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# Solidification and Improved Treatment of Low and Intermediate Level Radioactive Wastes at Hanford

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**SOLIDIFICATION AND IMPROVED TREATMENT  
OF LOW AND INTERMEDIATE LEVEL  
RADIOACTIVE WASTES AT HANFORD**

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Date: August 17, 1970  
To: Holders of SM-137/50  
From: *P. W. Smith*  
P. W. Smith  
Subject: ERRATA SHEET - SM-137/50

Please make the following change to SM-137/50, "Solidification and Improved Treatment of Low and Intermediate Level Radioactive Wastes at Hanford", by W. L. Godfrey, G. L. Hanson and P. W. Smith, dated August 5, 1970:

3.3, Page 8, last paragraph

"Overheads from the nitric acid recovery condenser represented only 25 percent of the Purex process condensates but contained approximately 95 percent of the total radioactivity, principally plutonium.  
This con-"

The underlined word should be ruthenium.

PWS:cb

cc: RP Corlew  
LM Richards  
JH Warren  
Central File

AEC

OJ Elgert, RLO  
JH Kane, DTIE *[Signature]*

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REVISED ABSTRACT

This paper discusses operational experience in large scale production techniques used to solidify intermediate level radioactive liquid wastes and to improve the treatment of low and intermediate level radioactive liquid wastes at Hanford.

Three types of evaporation units are used to concentrate intermediate level wastes to solids: a 650 C in-tank air sparger; a 4 megawatt in-tank electric immersion heater; and a thermosiphon evaporator. The hot air sparge unit operates semi-continuously to concentrate fresh feed to high viscosity and specific gravity for blending with feed to the electric immersion heater. Special off-gas treatment is required due to the large heated air flow. The electric immersion heater and the thermosiphon evaporator operate on a continuous recycle system, utilizing several  $2.8 \times 10^6$  litre storage tanks in series for air cooling and salt accumulation. Salts are deposited from the cooled mother liquors and the supernatant solutions are mixed with fresh feed for recycle to the evaporators. When a cooled tank is filled with solids the residual liquid is removed leaving an immobile salt cake.

In past practice at Hanford, cooling water effluents have been routed to artificial ponds and certain low and intermediate level wastes to underground tile fields where the soil's active ion exchange sites remove and retain the associated radioactivity. Several of the latter streams are being treated as part of a program to reduce the radioactivity routed to the ground to the lowest practicable quantity.

Monitoring systems have been installed for some liquid effluents, such as process cooling water and steam condensates

which activate diversion to intermediate level disposal in case of an increase in radioactivity due to equipment failure or an accident.

Intermediate level liquid wastes from laboratory facilities, nuclear reactor flushes and decontamination of nuclear fuel processing equipment are evaporated to remove radioactivity prior to discharge to the soil. The more radioactive process condensates in the Purex nuclear fuels processing plant are recycled to the process. Condensates from high level waste, self-boiling storage tanks and the in-tank solidification units are recycled or treated by ion exchange to remove cesium-137.



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1.0 INTRODUCTION

Radioactive wastes from the chemical processing of irradiated fuels have been accumulated at the Hanford site since the start of production in 1944. Intermediate-level liquid wastes have been stored as alkaline slurries in tanks with high-level wastes, or, along with certain low-level wastes, have been disposed of to soil storage sites through underground distribution systems called "cribs". Cooling water effluents have been sent to ponds where they percolate through the soil or evaporate. The remoteness of the Hanford site and its favorable geological and climatic conditions have permitted extensive use of the ground for disposal of these wastes. The control and management of the radioactive wastes at Hanford have been discussed in detail previously at international conferences at Geneva[1, 2], Monaco[3], Saclay[4], and Vienna[5].

The management of radioactive wastes at Hanford has continued to evolve and the program currently includes, (1) removal of the long-lived fission products ( $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ ) from the high-level wastes; (2) storage of the remainder for decay of the short-lived fission products; (3) solidification of stored high salt intermediate-level wastes in existing tanks; and (4) additional treatment of low and intermediate-level waste effluents.

Hanford low-level wastes range in radioactivity concentrations from undetectable up to  $5 \times 10^{-5} \mu\text{Ci/ml}$  which is the mid-point of IAEA Category 2 and intermediate-level wastes range from the mid-point of Category 2 through Category 4 which has a maximum concentration of  $10^4 \mu\text{Ci/ml}$ .

Process cooling water effluents which are routinely free of radioactivity have the potential for becoming contaminated in the event of

an equipment failure. This water is routed to surface ponds. Intermediate-level wastes with radioactivity concentrations up to  $100 \mu\text{Ci/ml}$  and low-level wastes having a high potential for contamination, are disposed to the ground through sub-surface distribution systems called "cribs". Intermediate-level wastes of concentrations between  $100$  and  $10^4 \mu\text{Ci/ml}$  are stored in underground tanks and are being solidified.

This paper discusses production experience with the techniques used at Hanford to solidify intermediate-level radioactive liquid wastes in tanks and with methods used in the pre-treatment of low and intermediate-level radioactive liquid wastes being sent to the ground. The treatment and storage of high salt intermediate-level waste and the land requirements and waste management policy at Hanford will be covered in two other papers presented at this conference.

## 2.0 IN-TANK SOLIDIFICATION PROGRAM

Large volumes of liquid radioactive wastes generated at Hanford during the past 25 years are stored in underground tanks with capacities from  $2 \times 10^5$  to  $3.8 \times 10^6$  litres. It is the goal of the Hanford Waste Management Program to solidify all of the stored, as well as the current wastes as they are generated. A concentration-crystallization process in which the underground tanks are utilized for solid salt-cake storage (Figure 1) was determined to be a safe and economical method of meeting the goal.

Waste solutions ranging in concentration from  $100 \mu\text{Ci/ml}$  to  $10^4 \mu\text{Ci/ml}$  are concentrated in evaporators until crystallization begins at the boiling temperature. The resulting slurry is continually transferred to air-cooled underground tanks where the slurried solids deposit together with the crystals which form as a result of cooling. Cooled supernatant is decanted from the crystallizer tanks, mixed with fresh feed, and recycled to the evaporators for further concentration. Solids deposit as wet sludges in the crystallizer tanks until the tanks are essentially filled; the small volume of residual supernatant is then pumped out and a desiccant added or air drying employed to "fix" any residual liquid.

Three different evaporator facilities are presently concentrating radioactive waste solutions to salt cakes and a fourth is in the planning stages: a hot air sparge evaporator, a steam-heated thermosiphon evaporator, and an electrical immersion heater evaporator are in operation and a vacuum evaporator-crystallizer is currently being designed. The use of several types of evaporator systems reflects the continued development of commercial evaporator technology and its application to the unique Hanford waste solidification operations.

The air sparge evaporator has been previously described in detail[6], and will be discussed here only with respect to its off-gas system. The steam-heated evaporator is similar to standard equipment in the chemical processing industry and since it has no particularly noteworthy features, it will be discussed only briefly. The electrical immersion heater is a unique facility designed to take advantage of recently developed technology and will be discussed in greater detail.

## 2.1 Hot Air Sparge Evaporator

The hot air sparge evaporator, termed the First In-Tank Solidification Unit (ITS-1), operates by introducing electrically heated compressed air through an airlift circulator located in a large underground tank. The off-gases leaving the high salt solutions in the concentration tank are unsaturated with respect to pure water and contain entrained radionuclides which must be removed prior to release of the non-condensibles to the atmosphere.

The original ITS-1 off-gas train consisted of a stainless steel mesh deentrainer, a deep bed fiberglass filter, a condenser, an exhaustor, and a short stack. Several months after startup, the fiberglass filter failed from caustic corrosion of the glass fibers due to entrainment from the sparger unit and an identical replacement filter failed in only a few weeks from the same cause. Operation of the ITS-1 unit without the fiberglass filter was not acceptable because of the potential for release of radionuclides to the environs. The addition of high efficiency filters downstream of the exhaustor effectively prevented radionuclide releases, but high dust loading in the process off-gas stream and/or high radiation readings (~500 mR/h) on the filters necessitated filter changes about every three days. A short-term solution was achieved by installing a Teflon pad deentrainer between the condenser and exhaustor. While this system was operable, it was not a long-term solution since the Teflon pads required frequent flushing and the filter life had only been extended from a few days to a few weeks.

A program was initiated to develop design data for an improved off-gas deentrainment system[7]. It was found that the entrained particulate loading was approximately  $5 \times 10^{-4}$  grams per 1000 actual litres of off-gas and the mean size of the entrained particles was below 3  $\mu$ m. On the basis of prototype field test information a full scale Brink Mist Eliminator<sup>R</sup> was installed ahead of the condenser.

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<sup>R</sup> Registered trade name of the Monsanto Chemical Company

The assembly, shown in Figure 2, contains 12 synthetic fiber elements in an insulated cylindrical carbon steel vessel equipped with a flanged top for installation and removal of the fiber elements. Each of the 12 fiber elements is composed of a cylindrical polypropylene fiber bed restrained between concentric stainless steel wire screens. The bed fibers are 20  $\mu\text{m}$  in diameter and are packed to a density of 185 g/l; each element is designed for a superficial gas velocity of 5 m/min with collection efficiencies independent of differential pressure. The design particle removal efficiency is 100 percent for particles above 3  $\mu\text{m}$  and 95 - 99 percent for particles below 3  $\mu\text{m}$ . The maximum recommended operating temperature is limited to 93 C by the polypropylene fibers and for extended operation the maximum recommended differential pressure across the fiber bed is  $2.5 \times 10^{-2}$  atm. Exceeding these limits can result in permanent bed deformation and subsequent channeling of the gas stream through the beds. During normal operations the differential pressure is less than  $1.5 \times 10^{-2}$  atm at flow rates of  $1.4 \times 10^5$  actual l/min.

The fiber elements are operated as self-cleaning units by adding water through spray nozzles at a rate of at least 1 l/min per  $3.0 \times 10^4$  actual l/min of saturated gas. Residual insoluble solids are removed by soaking the bed in a suitable solvent when fouling is encountered; however, only one soaking has been required since the unit was installed in March of 1968. Since startup, a minimum particle removal efficiency of 95 percent has been demonstrated.

The air sparge unit has a demonstrated space recovery rate of 15 - 20 l/min and since startup of the unit in early 1965, approximately  $37 \times 10^6$  litres of storage space have been recovered. Concentrate from this evaporator is routed to the electric immersion heater system for solidification.

## 2.2 Steam Heated Evaporator

The steam heated evaporator is a conventional short-tube thermosiphon evaporator installed in a shielded concrete structure. Vapor treating equipment including a deentrainer, a cyclone separator, a condenser, condensate catch tanks and off-gas filters is also installed in the shielded building. Chemical flushes are used when required to reduce radiation fields sufficiently to permit contact maintenance.

The steam heated evaporator has a demonstrated space recovery rate of about 50 l/min. Since the facility was started up in 1965, approximately  $47 \times 10^6$  litres of storage space have been recovered and  $6.0 \times 10^6$  litres of salt cake have been produced.

### 2.3 Electric Immersion Heater Evaporator

The electric immersion heater unit consists of 4 one-million watt electric immersion heaters installed in a  $2.8 \times 10^6$  litre underground waste storage tank which has been modified to serve as an evaporator vessel [8]. All equipment requiring routine maintenance is installed above ground and is readily accessible for contact maintenance after chemical flushing.

A mixture of fresh feed and recycled mother liquor is pumped from the feed tank to the evaporator tank and is concentrated. Steam is drawn from the evaporator vessel vapor space, which also serves as a large settling chamber for entrained droplets, and is passed through a wire mesh deentrainer and condensed. The non-condensibles are deentrained, filtered through high efficiency filters and discharged to the atmosphere. Concentrated waste solutions are continuously pumped from the concentrator tank to a cascade of air-cooled underground tanks for cooling and salt deposition.

The electric immersion heater consists of four separate one-million watt quadrants installed in a single 100 cm diameter airlift circulator. Quadrant construction was chosen to facilitate heater fabrication, transportation, installation, and maintenance. Four floating delta transformers supply 480 volt, 3-phase power to the heater quadrants, and each is operated independently to provide operating and maintenance flexibility. Each quadrant is an assembly of 90 hair-pin shaped low watt density heating elements, 1.3 cm in diameter and 12 m long. The lower 4 m of the hair-pin section contains a helical Nichrome<sup>R</sup> resistance wire held in a MgO matrix and encased in an Inconel<sup>R</sup> sheath. The airlift circulator generates high liquid flow rates past the heater elements which retards fouling and increases the heat transfer coefficients.

Though scale formation is retarded, elements do foul; however, the scale is water soluble. The heater quadrants are cleaned by removing them from the concentrator tank at about 90-day intervals and allowing them to soak for a few days in a tank filled with fresh water.

The electric heaters were initially started up in December, 1967, and operated without incident until July, 1968, when electrical burnout problems developed. The failure of large numbers of heater elements apparently occurred simultaneously without warning and was

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<sup>R</sup> Registered trade name of the International Nickel Co.

accompanied by arcing throughout the heater bundle. The cause was eventually traced to a combination of manufacturing variations, operating conditions, and electrical characteristics peculiar to floating delta electrical systems. As a result of these early failures three quadrants were eventually removed from service.

Ground fault detection equipment was installed which automatically disconnects the heaters when a ground fault current develops equivalent to 1/10 the normal current rating of a single element (approximately 2.3 amps current flow to ground, out of a total of 2100 amps per quadrant). Quick acting fuses were also installed on both legs of each element to limit current flow to 110 percent of the element's rated capacity. The addition of the protective equipment has been successful in preventing additional failures.

The demonstrated space recovery rate for the electric immersion heater facility is 50 t/min. Since startup in December of 1967, approximately  $41 \times 10^6$  litres of storage space have been recovered and  $4.5 \times 10^6$  litres of salt cake have been produced.

### 3.0 IMPROVED TREATMENT OF LOW AND INTERMEDIATE-LEVEL WASTES

Historically at Hanford, low-level waste effluents, consisting primarily of process cooling water, have been released to isolated surface ponds. Intermediate-level wastes of less than  $100 \mu\text{Ci/ml}$  such as process condensates, contaminated steam condensates, and organic solvents have been disposed of to the soil through sub-surface distribution systems. In such cases either the ion exchange properties of the soil or the soil's specific retention capacity is relied on to "fix" the radionuclides. A program to provide improved treatment of low and intermediate-level wastes was started in 1967 with initial emphasis on the most highly radioactive streams. Plant scale operating experience is now being gained.

#### 3.1 Cooling Water and Condensate Recycle and Diversion

The Waste Management Facility (B Plant) processes (a) high-level current acid waste from the Purex plant for recovery of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ ; (b) stored high-level waste supernatants for the recovery of  $^{137}\text{Cs}$ ; and (c) waste storage tank sludges for the recovery of  $^{90}\text{Sr}$ . The separated strontium and cesium fractions are concentrated and stored as solutions in in-canyon stainless steel tanks, pending completion of facilities for solidification and encapsulation. These product storage tanks are continuously cooled to dissipate the radioactive decay heat and the cooling water is collected

via six sub-header systems, which can be valved to either the plant sewer or to a by-pass system as shown in Figure 3.

Water exiting the by-pass system passes a continuous scintillation gamma monitor upstream of a diversion station. Normally the water overflows from the diversion station into the plant sewer and to retention basins before discharge to a surface pond. The diversion station contains a dump valve, activated by the gamma monitoring system which routes contaminated water to an emergency disposal trench. The system is sized with reference to the response time of the radiation monitor and dump valve arrangement to avoid routing contamination into the retention basins and disposal pond. It is checked for operability once each week. In case of an incident, the sub-header system is manually monitored and the sub-header found to be the source of the radioactivity is isolated until repairs can be made.

During the four years this system has been in operation, one coil has failed and the header was automatically dumped to the emergency disposal trench as designed. Similar systems are in service in other locations.

A coil or tube bundle used in alternate heating and cooling service is more susceptible to failure than a similar coil used only for cooling service; consequently, steam condensates and cooling waters from  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  product concentrator tube bundles, although uncontaminated, are discharged to underground cribs. During the first thirty minutes after startup of a concentrator the condensate is recycled to contain radioactivity which might result from a coil failure during the shutdown period.

The steam condensate is routed to a sampler tank which overflows to the crib. A continuous sampler-radiation monitor routinely monitors the stream for detection of abnormal radioactivity. If abnormal radioactivity is detected, the steam condensate inlet stream can be recycled and the concentrator shut down for repairs; however, no tube bundle failures have ever been experienced in this system.

### 3.2 Decontamination of Waste by Evaporation

Intermediate-level radioactive liquid wastes with concentrations of between  $5 \times 10^{-5}$  and  $100 \mu\text{Ci/ml}$  from Hanford operations other than the fuels reprocessing facilities, have been transported to a central location for discharge to underground cribs since 1953. In 1967, evaporators located in the Redox facility - a

shut down fuels reprocessing plant - were reactivated to add another stage of decontamination before discharge to cribs. A waste unloading station was provided at the Redox site for transfer of the liquid waste from rail tank cars or truck trailers to the evaporators and an existing waste system was used to route the evaporator bottoms to underground tank storage and subsequent solidification.

Reactor flush solutions are also transported to the Redox plant waste concentrators for evaporation prior to crib discharge as are dilute wastes from the decontamination of failed equipment.

Wastes intended for evaporation are neutralized to a pH of  $\geq 10$  by the generating facility. Dilute wastes are shipped to the Redox plant in 19,000-litre truck tankers or 76,000-litre rail tank cars from the laboratory and reactor areas while the decontamination facility wastes are pumped via existing lines. Two 60 t/min waste evaporators are available in the Redox facility, which can be operated separately or together as the demand requires. Vapors from both evaporators are routed to a common shell and tube condenser and the condensate is collected, sampled, analyzed, and discharged to an underground crib. If the condensate is above normal levels of radioactivity, it is recycled to the waste evaporator.

### 3.3 Purex Process Condensate Recycle

In Hanford's Purex fuels reprocessing plant, the high-level aqueous waste from the solvent extraction First Decontamination and Partition Cycle is evaporated in a high-level waste concentrator for volume reduction and recovery of the nitric acid. The concentrator feed solution is diluted with water to optimize nitric acid volatilization and the nitric acid concentration of the concentrated waste is reduced from  $\sim 6M$  to  $1.5M$  in a batchwise reaction with sucrose solution at elevated temperatures. The overheads from the waste concentrator and the nitrogen oxides from the sugar denitration operation are combined and routed to a nitric acid absorber for acid recovery. Vapors leaving the acid absorber are condensed and discharged to an underground crib together with other condensates from product and intercycle concentrators.

Overheads from the nitric acid recovery condenser represented only 25 percent of the Purex process condensates but contained approximately 95 percent of the total radioactivity, principally <sup>239</sup>plutonium and <sup>241</sup>americium. This condensate was recycled in process tests to determine the reduction which could be obtained in the activity released and to determine the effect of recycle on solvent



extraction plant performance. Testing showed a decreased fission product activity in the waste stream when 25 percent of the waste stream was recycled as reflux water, 40 percent as dilution water to the high-level waste concentrator feed solution and 15 percent to the "back-cycle" waste system. Fission product activity in the recovered acid increased somewhat, but remained at acceptably low levels. As was expected, no detectable adverse effects on either solvent extraction fission product decontamination or plant operating performance were observed. The recycle system has been in operation for over two years. Approximately 80 percent of the waste stream is now being recycled to the process accompanied by a corresponding reduction in radionuclide discharge via this stream. As an additional environmental benefit, the nitric acid discharged to the process condensate crib via this stream was also reduced by about 80 percent.

Approximately 30 percent of the total Purex process condensate volume is contributed by condensate from the first cycle uranium product concentrator. Collection and re-use of this condensate as a uranium stripping solution has been routine since October, 1969. No adverse process effects have been noted. Although this stream contains less than 5 percent of the total radioactivity previously discharged to the process condensate crib, the successful recycle of this stream has demonstrated that recycle of additional streams may be feasible.

### 3.4 Recycle of Waste Management Process Condensates

Aqueous salt wastes from B Plant processing are concentrated prior to storage in underground tanks. Condensates are routed to a sample tank which overflows to a crib. A continuous sampler-radiation monitor is installed in the sampler tank for routine sampling and continuous monitoring of radioactivity. Initial operation of the waste concentrator, using dilute stored wastes as feed, began in late October, 1967, and condensate-recycle capability was installed in August of 1968. Recycle of process condensate from the sampler tank to the waste concentrator feed tank is initiated by the detection of abnormal radioactivity in the condensate stream by the continuous sampler-radiation monitor. When the radiation alarm sounds, the recycle pump is turned on and a valve in the crib discharge line is closed. Condensate recycle to the concentrator is continued until the radioactivity decreases to normal operating levels. The first year's operation of the recycle system reduced the total beta activity discharges by a factor of 13. Further reduction of radioactivity in this stream, by ion exchange treatment for  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  removal, is planned.

### 3.5 Ion Exchange Treatment of Process Condensates

The large volumes of process condensate from the evaporation of wastes in the three In-Tank Solidification (ITS) units and condensates from the Hanford self-boiling high-level waste storage tanks are routinely routed to underground cribs. Recycle of the condensates to storage tanks can be accomplished during periods of high entrainment and production scale treatment systems to remove trace amounts of radioactivity prior to discharge have been and are currently being tested.

A shielded ion exchange column 45 cm diameter by 245 cm high, containing 285 t of a naturally occurring zeolite, was installed to process the condensate from the hot air sparge evaporator in In-Tank Solidification Unit No. 1. Zeolite minerals have a high affinity for cesium which is the primary isotope entrained in the waste evaporator streams. The condensate was filtered prior to entering the ion exchange unit to remove particulate matter. During the test period, approximately  $1.3 \times 10^6$  litres of condensate were processed and a  $^{137}\text{Cs}$  concentration reduction from a nominal  $3 \mu\text{Ci/t}$  to  $5 \times 10^{-3} \mu\text{Ci/t}$  was effected. This latter value is below applicable USAEC guidelines for maximum concentrations of  $^{137}\text{Cs}$  in drinking water ( $\text{MPC}_w$ ). The test was terminated when other process improvements reduced the  $^{137}\text{Cs}$  concentration in the ion exchange column feed to below  $\text{MPC}_w$ .

A second ion exchange process test is currently being conducted on the condensate from the self-boiling waste storage tanks. The test is primarily designed to determine whether entrained chemicals and/or organics would interfere with the ion exchange process. A 60 cm diameter by 300 cm shielded column containing 850 litres of a naturally occurring zeolite was installed in the boiling tank farm condensate system as shown in Figure 4. Initial results based on  $3.4 \times 10^6$  t throughput, indicate the condensate  $^{137}\text{Cs}$  concentration is maintained below  $\text{MPC}_w$  with reduction factors varying from 4 to 80 depending on the influent concentration. Some chemical degradation of the zeolite binder material has been observed and the test will be continued with a binderless synthetic zeolite. Following the  $^{137}\text{Cs}$  removal demonstration, a mixed-bed ion exchange system will be tested for removal of  $^{90}\text{Sr}$  and  $^{144}\text{Ce}$  as well as cesium.

#### 4.0 SUMMARY

The treatment of Hanford low and intermediate-level wastes is an integral part of the Hanford Waste Management Program. The high salt intermediate-level wastes are being evaporated to immobilized salt cakes and stored in existing underground tanks. Treatment methods including evaporation, ion exchange and recycle are being implemented to reduce radioactivity concentration in the intermediate-level effluent streams currently sent to underground cribs. Cooling water diversion systems have been installed on selected streams to prevent radioactivity discharge to surface ponds in the event of equipment failure.

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FIGURE 1  
SECOND IN-TANK SOLIDIFICATION UNIT

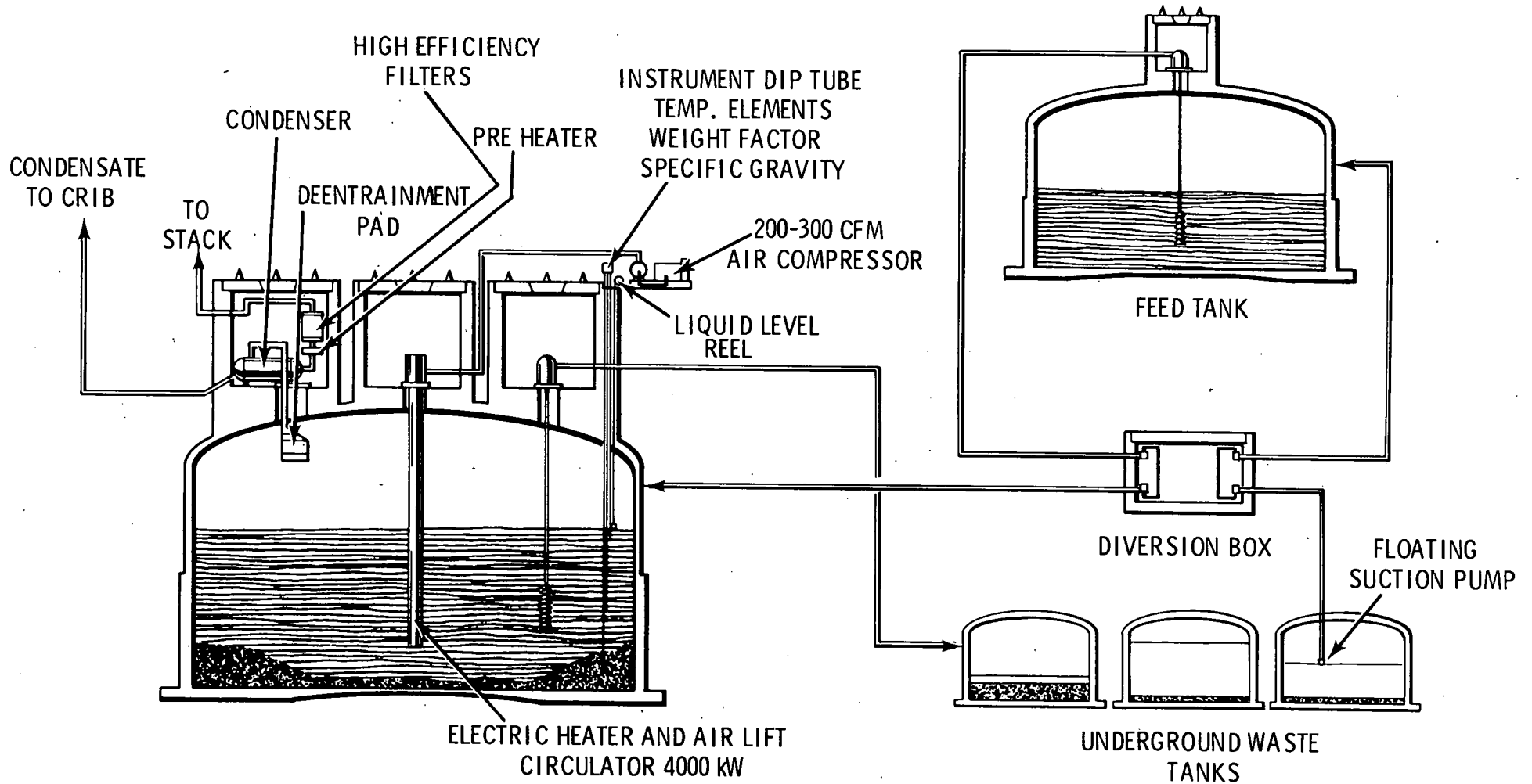


FIGURE 2  
BRINK MIST ELIMINATOR ASSEMBLY

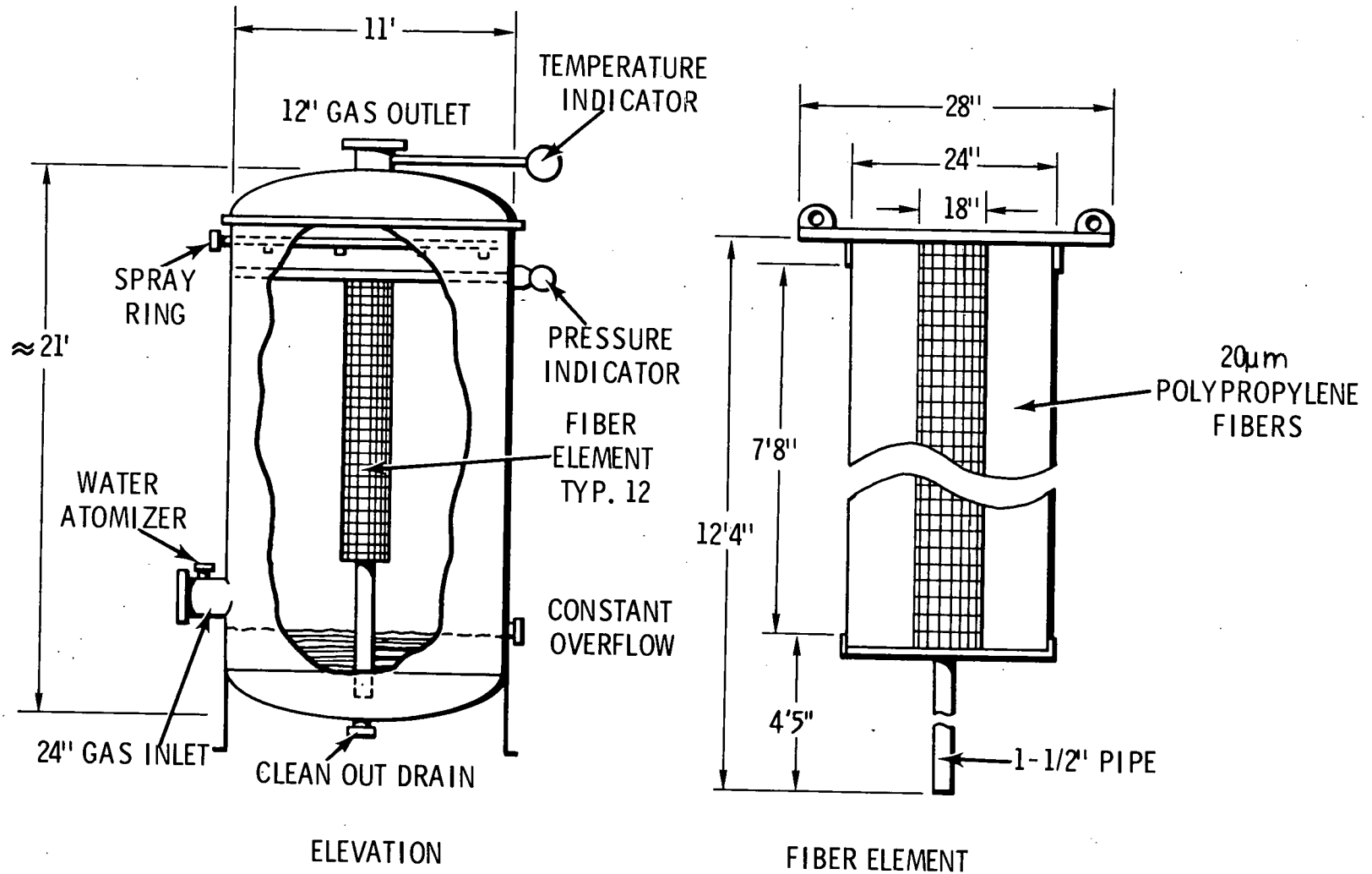
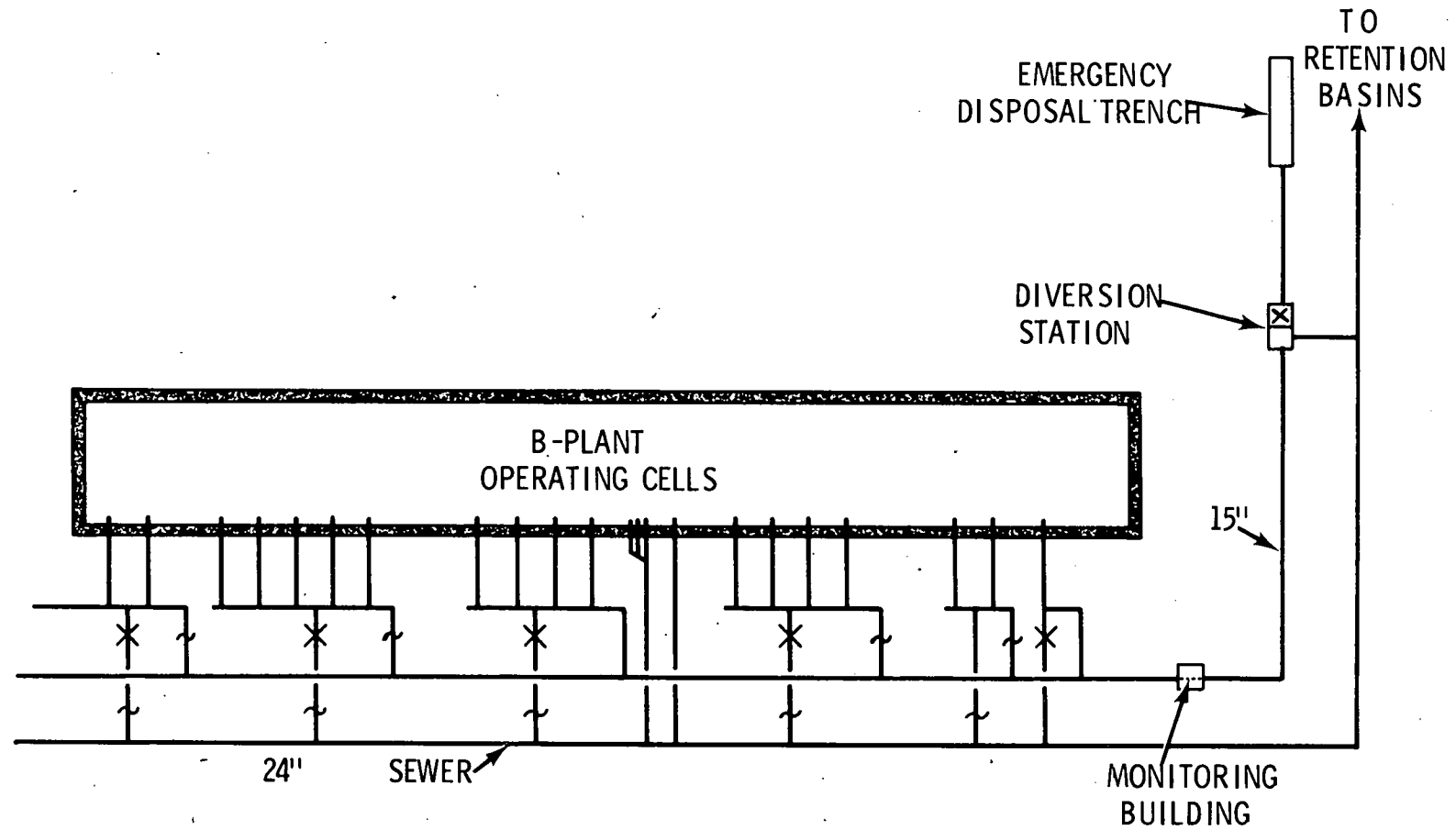


FIGURE 3  
B-PLANT COOLING WATER DIVERSION



LEGEND

X NORMALLY CLOSED VALVES

~ RADIATION MONITORING WELLS

FIGURE 4  
CONDENSATE ION EXCHANGE UNIT

