Pretreatment Engineering Platform - Reducing Technical Risks for the Waste Treatment Plant Pretreatment Facility through Scaled Process Testing - 8365

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ABSTRACT

The Hanford Waste Treatment and Immobilization Plant (WTP) will separate and vitrify (immobilize in glass) millions of gallons of radioactive and chemical wastes stored at the Hanford Site. Pretreatment of the waste by caustic and oxidative leaching processes will minimize the volume of high-level waste (HLW) to be vitrified, and cross-flow ultrafiltration will be used to remove liquids from the HLW solid slurry. An extensive and critical review of the WTP technical bases and design identified the need to demonstrate of the integrated leaching and ultrafiltration processes at greater than bench scale. To respond to this need, the WTP prime contractor, Bechtel National, Inc., and their principle subcontractor Washington Group International concluded a 1/4.5 scale facility to treat non-radioactive waste simulants was needed to demonstrate the process. This paper describes the technical bases and design of the scaled Pretreatment Engineering Platform (PEP) and the strategy to develop waste simulants to be used in the PEP.

INTRODUCTION

The Hanford Waste Treatment Plant (WTP) will receive, pretreat, and vitrify radioactive waste currently stored in underground tanks at the Hanford Site. The WTP consists of three major facilities: high-level waste (HLW) vitrification, low-activity waste vitrification (LAW), and the pretreatment facility. The pretreatment facility (PTF) is central to the strategy for treating Hanford tank waste and minimizing the number of HLW canisters to be disposed at the national repository. Pretreatment operations will separate the bulk of the Hanford chemicals from the highly radioactive components and concentrate the HLW fraction for vitrification. The remaining low activity waste (LAW) fraction contains the bulk of the waste chemicals. It will also be vitrified, but will be disposed at the Hanford site. The PTF separates the HLW and LAW fractions through a series of reactions designed to dissolve (leach) non-radioactive components from the “insoluble” HLW components and concentrate the insoluble materials using ultrafiltration. The soluble highly radioactive cesium is separated from the non-radioactive components by ion exchange and combined with the “insoluble” fraction for HLW vitrification downstream of ultrafiltration.

This paper presents the plans to prepare and conduct large-scale, confirmatory testing of the caustic leaching, oxidative leaching, and ultrafiltration pretreatment processing operations. Integrated testing of the WTP leaching processes has not been completed at larger than bench scale. Consistent with recommendations from an External Flowsheet Review Team (EFRT) and the Department of Energy Office of River Protection (ORP), the WTP Project concluded that lab- and bench-scale experiments are capable of defining the leaching chemistry, but they will not necessarily represent the integrated leaching, washing, and filtering process performance at full-scale [1]. Scale-up data is needed to improve the WTP productivity projections for treatment of the Hanford wastes. Integrated engineering scale test results will significantly reduce the technical risk to critical WTP pretreatment facility processes [2].

In response to the need for engineering scale, integrated process testing, the WTP Project is designing and constructing the Pretreatment Engineering Platform (PEP), developing simulants, and developing the testing matrix. The PEP will be a 1/4.5-scale facility with prototypic vessels and process equipment [3]. It has been designed to perform a demonstration on an engineering scale to confirm the PTF leaching and ultrafiltration process (UFP) equipment design and waste treatment process flowsheet. The unit operations to be tested include pumping, solids washing, chemical reagent addition and blending, heating, cooling, leaching, cross-flow ultrafiltration, and filter cleaning.

Overview of the WTP Leaching and Ultrafiltration Process

The purpose of the WTP PTF flowsheet is to concentrate high-level radioactive waste solids and leach (dissolve) non-radioactive solids (aluminum and chromium) that limit the waste loading in high-level waste glass. The PTF includes a caustic leaching process (for dissolution of aluminum solids), an oxidative leaching process (for dissolution of chromium solids), ultrafiltration processes (for washing and concentrating solids), evaporation process, and cesium removal by ion-exchange. The PEP includes the leaching and ultrafiltration processes of the PTF (see Fig. 1). Feed to the PTF can include HLW, LAW, and Feed Evaporation Process (FEP) concentrates. The PTF produces concentrated high-level radioactive solids for HLW glass immobilization and high-sodium solutions that are sent forward to LAW glass vitrification.

The PTF leaching and ultrafiltration processes (UFP) consist of two parallel systems of feed-preparation vessels, ultrafiltration feed vessels, ultrafilters, permeate collection vessels, and associated heat exchangers, pumps, valves, and piping. The PEP duplicates one line of the WTP system with prototypic feed vessels, feed preparation vessels, and ultrafilters. The PEP includes a second prototypic feed vessel to accommodate continuous operation of the leaching process.

The WTP design supports two alternative flowsheets that differ primarily in whether caustic leaching is conducted as a dilute slurry (in the ultrafiltration feed preparation vessels) or after initial dewatering by ultrafiltration (in the ultrafiltration feed vessel). In both flowsheet cases waste is combined with 19M caustic and the resulting slurry is heated by direct steam addition to 100ºC. This temperature is maintained until the required fraction of leachable aluminum solids have been

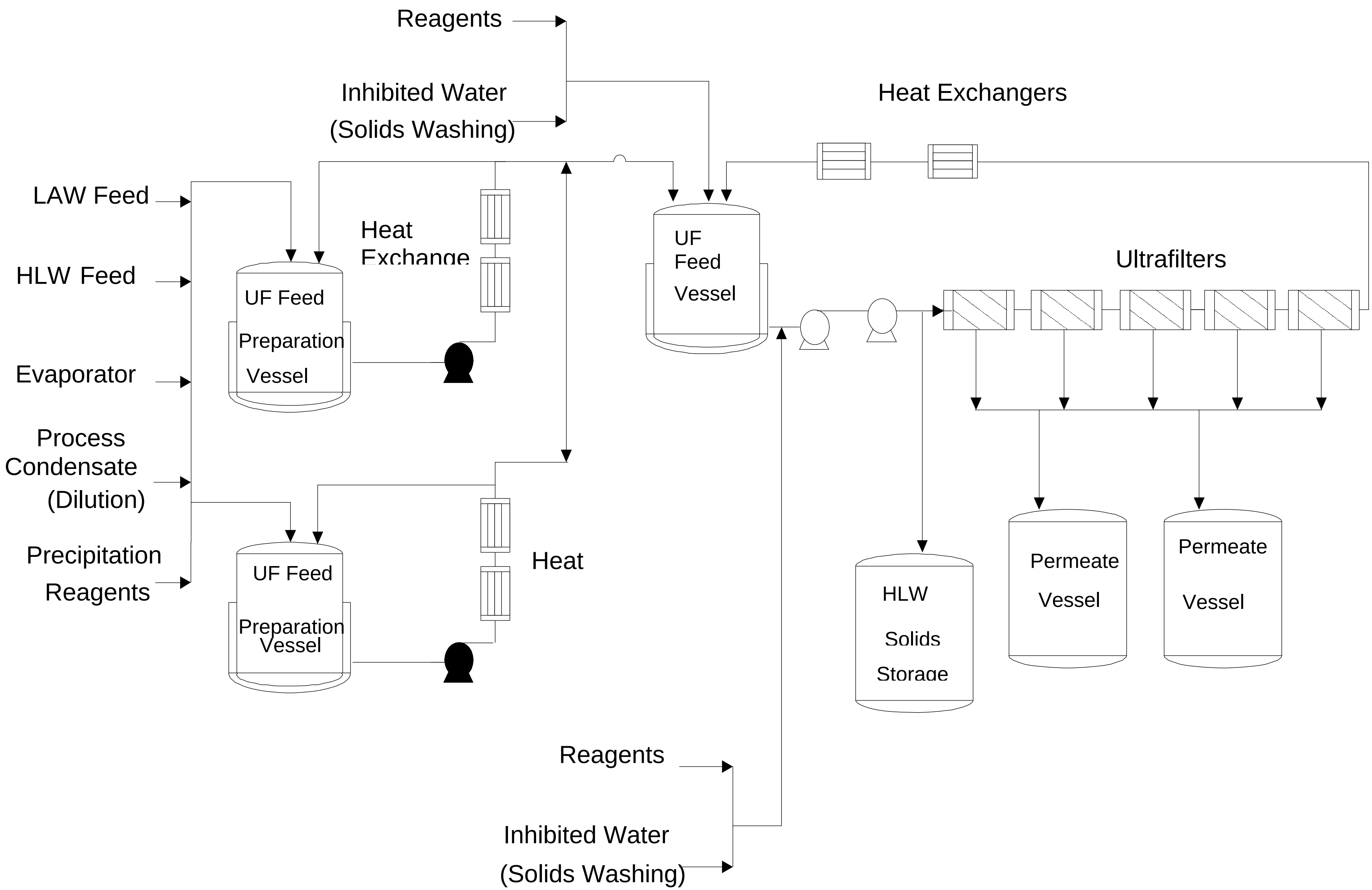

Fig. 1. Pretreatment Engineering Platform—Process Flow Diagram.

dissolved by caustic leaching. After caustic leaching, the slurry is cooled and a concentrated slurry washed to remove the dissolved aluminum and caustic. Sodium permanganate is then added to the slurry to oxidatively leach chromium solids. After oxidative leaching, the slurry is washed to remove dissolved chromium and concentrated to approximately 20 wt% solids. This concentrated slurry is finally pumped to the HLW vitrification process.

The primary advantage to conducting caustic leaching in the ultrafiltration feed vessel is that it allows the slurry to be concentrated to about 20 wt% solids before caustic leaching is conducted. This reduces the amount of caustic needed, which translates to less sodium eventually sent to the low activity waste melter. However, caustic leaching in the ultrafiltration feed vessel results in these vessels being used for the lengthy heat-up, caustic leach, and cool-down steps, as well as for feeding the ultrafiltration loops. Performing all of these process steps in the ultrafiltration feed vessels can limit throughput for some waste types. Caustic leaching in the ultrafiltration feed preparation vessels improves throughput by increasing the time the ultrafilters can be used to wash and concentrate solids, but has the disadvantage of increasing the amount of caustic used.

Testing Objectives

The objectives of the tests are to demonstrate the planned PTF leaching and ultrafiltration process. Specifically,

- Demonstrate the flowsheet and equipment design concept for separation and treatment of HLW sludge in integrated non-radioactive testing. Testing will demonstrate the
    - Caustic leaching process,
    - Oxidative leaching process, and
    - Ultrafiltration washing and solids concentration processes.
- Provide scaled system performance data to facilitate estimating WTP performance.
- Perform integrated operation of the processes using prototypic equipment to demonstrate
    - Operating modes
    - Process control approaches.

In addition, this test program is designed to evaluate proposed design or process changes intended to enhance the overall throughput capacity of the pretreatment facility and remove potential pinch-points identified by WTP process model assessments.

To achieve these objectives, a testing program was developed to determine key waste characterization and process performance data for the various nuclear wastes stored at Hanford, develop non-radioactive simulants for the integrated process tests, and design and construct the PEP. Each of these project tasks and the planned integrated tests are described below.

HANFORD WASTE CHARACTERIZATION

To support the simulant development program, six sludge and two salt cake waste types were identified that represent over seventy five percent of the waste mass to be processed at the WTP. The waste types and their relative abundances in the Hanford Best-basis Inventory are indicated in Fig. 2 [4]. Archived samples from multiple tanks judged representative of each waste type were obtained and composited. Compositing was necessary to obtain the quantities (liter/kilogram) of actual waste needed for testing.

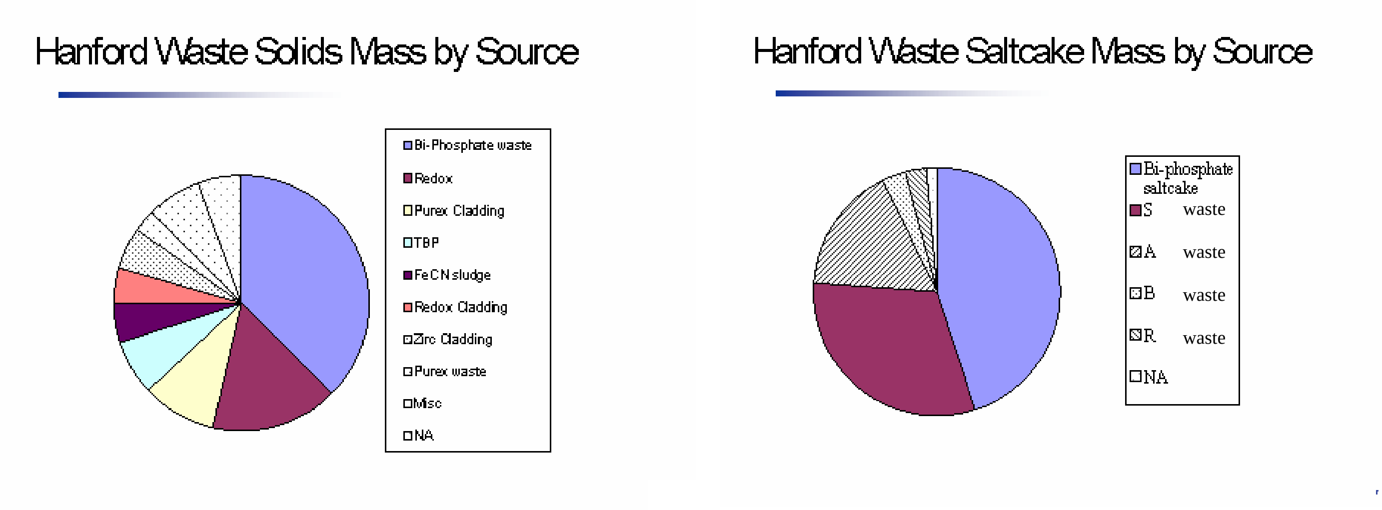

Fig. 2. Hanford Tank Wastes by Source.

Physical and chemical characterizations of the waste composites are being conducted. Physical characterizations will determine material properties (e.g., crystal form of the solids, crystal habit and morphology, particle size distributions, specific surface area subject to dissolution, rheology, solution and solids densities, and solids fractions). Chemical characterizations will include composition, dissolution kinetics, and equilibrium solubility. Dissolution kinetics studies will address the caustic leaching rates of aluminum and phosphates and oxidative leaching rates of chromium and the extent of plutonium dissolution. Filtration characteristics of the actual waste samples (if adequate waste composite quantities exist) will also be performed.

SIMULANT DEVELOPMENT AND TESTING

A basic premise of the simulant development approach is that component simulants can be developed to mimic specific characteristics of waste components, and then blended as appropriate to simulate a broad variety of Hanford wastes. The chemical component simulants for the sludge will include:

- Gibbsite, $Al(OH)_3$, to represent rapidly leached aluminum,
- Boehmite, AlOOH, to represent the slowly leached aluminum,
- Chrome hydroxide, $Cr(OH)_3$, to represent soluble chromium solids,
- Chrome oxyhydroxide, CrOOH, to represent less soluble chromium solids,
- Phosphates and oxalates, to represent relatively low solubility salts that tend to precipitate when solutions are cooled or sodium levels are increased, and
- Iron hydroxide (with minor amounts of Zr, Mn, Ca, Mg, Ni, Nd, Ce, and Pb) to represent insoluble solid fines.

The simulant development approach will be performed in two stages. The initial stage will provide component simulants by blending amounts of the listed sludge components based on existing waste characterization and laboratory experiments to match reaction mass loss performance. Specifically, the initial simulant should exhibit greater total solids mass loss (via leaching and washing) than 80% of the waste, and the batch processing time should also be greater than that for 80% of the waste. The component simulants from this stage will be used in the Phase 1 integrated demonstration. Note that this stage will only develop simulants for aluminum leaching, chromium leaching and filtration. The second simulant development stage involves developing simulants to represent ranges of tank waste behaviors and will be based on the results of the laboratory scale testing with actual wastes.

The objectives of the laboratory simulant testing and analyses are to:

- Develop data on caustic and oxidative leaching, filtration and mixing behavior over a range of wastes and processing conditions, and
- Develop scaling information required for the design of the integrated demonstration system.

These tests are carried out in laboratory scale equipment, and use liter quantities of simulant feeds under well-mixed conditions similar to the bench-scale tests with the actual wastes. The simulant tests may be limited to bounding conditions of the waste types that will be identified during the actual waste testing. The simulant tests will be broader in scope in that they will include a broader range of parametric testing for leaching and filtration.

ENGINEERING SCALE CONSIDERATIONS

The PEP will be used to demonstrate the individual and integrated WTP leaching and ultrafiltration processes. Process monitoring instrumentation similar to that of the full-scale facility has been incorporated in the PEP design, so the PEP will be operated and controlled similarly to the full-scale facility. The intent is that the PEP facility will perform in a similar manner to the full-scale facility, demonstrating those aspects of the processes that meet expectations and helping to identify potential problems in the full-scale facility [5].

The PEP also will be used to improve predictions of the full-scale facility performance. The improved full-scale facility performance predictions are based on process data from a single scale (the 4.5-scale) and on laboratory testing of simulants and actual wastes that have been performed under idealized conditions (uniform temperature, constant and uniform mixing, etc.). Scale-up is not, therefore, based on a series of simulation tests conducted at different scales. This approach requires the ‘scale-up’ of the 4.5-scale process data to full-scale to be well understood and quantifiable. Though the PEP has been designed to simplify this scale-up of process data and mimic the behavior of the full-scale facility to the extent possible, there are scaling inconsistencies between different aspects of the processes that can not be resolved *a priori*.

Scaling considerations for different process operations and the scaling inconsistencies between them are discussed in the following sections. A strategy to address these issues with the PEP is also presented, along with identification of specific integrated tests.

Scaling of Mixing Behaviors

Similarity principles have been used to scale the mixing behavior of pulse jet mixers (PJM) and other fluid jets, such as the filter loop return nozzle. As described in *Overview of the Pulse Jet Mixer Non-Newtonian Scaled Test Program* [6], scaled mixing of non-Newtonian materials can be accomplished by maintaining the same jet velocities in the full- and 4.5-scale vessels [3]. In the scale system, fluid jet nozzle diameters are scaled by 1/4.5 and the volumetric flow rate by $1/4.5^2$. It is currently assumed that this scaling law is also valid for Newtonian fluid mixing.

When considering the time required to homogenize the contents of the vessel to some arbitrary extent (referred to here as the vessel blend time) equal jet velocities in the 4.5- and full-scale vessels will result in shorter blend times in the smaller system (scale-time). When the jet velocities are equal, the blend time in the 4.5-scale system should be 4.5 times shorter than in the full-scale system. Vessel blending is important in the PEP testing whenever solids settling or solids redistribution are important.

Though vessel blending from PJMs and other fluid jets is accelerated in the 4.5-scale vessels, the turbulence associated with the jets will be approximately the same. This aspect is potentially important to the caustic and oxidative leaching operations, which may be affected by turbulence.

Air sparging will be used to mix the ultrafiltration feed vessels during caustic leaching. Air sparging rates will be scaled by matching the superficial velocity of the sparge air. When the superficial velocities of the sparge air are the same in the 4.5- and full-scale vessels, slurry velocities will be approximately the same, and the mixing behavior at the two scales is approximately the same.

Scaling of Ultrafiltration Operations

Performance of the cross-flow ultrafiltration tubes is highly dependent on their length, diameter, and permeability. The pressure decrease from the inlet to the outlet of the filter tube affects the pressure differential between the inside (where the solids-containing slurry flows) and the outside of the tube, which in turn affects the permeate flux. Slurry flow rate and tube diameter strongly affect the turbulence within the tube, which is itself important to limiting the buildup of solids on the inside of the tube. Expert recommendations are *not* to scale filter dimensions, cross-filter pressure differential, or permeate flow rate. Based on their recommendations:

- The PEP will use full-scale filter tubes. The filter tubes will be identical to the full-scale plant filter tubes in length, diameter, and permeability.
- The volumetric flow rate of fluid through individual filter tubes will be the same as that of the full-scale system.

Additionally, because the accumulation of particles on the filters affects the flux of permeate through the filters, and the accumulation of particles is affected by the total volume of fluid filtered per area of filter, the volume of filtrate/filter area ratio should be the same in the full-scale and the PEP. Given the total volume of slurry in the PEP will be reduced by a factor of 4.5^3, to properly mimic the permeate flux and accumulation of particles on the filters, the filter area also should be scaled by a factor of 4.5^3. This will preserve the (total mass of solids)/(filter area) ratio, and result in the filtration operation being conducted over the same time in the PEP as in the full-scale plant (plant-time).

The preferred scale for the filtration processes (plant-time) is thus inconsistent with the preferred time scale for vessel blending (4.5x faster). This is an issue because filtration rates can be affected by the solids content of the slurry, which in turn can be affected by the blending of solids in the feed vessel. This time scale inconsistency will be resolved by configuring the filter loop to allow different filter bundles to be used and by conducting ultrafiltration in two different operational modes.

The first operational mode will conduct both mixing and filtration at scale-time. This mode considers the fact that mixing in the 4.5-scale leach vessel is most accurately conducted at scale-time. The ultrafiltration loop would be configured with extra filter bundles to achieve a filtration rate that is approximately 4.5 times faster than in the full-scale facility. This mode, however, will not have the correct (filtrate volume)/(filter area) ratio. Instead, the (filtrate volume)/(filter area) ratio will be a factor of 4.5 higher than in the full-scale facility, leading to less pore-plugging of the filters and presumably a higher filter flux.

The second operational mode will conduct filtration at plant-time, but mixing at scale-time. This mode preserves the correct (filtrate volume)/(filter area) ratio, but the blending of solids in the ultrafiltration feed vessel is enhanced by a factor of 4.5 over the full-scale facility.

Scaling of Caustic and Oxidative Leaching

The kinetics of dissolution requires that the duration of the caustic and oxidative leaching processes be the same in the PEP and full-scale facilities (i.e., plant-time). The dependence of leaching rates on mixing has not been established, but preliminary testing suggests that leaching rates do not depend on mixing except in low mixing regimes. Testing in the PEP vessels will be done with scaled mixing and reduced mixing to determine the mixing effect.

Scaling of Thermal Distributions

Temperatures in the leaching and ultrafiltration processes will vary from about 25 to 100 °C. Most aspects of the processes do not depend heavily on temperature, so small deviations (i.e., several °C) from the specified process temperatures should not significantly affect the processes. The caustic leaching process, however, relies on a relatively high 100 °C temperature to dissolve the boehmite phase of aluminum at an acceptable rate. Thermal distribution differences between the full- and 4.5-scale leaching vessels are thus potentially important to the evaluation of caustic leaching performance.

Thermal distributions are expected to be geometrically scaled, however, because heat losses via conduction through the vessel walls and via evaporation of water scale by the square of the scale factor (4.5^2), the loss of thermal energy per unit volume of slurry per unit time will be higher in the PEP than in the PTF by approximately the scale factor. So, while the PEP can be made to develop thermal distributions very similar to the PTF, their development and evolution will occur 4.5 times faster than in the PTF, and will not be synchronous with the plant-time PEP leaching process. Of particular concern is the accumulation of steam condensate, which would occur in the PEP at 4.5 times the rate in the PTF and significantly dilute the slurry, affecting leaching rates. Computational fluid dynamics (CFD) modeling is being conducted to help evaluate these thermal issues. The goal of the CFD modeling is not to provide exact predictions of the thermal distributions, but to provide approximate values for system temperatures, boundary layer thicknesses, and thermal gradients at both scales for comparison. In addition to the CFD analyses, the thermal distributions of the leaching vessels will be examined during functional testing of the PEP.

Pretreatment Engineering Platform Design and Construction

Conceptual design of the PEP was initiated in November 2006. The conceptual design included development of the Process Flow Diagram, Functional Requirements for PEP [7], and PEP Phase I Process Description. The detailed design and fabrication contract was awarded to WGI-Engineered Products Department and Tessenderlo-Kerley Services in January 2007. PEP has been designed to operate using simulated waste; therefore no shielding or radiolytic design features are included on the PEP. The equipment has been designed as “commercial” with NQA-1 quality standards being applied to instrumentation that will collect data contributing to the WTP nuclear design. The design strategy was to construct the PEP in a modular configuration, so the PEP can be constructed off-site, shipped, assembled, and tested. The modular design allows not only integrated testing, but also unit operation testing. Each module was designed to be controlled by a separate PLC or integrated into the central control for integrated testing.

The detailed design, fabrication, and assembly were structured as a fast track project, therefore the fabrication and procurement was initiated upon completion of the 30% design. The PEP test results are needed by WTP to confirm the Ultrafiltration Process (UFP) design. UFP design confirmation is needed by January 2009, therefore the PEP must be designed, fabricated, installed, acceptance tested, and commence phase 1 testing in 2008 to support completion of the UFP design. The test results will also validate WTP model inputs for calculating the overall throughput of the WTP Pretreatment Facility and estimated quantity HLW and LAW canisters. As described above, the leaching and filtration performance will impact not only the number of glass canisters produced by WTP, but can also impact the waste throughput of the HLW and LAW vitrification facilities.

In parallel with the design of the process equipment, a facility that will house the PEP was also selected and prepared for installation and operations. Several facilities in the Hanford area were considered and reviewed for PEP operations [8]. The analysis concluded that the Battelle Processing Development Laboratory-West (PDL-West) is the best location to house and operate the PEP. The PDL-West facility includes a 20-ton overhead crane, existing environmental and building use permits that require minimum modifications, and conduct of operation procedures in place to conduct demonstration testing. The main process corridor in PDL-West is approximately 102-ft by 50-ft and 35-ft high. In addition to the process corridor, there are two adjacent rooms, electrical room and control room, which will be utilized to operate the PEP. Because the PEP requires the entire main corridor, utility skids are designed to be located outside the building to deliver steam, chilled water, compressed air, and off-gas blower/stack.

Figure 4 shows a rendering from the 3-D design model for the PEP. Waste simulant feed can be received and stored for processing in 4000 gallon non-prototypic tanks. Prototypic processing begins when the simulant is transferred to ultrafiltration feed preparation vessels. The composition of simulant, confirmed by sampling in the simulant storage vessels will determine the required treatment processes.

PRETREATMENT ENGINEERING SCALE TESTING

The scaled system is designed to simulate the full-scale WTP pretreatment processes including caustic leaching, oxidative leaching, solids concentration by ultrafiltration, and slurry washing. PEP testing will also be used to assess a variety of process steps, such as line and filter flushing, PJM operation at high temperature, filter back-pulsing and filter cleaning protocols. The initial PEP tests will involve two stages: functional process testing and integrated process testing. The tests will utilize a non-radioactive simulant formulated to mimic caustic and oxidative leaching and ultrafiltration behavior at the 80 percentile level of expected Hanford wastes.

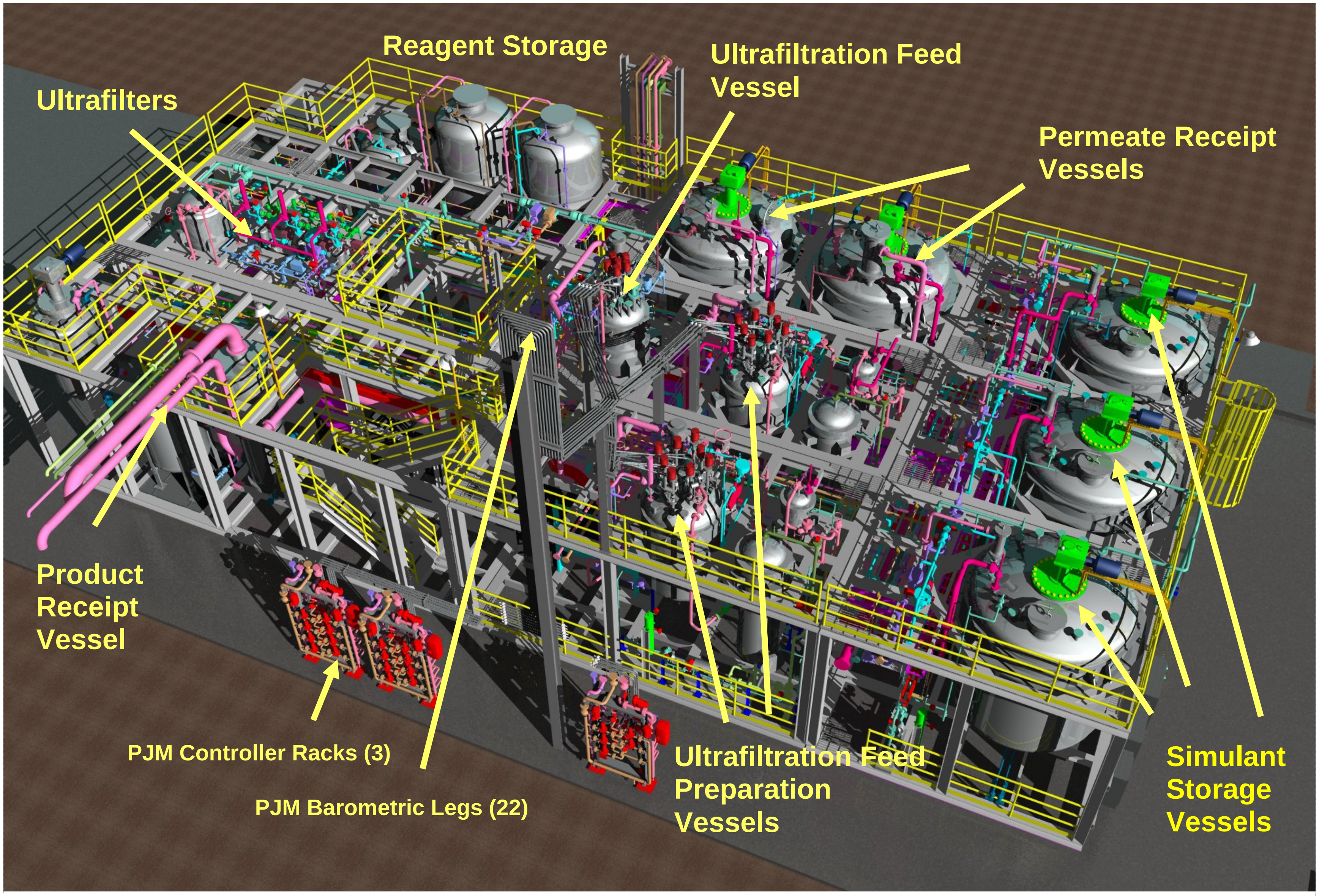

Fig. 3. Pretreatment Engineering Platform—3-D model Rendering.

Additional testing will be conducted at a later time to assess the specific impacts of different Hanford waste types on the PTF processing and optimize operations. Only the initial testing is discussed here.

Functional Process Testing

Functional process tests will be performed to confirm PEP equipment and instrument functionality and quantify performance, and determine limited baseline process/equipment performance needed to perform integrated testing. The functional process tests will include the following:

- Quantify mixing in prototypic vessels with PJMs.
- Characterize the heating of the ultrafiltration feed preparation and the ultrafiltration feed vessels with steam injection and quantify dilution of simulant with steam condensate.
- Determine vessel temperature profiles during the 100 °C caustic leaching process.
- Characterize ultrafiltration performance on slurry over the range 5% to 20 wt% solids.
- Fine tune operation of heating and cooling heat exchangers to maintain desired tank temperature; fine tune serial control of high-pressure centrifugal pumps in ultrafiltration system.
- Demonstrate ultrafiltration system control scheme performance during normal operating modes (e.g., fill and startup, operation, backpulsing, flush and drain, cleaning and return to service).

Integrated Process Testing

Three integrated process tests will be conducted to estimate the performance of the PEP integrated process. The PEP system is expected to have similar performance, but not necessarily identical to the full-scale system. Two parameters that will be tested in integrated process testing whose impacts on process performance are too complex to model *a priori* are PJM cycle time and plugging of filter tubes with fine particles.

Mixing (i.e., turbulence) by the PJMs is expected to be similar in the full-scale plant and the PEP when the PJMs are operated with the same discharge velocity and the same ratio of nozzle cross-sectional area to tank cross-sectional area. As discussed earlier, however, time is not the same in the two systems. Because the PEP system approaches steady-state more rapidly than the full-scale system, the net result is that the PEP will operate closer to its steady-state mixing condition than the full-scale plant. The effect of this time difference is not known and impossible to predict completely. The impact of changing mixing will be determined by conducting integrated tests operating PJMs with "scale-time" (4.5 times faster cycling) and with "plant-time."

The three integrated tests and their technical objectives are described below. In addition to collecting data for the modeling effects of PJM cycle time and ultrafilter area on system performance, the tests provide data to quantify the impacts of implementing caustic leaching in the ultrafiltration feed preparation vessels compared to caustic leaching in the ultrafiltration feed vessel. Abbreviated descriptions of the three integrated tests and conditions are shown below. The sequence of tests for leaching in the ultrafiltration feed preparation vessel versus the ultrafiltration feed vessel will be set based on project data needs.

- *Test 1—C*onduct leaching in the ultrafiltration feed vessel with scale-dependent process parameters selected to maximize similarities between the PEP and PTF.
- *Test 2*—Conduct leaching in the ultrafiltration feed vessel with scale-dependent process parameters selected to contrast those of Tests 1 and 2.
- *Test 3*—Conduct caustic leaching in the ultrafiltration feed preparation vessel to produce a full batch of 20 wt% solids in the ultrafiltration feed vessel after dewatering. Scale-dependent process parameters will be selected to maximize similarities between the PEP and PTF.

CONCLUSIONS

WTP Project, DOE, and external reviewers concluded that the PTF leaching and ultrafiltration processes lacked sufficient operational testing at a large scale, resulting in additional risk for the WTP project. Testing of an engineering-scale system would significantly reduce the overall risk of WTP and provide a test platform to validate the design and underpin the PTF flowsheet. The Project has developed a multi-step program to reduce the technical risk of the UFP process. The program includes laboratory testing of radioactive waste samples and simulants, bench testing of simulants that can be compared and validated against actual waste test results, and lastly perform engineering scaled testing using the PEP to demonstrate performance of the WTP PTF flowsheet.

The main purpose of the PEP is to validate the UFP flowsheet, but additional benefits during the design and fabrication process of the PEP are being incorporated into the WTP design. Design of the control systems and process systems of the PEP are being conducted in parallel with the WTP design, therefore improvements are being shared and incorporated into the design via the lessons learned programs. It is expected that the PEP will continue to reduce the WTP Project risk as the PEP proceeds through testing to confirm design. Contributions to improved performance and development of operating parameters of the PEP will pay great dividends to WTP from the investment in the PEP.

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