructed a 39-inch diameter by 35-feet deep tank within the hot semi-works (C Plant) to represent the core of one of the 75-feet diameter 241-A tanks. Wastes placed in the tank had a fission product heat generation rate of approximately 9,000 British thermal units (Btu) per hour. With continued boiling, the waste quickly separated into a solution layer (above) and a hotter sludge layer (below). Within the solution layer, successively higher temperatures were demonstrated at successively lower levels in the solution. When the temperature differential within the solution layers reached (20 to 25 °F), the tank "bumped." Steam was released and, in effect, the layers of waste solution were mixed by the forceful emission and temperatures within the layers equalized.[36]

To avoid bumping and to provide for the proper control of temperatures, each of the 241-A tanks was equipped with four air-lift circulators (mixers) that utilized forced air to lift and stir the wastes. Two of the circulators were 17.5 feet long, and two were 8.5 feet long. The other key feature designed for managing self-boiling tank wastes was the vapor system, through which all tanks were interconnected via a 24-inch vapor header. Vapors generated by self-boiling wastes flowed out the vapor header, through a cyclone separator where they contacted the condensers and a vent de-entrainer, and then escaped through a local stack. Additional cooling water could be supplied to the condensers from tank 241-A-201 and from other 241-A tanks (except for the last tank, 241-A-106). Each of the 241-A-tanks had a 6-inch line that connected it to the next tank in the series. HW scientists hoped these

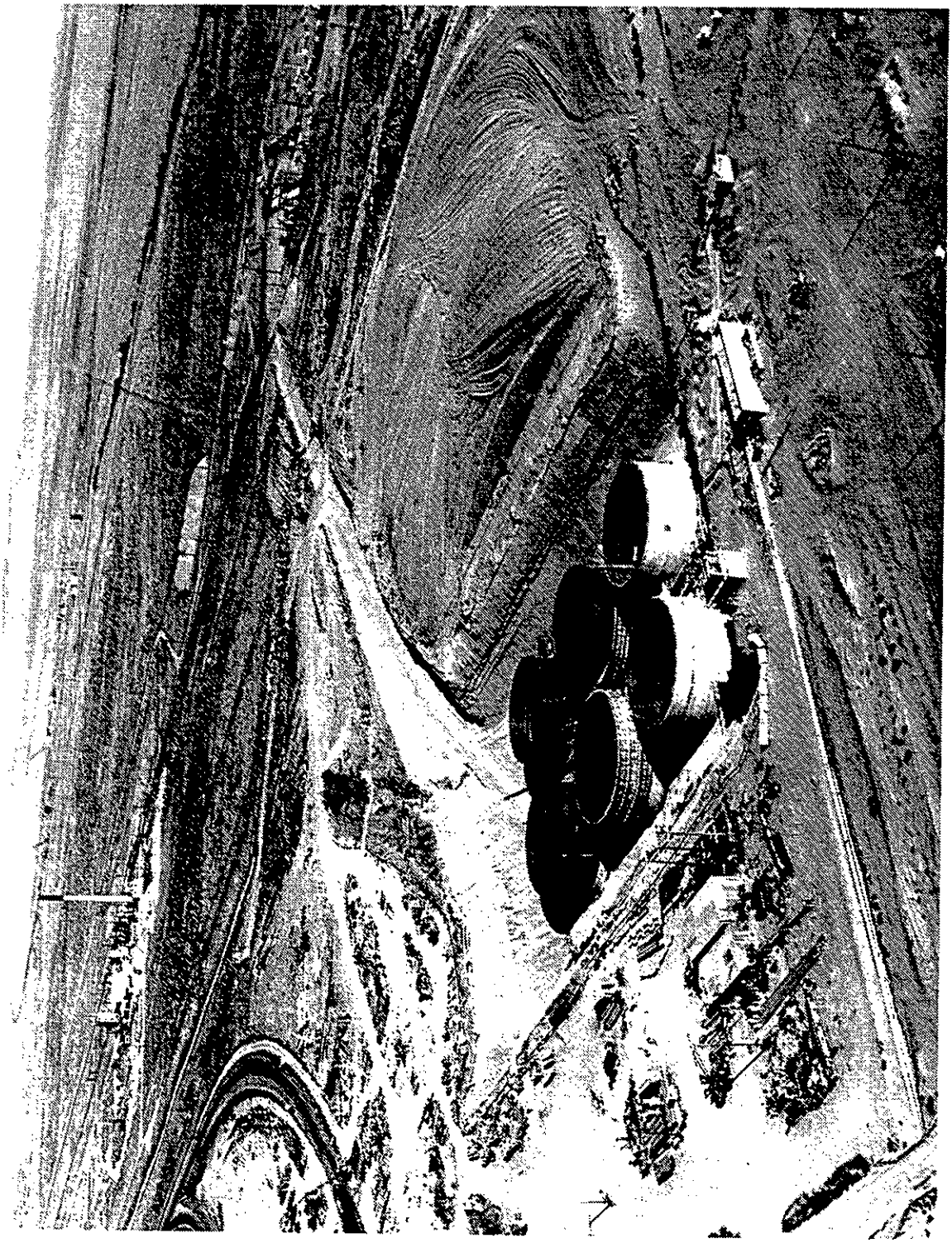

Figure 11. 241-A Tank Farm, for the storage of high-level wastes from the PUREX Plant, under construction, July 1954.

features could regulate the constantly changing temperatures within the high-level waste tanks.[37]

While the management of in-tank temperatures progressed well during the early years, the ubiquitous HW problem of shortfalls in high-level waste tank space soon affected PUREX. By mid-1956, the need for a new tank farm for the giant separations facility was predicted for as early as 1958. Coating removal wastes from PUREX, which averaged 284 gallons per ton of production, were sent to the World War II 241-C Tank Farm. Higher heat wastes from the extraction and decontamination steps of the PUREX process were routed to the newer 241-A Tank Farm. Finding a way to bind up or decontaminate the coating wastes so they would be eligible for disposal in soil or in underground caverns became a high priority during 1957 and 1958. However, although studies in the gelation of such wastes with chemical additives were initially promising, it was demonstrated by late 1958 that gels were unstable and did not provide good radionuclide retention over time.[38]

Studies in managing high-level liquid wastes at HW then broadened into several pathways. Additional leak-detection systems for self boiling tanks were developed; various tank construction materials, such as titanium, were explored for corrosion resistance and for other factors; and the question of the permanent ultimate disposal (as opposed to in-tank storage) of high-level wastes was initiated. The 1958 Anderson Report advocated the solidification of such wastes into glass-like, solid forms; others at HW favored partitioning the wastes into various isotopes for space power, medical, and other uses. The proliferation of studies and experimentation is too extensive to be described here. However, much of the PUREX high-level waste was partitioned, primarily for Sr-90 and Cs-137 recovery, for space power and other applications use. Large partitioning campaigns were conducted in B Plant from 1968 to 1978; and smaller campaigns to divide and utilize portions of PUREX wastes and used acids were conducted in C Plant during the 1960's. In the meantime, the rapidly expanding PUREX production capacity required more storage tanks. During 1963 and 1964, the AX Tank Farm, which comprised the last four single-shell, high-level waste tanks built at HW, was constructed. for the PUREX Plant.[39] The first two double-shell tanks (DSTs) were built for PUREX in the 241-AY Tank Farm from 1968 to 1970 and 15 more DSTs were constructed for PUREX during the 1972 to 1983 shutdown period in the 241-AZ, -AW, and -AN Tank Farms. Finally between 1983 and 1988, eight more DSTs were built for the giant plant in the 241-AP Tank Farm.[40]

The original PUREX Plant incorporated a unique feature for disposing of large pieces of radioactive solid waste, such as failed or outworn equipment. A 500-foot extension running southward was built onto the single-track rail tunnel to bring irradiated slugs to the east end of the PUREX building. Expendable flat rail cars were used to push intensely radioactive equipment up the tunnel for disposition. In 1964, a 1,500-foot tunnel was constructed after the first one had become full (see Figures 12 and 13).[41] Immense quantities of smaller pieces of solid radioactive wastes, including tools, special work permit (SWP) clothing, wipes, small equipment parts, etc., that had been generated by PUREX operations over the years were shipped to the burial grounds in standard 200 Areas burial boxes. Like other Hanford plants, PUREX responded to the 1970 joint contractor task force

Figure 12. Original contaminated equipment disposal tunnel, PUREX Plant, under construction, October 1955.

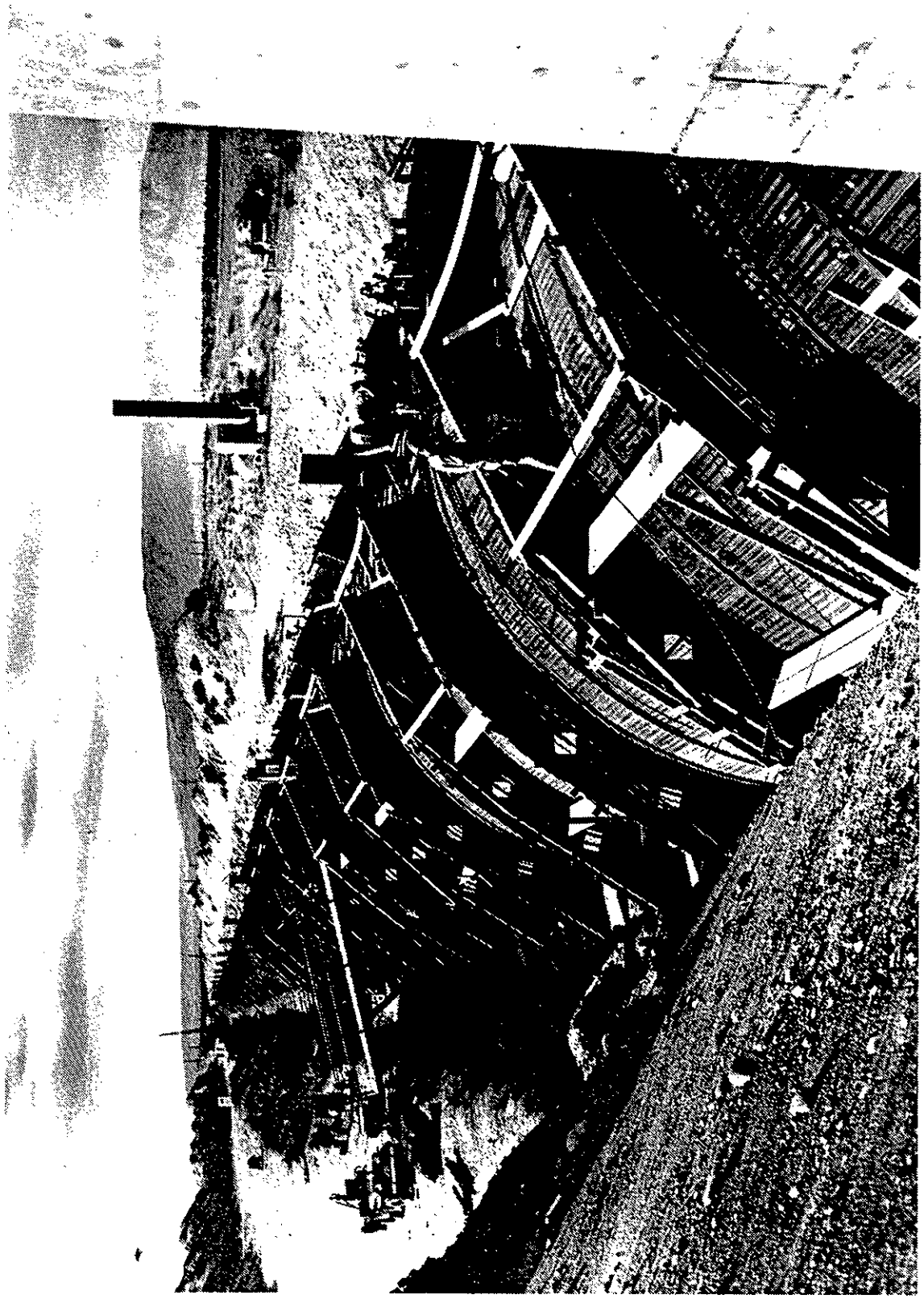

Figure 13. Second contaminated equipment disposal tunnel, PUREX Plant, under construction, August 1964.

report and directive that solid wastes be segregated by specific contents. Wastes contaminated with certain chemicals, transuranic elements, polychlorinated biphenyls, combustibles, and other contents were separated and labeled carefully. After that time, liquid contents of solid wastes (such as pumps) were drained stringently before burial, and strict packaging and other requirements were followed. Over the years much of the radioactive solid waste generated by PUREX has been placed in burial grounds near B Plant and in the northwest quadrant of the 200 West Area.[42]

1.7 CONTAMINATION EVENTS

While many incidents have occurred involving the loss of control of radioactive materials at PUREX throughout its history, few have been serious.

The first significant event occurred in the second month of operations (February 1956), when an instrument line leading to the L-6 tank (part of the final plutonium decontamination cycle) released about 20 gallons of plutonium-bearing solution into the west end of the P&O gallery. The tank had been subject to repeated plugging, and the incident occurred when operators attempted to open valves and lines. Liquid contamination was spread through the chemical sewer drain, the canyon lobby, the P&O gallery, and into R-Cell. Airborne contamination was drawn by exhaust fans throughout the P&O gallery, other 202-A Building locations, and into the environment. So many coats of sealant paint were applied to the immediate area of the spill in the west end of the P&O gallery that the area became known as the White Room. In 1957, a shielding wall and a separate ventilation system were installed to isolate the White Room, and the area remains a radiation zone today.[43]

The second serious radiation event occurred two years later (February 1958) when the bottom portion of the silver reactor filter in A-Cell exploded. The huge filters, 8-foot-thick beds of packing material coated with silver nitrate, absorbed and reacted with the radioiodine in the dissolver offgases. Once saturated, they regenerated with an ammonium hydroxide flush. It was presumed that the 1958 uncontrolled reaction (explosion) occurred when unstable products formed in the ammonia-silver salts mixture. Although there was little detectable spread of contamination to other portions of the PUREX building and none to the environment, cleanup and repair were difficult because the filter was located in a heavily shielded, inaccessible area.[44]

The third significant incident (June 1964) occurred when first-cycle acid waste from the PUREX Plant accidentally was discharged to the cooling water swamp southeast of the 202-A Building. An estimated 10,000 curies (Ci) of mixed fission products (primarily Zr/Nb-95, Ce-141/144, and Ru-103/106) were released and settled in the mud and algae. Corrective action included killing the algae, covering the contaminated ditch area that flowed to the swamp with backfill, and digging partial new ditch/swamp areas.[45]

The fourth serious incident (January 1965) occurred when a sudden release of steam from high-level waste storage tank 241-A-105 caused contamination of tank instrumentation and a nearby construction ditch. The contamination cleaned up readily, but probes showed that a large area of the steel tank liner bottom had bulged upward as much as 8.5 feet, creating a void capable of holding up to 85,000 gallons. It was surmised that a leak in the liner had caused water to accumulate, and that the temperature differential between the water and the hot tank sludge had caused the buckling. The tank continued to hold wastes until April 1967, but no new wastes were added and recirculator flow and surveillance were increased. In 1967, when liquid level fluctuations began to occur, the tank was taken out of service. All liquids were pumped out, and as much sludge as possible was removed in sluicing campaigns in the early 1970's. The remaining sludge still produced enough heat on a regular basis that cooling water needed to be sprinkled into the tank for several years until it was fully stabilized in 1978. Since that time no wastes have been added to the tank.[46]

The fifth serious contamination incident (November 1967) occurred when high-level contamination escaped from a shielded sample carrier and spread onto the clothing of an operator. Because it was not detected for 44 minutes, low-level contamination was spread by the employee into nonradiation zones. The incident was considered serious because of operating procedure violations rather than to the extent of contamination spread.[47] Another incident of operator contamination, accompanied by the spread of low-level contamination in areas of the 202-A Building, took place in June 1968. Radioactive liquid was sprayed onto an operator's coveralls during the removal of a broken pipette tip from the F-26 sample riser. This incident did not involve the violation of work procedures.[48]

In April 1982, a dilute nitric acid solution was misrouted during a routine transfer, and a 2,500-gallon spill occurred. Radioactivity was spread to various areas normally occupied by operations personnel. However, because the plant was not operating, the personnel exposures were limited. Loss of configuration control and poor tracking were blamed for the incident.[49]

In February 1986, a leak occurred at a pipe fitting downstream from a plutonium nitrate product solution storage tank, and solution dripped to the floor of the containment glovebox. A temporary transfer of solution to another tank was made, without entering the change into the formal documentation and configuration control procedures. Although a blank was installed in the piping to prevent inadvertent and subsequent transfer of the solution to a nonfavorable environment and the leak was repaired, the episode revealed several violations in proper documentation and control.[50]

2.0 UO_3 PLANT

The original Uranium Trioxide (UO_3) Plant was located in a World War II facility known as the 224-U Bulk Reduction Building (see Figures 14 and 15). The three-story, frame and concrete block structure of just under 12,000 square feet had been constructed to house a part of the plutonium separation and concentration process used in the 1940's. However, process improvements as early as 1945 demonstrated that the 224-U Building was not needed. Rather than contaminate it with plutonium, Hanford's management decided to use the building as a training facility until another use was found.

The basic UO_3 process, calcining, consisted of concentrating and then heating liquid UNH until it converted to a stable, orange-yellow powder. The nitric acid in the UNH solution could be recovered in the same process. The UO_3 powder was the base material needed for the manufacture of uranium hexafluoride (UF6), the primary feed material for the United States' gaseous diffusion plants. Because the largest of these plants was located in Ohio and Tennessee, it was considered safer to ship the material across the country in

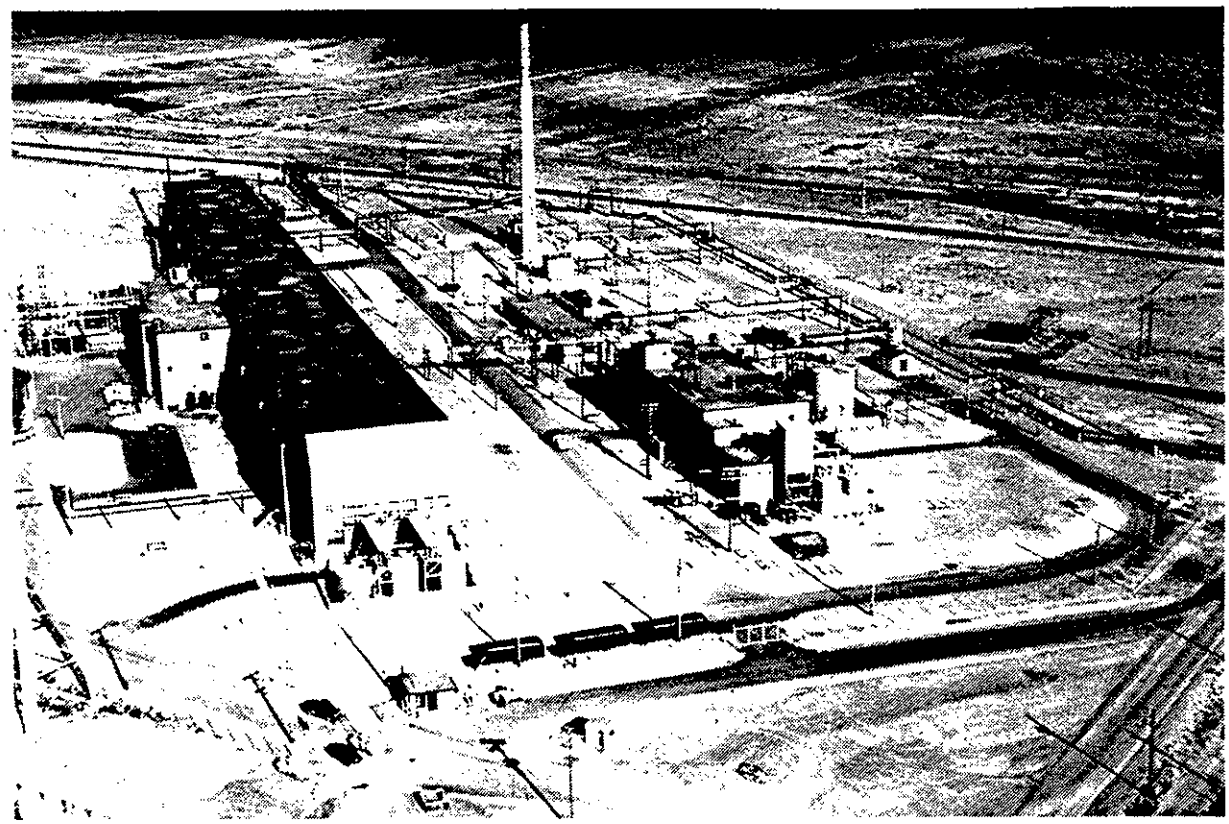

Figure 14. Original 224-U Building (upper center) operating as the UO_3 Plant, February 1953. The larger building is the 221-U canyon, and the small, one-story building is the 222-U structure.

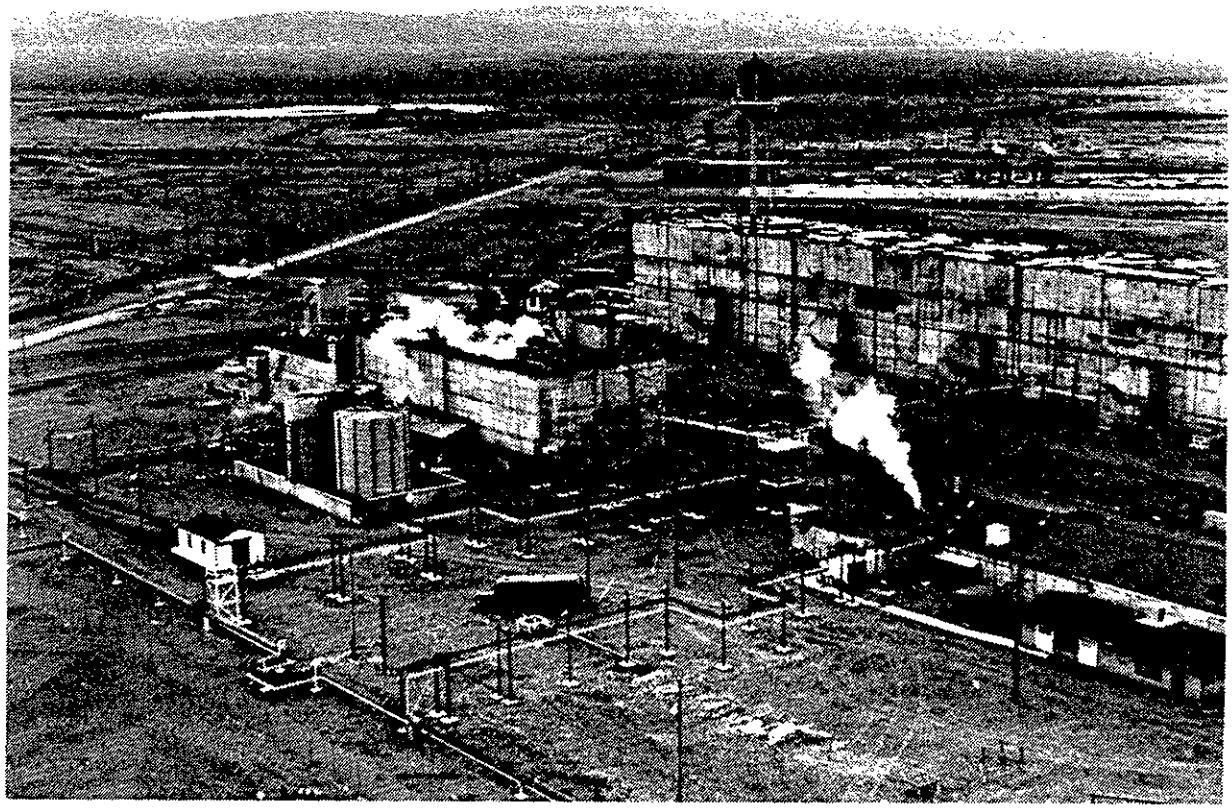

Figure 15. UO_3 Plant (center right), shown with the larger 221-U Plant and the one-story 222-U Building, February 1953.

powder rather than in liquid form. It was not possible to operate the UO_3 process at Hanford until a feed supply of UNH became available (1952) with the startup of the REDOX Plant and the Metal Recovery Plant (U Plant, also known as the TBP Plant during the Metal Recovery Mission). The earlier separations process at B and T Plants had lost the incoming uranium as waste. But the newer solvent extraction processes at REDOX and U Plant recovered the incoming uranium as a complex salt (UNH). Knowing the feed supply was imminent, HW officials undertook and finished the conversion of the 224-U Building to the UO_3 Plant in 1951.[51]

Facility test runs began in January 1952 with full operations the following month. For the first time, a full cycle of uranium into and out of HW was completed. The first UO_3 calciners were large pots that heated the UNH in batches. One early difficulty concerned the UNH feed from REDOX. It contained sufficient impurities that much of Hanford's UO_3 product "proved to be inadequate for successful processing in the continuous UF6 conversion process at Oak Ridge." Process improvements extending back into the REDOX Plant were made during 1952 and 1953 to correct this situation. The first feed supplies of UNH received from the TBP Plant in 1953 were also proved problematic. Metallic impurities,

nitric acid content, and the presence of TBP and its decomposition products meant that UO_3 from the TBP Plant stream had to be sent to an intermediate plant for further purification before shipment to Oak Ridge. Additionally, the TBP Plant UNH foamed, caked, and stuck in the pots during the calcining process, so that the UO_3 had to be vacuumed, even chopped out. This condition was undesirable from the standpoints of efficiency, operator safety, and contamination control. However, changes made in the TBP Plant rendered the UNH so pure that by the end of 1953 the UO_3 Plant product could be shipped directly to Oak Ridge without further purification. That year, the UO_3 Plant processed more than 1.5 times the total fuel inventory of uranium received at the Hanford Site.[52]

With the addition of two gas-fired calciners in 1954, UO_3 Plant capacity increased to 18 tons per day. The unit cost per ton dropped to 79 percent that of 1953. However, overall 1954 production was limited by the quantity of feed available from the REDOX and TBP Plants. REDOX experienced many equipment failure shutdowns, and the TBP Plant was closed for much of the autumn to modify the system so that it could process wastes aged for one year instead of three. Several problems were encountered and overcome during 1954. High radiation fields around the calciner pots (caused by the processing of UNH higher in zirconium, niobium, and ruthenium) were reduced by special shielding to protect operators during pot unloading. Sulfamic acid (added to the UNH prior to calcination) caused caking in the calciner pots, and renewed foaming (caused by the presence of organic decomposition products in the UNH) occurred. The latter problems were controlled by silicon anti-foaming agents and other sulfur-based additives. A key improvement was the use of vacuum cleaning techniques that enabled the plant to recover 90 to 95 percent of the oxide powder associated with failed filter bags. A large backlog of such bags, which had accumulated since startup, was cleaned and discarded. By the end of the year, plans were underway to expand the UO_3 Plant.[53]

During 1955, monthly UO_3 production averaged 106 percent over that of 1954 while unit costs per ton dropped to 92 percent. The plant shut down for part of the summer because of contamination problems resulting from high gamma feed at the TBP Plant. More persistent problems resulted from the gradual corrosion and mechanical wear of equipment and calciner seals and gaskets to the point where a vacuum could not be maintained in the pots. Airborne contamination around the pots increased until operators had to wear assault masks. In 1956, the following new equipment was installed to address the problem of contamination: new pot agitators, shafts, seals, bearings, and supports, as well as new pot cover assemblies, offgas systems, and gasketing.[54]

Meantime plans went forward to construct the 224-UA Building, a major addition to the UO_3 Plant (see Figures 16 and 17). The 224-UA Building was equipped with six large, new, continuous action calciners. The calciners, designed and developed at the Hanford Site, had large troughs with paddle agitators that turned and mixed the UNH throughout the process. Caking and clogging problems that had plagued the pots were obviated by the belowgrade valving of the continuous calciners. The new valves used air as an agitant and maintained a seal between the calciner and the powder pickup bin while passing a continuous stream of UO_3. The new calciners also produced a pebbly product consisting of spheres with an average diameter of 200 microns (about 1/100th of an inch), as opposed to the granular oxide product of the pots.

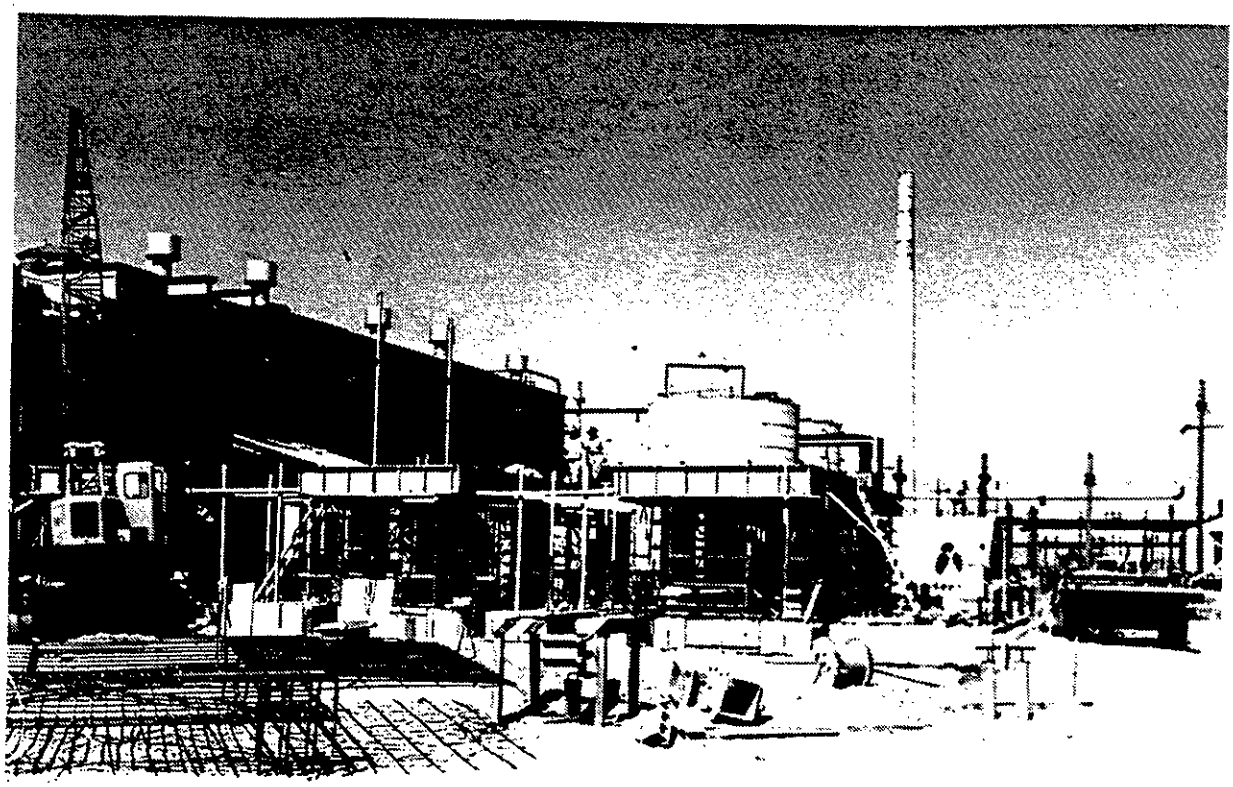

Figure 16. 224-UA Building under construction, August 1955.

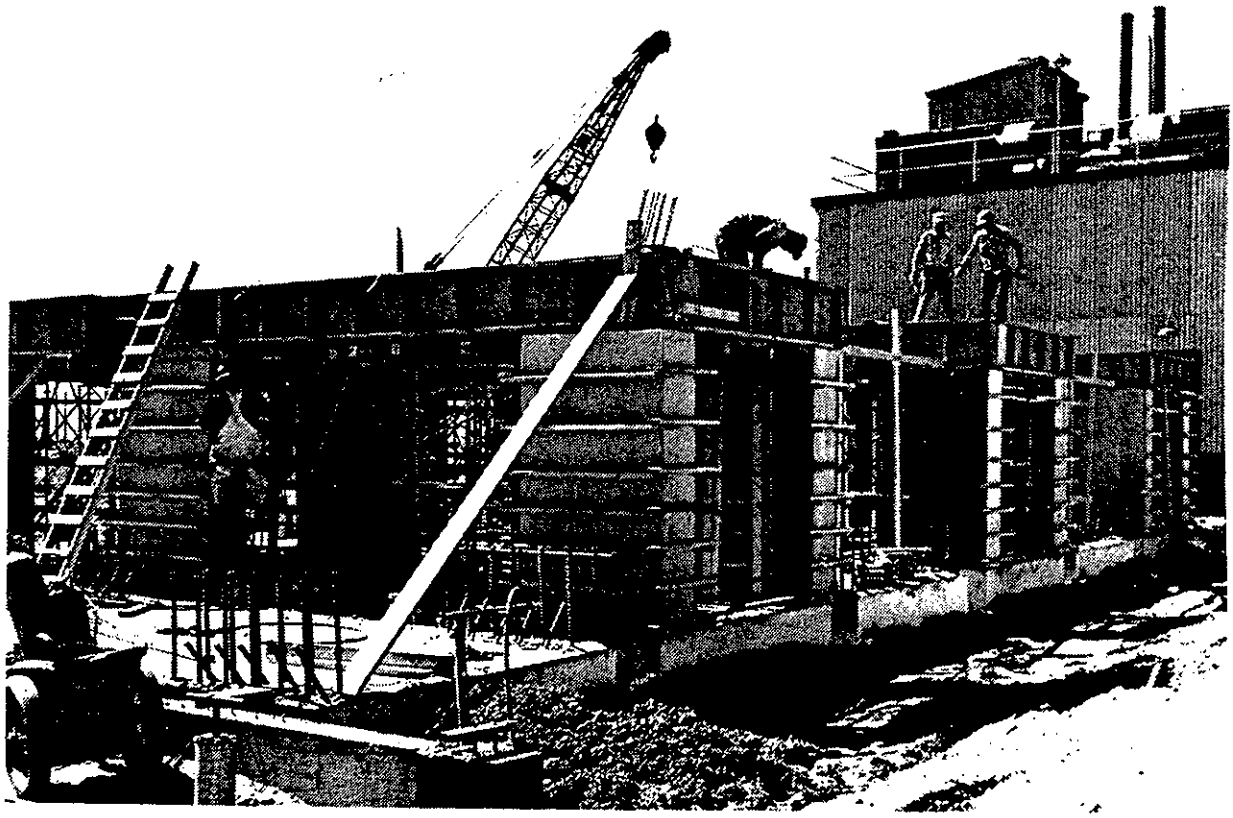

Figure 17. 224-UA Building under construction, August 1955.

Figure 18 shows a continuous calciner in the 224-UA Building. The first three continuous calciners began operations in the last quarter of 1956, and, despite early mechanical difficulties with their powder handling systems, they soon demonstrated their production potential. In December, UO_3 Plant throughput exceeded that of any previous month by 27 percent. Overall, the 1956 plant production exceeded that of 1955 by 57 percent.

Another salient improvement made during 1956 was the replacement of the old 900-pound drums used for offsite shipping of the UO_3 powder by new T-hoppers. These containers left the site on specially fitted rail flat cars and served as feed hoppers for the Oak Ridge Plant customer (see Figure 19).[55]

The last three of the continuous calciners were installed in early 1957. At that time, the 18 pot units were retired from service, and all processing was done through the continuous calciners. Late that year, design scoping was begun for new facilities needed to segregate regular UNH feed from that generated by the processing of E Metal in the REDOX Plant. Because the UO_3 powder that resulted from processing the two types of UNH streams had significantly different reactivity levels, separation was needed at Oak Ridge.

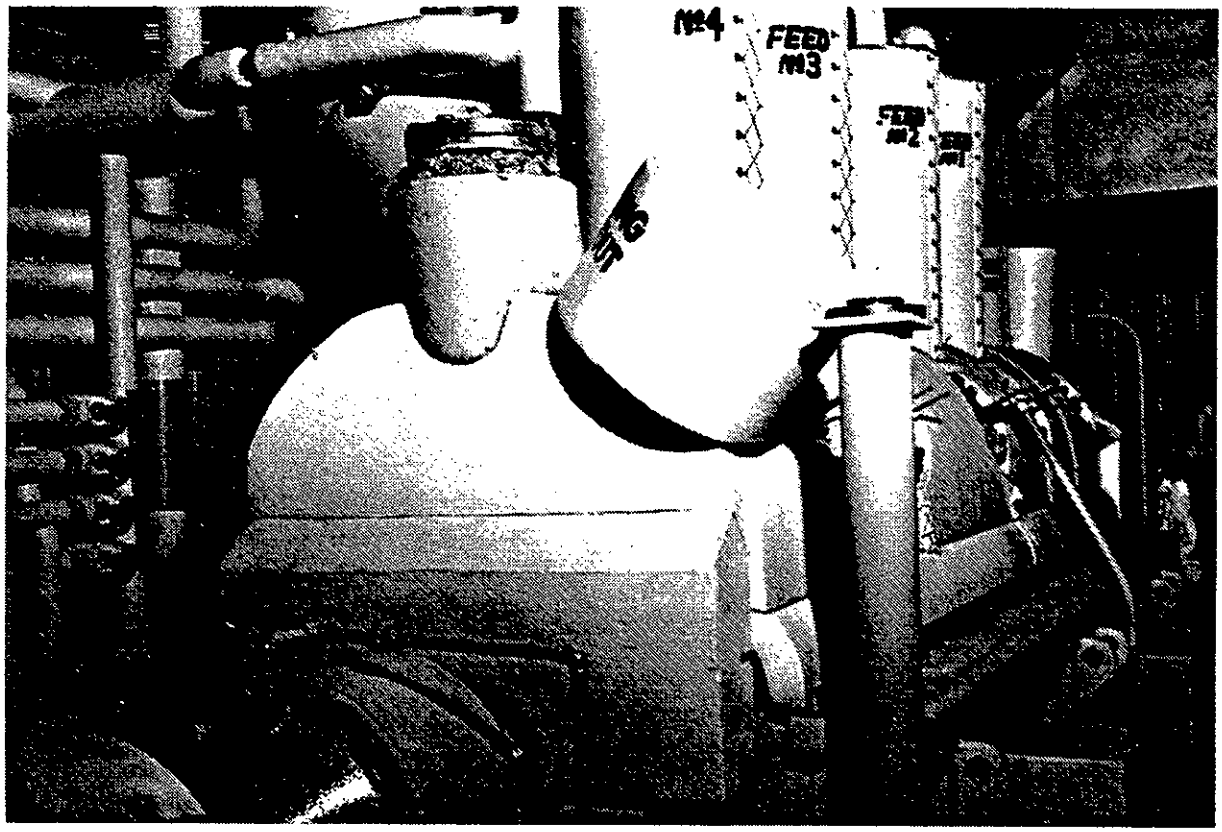

Figure 18. Continuous calciner, new in the 224-UA Building, September 1956.

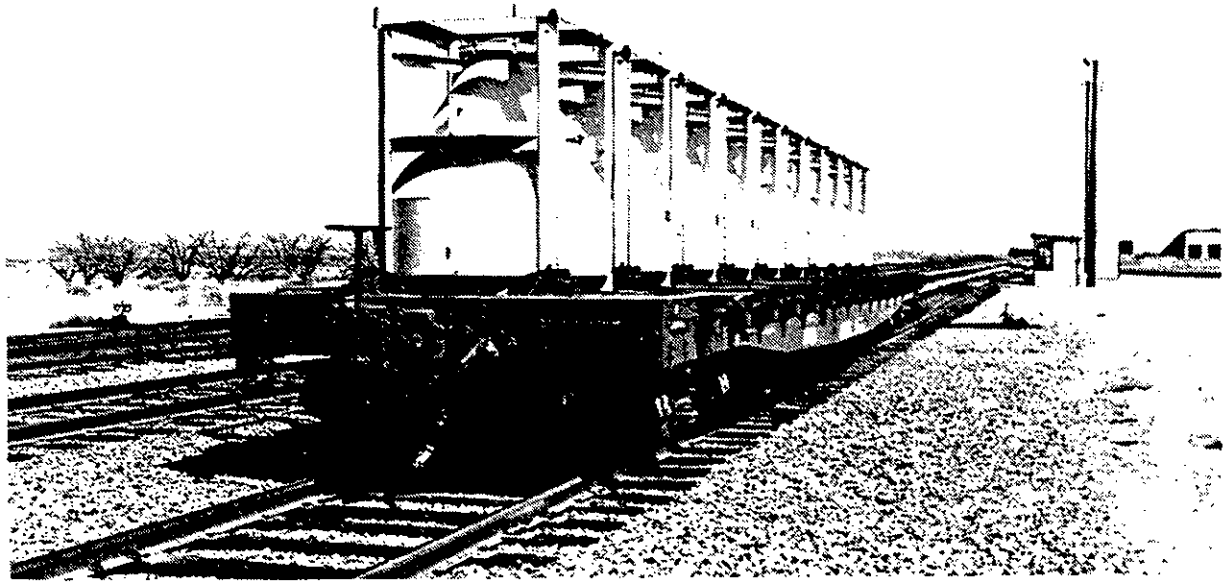

Figure 19. Shipping casks for UO_3 product, August 1956.

During 1958, two new tanks with associated piping and instrumentation were installed and fitted to receive the enriched UNH from the stainless steel, overhead pipeline bringing the stream from the REDOX Plant. Routine transfers of enriched UO_3 from the Hanford Site to Oak Ridge began in early 1959. Meanwhile in 1958, the UO_3 Plant increased production 5.5 percent over 1957 and almost double that of 1954. Metallurgical examination of the two-year-old continuous calciners showed their condition to be excellent.[56]

In 1958, the TBP Plant finished its mission and shut down. However, its feed to the UO_3 Plant was far surpassed by that coming from the PUREX Plant. Together, the UO_3 and PUREX Plants went on to set and surpass production records almost continually while the REDOX Plant continued to supply a small stream of enriched UNH until it shut down in 1967. During 1959, the concentration equipment and the acid recovery system at the UO_3 Plant were automated, and in 1960, for the first time, the calciners were automated so that they could be operated and shut down remotely. Programming of the calciners included preliminary air blowing and steam heating of the feed points, admission and regulation of the feed, and control of the UO_3 bed temperature. Steep production climbs continued through 1961 but dropped off slightly in 1962 because of mechanical and process difficulties at the PUREX and REDOX feed plants. By 1963, production was so high again that serious consideration was given to constructing a pipeline, or converting an existing one, to carry UNH from PUREX to the UO_3 Plant. However, the project did not go forward, and transfers continued by tanker truck. That same year, the production of UO_3 actually shipped offsite rose to about 12,000 tons per year, and the capability was added to process UNH derived from 125 Metal. During 1965 and 1966, an experimental processing of commercial thorium nitrate into thorium oxide powder was carried out in the UO_3 Plant using the old electric pots. The goal of this work was to produce thorium oxide powder suitable for fabrication into reactor target elements for U-233 production. However, for reasons unrelated to the UO_3 Plant, the use of thorium oxide powder was abandoned in favor of experiments with thorium wafer targets.[57]

Beginning in 1967, UO_3 Plant operations were tied exclusively to those of the PUREX Plant. UNH concentrator modifications in the UO_3 Plant improved heat distribution and allowed the calciner feed to maintain a uniformly higher specific gravity, thus producing more powder in the same operating time. The UO_3 and PUREX Plants closed in 1972; PUREX resumed operations in late 1983, closely followed by UO_3 in early 1984. Since that time, there have been 17 startups at the UO_3 Plant, averaging about 8 days each, as the plant can calcine UNH at a much faster rate than the PUREX Plant can produce it. Final deactivation orders came for the plants in 1992. In April 1993, the UO_3 Plant resumed operations to convert 200,000 gallons of remaining UNH to uranium oxide powder. This run was the plant's last (June). A final deactivation plan for the facility, written in the summer of 1993, will be carried out in the next few years.[58]

In general, waste management has been handled very simply and efficiently at the UO_3 Plant. Radiation levels in the UNH are relatively low compared with levels in the chemical processing feed plants; no high-level waste is generated by the UO_3 process. Process condensate has been sent to various U Plant cribs over the years, specifically the

216-U-12 crib (1980's) and the 216-U-17 crib for most recent operations. The U Pond and more recently the 214-U Ditch have taken the equipment cooling water and the steam condensate. Airborne UO_3 powder also has been trapped in primary bag filters, secondary bag filters, and then high efficiency air particulate (HEPA) filters added during the 1972 to 1984 shutdown. The addition of fire protection improvements, a backup power supply for instrumentation, an open faced hood and associated exhaust and filtration equipment for the UO_3 loadout, and a nitrous oxide (NOX) monitor for the 296-U-4 stack were also made in that period. Beginning in late 1950, NOX, the main offgas of the UO_3 process, was routed through wet scrubbers, devices that spray the gas with nitric acid to dissolve the powder within it. The acid mixture then was routed through an acid absorber (bubble-cap tower) and stored in chemical tanks for transfer to and reuse at PUREX. In the late 1980's, a Fiber Mist Eliminator was added to scrub the acid vapors from the UNH concentrator to remove the uranium.[59]

During the early years of operations, several incidents involving spills of UNH and airborne filter breakthroughs occurred at the UO_3 Plant. Additionally, lines within the plant sometimes plugged and ruptured as attempts were made to clear them with steam. At other times, powder emitted in bursts from the old calciner pots resulting in airborne contamination. Two of the more serious early incidents included the spill of 2,000 gallons of UNH on the main roadway east of the 200 East Area hill in December 1954, and a train derailment that spilled 113 barrels of UO_3 into the Clark Fork River near Thompson Falls, Montana in May 1956. The most serious known radiation event involving personnel exposure at the UO_3 Plant occurred in December 1960 when the fresh air mask hose being used by an employee developed leaks and gaps, and the employee inhaled 5,000 microcuries (uCi) of uranium. In 1986, 625 gallons of recovered nitric acid were spilled to the 241-U ditch; and in 1989, 4,420 gallons of slightly contaminated water were lost to the environment from the sump pit of C-Cell in the 224-U Building.[60]

ENDNOTES

1. Campbell, HW-38781, pp. 1-4.

2. Carr, HW-24800-105, pp. 2-21.

3. Ibid., pp. 2-7; and Working Committees RDA DC-4 and RDA DC-7, HDC-2529; and Courtney and Clark, HW-32413-DEL, pp. 5-7.

4. Carr, HW-24800-105, pp. 63-114; and Blaw-Knox Co., "Utilization Report..., pp. 45-49.

5. Ibid., pp. 22-23; and Courtney and Clark, pp. 48-49.

6. Carr, HW-24800-105, pp. 54-62, and Courtney and Clark, HW-32413-DEL, pp. 59-69.

7. Carr, HW-24800-105, pp. 40-42; and GE Hanford Co., HW-30000-DEL, pp. 29-30.

8. Carr, HW-24800-105, pp. 26-34.

9. Ibid., pp. 34-54; and GE Hanford Co., HW-34000-DEL, pp. 23-24.

10. Dow, HW-45102; and Courtney and Clark, pp. 8-9.

11. Felts, HW-33093-RD; and GE Hanford Co., HW-3400-DEL, pp. 23-24; and Felts, HW-35598; and GE Hanford Co., HW-50000-DEL, pp. 23-24; and GE Hanford Co., HW-54000-DEL, p. 31.

12. GE Hanford Co., HW-50000-DEL, pp. 23-24; and GE Hanford Co., HW-54000-DEL, pp. 25-27; and Tomlinson, HW-60892-RD; and Platt and Tomlinson, HW-46138; and GE Hanford Co., HW-60000-DEL, pp. 18-19.

13. GE Hanford Co., HW-64000-DEL, pp. 32-35; and GE Hanford Co., HW-67700-DEL, pp. 11, 22; and GE Hanford Co., HW-72000-DEL, p. 12; and GE Hanford Co., HW-75700-DEL, pp. 4-5, 13, 20; and Duckworth, HW-77678; and GE Hanford Co., HW-82330-DEL, p. 5, 29; and Gydesen, PNWD-2017 HEDR; and IPD and CPD, HW-78100, pp. 5-6, 26; Finch, HW-64162; and Roemer, SD-HS-SAR-001, p. B-7.

14. Bruns, HW-67674; and Kendall, Lambert and Nelson, HW-79665; and CPD, RL-SEP-650; and Harmon, HW-84317; and Denton, "Operating History... ", and PUREX Process Subsection, ISO-419.

15. Fremling, TID-26431; and Research and Engineering, ARH-2874; and "Hanford Mess Means Shake-Up Is Needed," August 5, 1973; and Thompson, "Everybody Will Be Happier When We Understand Atoms," September 2, 1973; and Christensen, Alford, Kelley, Schmidt and Smith, ARH-CD-450; and U.S. ERDA, ERDA-1538; and Brown, Routson, Price and Fecht, RHO-ST-1; and Routson, Price, Brown and Fecht, RHO-ST-14; and NAS, "Radioactive Wastes at the Hanford Reservation...".

16. Cahow, RHO-CD-704; and Roemer, SD-HS-SAR-001, Rev. 5, Section 1; and "PUREX Resumes Operation," April 27, 1988; and "Hanford's PUREX Plant Restarts After Year Closed," December 18, 1989; and "Watkins Orders Standby for PUREX, October 17, 1990; and PUREX Ready for Next Step," December 14, 1992.

17. Carr, HW-24800-105, pp. 54, 236-268; and Sege, HW-28690; and Shefcik, HW-40556.

18. Doud and Stoker, HW-31334; and G.E. Hanford Company, HW-31000-DEL, p. 427; and Michels and Zahn, HW-39265 RD; and Michels and Zahn, HW-40238; and Falk, HW-40730-DEL; and Roemer, and SD-HS-SAR-001, Rev. 5, Appendix B.

19. Hesson, HW-44974; and Finch, HW-49679; and Swift and Irish, HW-52389-DEL; and Fecht, Jaske, Lane and Ludlow, HW-38881; Ryan and Wheelwright, HW-55894; and Roemer, SD-HS-SAR-001, Rev. 5, Appendix B; and Carr, HW-24800-105, p. 40.

20. Richardson, HW-36371-RD; and Coppinger, HW-77080; and Roemer, SD-HS-SAR-001, Rev. 5, Appendix B; Fecht, HW-67783; and Geier and Rathvon, HW-69517; and Greenhalgh, BNWL-460.

21. GE Hanford Co., HW-67700-DEL, p. 11; and HW-75700-DEL, pp. 4-5, 13; and Michels, HW-75975; and Harmon, HW-84317; and Smith, HW-65979; and Lee and Walser, RHO-CD-256; and Clark, Davis, and Schmidt, ARH-SA-9; and Roemer, SD-HS-SAR-001, Rev. 5, Appendix B; and Platt, HW-42978; and Finch, HW-64162.

22. Walser, ISO-584; and Beary, ARH-SA-73.

23. Roemer, SD-HS-SAR-001, Rev. 5, Appendix B; and Clemans, WHC-CM-5-20.

24. Roemer, SD-HS-SAR-001, Rev. 5, Appendix B; and Project Records IAP-600, IAP-608, IAP-615, ICE-628, CAC-152 and IAP-605.

25. Cahow, RHO-CD-704; and Reberger, RHO-CD-1418.

26. Finch, ARH-2291; and Raile, RHO-CD-1262-CDR; and Danch and Schroeder, RHO-CD-268; and Shoemaker, RHO-CD-1259-FDC; and Pamell, RHO-CD-282, Rev. 2; and Prosk, SD-CP-SAR-006; and Vitro Engineering Corporation, RHO-CD-1269-CDR; and Van Meter, RHO-CD-1349; and Braun Hanford Company, B-379-Cl; and Prosk, SC-HS-SAR-013; and Dahlke, SD-CP-DCR-001.

27. Murphy, RHO IL 37400-86-124; and Bowman, RHO IL 65940-85-616; and Nolan and Lorenzini, "Use of 305 Building...", 1985; and Jacobs, WHC IM 12223-88-133; and Schofield, WHC IM 14300-99-110.

28. Kingsley, HW-63703; and Wiater, "Monthly Equipment Progress Reports," 1969; and Szulinski, ARH-SA-181.

29. Actually, eight waste cribs were designed for the PUREX facility, but 216-A-1 was abandoned before startup. It was to have received condensate from the 241-A waste tanks. However, when a contact-type condenser was chosen for the 241-A tanks, instead of a shell and tube-type, it was realized that the A-1 crib would be inadequate for the increased flow rates. See Courtney and Clark, HW-32413-DEL, p. 78; and Carr, HW-24800-105, p. 56.

30. Courtney and Clark, HW-32413-DEL, pp. 77-83; and Carr, HW-24800-105, pp. 56-57.

31. Bierschenck and McConiga, HW-51277; and Brown et al, HW-49465; Michels and Zahn, HW-44506; and Brown, et al, HW-50186; and Andersen, CIC-12765 (for December); and CETO, HW-50195.

32. Wood, HW-46396; and Bierschenck, HW-49728; and CETO, HW-53225; and CETO, HW-54655; and Clukey, HW-41440; and Roemer, SD-HS-SAR-001, Rev. 5, Appendix B; and CETO, HW-54848; and CETO, HW-58811-RD; and Klepper, Rogers, Hedlund and Schreckhise, PNL-1948-UC-70.

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37. O'Neill and Swift, HW-41791, Parts I and II.

38. Hanthom, HW-42230; and O'Neill and Swift, HW-41791-Part 11; and Roberts, HW-54750 and Linderoth, HW-50198; and Linderoth, HW-51682; and Haney, HW-58211.

39. Stivers, HW-57289; and Fraser, HW-43986; and Anderson, HW-58329; and G.E. Hanford Company, HW-67700-DEL, p. 22; and G.E. Hanford Company, HW-75700-DEL, pp. 4-5, 20; and G.E. Hanford Company, HW-82330-DEL, pp. 5, 29; and Buckingham, HW-80563; and Isaacson, ARH-1491; and Bowers and Foster, eds., ARH-ST-103.

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