INSTALLATION, START-UP AND OPERATION OF A PUREX HLW SLUDGE MOBILIZATION AND WASH SYSTEM AT THE WEST VALLEY DEMONSTRATION PROJECT

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ABSTRACT

Alkaline and acidic high-level radioactive wastes are contained at the Western New York Nuclear Service Center in two underground tanks. The purpose of the West Valley Demonstration Project is to retrieve the wastes from the tanks and to solidify them in borosillicate glass with a slurry-fed ceramic melter. The alkaline waste has formed two layers: a supernatant and an underlying sludge.

To prepare the sludge layer for vitrification and to reduce the volume of glass produced, West Valley Nuclear Services (WVNS), Inc., is washing the sludge and pretreating the resultant sludge wash solution. Washing the sludge involves adding a dilute caustic solution and resuspending the sludge. WVNS has designed and developed a Sludge Mobilization and Wash System (SMWS) to complete this task. Pretreating the resultant wash solution entails running the solution through four ion-exchange columns filled with Linde Ionsiv IE-96® zeolite treated with titanium. When sludge wash solution processing is complete, the SMWS will be used to resuspend the settled sludge solids in the tank so that they can be removed and transferred to the melter.

BACKGROUND

In 1980, the West Valley Demonstration Project Act directed the United States Department of Energy (DOE) to develop and demonstrate technology for the safe retrieval and solidification of alkaline PUREX (plutonium uranium reduction extraction process) and acidic THOREX (thorium extraction) high-level radioactive wastes contained at the Western New York Nuclear Service Center (WNYNSC). The 2,250.0 m^3 of PUREX and 50.0 m^3 of THOREX high-level waste (HLW) resulted from fuel reprocessing operations performed by Nuclear Fuel Services, Inc., from 1966 to 1972. Approximately 640 metric tons of commercial and government spent fuels were processed during that period. In 1981, the DOE selected West Valley Nuclear Services (WVNS) Company, Inc., a subsidiary of Westinghouse Electric Corporation, as the management and operating contractor for the West Valley Demonstration Project (WVDP). The DOE assumed responsibility for the site in February 1982.

WVNS has designed, developed, and tested a vitrification process for solidifying the HLW in borosillicate glass. The DOE has formally selected borosillicate as the terminal HLW form (1, 1). The vitrification process, which involves feeding the waste to a slurry-fed ceramic melter (SFCM), will be performed in the WVDP Vitrification Facility (VF), which presently is in the final stage of construction.

The high-level PUREX waste is contained at the WVDP in an underground carbon-steel tank, Tank 8D-2. Tank 8D-2 resides in a Waste Tank Farm comprised of four tanks enclosed in three underground concrete vaults (Tank 8D-3 and 8D-4 are contained in the same vault). Tank 8D-1, which is identical to 8D-2, serves as a spare to 8D-2. Tank 8D-4, which is constructed of stainless steel, contains the acidic THOREX waste. Tank 8D-3, which is identical to 8D-4, serves as a spare to 8D-4. Since Tank 8D-2 is carbon steel, the acid waste by-products of the PUREX process had to be neutralized. The neutralized waste formed two layers: a precipitated sludge, which contained insoluble oxides, hydroxides, and crystallized salts and an overlying alkaline supernatant comprised of concentrated salts at a pH of 10. Rykken et al. have characterized the radiochemical composition of the Tank 8D-2 waste (2).

The sludge layer had an average depth of 0.5 m. The predominant crystallized salt was sodium sulfate. Greater than 90 percent of the radioactivity in the insoluble solids within the sludge layer consisted of ^{90}Sr (7,700,000 Ci) and ^{90}Y (7,700,000 Ci)(3, 2). The supernatant solution contained approximately 40 weight percent salts. The salts consisted of 81 weight percent alkali metal nitrates and nitrites and about seven weight percent sulfate. Essentially, all the cations were sodium and potassium. ^{137}Cs (8,100,000 Ci) and its short-lived daughter ^{137}Ba (7,600,000 Ci) made up greater than 99 percent of the radioactivity in the supernatant (3, 2). The liquid THOREX acid waste consists of 71 weight percent nitrate salts and 29 weight percent water. Although the THOREX makes up only approximately two percent of the total high-level waste volume, it is an important consideration in the formulation of the reference borosilicate glass.

In order to prepare the waste for melter feed and to reduce the volume of glass produced, the WVDP is pretreating the HLW. Pretreatment involves decontaminating the supernatant by ion exchange, washing the sludge, decontaminating the sludge wash solution, and combining the ion-exchange material and the THOREX waste from Tank 8D-4 with the washed sludge in Tank 8D-2. After the wastes are combined, they will be homogenized and fed to the VF. To complete these tasks, WVNS has designed and developed an Integrated Radwaste Treatment System and a Sludge Mobilization and Wash System. The IRTS decontaminates, concentrates, solidifies, and safely stores the decontaminated supernate. The SMWS washes the salts from the insoluble solids by adding a dilute caustic solution and resuspending the sludge. After all the wastes have been combined in Tank 8D-2, the SMWS also will be used to resuspend the settled sludge solids so that they can be removed from Tank 8D-2 and transferred to the VF as SFCM feed.

DISCUSSION

Integrated Radwaste Treatment System

The WVDP started processing Tank 8D-2 supernate with the Integrated Radwaste Treatment System (IRTS) in 1988. The IRTS comprises the Supernatant Treatment System (STS), Liquid Waste Treatment System (LWTS), Cement Solidification System (CSS), and the Drum Cell. The STS removes ^{137}Cs from the supernatant by running it through four ion-exchange columns filled with the inorganic clay Linde Ionsiv IE-96® zeolite (4, 16). United Oil Production (UOP), Inc., formerly Union Carbide, manufactures the IE-96. WVNS selected IE-96 for cesium recovery because of its high exchange capacity, decontamination factors, and compatibility with glass formers.

After the supernatant is decontaminated, it is transferred to the LWTS. The LWTS reduces the volume of decontaminated waste by concentrating the liquid with a steam-heated evaporator to a predetermined salt concentration (5, 2).

Once the waste is concentrated, it is transferred to the Cement Solidification System (CSS), where high-shear mixers combine the concentrated liquid waste with cement and other additives-calcium nitrate, sodium silicate, and an antifoam agent (6, 4). Cement solidification stabilizes the decontaminated supernatant for disposal as low-level radioactive waste.

The cement-waste mixture is solidified in 0.29 m^3 square steel drums. Square drums are used because they maximize storage efficiency (35 percent greater than round drums) and provide enhanced storage stability. These drums are stored on site in an above ground Drum Cell with drum retrieval capabilities, pending a decision on final disposition.

Supernatant Processing Results

Supernatant processing was started in 1988 and completed in 1991. Separating the nonradioactive salt species from the ^{137}Cs contained in the supernatant reduces the volume of waste glass produced by a factor of six (approximately). The WVDP has processed approximately 1,704 m^3 of the original PUREX supernatant through the IRTS and produced 10,400 drums of low-level Class C cement. The Decontamination Factors (DFs) for ^{137}Cs averaged 40,000. 42,000 kgs of cesium-loaded zeolite was generated. The acceptance rate for the cement, based on U. S. NRC Acceptance Criteria, was in excess of 99.6 percent. All the cement drums produced during supernatant processing exceeded the 85 percent full acceptance criteria, contained no free water, and maintained increasing long-term strength as verified by compression tests with actual cement waste core samples. Compressive strength for cement during supernatant processing was on the order of 11,000 KPa. The LWTS consistently concentrated the liquid supernatant through the evaporator to meet the West Valley cement recipe criteria of 37 to 41 weight percent salt solution.

Sludge Mobilization and Wash System

The Sludge Mobilization and Wash System (SMWS) was designed to resuspend the insoluble solids in the sludge, wash the salts from the sludge, and decontaminate the wash solutions to ensure that the cement product does not exceed Class C limits. A series of five low-pressure, high-flow, long-shafted centrifugal pumps installed in Tank 8D-2 are used to resuspend the settled sludge solids. The mobilization pumps mix the solids and added caustic solution, accelerating dissolution of salt crystals. Once the desired salt concentration is attained, the mixing is stopped and the solids are allowed to settle. After the solids have settled, the wash solution is drawn from Tank 8D-2 with a floating suction removal pump and transferred to the IRTS for processing.

Sludge washing started in October 1991 and will continue through 1995. Washing the sludge and treating the resultant wash water solutions through the IRTS further reduces the HLW volume. Washing is necessary to remove the high concentration of crystallized sulfate salts, as well as the intersticial supernatant, from the sludge.

SFCM operations limit the solubility of sulfate in the borosillicate glass to 0.3 weight percent. Any sulfate-ion concentration in the melter feed that exceeds this limit will form a molten salt layer on the surface of the melt; this will result in an unacceptable glass product. Furthermore, if the sulfate is not reduced, the amount of waste solidified in the glass will have to be decreased and the number of glass canisters substantially increased. The sludge will be washed three times in order to reduce the sulfate concentration to the required level.

Waste projections for vitrified glass after pretreatment are 300 canisters, each containing 0.756 m^3 of stable borosilicate glass. After each sludge wash, the resulting wash solution is processed through the IRTS in the same manner as the supernate.

Sludge Wash Process Development

In 1987 and 1989, sludge samples were removed from Tank 8D-2 and used for sludge wash development testing and cement waste form qualification tests. A scaled experiment was performed using 200 gm of sludge at pH 10 and process water in the same ratio as that chosen for the actual sludge wash process. During this experiment, much of the plutonium (approximately 23 percent) and nearly all the uranium in the sludge went into solution. Processing this solution through the IRTS would have changed the status of the waste from Class C to Transuranic (TRU). It was determined that the high carbonate to bicarbonate content of the test solution was causing the increase in solubility of the plutonium and uranium content.

The WVDP was committed to maintaining the bulk of the radionuclides in the Tank 8D-2 sludge to permit solidifying of these radionuclides in glass rather than cement; therefore, after the initial experiment was performed, different wash chemistries and wash solution treatment technologies were evaluated. From the evaluations of the wash chemistries it was determined that adding a dilute caustic solution (10 g/L) to the sludge increased the pH to 12.5, which greatly reduces plutonium and uranium dissolution from the sludge. The exact chemical reaction that occurs in the pH controlled wash is not known: controlling the pH of the sludge wash water either prevents the plutonium and uranium compounds from dissolving in the wash solution or precipitates the plutonium and uranium as hydroxides. Although the plutonium concentration of the solution was lowered, additional plutonium removal capacity was desired to ensure a Class C cement waste. A special titanium treated ion-exchange medium with the capability of removing ^{137}Cs, ^{90}Sr, and plutonium was developed for use in the STS.

Ion Exchange Development

Battelle-Pacific Northwest Laboratory has developed a new ion-exchange material, Ionsiv IE-96 zeolite treated with titanium (TIE-96), with the capacity to remove plutonium from the WVDP supernate and sludge wash solutions. The use of titanates to decontaminate radioactive aqueous solutions is well documented in the chemical literature.

Although the primary function of the titanates is to remove ^{90}Sr, these compounds also are very effective at removing specific actinides. Extensive tests performed at the WVDP as well as Battelle-PNL demonstrated that TIE-96 could remove quantities of ^{137}Cs from the WVDP supernatant comparable to that removed with IE-96. In addition to removing ^{137}Cs from the solution, TIE-96 also removes trace quantities of plutonium, as well as ^{90}Sr, over a wide range of operating conditions: temperature, pH, and concentration. The exact mechanism responsible for plutonium removal is not entirely understood; however, it is known that the plutonium removed by the TIE-96 remains on the ion-exchange column under anticipated sludge wash processing conditions.

After successful lab-scale testing of titanium-treated zeolite prepared by Battelle-PNL, United Oil Production, Inc., was selected as the commercial supplier of the TIE-96 zeolite used for the sludge wash process.

Mobilization Equipment Development

In 1985, the WVDP performed on-site tests with a one-sixth scale model of Tank 8D-2 to validate that the sludge could be resuspended and removed from the tank bottom using multiple mixing pumps, as demonstrated with full-scale equipment at other DOE facilities (7, 3).

Tank 8D-2 is 21 m in diameter and 8.3 m in height. Tank 8D-2 has an elaborate internal gridwork consisting of I-beams and plates supported from the floor by 38 mm rods. The rods support the steel plates at least 0.25 m above the floor. The vault roof is supported by six concentric columns that penetrate the tank roof through six 1.22 m in diameter sleeves. This internal structure was designed to stiffen the tank floor and act as bearing support for the tank roof. The internal structure of Tank 8D-2 severely complicated the task of resuspending the insoluble sludge through one access opening. One-sixth scale tests demonstrated that five mixing or mobilizing pumps would be required to resuspend the entire sludge layer (7, 23). The tank, therefore, had to be modified to permit the installation of the required number of pumps.

A single access opening was originally constructed with Tank 8D-2. Eight new 0.61 m in diameter access risers were installed for mobilization and waste removal operations. Because of the level of radioactivity in Tank 8D-2 (> 400 R/hr at the tank roof), remote installation techniques had to be utilized (8, 3). Jackson and Gessner have described the remote riser installation at length (9). Installing the risers took approximately a year. Installation began in 1986 and was completed in 1987.

The SMWS uses five low-pressure, high-flow centrifugal pumps to resuspend the settled sludge. In Fig. 1, a pump installed in a tank access riser is shown. The pump volutes discharge beneath the bottom gridwork of Tank 8D-2, approximately 0.30 m from the tank floor through two opposed, tangential nozzles. Each nozzle discharges 38 L/s. Each pump is supported from a 15.25 m column constructed from pieces

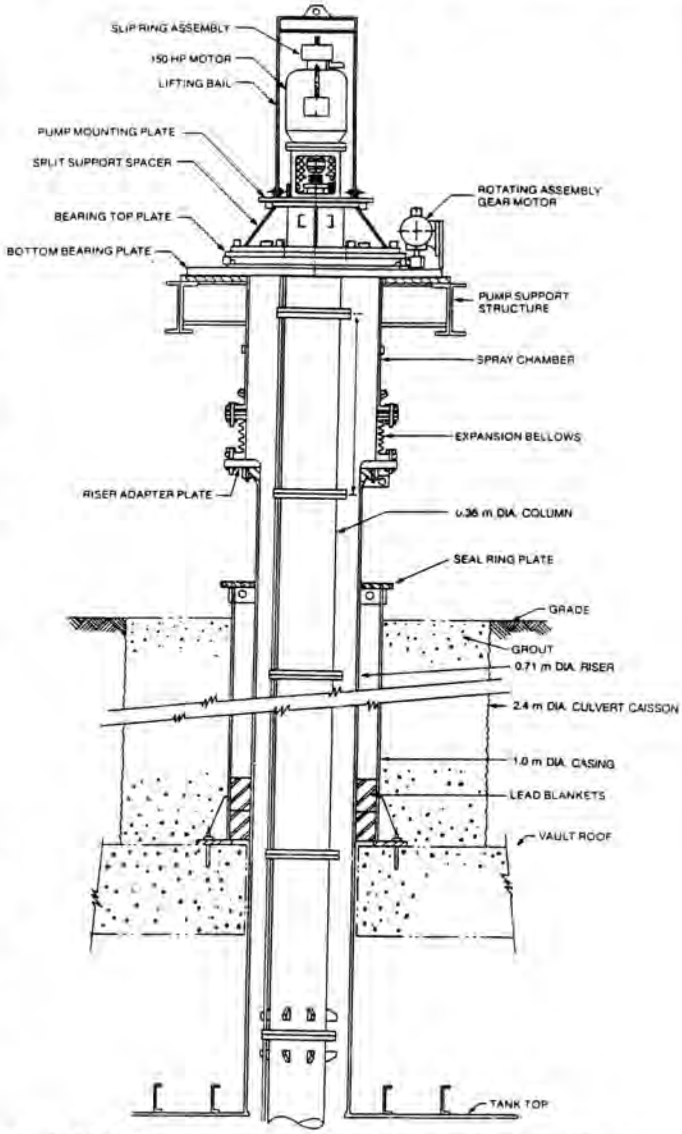

Fig. 1. Mobilization pump installed in a tank access riser.

of tubular stainless steel pipe. These columns house the pump drive shaft. The pumps are continuously rotated 360 degrees at a rotational speed of 0.5 rpm. A 110 kW motor vertically mounted at the top of the shaft drives the pump. The pump rotating assembly, upper mechanical seal, and motor are mounted out of the tank. Because the sludge layer obstructed the placement of the pumps, the mobilization pumps were placed on sets of split shim spacers to set the pump suction above the sludge layer. The mobilization pumps were custom fabricated to WVNS specifications by Floway Pumps of Fresno, California, in 1986.

Tank 8D-2 and its vault were not designed for the mobilization equipment loads; for this reason, an array of pump support trusses had to be constructed over the tank vault. The pump support structures consist of steel trusses that rest on concrete piers and spread footing foundations. After the trusses were placed, flexible expansion bellows and spray chambers were installed to complete the tank confinement barriers between the risers and trusses (Fig. 2). The expansion bellows enable the trusses to move slightly without imparting loads to the risers and tank vault. The spray chambers will be used during pump removal to facilitate decontaminating the pumps.

Conventional and semi-remote construction methods were used to install the sludge mobilization pumps. The first components installed were those that did not require removing the shield plugs from the access risers. These included the pump support components, such as the pump rotating assembly and split shims. Next, a containment tent was placed over the riser to provide contamination control and the shield plug was removed. The pump was then lifted with a crane from a horizontal to a vertical position, placed over the riser, and lowered into the tank. Mounting the vertical motor on the pump motor stand, which completed the installation, was performed using conventional methods. The pump installation was performed by WVNS operators using controlled procedures. A Radiation and Safety Specialist monitored the area for radiation levels to minimize operator exposure over the open riser.

SMWS Start Up and Equipment Check Out

During SMWS check out, WVNS Quality Assurance verified that the installation of the sludge mobilization components was accomplished in accordance with the system design drawings, installation specifications, and work procedures. The SMWS check out included a pre-operational test program, which consisted of three checkouts: factory, field, and start up. The pumps were run through a series of acceptance tests. A formal Operational Readiness Review (ORR) was conducted according to a controlled ORR Plan prior to start-up approval. Reviews were performed by both WVNS and the DOE West Valley Project Office (WVPO).

SMWS Operation

The sludge mobilization pumps had to be operated above the sludge layer before they could be lowered into their final position. The pumps were initially placed on sets of 76.0 mm thick split shims to set the pump suction approximately 150.0 mm above the sludge layer. Lowering the pumps into their final position involved operating the pumps above the settled sludge layer and gradually lowering them into the tank by removing split shim spacers. Aluminum split shims were used to reduce the amount of weight that had to be manually lifted by the operators during shim removal.

Buoyancy probe measurements were used to lower the pumps from their initial to their final position. The buoyancy probe consists of a stainless-steel weight suspended by a piece of flat tape. The probe is raised and lowered with an electrical motor through a spare riser in Tank 8D-2. The weight and location of the probe are constantly monitored by a load cell and mechanical totalizer. As the probe touches the sludge, the load cell readings decrease. The totalizer then indicates the depth of the probe relative to the riser. The height of the sludge layer was initially measured prior to running the pumps to establish a base line for the effectiveness of sludge resuspension.

While the mobilization pumps were running, approximately 40 m^3 of 20 weight percent caustic, followed by 225 m^3 of demineralized water, was added to the tank. The pumps were continually operated for eight hours to ensure that the sludge and caustic solution were thoroughly mixed. Mixing the solids and added caustic solution accelerates the dissolution of salt crystals. After the pumps were shut down, a new buoyancy probe measurement was taken. The difference in the sludge layer height was used to determine the number of split shims that could be removed after a run. Each pump was

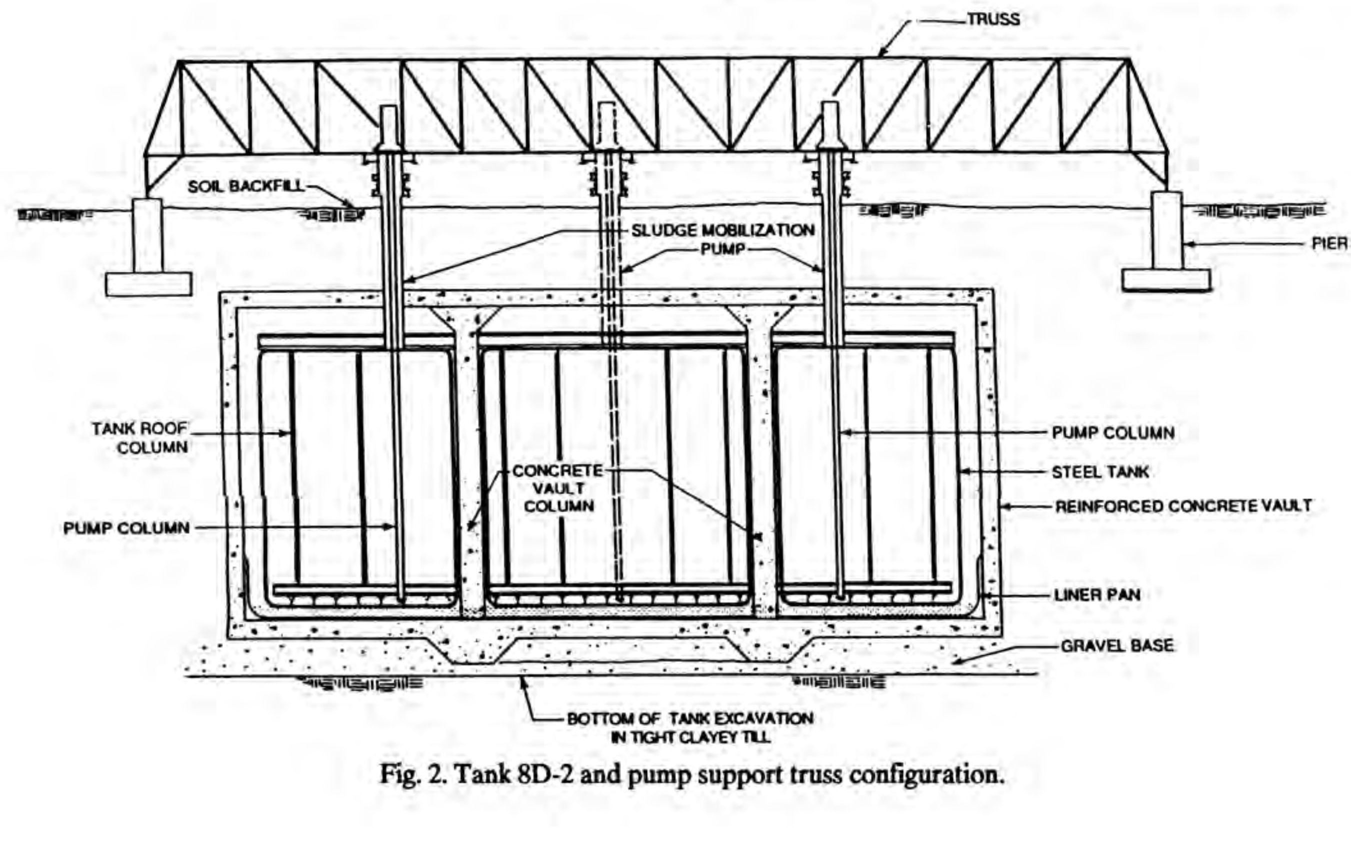

Fig. 2. Tank 8D-2 and pump support truss configuration.

lowered by slowly lifting the pump with the mobile crane, removing shims, and lowering the pump. This procedure was repeated until all the shims were removed and the pumps were mounted to the rotating assembly.

Once all the pumps were lowered into their final position, the tank contents were mixed for three days. The exact duration of mixing was determined by periodically sampling the liquid and tracking the change in salt concentration in the solution. After the salt concentration had leveled off, the mixing was stopped and the solids were allowed to settle. Once the solids had settled, the wash solution was drawn from Tank 8D-2 and transferred to the STS.

The major process components of the STS are installed in Tank 8D-1. These components include: a prefilter, feed tank, supernatant cooler, the four ion-exchange columns, and a filter for decontaminated supernatant (4, 25). The solution is cooled to less than 13°C to improve the cesium removal efficiency on the ion-exchange material. Once the solution was cooled, it was pumped, in series, through the ion-exchange columns. Each ion-exchange column contains 1.5 m^3 of zeolite (4, 10). It is estimated that decontaminating the three sludge washes will require eight columns of TIE-96. The decontaminated wash solutions will generate approximately 9,000 additional drums of low-level waste. Waste loading in the cement is limited to approximately 20 weight percent salts because of the increased level of sulfate in the wash solution; however, because of the low concentrations of fissile material in the cement, the waste form complies to Class A requirements.

Sludge Wash Results

Three washes are planned to remove the required sulfates from the PUREX sludge. The first sludge wash proved effective in resuspending the settled solids from the tank bottom. Lowering the mobilization pumps into the sludge layer by shim removal was accomplished in three separate 48 hour runs. The concentration of sodium sulfate salts in the wash solutions was close to the concentration achieved during the scaled experiment with the sludge samples. Excellent mixing was obtained using five mobilization pumps as predicted during the one-sixth scale model tests. Using pH control during washing maintained fissile material concentrations in the wash solutions at a level lower than predicted during the scaled experiment. The titanium-treated zeolite used to treat the wash solution was effective in capturing the plutonium without a reduction in cesium removal. Initial DF calculations for the processed sludge wash solution are as follows: between 30,000 and 40,000 for ^{137}Cs; approximately 12 for ^{90}Sr; and up to 400 for plutonium.

SUMMARY

When PUREX sludge washing is complete, the sludge will be mixed with the THOREX waste and cesium-loaded zeolite and transferred to the VF. At present, the cesium-loaded zeolite is held in Tank 8D-1 under water. Transferring these wastes will involve neutralizing and then mixing the THOREX waste from Tank 8D-4 with the washed sludge in Tank 8D-2 after the third sludge wash. Excess caustic will be used to neutralize this mixture; the resulting precipitate will be washed as part of a fourth sludge wash. Then, the cesium-loaded zeolite from Tank 8D-1 will be transferred to Tank 8D-2 and combined with the mixture. The final waste mixture will be homogenized using the mobilization pumps and then transferred to the VF.

Construction of the waste transfer and vitrification facilities are in progress. The waste transfer facilities, which includes piping, a containment trench, and pump pits will be completed in early 1994. Completion of the VF is scheduled for early 1994. The start of VF hot operations are scheduled for January 1996. Figure 3 summarizes the major activities associated with HLW pretreatment and solidification at the West Valley Demonstration Project.

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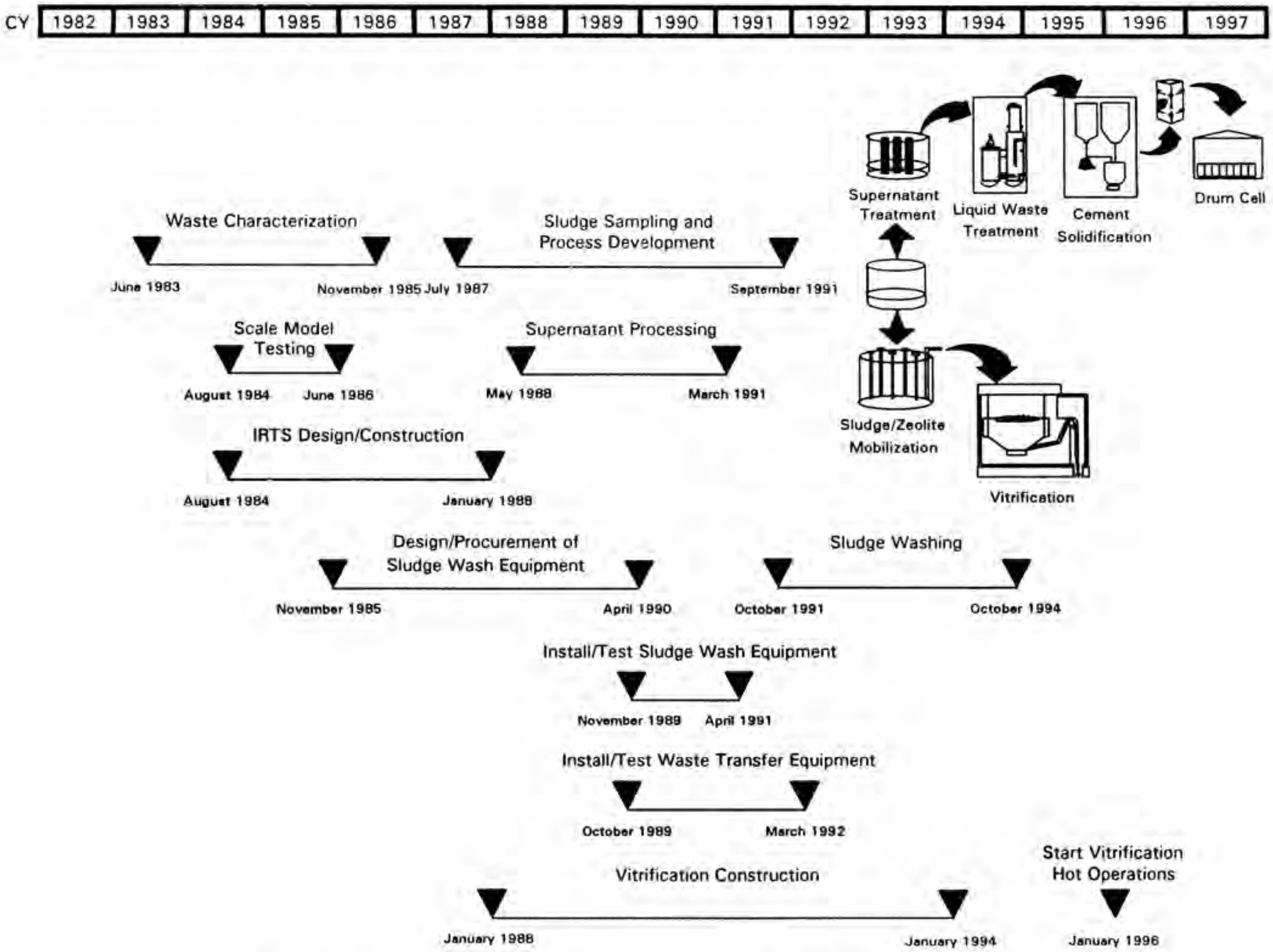

Fig. 3. Summary of WVDP HLW pretreatment and solidification activities.